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Deflagration-to-Detonation Transition and Detonation Propagation in H₂-Air Mixtures with Transverse Concentration Gradients

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to Kurt

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Kurzfassung

Explosionen in H₂–Luft Gemischen stellen ein zentrales Sicherheitsrisiko in Kernkraftwerken dar. Jüngstes Beispiel ist die nukleare Katastrophe in Fukushima Daiichi im Jahr 2011. Abhängig von Initialbedingungen sowie von der einschließenden Geometrie können verschiedene Explosionsregimes mit stark unterschiedlichen Auswirkungen erreicht werden. Der sogenannte Übergang von Deflagration zu Detonation (DDT) bedeutet hierbei den Schlimmstfall.

Bei realen Unfällen liegen vor der Explosion meist räumliche Gradienten der H_2 Konzentration vor, da H_2 aufgrund seiner geringen Dichte zur Schichtbildung neigt. Umfassendes Wissen zu Explosionen in homogenen Mischungen ist vorhanden, es existiert jedoch ein deutliches Wissensdefizit bezüglich inhomogener Gemische. Die vorliegende Arbeit greift dieses an. Es werden ein-dimensionale Konzentrationsgradienten betrachtet, welche senkrecht zur Hauptausbreitungsrichtung der Explosionsfront orientiert sind. Explosionsversuche in H_2 -Luft wurden in einem Kanal in Laborgröße durchgeführt. Sowohl DDT, als auch Detonationsausbreitung wurden untersucht. Zeitlich hochaufgelöste (laser-) optische Messtechniken kamen zur Anwendung.

Konzentrationsgradienten können verglichen mit homogenen Mischungen zu erheblich stärkeren Explosionen führen und damit höhere Überdrücke, Flammengeschwindigkeiten und höhere DDT-Wahrscheinlichkeit bewirken. Die Annahme homogener Mischung in Sicherheitsbetrachtungen ist daher meist nicht konservativ. Zugrundeliegende physikalische Mechanismen werden identifiziert und quantifiziert. Analytische Verbrennungsmodelle sowie Berechnungen kompressibler Strömung mit detaillierter chemischer Kinetik unterstützen den Aufbau eines umfassenden physikalischen Verständnisses.

Abstract

Explosion of H₂–air mixtures portrays a major hazard in nuclear reactors during severe loss-of-coolant accidents. A recent example is the Fukushima Daiichi nuclear disaster in 2011. Depending on initial conditions and features of the enclosing geometry, different explosion regimes with a wide range of explosion impact can occur. The so-called deflagration-to-detonation transition (DDT) represents the worst-case scenario.

Spatial gradients in H_2 concentration prevail in real-world scenarios, mainly because H_2 stratifies in air due to its low density. Extensive knowledge on explosions in homogeneous mixtures has been accumulated over the last decades. However, a significant knowledge gap exists regarding the influence of mixture inhomogeneity.

This knowledge gap is addressed in the present work. H_2 -air mixtures with one-dimensional concentration gradients, oriented normal to the main direction of explosion front propagation, were studied experimentally in a laboratory scale explosion channel. Both DDT and the detonation regime were investigated. Advanced (laser-) optical measurement techniques at high temporal resolution were applied.

It can clearly be stated that mixture inhomogeneity can lead to significantly stronger explosions in terms of overpressure, flame speed and probability of DDT than homogeneous mixtures. Assuming homogeneous H_2 distribution in explosion safety considerations is therefore often not conservative. The underlying physical mechanisms are identified and quantified in the present work. Besides experimental results, low-order combustion models and computations of compressible flow with detailed chemical kinetics support the development of a comprehensive physical understanding.

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Nomenclature

Latin Symbols

А	Pre-exponential Arrhenius constant	
А	Surface area	[m ²]
a	Speed of sound	[m/s]
a	Thermal diffusivity	$[m^2/s]$
b	Damping coefficient	[N-s/m]
С	Speed of light	[m/s]
c _p	Specific heat capacity	[J/kg-K]
D	Detonation velocity	[m/s]
D	Diffusion coefficient	$[m^2/s]$
d	Diameter	[m]
Ea	Activation energy	[J/mol]
F	Force	[N]
f	Focal length	[m]
g_m^0	Standard-state Gibbs enthalpy	[J/mol]
Н	Channel height	[m]
h	Enthalpy	[J/kg]
h	Planck constant	[J-s]
h	Obstacle height	[m]
k	Reaction rate coefficient	
k	Spring constant	[N/m]
L	Length	[m]
1	Length	[m]
Μ	Molar mass	[kg/kmol]
m	Mass	[kg]

n	Arrhenius constant	[-]
р	Pressure	[bar]
q	Specific heat	[J/kg]
R	Gas constant	[J/mol-K]
S	Burning velocity	[m/s]
Т	Temperature	[K]
t	Time	[s]
U	Voltage	[V]
u	Flow velocity	[m/s]
V	Flame speed	[m/s]
x, y, z	Cartesian coordinates	[m]

Greek Symbols

α	Absorption	[-]
α	Angle	[rad]
β	Zeldovich number	[-]
γ	Specific heat capacity ratio	[-]
ϵ	Viscous dissipation rate	$[m^2/s^3]$
η	Dynamic viscosity	[Pa-s]
θ	Reduced effective activation energy	[-]
λ	Detonation cell width	[m]
λ	Wavelength	[m]
ν	Kinematic viscosity	$[m^2/s]$
ν	Specific volume	[m ³ /kg]
ρ	Density	[kg/m ³]
σ	Expansion ratio	[-]
τ	Time	[s]
Φ	Circle of confusion	[Px]
Φ	Equivalence ratio	[-]
ϕ	Model parameter	[-]
ω	Angular frequency	[1/s]

Superscripts

* Excited state of a molecule' RMS value

Subscripts

а	Activation
abs	Absolute
С	Channel
CJ	Chapman-Jouguet
d	Diffusion
eff	Effective
F	Flame
grad	Gradient
hom	Homogeneous
ind	Induction
L	Laminar
Μ	Markstein
pr	Products
r	Reflected
re	Reactants
S	Stretched
stoich	Stoichiometric
Т	Turbulent
VN	Von Neumann
δ	Heat release zone
η	Kolmogorov parameters

Nondimensional Parameters

Dimensionless amplitude
Flame acceleration model parameter
Karlovitz stretch factor
Karlovitz number
Lewis number
Mach number
Markstein number
Mole fraction, Volume fraction

Acronyms

Obstacle blockage ratio
Computational fluid dynamics
Chapman-Jouguet
Deflagration-to-detonation transition
Flame acceleration
Field of view
Full width at half maximum
Hybrid multiple-prism grazing-incidence
Kelvin-Helmholtz
Loss-of-Coolant Accident
Minimum ignition energy
Neodymium-doped yttrium aluminium garnet
Neodynmium-doped yttrium orthovanadate
Optical segment
Photodiode
Planar laser-induced fluorescence
Rhodamine 6G, Rhodamine 590
Rotational energy transfer
Richtmyer-Meshkov
Rayleigh-Taylor

Run-up distance
Obstacle spacing
Signal-to-noise ratio
Shock wave amplification by coherent energy release
Tunable dye laser absorption spectroscopy
Vibrational energy transfer
Zeldovich-von Neumann-Döring

1 Introduction

Fuel release—flammable cloud formation—ignition—explosion. This sequence of events is commonly involved in explosion accidents, posing a major hazard in industry besides fires and natural catastrophies. Gas, vapor and dust explosions are most relevant leading to worldwide losses in the billions each year. Not only direct damage of industrial plants needs to be considered, but also secondary costs due to business interruptions in highly globalized value chain structures with a high degree of consolidation, which may exceed the direct explosion damage. A high number of severe injuries and fatalities each year shows the tragic side of explosion accidents. Large explosion accidents draw great public attention and can have unforseeable social and political impact.

In the 17th and 18th century, when coal mining developed in Europe, explosion of natural gas and coal dusts in coal mines occurred frequently, mostly with fatal consequences for miners involved. This motivated Sir Humphrey Davy to develop the explosion-safe Davy lamp that reduced the number of accidents drastically. However, still nowadays explosion in mines is a serious hazard which cost about 20,000 lives between 1900 and 1970 [38]. Similar to Sir Humphrey Davy's early scientific efforts, explosion accidents have always been a source of motivation—and funding—for research programs seeking to understand the nature of explosions and to derive preventive and mitigating measures. Besides mining, this particularly concerns process and energy industries. Research programs funded by nuclear energy industry and by onand offshore oil and gas industry have decisively shaped the scientific landscape.

A prominent example from nuclear energy industry is the Three Mile Island accident in 1979 in Pennsylvania, USA [128]. Loss of coolant (LOCA = loss-of-coolant accident) caused a partial core meltdown. A bubble of H_2 formed

in the dome of the reactor pressure vessel through oxidation of zirconium by water steam at high temperature. Zirconium is used as a cladding material to contain the nuclear fuel rods. Explosion of this bubble was apprehended, but no serious explosion occurred due to a lack of oxidizer in the pressure vessel. The imagination of a strong H_2 explosion that might have destroyed the reactor vessel was dismaying. This event raised great concern about explosion of H_2 during LOCAs and marked the beginning of intense international research campaigns. The Fukushima Daiichi nuclear disaster in 2011 in Japan is a similar, more recent example of a LOCA, where severe H_2 explosions in three reactor buildings occurred [138]. Since these explosions took place outside of the reactor pressure vessels, the effect in terms of release of radioactive material was fortunately limited. The disaster sealed the German nuclear power phase-out and evoked a stronger focus on H_2 safety in the explosion research community.

In the sector of oil and gas production, the Piper Alpha accident in 1988 with 167 fatalities demonstrated the hazard related to offshore platforms [113]. It was an initiator for extensive safety programs in Norway [38]. Nowadays, Norway is considered to have the highest safety standards worldwide.

Sound knowledge of explosion physics is of vital importance for the prediction of explosion consequences and for providing engineers with guidelines for implementation of preventive and mitigative measures. Depending on volume and composition of the explosive mixture and the degree of confinement and congestion, different explosion regimes with different hazard potentials can be reached. For a long time, a common approach has been to develop predictive semi-empirical criteria for limits between these explosion regimes. Nowadays, such criteria are more and more substituted by numerical explosion simulation tools, which can be embedded in a comprehensive hazard analysis chain comprising simulation of fuel release and dispersion, explosion and structural response of the confinement. However, the physical complexity of explosion processes leads to a high level of mathematical complexity and a large degree of modeling to keep computational costs at a reasonable level. Extensive modeling can cause significant uncertainty in explosion simulations on industrial scale. Experiments are therefore inevitable not only to further deepen physical insight into explosion processes, but also as a means of validation for numerical simulation tools. Both global parameters such as explosion overpressure or explosion front velocity, but also detailed information on explosion front structure resolved preferentially by optical measurement techniques is required. High temporal measurement resolution is mandatory due to the small time scales of explosion dynamics.

The present work makes a contribution to experimental explosion safety research, particularly focused on explosions in non-uniform H_2 -air mixtures. The research project behind this thesis was dedicated to H_2 safety in nuclear reactor containments, funded by the German Ministry of Economic Affairs and Energy (BMWi).

1.1 Deflagration-to-Detonation Transition

Aforementioned explosion regimes include so-called slow deflagrations, fast deflagrations and detonations. In this order, explosion severity in terms of resulting overpressure increases. Ignition in an explosive mixture usually does not directly initiate a detonation. A flame acceleration (FA) process is required to reach the fast deflagration regime as a precursor for potential onset of detonation. The entire process of FA and subsequent onset of detonation is termed deflagration-to-detonation transition (DDT). If transition to detonation occurs, catastrophic damage must be expected. This explains why the present work places a particular focus on DDT.

High degree of confinement supports DDT. Tube- or channel-like geometries without lateral openings are therefore most prone to DDT, especially if a high aspect ratio (ratio of length to diameter/width) and additional congestion is provided. The relevance of such geometries in industry is evident: tubes, tunnels, rooms highly congested by installations and chains of connected rooms are omnipresent. Also connecting tubes between larger volumes can pose an increased DDT hazard. A detonation formed in such a connecting tube can emerge into a larger volume and, under appropriate conditions, be sustained.

The present work investigates such a worst-case configuration of an entirely closed high-aspect-ratio channel with and without repeated obstacles.

1.2 Mixture Inhomogeneity in H₂–Air Explosions

Extensive knowledge is available on explosions in homogeneous gas mixtures. Mixtures of H_2 and air have been investigated particularly in the context of nuclear reactor safety [12]. However, a major current knowledge gap concerns the influence of mixture inhomogeneity as emphasized in a comprehensive OECD report on FA and DDT in nuclear reactor safety [12] and recently underscored by Bleyer et al. [6] and Kotchourko [82]. Spatial concentration gradients are omnipresent in real-world accident scenarios since H_2 is usually released from a finite region and stratifies in air due to its low density. Mixture homogenization through convective mixing may occur in industrial environments, but should not be presupposed in a worst-case based approach. Molecular diffusion also acts homogenizing, but is comparably slow and thus of minor relevance in large volumes.

In reality, concentration gradients in clouds of H₂ and air must be expected to be three-dimensional. Explosion in such mixtures is highly complex, which complicates the generation of fundamental physical understanding and isolation of dominating physical effects. The strategy in the present work is therefore to reduce complexity by investigating one-dimensional concentration gradients. Gradients are oriented perpendicular to the main direction of explosion front propagation, thus termed "transverse concentration gradients". Comparison to other studies addressing gradients parallel to explosion propagation will be made.

1.3 Goals and Structure of this Work

This work aims at generating a physical understanding of FA, onset of detonation and detonation propagation in H_2 -air mixtures with transverse concentration gradients in entirely closed channels. Since extensive knowledge on these processes has been built up over the decades for homogeneous mixtures, the strategy is to identify similarities and differences induced by concentration gradients. The focus is clearly placed on the influence of mixture, in contrast to the often investigated influence of confining geometry. Broad application of advanced optical measurement techniques at high temporal resolution in conjunction with conventional techniques is one of the major features of the present work.

Experiments have been conducted in a laboratory scale explosion channel. Local flame speed and overpressure were measured by conventional measurement techniques. Highly time-resolved optical measurement techniques provide images of explosion fronts at different stages of the explosion process. An approximate number of 3500 conventional and 1500 optical experiments has been performed to cover a large range of parameters at a high statistical reliability.

Theoretical approaches have been developed to describe the major experimental observations. The high complexity of explosion processes tends to limit the applicability of simplified models to the prediction of general trends. These models can however make an important contribution to understanding experimental results.

This work starts by providing the theoretical background of H_2 -air explosions in closed tubes in Ch. 2. By utilizing own experimental results already in this chapter, the reader is acquainted with typical measurement results obtained in the experiment. The chapter ends with a summary of available knowledge on explosion in mixtures with concentration gradients.

Chapter 3 presents the experimental setup, including the method of transverse concentration gradient generation and the investigated geometrical configurations.

In Ch. 4, applied measurement techniques are introduced. Both conventional and optical techniques are discussed. Besides providing theoretical background, hands-on information is given on the practical application of these techniques for explosion diagnostics. Results and their discussion are split into two chapters: DDT is analysed in Ch. 5. This includes the FA process and the onset of detonation. This chapter may be considered the most safety-relevant part of the present work. Detonation propagation is discussed separately in Ch. 6, rounding the picture of possible explosion regimes.

This work ends with a summary and outlook in Ch. 7.

2 Physics and Chemical Kinetics of H₂–Air Explosions in Tubes

This chapter gives an introduction to phenomena involved in confined H_2 -air explosions. Note that the scientifically unprecise term "explosion" is used in the first paragraphs to avoid denomination of all possible explosion regimes and will be refined progressively. The author focuses on entirely closed tube and channel geometries with a large aspect ratio (length to diameter/ width), filled with premixed H_2 -air mixture at initially ambient pressure and temperature. For broad background information on fluid mechanics, combustion and gasdynamics, the reader is referred to standard literature like [48], [91] and [98].

This chapter is particularly conceived to provide the scientific base for the analysis of experimental results presented in Ch. 5 and 6. Transformation of the herein discussed physics towards other types of fuels, geometries or initial conditions requires careful validation. Observations from experiments conducted within the scope of the present work are already included in this chapter, in particular images obtained by means of optical measurement techniques. The height of images presented here equals the explosion channel height of 0.06 m.

Section 2.1 provides a simplified overview of H_2 -air explosion processes and introduces common terminology. Fundamental relations of reactive compressible flow are presented in Sec. 2.2. Remarks on the chemical kinetics of the H_2 - O_2 system are given in Sec. 2.3. A discussion of ignition mechanisms and their relevance for explosion accidents follows in Sec. 2.4. Section 2.5 deals with FA and Sec. 2.6 introduces mechanisms of onset of detonation. Detonation properties are described in Sec. 2.7. The present state of knowledge on the role of mixture inhomogeneity is finally reviewed in Sec. 2.8.

2.1 Overview of H₂–Air Explosions

Explosions in closed tubes can be diverse in nature, depending on boundary and initial conditions. Phenomena observable in H_2 -air mixtures at initially ambient temperature and pressure range from slow deflagrations with maximum overpressures of around 1 bar and maximum flame speeds of the order of 100 m/s to detonations with maximum overpressures distinctly higher than 10 bar and supersonic propagation velocities of up to 2000 m/s.

Direct initiation of detonation in realistic H_2 -air explosion scenarios is highly improbable since the required ignition energy would be very high. This would only be achievable by very energetic ignition sources like high explosive charges. Thus, most explosions start with mild ignition by a weak ignition source like a spark.

Directly after ignition, a laminar flame front propagates into the mixture. The term deflagration is used for this type of flame propagation through diffusion of heat and species. Temporal evolution of the flame surface area plays an important role since enlargement results in an increased overall reaction rate, which is the integral of local burning velocity over the flame surface area. Flame surface area enlargement thus accelerates the flame. The laminar deflagration regime with a smooth, undistorted flame front is typically of short and thus negligible duration compared to the entire explosion process. Flame front instability arises quickly, distorting the flame surface and thereby further increasing the flame surface area. The result are so-called cellular flames. This type of instability is characteristic for H_2 -air mixtures, especially under lean conditions.

In a closed tube with end wall ignition, the flame acts like a piston due to thermal expansion of the reaction products, pushing fresh mixture ahead of the flame into the direction of flame propagation. Given that sufficiently high flow velocities and thus high Reynolds numbers are reached ahead of the flame, regions of enhanced turbulence appear. Such regions are typically wall boundary layers and the wake of obstructions. Interaction of flame and turbulence locally increases the burning velocity up to a maximum value, which is of the order of 10 times the laminar burning velocity. This leads to an increase in overall reaction rate in the tube and thus reinforces flow generation in the fresh mixture ahead of the flame.

During acceleration, flames continuously generate acoustic waves which propagate into the fresh mixture. These waves can coalesce and form shocks, which precompress and thus heat the fresh mixture. Interactions of shocks and the flame further increase the reaction rate.

This feedback cycle of flow, turbulence and shock generation accompanied by reaction rate enhancement, illustrated in Fig. 2.1, can allow deflagrations in H_2 -air mixtures to accelerate to the regime of fast deflagrations. Such an acceleration process is termed "strong", while "weak" acceleration only results in velocities of the order of 100 m/s. Fast deflagrations typically appear as a series of precursor shocks and the succeeding turbulent flame brush. Acceleration up to velocities of the order of 1000 m/s in reference to an external observer, accompanied by overpressures up to about 10 bar, is possible. However, flame front velocity relative to the gas ahead of the front is still subsonic.

At a critical flame velocity, often approximated by the speed of sound of the reaction products, onset of detonation may occur. This mostly involves a local explosion that causes high local overpressure and a sudden jump in explosion front velocity. Various mechanisms are known that can initiate onset of detonation. A sequence of criteria needs to be satisfied to allow for DDT. The wide range of effects depending on the respective mixture and geometry explains why it has not yet been accomplished to develop a comprehensive model for DDT. One could even disbelieve that such a generalized model would cope with the complexity of DDT processes at all.

After onset of detonation, a detonation wave propagates into the fresh mixture. Detonations are supersonic three-dimensional complexes of shocks and reaction zones, where the shocks lead to rapid auto-igntion by compressing and heating the mixture. Flame propagation due to heat and species diffusion is of minor importance. In contrast to deflagrations, detonations in H_2 – air propagate at velocities of up to 2000 m/s.



Figure 2.1: Overview of the DDT process.
2.2 Reactive Compressible Flow

This section provides equations of reactive compressible flow, referring to Anderson [1] and Lee [93]. Figure 2.2 shows the denotation of states used throughout the present work.



Figure 2.2: Definition of states in a shock-reaction zone complex propagating to the right.

State 0 denotes the initial gas state, defined as $p_0 = 1$ atm and $T_0 = 293$ K in the present work, resembling laboratory conditions. A shock travels at Mach number M_0 into the gas, yielding post-shock state 1. When shock reflection off a solid wall is considered, the post-reflected-shock state is denoted 1_r . Addition of specific heat q through chemical reaction leads to state 2. Specific heat q is calculated as the difference between standard enthalpies of formation of reactants and products according to Hess' Law for a multicomponent mixture:

$$q = \sum_{i} X_{i} h_{fi}^{\circ} - \sum_{j} X_{j} h_{fj}^{\circ}.$$
 (2.1)

 X_i and X_j denote molar concentrations of reactant and product species, respectively, and h_f° are the standard enthalpies of formation.

We employ the equations of conservation for mass, momentum and energy:

mass:
$$\rho_0 u_0 = \rho_1 u_1$$
, (2.2)

momentum:
$$p_0 + \rho_0 u_0^2 = p_1 + \rho_1 u_1^2$$
, (2.3)

energy:
$$h_0 + \frac{u_0^2}{2} + q = h_1 + \frac{u_1^2}{2}$$
. (2.4)

The normal shock relations can be derived, assuming calorically perfect gas:

$$\frac{\rho_1}{\rho_0} = \frac{(\gamma+1)M_0^2}{(\gamma-1)M_0^2+2},$$
(2.5)

$$\frac{p_1}{p_0} = 1 + \frac{2\gamma}{\gamma + 1} \left(M_0^2 - 1 \right).$$
(2.6)

Post-shock temperature can be calculated using the equation of state for perfect gas,

$$pM = \rho RT. \tag{2.7}$$

The adiabatic Hugoniot equation,

$$\mathbf{h}_1 - \mathbf{h}_0 = \frac{\mathbf{p}_0 - \mathbf{p}_1}{2} \left(\frac{1}{\rho_0} + \frac{1}{\rho_1} \right), \tag{2.8}$$

describes the change in thermodynamic state across a normal shock. Only thermodynamic properties are considered in contrast to the normal shock relations, Eq. (2.5) and (2.6), which relate post-shock conditions to the shock Mach number M_0 .

The Hugoniot equation can be readily extended for the case of a shock with specific heat addition q, which will allow for describing state 2 behind a complex of shock and reaction zone.

$$h_2 - h_0 + q = \frac{p_0 - p_2}{2} \left(\frac{1}{\rho_0} + \frac{1}{\rho_2} \right)$$
(2.9)

The combination of Eqs. (2.2) and (2.3), conservation of mass and momentum, yields the Rayleigh relation,

$$\frac{\mathbf{p}_1}{\mathbf{p}_0} = \left(1 + \gamma \mathbf{M}_0^2\right) - \left(\gamma \mathbf{M}_0^2\right) \frac{\rho_0}{\rho_1},\tag{2.10}$$

constituting the Rayleigh line in a p-v plane ($v = 1/\rho$).

When a normal shock is reflected off a solid wall, the reflected shock Mach number M_r is a unique function of the incident shock Mach number M_0 for a

given heat capacity ratio γ since flow velocity $u_{1,r}$ must vanish at the reflecting wall. Thus,

$$\frac{M_{\rm r}}{M_{\rm r}^2 - 1} = \frac{M_0}{M_0^2 - 1} \sqrt{1 + \frac{2(\gamma - 1)}{(\gamma + 1)^2} (M_0^2 - 1) \left(\gamma + \frac{1}{M_0^2}\right)}.$$
 (2.11)

Post-reflected-shock pressure and temperature can be calculated using the reflected shock Mach number M_r and the normal shock relations. This yields

$$p_{1,r} = p_1 \left[1 + \frac{2\gamma}{\gamma + 1} (M_r^2 - 1) \right]$$
(2.12)

and

$$T_{1,r} = T_1 \left[1 + \frac{2\gamma}{\gamma+1} (M_r^2 - 1) \right] \left[1 - \frac{2}{\gamma+1} \left(1 - \frac{1}{M_r^2} \right) \right].$$
 (2.13)

The Shock and Detonation Toolbox [133] is employed to solve relations of compressible reactive flow and compute zero- and one-dimensional explosion problems in the present work. It is coupled with Cantera [55], which is a software tool for simulating chemical kinetics, thermodynamics and transport processes. Thermodynamic data is taken from the Chemkin Database [73]. For simulations of chemical kinetics of H_2 -air, the reaction mechanism of Ó Conaire et al. [109] is applied, which is a well-accepted mechanism for H_2 -air. It was successfully used by Ettner [42] for CFD simulations of DDT and recently employed by Hasslberger et al. [61] for large scale explosion simulations. Its most important feature for the present work is the broad validation range: Validated pressure ranges from 0.05 to 87 atm, temperature from 298 to 2700 K and equivalence ratio from 0.2 to 6.

2.3 Chemical Kinetics of the H₂–O₂ System

Chemical reaction of H_2 and O_2 proceeds through a network of elementary reactions. For general explanations on combustion chemistry, please refer to [91, 147]. Elementary reactions that influence the overall reaction rate most significantly are listed subsequently [91, 92, 157]. For each elementary reaction, Arrhenius chemistry is assumed, so that reaction rate coefficients k are given by the modified Arrhenius equation

$$k = AT^{n}e^{-\frac{La}{RT}}$$
(2.14)

with A being the pre-exponential constant, T the thermodynamic temperature, n a constant describing temperature dependence, E_a the activation energy and R the gas constant.

Chain initiation:

$$H_2 + O_2 \stackrel{k_{2.15}}{\longrightarrow} HO_2 + H \tag{2.15}$$

Chain propagation:

$$H + O_2 \stackrel{k_{2.16}}{\longrightarrow} O + OH$$
 (2.16)

$$O + H_2 \stackrel{k_{2.17}}{\longrightarrow} H + OH$$
 (2.17)

$$OH + H_2 \stackrel{k_{2.18}}{\longrightarrow} H + H_2 O \tag{2.18}$$

Chain termination:

$$H + O_2 + M \stackrel{k_{2.19}}{\longrightarrow} HO_2 + M$$
(2.19)

H₂O₂ formation and decomposition:

$$HO_2 + HO_2 \xrightarrow{k_{2.20}} H_2O_2 + O_2$$
(2.20)

$$HO_2 + H_2 \xrightarrow{k_{2,21}} H_2O_2 + H$$
 (2.21)

$$H_2O_2 + M \stackrel{k_{2.22}}{\longrightarrow} OH + OH + M$$
 (2.22)

Reaction (2.15) represents the relatively slow chain initiation, forming H atoms which act as chain carriers for the subsequent chain-branching reaction. Dissociation of H_2 may portray another initiation reaction, but becomes relevant only at very high temperatures [92]. Once the concentration of chain carriers is sufficient, chain propagation takes over, which is of branching character in atmospheric H_2 – O_2 flames. Reactions (2.16) and (2.17) are chain-branching reactions since they increase the number of chain carriers H, O and OH. Repetition of chain-branching reactions leads to an exponential growth in reaction rate, termed branched-chain explosion.



Figure 2.3: P-T-explosion diagram of H₂-O₂, adapted from [23, 92].

The major heat is released through chain termination and recombination reactions [160]. Chain termination can follow multiple paths. Variants will be discussed here along with the introduction of the p-T-explosion diagram for H_2 – O_2 , shown in Fig. 2.3.

To obtain a p-T-explosion diagram, H_2-O_2 mixture is introduced into a closed hot vessel. Below about 650 K, chain branching does not occur since reaction (2.16) is endothermic and does not contribute at low temperature [91]. Thus, the character of the reaction is slow, termed non-explosive. Beyond 650 K, for instance at 740 K, variation of pressure leads to the observation of either explosive or non-explosive reaction. Limits between these regimes are depicted in the p-T-explosion diagram as pressure-temperature boundaries and can be interpreted as limits for auto-ignition¹. The time between exposure of the mixture to the specific pressure and temperature and ignition is called induction time τ_{ind} . Within the present work, ignition is defined as the moment of maximum heat release rate, coinciding with the point of maximum temperature gradient. Note the difference to approaches utilizing emission of radiation from excited molecules as a marker².

Below the first explosion limit, termination of chain reaction occurs due to diffusion of active species H, O and OH to vessel walls. Destruction of these species exceeds their formation by chain-branching reactions and the overall reaction rate is negligible. Raising pressure, wall termination is overcome since the higher gas density hinders diffusion of active species to walls. Thus, explosive reaction is observed. With a further increase in pressure, three-body reactions become more frequent. Reaction (2.19) can effectively terminate the chain reaction. Mainly the competition between chain-branching reaction (2.16) and chain termination through reaction (2.19) controls the second explosion limit. A simplified second limit criterion can be written as a balance of chain carriers:

$$2k_{2.16} = k_{2.19}[M], \qquad (2.23)$$

where k_i are the respective rate constants and [M] is the concentration of third body. An effective third body collision efficiency is employed here, taking into account all possible third body species. This simplified limit is shown in Fig. 2.3 as a black dashed line. Deviation of this simplified limit from the experimentally observed second explosion limit is due to the influence of H_2O_2 formation and decomposition reactions (2.20)–(2.22) [23].

Beyond the third explosion limit, thermal explosion with an exothermal induction period and a dominating straight-chain mechanism occurs. This

¹Auto-ignition is defined as ignition due to homogeneous heating of a mixture in abscence of an external ignition source.

²This is a common approach in shock-tube experiments. However, the temporal coherence of heat release and light emission requires careful analysis, as pointed out by Mével et al. for the excited hydroxyl (OH) radical [106].

stands in contrast to the branched chain explosion below the second explosion limit as discussed beforehand, where the induction period is nearly isothermal [23]. Reactions (2.20)–(2.22) gain in importance beyond the third limit. Recombination of HO₂ in reaction (2.20) and abstraction of H through reaction (2.21) form H_2O_2 , which is decomposed by reaction (2.22) into OH [92].

The three explosion limits discussed so far represent boundaries between regions in the T-p-plane where auto-ignition in a mixture occurs, from regions where it does not occur. In addition to these three limits, an extension of the second explosion limit can be observed in experiments [92]. This limit is indicated in Fig. 2.3 as a red dashed line. It extends towards high values of both pressure and temperature. This region of high pressure and temperature is highly relevant for the present work. When a shock at high Mach number, distinctly higher than M = 2, propagates into fresh mixture of H_2 and air at initially ambient conditions, post-shock temperature and pressure, Eq. (2.6) and (2.7), reach values that approach or cross this extended second explosion limit. The character of the extended second explosion limit differs from the three classical limits discussed beforehand: On both sides of the limit, autoignition occurs. However, reaction is distinctly faster on the right side of the limit (at higher T, dominating branched-chain mechanism [92]) than on the left side (dominating straight-chain mechanism [92]). This difference in ignition behavior and implications for the present work will be discussed in more detail in the next section.

2.4 Ignition Mechanisms

Two modes of ignition can be discriminated according to the ignition energy involved: mild ignition with a moderate ignition energy, leading to a deflagration and strong ignition with a high ignition energy, directly initiating a detonation. These two mechanisms are discussed in this section.

Through mild ignition, a flame is initiated locally, propagating from the point of ignition into the fresh mixture. The combustion mode is deflagration, thus

diffusion of heat and species dominates flame propagation. The minimum ignition energy (MIE) of H_2 -air mixtures at standard pressure and temperature depends on H_2 concentration and reaches a minimum of 0.017 mJ close to stoichiometry [84]. Minimum ignition energies of other combustible gases in air are typically in the range of 0.2-0.3 mJ [96]. It is obvious that H_2 can be ignited comparably easily.

Ignition can be investigated experimentally in shock tubes. A shock of defined strength, expressed in terms of Mach number, is generated and propagates into a measurement section filled with the test gas. At the closed end of the tube, the shock is reflected. At sufficient incident shock Mach number, auto-ignition behind the reflected shock occurs due to shock-induced compression and related heating of the mixture. Rather weak shocks cause **mild ignition** which is characterized by the occurence of randomly distributed flame kernels behind the reflected shock [110]. Mild ignition in shock tubes is crucially dependent on the homogeneity of mixture composition and temperature field since ignition occurs first at the most favorable points. Also formation of hot spots, for example by shock focussing or further non-ideal effects, influence results in shock tube experiments with mild ignition (cp. [115, 118]). Flames propagate as deflagrations in the post-reflected-shock mixture. The separation distance between reflected shock and flame front thus grows continuously due to the low flame speed.

Strong ignition occurs if the incident shock is strong enough to cause rapid auto-ignition after reflection, directly leading to explosion at the reflecting wall. A blast wave is produced that can overtake the reflected shock and form a detonation. This process of shock reflection, causing a local explosion that generates a detonation front, is typically observed during onset of detonation in obstructed geometries. Details are given in Sec. 2.6. The range of T and p where strong ignition is possible, can be roughly approximated by calculating post-shock conditions of Chapman-Jouguet detonations ³. Temperatures within assumed detonability limits of 12 and 70 vol. % H₂ range from 1050 K to 1550 K, whereas pressures range from 17 to 29 bar⁴. The extended second ex-

³Properties of detonations are discussed in Sec. 2.7.

⁴Calculated using the Shock and Detonation Toolbox [133], Cantera [55] and thermodynamic data from the Chemkin database [73].

plosion limit crosses this region. As argued by Lee and Hochgreb [92] based on experiments of several authors, this limit separates mild ignition after shock reflection on its left side from strong ignition on its right side. This finding will be employed in Sec. 5.2.3 to compute critical incident shock properties that are required to cause strong ignition and thus onset of detonation after shock reflection in H_2 -air mixture.

Belles [4] suggested to employ the extended second explosion limit as a limit for detonation initiation and propagation. He used the simplified description given by Eq. (2.23) to determine the extended second explosion limit. Ng et al. [108] recently followed this approach to study detonation hazards in H₂ at high pressure. However, the simplified extended second explosion limit criterion does not account for reactions of H_2O_2 formation and decomposition (reactions (2.20)–(2.22)), which play an important role at this limit. This has been concluded by several authors as summarized by Lee and Hochgreb [92], and also noted by Dove and Tribbeck [33] and Browne et al. [15] for instance. Shepherd [132] likewise shows that reaction around this limit is a coupled chain-branching and thermal explosion. The simplified criterion leads to an overprediction of temperature necessary to reach strong ignition at a given pressure, cp. Fig. 2.3. In the present work, the extended second explosion limit will be determined employing detailed chemical kinetics.

In detonation experiments, strong ignition and thereby direct initiation of detonation can be achieved for example by means of a high explosive charge. An energy per surface area of about 0.7 MJ/m² is required to cause direct initiation of a planar detonation in stoichiometric H_2 -air. As a comparison, propane demands 3.1 MJ/m² and methane 10 MJ/m² [8].

In addition to mild and strong ignition, a transient region can be observed in shock tube experiments. In the transient ignition regime, mild ignition initially produces flame kernels, but a subsequent DDT process⁵, often involving a violent secondary explosion, leads to detonation as observed optically by Wang et al. [156]. Since the flame kernels form at random positions behind the reflected shock, secondary explosions also emerge from random locations in the transient regime. Distinction between transient ignition involving DDT

⁵The DDT pocess is described in detail in Sec. 2.6.

and strong ignition is often not clearly stated in literature. One may describe the boundary between transient and strong ignition as the point when autoignition is firstly observed directly at the reflecting wall and not at random positions between wall and reflected shock as in the transient regime.

Evaluated from a safety perspective, two conclusions have to be drawn from the preceding discussion:

- Due to the low ignition energy required to ignite H₂-air mixture, ignition in H₂-air is highly probable in industrial environments as soon as flammable mixture is present. Potential ignition sources are omnipresent such as sparks of different origin (mechanical, electrostatic discharge, etc.), hot surfaces, mechanical friction or auto-ignition in hot environments [38].
- Strong ignition and thereby direct initiation of detonation is improbable in real-world accident scenarios. As it has been shown, the energy required for direct detonation initiation could in principle be provided by sources like high explosives, which are however seldomly involved in industrial explosions. Thus, a DDT process is typically required to reach detonation.

2.5 Flame Acceleration

A velocity plot of a flame accelerating in the experimental setup used in the present work, an entirely closed rectangular channel, is given in Fig. 2.4. The FA process can be divided into three characteristic phases. Phase (1) starts with laminar and cellular flame propagation, introduced in Secs. 2.5.1 and 2.5.2, respectively. The slow turbulent deflagration regime follows, elucidated in Sec. 2.5.3. Exponential FA is mostly observed in phase (1) [150].



Figure 2.4: Exemplary experimental flame speed plot with characteristic phases of FA. Measured speed (●) and illustrative fit (red line).

As flame speed exceeds a value around the sound speed of the reactants a_{re} , the fast deflagration regime (2) is reached, explored in Sec. 2.5.4. The importance of flow compressibility increases. Constant acceleration is often observed in this second phase [150]. Slow and fast turbulent deflagration can be distinguished as follows: In the slow regime, flame propagation is dominantly controlled by subsonic fluidmechanic processes. In contrast, the fast regime is characterized by the presence of gasdynamic discontinuities such as shocks, precompressing the fresh mixture and interacting with the flame.

Phase (3) shows velocity saturation close to the speed of sound of the reaction products a_{pr} . It is often termed the choked regime. At flame speeds in this range, onset of detonation is often observed in experiments.

2.5.1 Laminar Deflagration

Shortly after ignition, a laminar deflagration propagates from the point of ignition into the fresh mixture. Initial laminar flame propagation after end wall point ignition in a channel is illustrated in two dimensions in Fig. 2.5. Curvature and gravitational effects are neglected in the following generalized discussion.



Figure 2.5: Laminar initial flame propagation in 2D. Detail of the laminar flame front (right).

The laminar flame of thickness l_L is composed of preheat and heat release zone. The heat release zone thickness l_δ is

$$l_{\delta} = \frac{l_{\rm L}}{\beta},\tag{2.24}$$

where the Zeldovich number β [167] is defined as

$$\beta = \frac{\mathrm{E}_{\mathrm{a}}(\mathrm{T}_{\mathrm{pr}} - \mathrm{T}_{\mathrm{re}})}{\mathrm{R}\mathrm{T}_{\mathrm{pr}}^{2}}.$$
(2.25)

Since β is of the order of 10 for hydrocarbons and H₂ [54], the heat release zone thickness l_{δ} is often approximated as $l_L/10$.

Flame propagation velocity with respect to the mixture ahead of the flame equals the laminar burning velocity S_L , cp. Fig. 2.5, right. The velocity of prod-

ucts behind the flame equals $S_L \sigma$,

$$\sigma = \frac{\rho_{\rm re}}{\rho_{\rm pr}},\tag{2.26}$$

where ρ_{re} and ρ_{pr} are the densities of reactants and products, respectively, and σ is termed expansion ratio. Since the rear wall boundary condition at x = 0 m, namely stagnant flow, needs to be satisfied, the flame propagates with a velocity $S_L \sigma$, termed flame speed, with respect to an external observer. Flow is induced ahead of the flame at a velocity u. Thus,

$$S_{\rm L}\sigma = u + S_{\rm L}.\tag{2.27}$$

Flame surface areas A_F different from the channel cross-section A_C can be considered:

$$S_{\rm L}\sigma A_{\rm F} = uA_{\rm C} + S_{\rm L}A_{\rm F}.$$
 (2.28)

 A_F would be the flame front length and A_C the channel height H in the present two-dimensional example. It is obvious that an enlargement of A_F also results in an increase of visible flame speed, given by the left side of Eq. (2.28).

Within the present work, the unstretched laminar burning velocity at standard temperature and pressure S_L in [m/s] as a function of X_{H2} in [vol. %] is approximated from experiments, summarized by Konnov [81], as a 6th order polynomial:

$$\begin{split} S_{L} &= -1.55236 \cdot 10^{-9} \cdot X_{H2}^{6} + 3.49519 \cdot 10^{-7} \cdot X_{H2}^{5} - 2.82975 \cdot 10^{-5} \cdot X_{H2}^{4} \\ &\quad + 9.35840 \cdot 10^{-4} \cdot X_{H2}^{3} - 9.97510 \cdot 10^{-3} \cdot X_{H2}^{2} \\ &\quad + 5.00120 \cdot 10^{-2} \cdot X_{H2} - 8.32830 \cdot 10^{-2}. \end{split} \tag{2.29}$$

Comparison of this approximation with experimental data [81] is provided in Fig. 2.6.

Limits for flame propagation exist in H_2 -air mixtures, termed flammability limits. The direction of flame propagation has an effect on the flammability limits due to convection currents generated by the flame [24]. Table 2.1 provides numbers for flammability limits for upward, horizontal and downward flame propagation [24].



Figure 2.6: Unstretched laminar burning velocity S_L of H_2 -air at standard temperature and pressure [81]. Data sources: \bullet [34], \Box [162], \star [146], \triangle [72], + [149]. Red line: approximation by Eq. (2.29).

Table 2.1: Flammability	limits of H ₂ -air at standard temperature and	d pressure
[24].		

Propagation direction	Lower limit [vol. % H ₂]	Upper limit [vol. % H ₂]
Upward	4.1	74
Horizontal	6.0	n/a
Downward	9.0	74

2.5.2 Cellular Deflagration



Figure 2.7: OH-PLIF images of cellular flames in homogeneous H₂–air mixtures.

This section first shows the phenomenology of cellular flames, which is then explained based on two instability mechanisms. It can be experimentally observed that H_2 -air flames become unstable shortly after ignition although flow is still laminar. Figure 2.7 shows OH-PLIF images⁶ of flames in mixtures of 15–40 vol. % H_2 in air, obtained using the experimental setup described in Ch. 3. The image height equals the channel height of 0.06 m. Development of the cellular structure is a dynamic process, which has been widely described in literature, see for instance work of Hertzberg [62], Clavin [22] or Law [90]. After

⁶For a description of the OH-PLIF measurement technique, please refer to Sec. 4.2.2.



Figure 2.8: Illustration of hydromechanic (Landau-Darrieus) instability.

ignition, an initial decrease in cellular lengthscale, often termed wavelength, is typical. It is followed by a state of quasi-stationary topology, where cells grow and refine dynamically as described by Bradley et al. [10, 11]. This distortion of the flame front is known to enhance the overall reaction rate and thus support FA [20].

In lean mixtures (15 and 20 vol. %) separated flame islands with quenching in intermediate cracks are observed in Fig. 2.7. At 25 vol. %, no local quenching is observed anymore. With rising H_2 concentration the wavelength of the cellularity increases and flame fronts becomed more stable.⁷

An instability mechanism leading to cellular flame development is hydrodynamic instability, also termed Landau-Darrieus instability [26, 88, 114]. It is illustrated in Fig. 2.8. If a flame is locally perturbed (red solid line), left part of Fig. 2.8, forming convex and adjacent concave sections, flow behind the flame is deflected due to expansion across the flame as depicted in the right part of Fig. 2.8. Behind the convex section streamlines converge, whereas they diverge behind the concave section. This accelerates and decelerates the flame locally in the convex and concave sections, respectively, and thus amplifies flame wrinkling (red dashed line).

Additionally, diffusive instability [96] needs to be taken into account. It interacts with the hydrodynamic instability, either supporting or damping flame wrinkling. If the diffusivities of the limiting component of a mixture

 $^{^{7}}$ In addition, these images show the influence of buoyancy at low H₂ concentration. Lean flames are oriented towards the top of the channel, while stoichiometric and rich mixtures cause rather symmetric flames with respect to the channel centerline.



Figure 2.9: Illustration of preferential diffusion of limiting component.

(e.g. H_2 in lean H_2 -air) and the excess component differ, diffusive fluxes of these components across a wrinkled flame front follow different paths as illustrated in Fig. 2.9⁸. Preferential diffusion of the limiting species leads to local increase in limiting species concentration in convex flame sections. Vice versa, limiting species concentration in concave sections drops. This can even lead to local extinction of the flame as observed in the notches between reacting islands in the 15 and 20 vol. % mixtures in Fig. 2.7.

Diffusion of heat interacts with the described preferential species diffusion, which explains the widely used term diffusive-thermal instability. As argued beforehand, the concentration of limiting component can be increased locally in convex flame sections. Whether this in turn leads to higher reaction rates and high temperatures at these locations additionally depends on thermal diffusion. In case of a low thermal diffusivity, enhanced limiting species concentration combined with weak heat flux from this region to the ambient gas causes a region of high temperature, illustrated in Fig. 2.10, left side. Thereby, burning velocity is locally increased in convex sections and consequently reduced in concave sections. Flame wrinkling is enforced in this case. Otherwise, a high heat flux in case of high thermal diffusivity balances the burning velocity distribution between convex and concave flame sections along the flame front. Flame wrinkling is thus reduced, see Fig. 2.10, right side. The ratio

⁸This can be observed not only for reactants, but also for intermediate species and products, resulting in a complex pattern of diffusive fluxes.



Figure 2.10: Illustration of diffusive-thermal instability, adapted from [20].

of thermal diffusivity a and diffusion coefficient D of the limiting species in the mixture forms the Lewis number Le⁹:

$$Le = \frac{a}{D}.$$
 (2.30)

Lewis numbers smaller than about unity enhance flame wrinkling whereas Lewis numbers larger than unity damp it. Experimental values for the Lewis number in H_2 -air are given in Fig. 2.11. It can be seen that transition from stabilizing to destabilizing occurs close to stoichiometry.

Since most other flammable gas mixtures have a Lewis number close to or larger than unity, H_2 takes a special position with the highest propensity for cellular flame development due to the high diffusivity of H_2 .

Cellular flame propagation has been investigated by Markstein [102]. He defines the Markstein length L_M , describing the effect of flame stretch rate on local burning velocity. The unstretched burning velocity is further on termed S_L , whereas the local burning velocity of the stretched flame is $S_{L,S}$. Employing the Karlovitz stretch factor K [71],

$$K = \frac{1}{A_F} \frac{dA_F}{dt},$$
 (2.31)

describing the normalized rate of flame surface area change, yields the relation of flame stretch rate and burning velocity of the stretched flame depend-

⁹The following discussion neglects multicomponent diffusion, which would lead to the formulation of a separate Lewis number for each species. The Lewis number introduced here is an effective Lewis number for the entire mixture.



Figure 2.11: Experimentally determined Lewis number Le of H_2 -air mixtures as a function of equivalence ratio Φ [139].

ing on the Markstein length L_M [22]:

$$S_{L} - S_{L,S} = L_{M}K.$$
 (2.32)

This relation is often rewritten using the dimensionless Markstein number

$$Ma = \frac{L_M}{l_L},$$
 (2.33)

 l_L being the laminar flame thickness.

The Markstein number can be defined separately for the effect of flame curvature and strain [102]. Convex and concave sections of a flame front experience positive and negative stretch rates, respectively. Figure 2.12 gives values for Markstein length in H_2 -air determined experimentally through flame front velocity measurements of spherically expanding flames. For H_2 -air mixtures, L_M is negative below stoichiometry and positive beyond. For negative Markstein lengths, flame instability is amplified since positive (negative) stretch rates enhance (reduce) the local flame velocity as described by Eq. (2.32). Positive Markstein lengths damp instabilities. This behavior is well discernible in Fig. 2.7.



Figure 2.12: Experimentally determined Markstein length as a function of equivalence ratio Φ. Data sources: ● [14], □ [29].

2.5.3 Slow Turbulent Deflagration

Flow induced ahead of a flame in a closed tube interacts with the tube walls and with obstructions, if present. At sufficiently high induced flow velocity, or related Reynolds number, regions of turbulent flow form. The flame propagating through the tube thus experiences spatially varying flow regimes which may transform it locally or globally from a laminar into a turbulent deflagration. This section first elucidates the phenomenology of turbulent flows and deflagrations and subsequently presents experimental observations in unobstructed and obstructed tubes. Fundamentals of turbulent flow and turbulent combustion regimes are reviewed only in brief. The reader is referred to standard work by Pope [123], Turns [147] or Law [91] for more extensive explanations.

Statistical temporal and spatial velocity fluctuation is inherent to turbulent flow. Reynolds describes local flow velocity as the superposition of time-averaged flow velocity u and velocity fluctuation u' [125]. Turbulence is induced in a flow by its interaction with confinement. The largest turbulent eddy size is of the order of the confining geometry dimensions. The mean size of

large eddies is termed the "integral length scale" of turbulence l_T . One definition of l_T , assuming isotropic turbulence, is given in Eq. (2.34). The integral time scale t_T can be defined accordingly.

$$l_{\rm T} = \frac{u'^3}{\epsilon}, \quad t_{\rm T} = \frac{l_{\rm T}}{u'}. \tag{2.34}$$

Turbulent kinetic energy is continuously transported from eddies with mean size l_T to smaller eddies at a viscous dissipation rate ϵ . Eddy sizes between the integral length scale l_T and the Kolmogorov micro scale of length l_η , going back to the analysis of turbulent flow by Kolmogorov [78, 79], exist in fully developed turbulent flows. The Kolmogorov micro scale of length l_η and the Kolmogorov micro scale of time t $_\eta$ are

$$\mathbf{l}_{\eta} = \left(\frac{\nu^3}{\epsilon}\right)^{1/4}, \quad \mathbf{t}_{\eta} = \left(\frac{\nu}{\epsilon}\right)^{1/2}, \tag{2.35}$$

based on kinematic viscosity v and ϵ . At an eddy size of l_{η} , turbulent kinetic energy undergoes viscous dissipation [80]. Since l_{η} decreases with an increase in dissipation rate ϵ , which in turn rises with flow velocity u ($\epsilon \propto u^3/l_T$) [140], high flow velocity in a given fluid yields a lower smallest eddy size than flow at lower velocity.

With the definitions given so far, dimensionless parameters can be formed to characterize turbulence-flame interaction. Two Karlovitz numbers, Ka and Ka $_{\delta}$, based on Karlovitz's investigation of turbulent deflagration [70], are defined as

$$Ka = \left(\frac{l_L}{l_\eta}\right)^2, \quad Ka_\delta = \left(\frac{l_\delta}{l_\eta}\right)^2.$$
(2.36)

Ka is the ratio of laminar flame thickness l_L , including preheat and heat release zone, and the Kolmogorov length scale l_{η} . Ka_{δ} is defined based on the thickness of the heat release zone l_{δ} .

The combustion regime diagram, introduced by Borghi [7] and modified by Peters [116, 117], is used subsequently to summarize turbulence-flame interaction regimes. Figure 2.13 shows the regime diagram along with OH-PLIF images of flames in distinct phases of FA.



Figure 2.13: Combustion regime diagram with representative OH-PLIF images. Interpretation of FA process (red arrow).

Cellular flames as discussed in Sec. 2.5.2 must be categorized as laminar flames in the diagram since flow is laminar in this regime and flame front distortion is caused by instability mechanisms only.

Wrinkled and corrugated flamelet regimes are characterized by dominance of eddies larger than the laminar flame thickness, $l_T > l_L$, interacting with the flame front and causing macroscopic enlargement of the flame surface area. Turbulent burning velocity S_T can be defined as

$$S_{\rm T} = S_{\rm L} \frac{A_{\rm F,T}}{A_{\rm F,L}}$$
(2.37)

with flame surface areas $A_{F,L}$ and $A_{F,T}$ of the laminar and turbulent flame front, respectively. The microscopic structure of the flame front is however assumed to remain similar to that of a laminar flame. Local burning velocity along the flame front still equals the laminar burning velocity S_L . In other terms, local



Figure 2.14: OH-PLIF sequence of a slow turbulent deflagration, unobstructed channel, 15 vol. %, homogeneous mixture. $\bar{v} = 35$ m/s.

transport of heat and species is not altered by large eddies.

At Ka > 1, transport of heat and species within the flame front is enhanced, the local burning velocity thus exceeds the laminar burning velocity. Eddies can penetrate the flame front and increase transport of species and heat inside the flame front, which is otherwise only due to diffusion.

 Ka_{δ} describes the potential of turbulent eddies to penetrate the heat release zone of a laminar flame. If $Ka_{\delta} > 1$, chemical reaction cannot be terminated during one eddy circulation. Reacting portions of gas are mixed with cold reactants. Thus, local flame quenching can occur at $Ka_{\delta} > 1$. This poses an upper boundary to the turbulent burning velocity S_T . An approximation of maximum turbulent burning velocity frequently used in safety analysis is 10 times the laminar burning velocity [20]. Even global flame quenching has been observed in experiments where flames passed an obstacle with a blockage ratio > 90 % [67]. This regime is however not relevant for the present work.

Turbulent regions in the fresh mixture ahead of the flame in an **unobstructed tube** are restricted to the wall boundary layers. The flame front experiences wrinkling by turbulent eddies in these regions, visible in Fig. 2.14. This sequence of OH-PLIF images was obtained experimentally in an unobstructed channel configuration. It can be seen that the flame front is compact in the

center of the channel, similar to the flame fronts shown in Fig. 2.7. In contrast, turbulence near the walls interacts with the flame and leads to a locally corrugated flame front. Dorofeev [30] uses this observation to calculate a mean burning velocity in an unobstructed tube as the average of laminar burning velocity S_L , weighed with the height of the laminar flow region h_L , and the turbulent burning velocity S_T , weighed with the height of the turbulent flow regions $h_{T,1}+h_{T,2}$.

In an **obstructed tube**, three major effects and their interaction is relevant for FA:

- Macroscopic enlargement of flame surface area in the vicinity of obstacles,
- induction of instabilities and
- turbulence generation in the wake of obstacles and interaction with the flame.

In shadowgraph and OH-PLIF sequences¹⁰ with H₂ concentrations between 12.5 and 20 vol. %, these effects can be observed. Both macroscopic flame surface area enlargement and flow instability can be seen in Fig. 2.15, where the flame propagates in a very lean homogeneous mixture of 12.5 vol. % H₂ at an average flame tip velocity of $\bar{v} = 40$ m/s. Shadowgraph images show a flame with strong cellular instability and turbulent regions in the vicinity of the channel walls. Turbulence in the obstacle wake is weak and almost invisible. While passing the obstacle, the flame contracts strongly and forms a finger-shape leading tip, which enlarges the flame surface area locally. OH-PLIF images give detailed insight into the flame front topology. The flame surface is wrinkled and remains connected, not showing fragmentation. Wrinkling can be attributed to a combination of hydrodynamic, diffusive-thermal, Rayleigh-Taylor and Kelvin-Helmholtz instability.

At 15 vol. % H_2 , Fig. 2.16, the average flame tip velocity in the same field of view (FOV) rises to about 120 m/s. Flow ahead of the flame shows turbulent

¹⁰Shadowgraph and OH-PLIF images were taken in separate experiments with equal experimental conditions.



Figure 2.15: Shadowgraph and OH-PLIF sequences of a slow turbulent deflagration, obstructed channel, 12.5 vol. %, homogeneous mixture. $\bar{v} = 40$ m/s. Red box represents OH-PLIF FOV.

fluctuations, well visible in the shadowgraph images. Wall boundary layer turbulence being present uptream of the obstacle is transported into the channel center through flow deflection by the obstacle. Additional turbulence is generated by flow separation at the upstream obstacle edge. The flame surface is wrinkled on clearly smaller scale than in Fig. 2.15. The OH-PLIF sequence shows that the flame surface is now fragmented, including separated flame islands. The formation and re-connection of such islands is a dynamic process. Obviously the two-dimensional OH-PLIF depiction of the flame front is influenced by three-dimensional motion of flame front elements, so that flame islands can be sectional images of flame fingers reaching into the imaging plane.

At 20 vol. %, Fig. 2.17, where average flame tip velocity equals 300 m/s, the flow pattern ahead of the flame clearly differs from the two preceding H₂ concentrations. The most obvious difference is the formation of a turbulent shear layer, originating at the upstream edge of the obstacle plates. Black regions at the beginning of the shear layer are signs of expansion fans, indicating transsonic flow. Weak gasdynamic structures can be seen in the obstacle opening. Thus, following the definition of slow and fast regimes given incipiently, this case marks transition from slow to fast deflagration. The flame front is intensely wrinkled at very small scales and the reaction zone seems to be spatially extended. This suggests that turbulence interacts with the flame front and Ka_{δ} < 1 < Ka.



Figure 2.16: Shadowgraph and OH-PLIF sequences of a slow turbulent deflagration, obstructed channel, 15 vol. %, homogeneous mixture. \bar{v} = 120 m/s. Red box represents OH-PLIF FOV.



Figure 2.17: Shadowgraph and OH-PLIF sequences of a turbulent deflagration, obstructed channel, 20 vol. %, homogeneous mixture. $\bar{v} = 300 \text{ m/s}$. Red box represents OH-PLIF FOV.

2.5.4 Fast Turbulent Deflagration

As defined in Sec. 2.5.3, the fast turbulent deflagration regime differs from the slow deflagration regime in the occurence of gasdynamic discontinuities and their interaction with the flame front. Consistent with the preceding sections, experimental results are used to introduce the physics of fast deflagration. The FOV is equal to the one used in the previous section. To obtain fast deflagrations, H₂ concentration is increased. At a flame speed of the order of 300-400 m/s, shocks start to form. Images shown in Fig. 2.17 mark the transition from slow to fast deflagration. This is further underscored by analyzing the same experiment at higher temporal resolution. Figure 2.18 comprises a shadowgraph sequence. In the first frame, t = 0 μ s, turbulent flow in the obstacle wake as discussed in Sec. 2.5.3 is visible. Weak compression waves can be seen upstream of the obstacle continuously generated in the flame zone and being reflected off the obstacle. The second frame shows the formation of shocks by coalescence of reflected pressure waves. They appear at the upper and lower channel wall upstream of the obstacle. Since each pressure wave travelling at the local sound speed raises pressure incrementally, the following pressure wave propagates in mixture of incrementally higher sound speed and catches up with the pressure wave running ahead. Subsequent frames in Fig. 2.18 display the strengthening of the shocks and eventually their intersection at the channel center line in the last frame. Since pressure wave reflection off an obstacle is involved here, formation of shocks in a channel occurs earlier if obstacles are present compared to an unobstructed channel. The resulting difference in shock pattern ahead of the turbulent flame brush will be discussed at the end of this section.

At an H_2 concentration of 25 vol. %, FA is more advanced in the FOV. Figure 2.19 shows a great number of shocks, increasing in strength as the flame approaches the obstacle. Reflected shocks interact with the flame. This shock-flame interaction and precompression of the fresh mixture by shocks is of crucial importance for the FA process within the fast deflagration regime. Recently Ciccarelli et al. [21] and Johansen [65] discussed the role of shock-flame interaction, stating that turbulence may not be sufficient for experimentally observed strong FA to values of the order of 1000 m/s. Shock-flame interaction



Figure 2.18: Shadowgraph sequence of shock formation process, obstructed channel, 20 vol. %, homogeneous mixture. $\bar{v} = 300$ m/s.



Figure 2.19: Shadowgraph sequence of shock-flame interaction, obstructed channel, 25 vol. %, homogeneous mixture. $\bar{v} = 440$ m/s.

is assumed to make an important contribution by distortion and wrinkling of the flame through Richtmyer-Meshkov (RM) instability. The RM instability arises when a shock interacts with an interface between two fluids, or as in case of flames, the interface between reactants and products [13, 105, 126]. Baroclinic vorticity generation due to non-parallel gradients in pressure and density leads to flame wrinkling on small scales and macroscopic flame distortion on large scales. Thomas et al. [141] experimentally demonstrated the great potential of shock-flame interaction to accelerate flames. Kholkhlov et al. [76] likewise conclude that shock-flame interaction is important to accelerate flames to critical conditions for onset of detonation. They state that large scale RM instability is the primary mechanism increasing the heat release rate during interaction of a flame with a single shock through macroscopic flame surface area growth. Small-scale instability decays quickly and thus only contributes for a short time. However, in an FA process, shock-flame interactions take place continuously as seen in Fig. 2.19. This suggests that a high level of small scale shock-induced turbulence can be maintained. Gamezo et al. [52] analyze the overall reaction rate development across a DDT process in H₂-air mixtures by numerical simulation. They find that flame surface area enlargement causes a 100-200-fold growth in overall reaction rate, whereas precompression of the mixture by shocks contributes another factor of 10–20.

At an H₂ concentration of 30 vol. %, Fig. 2.20, a group of shocks is observed passing the FOV long before flame arrival. These shocks diffract around the obstacle inducing flow and enhancing shear layer turbulence behind the obstacle plates. The typical vortex street due to Kelvin-Helmholtz instability is observed. 362.5 μ s after leading shock arrival, the flame is visible in the FOV. Similar to the 25 vol. % sequence, a large number of shocks precede the flame front and precompress the mixture. However, these shocks are not strong enough to cause auto-ignition by reflection at the obstacle yet.

In conclusion, continuously repeated flame interaction with a multiplicity of shocks, in conjunction with a high level of flow-induced turbulence, enforces FA and can culminate in fast deflagration propagation at a velocity of the order of 1000 m/s. A maximum deflagration velocity close to the speed of sound of the reaction products a_{pr} is often observed experimentally.

2.5 Flame Acceleration



Figure 2.20: Shadowgraph sequence of shocks ahead of a flame approaching an obstacle, obstructed channel, 30 vol. %, homogeneous mixture. $\bar{v} = 660$ m/s.

Such high-speed flames either continue propagating at this quasi-steady velocity or undergo transition to detonation. The structure of high-speed deflagrations at velocities close to a_{pr} is shown in Figs. 2.21 and 2.22 for an obstructed and unobstructed channel configuration, respectively. A series of precursor shock waves precompress the mixture. In the channel with obstructions, Fig. 2.21, the flame¹¹ follows a strong precursor shock at a small separation distance at a velocity similar to the shock velocity. The shock is not yet strong enough to cause auto-ignition in the mixture at an induction time τ_{ind} lower than the time difference between shock and flame arrival. As can be seen in Fig. 2.22, the presence of a single strong precursor shock is not necessary for fast flame propagation. In the unobstructed channel a series of shocks ahead of the flame precompresses the mixture incrementally.



Figure 2.21: Shadowgraph image of a fast turbulent deflagration, obstructed channel, 15 vol. %, homogeneous mixture. $\bar{v} = 880$ m/s.



Figure 2.22: Shadowgraph image of a fast turbulent deflagration, unobstructed channel, 30 vol. %, homogeneous mixture. $\bar{v} = 860$ m/s.

¹¹The flame manifests as a blurred dark area on the left side of the obstacle.

2.6 Onset of Detonation

FA as described in the previous sections can create critical conditions for onset of detonation. Ciccarelli and Dorofeev [20] divide onset mechanisms that can be observed experimentally and numerically into the following groups:

- Detonation initiation through shock reflection or shock focusing and
- onset of detonation caused by instabilities and mixing processes (e.g. shock-flame interaction, explosion of a quenched mixture pocket, pressure and temperature fluctuations in flow and boundary layers).

An important similarity of all mechanisms has been discovered in numerical simulations by Oran and co-workers, summarized in [111]. In each DDT simulation they performed, the origin of detonation was a localized hot spot in a sensitized mixture region, forming a local explosion which eventually evolved into a detonation wave. Urtiew and Oppenheim were the first to recognize this phenomenon of "explosion in the explosion" [148]. The following discussion is structured by answering three questions, which reflect the sequence of sub-processes during the onset of detonation:

- 1. How can a hot spot be formed?
- 2. How can a detonation wave emerge from a hot spot?
- 3. How does a locally initiated detonation transition into the macroscopic confining geometry?

The broad spectrum of answers to **question (1)** is outlined by Ciccarelli and Dorofeev [20]. Each of the incipiently named two groups of mechanisms can involve the formation of a hot spot. A universal theory to predict the development of hot spots as a first crucial requirement for the onset of detonation is currently not available. It is typically observed that a change in geometrical configuration, for instance obstacle spacing or blockage ratio, changes the dominant onset mechanism. Two examples which are relevant for the present



Figure 2.23: Simulated temperature fields for shock-induced ignition in stoichiometric ethylene-air mixture [111].

work are introduced here, namely shock reflection off a solid wall or obstacle and creation of a hot spot in the turbulent boundary layer behind a leading shock in an unobstructed tube.

Oran and Gamezo [111] present simulations of ignition after shock reflection at a solid wall in a stoichiometric ethylene-air mixture with non-slip boundary conditions, Fig. 2.23. The near-wall region is depicted after shock reflection. The reflecting wall is located at the righthand image boundaries. Incident shock Mach number is varied from 2.5 to 2.2. Due to high shock Mach numbers, shock bifuration occurs after reflection¹², which leads to the formation of an oblique shock close to the lower wall (lower image boundaries). This is clearly visible in case of $M_S = 2.5$. For the highest incident shock Mach number of $M_S = 2.5$, ignition occurs directly along the reflecting wall, discernible as a white area. It is independent of shock bifuration in this case. This corresponds to the strong ignition regime as discussed in Sec. 2.4. For lower shock Mach numbers, the ignition location first relocates to a higher point at the end wall ($M_S = 2.4$ and 2.3), which is due to the wall jet behind the bifurcating shock [111]. At $M_S = 2.2$, a hot spot forms at the lower wall in the turbulent wall boundary layer behind the reflected shock. From this hot spot a detonation may still evolve, cp. to the transitional regime between mild and strong ignition, Sec. 2.4. Since spatial fluctuations in temperature and pressure are

¹²Shock bifuration occurs as an interaction of reflected shock and wall boundary layer. The higher the incident shock Mach number and the lower the mixture heat capacity ratio γ , the more pronounced the bifurcation. It is an important non-ideal effect in shock tubes that can falsify measurements of induction times. Background information is given by [101].
present behind the reflected shock and since induction time is highly sensitive to temperature in the high temperature regime under consideration, the location of hot spot formation is random.

In the wall boundary layer behind a shock travelling in an unobstructed channel, a hot spot may form and cause onset of detonation. It is known that a local explosion initiating detonation may either occur between the leading shock and the turbulent flame brush or in the direct vicinity of the flame. Recently Dzieminska and Hayashi [37] showed a numerical investigation of auto-ignition and DDT by shock-wave boundary layer interaction in H_2 – O_2 . They found that the wall boundary layer is continuously compressed by multiple weak shock waves between a leading strong shock and the trailing flame. Auto-ignition occurs and a flame travels in the boundary layer towards the leading shock. At a certain point, a local explosion is observed that initiates a detonation. It is also possible that the initial auto-ignition triggers a detonation directly.

To address **question (2)**, Zeldovich et al. [165, 166] proposed the mechanism of spontaneous wave formation in a region with a gradient in temperature and thus induction time $\nabla \tau_{ind}$. They show that the spontaneous wave moves with a velocity D_{SP} that is determined by the induction time gradient:

$$D_{SP} = \frac{1}{\nabla \tau_{ind}}.$$
 (2.38)

Bartenev and Gelfand [3] give an overview of the large number of investigations related to the Zeldovich gradient mechanism. Oran and Gamezo [111] present numerical simulations of hot spot formation involving spontaneous wave development. If the initial spontaneous wave velocity is higher than the Chapman-Jouguet (CJ) velocity D_{CJ}^{13} , the wave relaxes towards the CJ state and can transform into a CJ detonation.

Lee et al. [93–95] proposed the SWACER (Shock Wave Amplification by Coherent Energy Release) mechanism, which might be seen as a generalization of the Zeldovich mechanism. The SWACER mechanism does not only consider gradients in temperature as a cause for gradients in induction time, but

¹³The concept of Chapman-Jouguet detonations is introduced in Sec. 2.7.

also allows for other origins. Comparable to the well-known Rayleigh criterion [124], which is used in thermo-acoustics to describe the feedback of fluctuations in pressure and heat release rate, Lee et al. state that continuous explosion of infinitesimal mixture volumes behind a shock needs to satisfy a coherence criterion in order to amplify the shock. Similar to the spontaneous wave concept, a gradient in induction time leads to the proper synchronization of energy release and shock wave motion. Theoretical studies can reproduce the SWACER mechanism, whereas its unambiguous experimental observation is still missing. Experiments conducted within the scope of the present work do not deliver information on the relevance of the SWACER mechanism.

When a sufficiently strong shock capable of causing rapid auto-ignition is produced in a hot spot explosion, it interacts with confining geometry and the transient flow field in the vicinity of the hot spot. Initiation of detonation may still fail at this early stage. Question (3) is treated by giving an example. For the present work, detonation initiation by shock reflection off an obstacle surface is highly relevant. This problem was studied for instance by Thomas [142] and Kellenberger and Ciccarelli [74]. Their results show the formation of a hot spot at the obstacle surface after shock reflection and an emerging detonation wave. This is in good agreement with the beforehand outlined mechanisms. Using the terminology introduced in Sec. 2.4, this process can also be described as strong ignition. Thomas [142] provides detailed experiments and simulations of the interaction of detonation waves produced by strong ignition after shock reflection at an obstacle with the flow field in the obstacle vicinity. He concludes that the expansion fan originating at the obstacle edge, as observed in Fig. 2.20 after shock passage at 75 μ s, can interact with the detonation wave emerging from the obstacle surface such that the detonation is substancially mitigated and eventually fails. This observation leads to a criterion for detonation initiation by shock reflection at an obstacle, based on comparison of obstacle height and induction time. If φ in Eq. (2.39) is lower that unity, detonation initiation fails.

$$\varphi = \frac{h}{a_{1r}\tau_{ind,1r}}$$
(2.39)

Here, a_{1r} is speed of sound and $\tau_{ind,1r}$ induction time behind the reflected shock. Note that this geometrical criterion does not answer questions (1) and

(2), but only addresses transition of a local explosion into the macroscopic geometry.

A more empirical approach has been presented by Dorofeev et al. [31]. The authors suggest the so-called 7λ criterion which expresses that a characteristic length scale L of the confining geometry needs to be at least seven times larger than the detonation cell width λ . Again, this geometrical interpretation does not address questions (1) and (2) and thus does in principle not include the formation of a local explosion as a requirement. Chemical kinetics of local explosions is not considered.

To complete the discussion about onset of detonation at this point, it is important to note that characteristic length scales of confining geometry need to be sufficiently large to allow for sustained detonation propagation. This is further discussed in Sec. 2.7.

2.7 Detonation

Detonations are nowadays known to be three-dimensional, highly dynamic complexes of shock waves and reaction zones propagating at supersonic velocity. In contrast to deflagrations, reaction is controlled by auto-ignition through shock heating. Despite their three-dimensionality and irregularity, one-dimensional description is surprisingly successful in predicting global detonation properties.

2.7.1 One-Dimensional Analysis

The simplest model to describe detonations in one dimension has been suggested by Chapman [18] and Jouguet [68], termed CJ model. The detonation front is treated as a single discontinuity. This model only differentiates between fresh mixture and the equilibrium state behind the detonation. The lowest possible detonation velocity in this model is equal to the stable solution of a one-dimensional detonation without losses. Such detonations are referred to as CJ detonations. Sonic flow of products behind the detonation wave characterizes this point. Taking into account laws of mass (Eq. (2.2)), momentum (Eq. (2.3)) and energy (Eq. (2.4)) conservation across the detonation front yields the product Hugoniot curve in a p-v diagram, Fig. 2.24. Conservation of mass and momentum form the Rayleigh line. In the Hugoniot diagram, the tangency point between Rayleigh line and product Hugoniot depicts the upper CJ point, the CJ detonation solution. Obviously, neither chemical kinetics nor the detailed structure of the detonation front play a role here since the CJ solution can be obtained by consideration of equilibrium states only. It is known from experiments that detonation velocity in a sufficiently large tube with low wall roughness agrees well with the CJ model, typically with a small (e.g. 2%) velocity deficit with respect to the ideal CJ value.

The ZND (Zeldovich, Von Neumann, Döring [35, 155, 164]) model splits the discontinuity of the CJ model into a shock and a trailing reaction zone, similar to the structure introduced in Sec. 2.2. Thus, this model can reproduce the interaction of shock and reaction zone. The Hugoniot diagram, Fig. 2.24,



Figure 2.24: Hugoniot diagram with Rayleigh lines (blue; red) and CJ tangency solutions.

yields the post-shock state as the intersection of Rayleigh line and adiabatic shock Hugoniot (q = 0), referred to as the Von Neumann (VN) state. From this state, reaction with specific heat release q leads to the upper CJ point. Non-equilibrium states of weak (intersection of Rayleigh line and product Hugoniot below the upper CJ point) and strong detonations (intersection of Rayleigh line and product Hugoniot above the upper CJ point) are not discussed here. Reaction behind the shock can be modeled by following the Rayleigh line from the VN state to the product Hugoniot, which represents chemical equilibrium. On the way, Hugoniot curves for partial heat release are crossed. Transition from the VN state to the upper CJ point can be approximated very accurately by a constant volume explosion. Shepherd [131] shows that induction times calculated with a ZND model and the constant volume explosion approximation are almost identical at H₂ concentrations between 13 and 70 vol. %.



Figure 2.25: ZND structure of a CJ detonation, 30 vol. % H₂ in air. Temperature and dimensionless heat release rate (a), mole fractions of major species (b) and minor species (c).

A ZND calculation of a CJ detonation in 30 vol. % H_2 in air is shown in Fig. 2.25. The precursor shock propagates at M_{CJ} = 4.9 and heats the gas at t = 0 s to a post-shock temperature of about 1540 K. The induction period is nearly isothermal. However, chain carriers (minor species) build up exponentially (c). Reaction is of combined thermal and chain-branching character. Induction time $\tau_{ind,CJ}$ is defined as the time difference between shock heating and the maximum temperature gradient, which coincides with the point of maximum heat release, shown as a dashed line in (a) (dimensionless depiction).

2.7.2 Three-Dimensional Structure

The structure of real detonation fronts greatly differs from beforehand discussed one-dimensional models. A real detonation exhibits transverse instability, which is typically required to sustain detonation propagation. A twodimensional illustration of a detonation front is shown in Fig. 2.26. Transverse waves oscillate perpendicularly to incident shock sections. Detonations can be classified according to their number of transverse waves in a given geometry as single-headed (one transverse wave) or multi-headed (more than one transverse wave) detonations. Intersection of incident shocks and transverse waves forms Mach stems and triple points. Triple point trajectories create a cellular pattern. Note that a regular pattern as shown in Fig. 2.26 as an instructive example is only observed in highly stable detonations, for instance achieved through high dilution with a monoatomic gas like argon [2]. The width of a cell is termed detonation cell width λ . Mach stems exhibit a higher post-shock pressure and temperature compared to the incident shock. Consequently, induction time behind Mach stem sections is lower than behind the incident shock. A shear layer emerges from the triple point due to gas velocity differences behind Mach stem and incident shock. Following the reaction zone progressing through a detonation cell, it starts behind a Mach stem originating at a triple point, which represents the point of transverse wave collision. Mach stem and reaction zone initially propagate at overdriven conditions with respect to CJ velocity D_{CI}. Propagation velocity ranges around 1.2 D_{CI} [131]. The Mach stem subsequently decays in strength and thus velocity and transforms into the incident shock. Towards the end of a cell, velocity re-



Figure 2.26: Two-dimensional detonation pattern. Shock waves and shear layer (blue lines); reaction zones (red regions); cellular pattern (grey lines). Adapted from [93] and [2].

duces to about 0.8 D_{CJ} [131]. Hence, reaction is initially closely coupled to the Mach stem (low induction time) and progressively distances. Approaching the end of a cell, shock and reaction zone can decouple significantly. Detonation propagation can thus be interpreted as a continuous sequence of initiation at hot spots formed by transverse wave collision at triple points and, depending on the mixture, failure by decoupling of incident shock and reaction zone. CJ and ZND model are thermodynamically equivalent to a three-dimensional detonation without losses, but cannot describe transient conditions within detonation cells. For instance, induction time $\tau_{ind,CJ}$ only exists in reality at one specific state within a detonation cell. Induction times at the beginning (the end) of a cell are orders of magnitude lower (higher) than $\tau_{ind,CI}$.

Detonation cell width λ has been found to correlate with mixture properties, in particular with effective mixture activation energy and reaction zone length [53, 131, 151]. Experimentally determined cell widths in H₂–air are shown in Fig. 2.27 [69]. For calculations presented in Ch. 6 experimental cell widths are



Figure 2.27: Experimental detonation cell width λ as a function of equivalence ratio Φ for H₂–air [69]. Data sources: \Diamond [57] (293 K, 101.3 kPa), + [143] (298 K, 101.3 kPa), × [5] (293 K, 82.7 kPa). Red line: approximation by Eq. (2.40).

approximated by:

$$\lambda = 1.706 \cdot 10^4 \cdot \exp\left(-9.755 \cdot \Phi\right) + 5.179 \cdot \exp\left(0.973 \cdot \Phi\right).$$
(2.40)

The cell width can be used to determine detonability limits in terms of mixture composition in a given geometry. Self-sustained detonation propagation in unobstructed channels of height H is typically possible if $H \ge \lambda$ [20]. For larger detonation cell widths, transverse instability cannot build up and detonation fails. Detonation propagation in flat mixture layers of height H (layer of reactive mixture bounded by a solid wall on one side and by an inert on the other side) has been investigated recently by Rudy et al. [129] and Gaathaug et al. [50]. A layer height of about $H \ge 3 \lambda$ is required for self-sustained detonation propagation.

2.8 Mixture Inhomogeneity

Although inhomogeneous mixtures are likely to prevail in real-world explosion accident scenarios, their scientific investigation has drawn surprisingly little attention yet. This knowledge gap has been noticed by various authors and renowned reports, e.g. [6, 12, 82]. Available studies on the topic can be categorized in terms of the relative orientation between concentration gradients and main direction of flame propagation, parallel or transverse. To study either the former or the latter case is an appropriate means of reducing complexity compared to directly addressing three-dimensional gradients in explosive clouds. Although only transverse gradients are investigated experimentally and theoretically in the present work, inclusion of parallel gradients into the discussion yields important conclusions regarding worst-case and realistic three-dimensional accident scenarios.

2.8.1 Parallel Concentration Gradients

Parallel concentration gradients are of particular interest if oriented vertically, thus additionally interacting with gravitational effects. This setting is for example highly relevant for nuclear reactors where the steam generator with a high degree of confinement resembles a long vertical tube, thus being exposed to a high DDT propensity [12]. As shown in Sec. 2.5.1, flammability limits for upward and downward flame propagation differ and can be substancially altered for a given volume by the presence of concentration gradients.

In case of globally very lean mixtures, ignition may be possible in a region of elevated fuel concentration also if the average concentration is below the flammability limit. Combustion can potentially consume a larger share of mixture and thus cause higher overpressure in an enclosure. Resulting peak overpressure has been found to increase in globally lean mixtures of H_2 and air [17, 158, 159].

It has furthermore been observed that maximum flame speed measured during flame passage through the gradient field of a mixture entirely within the flammability limits can be higher than the velocity observed in a homogeneous mixture at equal average concentration [19, 161]. This depends on the location of ignition and the gradient orientation. Positive vertical gradients, lean at the bottom, with bottom ignition seem to cause stronger acceleration than negative gradients [19]. Since only very few globally lean mixtures have been tested, generalized conclusions should however not be drawn precipitately. Detailed insight into the underlying physics has not been provided yet.

Sochet et al. published a series of papers on flame propagation in nonuniform clouds and vertical concentrations gradients [136, 137]. In their study on vertical gradients in a tube [27], the authors observe potentially stronger FA with gradients, supporting previously outlined results. Only single observations and rough trends are reported.

Sound quantification and modeling of explosions in mixtures with parallel concentration gradients is not available yet. A conservative approach for safety considerations is to predict explosion characteristics based on a homogeneous mixture of maximum locally existing reactivity. This however leads to unnecessarily high costs due to an overprediction of explosion consequences and thus very conservative design.

2.8.2 Transverse Concentration Gradients

Major knowledge on the **influence of transverse concentration gradients on DDT** in H₂–air has been obtained by experimental and numerical work at the Institute of Thermodynamics, Technical University of Munich, in cooperation with ProScience GmbH and Karlsruhe Institute of Technology. While the former group focused on entirely confined configurations at laboratory scale, the latter performed large and laboratory scale semi-confined tests.

Vollmer et al. [153] presented first experiments from the same setup as used in the present work. They showed that there can be a strong enforcing effect of concentration gradients on FA, particularly in a channel without obstructions. A comprehensive quantitative characterization was not carried out at that stage. It has been shown by the same authors [154] that probability of DDT can be increased by concentration gradients. A clear conclusion could not be drawn at this time since results for different geometrical configurations were ambiguous. The final report of the corresponding research project [40] as well as the respective PhD thesis [152] include first optical observations of flames in gradient mixtures. It can be seen that flames tend to elongate in the unobstructed channel and thus considerably change their macroscopic shape in gradient mixtures.

Kuznetsov et al. [85] recently reported that DDT in mixtures with transverse gradients in semi-confined geometries might be governed by the maximum local H_2 concentration. These studies were carried out in the ProScience large scale explosion experiment. Average H_2 concentrations were increased until onset of detonation occurred. The experimental approach was to compare homogeneous and inhomogeneous mixtures with equal maximum local hydrogen concentrations. The overall amount of hydrogen in the explosion volume was therefore different for homogeneous and inhomogeneous mixtures. Grune et al. [56] contribute experiments from a semi-confined small scale experiment, confirming the idea that the maximum local H_2 concentration dominates DDT propensity.

The following studies addressed **detonation propagation** in mixtures with transverse concentration gradients. Ishii and Kojima [64] examined fuel-lean H_2-O_2 and $H_2-O_2-N_2$ mixtures with transverse concentration gradients experimentally in a detonation channel of 40 mm height. Relatively weak gradients were used. Local equivalence ratio ranged from about 0.7 to 1 in case of the steepest gradient in H_2-O_2 . Tilted detonation fronts were observed in schlieren measurements. Soot foils showed detonation cells adapting dynamically to the local mixture composition. The authors furthermore found a velocity deficit of detonations in gradient mixtures compared to homogeneous mixtures. The average equivalence ratio was not kept constant between different gradients which complicates the quantitative interpretation of results.

Ettner et al. [41] performed Euler simulations of detonations in H_2 -air mixtures with transverse gradients. Curved multi-headed detonation fronts with a Mach-stem in the fuel-lean region were observed. The macroscopic detonation front shape remained constant over the propagation distance. Asymmetric wall pressure loads occurred, being highest in the region of lowest fuel concentration due to Mach-stem formation.

Kessler et al. [75] presented simulations in mixtures with varying activation energy and transverse gradients. They found a complex structure of the reaction zone including regions with delayed deflagrative combustion behind the detonation front. A deficit in propagation velocity of about 5–10 % was observed compared to the Chapman-Jouguet velocity D_{CJ} . This was compared to results by Calhoon and Sinha [16] who computed detonation velocities of about 94 % D_{CJ} before the gradients caused failure of the detonation. Local decoupling of shock and reaction zone was observed.

As will be shown in Ch. 6, detonations in transverse concentration gradients can exhibit similar characteristics as detonations propagating in two layers of mixture with different reactivity. More literature exists on such configurations. Dabora et al. [25] reported a velocity deficit of detonations in layers of H_2 – O_2 bounded by N_2 . A velocity deficit beyond 8–10 % lead to failure of detonation. Near this limit spinning detonations were observed.

Oran et al. [112] numerically studied detonation transmission in H_2 – O_2 from a primary to a secondary mixture. The authors compared their results to experimental work by Liu et al. [99]. Characteristic detonation patterns formed depending on the relative values of Chapman-Jouguet velocities of primary and secondary mixture. Detonations either failed or re-initiated in the secondary mixture. The authors pointed out that the unsteadiness of detonation transmission needs to be considered for predicting the detonation pattern.

Tonello et al. [145] investigated layered H_2 – O_2 mixtures experimentally. Similar to the aforementioned studies different types of diffraction patterns were observed depending on the respective reactivities of primary and secondary mixture. The detonation velocity in the mixture of higher reactivity was decreased while that in the other mixture was increased.

Lieberman and Shepherd [97] investigated detonation interaction with a diffuse interface between two mixture layers. They concluded that detonations curve and decoupling of shock and reaction zone may occur, depending on the local mixture dilution. In a more recent study Rudy et al. [129] investigated critical conditions of layered H_2 -air detonations in a semi-confined large-scale experiment. The mixture was bounded by a solid wall on the top and by air on the bottom. For homogeneous layers a minimum layer height for detonation propagation corresponding to 3 times the detonation cell size was found. They also examined mixtures with nearly linear transverse concentration gradients. Local concentrations were kept below stoichiometry. The mean H_2 concentration within the detonation layer needs to exceed approximately 16.6 vol. % to allow for detonation propagation. Locally, no detonation is observed if the local H_2 concentration falls below 14 vol. %.

Numerical simulations of detonations in layers of generic mixtures were recently presented by Gaathaug et al. [50] with a particular focus on the role of detonation front stability. A critical layer height of about 3 detonation cells for low activation energy mixtures (moderately stable) was determined. Failure and re-initiation of detonation was observed for high activation energy mixtures (unstable).

3 Experimental Setup

The experimental setup used in this work is a classical explosion channel. It was developed by and manufactured under supervision of K.G. Vollmer. Details are given in the respective PhD thesis [152] and the final project report [40], but shall be reviewed here for completeness. Besides information on design philosophy and geometry, Sec. 3.1 provides a list of geometrical configurations discussed in this work. Section 3.2 describes mixture preparation. The experimental procedure is outlined in Sec. 3.3. Measurement techniques applied are seperately introduced in Ch. 4.

3.1 Overview, Geometry and Configurations

The explosion channel operated at the Institute of Thermodynamics, Technical University of Munich, is comparable to typical explosion test facilities with high aspect ratio (length-to-diameter/width ratio). The basic idea to reach fast combustion regimes on laboratory scale is to provide a high degree of confinement and congestion realized as an entirely closed channel equipped with evenly spaced obstacles. This type of experiment was extended by a mechanism for the generation of transverse concentration gradients. Mild ignition is implemented by means of an electric spark.

Figure 3.1 provides a schematic of the setup. The channel is composed of six segments. Solid plates, referred to as ignition and end plate, close the channel at both sides. At this point, the coordinate system used throughout the present work is introduced: axial direction x, vertical direction y and lateral direction z as marked in Fig. 3.1. Standard segments with a length of 0.9 m and one optical segment (OS) with a length of 0.6 m are available. The channel can be operated with six standard segments, resulting in a total length of 5.4 m,



Figure 3.1: Schematic of experimental setup, exemplary configuration OS5. Facility top view (top) and explosion volume cross section (bot-tom).

or with five standard segments and the optical segment, giving a total length of 5.1 m. The optical segment can be placed at arbitrary positions along the channel, denoted OS1 (optical segment at position 1, x = 0 m to x = 0.6 m), OS2 etc.

The explosion volume cross-section has a width of 0.3 m and a height of 0.06 m. Underneath this volume, an additional volume intended for investigation of transverse venting is located. Explosion volume and venting volume are separated by solid plates. Only experiments without venting are discussed within the present work, thus the separation plates are installed at any time.

Notches in the top and separation plates at an equal spacing of 0.1 m allow for installation of obstacles. The channel either remains unobstructed (notches are covered with H_2 injection manifolds at the top and with flat inlays at the



Figure 3.2: Obstacle geometry. Side view.



Figure 3.3: Top obstacles BR60 (a), BR30 (b) and injection manifold (c). Cross sectional cut through injection plane.

bottom) or flat plate obstacles with a blockage ratio (BR) of 30 or 60 % are employed. Blockage ratio is defined as BR = 2h / H, cp. Fig. 3.2. Obstacle thickness is 0.012 m. Figure 3.3 shows the three types of channel top installations.

In the predecessor project conducted by Ettner and Vollmer [40], the channel was divided into an obstacle section beginning at x = 0.25 m and ending at x = 2.05 m and an unobstructed subsequent section. This setup was initially adopted in the present study to ensure comparability with previous results. Limitations due to the short obstacle section length were encountered and overcome by an additional configuration with an extended obstacle section beginning at x = 0.25 m and ending at x = 4.95 m.

The following notation is used to identify geometrical configurations:

$$BRxxSyyy(L). (3.1)$$

BR stands for blockage ratio, xx is the respective value in %, S for obstacle spacing, yyy is the spacing value in mm and the long obstacle section con-

Notation	Blockage ratio [%]	Spacing [m]	Obstacle section [m]
BR00	2		
BR60S300	60	0.3	0.25-2.05
BR30S300	30	0.3	0.25-2.05
BR30S300L	30	0.3	0.25-4.95

Table 3.1: Geometrical configurations discussed in the present work.

figuration is identified by the character L. Configurations discussed within the present work are listed in Tab. 3.1. Note that the subsequently described H_2 injection mechanism effects a small blockage ratio of 2 % in the unobstructed configuration BR00.

3.2 Generation of Transverse Concentration Gradients

Figure 3.4 illustrates the generation of concentration gradients. The last obstacle of BR60S300 (x = 2.05 m) and the first injection manifold of the unobstructed channel section (x = 2.15 m) are depicted. First, the channel is filled with ambient air. Using a vacuum pump, the volume is partially evacuated. Initial pressure prior to H₂ injection depends on the requested H₂ concentration and is calculated by the method of partial pressures. Then, H₂ at a pressure of 8 bar from a gas cylinder is injected through a regular pattern of injection ports in the facility top plate (1). This pattern is shown in Fig. 3.5 for a standard segment. Each row of ports comprises three ports across the channel width. Axial positions of rows coincide with notch positions in the top plate. Distributors connect 9 injection ports each to the H₂ supply system. In each distributor, a 0.9 mm aperture controls the flow rate.

The H_2 flow is deflected inside the channel, forming a compact horizontal H_2 layer along the channel top (2). At obstacle positions in the obstructed channel section, H_2 deflection is achieved by slots in the upper obstacles. Positions between obstacles as well as top plate notches in the unobstructed section are equipped with manifolds protruding into the channel at the upper wall (cp.



Figure 3.4: Creation of transverse concentration gradients. Gas injection (1), deflection (2), diffusion (3), formed gradients (4). Side view.



Figure 3.5: H₂ injection port pattern in a standard channel segment. Top view.

Fig. 3.3 (c)). These manifolds do not significantly influence the DDT process as will be discussed in Ch. 5. Likewise, they are not responsible for detonation phenomena observed in this work, see Ch. 6. Vertical concentration gradients form due to diffusion (3). The orientation of resulting gradients (4) is thus normal to the main direction of explosion front propagation.

Gradients of defined slope can be generated by controlling the diffusion time t_d between H_2 injection and ignition. A diffusion time of 60 s yields a homogeneous mixture, whereas a diffusion time of 3 s results in a steep concentration gradient. Further diffusion times of 10 s, 7.5 s and 5 s are investigated in the



Figure 3.6: Exemplary concentration gradient profiles from CFD simulations [42]. Variation of t_d between 3 and 60 s at 20 vol. % (a); variation of average H₂ concentration between 12.5 and 30 vol. % at t_d = 3 s (b).

present work. The described method for concentration gradient generation has been characterized experimentally and numerically in previous work by Vollmer et al. [154] and Ettner et al. [42], respectively. Profiles computed in the latter work are used within the present work to calculate local and integral mixture properties. Figure 3.6 gives a first impression of concentration gradients profiles. Please refer to App. A for a compilation of further concentration gradient profiles relevant for this work.

3.3 Summary of Experimental Procedure

Each experiment begins with mixture preparation according to the procedure outlined beforehand. Thus, the channel, filled with ambient air, is first partially evacuated. H₂-air mixture with the requested average concentration and

transverse gradient is prepared. Subsequently, the mixture is ignited at x = 0 m by an electric spark. Measurement systems are triggered off the ignition signal. After explosion, the channel is flushed with air for about 5 min to exhaust combustion products. The setup is then ready for the next experiment. Following this experimental procedure, the setup provides excellent reproducibility and a high test repetition rate with a total time for one experiment below 10 min.

4 Measurement Techniques

Measurement techniques used within the present work can be grouped into conventional and optical techniques. The former include time-of-arrival photodiodes, piezoelectric pressure transducers and soot-foils. The latter comprise shadowgraphy, OH planar laser-induced fluorescence (OH-PLIF) and OH* luminescence imaging. This chapter introduces these techniques and elaborates on their characteristics in application to explosion diagnostics.

4.1 Conventional Measurement Techniques

Figure 4.1 shows the photodiode (PD) and pressure transducer (p_1-p_7) layout in an exemplary configuration with the optical segment at position OS5. Each segment provides one pressure transducer and either eight (segments 1–3) or three (segments 4–6) photodiodes. The end plate is equipped with one additional pressure transducer (p_7) . Tables 4.1 and 4.2 contain positions of photodiodes and pressure transducers for the different segment types, respectively.



Figure 4.1: Schematic of photodiode (PD, red symbols) and pressure transducer (p_1 – p_7 , green symbols) locations. Configuration OS5. Top view.

Table 4.1: Photodiode positions for standard	l and optical segments, relative to
upstream segment edge.	

Segment type	Photodiode positions [m]
Standard 1–3	0.1 - 0.2 - 0.3 - 0.4 - 0.5 - 0.6 - 0.7 - 0.8
Standard 4–6	0.2 - 0.5 - 0.8
Optical	0.2 - 0.3 - 0.4

Table 4.2: Pressure transducer positions for standard and optical segments,relative to upstream segment edge.

Segment type	Pressure transducer position [m]
Standard 1	0.4
Standard 2–6	0.5
Optical	0.3

4.1.1 Time-of-Arrival Photodiodes

The explosion channel is equipped with UV-sensitive photodiodes in the top plates close to the center line (z = 0.135 m), type Hamamatsu S1336-18BQ. Since these diodes register the broadband luminescence of passing combustion waves, they can be used to determine arrival times of both deflagrations and detonations. Diode mounting is depicted in Fig. 4.2. By setting back the diode from the channel wall and optically connecting it through a hole, the resulting narrow 10° angle of view raises the resolution of combustion wave arrival time masurement. A quartz glass window with a high transmittance in the UV protects the diode from high temperature and overpressure. The diode current is amplified by a custom-made signal amplifier. This amplifier has two internal settings for the amplification factor. For lean mixtures in the range of 15 vol. % and below, a particularly high amplification factor is required due to low flame luminescence intensity. For higher H₂ concentrations, the lower amplification factor setting can be used, yielding a better signal-tonoise (SNR).



Figure 4.2: Mounting of photodiodes, adapted from [40].

Data acquisition is performed using a 50 channel A/D converter at a sampling rate of 250 kS/s.

Typical measured voltage profiles from a representative experiment are shown in Fig. 4.3. It can be seen that a gradual rise in voltage occurs when the flame tip passes the first photodiode at x = 0.1 m. The faster the flame propagates, accelerated by obstacles in the exemplary configuration, the steeper the signal rise at respective photodiodes. In signal post-processing, flame arrival is defined as the point where the measured voltage rise exceeds 1 V. This definition is adopted from the work of Vollmer [40]. For the derivation of profiles of flame tip velocity over x-coordinate, linear interpolation between the photodiode positions is performed. The velocity plot corresponding to photodiode signals shown in Fig. 4.3 is given in Fig. 2.4.

4.1.2 Piezoelectric Pressure Transducers

Explosion overpressure is captured by dynamic piezoelectric pressure transducers, type Kistler 601A, combined with Kistler 5011B electrostatic charge amplifiers. Data acquisition at a maximum rate of 250 kS/s is performed by means of a NI PCI-6133 internal multifunction board which allows for simultaneous measurement of 8 channels. All transducers are mounted flush with the walls close to the channel center line (z = 0.165 m). All channel segments and the end plate alike provide one measurement position. Segment transducers are mounted in the channel top plate.



Figure 4.3: Voltage signals from photodiodes corresponding to velocity plot Fig. 2.4.

The pressure transducers have a measurement range of 0–250 bar overpressure and a natural frequency of 150 kHz. A high natural frequency is required for the measurement of highly dynamic pressure loads inherent to explosion processes. However, the finite value poses a limitation regarding the maximum useful measurement frequency. This will be explained in the following.

In general, a considerable amplitude overprediction occurs when the physical pressure signal frequency approaches the transducer's natural frequency as demonstrated in a NASA report for several frequently used transducer types [127]. The acceptable amplitude overprediction poses an upper limit to the useful frequency range of a specific transducer. Since this contradicts the requirement of high temporal resolution to accurately capture explosion dynamics in fast regimes, a tradeoff needs to be made. Theoretical considerations to define a meaningful upper frequency limit are outlined here.

The transducer behaves similar to a 1-D underdamped driven harmonic oscillator. The dynamic response of a flush-mounted¹⁴ pressure transducer can be described in terms of amplitude overprediction by the steady-state under-

¹⁴Due to flush mounting, no additional consideration of transmission volumes is required. The dynamic response of the measurement system is that of the transducer itself [135].



Figure 4.4: Dimensionless amplitude A over dimensionless angular frequency ω for the underdamped driven harmonic oscillator.

damped driven harmonic oscillator solution in complex notation:

$$A = \frac{F}{-m\omega^2 + i\omega b + k}.$$
(4.1)

A is the oscillation amplitude, normalized by the amplitude which would occur for $\omega \rightarrow 0$, F is the external driver force, m is mass, ω is the angular driver frequency normalized by the transducer resonance frequency, b is the damping ratio and k is the spring constant. Calculation of this relation assuming a small damping ratio (b = 0.1)¹⁵ yields the graph in Fig. 4.4, showing the dimensionless amplitude A as a function of dimensionless angular frequency ω . The upper useful frequency is defined in the present work such that the maximum amplitude overprediction is 10 %. This is given at frequencies lower than 0.3 times the natural frequency, cp. Fig. 4.4, red line. This restriction is realized by retaining a maximum data acquisition rate of 250 kHz and deploying a low-pass filter with a cut-off frequency of 45 kHz within the digital signal post-processing. Highly time-resolved raw data thereby remains available for the determination of shock or detonation arrival times.

¹⁵Increase of b from 0.1 to 0.5 raises the upper useful dimensionless frequency only by 0.02. Thus, the exact value of b is of minor influence for the argumentation outlined here.



Figure 4.5: Example for thermal shock. Transducers 1 (green line), 2 (blue line) and 3 (red line, with thermal shock causing a negative offset indicated by arrows).

Another important source of pressure signal falsification is the effect of thermal shock [36]. This effect is caused by non-uniform transient heating of the transducer during combustion wave passage. The transducer diaphragm has a low thermal capacity compared to the housing. Since the thin diaphragm heats up quickly and thereby expands, the preloading imposed on the quartz cristal stack inside the transducer at neutral conditions is reduced. This causes an underprediction of overpressure and potentially even negative measured overpressure in the long-term response behind a combustion wave. Such results need to be carefully analyzed and their accuracy scrutinized. Since the effect builds up dynamically over time, beginning with the first contact of the transducer with hot gas, it is nearly impossible to correctly readjust the overpressure trace in post-processing. However, the short-term response is observed not to be significantly altered in many experiments so that measured maximum peak pressures in fast combustion regimes may still be accurate.

Figure 4.5 shows an experiment in which valid pressure curves are obtained from transducers 1 (green line) and 2 (blue line). Transducer 3 (red line) shows obvious signs of thermal shock well visible at the arrow position where the

measured pressure of all transducers should assimilate since pressure differences within the channel decrease after multiple passage of longitudinal shocks and decay of shocks towards the acoustic limit.

This problem is commonly overcome by covering the transducer diaphragm with a thin flat layer of high-temperature grease or silicone. In the present work, both materials were tested and no major difference in the resulting pressure signals was detected. Since high-temperature silicone forms a more resistant layer and thus remains intact for a larger number of experiments, this material was selected.

Note that data plotted in Fig. 4.5 is unfiltered and thus also represents an illustrative example of amplitude overprediction at transducer 3 and t = 13.5 ms by a factor of approximately 1.5. In this experiment a local explosion close to transducer 3 occurs, causing the high local peak overpressure. The rapid pressure rise due to the blast wave impinging on the transducer causes amplitude overprediction and overshoot into the negative pressure range. The overshoot clearly supports the assumption of underdamping in the estimation of the upper useful measurement frequency.

4.1.3 Soot-Foils

Recording the cellular pattern of detonations as introduced in Sec. 2.7 on sooted plates installed at channel side walls helped to discover the threedimensionality of detonation fronts [28]. This technique is utilized in the present work to compare the cellular pattern of detonations in homogeneous and inhomogeneous mixtures. Application is imaginably simple: Thin steel plates are covered with soot, preferrably from a heavy hydrocarbon flame, and installed flush on the channel walls. Trajectories of detonation front triple points correlate with the observed soot foil traces as demonstrated by Urtiew and Oppenheim [148]. The exact mechanism of soot removal has not yet been identified unambiguously. Mechanisms like sheer stress orientation differences behind triple points may be responsible as shown experimentally [87] and numerically [63].

4.2 Optical Measurement Techniques

Broad application of advanced optical measurement techniques is one of the major features of the experimental approach pursued in the present work. Three highly time-resolved optical measurement techniques were chosen for the visualization of explosion processes: Shadowgraphy, OH planar laser-induced fluorescence (OH-PLIF) and OH* luminescence imaging. Optical information is used complementary to conventional measurement data. Since each of the three techniques contributes with its specific properties and associated potentials and limitations, especially the joint or even simultaneous application provides valuable insight.

All named techniques depend on scientific high-speed cameras as imaging devices. Two non-intensified CMOS cameras (Photron SA-5 and SA-X), one modular external image intensifier (Hamamatsu C10880-03) and one image-intensified camera (Photron APXI²) were used. The non-intensified cameras can be combined with the external image intensifier to form an intensified high-speed system. Technical specifications can be found in the respective datasheets [59, 119–121]. Three different camera lenses were employed, providing transmittance in the visible (Nikkor 85 mm 1:1.4) and ultraviolet (UV-Nikkor 105 mm 1:4.5 and UV-CERCO-SODERN 45 mm 1:1.8) spectrum.

4.2.1 Shadowgraphy

Since shadowgraphy and the schlieren technique are closely related, they are often described collectively. Toepler [144] first introduced the schlieren technique as a method to visualize spatial non-uniformities in refractive index in transparent media. The irregular deflection of a collimated beam passing a transverse gradient in refractive index is used in a manner that the non-deflected rays serve as a reference, whereas deflection leads to the depiction of non-uniform regions. Since density gradients effect refractive-index gradients via alteration of speed of light, shadowgraphy and schlieren allow for visualizing gradients in density and thereby in pressure and temperature of gases. Line-of-sight integration is inherent to the techniques. The fundamen-



Figure 4.6: Principle of schlieren visualization. Light rays are deflected by spatial gradients in refractive index and blocked by a knife edge.

tal physics shall not be further reviewed here since the techniques have been representing a scientific standard for many decades and are widely used. For background information, the reader is referred to [130]. A basic in-line optical setup for schlieren visualization is shown in Fig.4.6. The schlieren technique requires a schlieren knife edge to block deflected light. The setup sensitivity can be adjusted by moving the schlieren knife edge as indicated in Fig.4.6. For shadowgraphy, the schlieren knife edge is removed. The difference between shadowgraphy and schlieren is that the former displays the second spatial derivative of the refractive index, while the latter visualizes the first derivative. Replacing the lenses in Fig. 4.6 by a collimating and a focusing mirror and adding two planar mirrors leads to the setup employed in the present work, Fig. 4.7. An LOT Oriel 350 W Xe light source is used. The depth-of-field Δz of such a setup can be expressed as the ratio of the acceptable circle of confusion Φ over the aperture angle of the light source α , where $\alpha = d/f_1$ for small α . d is the light source diameter and f_1 is the focal length of the collimating mirror.

$$\Delta z = \frac{\Phi}{\alpha} = \frac{\Phi f_1}{d} \approx 500 \text{mm}$$
(4.2)

For the specific setup used here with $f_1 = 2500 \text{ mm}$, $d \approx 5 \text{ mm}$ and $\Phi = 1 \text{ mm}$, the depth-of-field accommodates a value of 500 mm. Since the depth-of-field is larger than the 300 mm width of the explosion channel, the shdaowgraph setup is "unfocused". Gradients in refractive index along the lateral dimension of the test section contribute equally to the integral light deflection, so that the lateral position of a disturbance does not affect the resulting image.

4.2 Optical Measurement Techniques



Figure 4.7: Schematic of the shadowgraphy setup used in the present work, top view.

Shadowgraphy has an advantage over the schlieren technique in the present application. Since the sensitivity of a schlieren setup is linearly dependent on the distance between the schlieren object and the imaging plane, termed optical lever arm, the large effective optical lever-arm of approximately 4 m of the present setup yields a very high sensitivity. Along with the large width of the explosion channel this leads to detailed images throughout the entire range of investigated combustion regimes even without a schlieren knife edge. Using a schlieren knife edge decreases the visibility of details in regions with steep density gradients.

4.2.2 High-Speed OH Planar Laser-Induced Fluorescence

This section first provides an introduction to the purpose and fundamentals of OH planar laser-induced fluorescence (OH-PLIF) measurement, then describes the utilized OH-PLIF laser and camera system including details of component synchronization and finally presents exemplary OH-PLIF images to discuss opportunities and limitations of the high-speed OH-PLIF technique applied to explosion processes. One goal within the project behind this thesis was to develop a pulsed OH-PLIF system operating at repetition rates of up to 40 kHz, suitable to resolve fast combustion processes. Components of a pulsed laser system were procured, installed, tested at laboratory flames and finally applied to the explosion channel.

OH-PLIF allows for capturing two-dimensional images of flame fronts by visualizing OH radicals, introduced in Sec. 2.3 as an intermediate species of H_2 – O_2 reaction. Since the technique was only used for visualization purposes in the present work, the subsequent discussion will be confined to qualitative OH-PLIF. For further information on quantitative measurements for the determination of local species concentrations, including in particular relevant quenching effects, please refer to [77, 86, 100, 122].

Laser-induced fluorescence comprises two major steps: excitation of OH radicals by absorption of a photon of specific energy and subsequent emission of a photon of lower energy from an excited singlet state. This process is termed fluorescence. The observed shift in photon energy ΔE , known as Stokes Shift, is equivalent to a shift in wavelength $\Delta \lambda$. The relation is given by Planck's law:

$$\Delta E = \frac{hc}{\Delta \lambda}.$$
 (4.3)

Here, h is the Planck's constant and c the speed of light. Thus, emission occurs at a higher wavelength compared to excitation. The fluorescence signal can thus be separated from the excitation wavelength by means of an interference filter.

Energy transitions between excitation and emission can be visualized in a Jablonski diagram as shown in Fig. 4.8. The Jablonski diagram is an intuitive means of illustrating energy states of a molecule and respective transitions.



Figure 4.8: Jablonski diagram for the $A^2\Sigma^+ - X^2\Pi_i$ electronic band system of OH with fluorescence in (0,0), adapted from [77].

The Q₁(6) transition used in the present work for OH-PLIF measurements is visualized. The number in parenthesis corresponds to the rotational quantum number of the ground state, whereas the letter Q denotes a transition at constant rotational quantum number. Starting from the ground state of the OH molecule at the lowest vibrational state $X^2\Pi_i$ (v"=0), excitation to the vibrational state v'=1 of the first electronically excited state $A^2\Sigma^+$ is achieved by absorption of a photon. Depending on the photon's energy, different rotational energy states within the vibrational state v'=1 can be reached. After excitation, internal conversion by rotational energy transfer (RET) or vibrational energy transfer (VET) rapidly leads to redistribution to lower levels within the electronically excited state. Transitions to the ground electronic state through fluorescence thus start from various energy states within the electronically excited state. Only a small share of molecules transitions from the highest ex-

cited state, which would be termed resonant fluorescence. Besides fluorescence, several non-radiative possibilities exist for the return. The fluorescence quantum yield, which is the number ratio of emitted fluorescence photons to absorbed photons, is consequently smaller than unity. For quantitative applications detailed knowledge of non-radiative de-excitation processes, collectively termed quenching, is necessary. For the present work mainly the dependency between quantum yield and pressure is of interest. As Pintgen [122] shows, quenching increases with an increase in pressure, which can be explained by the higher frequency of intermolecular collisions leading to an enhanced collisional quenching rate. This relationship will be of importance for the discussion of OH-PLIF applicability to explosion experiments.

Transitions to the ground electronic state by fluorescence emit photons, which can finally be recorded. Dependent on the employed optical camera filter, several emission wavelengths are detected at once. Transitions to a state at higher energy than the ground state is likely. The absolute ground state can be reached again through internal conversions between vibrational and rotational states. Energy losses in internal conversions both in the excited and in the ground state cause a Stokes Shift for each single transition.

For a particular molecule like OH, an excitation scan delivers information on the detectable fluorescence intensity as a function of excitation wavelength [77]. Such spectra can be calculated using the LIFBASE spectral simulation tool [100]. This tool has been developed at SRI International with the goal to compile available information on spectral properties of diatomic molecules most relevant for LIF measurements. Figure 4.9 shows such a calculated spectrum for the $A^2\Sigma^+ - X^2\Pi_i$ electronic band system of the OH molecule around 283 nm. Each line represents a possible rotational state or an overlap of several lines below the wavelength resolution of the simulation. The Q₁(6) line offers the highest theoretical fluorescence intensity at an exemplary temperature of 2000 K. A practical approach to achieve maximum fluorescence intensity is to perform a wavelength scan over the tunability range of the deployed laser system, whereby a calculated optimal wavelength can provide a first estimate. For the present work, an optimum excitation wavelength setting of 282.945 nm (calibrated readout of dye laser control software, hereinafter referred to as



Figure 4.9: Excitation scan for OH, LIFBASE [100], T = 2000 K. Wavelength conversion from vacuum to air according to Morton [107].

283 nm) was determined experimentally using an atmospheric premixed H_{2} -air test flame. The wavelength setting of the dye laser has an accuracy of 0.03 nm [134]. Thus, the experimental and theoretical optima coincide within the hardware precision.

In most combustion diagnostics applications of OH-PLIF a pulsed laser system is used, providing high intensity during the short laser pulses [77]. This allows for suppressing the contribution of flame luminescence through choosing a short camera exposure time. The pulsed laser system used in the present work is schematically illustrated in Fig. 4.10. The essential component of the system is a pulsed, frequency doubled Nd:YVO4 pumplaser (Edgewave IN-NOSLAB IS8II), emitting light at 532 nm from two cavities. Each of the cavities can be individually triggered at repetition rates of up to 20 kHz with a pulse length of 8 ns. The design pulse energy is 2 mJ, resulting in an average power of 80 W. This laser is combined with a tunable dye laser (Sirah Credo) designed for the respective repetition rates and pulse energies. It consists of a resonator stage, an amplifier stage, a second harmonics generator (SHG) and



Figure 4.10: Schematic of the OH-PLIF laser system, adapted from [134].

a wavelength separation unit. Through this setup, the pumplaser wavelength is first converted to the fundamental wavelength of the resonator (setpoint is 565.8892 nm in the present work, hereinafter referred to as 566 nm), which can be adjusted by moving the tunable resonator end mirror. A hybrid multipleprism grazing-incidence (HMPGI) resonator design with one-dimensional intracavity beam expansion is used. An additional amplifier stage is required to obtain sufficient power at the fundamental wavelength. The SHG, also called frequency doubling unit, with a nonlinear temperature-stabilized Beta Barium Borate (BBO) crystal and a compensator generates a share of photons with twice the frequency compared to the incident photons and thus 283 nm wavelength. Both 566 and 283 nm exit the SHG due to incomplete conversion. The 283 nm wavelength is separated from the 566 nm wavelength in the separation unit, using an arrangement of four Pellin-Broca prisms and a beam stop. In principle, one prism would already fulfill this function, but the four
prism arrangement avoids variation of the 283 nm beam output position and angle when the wavelength is varied.

Rhodamine 6G (R6G), also termed Rhodamine 590, dissolved in Ethanol is used as a lasing medium in the dye laser. The spectroscopic characteristics of solutions of R6G at different concentrations were recently studied by Zehentbauer et al. [163]. Tunability of laser emission ranges from about 560 nm to 610 nm with a peak around 575 nm. This covers the desired fundamental dye laser wavelength of 566 nm. Maximum absorption is achieved at a pumplaser wavelength of 530 nm, very close to the second harmonics of Nd:YAG and Nd:YV04 lasers of 532 nm. The highest laser emission intensity can be expected at R6G concentrations around 0.1 g/l. Concentrations of 0.135 and 0.09 g/l in the resonator and amplifier, respectively, were used in the present work following recommendations of the laser manufacturer. Information on different laser dye solutions, their range of applicability and conversion efficiencies is summarized in [9].

The dye laser is optimized for being pumped with vertically polarized laser light (s-pol). Since the two pumplaser cavities have different polarizations (spol and p-pol) to allow for beam overlap, the conversion efficiency for s-pol is as desired, but the efficiency for p-pol is inadequate. To use both cavities and thus obtain a 40 kHz repetition rate, a $\lambda/2$ waveplate can be introduced between pump and dye laser to rotate the polarization orientation of both beams at an angle of 45°. This is achieved by an angle between the axes of the wave plate and the polarization planes of the incident beams of $22.5^{\circ 16}$. The rotational position of the $\lambda/2$ plate needs to be precisely adjusted to reach equal dye laser output pulse energies with both pump laser cavities. Utilization of the $\lambda/2$ plate leads a reduction in pulse energy of about 30–40 % compared to the performance of the s-pol cavity alone. Since application of the OH-PLIF system to fast combustion regimes requires the maximum achievable pulse energy, only the s-pol cavity could be used for the measurements presented in this thesis. If a smaller FOV is sufficient and self-luminescence of the investigated flame is low, the double-cavity option with $\lambda/2$ plate is ap-

¹⁶The Jones calculus [66] allows for description of polarized light passage through a wave plate. It illustrates that birefringent material of thickness $\lambda/2$ rotates the plane of polarization of a linearly polarized incident beam by twice the angle that incident beam polarization plane and the plate's fast axis confine [60].

plicable. The requirement for very exact overlap of the two pumplaser beams however complicates the adjustment procedure. Thermal steady state of the pumplaser needs to be reached (after approximately 20 min of operation) before adjustment of the beam overlap unit can be performed. The pumplaser power during the adjustment procedure needs to equal the final power during OH-PLIF operation (typically maximum power setting of 80 W). Therefore, highest caution should be exercised and the pumplaser beam intensity reduced by multiple reflection off wedged windows, before passing the laser beam to an observation target. The target is preferentially placed at a distance from the pump laser equal to the distance between pumplaser and amplifier cuvette since precisely parallel beam alignment along the entire beam propagation distance is difficult to achieve.

Due to the limited maximum dye efficiency of about 28 % for R6G and considerable losses during frequency doubling (maximum SHG efficiency of 20 %), the achieved output pulse energy is 120 μ J or less depending on the system configuration. Well-established low speed OH-PLIF systems deliver pulse energies ranging from several mJ (pumped dye lasers) up to a few 100 mJ (excimer lasers) [103]. However, the low pulse energy of the high-speed system designed here was found to be sufficient for OH-PLIF imaging in a FOV width of up to 100 mm. A short camera exposure time of the order of 30 ns is mandatory to suppress flame luminescence which originates from the same energy transitions as the fluorescence signal and thus cannot be eliminated by means of optical filters. Measures need to be taken to avoid losses during beam guidance (use dielectric UV mirrors with a high reflectance), laser sheet formation (use quartz glass lenses with UV AR coating) and delivery of the laser sheet into the test section (provide high transmittance at 283 nm, thus low OH concentration, keep windows clean), as well as to optimize the laser output power, the beam profile and the sensitivity of the imaging system.

The 283 nm dye laser output beam is guided towards the test section by mirrors with a dielectric coating designed for a reflectance > 98 % at 283 nm. Only redirections of 90° are permitted to avoid deformation of the oval-shaped UV laser beam. The vertical dimension of the beam is then expanded by a UV AR coated -30 mm cylindrical lens and collimated by a UV AR coated 500 mm

spherical lens. The formed light sheet is introduced into the optical segment through a window in the channel top plate. The distance between spherical lens and symmetry line of the explosion channel (y = 0.03 m) is equal to the 500 mm focal length of the spherical lens to produce a thin light sheet (about 0.1–0.3 mm thick).

For image acquisition, an external image intensifier (Hamamatsu C10880-03) is combined with a high-speed camera (Photron SA-X or SA-5). A Semrock BrightLine HC 320±20 nm bandpass filter is applied to the 45 mm UV camera optics for detection of the OH-PLIF signal, which appears mainly between 306 and 324 nm due to fluorescence from (0,0) and (1,1) transitions. Synchronization of the pump laser cavities, camera and image intensifier as well as optional further cameras for simultaneous application of other measurement techniques like OH* luminescence imaging is accomplished by two Stanford Research Digital Delay Generators (DG535 and DG645). Control signals, laser pulses, camera and image intensifier exposure windows and respective delays are shown in Fig. 4.11. A first estimation of delays was achieved by using a photodiode detecting laser emission and comparing against the camera exposure output signal. Fine tuning of the delays in the ns range needs to be performed by adjusting the image intensifier delay at maximum pumplaser power until stable maximum fluorescence from a test flame is visible on camera images. In this way, the delay between laser pulse and fluorescence emission is accounted for. Sufficiently low jitter of the laser pulse output is only reached at maximum pumplaser power. Image intensifier exposure times lower than 30 ns resulted in fluctuations in recorded fluorescence intensity due to jitter.

The raw OH-PLIF images are post-processed in order to reduce image intensifier noise and facilitate interpretation. An algorithm for noise reduction has been implemented based on the MATLAB bwboundaries method. This method identifies small isolated structures with a boundary vector including less than 20 pixel-size elements and eliminates them by masking. In addition, intensities lower than 5 % of the maximum intensity within a series of images are removed. Images are finally shown in false-color depiction for better visibility. Examples can be seen in Figs. 4.12, 4.13 and 4.14.



Figure 4.11: OH-PLIF synchronization scheme. Negative edges of camera exposure and trigger signals not depicted.

In this work, the first application of time-resolved OH-PLIF to an FA experiment reaching fast regimes—to the best of the author's knowledge—was performed. Therefore, achievable results of high-speed OH-PLIF application to explosion diagnostics will be discussed subsequently to provide practical guidance. Images presented here will not be interpreted from the explosion physics point of view, but only in terms of experimental image quality. An FOV width of up to 100 mm at pulse energies around 100 μ J was achieved. In all experiments, the highest image intensifier gain was chosen to achieve exposure covering the entire dynamic range of the camera sensor. The major challenge is to obtain a high signal-to-noise ratio (SNR), defined here as the ratio of fluorescence signal to background noise mainly generated by flame luminescence.



Figure 4.12: OH-PLIF sequence of a slow flame, $\bar{v} = 50$ m/s, BR00, OS5, 20 vol. %, homogeneous, FOV width 98 mm, SNR 20–50.

Exemplary OH-PLIF images of a slow deflagration propagating at an average velocity of 50 m/s are presented in Fig. 4.12 including raw and processed images. The images are of high quality regarding the SNR, which is in a range of 20–50. This value can be estimated by comparing intensities in the region where the light sheet is present (right to the white dashed line) to intensities in the region where only flame luminescence is recorded (left to the white dashed line). Temporal resolution is reduced for visualization in Fig. 4.12. These results are well comparable to OH-PLIF measurements of slow flame propagation from other groups, e.g. [58].



Figure 4.13: OH-PLIF sequence of a fast flame, $\bar{v} = 380$ m/s, BR00, OS5, 25 vol. %, homogeneous, FOV width 91 mm, SNR 2–5.

Reaching velocities around the speed of sound of the reactants a_{re} , the SNR is distinctly lower than in the slow regime, around 2–5. About five images of the leading flame tip can be taken within the FOV. Figure 4.13 gives an example. The region to the left of the light sheet shows flame luminescence, while sharp structures along the leading flame front originate from OH-PLIF. The flame front is still well detectable under these conditions. Absorption of the laser light, passing the flame from the top, is considerable and reduces the fluorescence intensity by a factor of about 2, which can be seen by comparing the upper and lower part of the leading flame front.

When flame speed approaches the speed of sound of the reaction products a_{pr} , the SNR decreases sharply to values of unity and below, Fig. 4.14. This makes high-speed OH-PLIF with the hardware specifications described beforehand inapplicable. The flame front can no longer be detected in this regime. Naturally, this also applies to the detonation regime. In Fig. 4.14, a local explosion at the channel top additionally overexposes the image. The image intensifier reacts with extensive blooming. Three effects are believed to be mainly responsible for the limitation of OH-PLIF towards fast flame propagation with high local pressures and temperatures:

- Increased flame luminescence intensity due to thermal production of OH*. This is further discussed in Sec. 4.2.3.
- Reduction of fluorescence intensity due to increased collisional quenching rates [122].
- Strong absorption of the 283 nm laser sheet already in the upper region of the channel. The effect of pressure on absorption was investigated separately in a high pressure flame absorption experiment, presented in App. B.

Additionally, only about two images can be expected within the FOV. This clearly suggests to favor single-shot OH-PLIF for regimes close to onset of detonation and beyond. Examples of single-shot detonation investigations have been published by Eder [39] and Pintgen [122]. The author is not aware of successful attempts to capture onset of detonation with OH-PLIF.



Figure 4.14: OH-PLIF sequence of a fast flame, $\bar{v} = 900$ m/s, BR00, OS5, 30 vol.%, homogeneous, FOV width 97 mm, SNR < 1.</td>

4.2.3 OH* Luminescence

Like shadowgraphy, OH* luminescence imaging is an optical measurement technique integrating along the optical path. It is widely used in combustion diagnostics being simple to apply and cost-effective. Photons emitted during transitions of OH radicals from excited states $A^2\Sigma^+$ (denoted OH*) to ground states $X^2\Pi_i$ can be captured with an image-intensified camera combined with a UV transmissive camera lens and a 307 ± 5 nm bandpass filter. Although the location of light emission may not exactly coincide with the region of heat release and neither correlate linearly with heat release rate in turbulent flames as discussed by Lauer [89], it is still applicable for the simple visualization of reaction zones within the present work.

The particular advantage over the OH-PLIF technique is that no laser is required and that the imaging rate and FOV is therefore only limited by the utilized camera. On the other hand, the drawback is line-of-sight integration, hiding details of flames and detonations that can only be recognized in twodimensional depictions. The technique is therefore used to complement OH-PLIF measurements and replaces it beyond the limits of OH-PLIF applicability.

Fiala and Sattelmayer [47] recently discussed details of OH* luminescence imaging. While the technique is commonly referred to as OH* chemiluminescence in investigations of deflagrations indicating that excitation of OH radicals is of chemical origin, this assumption does not necessarily prove true in application to fast combustion regimes. As Fiala and Sattelmayer demonstrated by non-premixed counterflow flamelet simulations, thermal excitation dominates over chemical excitation in high temperature flames. Thermal excitation exceeds chemical excitation at flame temperatures above 2700 K. The thermal excitation rate depends exponentially on temperature in the relevant range, whereas chemical excitation remains fairly constant at a given pressure. According to [45] the molar concentration of OH*, [OH*], can be written as

$$[OH^*] \propto [OH] \cdot e^{\frac{-\Delta g_m^0}{RT}}, \qquad (4.4)$$

assuming thermal equilibrium. [OH] is the molar concentration of OH in the



ground state, R_m the universal gas constant, T the flame temperature and Δg_m^0 the difference in standard-state Gibbs enthalpy between OH^{*} and OH.

Figure 4.15: Temperature field shortly before onset of detonation. CFD simulation [42]. 25 vol. %, inhomogeneous mixture, t_d = 3 s.

Matching this understanding with simulations of the DDT process by Ettner [42], local temperatures of about 2700 K are indeed reached in the turbulent flame brush shortly before the onset of detonation as seen in Fig. 4.15. This supports the hypothesis constructed in Sec. 4.2.2 regarding the limitation of OH-PLIF applicability towards high flame speeds. Thermal excitation of OH causes strong OH* luminescence, exceeding the laser-induced fluorescence signal. In case of detonations, where local temperatures at CJ state of up to 3000 K are obtained, thermal production of OH* can be assumed to clearly dominate. At locations of local explosions during the onset of detonation, even higher temperatures are expected due to overdriven conditions. The distinct temperature dependency of OH* luminescence in the high temperature range can on the other hand be used to gain estimates of the local mode of combustion and discriminate deflagration, CJ detonation and local explosions according to their luminescence intensity in ascending order. Two optical setups have been developed to record OH* luminescence at the experiment, presented in Fig. 4.16. Firstly, the Photron APXI² camera was used to record the entire window section of the optical segment simultaneously to OH-PLIF deflagration measurements. A planar mirror with a high reflectance in the UV range was used to reduce the viewing angle by increasing the distance between camera and measurement section, Fig. 4.16 (b).

4.2 Optical Measurement Techniques



Figure 4.16: Schematic of OH* luminescence imaging setups used for detonation (a, top) and deflagration (b, bottom) experiments. Top view.

For further reducing the viewing angle for detonation investigations and thus gaining a parallel perspective of the detonation front within the entire FOV, one parabolic mirror from the shadowgraphy setup combined with one planar mirror was used, Fig. 4.16 (a). By placing a 5 mm aperture into the focal point of the parabolic mirror, non-parallel light from the measurement section is blocked. Similar to the unfocused shadowgraphy system, cp. Sec. 4.2.1, this OH* luminescence imaging system yields a very small camera aperture angle. In this setup, the Photron SA-X camera was combined with the external Hamamatsu image intensifier. Compared to the Photron APXI² camera, higher image resolution at high frame rates can be obtained.

5 DDT in H₂–Air with Transverse Concentration Gradients

This chapter aims at developing a comprehensive picture of DDT in H_2 -air mixtures with transverse concentration gradients. Similar to Ch. 2, DDT is split into flame acceleration (FA) and onset of detonation. This division, in contrast to treating DDT as one subject, is of vital importance in particular in the context of transverse concentration gradients. The following short summary of major findings is intended to provide orientation and outlines the research strategy.

FA is characterized in Sec. 5.1 by means of optical and conventional measurements. Flame speed and deduced parameters are used to quantify the influence of concentration gradients. The FA process is strongly influenced by concentration gradients, in some cases leading to distinctly stronger FA in gradient mixtures. Two effects are of major importance: influence of integral mixture properties and development of macroscopic flame shape. The latter contributes differently in unobstructed and obstructed channel configurations. These two geometries are compared throughout the entire chapter.

Onset of detonation is analyzed in Sec. 5.2. Shadowgraph sequences and simultaneous pressure measurements allow for detailed description of onset mechanisms. Detonation onset occurs as soon as the preceding FA process creates critical conditions in terms of local overpressure and temperature. Onset mechanisms are similar in homogeneous and gradient mixtures. Simulation of detailed chemical kinetics of strong ignition is used to define critical conditions for onset of detonation.

By finally relating flame speed to overpressure in Sec. 5.3, a comprehensive explanation of DDT in H_2 -air mixtures with transverse concentration gradients is given in Sec. 5.4.

This section is divided into two parts: optical characterization of deflagrations (Sec. 5.1.1) and determination of flame velocities and run-up distances (Sec. 5.1.2).

The height of all shadowgraph, OH-PLIF and OH* images shown within this chapter equals the channel height H = 0.06 m. Only in case of simultaneous measurements, an absolute time scale is used where t = 0 s represents the moment of ignition. Otherwise, for the sake of simplicity, t = 0 s corresponds to the first image of a sequence. Data points (except in v-x-plots) represent the average of five experiments with error bars showing standard deviation. For infrequent cases, less than five valid experiments are available. However, this does not pose a constraint to data interpretation since reproducibility of tests was generally high.

5.1.1 Flame Shape and Structure

Optical observations provide a sound base for understanding effects that emerge in conventional measurement data. About 1500 experiments have been conducted involving the optical measurement techniques introduced in Sec. 4.2. The author reduces this large amount of optical data to the major results in the context of transverse concentration gradients and jointly discusses the two following questions:

- How is the macroscopic flame shape influenced by transverse concentration gradients?
- How do flames in gradient mixtures interact with obstacles?

Mainly the steepest concentration gradients ($t_d = 3 \text{ s}$) that can be generated in the experimantal setup are compared to homogeneous mixtures. First, results from the unobstructed channel configuration BR00 are presented. Second, representative examples are given for obstructed configurations BR30S300

and BR60S300 and compared to the former configuration. To cover both slow and fast deflagration regimes in each configuration, mainly an average H_2 concentration of 20 vol. % is discussed in case of BR00, and 15 vol. % in BR30S300 and BR60S300. Since characteristics of gradient mixtures are of interest, in-depth explanation of more general phenomena that are known from studies on homogeneous mixtures emerging in the images are omitted.

 H_2 concentration profiles across the channel height at $t_d = 3$ s, corresponding reactant density ρ_{re} , reactant sound speed a_{re} , expansion ratio σ , laminar burning velocity S_L and flame speed $S_L\sigma$ are given in Fig. 5.1. Observations on flame shapes will be linked to these parameters in the following.

In the **unobstructed channel**, flames in gradient mixtures are observed to elongate over propagation distance. Figure 5.2 gives a first impression of this effect. It depicts flames shortly after ignition in OS1 (FOV centered at x = 0.3m) in 20 vol. % mixtures, $t_d = 60$ s and 3 s. While the homogeneous mixture shows an almost symmetric flame with respect to the channel centerline, the flame front is inclined in the gradient mixture. The flame does not propagate into mixture below a certain local H₂ concentration at the channel bottom. OH-PLIF images (Fig. 5.3) confirm the observations. The flame front in the homogeneous mixture is not entirely symmetric. Buoyancy may contribute to this shape. However, a distinct difference is visible between homogeneous and inhomogeneous mixture also in the OH-PLIF images. The wavelength of flame front cellularity, discernible both in shadowgraph and OH-PLIF images, varies along the gradient flame from large cells at the top to smaller cells at the bottom, which is in good accordance with the expected change in local Markstein length along the concentration gradient profile. Figure 5.4 further investigates the lower flammability limit in gradient mixtures ($t_d = 3$ s). Shadowgraph images show rear parts of flames where the lower flame boundary is horizontal and does not propagate downwards significantly anymore. At H_2 concentrations of 12.5, 15 and 20 vol. % combustion is incomplete and the lower flammability limit can be estimated at 6–8 vol. %, which is inbetween the limits for horizontal and downward flame propagation as given in Tab. 2.1. Please note that these values only portray estimates. Measurement of local H_2 concentrations would be mandatory for precise statements.



Figure 5.1: Simulated concentration gradient profiles and derived parameters corresponding to experiments presented in Sec. 5.1.1 with $t_d = 3$ s. Flammability limits not considered.



Figure 5.2: Shadowgraph images, 20 vol. %, OS1 (FOV centered at x = 0.3 m), BR00. Red dashed line represents FOV of OH-PLIF images (Fig. 5.3).



Figure 5.3: OH-PLIF images, 20 vol. %, OS1, BR00.



Figure 5.4: Rear parts of flames, $t_d = 3$ s, OS1 (FOV centered at x = 0.3 m), BR00. Variation of X_{H2}. Red dashed line represents approximated lower bound of flammable region.

Figure 5.5 compares flames in OS3 (FOV centered at x = 2.1 m) at $t_d = 60$ s and 3 s. Again, the flame in the homogeneous mixture is nearly symmetric, which emphasizes the negligible influence of top plate injection manifolds on flame propagation. The flame in the $t_d = 3$ s mixture elongates progressively between OS1 and OS3. Elongation causes an increase in overall flame surface area and thus a strong incerease in overall reaction rate. As will be shown in Sec. 5.1.2, this allows flames in gradient mixtures in BR00 to accelerate significantly faster compared to homogeneous mixtures. Comparable surface area enlargement in homogeneous mixtures can typically only be caused by obstacles as discussed in Sec. 2.5.3.

Figure 5.6 compares flame tip geometries in OS3 for different gradient slopes (diffusion times t_d) at 20 vol. % H₂. As can be expected, the flame shape is a strong function of gradient slope. The steeper the gradient, the more flames elongate.

In Fig. 5.7, the gradient slope is kept similar (within the experimental limitations by keeping t_d constant), and H_2 concentration is varied from 12.5 to 30 vol. %. At 12.5 vol. %, the flame is roughly symmetrical within the flammable

region. In such very lean mixtures, flames do not accelerate monotonously, but oscillate back and forth. Flow ahead of the flame comes to rest periodically and even reverses, which may inhibit flame elongation. Already in OS3, reflection of acoustic waves at the channel back wall and their interaction with the flame can mitigate flame elongation. The flame shape in the 12.5 vol. % mixture can thus be considered to be dominated by the experiment geometry. Between 15 and 25 vol. %, flame shapes are very similar. This can be explained by considering profiles of calculated flame speed $S_L \sigma$ (Fig. 5.1). In this concentration range, the maximum local calculated flame speed $S_{\rm L}\sigma$ is located at the channel top, so that flame propagation is expected to be particularly promoted there. In the 30 vol. % mixture the flame tip is broader. This is in good agreement with the relocation of maximum calculated flame speed towards the channel center at this concentration. The leading tip propagates closer to the top wall than suggested by the flame speed profile, which indicates that flame-boundary layer interaction is important. A frequent side-observation is Kelvin-Helmholtz instability along the lower flame boundary due to a vertical gradient in axial reactant flow velocity, shown in Fig. 5.8. Comparing influences of gradient slope and average H₂ concentration on macroscopic flame shape, flame images suggest that the former is more influencial than the latter. This hypothesis will be supported by conventional data in Sec. 5.1.2.

Besides flame speed $S_L \sigma$, further parameters plotted in Fig. 5.1 may influence flame elongation. First, reactant density ρ_{re} is considerably lower in regions of high local H₂ concentration. This implies that mixture ahead of the flame in these regions can be accelerated more readily which leads to enhanced flame elongation [49]. Second, an increase in local reactant sound speed with increasing H₂ concentration means that the flame regime may for example resemble a slow deflagration at the channel top, while it already exhibits features of a fast deflagration at the channel bottom at a given flame speed. This effect can be observed in Fig. 5.9, t_d = 3 s. At t = 0 μ s, curved shocks and their reflections appear ahead of the flame at the channel bottom. Towards the channel top, these shocks disappear due to lower local shock Mach number.

In OS5 (FOV centered at x = 3.9 m), Fig. 5.9, flame elongation seems less distinct than at position OS3. OH-PLIF images of flame tips, Fig. 5.10, support

this observation. Note that the flame image for $t_d = 3$ s is an OH* luminescence image, since OH-PLIF delivered an insufficient SNR in this case (cp. reasons given in Sec. 4.2.2). Flame tips appear broader than in OS3, especially at higher diffusion times. This suggests again that interaction of flames with pressure waves reflected at the channel end plate reduces the tendency of flame elongation.

In advance of writing this thesis, the option of developing an analytical model to predict flame elongation based on parameters of the mixture field has been discussed. After careful deliberation, the author refrains from pursuing this idea, since the complexity of the transient elongation process requires further research. Influences of the specific experimental setup need to be taken into account. An adequate experimental approach should furthermore be supported by numerical simulation.

In **obstructed configurations**, flame elongation is mitigated by obstacles. Figure 5.11 shows flame passage through an obstacle opening in BR30S300, OS2, in a homogeneous 15 vol. % mixture. OH-PLIF images upstream and downstream of the obstacle (green and red FOV), taken in two separate experiments, complement the shadowgraph images, Fig. 5.12. Upstream of the obstacle the flame is symmetric. Only behind the obstacle, weak flame asymmetry is observed in the shadowgraph sequence. This might be caused by the H₂ injection slits in the upper obstacle. OH-PLIF images upstream of the obstacle show the formation of a notch in the flame tip, which is typical for H₂ flames [67]. In comparison, the flame in a transverse gradient mixture (t_d = 3 s), Figs. 5.13 and 5.14, is inclined upstream of the obstacle. OH-PLIF images show that the highly fragmented flame tip passes the obstacle in the upper half of the opening. However, the flame orients towards the channel bottom behind the obstacle, so that a nearly symmetric flame front can be observed in the last shadowgraph image, t = 500 μ s.

As can be seen in Figs. 5.15, 5.16 and 5.17, flame elongation is not discernible in OS3 anymore. OH-PLIF images are taken upstream of the last obstacle of BR30S300, showing nearly planar flame fronts both in the homogeneous and the gradient mixture. Flame pathways are mainly determined by flow streamlines, which are very similar for homogeneous and inhomogeneous mixtures in the vicinity of obstacles. In summary, already a low blockage ratio of 30 % at a wide obstacle spacing of 300 mm suppresses flame elongation considerably compared to the unobstructed configuration.

Likewise, a higher blockage ratio of 60 % in configuration BR60S300 inhibits flame elongation entirely. This can be seen comparing Figs. 5.18 and 5.19, homogeneous and gradient mixture at 15 vol. % in OS2. Flame shapes are similar in the homogeneous and the gradient mixture already at this early position in the channel. This similarity of flame shapes will allow for an isolated evaluation of mixture field influence on FA in this configuration, presented in Sec. 5.1.2. In contrast, FA in the unobstructed channel (BR00) is influenced both by mixture properties and macroscopic flame geometry. BR30S300 can be understood as an intermediate configuration, rather resembling BR60S300 than BR00 regarding flame shape evolution.



Figure 5.5: Shadowgraph images, 20 vol. %, OS3 (FOV centered at x = 2.1 m), BR00. Red dashed line represents FOV of OH-PLIF images (Figs. 5.6, 5.7, 5.8).



Figure 5.6: OH-PLIF images, 20 vol. %, OS3, BR00. Variation of t_d .



Figure 5.7: OH-PLIF images, $t_d = 3$ s, OS3, BR00. Variation of X_{H2} .



Figure 5.8: OH-PLIF images, 30 vol. %, t_d = 3 s, showing Kelvin-Helmholtz instability.



Figure 5.9: Shadowgraph images, 20 vol. %, OS5 (FOV centered at x = 3.9 m), BR00. Red dashed line represents FOV of OH-PLIF images (Fig. 5.10).



Figure 5.10: OH-PLIF images, 20 vol. %, OS5, BR00. Variation of t_d .



Figure 5.11: Shadowgraph images, 15 vol. %, $t_d = 60$ s, OS2 (FOV centered at x = 1.2 m), BR30S300. Red and green dashed lines represent FOVs of OH-PLIF images (Fig. 5.12).



Figure 5.12: OH-PLIF images, 15 vol. %, $t_d = 60$ s, OS2, BR30S300. Upstream (left) and downstream (right) of obstacle.



Figure 5.13: Shadowgraph images, 15 vol. %, $t_d = 3$ s, OS2 (FOV centered at x = 1.2 m), BR30S300. Red and green dashed lines represent FOVs of OH-PLIF images (Fig. 5.14).



Figure 5.14: OH-PLIF images, 15 vol. %, $t_d = 3$ s, OS2, BR30S300. Upstream (left) and downstream (right) of obstacle.



Figure 5.15: Shadowgraph images, 15 vol. %, t_d = 60 s, OS3 (FOV centered at x = 2.1 m), BR30S300. Green dashed line represents FOV of OH-PLIF images (Fig. 5.17).



Figure 5.16: Shadowgraph images, 15 vol. %, t_d = 3 s, OS3 (FOV centered at x = 2.1 m), BR30S300. Green dashed line represents FOV of OH-PLIF images (Fig. 5.17).



Figure 5.17: OH-PLIF images, 15 vol. %, OS3, BR30S300. FOV upstream of obstacle.



Figure 5.18: Shadowgraph images, 15 vol. %, $t_d = 60$ s, OS2 (FOV centered at x = 1.2 m), BR60S300.



Figure 5.19: Shadowgraph images, 15 vol. %, $t_d = 3$ s, OS2 (FOV centered at x = 1.2 m), BR60S300.

5.1.2 Flame Speed and Run-Up Distances

After the presentation of optical observations revealing characteristic differences between unobstructed and obstructed channel configurations, this section employs data from time-of-arrival photodiodes to quantify differences in FA between homogeneous and gradient mixtures. The **unobstructed channel** (BR00) is examined first. Flame speeds in gradient mixtures are compared to their homogeneous counterparts at equal average H₂ concentration. Figure 5.20 (a) shows flame speed profiles at 22.5 vol. % average H₂ concentration to give a first impression of gradient's effects in BR00.

Evidently, gradients cause stronger FA than the homogeneous mixture ($t_d = 60$ s). Note the significant difference between the homogeneous mixture and a steep gradient mixture ($t_d = 3$ s): The former shows slow flame propagation without any sign of significant FA, whereas the latter allows for FA up to choked conditions and onset of detonation at around x = 4 m.

Viewing Fig. 5.20 (a), which includes all possible regimes of flame propagation, it becomes clear that specific parameters need to be defined that allow for separate characterization of different phases of FA and ultimately DDT. Besides the broad application of optical techniques, this is one of the important steps that is taken to cope with the complexity of DDT in gradient mixtures. Two parameters will be evaluated: First, flame speed at a given position in the channel. This position is chosen as x = 2.05 m, which represents the end of the obstacle section in obstructed configurations BR60S300 and BR30S300. Comparability between unobstructed and obstructed configurations is thereby provided. The result for BR00 is presented in Fig. 5.20 (b). In accordance with Fig. 5.20 (a), flames accelerate to higher speed at x = 2.05 m if a concentration gradient is present. The maximum local flame speed in homogeneous mixtures is reached at about 35 vol. %, whereas this maximum is clearly shifted towards higher H₂ concentrations in gradient mixtures. Second, run-up distances (RUDs) to specific flame speed values are determined. Low RUDs relate to strong FA. RUD_{re} is defined as the distance x from the point of ignition, where the flame tip reaches a velocity equal to the speed of sound of the reactants at initial conditions are. For a homogeneous mix-


+ t_d = 60 s, × t_d = 10 s, □ t_d = 7.5 s, $\diamond t_d$ = 5 s, $\star t_d$ = 3 s

Figure 5.20: Flame velocity along the channel at 22.5 vol. % and varying t_d (a); local flame speed at x = 2.05 m at varying X_{H2} and t_d (b). BR00.



+ t_d = 60 s, × t_d = 10 s, □ t_d = 7.5 s, $\diamond t_d$ = 5 s, $★ t_d$ = 3 s

Figure 5.21: Run-up distances to a_{re} (a) and 0.95 a_{pr} (b). BR00.

ture, this marks the velocity region where transition to the fast flame regime occurs. For gradient mixtures, a_{re} is calculated using the average H_2 concentration, thus being equal to the flame speed threshold value for homogeneous mixtures. RUD_{re} in summary characterizes early FA. RUD_{pr} is defined as the distance from the ignition source, where the flame tip reaches a velocity equal to 95 % of the speed of sound of the reaction products a_{pr} , which is calculated assuming adiabatic isobaric complete combustion. For the moment, RUD_{pr} can be interpreted as a first indicator for the potential of flames to reach critical conditions for onset of detonation. The prefactor of 95 % is chosen since flame speed saturation at around a_{pr} , introduced as the third characteristic phase of FA in Sec. 2.5, leads to large scatter in RUD_{pr} if the velocity threshold is set directly to a_{pr} .

Figure 5.21 shows that FA in BR00 is always enforced when a transverse concentration gradient is present. Since both RUD_{re} and RUD_{pr} support this observation, FA is stronger in gradients in all phases of the FA process. The minimum RUD_{pr} that can be achieved in a mixture at $t_d = 3$ s (at about 40 vol. %) is 32 % shorter than in homogeneous mixtures (at about 35 vol. %). Note that this enforcement of FA through concentration gradients in terms of flame speed is not directly transferrable to DDT propensity. This will be discussed in detail in the following Secs. 5.2–5.4. However, evaluation of RUDs as parameters linked to flame speed portrays an important first step for ultimately understanding DDT.

FA in **obstructed channel configurations** follows different trends. Plots of local flame speed at x = 2.05 m for BR60S300 and BR30S300 are shown in Fig. 5.22 (a) and (b), respectively. Since DDT occurs in both configurations at average H₂ concentrations higher than about 22.5 vol. %, all data beyond this concentration reveals rather characteristics of onset of detonation and detonation propagation than of FA. Analysis of RUDs is again highly beneficial to gain insight into the FA phase. This data can then be compared to results from BR00, Fig. 5.21.

RUDs for BR60S300 are shown in Fig. 5.23. At average H_2 concentrations lower than about 22.5–25 vol. %, concentration gradients enforce FA. RUD_{re} (a) shows the same trend as RUD_{pr} (b). Above an average H_2 concentration of



+ t_d = 60 s, × t_d = 10 s, □ t_d = 7.5 s, $\diamond t_d$ = 5 s, $\star t_d$ = 3 s

Figure 5.22: Local flame speed at x = 2.05 m. BR60S300 (a); BR30S300 (b).

22.5–25 vol. %, gradients lead to higher RUDs. FA is clearly retarded by gradients in this region beyond this average H_2 concentration, hereinafter termed flame speed cross-over concentration. It is marked in Fig. 5.23 by a red arrow.

A second obstacle configuration (BR30S300) was investigated in order to check the universality of findings for obstructed channels. RUDs are shown in Fig. 5.24. In general, FA is less effective at such low blockage ratio compared to BR60S300. However, the same effect of average H_2 concentration combined with concentration gradients on RUDs can be seen. The flame speed cross-over concentration is located at around 25 vol. %, very similar to the result in BR60S300

The major difference in RUDs between unobstructed and obstructed configurations is caused by a different development of macroscopic flame shape. As it was shown in Sec. 5.1.1, flames in gradient mixtures only elongate significantly in BR00. Obstacles in contrast hinder the elongation process. Thus, relevant flame surface area enlargement causing a strong increase in overall reaction rate and thereby enforcing FA, only takes place in BR00. In BR60S300



+ t_d = 60 s, × t_d = 10 s, □ t_d = 7.5 s, $\diamond t_d$ = 5 s, $\star t_d$ = 3 s





+ t_d = 60 s, × t_d = 10 s, $\Box t_d$ = 7.5 s, $\diamond t_d$ = 5 s, $\star t_d$ = 3 s

Figure 5.24: Run-up distances to a_{re} (a) and 0.95 a_{pr} (b). BR30S300.

and BR30S300, differences in FA are mainly caused by mixture properties.

The following theoretical approach primarily aims at explaining the occurence of a flame speed cross-over concentration in obstructed configurations, based on the analysis of mixture properties. It will be shown that simple analytical considerations can reproduce differences in RUDs between homogeneous and gradient mixtures at a surprisingly high accuracy. However, please keep in mind that this approach is not yet sufficient to describe the entire process of DDT but only focuses on FA as the first phase of DDT.

A central mixture property, the expansion ratio $\sigma = \rho_{re}/\rho_{pr}$, is examined first. The expansion ratio can be employed to describe FA and is especially useful to predict potential for strong FA [32]. It can be calculated for a homogeneous H₂-air mixture as shown in Fig. 5.25 (a), diffusion time t_d = 60 s. Adiabatic isobaric complete combustion is assumed as a simplification for the deflagration regime to obtain the burnt state and thus ρ_{pr} .

An integral approach is suggested here to account for the influence of transverse concentration gradients. The effective expansion ratio $\sigma_{\rm eff}$ is defined as the average across the channel height H for a given H₂–air distribution, thus

$$\sigma_{\rm eff} = \frac{1}{\rm H} \int_0^{\rm H} \sigma(y) dy, \qquad (5.1)$$

where $\sigma(y)$ is the local expansion ratio calculated for the corresponding local mixture composition at a vertical position y. Concentration profiles from numerical simulations of the injection process in the facility by Ettner [42] as described in Sec. 3.2 are used for these calculations.

In Fig. 5.25 (a), the effective expansion ratio σ_{eff} as a function of average H₂ concentration is shown for different gradients labeled by corresponding diffusion times t_d. It remains below the values for homogeneous mixtures at all average concentrations due to the non-linear dependency between expansion ratio and H₂ concentration. As long as nearly point-symmetric concentration gradients are examined, this trend is principally not dependent on the gradient shape and would be qualitatively similar for linear concentration gradients, for example.

As a second basic parameter for FA, the laminar burning velocity S_L , is examined analogously by defining an effective laminar burning velocity $S_{L,eff}$. It is written as

$$S_{L,eff} = \frac{1}{H} \int_0^H S_L(y) dy.$$
 (5.2)

 $S_{L,eff}$ is plotted in Fig. 5.25 (b). The polynomial given by Eq. (2.29) yields the curve for the homogeneous mixture, $t_d = 60$ s. Evaluation of gradient profiles using Eq. (5.2) shows that the effective burning velocity directly reproduces the flame speed cross-over point, marked by a red arrow. Only in the region of average H₂ concentrations lower than about 24 vol. %, gradients lead to higher effective burning velocities. Beyond this concentration, effective burning velocity is lower in gradient mixtures than in homogeneous mixtures. The cross-over concentration observed experimentally is reproduced accurately. Note that the shape or the slope of the concentration gradients does not influence the flame speed cross-over concentration, as long as nearly point-symmetric profiles are considered. All gradient profiles investigated show a common intersection point.

Two important conclusions can be drawn at this stage:

- The integral approach is verified by the accurate reproduction of flame speed cross-over concentration. In comparison, consideration of maximum H₂ concentration only as suggested by Kuznetsov et al. [85] and Grune et al. [56] cannot reproduce this very basic effect of concentration gradients in an entirely closed tube. The utilization of numerically determined concentration profiles does not influence this conclusion. It was possible to gain this insight by separating the influence of mixture from the influence of flame elongation by comparing unobstructed and obstructed configurations.
- The relevance of laminar burning velocity for accurate description of FA in inhomogeneous mixtures is shown. Expansion ratio alone is evidently insufficient to account for mixture inhomogeneity in H₂-air.



Figure 5.25: Effective expansion ratio σ_{eff} (a); effective laminar burning velocity S_{L,eff} (b); effective flame speed (S_L σ)_{eff} (c).

The last step of this analysis is the combination of expansion ratio σ and laminar burning velocity S_L. This is motivated by the 1-D balance of mass across a flame front, given in Eq. (2.27), which yields flame speed S_L σ . The parameter can again be treated as an effective property according to Eq. (5.3), plotted in Fig. 5.25 (c).

$$(S_{\rm L}\sigma)_{\rm eff} = \frac{1}{\rm H} \int_0^{\rm H} [S_{\rm L}(y) \cdot \sigma(y)] dy$$
 (5.3)

The following summary contains assumptions inherent to the presented approach:

- The combustion behavior of the entire mixture is taken into account in the integral approach. Understanding the FA process as a self-enforcing gasdynamic and fluiddynamic feedback cycle justifies this approach since reaction of the entire flammable mixture is the driver for FA in a closed channel. Consequently, it must be the overall reaction rate in the channel that governs the process, in contrast to an exclusive consideration of distinct regions within gradient profiles.
- Alteration of burning velocity due to turbulence, shock-flame interactions and preconditioning of the mixture by shocks is assumed to contribute in a similar manner in homogeneous and gradient mixtures. A relation between laminar and turbulent burning velocity as well as temperature and pressure dependence is not included. Observations like Kelvin-Helmholtz instability at the lower flame boundary are not taken into account. The concept of comparing RUD_{pr} is highly beneficial here since flame propagation modes are similar at RUD_{pr} in different types of mixtures.
- Macroscopic convective mixing in the flame-induced reactant flow is neglected. Thus, the flame is assumed to propagate in the initial, undisturbed gradient field at any time. Ettner [42] provides a numerical study on mixing ahead of the flame front for the channel configurations investigated in the present work. He concludes that gas is mainly mixed within pockets in the wake of obstacles. The difference between maximum and minimum local concentrations remains fairly unaltered. Mixing is not significant in BR00 and is more intense in BR30S300 than in BR60S300.

5.1 Flame Acceleration

- Unreacted portions of H₂ behind the flame front in fuel-rich regions are assumed not to react with excess oxygen from fuel-lean regions. This seems reasonable since vertical transport of reactants behind the flame front can be considered much slower than the flame speed. Especially for fast combustion regimes the time available for mixing is very short. The concept therefore inherently portrays the effect of incomplete combustion due to concentration gradients.
- As a first conservative approach, mixture is considered inert below the lower flammability limit for upward flame propagation. This could be adapted to observations from Sec. 5.1.1, which suggest a lower flammability limit between the limit for horizontal and downward propagation. The choice of flammability limit does not decisively alter effective properties for rather high average H₂ concentrations relevant for the present work. In very lean mixtures, this point requires reconsideration.
- Perfect molecular mixing is assumed at any position across the channel height. Validity of this assumption is difficult to prove. However, good quantitative agreement between model and experiment as shown subsequently supports this assumption.

The relative difference in effective flame speed between homogeneous and inhomogeneous mixtures can be directly compared to measured RUD_{pr} . This will emphasize the quantitative validity of the analytical approach in addition to the correct prediction of flame speed cross-over concentration. Data from BR60S300 is chosen for this comparison since flame elongation and thus macroscopic flame surface area enlargement was found to be negligible (Sec. 5.1.1). Two dimensionless parameters, M (Eq. (5.4)) and E (Eq. (5.5)), are defined representing results of theoretical model and experiments, respectively.

$$M = \frac{1/(S_L\sigma)_{eff,grad} - 1/(S_L\sigma)_{hom}}{1/(S_L\sigma)_{hom}}$$
(5.4)

$$E = \frac{RUD_{pr,grad} - RUD_{pr,hom}}{RUD_{pr,hom}}$$
(5.5)



 \times t_d= 10 s, \Box t_d= 7.5 s, \diamond t_d= 5 s, \star t_d= 3 s

Figure 5.26: Comparison of dimensionless experimental run-up distance E and calculated effective flame speed M. BR60S300 (a); BR00 (b).

The result is shown in Fig. 5.26 (a), where experiments are plotted on the abscissa and model results on the ordinate. Experimental error bars as shown in previous RUD plots are omitted here since the model cannot account for statistical scatter in RUDs. Despite the uncertainty regarding concentration profiles taken from numerical simulations and neglection of mixing ahead of the flame, the model provides an accurate prediction of relative differences in RUD_{pr} between homogeneous and inhomogeneous mixtures. It directly reproduces the fact that concentration gradients can cause stronger or weaker FA depending on the average H₂ concentration. For most average H₂ concentrations and gradients, the model character is conservative, meaning that the model predicts lower RUD_{pr} than experimentally determined. Single data points show a non-conservative behavior with a maximum prediction error of 11.5 %. However, this accuracy is remarkably high considering the simplicity of the model without any calibration constants. The key finding from this analysis is again, that the presented integral approach and the consideration of effective flame speed represents a valid way to characterize the potential for

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Diffusion time t _d	10 s	7.5 s	5 s	3 s
Model constant C	1.25	1.45	1.80	2.57

Table 5.1: Model constants C for RUD_{pr} in BR00.

FA in H₂-air mixtures with concentration gradients.

Extension of the approach for the unobstructed channel (BR00) is possible if flame surface area enlargement is taken into account. This is accomplished by an experimentally determined model constant $C = f(t_d)$ for each diffusion time, yielding the parameter M_C ,

$$M_{\rm C} = \frac{1/[C \cdot (S_{\rm L}\sigma)_{\rm eff,grad}] - 1/(S_{\rm L}\sigma)_{\rm hom}}{1/(S_{\rm L}\sigma)_{\rm hom}}.$$
(5.6)

Comparison of model and experiments is plotted in Fig. 5.26 (b). Again, very good agreement with an accuracy better than 8.2 % difference is obtained. Model constants C determined by the least squares method for the investigated concentration gradients are given in Tab. 5.1, reproducing the effect of flame surface area enlargement well. Since no clear dependency between C and the average H_2 concentration could be found, C is chosen as a function of diffusion time only. This supports the hypothesis stated in Sec. 5.1.1, that flame shape is primarily a function of gradient slope.

5.2 Onset of Detonation

Following the analysis of FA, this section investigates onset of detonation in both unobstructed and obstructed channel configurations. The importance of separating this discussion from FA considerations will become clear. First, onset of detonation in the unobstructed configuration BR00 is discussed in Sec. 5.2.1. Subsequently, results for obstructed configurations BR30S300 and BR60S300 are presented in Sec. 5.2.2. In order to understand the physics of onset of detonation it will be necessary to look at the problem from different perspectives and apply different methods. Simulation of detailed chemical kinetics at the extended second explosion limit is eployed to describe the onset from a chemical kinetics point of view. This approach is presented in Sec. 5.2.3. Concluding explanations of experimental results are shifted to Sec. 5.4 since a more comprehensive view involving results on FA from Sec. 5.1 and measurements of overpressure related to flame speed, presented in Sec. 5.3, is required.

5.2.1 Unobstructed Channel

In the unobstructed channel configuration (BR00), onset of detonation is observed at the channel walls in the vicinity of the turbulent flame brush. High average H_2 concentrations in conjunction with a concentration gradient are required to provoque DDT in BR00. Only few experiments of this kind involving the optical segment and shadowgraphy were performed due to safety reasons. The side windows of the facility can easily be damaged, resulting in hairline cracks intruding the windows within a depth of about 10 mm, after only one DDT event in the window vicinity.

Figure 5.27 shows a shadowgraph sequence of onset of detonation in OS5 (FOV centered at x = 3.9 m) in a 35 vol. % mixture at $t_d = 7.5$ s complemented by simultaneous pressure traces at transducers p_4 (a) at x = 3.2 m, p_5 (b) at x = 3.9 m and p_6 (c) at x = 4.7 m. Transducer p_5 is located in the FOV, marked by blue triangles. By choosing this particular H_2 concentration and concentration gradient, onset of detonation could be observed within the FOV.

The mechanism of onset of detonation is well comparable to observations by Urtiev and Oppenheim [148]. The flame enters the FOV with the leading flame tip at the channel top at a flame tip velocity of $v \approx 1200$ m/s. This equals a local flame Mach number of 2.7 at the channel top. The fresh mixture ahead of the flame is compressed up to $p \approx 11.5$ bar overpressure (Fig. 5.27, (1)). At t = 18.3 ms, a local explosion emerges at the upper channel wall (2). Note that the location of explosion origin does not coincide with any H₂ injection manifold, thus onset of detonation is not caused by shock reflection off a manifold. The generated blast wave forms a forward-propagating detonation wave and a backward-propagating so-called retonation wave. These waves can be further



Figure 5.27: Shadowgraph sequence and pressure traces of onset of detonation, 35 vol. %, t_d = 7.5 s, OS5 (FOV centered at x = 3.9 m, x_{p_4} = 3.2 m, x_{p_5} = 3.9 m, x_{p_6} = 4.7 m), BR00.



Figure 5.28: Probability of DDT in BR00 as a function of average (a) and maximum (b) H₂ concentration.

tracked in the pressure records. The blast wave is reflected off the channel bottom wall, t = 18.317 ms. Point (3) in the pressure record of p_5 marks the arrival of the reflected wave at the top plate. Two pressure peaks appear. The second peak may either originate from a secondary local explosion triggered by the primary one, or by three-dimensional effects. The frame at t = 18.333 ms indeed shows two waves moving upwards towards the pressure transducer. Point (4) marks the arrival of the forward-propagating detonation wave at p_6 , while point (5) depicts the arrival of the scattered retonation wave at p_4 . Finally, the detonation is reflected off the channel end plate at x = 5.1 m and propagates backwards through burnt mixture (points (6) and (7), not visible in Fig. 5.27 (b) due to higher temporal resolution).

Besides optical investigation, DDT has been studied by means of conventional measurement techniques in configuration BR00. The particular goal was to determine conditions in terms of average H_2 concentration and concentration gradient that allow for DDT. DDT can be discerned by manually analysing pressure traces. Local explosion followed by a typical detonation pressure profile at the next downstream pressure transducer serves as a criterion. Local ex-

plosion at the channel end plate was not considered a relevant DDT event. The result is shown in Fig. 5.28 (a). Each pair of average H_2 concentration and diffusion time t_d was repeated five times. Shares of experiments with DDT are depicted as pie charts. DDT in homogeneous mixtures only occurs infrequently at 35–40 vol. % H₂. The channel length is insufficient to allow for DDT in leaner homogeneous mixtures. As shown in Sec. 5.1.2, FA is most pronounced in homogeneous mixtures around 35-40 vol. % which consistently leads to DDT in this concentration range. Gradients lead to earlier DDT in terms of average H₂ concentration compared to homogeneous mixtures. This is in accordance with the trend in FA, which is enforced by gradients through flame elongation and additionally supported by increased effective flame speed $(S_L \sigma)_{eff}$ in mixtures below 24 vol. % H₂. Mixtures at $t_d = 3$ s allow for DDT already in a 20 vol. % mixture, which is remarkable keeping in mind that the channel is unobstructed. However, the results also show that an effect counteracting the FA enforcement by gradients must exist, since DDT is evidently suppressed in mixtures with steep gradients ($t_d = 3$ and 5 s) at high H₂ concentrations. This poses an upper DDT limit for gradient mixtures in the channel investigated here. The effect can be seen most clearly at $t_d = 3$ s where no DDT occurs at average H₂ concentrations of 30 vol. % and higher. The reason for the deviation between DDT trends and FA behavior will be given in Sec. 5.4 since further considerations presented in the following sections are required to give a concluding explanation. At $t_d = 7.5$ s, the range of H_2 concentrations with DDT is remarkably wide. The observed effect that suppresses DDT in rich mixtures with steeper gradients can apparently be overcome by strong FA here.

From a practical perspective, the lower DDT limit is more relevant than the upper limit if, like in nuclear reactor accident scenarios, low H_2 concentrations prevail. Lower DDT limits in homogeneous and inhomogeneous mixtures can be approximated by using the maximum local H_2 concentration at the channel top, cp. Fig. 5.28 (b), as suggested by Kuznetsov et al. [85] and Grune et al. [56]. Results for different gradients coincide within a band of maximum H_2 concentration at the channel top (grey band in Fig. 5.28 (b)). However, the present work shows that such an approach does not reflect the physics of DDT in mixtures with transverse concentration gradients and thus represents an empirical criterion only.

5.2.2 Obstructed Channel Configurations

Figure 5.29 shows onset of detonation in BR30S300L, OS5 (FOV centered at x = 3.9 m), in a homogeneous 16.5 vol % mixture. A shock at a velocity of 1000 m/s propagates towards the BR30 obstacle, already causing auto-ignition in the wall boundary layers. This is a first indication for conditions very close to possible onset of detonation. The shock is reflected off the obstacle at t = 61.438 ms. Primary local explosions can be observed both at the upper and lower obstacle upstream surfaces, t = 61.450 ms. The image at t = 61.463ms shows collision of the explosion fronts at the channel center line. Subsequent images do not allow for clear tracking of the fronts during diffraction around the obstacle. Eventually, the detonation front emerges clearly at t = 61.525 ms at the channel bottom. Detonation is thus initiated by the formation of a secondary hot spot at the lower channel wall. Asymmetry (initiation only at the bottom wall) may be supported by the high sensitivity of chemical reaction rates on temperature in the relevant high temperature and pressure range. This observed mechanism stands in contrast to recent results by Kellenberger and Ciccarelli [74] who observed detonation initiation by center line collision of the diffracting primary explosion fronts in the obstacle opening. In that case, the leading point of the detonation front downstream of the obstacle is located at the channel center line. Presumably, the location of final detonation initiation depends on the obstacle spacing and size. If a stronger shock than observed in the present work incides on the obstacle, detonation may be initiated at a secondary hot spot at the center line. This suggests that the mechanism of onset of detonation with secondary hot spots at the walls can occur already at lower incident shock Mach number compared to the mechanism involving center line secondary hot spot explosion. At an even higher incident shock Mach number, the primary hot spot may already be sufficient to initiate a detonation that can successfully diffract around the obstacle. Different geometries are also discussed by Gamemzo et al. [51]. Variation of geometry is however not topic of the present work.

Pressure traces of p_4 , p_5 (blue triangle in the FOV) and p_6 suggest that the shock-flame complex arrives at the obstacle in the FOV at a Mach number that may be already beyond the necessary Mach number to cause onset of



Figure 5.29: Shadowgraph sequence and pressure traces of onset of detonation, 16.5 vol. %, t_d = 60 s, OS5 (FOV centered at x = 3.9 m, x_{p_4} = 3.2 m, x_{p_5} = 3.9 m, x_{p_6} = 4.7 m), BR30S300L.

detonation by shock reflection. Already at p_4 , a high pressure level of about 10 bar and three distinct pressure spikes can be observed. This suggests that local explosions already occur at this position, but do not successfully lead to onset of detonation. High peak pressure at p_5 corresponds to the location of onset of detonation, while the typical detonation pressure profile with a sharp rise in pressure and subsequent expansion is recorded at p_6 . Peak pressure exceeds CJ pressure here, which may be due to three-dimensional effects or due to an overdriven state at the pressure transducer location.

Summarizing the observations, the onset process begins with strong ignition in post-reflected-shock mixture at the upstream obstacle surfaces. The explosion fronts diffract around the obstacle and eventually transform into a detonation after secondary hot spot generation at the channel walls. This process reflects well the sequence of events during onset of detonation as discussed in Sec. 2.6. Local explosions represent the first step and thus the first crucial requirement for this process. Secondary hot spots generated by reflection at channel walls, finally forming the detonation, may be seen as a consequence of the primary local explosions. The H_2 concentration of 16.5 vol. % is close to the lower detonability limit in a homogeneous mixture at initial ambient conditions in the experimental setup, since the detonation cell width quickly exceeds the channel dimensions at lower concentrations, cp. Fig. 2.27.

As will be shown subsequently, **transverse concentration gradients** did not lead to systematically earlier DDT in terms of average H_2 concentration in obstructed configurations, which stands in evident contrast to the findings for BR00. A diffusion time of $t_d = 3$ s will be compared to homogeneous mixtures.

In gradient mixtures, the onset mechanism is similar to the mechanism in homogeneous mixtures. It begins with strong ignition in post-reflected-shock mixture and proceeds through diffraction of emerging blast waves around the obstacle. Detonation initiation finally occurs at a channel wall. Mixture inhomogeneity influences the preferred location of primary local explosions. To capture onset of detonation in the largest range of H₂ concentration possible—17–35 vol. %—obstacle configurations and the position of the optical segment were varied. Onset events will be discussed beginning with low average H₂ concentrations. Figure 5.30 shows onset of detonation in a 17 vol. % mixture at $t_d = 3$ s in BR30S300L, OS5. Note that the average H₂ concentration is very close to the corresponding homogeneous mixture experiment, Fig. 5.29. Precursor shock speed is 1100 m/s upstream of the obstacle. After precursor shock reflection, local luminescence marks a local explosion at the upper obstacle at t = 27.738 ms. No explosion occurs at the lower obstacle. Final detonation initiation may be caused by merging of two shocks in this experiment, which can be seen in the last three images of this sequence. The last frame shows the forward propagating detonation front. The pressure trace of p_5 (blue triangle in the FOV) shows the sharp pressure peak typical for the location of onset of detonation. P_4 by contrast still shows the typical fast flame profile. The detonation might fail inbetween p_5 and p_6 since p_6 shows a double peak pressure trace, suggesting a very unstable or currently failing detonation.

For discussion of higher H_2 concentrations, selected images from shadowgraph sequences will be used. Images are selected to show the nature of local explosions and the final formation of detonation. Note that the images are not equidistant in time.

In BR30S300, OS3, onset of detonation can be first observed at 22.5 vol. % both in homogeneous and gradient mixtures. Figure 5.31 comprises four images of the process in the $t_d = 3$ s mixture. A strong local explosion, causing clearly visible luminescence, can be discerned at the upper obstacle at $t = 50 \ \mu$ s. Reflected shock velocity at the lower obstacle remains low. Thus, no local explosion takes place at this position. This is similar to the experiment at 17 vol. %, Fig. 5.30.

To obtain onset of detonation in richer mixtures at the next possible optical segment position OS2, configuration BR60S300 needs to be employed since BR30S300 allows for onset only further downstream due to weaker FA, cp. Sec. 5.1.2. To demonstrate the similarity between BR30S300 and BR60S300, an onset process in BR60S300, OS3, is shown in Fig. 5.32. Average H_2 concentration is equal to the BR30S300 case. The driving local explosion again occurs at the upper obstacle and final detonation initiation is observed at the channel top wall downstream of the obstacle.



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Figure 5.30: Shadowgraph sequence and pressure traces of onset of detonation, 17 vol. %, $t_d = 3$ s, OS5 (FOV centered at x = 3.9 m, $x_{p_4} = 3.2$ m, $x_{p_5} = 3.9$ m, $x_{p_6} = 4.7$ m), BR30S300L.

5.2 Onset of Detonation



Figure 5.31: Shadowgraph sequence of onset of detonation, 22.5 vol. %, t_d = 3 s, OS3 (FOV centered at x = 2.1 m), BR30S300.



Figure 5.32: Shadowgraph sequence of onset of detonation, 22.5 vol. %, t_d = 3 s, OS3 (FOV centered at x = 2.1 m), BR60S300.



Figure 5.33: Shadowgraph sequence of onset of detonation, 26 vol. %, $t_d = 3$ s, OS2 (FOV centered at x = 1.2 m), BR60S300.

Onset of detonation in homogeneous mixtures occurs at 25 vol. % in BR60S300 OS2. In the $t_d = 3$ s mixture, a slightly higher concentration of 26 vol. % is required. A corresponding experiment is presented in Fig. 5.33. Local explosions now form both at the channel top and bottom. The strength of the upper explosion can be estimated higher than the lower counterpart due to a higher propagation velocity. Final detonation initiation still occurs at the channel top downstream of the obstacle. In this series of images, it can be seen that gas is pushed through the upper obstacle H_2 injection slits, forming an oblique shock propagating in the upper wall boundary layer. This effect however does not decisively influence the onset of detonation.

Before showing experiments at 30 and 35 vol. %, expected behavior is derived from the effective flame speed model presented in Sec. 5.1.2. For this purpose, please compare trends of effective flame speed in Fig. 5.25 (c) in homogeneous and $t_d = 3$ s mixtures at H₂ concentrations beyond the flame speed cross-over concentration of 24 vol. %. Flame speed increases with increasing H₂ concentration in the homogeneous mixture, suggesting enforcement of FA. By contrast, the effective flame speed profile for $t_d = 3$ s flattens, effective flame speed does not increase considerably towards higher H₂ concentrations. Consequently, FA and thus also the potential for onset of detonation in the current FOV, OS2, should stagnate for $t_d = 3$ s mixtures.

5.2 Onset of Detonation



Figure 5.34: Shadowgraph sequence of onset of detonation, 30 vol. %, $t_d = 3$ s, OS2 (FOV centered at x = 1.2 m), BR60S300.



Figure 5.35: Shadowgraph sequence of onset of detonation, 35 vol. %, $t_d = 3$ s, OS2 (FOV centered at x = 1.2 m), BR60S300.

Figures 5.34 and 5.35 comprise experiments at 30 and 35 vol. %, $t_d = 3$ s, respectively. Evidently, the state of FA upstream of the obstacle is similar to the 26 vol. % experiment, Fig. 5.33. The best indicator is the similar separation distance between precursor shock and flame. This observation again clearly underscores the validity of the integral approach for FA utilizing effective flame speed to characterize FA. In homogeneous mixtures, only detonations are observed in OS2 at H₂ concentrations beyond 25 vol. %, which is consistent to the prediction based on effective flame speed. Regarding local explosions, strong ignition at the lower obstacle becomes increasingly important in gradient mixtures at such high average H₂ concentrations. At 30 vol. %, Fig. 5.34, the frame at t = 50 μ s suggests similar strength of upper and lower local explosions. At 35 vol. %, Fig. 5.35, the lower local explosion exceeds the upper one in strength, discernible at t = 62.5 μ s where the lower front has already reached the upper channel wall upstream of the obstacle, while the upper front is currently interacting with the top surface of the lower obstacle.

Experimental observations of onset of detonation in obstructed configurations are summarized as follows:

- Onset of detonation is initiated by local explosions (strong ignition, cp. Sec. 2.4) in post-reflected-shock mixture at upstream obstacle surfaces.
- Diffraction of blast waves emerging from local explosions around the obstacle and subsequent reflection of the diffracted waves at the channel walls finally initiates detonation.
- In gradient mixtures, local explosions preferentially occur at the upper obstacle, thus in the most fuel-rich region, which is consistent with the local explosion location in BR00 in gradient mixtures.
- At average H_2 concentrations of 22.5 vol. % and below, no local explosions are observed at the lower obstacle. At 26 vol. % and beyond, where the local H_2 concentration at the lower obstacle exceeds about 10 vol. %, local explosions occur both at upper and lower obstacle. An average H_2 concentration of 30 vol. % shows similar explosion strengths at both locations. At 35 vol. % the lower explosion exceeds the upper one in strength.

No case has been observed within the investigated range of H_2 concentrations, where local explosion only occurs at the lower obstacle.

• Beyond the flame speed cross-over concentration of 24 vol. %, calculated effective flame speed predicts DDT propensity well. On the lean side of cross-over, promotion of DDT by concentration gradients would be expected. This has, however, not been observed. Homogeneous and gradient mixtures at average H₂ concentrations between 17 and 25 vol. % show a similar potential for DDT.

The unexpected DDT behavior below the flame speed cross-over concentration requires further investigation. An understanding of the relation between detonation initiation and parameters such as flame speed and local overpressure generated by FA needs to be established, which will also help to explain DDT trends observed in BR00, Sec. 5.2.1. This motivates the theoretical approach subsequently presented in Sec. 5.2.3, exploring detailed chemical kinetics of shock-induced strong ignition in the vicinity of the extended second explosion limit.

5.2.3 Chemical Kinetics of Shock-Induced Strong Ignition

Onset of detonation by shock reflection off a flat obstacle surface portrays an attractive case to be analyzed by means of post-reflected-shock detailed chemical kinetics simulations. In comparison, onset in an unobstructed channel depends crucially on small-scale phenomena such as interaction of shocks and wall boundary layer and is therefore difficult to describe analytically. Depending on blockage ratio and spacing, shock focusing and reflection of Mach stems may occur in obstructed channels. Due to the large spacing of 0.3 m in configurations discussed in the present work, these effects are not of importance. Nearly normal shocks interact with obstacles here. Thus, reflection of a normal shock off a solid wall is considered in one dimension. Melguizo-Gavilanes and Bauwens [104] have shown that strong ignition behind a reflected shock can be modeled by a constant volume explosion in a homogeneous reactor at a high accuracy, compared to spatially resolved modeling. This is in good accordance with the similarity between ZND and constant volume explosion modeling of detonations outlined by Shepherd [131].

In experiments in the present work, but also in the work of other authors [74, 111, 142, 156], strong ignition behind a reflected shock can be identified as the typical first step of onset of detonation in obstructed channels. Analyzing this first step delivers a necessary, but not entirely sufficient criterion for onset, cp. Sec. 2.6. Propagation of a detonation emerging from a local shockinduced explosion into the confining geometry, which then needs to allow for self-sustained detonation propagation, cp. Sec. 2.7, is the subsequent necessary step for successful onset of detonation. Since the present work focuses on mixture properties in contrast to the influence of geometrical characteristics, analysis of the initial local explosion is meaningful here. This yields a conservative boundary for onset of detonation by shock reflection since the initial and thus crucial step is considered. Geometrical criteria like the well-known empirical 7λ criterion by Dorofeev et al. [31] cover geometrical influences only, but do not consider the actual first requirement in the chain of necessary criteria for onset. Also the criterion expressed in Eq. (2.39) by Thomas [142] is based on geometrical considerations and does not answer the question what the requirement for the initial necessary local explosion is.

The extended second explosion limit can be interpreted as a boundary between mild and strong ignition as shown by Lee and Hochgreb [92], cp. Secs. 2.3 and 2.4. In the following analysis, the limit is determined through detailed chemical kinetics simulations. Discussion is first confined to homogeneous mixtures and eventually transfers insights to mixtures with transverse concentration gradients. The major goal is the deduction of a criterion for critical conditions of a fast deflagration precursor shock, that will cause strong ignition after reflection at a solid wall such as an upstream obstacle surface.

As shown by Shepherd [132], an approach to determine the location of the extended second explosion limit in terms of temperature and pressure is to compute the reduced effective activation energy

$$\theta = \frac{E_a}{RT}$$
(5.7)

with T being the initial mixture temperature, by numerical differentiation:

$$\theta = \frac{1}{T} \frac{\ln(\tau_{\text{ind},+}) - \ln(\tau_{\text{ind},-})}{(1/T_{+}) - (1/T_{-})}.$$
(5.8)

 $\tau_{ind,+}$ and $\tau_{ind,-}$ are induction times computed for temperatures T₊ and T₋, respectively. Temperatures T₊ and T₋ are gained by varying T by a factor of 1±0.01. The T-p plane for θ is depicted in color in Fig. 5.36 for a 30 vol. % mixture. Absolute pressure is used here to be consistent with literature on explosion limits and the classical explosion limits diagram, Fig. 2.3, which covers lower T and p compared to Fig. 5.36 as explained in Sec. 2.4. Note that the axis of ordinates is linear in Fig. 5.36 as opposed to the logarithmic scale in Fig. 2.3. The region of maximum θ corresponds to the extended second explosion limit region [132] (dotted black line marks the line of maximum θ). Induction time is particularly sensitive to variations in temperature around the extended second explosion limit, cp. Eq. (5.8). Note that the location of the extended second explosion limit is almost independent of H₂ concentration as will be shown subsequently in Fig. 5.37.

So far, only a general T-p-plane for θ has been determined. However, in the context of shock-induced auto-ignition, only specific T and p can be caused by an incident shock or a reflected shock. Possible post-shock (state 1) and post-reflected-shock states (state 1_r) are described by the shock equations, Sec. 2.2. They are depicted in Fig. 5.36 for the 30 % vol. mixture as dashed black (incident shock) and dashed red (reflected shock) lines. These lines are obtained by variation of shock Mach number. Similar to the location of the extended second explosion limit, these lines are almost independent of H₂ concentration since H₂ as a diatomic gas has a specific heat capacity ratio $\gamma_{H2} = 1.41$, being very close to $\gamma_{Air} = 1.40$ of air¹⁷.

¹⁷Heat capacity ratio values at standard conditions.



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Figure 5.36: Reduced effective activation energy θ (color plot), post-incident (black dashed line) and post-reflected-shock states (red dashed line), extended second explosion limit (black dotted line). 30 vol. % H₂-air mixture.

The diagram can thus be interpreted in the following way: On the left side of the extended second explosion limit, post-shock conditions behind incident shocks (black dashed line, T_1 and $p_{1,abs}$) and reflected shocks (red dashed line, $T_{1,r}$ and $p_{1,r,abs}$) lead to mild ignition. On the right side of the limit, by contrast, strong ignition occurs. Post-reflected-shock conditions that lie on the right side of the limit thus allow for local explosions at the reflecting wall as observed in the experiments presented in Sec. 5.2.2, identified as the first step of onset of detonation.



Figure 5.37: Reduced effective activation energy θ as a function of post-reflected-shock ($p_{1r,abs}$) and post-incident-shock ($p_{1,abs}$) pressure.

Since the location of the extended second explosion limit as well as possible post-reflected-shock states only differ in a negligible manner over H₂ concentration, the required post-reflected-shock state to cross the extended second explosion limit is very similar within the entire detonable range of H₂ concentrations. Curves for reduced effective activation energy θ as a function of post-reflected-shock pressure p_{1r,abs} can be obtained by varying shock Mach number along the post-reflected-shock line. Figure 5.37 shows the result for mixtures between 15 and 45 vol. % H₂. The secondary axis of abscissas provides pressure p_{1,abs} behind incident shocks that lead to corresponding postreflected-shock pressures p_{1r,abs} after reflection. Pressure is analyzed in this argumentation since local temperature is difficult to determine experimentally, but wall pressure can be measured. An incident shock that causes strong ignition after reflection needs to provide a certain post-incident-shock pressure p_{1.abs} that is almost independent of H₂ concentration. Between H₂ concentrations of 15 and 45 vol. %, the post-incident-shock pressure $p_{1,abs}$, which yields maximum θ after reflection, only varies between 11.5 and 11.8 bar (10.5–10.8

bar overpressure). Even if the extended second explosion limit is interpreted as a band rather than a sharp boundary, critical post-incident-shock pressure lies in a narrow range of 10–11 bar overpressure.

Due to the minor influence of H_2 concentration on gasdynamic relations and chemical kinetics of strong ignition, conclusions can be directly transferred to mixtures with concentration gradients Also here, post-reflected-shock pressure and temperature need to exceed the extended second explosion limit to cause local explosions that can lead to onset of detonation.

In conclusion, the presented analysis of strong post-reflected-shock ignition using detailed chemical kinetics simulations shows that:

- The pressure ratio (or Mach number) of the precursor shock of a fast deflagration needs to exceed a critical value to allow for strong ignition after shock reflection. Post-incident-shock overpressure will be used as a criterion since it is well measurable in experiments and at the same time the crucial parameter from a safety point of view.
- Critical overpressure values are nearly independent of H₂ concentration.
- Critical post-incident-shock overpressure required to cause local explosions (strong ignition) after shock reflection as a first and thus crucial step during onset of detonation ranges between 10 and 11 bar.

Finally, it is interesting to combine the criterion formulated here for strong ignition with the criterion for successful transmission of a local explosion into the macroscopic confining geometry as formulated by Thomas [142], Eq. (2.39). Since this is only a geometrical criterion as explained in Sec. 2.6, a criterion for critical incident shock strength to cause strong ignition is not included. Combination with the kinetics-based criterion found in the present work allows for consideration of both chemical kinetics and obstacle geometry. Relatively independent of H₂ concentration, it can be calculated that induction time at the extended second explosion limit is of the order of $2 \cdot 10^{-6}$ s. At critical post-reflected-shock states, sound speed ranges from 780 m/s (15 vol. %) to 860 m/s (30 vol. %). For these conditions, Eq. (2.39) yields a minimum obstacle height of the order of 1 mm for successful onset of detonation.

This result suggests that even very small obstacles hold potential for causing onset of detonation in H_2 -air at initially ambient conditions.

5.3 Relation Between Flame Speed and Peak Overpressure

Flame speed measurements have been used to characterize FA in mixtures with transverse concentration gradients in Sec. 5.1.2. Simulation of detailed chemical kinetics in post-reflected-shock mixture in Sec. 5.2.3 revealed that post-incident-shock overpressure is a useful and physically meaningful parameter to describe critical conditions for onset of detonation both in homogeneous and gradient mixtures. The present section investigates the relation between flame speed and peak overpressure in mixtures with transverse concentration gradients. This information will then be merged with results on FA and the physics of onset of detonation in Sec. 5.4.

As discussed in Sec. 4.1.2, accurate quantitative explosion overpressure measurement in fast regimes is demanding, in particular if a large number of tests is conducted. Manual validation of pressure traces is mandatory. Thus, investigation of overpressure has been confined to two representative configurations, BR00 and BR30S300L, with a total number of 346 and 312 tests, respectively. Only $t_d = 60$ s and $t_d = 3$ s are compared to each other. The long obstacle path in BR30S300L allows for more efficient acceleration of lean mixtures compared to BR30S300. In addition, more pressure transducers within the obstacle path become available for evaluation.

To obtain a relation between flame speed and peak overpressure, pressure transducers p_2-p_5 are employed to determine peak pressure and adjacent photodiodes deliver local flame speed at respective pressure transducer positions. Transducer p_1 is omitted since no significant acceleration takes place at this early position. Measured peak pressure at p_1 is dominated by FA that takes place further downstream in the channel and does therefore not correlate with local flame speed. Transducer p_6 is also omitted since it showed signs of thermal shock because the protective silicone coating repeatedly peeled off due to generally high pressures and flow velocities at this location.

The approach to cover a wide range of flame speed and peak overpressure was to conduct experiments at different average H_2 concentrations between 12.5 and 40 vol. %. To make these experiments comparable, peak overpressure needs to be related not to flame speed, but to flame Mach number M_F . In a first approach, average H_2 concentration is employed for the calculation of average reactant sound speed a_{re} . Thus,

$$M_{\rm F} = \frac{\rm v}{\rm a_{\rm re}}.$$
(5.9)

 M_F will be referred to as the global flame Mach number. Figure 5.38 (a) presents experimental results for $t_d = 60$ s and 3 s in BR00. Overpressure is generally higher at a given global flame Mach number in homogeneous mixtures than in gradient mixtures. In other words, flames need to propagate faster (higher speed of the leading flame tip) in gradient mixtures than in homogeneous mixtures to reach equal overpressure at the pressure measurement position (channel top). For a more meaningful comparison, the location of pressure measurement needs to be taken into account. Since overpressure is measured at the channel top, local mixture properties at this position need to be considered by correlating overpressure data not to a global, but to a local flame Mach number at y = 0.06 m, thus employing the local reactant sound speed $a_{re,v=0.06m}$. Local flame Mach number at the channel top is calculated as

$$M_{F,y=0.06m} = \frac{V}{a_{re,y=0.06m}}.$$
(5.10)

For homogeneous mixtures, M_F equals $M_{F,y=0.06m}$. This approach yields a common correlation of flame speed and peak overpressure for homogeneous and inhomogeneous mixtures, Fig. 5.38 (b), solid line, with a quadratic fit

$$p = M_{F,y=0.06m}^2 \cdot 0.48bar + M_{F,y=0.06m} \cdot 2.8bar + 0.22bar.$$
(5.11)

Data from BR30S300L can be correlated similarly, see Fig. 5.38 (c). The correlation from BR00 (Eq. (5.11)) is again plotted as a solid line in this figure for comparison. In general, scatter of data points is more distinct. At local flame Mach numbers higher than unity, peak overpressure systematically exceeds values from BR00 since transverse shock waves are generated by shock diffraction around obstacles, impinging on the pressure transducers and thus increasing measured peak overpressure.



+t_d= 60 s, **☆**t_d= 3 s

Figure 5.38: Correlation between overpressure p and global flame Mach number M_F in BR00 (a), p and local flame Mach number $M_{F,y=0.06m}$ in BR00 (b) and p and $M_{F,y=0.06m}$ in BR30S300L (c). Black line: quadratic fit (Eq. (5.11)). Blue dotted line: model by Krok [83].

Since the assumption of quadratic data fit is not directly physically motivated, experimental data is compared to an analytical model of a flame driving a shock, propagating from the closed end of a tube. This model is based on the work of Krok [83]. It was implemented in MATLAB, using Cantera [55] and the Shock and Detonation Toolbox [133]. Three states are considered:

- Undisturbed state 0, quiescent mixture at initial conditions.
- Post-shock state 1, chemically frozen.
- Post-flame state 2, assuming $u_2 = 0$ (laboratory frame of reference).

As long as flame speed is lower than post-flame sound speed, expansion across the flame front can bring the flow to rest, thereby satisfying the rear wall boundary condition. No expansion wave forms behind the flame front [83]. This is valid within the entire range of flame Mach numbers investigated here.

The change of state across the flame front is determined by the intersection of Rayleigh line and product Hugoniot for given γ and specific heat release q (Eq. (2.1)). These relations are solved using a Newton-Raphson method. Finally, flame speed and flame Mach number are calculated from conservation of mass across the flame front.

Blue dotted curves in Fig. 5.38 depict the model result, assuming a 30 vol. % mixture¹⁸. Experimentally observed peak overpressure is generally underpredicted by the model. Since the model is based on a 1D approach, thus neglecting locally increased pressure due to three-dimensional effects like shock focusing, the result represents a lower bound to the experimental data. Towards this bound, experiments approach 1D behavior. Agreement of the model with experimental data, following this understanding, is very good. In the following section, the relation between overpressure and flame speed will be merged with the beforehand developed understanding of onset of detonation.

¹⁸The result varies in a negligible range with a variation of H_2 concentration, since γ is very similar in H_2 (1.41) and air (1.40), values at standard conditions, and variations of q are of minor influence.

5.4 Discussion

This section merges results from experimental and theoretical studies on FA and onset of detonation presented in Secs. 5.1–5.3. A comprehensive picture of the influence of transverse concentration gradients on DDT is developed. First, critical conditions for onset of detonation in terms of overpressure and flame Mach number are defined in Sec. 5.4.1. Then, the influence of transverse concentration gradients in unobstructed and obstructed channels is discussed in Sec. 5.4.2. This section ends with comments on the impact of concentration gradient orientation, Sec. 5.4.3.

5.4.1 Critical Flame Mach Number for Onset of Detonation

In a fast deflagration at a state close to critical conditions for the onset of detonation the highest overpressure within the shock-flame complex occurs behind the precursor shock (p_1). Across the flame, expansion subsequently leads to pressure decrease. Thus, the relation between peak overpressure ("postincident-shock overpressure") and local flame Mach number, Sec. 5.3, can be linked to the model for strong ignition behind a reflected shock, Sec. 5.2.3. Since critical conditions for strong ignition can be expressed in terms of postincident-shock overpressure, a critical local flame Mach number must be reached to achieve strong ignition and thus potentially onset of detonation. This applies to homogeneous and gradient mixtures.

The 1D model of a shock-flame complex by Krok [83] predicts a critical flame Mach number of 2.6–2.7 to reach a peak overpressure of 10–11 bar, cp. Fig. 5.38. Since this 1D model yields only a lower bound for realistic local overpressure, these Mach numbers cannot portray a conservative boundary. Real local peak pressures tend to be higher. Employing the experimental correlation, Eq. (5.11), critical flame Mach number can be estimated slightly lower at 2.4–2.6. The shadowgraph sequences of onset of detonation presented in Sec. 5.2.2 support these values of critical flame Mach number. It is more accurate to determine the precursor shock Mach number as opposed to flame Mach number from these shadowgraph sequences since the flame is visible within

Figure	X _{H2} [%]	t _d [s]	M _{S,y=0.06m} [-]
5.29	16.5	60	2.6
5.30	17	3	2.5
5.31	22.5	3	2.5
5.32	22.5	3	2.6
5.33	26	3	2.4
5.34	30	3	2.5
5.35	35	3	2.4

Table 5.2: Local precursor shock Mach number shortly before onset of deto-
nation, corresponding to shadowgraph sequences in Sec. 5.2.2.

the FOV only for a very short time. Shock and flame propagate at equal velocity within the measurement accuracy (primarily limited by the camera resolution) at that state so that these Mach numbers can be used equivalently. Table 5.2 provides values of local Mach number $M_{S,y=0.06m}$ of the precursor shock at the channel top shortly before onset of detonation, corresponding to Figs. 5.29–5.35. It can be seen that local shock and thus local flame Mach number $M_{F,y=0.06m}$ was higher than 2.4 in all of these experiments. Overpressure in the range of 10–11 bar is often observed shortly before onset of detonation as can be seen in Figs. 5.29 (a) and 5.30 (a), for instance. These values are in very good agreement with the theoretical predictions.

Distinct scatter in experimental data, in particular visible in Fig. 5.38, clearly shows the stochastic nature of DDT: In single experiments—and of course in real-world explosions—local pressure can exceed values predicted by theoretical or experimental correlations. Shock focusing may be a major reason. Critical flame Mach numbers determined here thus need to be understood as statistic mean values and be used only with an appropriate safety margin. Note that the approach using the extended second explosion limit could be extended for more complex scenarios of shock-induced strong ignition, for example involving shock focusing.

It is a common assumption in DDT research that the speed of sound of the reaction products a_{pr} can be used to estimate critical conditions for the onset of detonation. In that argumentation, flame speed needs to reach this sound
speed, thus $v = a_{pr}$. This approach does not take into account the physics of onset of detonation, but is rather of empirical origin. The important question is how a_{pr} should be calculated. Isobaric or isochoric combustion may be assumed. Neither of them yields realistic post-flame equilibrium states that are observed in a fast deflagration. Considering a shock-flame structure like in Krok's model [83] might give more realistic values. In comparison, the approach to determine critical conditions for onset of detonation presented in the present work is based on a combination of experimental observations and simulation of detailed chemical kinetics. Since the sound speed of unburnt mixture can be readily calculated, no uncertainty is related to this step here. An uncertainty is however the exact value of critical local flame Mach number for real-world situations. This uncertainty originates from three-dimensional effects in the first place as outlined beforehand. This poses a general problem for every analytical criterion that defines critical conditions for the onset of detonation.

5.4.2 Comparison of Unobstructed and Obstructed Channels

In experiments presented in Secs. 5.1–5.2, it has been found that mixtures with transverse concentration gradients can pose a considerably higher explosion hazard in closed channels than homogeneous mixtures at equal average H_2 concentrations. This hazard manifests through stronger FA and earlier DDT in terms of average H_2 concentration.

Clearly different conclusions must be drawn for unobstructed and obstructed configurations. In the former, flame elongation leads to flame surface area enlargement, which allows flames in gradient mixtures to accelerate much stronger than flames in homogeneous mixtures. Flame elongation likewise leads to earlier DDT in terms of average H_2 concentration. Already a mixture of 20 vol. % with a steep concentration gradient was observed to undergo DDT, which would be the case in a homogeneous mixture only if obstructions were present. In contrast to this shift of the lower DDT limit towards lower H_2 concentrations, gradients were found to suppress DDT at higher concentrations and thus pose an initially unexpected upper DDT limit. This can now

be explained by merging findings from Secs. 5.2.3, 5.3 and 5.4.1. It has been shown that flames in gradient mixtures need to accelerate to an equal local flame Mach number as homogeneous mixtures to cause certain values of local overpressure. Critical overpressure for the onset of detonation was derived for obstructed configurations. A similar argumentation may also apply to unobstructed channels. Local explosions in gradient mixtures, leading to the onset of detonation, were observed at the upper channel wall in BR00. Local pressure and temperature is increased by a series of shocks preceding the flame. When critical values are reached, the mixture can auto-ignite. This mechanism was introduced in Sec. 2.6 based on the work of Dzieminska and Hayashi [37]. Details of mixture preconditioning may be complex involving interaction of shocks and boundary layer, but eventually the buildup of overpressure dominates this process. In Fig. 5.27, it was observed that continuous mixture compression leads to a local overpressure of about 11.5 bar before a local explosion occurs. This value is only slightly higher than the critical overpressure determined for the onset of detonation in obstructed channels (10–11 bar). A local flame Mach number of 2.7 was measured in BR00 shortly before onset, which is in good accordance with the determined relation between overpressure and local flame Mach number, Eq. (5.11).

In summary, two mechanisms compete regarding DDT propensity:

- Enforcement of FA through flame surface area enlargement and, below the flame speed cross-over concentration of 24 vol. %, due to increased effective flame speed $(S_L\sigma)_{eff}$.
- Requirement for acceleration of the leading flame tip, which propagates at the channel top, to a critical local flame Mach number. Since reactant sound speed increases with increasing H₂ concentration, higher flame speed must be reached in gradient mixtures to allow for onset of detonation, which retards onset by causing a need for further acceleration distance.

The upper DDT limit in experimentally observed probabilities of DDT in BR00, Fig. 5.28, can be explained by this competition. In mixtures of $t_d = 3$ s, effective flame speed ($S_L \sigma$)_{eff} remains fairly constant beyond the flame speed

cross-over concentration, cp. Fig. 5.25. At the same time, the required flame speed for onset of detonation continuously increases with increasing H₂ concentration. Critical flame speed, or flame Mach number, cannot be reached within the channel length in mixtures of 30 vol. % and higher at $t_d = 3$ s. In comparison, effective flame speed still increases considerably in $t_d = 7.5$ s mixtures beyond the flame speed cross-over concentration, so that this increase overcomes the requirement for higher critical flame speed and leads to a broad range of concentrations that undergo DDT.

From a practical perspective, DDT propensity at low H_2 concentrations in an unobstructed channel can be estimated by using the maximum local H_2 concentration at the channel top. On the one hand, this confirms global observations of Kuznetsov et al. [85] and Grune et al. [56]. On the other hand, the present work shows that such an approach does not reflect the physics of DDT in mixtures with transverse concentration gradients and thus represents an empirical criterion only.

Differences between the underlying physics and the simple criterion of maximum concentration become most obvious in experiments in obstructed channel configurations. Even at a low blockage ratio of 30 % and a large obstacle spacing of 300 mm, DDT is not promoted by gradients in the entire detonating range of H₂ concentrations tested (17–40 vol. %). It was found that mitigation of flame elongation by obstructions is responsible for this major difference to BR00. Comparison of unobstructed and obstructed configurations allowed for separating the role of mixture properties and the flame elongation process. It was thereby understood that only global consideration of mixture properties, in contrast to local properties, yields an accurate description of the FA process in entirely closed channels. At average H₂ concentrations of 24 vol. % and lower, FA in terms of flame speed is stronger in mixtures with gradients due to higher effective flame speed (S_L σ)_{eff}. Beyond this concentration, FA is weakened by gradients due to lower (S_L σ)_{eff}.

In mixtures with transverse concentration gradients, local explosions that are responsible for the onset of detonation were mainly observed at the upper obstacles, thus in the most fuel-rich region in the channel. Only if local H_2 concentrations at lower obstacles exceed about 10 vol. %, local explosions

originating from lower obstacles additionally support the onset of detonation. No situation has been observed where only a local explosion at the lower obstacle, but none at the upper one occurred. The strength of local explosions, estimated based on their propagation velocity in shadowgraph sequences, is higher at the channel top than at the bottom in a wide range of average H_2 concentrations. This is presumably due to the higher energy release in these explosions at locations of high local H_2 concentration. Thus, these upper explosions dominate the process of onset of detonation in mixtures with transverse concentration gradients.

Evidently, the flame speed cross-over concentration of 24 % can only predict a rough trend for DDT propensity of gradient mixtures in obstructed configurations where flame elongation is mitigated. Beyond the cross-over concentration, FA is slowed down by gradients. DDT propensity is clearly reduced. Below the cross-over concentration, gradients cause stronger FA in terms of flame speed, but not necessarily a higher propensity for DDT. No case of significantly earlier DDT in gradients in terms of average H₂ concentration has been found in obstructed configurations at H₂ concentrations as low as 17 vol. %. It might be expected that this changes at even lower average H₂ concentrations, where enforced FA may overcome the requirement for higher critical flame speed for onset of detonation. This could however not be substantiated in the present work since larger channel dimensions, both in terms of cross-section (cp. detonation cell width, Fig. 2.7.2) and length (high RUD), would be necessary to reach DDT in such mixtures.

In conclusion, concentration gradients primarily lead to higher DDT propensity in closed channels if flame elongation is possible. This mechanism requires a low degree of blockage and is most effective in entirely unbstructed channels. Already a low degree of blockage (BR30S300) effectively mitigates flame elongation and leads to a similar lower DDT limit in homogeneous and gradient mixtures. It is not yet clear what minimum degree of blockage is sufficient to achieve this effect. Flame elongation is believed to proceed differently in different unobstructed channel configurations (e.g. dimensions, cross-section shape, wall roughness). A generalized model for the elongation process cannot be deduced from only one experimental setup, particularly if a limited channel length as in the present work suggests that effects of the end plate might not be negligible. Deeper insight into the process of flame elongation in various geometries is required, ideally supported by numerical simulations. Unfortunately, unobstructed channels impose the highest requirements on numerical simulations since flame instablilty, turbulence generation in wall boundary layers and turbulence-flame interaction need to be captured at highest accuracy. DDT simulations in unobstructed channels are often less accurate than such in obstructed configurations.

5.4.3 Comments on the Orientation of Concentration Gradients

As discussed in Sec. 2.8, three-dimensional concentration gradients may prevail in explosive clouds in real-world explosion scenarios. To study only transverse or parallel gradients is a helpful simplification that allows for scientific investigation and determination of underlying physical phenomena. In specific scenarios like explosions in horizontal or vertical tubes, one of these orientations might indeed dominate. Knowledge on both types of gradients may be merged to identify worst-case scenarios. This is only possible to a limited extent at present since only a small number of published studies on inhomogeneous mixtures is available. Quantification of the gradient effect over a sufficient range of parameters such as mixture composition, enclosing geometry or initial conditions is still missing. Interpretation of measurement data is often bounded to qualitative statements. Based on the information currently available in literature and knowledge generated within the present work, the following trends can be formulated:

- Both transverse and parallel concentration gradients can increase the hazard of confined explosions, compared to homogeneous mixtures of equal average H₂ concentration. The physics behind FA and DDT has been investigated in more detail for transverse gradients compared to parallel gradients so far.
- Transverse gradients might pose a more severe additional hazard in geometries with a low degree of congestion than parallel gradients due to the potential for flame elongation. Flame elongation does not occur in

parallel gradients where the macroscopic flame shape is similar in homogeneous and gradient mixtures throughout the FA process.

- While the maximum local H₂ concentration might be used for a first rough estimation of DDT propensity in transverse gradients in an unobstructed tube, validity of this method for parallel concentration gradients cannot be assumed. In parallel gradients, the explosion front is in contact with the region of maximum H₂ concentration only for a limited time during the explosion.
- Overall explosion severity can highly depend on the specific process taking place inside a localized region of high H₂ concentration. For example, the onset of detonation might be facilitated in such a region, depending on local mixture reactivity and geometry. A detonation initiated there could propagate into regions of lower H₂ concentrations and potentially be sustained.

6 Detonation in H₂–Air with Transverse Concentration Gradients

This chapter investigates detonation propagation in H_2 -air with transverse concentration gradients and thereby completes the range of possible explosion regimes. Detonations in homogeneous mixtures are characterized as a reference in Sec. 6.1 before detonations in gradient mixtures are discussed. Two series of measurements are presented: Variation of gradient slope at 25 vol. % average H_2 concentration (Fig. 6.1 (a)) and variation of average H_2 concentration at $t_d = 3$ s (Fig. 6.1 (b)).



Figure 6.1: Concentration gradient profiles of detonation experiments from CFD simulations [42]. Variation of t_d at 25 vol. % (a); variation of average H₂ concentration at t_d = 3 s (b).

Detonations were produced in the obstacle section of configuration BR60S300 and studied in the unobstructed channel section. Pressure transducers p_4 and p_6 in the unobstructed section of the channel composed of six standard segments were used to measure detonation arrival time and thereby determine the detonation velocity. Compared to photodiodes, pressure transducers proved more accurate due to a steeper signal rise. Shadowgraphy, OH* luminescence imaging and soot foils were applied to configuration BR60S300 OS5. The FOVs of all optical measurements and also the soot foils were centered at x = 3.9 m.

For detonation velocity measurements it is essential that a quasi-steady velocity is reached upstream of the velocity measurement section. Among all average H₂ concentrations examined subsequently the case 22.5 vol. % yields the highest run-up distance to the onset of detonation. Onset occurs at the last obstacle of BR60S300 both in homogeneous and $t_d = 3$ s mixtures as already shown in Sec. 5.2, Fig. 5.32. Two insights from this shadowgraphy sequence are important in the context of detonation characterization: The onset of detonation occurs directly behind the obstacle. The detonation structure is established quickly. Second, the detonation propagates into undisturbed, quiescent mixture. The first two images of the sequence show a vortex pair behind the upper and lower obstacle which is a marker for the first significant fluid displacement at this location due to the approaching deflagration. Already in the third image the combustion wave catches up with the leading part of this region. The concentration gradient ahead of the detonation thus maintains the initially generated profile in the downstream measurement section. Detonation velocity attains a stable value already behind pressure transducer p₃ as can be seen in the pressure trace diagram of an exemplary experiment with a 22.5 vol. %, $t_d = 3$ s mixture, Fig. 6.2. Each pressure signal is normalized by its maximum value. Transition to detonation can be localized between p₂ and p_3 (secondary pressure spike at p_2 ; typical detonation pressure signal at p_3). The fluctuation of detonation velocity between adjacent pressure transducers is lower than 0.6 % of the finally evaluated velocity between p_4 and p_6 .



Figure 6.2: Pressure trace diagram, 22.5 vol. %, t_d = 3 s, BR60S300. Detonation velocity measurement between p_4 (blue, x_{p_4} = 3.2 m) and p_6 (orange, x_{p_6} = 5.0 m).

6.1 Reference Experiments in Homogeneous Mixtures

This section begins with detonation velocity measurements shown in Fig. 6.3, $t_d = 60$ s. Data points represent the average of five experiments each. Standard deviations are smaller than 1 % for all data points. A dimensionless depiction in Fig. 6.3 (b) will allow for comparison with literature. Velocities in homogeneous mixtures are close to the Chapman-Jouguet velocity D_{CJ} of the respective mixtures. Values beyond D_{CJ} are unexpected, but might be explained by an inaccuracy of the CJ model.

Shadowgraph images, Fig. 6.4, show the well-known structure of detonations in homogeneous mixtures, such as the coupled leading shock and reaction zone as well as triple points (kinks in the leading front) moving in a vertical direction. The richer the mixture, within the range discussed here, the less these triple points emerge in the shadowgraph images. The reaction zone, which is visible as a dark area behind the leading shock, becomes narrower. This corresponds well to the reduction of induction time with an increase in H_2 concentration.



Figure 6.3: Detonation velocity at different average H₂ concentrations and gradients. Dimensional (a) and dimensionless (b) depiction.



Figure 6.4: Shadowgraph images of detonations in homogeneous mixtures.

6.2 Overview of Propagation Regimes

In this section, a general overview of experimental observations on detonation propagation in mixtures with transverse concentration gradients is given. For this purpose, detonation velocities are examined first. Afterwards, a series of measurements at a constant average H_2 concentration of 25 vol. % with a variation of concentration gradient slope is presented.

6.2.1 Detonation Velocity

Figure 6.3 (a) shows that detonations in mixtures with concentration gradients ($t_d < 60$ s) propagate slower than in homogeneous mixtures at equal average H₂ concentrations. This is a first obvious difference to the deflagration regime. This velocity deficit is further quantified in the dimensionless depiction in Fig. 6.3 (b). The steeper the gradient, the larger the velocity deficit. Mixtures with average concentrations equal to and lower than 30 vol. % show similar normalized velocity deficits, while richer mixtures yield smaller deficits. Nevertheless, even the steepest concentration gradients investigated ($t_d = 3$ s) do not suppress detonation propagation but cause only moderate velocity deficits of less than 9 % compared to D_{CJ} , which is calculated for the average H₂ concentration here. Kessler et al. [75] found velocity deficits of about 5–10 % compared to D_{CJ} in gradient mixtures and Calhoon and Sinha [16] determined a maximum deficit of 6 % before detonation failure occurred.

It has not been possible in the present work to determine whether complete failure of detonation can be caused by a concentration gradient, because also the DDT process in the experiment is strongly influenced by the gradient as shown in Ch. 5. This means that the differentiation if a limit for detonation propagation or for DDT is observed cannot be reliably achieved in this type of experiment. Formation of a detonation wave in a detonable mixture (for instance strong ignition with an exploding wire in a driver gas cloud) and subsequent exposure of the wave to the gradient mixture would lead to reliable results here.

6.2.2 Shadowgraph and OH* Luminescence Images

Figure 6.5 shows detonation fronts at an average H_2 concentration of 25 vol. % and varying gradient slope. Note that shadowgraph and OH* luminescence images were taken in different experiments and thus only show similar, but not identical detonation fronts. The homogeneous mixture (Fig. 6.5, $t_d = 60$ s) at 25 vol. % allows for multi-headed detonation propagation. Increasing the slope of the concentration gradient, the front gets progressively inclined (Fig. 6.5, $t_d = 10$ and 7.5 s). The macroscopic structure of the fronts remains similar to the homogeneous reference, the front is still multi-headed. The reaction zone seems to widen and becomes more diffuse in the OH* images.

This multi-headed regime has also been observed by Ishii and Kojima [64]. Gradient profiles in [64] and the present study are not directly comparable due to different mixture composition and gradient shapes. As a first orientation one may compare the average slope of the concentration gradient in terms of equivalence ratio. The steepest gradient examined in [64] has an average equivalence ratio slope of 0.0075 1/mm, whereas the average gradient slopes for the profiles in Fig. 6.5 are 0.0065 1/mm ($t_d = 10 \text{ s}$), 0.011 1/mm ($t_d = 7.5 \text{ s}$), 0.019 1/mm ($t_d = 5 \text{ s}$) and 0.028 1/mm ($t_d = 3 \text{ s}$). The average gradient slope is thus comparable between experiments in [64] and cases $t_d = 10 \text{ s}$ and 7.5 s in the present work.

Between $t_d = 7.5$ and 5 s a fundamental change in propagation mechanism occurs. In mixtures with $t_d = 5$ and 3 s, one strong single transverse wave appears, oscillating over the entire channel height. Following the classical understanding of cellular detonations, there exists only half a detonation cell within the channel height here. Such a propagation mode has not yet been observed experimentally in the context of inhomogeneous mixtures. Single-headed detonations typically occur only in circular or near-circular cross-sections. This regime is subsequently referred to as the single-headed detonation regime. Typical terminology does not classify the single-headed propagation as a separate regime. However, this distinction will be used to structure this work due to the distinct differences that can be observed experimentally.

6.2 Overview of Propagation Regimes



Figure 6.5: Shadowgraph (left) and OH* luminescence (right) images of detonation fronts at varying t_d and an average H_2 concentration of 25 vol. %.

6.3 Single-Headed Propagation

This section focuses on the single-headed detonation regime in mixtures with the steepest gradients examined ($t_d = 3$ s). It is characterized by means of highly time-resolved shadowgraphy, Sec. 6.3.1, and OH* luminescence imaging, Sec. 6.3.2, as well as by soot foils, Sec. 6.3.3.

6.3.1 Shadowgraph Images

At an average H₂ concentration of 25 vol. %, highly dynamic detonation propagation can be observed. Figure 6.6 shows a corresponding shadowgraph sequence. Two parts of a characteristic cycle can be seen, recorded in two experiments (two columns). This cycle occurs in most of the experiments at average concentrations up to 30 vol. % at $t_d = 3$ s. The structure of the detonation front resembles a single-headed detonation. One strong transverse wave appears which is periodically reflected off the channel walls. Comparable to the formation of transverse waves in homogeneous mixtures, this wave forms in order to equilibrate pressure differences behind the leading detonation front, which are intensified by the H₂ concentration gradient. Reflection of this transverse wave at the channel top causes strong local explosions and thereby periodic re-initiation of detonation. Note that there is no injection manifold installed near the location of the local explosion. Thus, the single-headed regime is evidently not caused by manifolds but by the concentration gradient. Reaction is coupled with the shock within the first frames after the local explosion (Fig. 6.6, left column). When this wave is reflected off the channel bottom, reaction is still coupled behind the Mach-stem but progressively decouples behind the incident shock (right column, t = 25–62.5 μ s). Arriving at the channel top, transverse wave reflection again causes a local explosion, which completes one cell cycle. Local detonation front velocity oscillates between approximately 1.2 and 0.8 times the average propagation velocity over one oscillation cycle, which is very similar to the range observed in detonation cells in homogeneous mixtures [131]. The grey blurred area behind the leading shock, identified as the reaction zone, extends across the entire shadowgraph image.



Figure 6.6: Shadowgraph sequences of detonations at an average H_2 concentration of 25 vol. % and $t_d = 3$ s. Columns represent two separate experiments.

This suggests that significant portions of mixture react in unburnt pockets as a turbulent deflagration or combined auto-ignition and deflagration downstream of the leading detonation front.

6.3.2 OH* Luminescence Images

OH* luminescence imaging is useful to distinguish between local explosions, regular detonations and deflagrations as explained in Sec. 4.2.3. OH* images show local explosions as bright spots at distinctly higher luminosity as compared to the reaction zone behind a detonation that propagates close to CJ conditions. Deflagration manifests as regions of even lower luminosity than CJ detonations.

The OH* images in Fig. 6.7 show one entire cell cycle as described beforehand in Sec. 6.3.1. Red rectangles mark the positions of injection manifolds. Beginning with the strong local explosion at the channel top, clearly upstream of the manifold, with a high local luminescence in Fig. 6.7, t = 0 μ s, the overdriven front propagates towards the channel bottom (Fig. 6.7, t = $0-50 \mu$ s). The front interacts with an injection manifold in Fig. 6.7, t = 25 μ s, but no influence on the overall propagation mechanism can be discerned. As the propagation velocity of the expanding front decreases, luminosity decreases accordingly. The enhanced rate of OH* production behind the Mach-stem after reflection of the transverse wave at the bottom wall can be clearly seen (Fig. 6.7, t = 62.5 μ s). Figure 6.7, t = 62.5–137.5 μ s, comprises the upward propagation phase. As the shadowgraph images already showed, decoupling of shock and reaction zone occurs in the upper channel region. Luminosity decreases sharply and the separation distance between the assumed shock front and the reaction zone increases at the channel top. Shock fronts are reconstructed from shadowgraph images and marked as dashed white lines. Behind the decaying Mach-stem the images show no significant reaction towards the end of the cycle. In Fig. 6.7, t = 112.5 μ s, the front interacts with the second manifold in the FOV. Reflection causes elevated luminescence due to locally increased temperature, but a local explosion is not observed. In the last frame (Fig. 6.7, t = 137.5 μ s), reflection of the transverse wave at the channel top triggers the



Figure 6.7: OH* luminescence sequence of a detonation at an average H₂ concentration of 25 vol. % and $t_d = 3$ s.

local explosion. Detonation propagation is thereby sustained and the next cell cycle begins. This image sequence suggests that only about half the propagation cycle is driven by shock-induced auto-ignition. A significant share of the mixture seems to be consumed rather by deflagration than through auto-ignition.

6.3.3 Soot Foils

Soot foil measurements were performed to examine if traces at the channel side walls confirm the previous observations. This is particularly important since the channel width of the explosion channel is large (0.3 m), which means that line-of-sight integration inherent to shadowgraphy and OH* lumines-cence imaging may lead to doubtful conclusions. Sooted plates were therefore installed at the side walls of the optical segment. Clear traces on these sooted



Figure 6.8: Soot foils of detonations at an average H_2 concentration of 30 vol. %. Homogeneous (top) and gradient mixture (bottom, $t_d = 3$ s).

plates were obtained at an average H_2 concentration of 30 vol. %. Results are presented in Fig. 6.8. The soot foil gained from a homogeneous mixture (Fig. 6.8, $t_d = 60$ s) serves as a reference. Exactly one cycle of transverse wave oscillation over the channel height was captured on one soot plate in the 30 vol. % mixture at $t_d = 3$ s. The upward-leading trajectory with low inclination corresponds to the triple point formed by incident shock, Mach-stem and upward-propagating transverse wave. The transverse wave propagates slowly in this part of the cycle compared to the incident shock. The steep downward-leading trajectory corresponds to the triple point after local explosion at the channel top. The arrow in Fig. 6.8 highlights the estimated location of local explosion. Furthermore, the soot foils underscore that the injection manifolds are not the cause of the single-headed regime. From the foil, the cell length can be estimated at about 0.18 m, not being a multiple of the manifold spacing. There may be a second transverse wave visible on this plate, but the interaction with transverse waves moving in the spanwise direction of the channel, which are visible as vertical wavy imprints on the plate, complicates the analysis.

6.4 Multi-Headed Propagation

In mixtures richer than 35 vol. % H_2 at $t_d = 3$ s, detonation propagation is multiheaded. Transverse waves are continuously regenerated by collisions with oncoming transverse waves and with the channel walls. This regime equivalently appears in leaner mixtures with weaker gradients as shown already in Fig. 6.5.



Figure 6.9: Shadowgraph (left) and OH* (right) images of detonation fronts at $t_d = 3 \text{ s}, 35$ (top) and 40 vol. % (bottom).



Figure 6.10: Soot foils of detonations at an average H_2 concentration of 40 vol. %. Homogeneous (top) and gradient mixture (bottom, $t_d = 3$ s).

Figure 6.9 shows detonation fronts in 35 vol. % and 40 vol. % H_2 at $t_d = 3$ s. The major macroscopic difference compared to lower average H_2 concentrations is a constant front curvature over time without visible Mach-stem formation on the upper or lower wall. The reaction zone (dark zone in shadowgraph images) is much narrower. This indicates a higher portion of mixture being directly consumed by auto-ignition, which may serve as an explanation for the lower velocity deficit compared to single-headed detonations. A singular strong transverse wave as seen in Fig. 6.6 does not form.

Soot foil measurements (Fig. 6.10) show curved traces similar to observations of Ishii and Kojima [64]. Detonation cells are asymmetric compared to the pattern in the homogeneous mixture with a higher portion of substructures. Near the walls, large cells would be expected, considering the local H₂ con-

centrations. It is thus surprising that cells remain very small even far away from the center line. The dynamics of transverse waves thus cannot be derived directly from local mixture composition, but requires consideration of the entire transverse wave oscillation cycle between the channel walls. Mixtures beyond 40 vol. % have not been investigated optically. Single-headed detonations might occur again when the upper detonation limit for the channel geometry is approached.

6.5 Discussion

It has been shown that detonation propagation is possible even in mixtures with very steep concentration gradients. Propagation velocity is generally lower in a gradient mixture than in a homogeneous mixture at equal average H_2 concentration.

Two detonation regimes were observed experimentally: single-headed propagation with one strong transverse wave and multi-headed propagation with a constant macroscopic front curvature over time and numerous weak transverse waves. The single-headed regime can be interpreted as a near-limit phenomenon similar to the spinning detonation observed by Dabora et al. [25]. It is also comparable to detonations in mixtures with high activation energy discussed by Gaathaug et al. [50]. The channel height of 0.06 m allows for detonation propagation in homogeneous mixtures with an H₂ concentration down to 16–17 vol. %. Single-headed propagation already occurs in mixtures with gradients at significantly higher average H₂ concentrations. It was induced by either steepening the gradient at constant average H₂ concentration or by decreasing the average H₂ concentration while maintaining the gradient slope (within the experimental limitations by keeping t_d constant).

Detonation cell width data as shown in Sec. 2.7 is used subsequently to interpret the investigated concentration gradient profiles physically. Note that these calculated cell widths cannot directly be expected in reality since the dynamics of transverse waves not only depends on local conditions, but on the entire oscillation cycle of transverse waves between the walls as seen in



Figure 6.11: Detonation cell width profiles corresponding to Fig. 6.1. Only cases with available optical data are displayed.

Fig. 6.10. Figures 6.11 (a) and (b) provide an analysis of detonation cell width as a function of local H_2 concentration corresponding to the concentration gradient profiles in Fig. 6.1 (a) and (b), respectively, that were charcacterized optically. Equation (2.40) delivers the relation between local H_2 concentration and cell width.

In case of a constant average H_2 concentration of 25 vol. %, Fig. 6.11 (a), it can be seen that the non-linear dependency between local H_2 concentration and cell width and the strong increase in cell width towards low local H_2 concentrations causes a sharp transition from small cells in the upper channel region to cells larger than the channel height in the lower part of most gradient profiles. Differences in cell width at the channel top are comparably small. Cases where single-headed propagation occurred, marked with "S", show the strongest increase in cell width towards the bottom as compared to multi-headed detonations, "M". This suggests that single-headed propagation occurs as soon as the minimum concentrations in the lower channel region

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	X _{H2} [vol. %]	t _d [s]	Avg. cell width [mm]	Detonable height [mm]	Regime
	25	3	25.3	34.2	single-headed
	25	5	21.1	37.8	single-headed
	25	7.5	21.6	43.8	multi-headed
	25	10	26.2	60.0	multi-headed
	25	60	18.5	60.0	multi-headed

Table 6.1: Average detonation cell width, detonable height and observed det-
onation regimes, corresponding to Fig. 6.11 (a).

reach sufficiently low values, causing a sharp increase in local cell width.

An evident qualitative similarity between the first group of experiments, Fig. 6.11, to detonation propagation in flat layers is the sharp increase in cell width in the fuel-lean region at the channel bottom. Thus, a question is whether an effective detonable layer height required for multi-headed propagation can be defined. For semi-confined configurations with homogeneous mixtures, a layer thickness of about 3 times the cell width is required for self-sustained multi-headed detonation propagation [50, 129]. The required number of detonation cells might be lower in the entirely confined configuration because reflection of transverse waves at the lower wall supports detonation propagation. For the following analysis only cells smaller than the channel height of 0.06 m are considered since this would pose the lower limit for detonation propagation in a homogeneous mixture. Table 6.1 shows the overall height within the channel where cells are smaller than the channel height, referred to as the detonable layer height in the following, and the average cell width in this region. Cases with single-headed detonation show detonable layer heights lower than 40 mm with less than 2 cells of average width in this region. Transition from single- to multi-headed detonation occurs when the detonable layer height is larger than about 40 mm. This corresponds to about 2 detonation cells being present in the detonable region. This value is close to the critical layer height of 3 cells found by Rudy et al. [129] and Gaathaug et al. [50]. While detonation fails in layers of smaller height in semi-confined configurations, the entirely confined channel in the present work still allows for detonation propagation in the single-headed regime.

[VOI. /0] [[5]	[mm]	[mm]	0
22.5	3	22.2	31.2	single-headed
25 3	3	25.8	34.2	single-headed
30 3	3	30.3	33	single-headed
35 3	3	28.1	28.8	multi-headed
40 3	3	28.2	28.9	multi-headed

Table 6.2: Average detonation cell width, detonable height and observed det-
onation regimes, corresponding to Fig. 6.11 (b).

Detonation cell width profiles in Fig. 6.11 (b) refer to experiments with steep gradients ($t_d = 3$ s) at varying average H₂ concentration. Table 6.2 shows the corresponding analysis of detonable layer height and average cell width. Similar to the group of profiles in Fig. 6.11 (a), cases with single-headed detonation show a detonable layer height of about 30 mm with about 1–1.5 detonation cells of average width in this region. Cell width increases sharply towards the fuel-lean region. In contrast to the group of profiles in Fig. 6.11 (a), high average concentrations and the steep gradients cause regions of large cells also at the channel top, in particular at $X_{H2} = 35$ and 40 vol. %. Despite this increase in cell width in the fuel-rich region, these two cases allow for multi-headed, very stable detonation propagation, cp. Fig. 6.9. Detonable layer height is also about 30 mm here. Only about one cell of average width is present in this region. The theoretical analysis of local cell width obviously does not deliver useful information on the detonation propagation mechanism in these two cases with globally fuel-rich mixtures.

In conclusion, interpreting concentration gradient profiles in terms of local detonation cell width seems to provide a useful tool to predict the stability and thus the propagation regime of detonations in globally fuel-lean mixtures with transverse concentration gradients. Cell widths of the investigated steep gradients increase sharply towards low local H₂ concentrations at the channel bottom. This poses a similarity to layers of reactive mixture bounded by an inert gas or a mixture of distinctly lower reactivity. A detonable layer height was introduced as a theoretical parameter. It was calculated as the region where detonation cells are smaller than the channel height. According to the experi-

ments, about 2 detonation cells need to be present in the detonable layer to allow for multi-headed detonation propagation. If the detonable height is lower, single-headed, unstable detonations occur. By further reducing the detonable layer height by even steeper transverse concentration gradients, failure of detonation will presumably occur. This could however not be investigated in the present work. In globally fuel-rich mixtures an increase in cell size also occurs at the channel top. Such profiles are not comparable to a layer of reactive mixture anymore. Multi-headed, very stable detonation propagation is possible even if the theoretically determined detonable region is rather narrow. Energy transfer between regions of higher and lower reactivity, realized by transverse waves, needs to be investigated in more detail to gain a better understanding of such scenarios.

7 Summary and Outlook

The present work investigated the influence of transverse concentration gradients on deflagration-to-detonation transition and detonation propagation in H_2 -air mixtures in a closed rectangular channel. It was part of a German nuclear reactor safety research program. Only little knowledge is available on this topic although inhomogeneous mixtures prevail in real-world explosion accident scenarios. This work makes a contribution to explosion safety research through providing a comprehensive experimental study on laboratory scale. This includes all possible explosion regimes from slow deflagration to detonation. A particular focus has been placed on the phenomenon of deflagrationto-detonation transition due to its worst-case character. Broad application of advanced (laser-) optical measurement techniques at high temporal resolution in conjunction with conventional techniques was one of the major features of the experimental approach. Separation of the characteristic phases of deflagration-to-detonation transition helped to identify the particular influence of gradients in each specific phase. Theoretical approaches supported the interpretation of experimental results.

High practical relevance of transverse concentration gradients for deflagration-to-detonation transition has been found and is summarized as follows:

 Deflagration-to-detonation transition can be significantly promoted by transverse concentration gradients. It can occur earlier in terms of average H₂ concentration compared to homogeneous mixtures. This means, that an inhomogeneous distribution of a given amount of H₂ in air in a given volume may pose a considerably higher explosion hazard than homogeneous distribution. Criteria for deflagration-to-detonation transition that are available for homogeneous mixtures can therefore not be considered conservative in many real-world situations. • Whether gradients cause stronger explosions mainly depends on the enclosing geometry. It was shown that transverse gradients promote deflagration-to-detonation transition significantly in an unobstructed channel. In such a geometry, the maximum local H₂ concentration can be used to compare deflagration-to-detonation transition propensity in homogeneous and gradient mixtures. By contrast, already a low degree of obstruction leads to similar lower limits for deflagration-to-detonation transition in terms of average H₂ concentration in gradient and homogeneous mixtures. The concept of maximum concentration fails in this case.

The entire process of deflagration-to-detonation transition was split up into the flame acceleration phase and the final onset of detonation. These processes are influenced differently by transverse concentration gradients.

- Transverse concentration gradients influence flame acceleration through two major effects: enlargement of macroscopic flame surface area by flame elongation and variation of effective (integral) flame speed of the mixture. The former effect mainly occurs in unobstructed channels and explains the higher propensity for deflagration-to-detonation transition caused by gradients there. Obstructions by contrast hinder flame elongation. The latter has an effect in both unobstructed and obstructed configurations.
- When flame elongation is suppressed by the enclosing geometry, like in obstructed configurations, a flame speed cross-over concentration appears at around 24 vol. % H₂. This was reproduced analytically by taking into account the effective (integral) flame speed of the mixture. Flame speed is defined as the product of laminar burning velocity and expansion ratio. Only below the average H₂ concentration of 24 vol. %, gradients enforce flame acceleration in terms of flame speed. However, it was not observed that gradients promote deflagration-to-detonation in this concentration range in obstructed channels. Beyond this point, gradients cause weaker flame acceleration. This automatically reduces the propensity for deflagration-to-detonation transition at high H₂ concentrations.

Trends in deflagration-to-detonation transition propensity were eventually explained by combining insight into the flame acceleration phase, physics and detailed chemical kinetics of the detonation onset process and the relation between flame Mach number and peak overpressure.

- Onset of detonation by shock reflection at obstacles was studied. Shadowgraph sequences show that the first and thus crucial step of onset of detonation is a strong local explosion at the upstream obstacle surface, caused by reflection of the fast deflagration precursor shock. A 1D model of shock reflection including detailed chemical kinetics around the extended second explosion limit was used to determine critical conditions for the onset of detonation. This showed that local overpressure behind the fast deflagration precursor shock at the channel top (in the most fuelrich region) is a crucial parameter that can be used for defining critical conditions for onset of detonation by shock reflection in an obstructed channel. It was found that critical local overpressure ranges between 10 and 11 bar. This value is nearly independent of H₂ concentration and therefore applies to both homogeneous and gradient mixtures.
- Local overpressure was experimentally correlated with local flame Mach number at the channel top where the leading tip of flames in gradient mixtures is located. A common correlation for homogeneous and inhomogeneous mixtures was obtained.
- The preceding steps revealed that flames both in homogeneous and gradient mixtures need to accelerate to the same critical flame Mach number to allow for onset of detonation. For gradient mixtures, the local flame Mach number at the channel top needs to be considered. Increased H₂ concentration at the channel top leads to higher local sound speed. Consequently, higher flame speed needs to be reached in gradient mixtures compared to homogeneous mixtures before the onset of detonation occurs. This can retard the onset of detonation even if flame acceleration is enforced by a concentration gradient. This finding directly explains why concentration in mixtures below the flame speed cross-over concentration of 24 vol. % H₂ in obstructed configurations.

In addition, transverse concentration gradients strongly influence detonation propagation:

- Self-sustained detonation propagation is possible even in mixtures with steep concentration gradients. Global propagation velocity decreases by up to 9 % in the transverse concentration gradient mixtures under investigation, compared to homogeneous mixtures at equal average H₂ concentration.
- Two detonation regimes were observed experimentally: single-headed propagation with one strong transverse wave and multi-headed propagation with a constant macroscopic front curvature over time and numerous weak transverse waves. The single-headed regime can be interpreted as a near-limit phenomenon similar to spinning detonations. Distinct phases of detonation failure and re-initiation occur, which may explain the observed global detonation velocity deficit. Low average H₂ concentrations and steep gradients foster the single-headed regime, whereas mixtures at high average H₂ concentrations and weak gradients allow for multi-headed detonation propagation.
- A qualitative similarity between detonations in globally fuel-lean mixtures with transverse concentration gradients to layered mixtures described in literature was found, which is the sharp increase of theoretically determined local detonation cell width in the most fuel-lean gradient regions. Only if two or more detonation cells are present within the detonable region (defined as the region where cells are smaller than the channel height) of a gradient in a globally fuel-lean mixture, detonations are multi-headed. Globally fuel-rich gradient mixtures cannot be treated like layered mixtures anymore since detonation cell width increases both in the lean and rich parts of the mixture.

Further steps are required to deepen the understanding of transverse concentration gradient effects. Since flame elongation has been identified as the dominant and thus most hazardous mechanism that enforces flame acceleration in gradient mixtures, a particular focus should be placed on this process. Understanding the physics of flame elongation and the transition from elongation in unobstructed channels to suppression of elongation in obstructed configurations is essential. Small degrees of obstruction should be investigated. This work dealt with H_2 -air mixtures only. A similar influence of transverse concentration gradients can be expected for other gases. The final steps would be to study parallel and multi-dimensional concentration gradients to further approach real-world conditions. Appendix

A Concentration Gradient Profiles

Profiles of H_2 concentration gradients used in the present work were determined by Ettner [42] by CFD simulation. This appendix provides 4th order polynomials describing these profiles for diffusion times $t_d = 3, 5, 7.5$ and 10 s. The general notation is as follows:

$$X_{H2}(y) = p_1 \cdot y^4 + p_2 \cdot y^3 + p_3 \cdot y^2 + p_4 \cdot y + p_5$$
 (A.1)

where y is the vertical position in the channel in [m] according to the coordinate system introduced in Sec. 3.1 and p_i are polynomial coefficients given in Tab. A.1.

X _{H2} [-]	t _d [s]	p_1 [1/m ⁴]	p ₂ [1/m ³]	p ₃ [1/m ²]	p ₄ [1/m]	p ₅ [-]
0.1	3	-2.73E+04	1585	49.48	0.241	0.01765
	5	-6186	-426.5	81.55	-0.09072	0.0425
	7.5	-964.3	-594.2	60.19	-0.1218	0.0645
	10	-172.9	-407.3	37.85	-0.08276	0.07806
0.2	3	-4.18E+04	1769	133.1	0.1339	0.04706
	5	-8604	-1176	165.9	-0.2946	0.09362
	7.5	-1315	-1166	114.1	-0.2416	0.134
	10	-246.5	-762.9	70.35	-0.1542	0.16
0.3	3	-3.66E+04	95.76	247.8	-0.1624	0.0949
	5	-7646	-1985	232	-0.4398	0.1595
	7.5	-1318	-1577	151	-0.3158	0.2143
	10	-306.5	-1002	92.3	-0.2002	0.2476
0.4	3	-1.88E+04	-2651	370.4	-0.5813	0.163
	5	-4417	-2732	276.6	-0.5312	0.241
	7.5	-1006	-1828	171.4	-0.3543	0.3041
	10	-315.5	-1127	103.6	-0.2223	0.3419

Table A.1: Polynomial coefficients of Eq. (A.1) describing concentration gradi-
ent profiles, deduced from CFD simulations by Ettner [42].

B Tunable Dye Laser Absorption Spectroscopy of the OH $Q_1(6)$ Line

Spectrally resolved absorption of radiation by OH radicals in an H_2 – O_2 diffusion flame at elevated pressure has been investigated by tunable dye laser absorption spectroscopy (TDLAS) by the author in cooperation with Dipl.-Ing. Thomas Fiala, Institute of Thermodynamics, TUM. A laminar jet flame established in an O_2 flow and an H_2 co-flow was used. The experimental setup allows for raising pressure up to 40 bar. For further details please refer to [43, 44, 46].

The laser system described in Sec. 4.2.2 was employed as source of radiation, delivering a focused beam which was guided through the center of the flame. In this context, the 1.8 pm Full Width at Half Maximum (FWHM) of the UV radiation is of importance since it defines the wavelength resolution of the TDLAS measurement. Wavelength was scanned continuously in the vicinity of the $Q_1(6)$ transition of the OH radical used in the OH-PLIF measurement in the present work. Absorption of radiation through the flame was measured simultaneously by means of UV-sensitive SiC photodiodes, including one reference diode for correction of variations in laser output power during the wavelength scanning process.

The purpose of these measurements was to obtain information on the effect of pressure on absorption and furthermore validate simulations presented in [43]. As stated in Sec. 4.2.2, one challenge in conducting OH-PLIF measurements of fast flames or even detonations is strong absorption of laser light within the flame.

Figure B.1 shows measured absorption α through the laminar H₂–O₂ flame as a function of wavelength λ in air and pressure. Absorption strongly depends on pressure. At ambient pressure the absorption characteristics are in good



Figure B.1: TDLAS measurement of a laminar H_2 – O_2 diffusion flame at elevated pressure. Absorption α as a function of wavelength λ and pressure.

accordance with theoretical predictions, cp. Fig. 4.9. Maximum absorption is measured close to the $Q_1(6)$ wavelength of 282.945 nm within the 30 pm precision of the dye laser. With increasing pressure, a growing amount of radiation is attenuated by the flame. At 10 bar, 98 % of the radiance at the $Q_1(6)$ line is absorbed. Absorption lines broaden through collisional broadening (also termed pressure broadening) and Doppler broadening due to increased temperature. The central wavelength of absorption seems to increase slightly with an increase in pressure. For example, a change in pressure from 1 bar to 10 bar results in a shift in maximum absorption wavelength by about 1.6 pm for all measured absorption lines. This shift is similar to the FWHM of the laser. The measurement accuracy thus clearly reaches a limit here. The question arises if the wavelength of an OH-PLIF laser system operating at the $Q_1(6)$ line needs to be set differently for low and high pressure flames (slow and fast flames in an FA experiment). Figure B.1 shows that line broadening leads to almost optimal absorption at an exemplary pressure of 10 bar at the $Q_1(6)$ wavelength for an atmospheric flame, although the maximum is minimally shifted towards higher wavelength. This is in good agreement with the author's experience who conducted all OH-PLIF experiments at equal laser wavelength. This conclusion also applies to detonation investigations.

Please note that absolute numbers of absorption are specific to the particular flame under investigation. General trends are however valid for all flames where OH occurs as a minor species. Compared to fast deflagrations in H_2 -air investigated in the present work, the local OH concentration is about one order of magnitude higher in the laminar diffusion flame. Absorption has two effects on OH-PLIF measurements: First, laser light is attenuated along its path through the flame. This is particularly problematic if the laser light sheet is introduced transversely into the explosion channel. In the pressure range up to 10 bar, which covers slow and fast deflagrations (cp. Fig. 5.38), absorption changes strongly. A clear recommendation is thus to introduce the OH-PLIF light sheet from the rear wall of an explosion channel, thus in opposite direction of flame propagation. This should help to extend the applicability of OH-PLIF towards faster regimes compared to the present work.

Secondly, fluorescence light is absorbed on its way to the camera (selfabsorption), leading to lower signal intensity and quantitative uncertainty because of the unknown three-dimensional absorptivity distribution. A thin channel design or employment of vertical local cookie-cutter plates could be considered to decrease self-absorption between the PLIF laser sheet and the camera. These measures also reduce recorded line-of-sight integrated flame luminescence intensity which tends to exceed the fluorescence intensity in fast flame regimes.
Previous Publications

Wesentliche Teile dieser Dissertation wurden vom Autor bereits standardmäßig vorab als Konferenz- und Zeitschriftenbeiträge sowie im Rahmen von Berichten veröffentlicht. Alle relevanten Vorveröffentlichungen sind entsprechend der gültigen Promotionsordnung ordnungsgemäß gemeldet und anschließend aufgeführt. Sie sind deshalb nicht zwangsläufig im Detail einzeln referenziert. Vielmehr wurde bei der Referenzierung eigener Vorveröffentlichungen Wert auf Verständlichkeit und inhaltlichen Bezug gelegt.

Significant parts of this Ph.D. thesis have been published by the author beforehand in conference proceedings, journal papers, and reports. All of these relevant prior printed publications are registered according to the valid doctoral regulations and listed below. However, not all of them are quoted explicitly everywhere as they are part of this present work being official documents. Whether these personal prior printed publications were referenced, depended on maintaining comprehensibility and providing all necessary context.

Boeck, L.R., Berger, F.M., Hasslberger, J., Sattelmayer, T., Detonation Propagation in Hydrogen-Air Mixtures with Transverse Concentration Gradients; Shock Waves, Submitted 14/07/2014.

Boeck, L.R., Hasslberger, J., Sattelmayer, T., Flame Acceleration in Hydrogen/Air Mixtures with Concentration Gradients; Combustion Science and Technology, Vol. 186, No. 10-11, pages 1650-1661, 2014. Boeck, L.R., Berger, F.M., Hasslberger, J., Sattelmayer, T., Detonation Propagation in Hydrogen-Air Mixtures with Concentration Gradients; Torus Press, Transient Combustion and Detonation Phenomena: Fundamentals and Applications, 2014.

Boeck, L.R., Msalmi, M., Koehler, F., Hasslberger, J., Sattelmayer, T., Criteria for DDT in Hydrogen-Air Mixtures with Concentration Gradients; 10th International Symposium on Hazards, Prevention and Mitigation of Industrial Explosions, Bergen, Norway, 2014.

Boeck, L.R., Hasslberger, J., Ettner, F., Sattelmayer, T., Investigation of Peak Pressures during Explosive Combustion of Inhomogeneous Hydrogen-Air Mixtures; Proceedings of the 7th International Seminar on Fire and Explosion Hazards, Providence, RI, USA, 2013.

Boeck, L.R., Primbs, A., Hasslberger, J., Ettner, F., Sattelmayer, T., Influence of Concentration Gradients on Detonation Velocities in Hydrogen-Air Mixtures; International Conference on Combustion and Explosion, Ramsau, Austria, 2013.

Boeck, L.R., Hasslberger, J., Sattelmayer, T., Explosive Combustion of Homogeneous and Inhomogeneous Hydrogen-Air Mixtures – Experimental Observations and Conclusions for Safety Applications; 3rd Colloquium of the Munich School of Engineering, Garching, Germany, 2013.

Boeck, L.R., Berger, F.M., Hasslberger, J., Ettner, F., Sattelmayer, T., Macroscopic Structure of Fast Deflagrations and Detonations in Hydrogen-Air Mixtures with Concentration Gradients; 24th International Colloquium on the Dynamics of Explosions and Reactive Systems, Taipei, Taiwan, 2013.

Boeck, L.R., Hasslberger, J., Ettner, F., Sattelmayer, T., Flame Acceleration in Hydrogen-Air Mixtures with Concentration Gradients; 24th International Colloquium on the Dynamics of Explosions and Reactive Systems, Taipei, Taiwan, 2013.

Boeck, L.R., Primbs, A., Hasslberger, J., Sattelmayer, T., Investigation of Flame Acceleration in a Duct Using the OH PLIF Technique at High Repetition Rate; Lasermethoden in der Strömungsmesstechnik, Munich, Germany, 2013. Boeck, L.R., Hasslberger, J., Sattelmayer, T., Flammenbeschleunigung und Deflagrations-Detonations-Übergang in Wasserstoff-Luft Gemischen mit Konzentrationsgradienten; 26. Deutscher Flammentag, Duisburg, Germany, 2013.

Supervised Student Theses

Im Rahmen dieser Dissertation entstanden am Lehrstuhl für Thermodynamik, Technische Universität München, in den Jahren 2011 bis 2014 unter wesentlicher wissenschaftlicher, fachlicher und inhaltlicher Anleitung des Autors die im Folgenden aufgeführten studentischen Arbeiten. In ihnen wurden verschiedene Fragestellungen zur Explosionssicherheit untersucht. Ergebnisse aus diesen Arbeiten sind in Teilen in das vorliegende Dokument eingeflossen. Der Autor dankt hiermit explizit allen ehemals betreuten Studenten für ihr Engagement bei der Unterstützung des hier behandelten Forschungsprojekts sowie der damit verknüpften Dissertation.

Several "student theses" emerged from the research project behind the present work, listed below. These student projects were conducted at the Lehrstuhl für Thermodynamik, Technische Universität München, in the years 2011 through 2014 under close supervision of the author of this Ph.D. thesis with regard to academic, professional, and context-related concerns. Various issues were investigated contributing to explosion safety research. The author would like to express his sincere gratitude to all formerly supervised students for their commitment and support of this research project and of the Ph.D. thesis at hand.

SUPERVISED STUDENT THESES

Student	Thesis title, submission date
Frederik Berger	Fast Deflagration and Detonation Phenomena in Hydrogen-Air Mix- tures with Concentration Gradients, 11/06/2013
Frederik Berger	Evaluation of High Pressure Water Atomisation for Detonation Cham- bers Using Fuel-Injection Nozzles, 28/10/2012
Ben Flocke	Modellierung von Flammenbeschleunigung in H ₂ -Luft Gemischen mit Konzentrationsgradienten, 01/07/2013
Juan Hasbun	Optische Untersuchung des Einflusses von Blockierrate und Konzen- trationsgradienten auf Flammenbeschleunigung und DDT in Wasserstoff-Luft-Gemischen, 02/12/2013
Andreas Kink	Explosion of Wet Hydrogen-Air Mixtures in a Closed Channel, 20/09/2014
Thomas Knapp	Entwicklung einer Eindüsungsmethode für Wassertröpfchen in einen Explosionskanal, 20/06/2013
Franziska Köhler	Übergang von Deflagration zu Detonation in einem glatten Kanal unter Einfluss von Konzentrationsgradienten, 27/09/2013
Stanislav Mironov	Numerische Simulation der Eindüsung flüssigen Wassers in einen Ex- plosionskanal, 15/06/2013
Soumaya Msalmi	Velocity and Overpressure of Confined Flame Propagation in Hydrogen-Air Mixtures with Concentration Gradients, 04/12/2013
Denizhan Oezdin	Theoretische und experimentelle Analyse von Explosionen in H_2 -Luft Gemischen mit flüssigem Wasser, 05/10/2014
Andreas Primbs	Anwendung der High-speed OH-PLIF Technik zur Charakterisierung von Verbrennungsvorgängen, 15/06/2013
Andreas Primbs	Implementierung und Anwendung einer Methode zur Auswertung von Explosionsversuchen, 31/10/2012
Markus Scholz	Untersuchung der Flammenausbreitung in H ₂ -Luft-Gemischen mit Mischungsgradienten, 30/06/2012
Felix Will	Konzeptfindung für die Einbringung disperser Wassertropfen in einen Explosionskanal mit anschließender experimenteller und nu- merischer Untersuchung, 30/06/2012

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