Resonant holographic interferometry application to NH and OH concentration measurements in a 2D diffusion NH₃-O₂ flame

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RESONANT HOLOGRAPHIC INTERFEROMETRY

Application to NH and OH Concentration Measurements in a 2D Diffusion NH₃-O₂ Flame

A dissertation submitted to the
SWISS FEDERAL INSTITUTE OF TECHNOLOGY ZURICH
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DOCTOR OF NATURAL SCIENCES

presented by
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Paul Scherrer Institute, 1997
As you set out for Ithaka
hope your road is a long one,
full of adventure, full of discovery.
Laistrygonians, Cyclops,
angry Poseidon-don’t be afraid of them:
you’ll never find things like that on your way
as long as you keep your thoughts raised high,
as long as rare excitement stirs your spirit and your body.
Laistrygonians, Cyclops,
wild Poseidon-you won’t encounter them
unless you bring them along inside your soul,
unless your soul sets them up in front of you.

Hope your road is a long one.
May there be many summer mornings when,
with what pleasure, what joy,
you enter harbours you’re seeing for the first time;
may you stop at Phoenician trading stations
to buy fine things,
mother of pearl and coral, amber and ebony,
sensual perfume of every kind-
as many sensual perfumes as you can;
and may you visit many Egyptian cities
to learn and go on learning from their scholars.

Keep Ithaka always in your mind.
Arriving there is what you’re destined for.
But don’t hurry the journey at all.
Better if it lasts for years,
so you’re old by the time you reach the island,
wealthy with all you’ve gained on the way,
not expecting Ithaka to make you rich.

Ithaka gave you the marvellous journey.
Without her you wouldn’t have set out.
She has nothing left to give you now.
And if you find her poor, Ithaka won’t have fooled you.
Wise as you will have become, so full of experience,
you’ll have understood by then what these Ithakas mean.

Konstantinós Π. Καβάφης / Constantinos P. Cavafis (1911)
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Alexios-Paul Tzannis

Paul Scherrer Institute,

December 1997
Abstract

This work presents an experimental investigation on Resonant Holographic Interferometry as a suitable diagnostic technique for species density measurements in a combustion environment. Resonant Holographic Interferometry is based on the dispersion of light close to an electronic transition of a molecule. A novel single-laser, two-color set up for the recording of resonant holograms is applied to two dimensional species concentration measurements in a laminar diffusion NH₃-O₂ flame.

The refractive index in a flame can be characterized by the bulk refractive index which is a function of the temperature and of the local molar fractions of the combustion species. In the vicinity of a resonant molecular transition the refractive index is complex. The complex refractive index is described in detail and the spectroscopic parameters are taken into account. It is also shown that the real part of the complex refractive index adds to the bulk refractive index and the imaginary part is responsible for the attenuation of the intensity of the probe wave.

The frequency dependent change of the refractive index is used to generate a phase shift between two waves of slightly different frequencies as they pass through the flame. In fact, the phase velocity of the two waves will be altered according to the total refractive index, $n_0 + n(v)$. By interferometric subtraction, the bulk refractive index $n_0$ of the flame is eliminated yielding the net contribution of the resonance effect to the refractive index $n(v)$. Holography is applied to perform the interferometric subtraction. The two waves are recorded on the same holographic plate and subsequently reconstructed simultaneously to interfere. Fringe shifts are observed in areas where the resonant species have introduced a refractive index difference for the two exposure frequencies.

RHI is applied to measure two important species, the NH and OH radicals. The measurements are performed in a laminar diffusion NH₃-O₂ flame stabilized over a Wolfhard-Parker slot burner. The burner facilitates the interpretation of the recorded interferograms since it provides a 2D flame structure. A near interefometrically stable
2D flame is achieved by providing top hat flow velocity profiles for the three burner slots and carefully controlling the total and the ratio of the flow velocities of fuel and oxidizer.

Generally, two lasers are required in order to generate the two colors, thus increasing the costs and the complexity of the system. In this work, we propose a novel scheme based on one laser only. The generation of the second color is achieved by optical phase-conjugation from Stimulated Brillouin Scattering (SBS) in a cell filled with n-hexane. Phase-conjugation is optimized to provide a high reflectivity of the SBS process and a high phase-conjugate fidelity. The frequency shift of ~8.5-9.0 GHz introduced by the SBS process in n-hexane, matches approximately the linewidth of many molecular transitions at typical flame temperatures. Phase-conjugate RHI (PC RHI) has been implemented to produce holograms of good contrast and diffraction efficiency for the first time to our knowledge.

Image plane holography is applied in order to permit white light read-out of the recorded holograms. Image plane holography has an additional advantage: The object plane of the imaging lens is carefully chosen to lay at the 2/3's of the flame-probe wave interaction length. At that position of the object plane, the fringe count may be considered in the straight line approximation, as if no refraction occurred. A single reference arm holographic set up is built due to energy constrains. Therefore, phase stepping techniques cannot be applied and a relatively low resolution fringe measuring technique is used.

Phase-conjugate RHI is applied for concentration measurements of the NH radical. It is shown that RHI can indeed provide species selective interferograms. Deformations on the phasefronts induced by the bulk refractive index, the steering optics, or the refractive index gradients are successfully canceled.

The experiments are repeated for the second target molecule, the OH radical. A different laser-system is used to address the resonant frequencies of OH. This laser does
not provide a beam that can be phase-conjugated successfully. Consequently for these experiments the phase-conjugate RHI set up cannot be applied. Strong intensity modulations of the expanded phase-conjugate beam profile prohibited 2D measurements. These modulations are attributed to the pulse characteristics. Consequently, a two laser system is employed for the RHI measurements on OH.

The experiments manifest the advantages of utilizing SBS to create the second color in comparison to the two laser system. The statistical errors, when applying the two laser system, increase dramatically. This is due to the fact that the two probe waves are not frequency locked in respect to each other, in contrary to SBS technique. The frequency jitter of the applied lasers increase the uncertainty of the measurement. The possibility of tuning one laser off the optimal frequency increases as well. Using SBS to create the second color features an additional advantage. Small phase deviations on the phasefront are canceled during the interferometric read-out, since the second beam is the phase-conjugate of the first.

A theory supporting this work is developed. The interaction of light waves with a transparent object is described. The model describes the propagation of a wave through a refractive index medium. It is shown that the wavefront of a probe beam is deformed by refractive index variations in the transparent medium. This is performed in terms of a parabolic trajectory for an individual ray pencil. It is also shown that this deformation includes the integration of the local refractive index along the path of the ray through the probe medium. Therefore, RHI will be a line-of-sight technique yielding integral information. The wavefronts are reconstructed by the ensemble of these rays.

A numerical model of the complex refractive index is developed. This model is extended to calculate the fringe shift for a two-color RHI experiment and the intensity attenuation. Two combustion related parameters are the determined in the model. The temperature and the probe species concentration.
For comparison, 2D absorption measurements are also applied on the NH$_3$-O$_2$ flame. The 2D absorption measurements are performed for the same flame conditions. CARS measurements provide absolute temperatures in the flame.

Using these temperatures, the fringe shift and the attenuation is converted by the model to the NH concentration. Unexpectedly, the absolute concentrations obtained by the two techniques differ by a factor of 2-4. RHI appears to overestimate the species concentration. It is also observed that the discrepancy is species concentration dependent. For a given absorption coefficient, a high species abundance is leading to a higher deviation of the results obtained by RHI in respect to absorption.

Trying to investigate the discrepancy between the two measuring techniques, three effects were studied. First, the finite fringes introduced to facilitate the interpretation of the RHI measurements. It was theoretically shown that the error introduced by the slightly longer path of the tilted beam is negligible. Second, the refraction of the beams caused by the refractive index gradients in the flame was theoretically investigated. The gradients from the temperature distribution and from the resonant refractive index were included in the model. Calculations showed that for the positions where the measurements are performed, i.e. the position of maximum species concentration, the effect is again negligible. Finally, it was assumed that the bulk refractive index as probed by the second probe pulse is changed by heat dissipation introduced by the high absorption during the passage of the first pulse. Experimental investigations could not verify any refractive index changes induced by the first probe pulse. Consequently, the discrepancy in measuring species concentrations by RHI and absorption have not been explained.

This work has left unanswered questions on respect to absolute concentration measurements of species in flames environments by RHI. On the other hand, a better understanding of RHI has been obtained by comparing it with a more reliable laboratory technique. Additionally, phase-conjugate waves have been applied to
holography for the first time to our knowledge. This has a beneficial effect on the applicability of RHI in many laboratories, since it significantly lowers the costs by eliminating one laser. Furthermore the two probe frequencies are locked in respect to each other, by the acoustic frequency. The experimental constrains of using two lasers are thus relaxed and more reliable results are obtained. Extension of RHI with heterodyne techniques will increase the sensitivity of the technique. The high sensitivity of RHI, the insensitivity to background disturbances and the high spatial resolution it offers can yield PC RHI competitive to more standard techniques such as PLIF.
Resonant Holographic Interferometry on NH and OH
Zusammenfassung


Der Brechungsindex eines Mediums kann in zwei Anteile aufgespalten werden: der erste, nichtresonante Anteil (n₀) hängt von der Temperatur und der Teilchendichte ab. Der zweite ist durch den komplexen Brechungsindex in der Nähe eines elektronischen Überganges eines Moleküls beschrieben. Dieser Brechungsindex wird in der vorliegenden Arbeit detailliert behandelt, wobei die spektroskopischen Daten berücksichtigt werden. Während sich die Realteile sich zu einer Phasenänderung addieren, führt der Imaginärteil zur Abschwächung des Messstrahles.

erfahren, zeigt das Interferenzmuster Verschiebungen.


„Image plane holography“ (IMH) kommt zum Einsatz, da die aufgenommenen Hologramme mit Weisslicht ausgelesen werden können. IMH hat einen weiteren Vorteil: die Objektebene der abbildenden Linse liegt bei 2/3 der Interaktionslänge des Probestrahles mit der Flamme. Es wird gezeigt, dass Brechungseffekte vernachlässigt werden können, d.h. der Messstrahl kann als gerade angenommen werden. Da die Energie der Laserpulse in unserem Experiment beschränkt ist, wird nur ein Referenzarm für die holographische Aufnahme verwendet. Auslesetechniken, welche auf aktive Phasenverschiebungen beruhen, sind daher ausgeschlossen. Die Auswertung erfolgt deshalb konventionell, was die Auflösung einschränkt.

Mit PC RHI werden die Konzentrationen der NH-Radikale gemessen und so wird
aufgezeigt, dass RHI tatsächlich speziesselktive Interferogramme liefert. Verzerrungen, verursacht durch Gradienten des Brechungsindexes und durch optische Elemente, werden erfolgreich ausgelöscht.


Ein numerisches Modell des komplexen Brechungsindexes wird entwickelt, welches es erlaubt, die Verschiebung des Beugungsmusters und die Lichtabsorption in einem zweifarbigen RHI-Experiment zu simulieren. Aus dem Vergleich mit dem Experiment kann damit die lokale Temperatur und die Konzentration des geprobenen Moleküls bestimmt werden. Diese Resultate werden mit unabhängigen Messungen überprüft,
welche unter denselben Bedingungen in der NH₃-O₂ Flamme durchgeführt werden: 2D-Absorptionsmessungen liefern die Spezieskonzentration; CARS-Messungen die Temperatur. Mit den gemessenen Temperaturen, der Verschiebung des Beugungsmusters und der Absorption berechnet das Modell die Molekülkonzentration. Die absoluten Spezieskonzentrationen gemessen mit den zwei verschiedenen Techniken unterscheiden sich um einen Faktor 2 bis 4, wobei RHI die Spezieskonzentration zu überschätzen scheint. Der Unterschied zwischen diesen beiden Methoden ist konzentrationsabhängig. Für einen gegebenen Absorptionskoeffizienten führt eine höhere Spezieskonzentration zu einer größeren Diskrepanz der gemessenen Resultate.


Diese Arbeit lässt Fragen bezüglich der Messgenauigkeit von RHI zur Bestimmung der absoluten Spezieskonzentration offen. Der hier durchgeführte Vergleich mit einer bestehenden Methode trägt jedoch zum besseren Verständnis der RHI-Technik bei.
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Introduction

Combustibles are the most versatile and readily available energy storage with a good energy to weight ratio. As a consequence 90% of the energy consumption is based on combustion. In spite of this fact, an insight of many areas of combustion science and technology has not been obtained. However, regarding the dependence on fossil fuels and the increasing constraints with respect to the environment, a detailed understanding of combustion processes is compulsory. A better knowledge of the combustion process will increase its efficiency and cleanliness and contribute to the development of new promising combustion schemes.

A multitude of interdependent parameters determines the performance of technical combustion systems. Mathematical models can help to find the optimal conditions of various combustion parameters. Experimental data can be directly used to improve the combustion process. More often, the development procedure is more indirect and experimental data obtained under controlled conditions, serve the validation of simulations models.

A limitation for obtaining valuable information is the lack of readily available measuring techniques. Two prime reasons are the hostility and delicacy of combustion processes. The high temperatures and heat transfer rates make physical probing with high spatial and temporal resolution a difficult task. Additionally combustion processes are easily altered by physical intrusion.

Laser diagnostic techniques have long been recognized as suitable combustion diagnostic tools overcoming these limitation due to the non intrusive nature of probing the combustion processes. Such laser based techniques can provide in situ, real-time information on technical combustion processes and furnish data for a thorough validation of current theoretical models. They have replaced more traditional probe methods due to their intrinsic high temporal and spatial resolution, along with a high species selectivity and sensitivity.
The understanding of the chemical processes are of vital importance. For parameters such as ignition, heat release, quenching or pollutant formation, the measurement of temperature and of species, radicals and intermediate products involved in the chemical process is necessary. They can be measured by applying established techniques such as Raman, laser absorption, laser-induced fluorescence, [Refs. 1, 2], or other emerging techniques such as Four-wave-mixing [Ref. 3], stimulated emission pumping [Ref. 4] or multi-photon spectroscopy. Coherent Antistokes Raman Scattering (CARS) is widely applied for Temperature measurements [Refs. 2, 5].

Holography has also been proposed as a diagnostic tool in combustion environments. Two-color Holographic Interferometry was used to simultaneously measure the heat and mass transfer on a fuel droplet [Ref. 6]. Taking advantage of the high resolution abilities of Heterodyne Holographic Interferometry, the temperature and species concentration was measured in natural convection boundary layers [Ref. 7]. In general, in order to apply Holographic Interferometry assumptions are required on the composition of the probed combustion environment, since the technique measure the refractive index that itself is a function of all combustion components. Therefore a self consistent holographic technique exploiting the resonant refractivity, i.e. the change of the refractive index close to a spectral line, was suggested for enhancing visualization of flow phenomena [Ref. 8] and selectively measuring changes in the concentration of species [Ref. 9].

It wasn't long after that Resonant Interferometry combined with holography was suggested by Dreiden et al [Ref. 10] for plasma diagnostics determining atomic and electron densities. Sirota and Christiansen [Ref. 11] proposed Resonant Holographic Interferometry (RHI) as a laser diagnostic technique for obtaining full-field species-selective measurements in combustion environments.

RHI can provide a background-free, species-selective hologram by recording two simultaneous holograms at two different laser wavelengths: one tuned in the proximity
of a resonant transition of a target species and the other tuned off any resonance. Since
the relative difference in the index of refraction between these two wavelengths is
significant, the phase difference between these two wavefronts, after passing through
the medium containing the target species, will be non-zero. Reading-out the holograms
forms an interference pattern of fringes. This pattern can be directly attributed to the
target species density since all phase contributions from background species, thermal
and pressure gradients or optical aberrations from the steering optics will be
interferometrically subtracted out during the reconstruction. Sirota and Christiansen
also showed that RHI can be used for the measurement of flow parameters such as
velocity, pressure and temperature.

RHI has been gaining attention due to the fact that in contrast to laser induced
fluorescence it is not subject to excited state redistribution phenomena such as
quenching since the species is probed in the ground state level. RHI is not only
insensitive to effects from thermal and pressure gradients or high background
luminosity and scattering but offers some significant advantages over absorption
spectroscopy, both in sensitivity and in data-handling capability. Combination of RHI
with tomographic techniques can provide three-dimensional characterization of the
index of refraction which can be converted to species density. The utilization of RHI for
hydroxyl density measurements in simple laboratory flames and in a reactive flow
produced in a shock tube facility has been reported [Refs. 12, 13].

The scope of this work is to present a detailed investigation of the RHI technique as a
diagnostic tool for species concentration measurements in combustion environments.
First the underlying theory is presented. The interaction of light wavefronts with a
transparent medium is described in terms of the deformation of the phasefronts of the
probe light by the refractive index of the transparent medium. Since a wavefront can be
considered as an ensemble of individual light rays, the influence of the refractive index
gradients on an light ray is described.
As absorption, RHI is a ‘line of sight’ technique. The information is integrated along the probe path. This has the consequence that the hologram recorded does not contain 3D information of the species density, but rather the integration along a path.

The resonant refractivity is theoretically derived for the case where the target species is probed in a flame, taking into account relevant parameters of the flame. The resonant refractivity is complex, described by the Voigt profiles. The real part describes the contribution of resonance to the refractive index. The imaginary part is related to the absorption taking place at resonant frequencies. The resonant refractive index is a function of known spectroscopic parameters but primarily of two combustion-related parameters, the temperature of the flame and the density of the probed species. The refractive index and the absorption are interrelated, and can be individually measured, the first by RHI and the second by standard absorption techniques. A system is therefore proposed to derive both parameters, the temperature and the density of the species.

An experimental set up is built to perform two-color RHI. The need of two colors necessitates the implementation of two tunable dye lasers. Utilization though of Stimulated Brillouin Scattering (SBS) enable us to create a second, phase-conjugate to the first, beam. The frequency difference introduced by the SBS process matches ideally typical widths of resonant transitions of interesting species in flames, thus allowing an optimal utilization of the RHI technique.

Taking advantage of basic features of this set up, 2D absorption measurements are performed by recording the wavefront at the hologram plane. In this way a direct comparison of the two methods can be made. Using temperature measurements performed with the transportable CARS system developed at Paul Scherrer Institute, the species concentrations arriving from both measuring techniques are compared. The calculations are performed using a numerical approximation to the Voigt profiles. The problems in correctly evaluating the obtained data are discussed and simulation
programs are developed in order to investigate the influence of the refractive index gradients on the propagation of the probe wavefronts through the flame.

Finally, the results are discussed and conclusions are made concerning the applicability of RHI as a method for obtaining species concentration measurements. The advantages and disadvantages of implementing RHI are discussed. Schemes are described that increase the resolution of RHI by implementation of phase stepping techniques. Additionally an experimental set up for performing simultaneously RHI and 2D absorption measurements is presented.
Theory

In the following chapter the interaction of light with matter will be presented. The description is focused on resonant holographic interferometry, and on the characteristics of the subject of investigation, the flame.

Basic properties of the EM wave

From EM theory we know that the wave-number is defined as [Ref. 14, p. 16]

$$k_0 = \frac{2\pi}{\lambda_0} = \frac{\omega}{c}$$

in vacuum and in a medium

$$k = \frac{2\pi}{\lambda} = \frac{n\omega}{c} = \frac{\omega}{\nu}.$$

$\omega$ is the angular frequency of the wave, $\lambda$ the wavelength, $c$ the speed of light in vacuum. The subscript 0 (zero) denotes values for the vacuum, i.e. no medium present. The vacuum velocity of light is defined as $c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$ and $n = \sqrt{\varepsilon \mu}$ denotes the refractive index of a medium with $\varepsilon$ and $\mu$ the dielectric constant and the magnetic permeability respectively. Since for most substances $\mu=1$, we get that $\varepsilon = n^2$. The local propagation velocity of light in a medium is given by $\nu = c_0/n$.

A solution to Maxwell’s wave equation is a plane polarized transverse wave [Ref.15, p. 88]

$$\vec{E}(\vec{r}, t) = \vec{E}_0 \exp\{i(\omega t - \vec{k} \cdot \vec{r})\} \hspace{1em} \text{and} \hspace{1em} \vec{H}(\vec{r}, t) = \vec{H}_0 \exp\{i(\omega t - \vec{k} \cdot \vec{r})\},$$

with $\vec{r}$ the position vector and $\vec{k}$ the propagation wave-vector, $\vec{E}(\vec{r}, t)$ and $\vec{H}(\vec{r}, t)$ are the electric and magnetic field vectors respectively. They are mutually perpendicular and lie in a plane normal to $\vec{k}$: $\vec{E} \cdot \vec{H} = 0$, $\vec{E} \cdot \vec{k} = 0$ and $\vec{H} \cdot \vec{k} = 0$. The unit propagation wave-vector $\vec{s}$ is defined as $\vec{k} = k\vec{s}$ and therefore [Ref.14, p.14-17]

$$\vec{E}(\vec{r}, t) = \vec{E}_0 \exp\{i(\omega t - k\vec{s} \cdot \vec{r})\} \hspace{1em} \text{(1)}$$

Equation (1) describes a plane wave since for each instant $\vec{E}(\vec{r}, t)$ is constant over each of the planes $\vec{s} \cdot \vec{r} = \text{constant}$. These planes are called wavefronts, surfaces of constant phase (iso-phases).
For a more general type of fields we can write [Ref. 14, p. 111-112]
\[ \hat{E}(\vec{r},t) = \hat{E}_0 \exp[i(\omega t - k_0 \Phi(\vec{r}))], \]
where \( \Phi(\vec{r}) \), the optical path, is a real scalar function of position and obeys the eiconal equation:

\[ (\nabla \Phi(\vec{r}))^2 = n^2. \]

Again the surfaces \( \Phi(\vec{r}) = \text{constant} \) are called the geometrical wavefronts.

The above illustrates the importance of the refractive index on the propagation of EM waves. For a homogeneous medium, \( n = \text{constant} \), it follows that the local light velocity \( v \) remains constant along the trajectory and consequently \( k \) as well. The wavefronts propagate unchanged through the medium. For the case of a non-homogeneous medium, the wavefronts of the propagating light will be distorted since the light wave, depending on the local refractive index, will propagate with different velocities.

Light rays

Figure 1: Illustrating the relation between wavefronts and rays.
It is convenient here to introduce the concept of light rays in order to describe the effect of refractive index inhomogeneities on the propagation of light. A light ray is defined as the orthogonal trajectory to the geometrical wavefronts $\Phi(\vec{r}) = \text{constant}$. We consider them as oriented curves whose direction coincides everywhere with the unit propagation vector $\vec{s} = \nabla\Phi(\vec{r})/|\nabla\Phi(\vec{r})|$. If $\vec{r} (\vec{s})$ denotes the position vector of a point $P$ on a ray, for an illustration see Figure 1, with $s$ the travelled distance along the trajectory, then $d\vec{r}/ds = \vec{s}$ and the equation of the ray may be written as [Ref. 14, p. 121-124]

$$n \frac{d\vec{r}}{ds} = \nabla\Phi(\vec{r}) \tag{2}$$

The meaning of this equation may be made clearer considering two neighboring wavefronts $\Phi(\vec{r}) = \text{constant}$ and $\Phi(\vec{r}) + d\Phi(\vec{r}) = \text{constant}$. Then

$$\frac{d\Phi(\vec{r})}{ds} = \frac{d\vec{r}}{ds} \nabla\Phi(\vec{r}) = n \tag{3}$$

Hence the distance between the points where a normal intersects the two wavefronts is inversely proportional to the refractive index, i.e. directly proportional to the local light velocity $v$.

The integral $\int nds$ along a curve $Q$ is called the optical path length of the curve. Denoting by square brackets the optical length of the ray which joins points $P_1$ and $P_2$ we have

$$[P_1P_2] = \int_{P_1}^{P_2} nds = \Phi(P_2) - \Phi(P_1) \tag{4}$$

These equations specifies the rays by means of the function $\Phi(\vec{r})$. They can be transformed into a differential equation which specifies the rays directly in terms of the refractive index function $n(\vec{r})$. Differentiating equation (2) with respect to $s$ we obtain,

$$\frac{d}{ds} \left( n \frac{d\vec{r}}{ds} \right) = \frac{d}{ds} \nabla\Phi(\vec{r})$$

and after some calculations we obtain the vector form of the differential equation of a light ray
For a homogeneous medium $Vn = 0$, the light rays have the form of straight lines, since a solution of equation (5) has a form $\vec{r}(s) = s\vec{a} + \vec{b}$, $\vec{a}$ and $\vec{b}$ being constant vectors. In non-homogeneous media it is more difficult to find solutions for the ray trajectories [Ref. 16, p. 2-3].

However, under some circumstances certain simplifications are possible. The simplest case of a non-homogeneous medium, is the case in which the refractive index depends only on one co-ordinate. This is the so called stratified medium. Figure 2 shows the situation where the refractive index varies along the $y$-axis. The light rays enter the medium in the direction normal to the refractive index gradient, i.e. along the $z$-axis.

Figure 2: Refraction in a stratified medium $n = n(y)$. The stratified medium is confined in $z \in [0..L]$, with a surrounding refractive index of $n_0$. The beam enters the stratified medium parallel to the $z$-axis at a height of $y_e$.

A light ray incident at $(0, y_e)$ denoted by $y(z, y_e)$ lies in a plane formed by its initial propagation direction and the direction of the refractive index variation. In this case the general equation (5) simplifies to:
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\[
\begin{align*}
\frac{d}{ds}\left\{ n(y) \frac{dy}{ds} \right\} &= \frac{dn}{dy} \\
\frac{d}{ds}\left\{ n(y) \frac{dz}{ds} \right\} &= 0 
\end{align*}
\] (6)

It follows from Figure 2 that \( ds = \sqrt{1 + \left( \frac{dy}{dz} \right)^2} \) dz. Combination of these two equations gives

\[
\frac{y'' dy}{1 + y'^2} = \frac{dn}{n(y)}
\] (7)

The prime denotes differentiation with respect to \( z \). Using the boundary conditions of the ray trajectory \( y(z, y_e) \) at \( z = 0 \): \( y(0, y_e) = y_e \), \( y'(0, y_e) = 0 \) and \( n(y_e) = n_e \), we can integrate the second order differential equation to yield:

\[
1 + y'^2 = \left( \frac{n}{n_e} \right)^2
\] (8)

The exact knowledge of the refractive index distribution would allow us to calculate the propagation of each individual ray through this inhomogeneous medium and that consequently enabling the prediction of the deformations of the wavefronts.

A wave propagating through a inhomogeneous medium will experience locally different refractive indices. Each ray will register the passage through the medium in two ways: The actual passage through the medium, i.e. the optical path length will be an “integration of the refractive index along its path” as shown in equation (4) since the ray will travel with different velocities through the medium. Additionally the path will be deformed due to the refractive index gradients. The wavefront can be considered as an ensemble of individual rays. Describing the deformation of individual rays describes therefore the deformation of the phasefronts \( \Phi(\mathbf{r}) \) as they propagate through a phase medium, \( n = n(y) \).

To assess information on an inhomogeneous medium, an experimental technique is requested that can detect these deformations of the wavefronts caused by the medium. The technique used in this work is holographic interferometry. In the following a conceptual description will be given to illustrate the method.
Holography

Holography is a well established technique used to record and reproduce a true three-dimensional image [Ref. 17]. Holography differentiates itself from classical photography by its ability to record and reproduce the complete wave-field, i.e. the intensity and the phase of the light waves 'scattered' by the object. The information are usually recorded on a photographic emulsion. Since these media respond to radiation intensity, it is necessary to convert the phase information, that contain the information of the refractive index distribution, into variations of intensity. This is done by using coherent illumination and adding a reference wave to the wave scattered by the object, see Figure 3.

The interference pattern of the two waves is recorded on the emulsion and the intensity at each point of this pattern depends on both the intensity and the phase of the original object wave. This encoded pattern can reproduce the original object wave by illuminating it with the reference wave, see Figure 3. Actually, the hologram will not only reproduce the original object wave but three other waves as well. In this work, only
the recording and subsequent reconstruction of the original object wave is of interest. For a more detailed analysis of the holographic process, the reader is referred to the detailed work of P. Hariharan [Ref. 17] or R. R. A. Syms [Ref. 18].

In the following the method is described in more detail for the case of plane waves, without loss of the generality. Below, the utilisation of volume transmission holograms will be described [Ref. 17, p. 41-62]. The volume nature of the hologram can be justified when considering the hologram to have an emulsion thickness of hundreds of micrometers, i.e. orders of magnitude bigger than the characteristic parameters of holography such as the wavelength of the light or the spacing of the recorded interference pattern [Ref. 18, p. 21-27 and p. 35-64].

Consider two plane monochromatic waves as formulated in equation (1), originating from the same source, impinging on a holographic emulsion as illustrated on Figure 4. The first wave is assumed carrying the information of the object and the second is the reference wave. Both are defined by their wave-vectors $\mathbf{k}_o$ and $\mathbf{k}_R$. If $\mathbf{r}$ is a position in the hologram volume the registered intensity is given by (the asterisk * depicts the phase-conjugate wave):

$$I(\mathbf{r}) = |E_o(\mathbf{r}) + E_R(\mathbf{r})|^2 = |E_o(\mathbf{r})|^2 + |E_R(\mathbf{r})|^2 + \bar{E}_R^*(\mathbf{r})E_o(\mathbf{r}) + \bar{E}_o^*(\mathbf{r})E_R(\mathbf{r})$$

, or analytically

$$I(\mathbf{r}) = |E_o|^2 + |E_R|^2 + 2|E_o||E_R|\cos((\mathbf{k}_o - \mathbf{k}_R) \cdot \mathbf{r})$$

Integration over the exposure time/pulse duration averages out the time dependent part of the two waves. The so-called grating-vector $\mathbf{K} = \mathbf{k}_o - \mathbf{k}_R$ determines not only the orientation of the interference surfaces, since it is normal to them, but the distance between two of them as well ($\lambda_0$ the recording wavelength):

$$\mathbf{K} \cdot d = 2\pi \Rightarrow d = \frac{\lambda_0}{2 \sin \frac{\theta}{2}}$$
Figure 4: Vector presentation of the recording of a hologram. The two interfering waves represented by the wave-vectors $\mathbf{k}_O$ and $\mathbf{k}_R$ create an interference pattern with a spacing $d$ between the interference surfaces. Their orientation in the emulsion is defined by the half interference angle $\psi = \theta/2$.

The interference pattern must be transformed into a variation of transmittivity or refractive index of the photographic emulsion. This is obtained by chemical processing the holographic emulsion in a similar manner as in photography. If it is assumed that the amplitude transmittance of the emulsion, after chemical processing, has a linear

$$
\mathbf{K} = \mathbf{k}_O - \mathbf{k}_R
$$

$$
k_O = k_R = \frac{2\pi}{\lambda_0}
$$

$$
K = 4\pi \frac{\sin(\theta/2)}{\lambda_0}
$$
relation to the recorded interference pattern, the amplitude transmittance can be written as:

\[ t(\vec{r}) = t_0 + \beta I(\vec{r}) = t_0 + \beta \left( |\vec{E}_o(\vec{r})|^2 + |\vec{E}_r(\vec{r})|^2 + \vec{E}_r^*(\vec{r})\vec{E}_o^*(\vec{r}) + \vec{E}_r(\vec{r})\vec{E}_o(\vec{r}) \right) \]

Illuminating the developed hologram with a read-out wave, depicted by its wave-vector \( \vec{k}_{RO} \) we obtain two interesting for us images:

\[ \vec{E}_{im}(\vec{r}) = \vec{E}_{RO}(\vec{r})t(\vec{r}) \]

\[ \approx \left\{ \begin{aligned} & \vec{E}_{RO}(\vec{r})\vec{E}_r^*(\vec{r})\vec{E}_o^*(\vec{r}) = |\vec{E}_{RO}(\vec{r})\vec{E}_r(\vec{r})\vec{E}_o(\vec{r})| \exp\left[ i(\vec{k}_{RO} + \vec{K}) \cdot \vec{r} \right] \\ & \vec{E}_{RO}(\vec{r})\vec{E}_r(\vec{r})\vec{E}_o(\vec{r}) = |\vec{E}_{RO}(\vec{r})\vec{E}_r(\vec{r})\vec{E}_o(\vec{r})| \exp\left[ i(\vec{k}_{RO} - \vec{K}) \cdot \vec{r} \right] \end{aligned} \]  

(11)

The first image is the reconstruction of the original object wave while the second is the phase-conjugate wave of the object wave. The object wave is reconstructed if \( \vec{k}_{im} = \vec{k}_{RO} \pm \vec{K} = \vec{k}_{RO} \pm \vec{k}_o + \vec{k}_r \). This is the Laue-condition, known from x-ray diffraction experiments, for constructive interference of a spatially periodic structure described by the grating vector \( \vec{K} \). The Laue-condition is equivalent to Bragg’s condition for constructive interference of the reflections from several layers. Taking into account that the recorded holograms are thick volume holograms, re-illumination with a read-out wave will reconstruct only one of the two waves: the original object wave or its phase-conjugate, depending on the illumination angle. In Figure 5, the reconstruction scheme of the original object wave is shown. We get directly Bragg’s condition:

\[ \frac{2\pi}{\lambda_{RO}} \sin \alpha = \frac{K}{2} = \frac{\pi}{d} \Rightarrow 2d \sin \alpha = \lambda_{RO} \]

Combination of this equation with equation (10) results in:

\[ \sin \alpha = \frac{\lambda_{RO}}{\lambda_0} \sin \frac{\theta}{2} \]

Thus for \( \lambda_{RO} = \lambda_0 \) we obtain a perfect reconstruction of the original object wave, since \( \vec{k}_{im} = \pm \vec{k}_o \).
The wave can be considered as an ensemble of light rays. Each individual ray is described by equation (8) relating the position and propagation vectors \( \vec{r} \) and \( \vec{s} \). Since the wave-vector is defined as \( \vec{k} = k \vec{s} \), we see that the hologram will record and reconstruct each of the individual rays and consequently the whole distorted wavefront.

![Diagram](image)

**Figure 5:** Vector presentation of the reconstruction of a hologram. The wave defined by its wave-vector \( \vec{k}_{RO} \) is reflected on the interference pattern. With angles satisfying Bragg's condition the image wave, \( \vec{k}_{im} \), will emerge from the hologram.
Holographic Interferometry

In general we can define Holographic Interferometry (HI) as the interferometric comparison of two or more waves, where at least one of them is holographically reconstructed. The composite of these two or more waves will be referred as a holographic interferogram. The term interferogram denotes a pattern of interference fringes recorded on photographic film or formed on a two-dimensional viewing screen or the retina of the eye. A detailed discussion on the subject can be found in [Ref. 19, Ref. 20]. Worth noting is the fact that, in contrary to classical Interferometry, HI can compare two wavefronts of different wavelength or that interacted with the test object at different times, thus permitting the comparison of two states of the same object. Here, the description of holographic interferometry is presented following the outlines given above and with special attention to the investigation of inhomogeneous refractive index distributions.

We consider the case where the holographic recording is performed twice on the same holographic plate [Ref. 18, p.194-222, Ref. 21, 22]. After development of the holographic plate two grating-vectors will define the two holograms recorded as shown on Figure 6. In Figure 6 the two grating-vectors are depicted with slightly different directions to illustrate a change between the two holographic exposures.

Upon illumination of the hologram with a read-out beam \( \vec{k}_R \), two waves evolve each in angles that satisfy the Bragg’s condition.

\[
\tilde{E}_{im1}(\vec{r}) = E_{im1}(\vec{r}) \exp[i(\vec{k}_{im1} \cdot \vec{r})], \quad \text{and} \\
\tilde{E}_{im2}(\vec{r}) = E_{im2}(\vec{r}) \exp[i(\vec{k}_{im2} \cdot \vec{r})]
\]

Their coherent read-out will result in their interference:

\[
I_{int}(\vec{r}) = \left| E_{im1}(\vec{r}) \exp[i(\vec{k}_{im1} \cdot \vec{r})] + E_{im2}(\vec{r}) \exp[i(\vec{k}_{im2} \cdot \vec{r})] \right|^2
\]

\[
= \left| E_{im1}(\vec{r}) \right|^2 + \left| E_{im2}(\vec{r}) \right|^2 + 2 \left| E_{im1}(\vec{r}) \right| \left| E_{im2}(\vec{r}) \right| \cos(\vec{K}_{int} \cdot \vec{r})
\]

\( \vec{K}_{int} = \vec{k}_{im1} - \vec{k}_{im2} \) and the subscript ‘int’ denoting the interferogram created. Equations
(9)-(12) describe the case of recording and reconstructing two plane wavefronts. The description can be generalized to the case of arbitrary fields of the type

\[ \tilde{\mathbf{E}}_i (\mathbf{r}, t) = \tilde{e}_i (\mathbf{r}) \exp[i(\omega t - \mathbf{k}_0 \cdot \Phi (\mathbf{r})]. \]

\[ k_{im} = \mathbf{k}_R O + K_i \]

\[ d_i = \frac{\lambda_0}{2 \sin(\theta_i/2)} \]

\[ k_{im_1} = k_{RO} = \frac{2 \pi}{\lambda_{RO}} \]

\[ K_i = \frac{2 \pi}{d_i} \]

\[ \mathbf{K}_1 = \mathbf{K}_2 \]

\[ Hologram \]

Figure 6: Vector presentation of the reconstruction of a holographic interferogram. The wave defined by its wave-vector \( \mathbf{k}_{RO} \) is reflected on the two interference patterns. With angles satisfying Bragg’s condition for each case the two image waves, \( \mathbf{k}_{im_1} \) and \( \mathbf{k}_{im_2} \), will emerge from the hologram.

A double exposure and subsequent reconstruction with two general-field object-waves would yield an interferogram with an intensity distribution of

\[ I_{im} (\mathbf{r}) = |e_1|^2 + |e_2|^2 + 2|e_1| |e_2| \cos \left( k_{02} \Phi_2 (\mathbf{r}) - k_{01} \Phi_1 (\mathbf{r}) \right) \]

(13)

Observing that the optical path length is defined by equation (4) as
\[ [P_1P_2] = \int_{P_1}^{P_2} n ds = \Phi(P_2) - \Phi(P_1), \] the accumulated phase from \( P_1 \) to \( P_2 \) is

\[ \Delta \Phi(\bar{r}) = \int_{P_1}^{P_2} n(\bar{r}) ds. \]

That the intensity distribution therefore is a function of the difference of the optical path length between the pair of rays and equation (13) becomes

\[ I_{in}(\bar{r}) = |e_1|^2 + |e_2|^2 + 2|e_1| |e_2| \cos \left( k_{02} \int_{P_1}^{P_2} n_2(\bar{r}) ds - k_{01} \int_{P_1}^{P_2} n_1(\bar{r}) ds \right) \] (14)

In the case where no ray bending takes place and the rays propagate in straight lines the integrals reduce to products and we get, \([P_1P_2] = L:\)

\[ I_{in}(\bar{r}) = |e_1|^2 + |e_2|^2 + 2|e_1| |e_2| \cos \left( k_{02} n_2(\bar{r}) - k_{01} n_1(\bar{r}) \right) L \] (15)

If the two waves experience a difference in refractive index, \( n(\omega_1) \neq n(\omega_2) \), the interferogram will bare this difference in the form of fringes.

In the following a flame in described as a medium with variable refractive index distribution.
Refractive Index and Flames

In a flame where heat and mass transfer take place, the refractive index, \( n \), is related to the local density \( \rho \) by the Gladstone-Dale equation [Ref. 19, p. 344-346, Ref. 23, p. 116-120]:

\[
n - 1 = K \rho
\]

where \( K \), the Gladstone-Dale constant can be calculated as a mass-weighted average of the values of \( K \) for the component gases of the flame:

\[
K = \sum \alpha_i K_i,
\]

where \( \alpha_i \) is the mass fraction and \( K_i \) the Gladstone-Dale constant of the \( i^{th} \) component. The Gladstone-Dale constant is a slow varying function of wavelength and is nearly independent of temperature and pressure under moderate physical conditions. Therefore \( n \) is in essence only dependent on density and gas composition which are a function of the local temperature and local chemical reactions. A flame has normally a three-dimensional structure. Therefore the refractive index has a three-dimensional structure as well. The phase recorded on a hologram is in essence the integration of the refractive index along the wave's path. The hologram records a "projection" of the three-dimensional refractive index. Plainly speaking we lose information on one dimension.

This work is concentrated on holographic interferometry for obtaining quantitative information on the distribution of one flame species. The refractive index as described above, cannot be used to identify an individual species, since its information is buried in the sum of contributions of all species. Utilizing holographic interferometry a one equation, \( i \) unknowns system is obtained. Additional measurements are required to solve the system.

Some additional information is required that will be directly related to the specific component under investigation. The preceding analysis of the refractive index does not account for the range of probe frequencies near a molecular transition of one of the components of the flame. Till now the refractive index has been considered a real function. This is indeed the case for non resonant frequencies. In the proximity of a
molecular transition the imaginary part of refractive index becomes significant. The imaginary part describes the absorption of the transmitted light in the flame. Additionally, when tuning over a molecular transition, the real part of the refractive index will increase rapidly with frequency then decrease below the average value for frequencies above resonance. This decrease of the refractive index in the vicinity of a molecular resonance is termed "anomalous dispersion". When operating in the region of a transition of a molecule, in this case one of the species of the flame, the frequency dependence of the real part of the refractive index can be exploited to identify the probed flame species. Below the complex refractive index is derived for resonant frequencies.

The classical model of the Complex Refractive Index

A system of an assembly of bound electrons driven by an electric field, can be considered as a damped, forced oscillation. Once again plane-polarized EM wave propagating in the z-direction is considered. The equation of electron motion around the nucleus can be written as [Ref. 2, 24]

$$m_e \frac{d^2 x}{dt^2} + m_e \gamma \frac{dx}{dt} - eE_0 \exp(i\omega t) - m_e \omega_0^2 x = 0$$

x defines the displacement of electrons in the x-direction, $m_e$ and $e$ is the electron mass and electron charge respectively, $\gamma$ a damping constant, $\omega_0$ the natural circular frequency of oscillation and the magnetic permeability $\mu$ is set equal to one, $\mu=1$.

Assuming a solution of the form $x = x_0 \exp(i\omega t)$, $x_0$ can be shown to be

$$x_0 = \frac{eE_0}{m_e (\omega_0^2 - \omega^2 + i\gamma \omega)}.$$  

The induced polarization is the dipole moment per unit volume, thus for the ensemble of $N_e$ bound electrons the polarization is $\bar{P} = N_e e \bar{x}_0$. From Maxwell’s equations we have that the polarization is
\[ \vec{P} = \varepsilon_0 (\varepsilon - 1) \vec{E}. \] Since \( \varepsilon = n^2 \) we get that \( n^2 = 1 + \frac{\varepsilon N_e}{\varepsilon_m c_m (\omega_0^2 - \omega^2 + i\gamma \omega)} \).

Separating the real and imaginary part of the complex refractive index \( n = 1 + n(\omega) - i\kappa(\omega) \), we get after some rearrangements that:

\[
\kappa(\omega) = \frac{N_e e^2}{4 \varepsilon_0 m_0 \omega_0} \frac{\gamma/2}{(\omega_0 - \omega)^2 + \left(\frac{\gamma}{2}\right)^2}
\]

and

\[
n(\omega) = \frac{N_e e^2}{4 \varepsilon_0 m_0 \omega_0} \frac{\omega_0 - \omega}{(\omega_0 - \omega)^2 + \left(\frac{\gamma}{2}\right)^2}
\]

Figure 7 show the frequency dependence of \( \kappa(\omega) \) and \( n(\omega) \) in the vicinity of an eigenfrequency \( \omega_0 \) of an atomic transition.

**Figure 7:** The complex refractive index as a function of frequency in the vicinity of a molecular transition, \( n = 1 + n(\omega) - i\kappa(\omega) \). \( \gamma \) is the FWHM of the \( \kappa(\omega) \) curve defining the two turning points of the dispersive curve \( n(\omega) \).
Spectroscopic Considerations

*The oscillator strength*

Because of the many possible energy levels, molecules exhibit many eigenfrequencies at which they can absorb radiation, starting from an initial level $E_i$. The total absorption of the atom in the level $E_i$ is distributed among all possible transitions $E_i \rightarrow E_k$ to all higher levels $E_k$ which are optically connected with $E_i$. Each of these transitions contributes only a fraction $f_{ik}$ to the total absorption. This number $f_{ik} < 1$ is called the oscillator strength or transition probability, since in essence it measures the likelihood that a given transition will occur. The absorption of $N$ atoms on the transition $E_i \rightarrow E_k$ is equal to that of $f_{ik} N$ classical oscillators. The absorption oscillator strength can be related to the Einstein $A_{ki}$ coefficient of spontaneous emission by

$$f_{ik} = \frac{g_k}{g_i} \frac{2\pi e^2 m c^3}{\alpha_i^2 c^2} A_{ki}$$  \hspace{1cm} (18)$$

where $g_i$ are the degeneracy factors of the two energy levels [Ref. 25].

*The partition function*

The molecules can attain many energy states by vibration, rotation and electronic excitation. It is therefore important to take into consideration the partition of the ensemble of molecules over these states. The distribution derives from statistical mechanics and is described by the Boltzmann equation

$$N_j = N \frac{g_j \exp \left(- \frac{E_j}{k_B T} \right)}{\sum_j g_j \exp \left(- \frac{E_j}{k_B T} \right)}$$  \hspace{1cm} (19)$$

where $N_j$ is the number density of particles in the $j$th state of energy $E_j$, $N$ the total number density, $k_B$ the Boltzmann's constant, $T$ the temperature and $g_j$ the degeneracy of the state. The denominator is termed the partition function and is denoted by $Q$. 
The internal molecular energy can be written as a sum of the electronic, vibrational and rotational energies \( E_j = E_{el} + E_{vib} + E_{rot} \) and the total partition function can be expressed as the product of a partition function of each internal energy mode [Ref. 2].

The total population distribution factor \( F \) can thus be described as

\[
F = \frac{1}{Q_e} \exp\left(-\frac{\nu \hbar \omega_e}{k_B T}\right) \left(1 - \exp\left(-\frac{\hbar \omega_e}{k_B T}\right)\right) \frac{g_J (2J + 1) \exp\left(-\frac{BJ(J+1)\hbar}{k_B T}\right)}{Q_{rot}}
\]  

(20)

where

\[
Q_{rot} = \sum_{J=0}^{\infty} (2J + 1) \exp\left(-\frac{\hbar BJ(J+1)}{k_B T}\right).
\]

\( Q_e \) is the electronic partition, \( \nu \) the vibrational quantum number, \( \omega_e \) the molecular vibration frequency (cm\(^{-1}\)), \( J \) the rotational angular momentum quantum number, \( B \) the molecular rotation constant (cm\(^{-1}\)), \( g_J \) the nuclear spin degeneracy and \( \hbar \) Planck’s constant. The number of molecules taking part in the interaction is therefore \( N_J = FN_{tot} \), where \( N_{tot} \) are the molecules in the probed sample.

**Frequency broadening of a transition line**

In the formulation of the complex refractive index \( \gamma \) was introduced as a damping parameter. This frequency spread is termed natural linewidth and is usually negligible in compare with other broadening effects.

Spectral lines are broadened, and in many cases shifted, by increasing pressure and by the presence of ions and electrons. These perturbations -widening, shifting and mixing of energy levels, are due to interactions of the emitting or absorbing molecules with other particles. The form of the pressure broadened line is the known Lorentzian line shape, shown in Figure 7. So far, the natural and the collisional broadening can be considered as two contributions to the damping parameter \( \gamma \), \( \gamma = \gamma_{nat} + \gamma_{col} \).

The line will also broaden due to the thermal motion of the molecules. The movement
of the molecules gives rise to a Doppler shift. For a given temperature the distribution of velocities is described by the Maxwell-Boltzmann expression. The Doppler line shape can be derived by calculating the Doppler shift for each velocity component and averaging over the distribution of velocities. When the resonance frequency $\omega_0$ of the absorbing molecular transition has a homogeneous linewidth of $\delta\omega$ the velocities of the absorbing molecules must fall into the interval $\Delta u$ defined by $\omega_0 - k \cdot (u_z \pm \Delta u_z)$. In this expression the wave-vector $k$ is assumed parallel to the z-axis. At Temperature $T$, the number of molecules $n_i(u_z)du_z$ in the probe level $E_i$ per unit volume with a velocity component between $u_z$ and $u_z + du_z$ is

$$n_i(u_z)du_z = \frac{N_i}{v_p} \sqrt{\pi} \exp\left[-\left(\frac{u_z}{v_p}\right)^2\right]du_z,$$

where $N_i = \int n_i(u_z)du_z$ is the density of all molecules in level $E_i$, $v_p = \sqrt{2k_B T / M}$ the most probable velocity, $M$ the mass of the molecule and $k_B$ Boltzmann's constant. The number of molecules with absorption frequencies shifted from $\omega_0$ into the interval $[\omega, \omega + \delta\omega]$ is

$$n_i(\omega)d\omega = \frac{N_i}{v_p} \sqrt{\pi} \exp\left[-\left(\frac{c}{v_p} \left(\frac{\omega - \omega_0}{\omega_0}\right) \right)^2\right]d\omega.$$

The FWHM of a Doppler broadened line can be calculated to

$$\Delta \omega_D = \frac{2\omega_0 c}{\sqrt{2k_B T \ln 2 / M}}.$$

**Saturation considerations**

In the process of exiting the atoms from level $E_i$ to $E_k$ consideration has to be taken on the intensity of the probe wave. The population densities $N_i$ and $N_k$ of the two levels $E_i$ and $E_k$ are given by the Boltzmann distribution. They can be regarded as constant as long as they are not noticeably influenced by the interaction with the electric field. The absorbed intensity $dI$ is proportional to the incident intensity $I$. But as the intensity increases the lower state density $N_i$ can noticeably decrease while the upper $N_k$ increases. The absorbed intensity $dI$ is then no longer proportional to the incident intensity $I$ and the transition is saturated. It can be shown [Ref.24, p. 44] that the population difference in this case is

$$\Delta N = \frac{\Delta N_0}{\sqrt{\left(1 + I/I_{sat}\right)}},$$

where $\Delta N_0$ is the zero-
intensity population difference and $I_{\text{sat}} = cR/B_{12}$ stands for the intensity required to decrease $\Delta N$ to $\Delta N_0/2$. $B_{12}$ is the Einstein coefficient for induced absorption, $I(\omega) = c\rho(\omega)$ with $\rho(\omega)$ frequency dependent energy density. $\overline{R}$ is the mean relaxation probability for transitions back to the lower state. An intensity above the saturation level has an impact on the lineshape of the transition. According to Demtröder [Ref. 24, p. 106] the saturated linewidth is given as $\gamma_s = \gamma\sqrt{1+S_0}$. With $S_0 = B_{12}\rho(\omega_o)/\overline{R}$, the saturation parameter is

$$S_0 = S_0 \frac{(\gamma/2)^2}{(\omega - \omega_0)^2 + (\gamma/2)^2}$$

The saturated extinction coefficient profile is then

$$\kappa_s(\omega) = \frac{\hbar \Delta N_0 B_{12}}{\pi \gamma (1+S_0)} \frac{(\gamma_s/2)^2}{(\omega - \omega_0)^2 + (\gamma_s/2)^2}$$

in comparison to the unsaturated extinction coefficient profile

$$\kappa(\omega) = \frac{\hbar \Delta N_0 B_{12}}{\pi \gamma} \frac{(\gamma/2)^2}{(\omega - \omega_0)^2 + (\gamma/2)^2}.$$ 

The saturation decreases the extinction coefficient by a factor of $(1+S_0)$ and the transition line is broadened by a factor $\sqrt{1+S_0}$. A similar relation can be derived for the real part of the refractive index as well.

The value of the saturation intensity $I_{\text{sat}}$ is computed by using [Ref. 3]

$$I_{\text{sat}} = \frac{1}{2} \frac{\varepsilon_0 c \hbar^2 \gamma_{12} \Gamma_0}{|\mu_{12}|^2}$$

where $\gamma_{12}$ refers to the coherence dephasing rate and $\Gamma_0$ refers to the population decay rate. $\mu_{12}$ is the one-photon transition dipole moment and relates to the induced absorption coefficient $B_{12}$ by

$$\mu_{12}^2 = 6\varepsilon_0 \hbar^2 B_{12}.$$
The Voigt profiles

When both the Doppler and the collisional broadening are of importance the line profile corresponds to a convolution of the Gaussian and the Lorentzian profiles. The effective line center frequency is \( v^* = v_0 \left( 1 + \frac{y}{c} \right) \) and therefore \( u = c \left( v^* - v_0 \right) / v_0 \). By integrating over all the possible velocities of the molecules and by using equations (17), (18) and (19) we obtain:

\[
\kappa(v) = \frac{N_j f_j e^2}{4(2\pi)^3 \varepsilon_0 m_e} \sqrt{\frac{M}{2k_B T v_0}} \int_{-\infty}^{\infty} \frac{\Delta v_L/2}{\left( v^* - v \right)^2 + \left( \Delta v_L/2 \right)^2} \exp \left[ -\frac{M}{2k_B T} u^2 \right] du
\]

and

\[
n(v) - 1 = \frac{N_j f_j e^2}{4(2\pi)^3 \varepsilon_0 m_e} \sqrt{\frac{M}{2k_B T v_0}} \int_{-\infty}^{\infty} \frac{v^* - v}{\left( v^* - v \right)^2 + \left( \Delta v_L/2 \right)^2} \exp \left[ -\frac{M}{2k_B T} u^2 \right] du
\]

The detailed derivation of the complex refractive index can be found in Appendix A. Here the function are presented with the frequency \( v \) as variable instead of the angular frequency \( \omega \). Finally the frequency dependence of the complex refractive index, \( n'(v) = [n_0 + n(v)] - i\kappa(v) \), in the vicinity of a molecular transition, can be represented by:

\[
\kappa(v) = \frac{\sqrt{\pi \ln 2}}{8\pi^2} \frac{N_j f_j e^2}{\varepsilon_0 m_e \Delta v_D v_0} \text{Re}\{W(z)\}
\]

\[
n(v) = \frac{\sqrt{\pi \ln 2}}{8\pi^2} \frac{N_j f_j e^2}{\varepsilon_0 m_e \Delta v_D v_0} \text{Im}\{W(z)\}
\]

\( W(z) = \exp(-z^2)\text{erfc}(-iz) \) and \( z = \beta + i\alpha \). \( \beta \) corresponds to \( 2\sqrt{\ln 2} (v - v_0) / \Delta v_D \) and \( \alpha = \sqrt{\ln 2} \Delta v_L / \Delta v_D \) is the Voigt parameter. The real part of \( W(z) \) is the convolution of a Gaussian and a Lorentzian and is known as the Voigt function. \( v_0 \) denotes the resonance frequency of the transition, \( \kappa(v) \) the extinction coefficient, \( n(v) \) the contribution of resonance to the refractive index, \( n_0 \) the refractive index of the surroundings (given by equation (16) as \( n_0 = 1 + K\rho \)), \( N_j \) the species concentration in
the specific state and \( J \) the oscillator strength of the transition. \( \Delta \nu_D \) and \( \Delta \nu_L \) are the Doppler and the Lorentzian full widths at half maximum (FWHM) of the transition, respectively. The function \( W(z) \) does not have analytical solutions, and therefore numerical approximations have to be applied. The solution chosen in this work can be found in Appendix A.

In Figure 8 the plots of the calculated real, \( n(v) \) and imaginary, \( \kappa(v) \), part of the complex refractive index of the \( Q_2(5) \) transition in the \( (0,0) \) vibrational band of the \( A^3\Pi - X^3\Sigma^- \) electronic system of NH are depicted. Input parameters for these plots are \( A_21 = 2.5 \times 10^6 \) \( \text{s}^{-1} \), \( B = 16.667 \) \( \text{cm}^{-1} \), \( \omega_0 = 3300 \) \( \text{cm}^{-1} \), \( f_J = 0.0423 \), \( T = 2345 \) K and an assumed total NH concentration of \( 10^{20} \) \( \text{m}^{-3} \). As a Voigt FWHM the experimentally determined value of \( \Delta \nu_v = 0.3 \) \( \text{cm}^{-1} \) was used, yielding \( \Delta \nu_L = 0.065 \) \( \text{cm}^{-1} \) by calculating \( \Delta \nu_D = 0.266 \) \( \text{cm}^{-1} \) at this temperature.

![Figure 8: Plots of the calculated real, \( n(v) \) and imaginary, \( \kappa(v) \), part of the complex refractive index of the \( Q_2(5) \) transition in the \( (0,0) \) vibrational band of the \( A^3\Pi - X^3\Sigma^- \) electronic system of NH. The Voigt width is \( \Delta \nu_v = 9.067 \times 10^9 \) \( \text{Hz} \), \( \alpha = 0.203 \). Here plotted as a function of the frequency difference from the center frequency of the transition, \( v_0 = 8.9243 \times 10^{14} \) \( \text{Hz} \).]
Resonant Holographic Interferometry

In the vicinity of a molecular transition light waves of different frequencies will experience a different refractive index $n_0 + n(v)$. (In the following all expressions will be given in frequency $v$ instead of the angular frequency $\omega$). This difference in the refractive index can be used to generate a relative phase shift between two waves of slightly different frequencies as they pass through the flame. In fact, the phase velocity of the two waves will be altered according to the total refractive index, $n_0 + n(v)$. By interferometric subtraction, the bulk refractive index $n_0$ of the flame will be eliminated yielding the net contribution of the resonance effect to the refractive index $n(v)$. Holography is applied to perform the interferometric subtraction. The two waves are recorded on the same holographic plate and subsequently reconstructed simultaneously, to interfere. Fringe shifts will be observed in areas where the resonant species have introduced a refractive index difference for the two exposure frequencies.

For the plane monochromatic wave propagating in the $z$-axis

$$E_{in} = E_0 \exp\left(i(2\pi v_i t - k_{0j} n_0 z)\right),$$

with a frequency $v_i$ in the vicinity of a transition of a molecule, immediately after exiting the flame we will have

$$E_{out} = E_0 \exp\left[i\left(2\pi v_i t - k_{0j} \left(n_0 + n(v) - i\kappa(v)\right)L\right)\right].$$

$L$ now denotes the interaction length of the wave with the flame. We have considered the simple case where no ray bending takes place and the wave propagates in a straight line. Recording this wave on a “screen” we obtain an intensity distribution of

$$I(r) = \left|E_0 \exp\left(i\left(-k_{0j} \left(n_0 + n(v) - i\kappa(v)\right)L\right)\right)\right|^2$$

$$= \left|E_0^2 \exp(-2k_{0j}\kappa(v)L)\right| = \left|E_0^2 \exp(-\alpha(v)L)\right|$$

with $\alpha(v) = 2k_{0j}\kappa(v)$, the absorption coefficient. What we have here is in essence the classical “line-of-sight” absorption measurement in two dimensions.

For two waves with different frequencies in the vicinity of a molecular transition, $v_1$ and $v_2$ the corresponding refractive indexes are $n'_{1}(v)=[n_0+n(v_1)]-i\kappa(v_1)$ and
n'_2(v)=[n_0+n(v_2)]-i\kappa(v_2), respectively. The intensity recorded in a two color interferogram is given according to equation (15):

\[ I(\vec{r}) = |E_0 \exp(i[-k_{01}(n_0 + n(v_1) - i\kappa(v_1))L]) + E_0 \exp(i[-k_{02}(n_0 + n(v_2) - i\kappa(v_2))L])|^2 \]

or analytically,

\[ I(\vec{r}) = |E_0|^2 [\exp(-\alpha_1(v_1)L) + \exp(-\alpha_2(v_2)L)] + 2\exp\left(-\frac{[\alpha_1(v_1) + \alpha_2(v_2)]L}{2}\right)\cos[n_0(k_{01} - k_{02})L + \{k_{01}n(v_1) - k_{02}n(v_2)\}L] \]

In the argument of the cosine we recognize here the accumulated phase along the path \( L: \phi_i = k_i(n_0 + n(v_i))L \). The left term is a constant due to the frequency difference between the two exposure waves. The second is dependent on the resonant contribution to the refractive index at the two exposure frequencies. This term carries the information on the species concentration in the flame. The intensity of the interferogram is attenuated due to the imaginary part of the refractive index \( \kappa(v_i) \), just in the way for the single wave passing through the flame. The visibility of the recorded fringes will remain equal to one in the case where the input intensities of the two exposure waves are equal and \( \alpha(v_1) = \alpha(v_2) \), even in areas of the flame where strong absorption takes place. The visibility \( V \) of the interferogram is defined by Michelson as

\[ V = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} = \frac{2I_1^{1/2}I_2^{1/2}}{I_1 + I_2} \]

where \( I_1 \) and \( I_2 \) are the intensities due to each beam alone (Ref. 20, p. 46). Thus

\[ V = \frac{2I_1^{1/2}I_2^{1/2}}{I_1 + I_2} = \frac{2\exp\left(-\frac{\alpha(v_1) + \alpha(v_2)}{2}L\right)}{\exp(-\alpha(v_1)L) + \exp(-\alpha(v_2)L)} \]

For the case that \( E_{01} = E_{02} \Rightarrow I_1 = I_2 \) and \( \alpha(v_1) = \alpha(v_2) \) the visibility is \( V = 1 \).

A link has been achieved between the distribution of a species in the flame and the intensity recorded in a double exposure two color hologram by utilizing the complex refractive index in the vicinity of a transition of the species. The information is
contained in the fringe shift (F.S.) that is determined, assuming a uniform species distribution along the path i.e. a *stratified medium*, as

$$F.S. = \frac{k_{01} n_1(v_1) - k_{02} n_2(v_2)}{2\pi} L = \frac{v_1 n_1(v_1) - v_2 n_2(v_2)}{c} L$$

(24)
Experimental

The subject of investigation: the NH\textsubscript{3}-O\textsubscript{2} diffusion flame

For a first demonstration of the measuring technique that has been outlined in the previous section, a NH\textsubscript{3}-O\textsubscript{2} diffusion flame was chosen for following reasons. First, the flame temperature is high. 1D numerical simulations, see Appendix B, indicate a maximum temperature on the order of 2700 K. A high flame temperature is preferable for these initial experiments since the width (FWHM) of molecular transitions in atmospheric pressure flames is enhanced significantly. Second, two combustion relevant species, NH and OH, are produced in adequate concentrations to facilitate their experimental investigation by the new technique. The vibronic 0-0 bands of NH (A \textsuperscript{3}Π - X \textsuperscript{3}Σ) and OH (A \textsuperscript{2}Σ\textsuperscript{+} - X \textsuperscript{2}Π) are accessible in the UV, thus enabling the utilization of commonly available dye lasers.

The Wolfhard-Parker burner

From theory it is clear that Resonant Holographic Interferometry and the absorption measurements are "line of sight" techniques, i.e. the information are obtained as an integration of the signal along the path. Equations (23) and (24) illustrate this integration of the signal for the simple case of a straight beam propagating through the target medium. The integration along the path is substituted by a simple multiplication with the length of the target medium.

Fringe interpretation is a known problem in Holographic Interferometry and a lot of schemes have been developed in order to achieve the full 3D information contained in a typical flame structure. Theoretical schemes to derive the 3D information of the refractive index distribution in an axial symmetric flame by utilizing the Abel transformation have been developed [Ref. 19, p. 316, Ref. 26, 27]. Also, tomographical
techniques to reconstruct the full 3D structure from multiple ‘projections’ are found in
the literature [Ref. 28, 29, 30, 31, 32, 33, 34]. In principle RHI can be extended to 3D
measurements, however it is not the topic of this work.

Figure 9: The NH3-O2 diffusion flame stabilized over the Wolfhard-Parker
burner.

For a conclusive demonstration of the applicability of the proposed measuring
technique to quantify a single species in the flame, a combustion environment that
allows a simple interpretation of the results is advantageous. Therefore, a Wolfhard-
Parker slot burner is implemented for the preliminary RHI measurements since it
provides a 2D flame structure that simplifies the interpretation of the recorded
interferograms [Ref. 35], see Figure 9. In appendix B, detailed information and
drawings of the burner are presented. The burner consists of three parallel slots of the
same length. The fuel (NH₃) is provided through the middle slot while the two outer
slots provide the oxidizer (O₂). The burner produces a saddle shaped laminar diffusion
flame, consisting of two identical, two-dimensional flame sheets, which greatly
facilitate precise lateral profile measurements and maintain an optimal optical accessibility. Alternatively a premixed NH$_3$-O$_2$ flame stabilized on a flat flame burner could be chosen. However, such a burner produces a flame front that is parallel and close to the burner surface. The correct interpretation of the experimental results is difficult for this case. Diffraction patterns originating from the burner edge that spatially overlap with the imaged area have to be taken into account. Additionally ray bending might force the refracted beams to collide on the burner surface and result in a loss of valuable information.

Let us assume that the probe beams propagates along the $z$-axis. The flame is aligned with the slots parallel to the $z$-axis, see Figure 10. The bulk refractive index due to the combustion process and the resonant refractive index due to molecular absorption is constant along the $z$-axis but varies along the two other axes. The interpretation of the interferograms is thus straightforward and equation (23) and (24) can be applied without further modifications.

![Figure 10: Top view of the Wolfhard-Parker burner in the optical set up in a typical line absorption measurement set-up. Note that the aperture selects only a part of the expanded probe beam to perform the measurement.](image-url)
K. Smyth et al. [Ref. 35] supplied high flow velocities to the burner, thus forcing the two flame-sheets parallel to the x-axis. The flame is additionally stabilized by a stainless steel V-shaped mesh above the burner. The mesh serves as a heat reservoir with a beneficial effect on flame stability. The effects of the mesh on the flame kinetics are, however, unpredictable. Therefore, a substantial effort has been imposed in this work to generate a stable 2D flame without any stabilization structures. A stable flame is achieved by controlling the velocity profile at the exits of the three slots by adjusting the total and the ratio of the flow velocities of fuel and oxidizer.

To obtain a fully developed parabolic velocity profile, for the case of the two oxidizer slots, a length of ~50 cm would be required, taking into account typical velocities of the gases used in this work [Ref. 36]. The length of the burner is therefore too short to allow the development of a full parabolic velocity profile. Considering the rectangular shape of the three slots, see Appendix B, the parabolic velocity profiles at the long and the short side will be quite different. Therefore, efforts were placed to achieve a top hat velocity profile in the following way. Different fillings of the slots (i.e. glass beads) were tried in order to diffuse the gas in the slot-tubes and create a laminar flow. In Appendix B the fillings of the burner as used in all our experiments are described. For every filling trial the stability of the flame was investigated interferometrically, placing the burner into one of the arms of a Mach-Zehnder interferometer.

A perfectly stable flame can not be achieved. For some flow velocities and velocity ratios however, a sufficient stability is achieved for the imaged area, i.e. the primary flame zone exhibiting the form of a saddle. In fact, only small deviations can be observed by applying the Mach-Zehnder interferometer. Moreover, after thermal stabilization with the surroundings a stable flame is obtained up to a height of three to four times the position of the 'saddle'. In summary, this characterization shows that the burner stabilizes a reproducible laminar flame structure meeting all requirements to perform the experimental investigations.
The burner parameters used for the experiments performed in this work are summarized in Table 1:

<table>
<thead>
<tr>
<th></th>
<th>Slot Area (cm²)</th>
<th>Input Flow (lt/min)</th>
<th>Flow velocity (cm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH₃</td>
<td>3.1</td>
<td>2.5</td>
<td>13.44</td>
</tr>
<tr>
<td>O₂</td>
<td>13.5 (2 x 6.75)</td>
<td>30</td>
<td>37.04</td>
</tr>
</tbody>
</table>

Table 1: Burner parameters for the performed experiments.

The velocity ratio of oxidizer to fuel is then $V(\text{O}_2)/V(\text{NH}_3) = 2.75$. The flows are controlled by two flow meters, Sho-Rate by Brooks Instrument, calibrated for NH₃ and O₂. An accuracy of ±5% with an uncertainty of ±0.5% for an input pressure of 4 bar absolute is specified. During the experiments the setting on the flow meters is fixed and the shut off valves only are used to turn the flows on and off. In this way an increased repeatability of the inlet flows is obtained. During measurements no visible change in the flame is observed, i.e. the height of the flame-saddle and the flame remains stable.

### Laser requirements

For the correct implementation of Resonant Holographic Interferometry several considerations on the laser system have to be taken into account. First, a constant phase shape of the individual pulses is required. If we are to utilize the distortion induced on a wavefront to achieve information on the spatial distribution of a resonant species, it is important that the phase distribution is repeatable from pulse to pulse. Second, to achieve a good interference between the object and the reference beam a long coherence length is preferable, since constraints to set up the holographic experiment are relaxed. The coherence length can be roughly estimated as the inverse function of the laser bandwidth $l_{\text{coh}} = c/\Delta v_{\text{laser}}$, where $\Delta v_{\text{laser}}$ is the laser bandwidth and $c$ the speed of light. Third, a narrow laser bandwidth is of great importance when probing the real part of the complex refractive index. The formulation derived in the theoretical chapter, equations...
Resonant Holographic Interferometry on NH and OH

(23) and (24), is based on the assumption that a laser with an infinitely narrow bandwidth is used. Since this is obviously never the case, the refractive index frequency distribution must be convoluted with the frequency profile of the laser to correctly derive the fringe shift information acquired from an RHI experiment. By assuming a refractive index distribution as shown in Figure 8 and a gaussian frequency profile, we computed a resulting Fringe Shift as a function of the laser bandwidth by using equation (23). The results are depicted in Figure 11. The horizontal axis indicates the ratio of the laser width to the Voigt width of the Q5(5) transition of NH. All other data are summarized in the caption of Figure 8. It can be seen, that a broader laser width produces smaller fringe shifts, since the steep refractive index near resonance is averaged.

![Graph showing Total Fringe Shift as a function of the laser bandwidth.](image_url)

**Figure 11:** Total Fringe Shift as a function of the laser bandwidth. The laser width is normalized with the Voigt width of the transition line. For this calculation the refractive index distribution plotted in Figure 8 is used. Two cases are depicted. First, the bandwidth of both probe waves is increased. Second, the width of one probe wave is kept constant at 0.04 cm⁻¹.
Therefore, a narrow bandwidth laser is highly desirable. If the laser width meets the width of the transition ($\Delta v_{\text{laser}} \approx \Delta v_{\text{T}}$), fringe shift evaluation without deconvolution will yield an signal reduction of 30%. However, this can be corrected under the assumption that the laser bandwidth and the frequency distribution of the laser intensity are known. The same holds for the absorption measurements as described by equation (24).

Substantial work has been performed to circumvent the disadvantages of using broad band lasers [Ref. 37] as long as narrow bandwidth lasers were not readily available.

Fortunately, a Lambda Physik 3002 pulsed dye laser is available in our laboratories which is equipped with an intracavity etalon providing a laser beam exhibiting a width of 0.04 cm$^{-1}$. In comparison with typical absorption line widths encountered in NH$_3$-O$_2$ flame on the order of 0.25-0.3 cm$^{-1}$ the measurements will result in an error on the fringe shift information of about 2%. As will be shown later, this error is negligible compared to other intrinsic errors. Therefore, a monochromatic light source is considered in the following and equations (23) and (24) need not be modified. When using the intracavity etalon of 0.04 cm$^{-1}$, a coherence length of $l_{\text{coh}} \approx 25$ cm is expected. To address the two target molecules, NH and OH, the laser system is operated with two different dyes and two different pump lasers. Table 2 summarizes the two modes of the laser operation.

<table>
<thead>
<tr>
<th>Dye (nm)</th>
<th>Doubling</th>
<th>Pump laser</th>
<th>Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH</td>
<td>PTP</td>
<td>Excimer @308nm</td>
<td>$\sim 2$ mJ/pulse</td>
</tr>
<tr>
<td></td>
<td>330-353</td>
<td>$\sim 250$-300 mJ/pulse, 25ns pulse duration</td>
<td>$\Delta \tilde{v} = 0.04$ cm$^{-1}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Max. Rep. rate: 500 Hz</td>
<td></td>
</tr>
<tr>
<td>OH</td>
<td>Rhodamine 101</td>
<td>Nd:YAG @532nm</td>
<td>$\sim 3$ mJ/pulse</td>
</tr>
<tr>
<td></td>
<td>600-620</td>
<td>$\sim 200$ mJ/pulse, 6 ns pulse duration</td>
<td>$\Delta \tilde{v} = 0.04$ cm$^{-1}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Fixed Rep. rate: 10 Hz</td>
<td></td>
</tr>
</tbody>
</table>

Table 2: The two modes of the Lambda Physik 3002 operation used to probe the two target species NH and OH.
The single fringe pattern indicating a single mode operation of the laser is observed with a monitor etalon and a CCD camera, see Figure 27. In the following, the operation mode of the laser for NH and OH absorption frequencies will be called the NH mode and the OH mode, respectively.

**The holographic medium**

For the holographic recordings an emulsion has to be chosen with a high sensitivity in the UV where the experiments are performed. Since little holographic work has been performed in the UV [Ref. 38], the characteristics of different emulsions are not well documented in this wavelength region. Moreover, in contrast to standard holographic work pulsed lasers are utilized, thus the exposure energy cannot be controlled compared to the use of CW lasers. The pulse to pulse energy variations of the laser configurations are typically 10% and 15% for the NH mode and the OH mode, respectively. The beams are expanded in order to create plane wavefronts covering the whole flame zone. The intensity distribution across the expanded beam is also subject to intensity fluctuations from pulse to pulse. As stated in the theoretical part, the emulsion response is supposed to depend linearly on the exposure energy. In fact, the emulsion response is only linear in a limited range of exposure energies. For fixed developing parameters, a curve of emulsion response *versus* exposure energy, the so-called characteristic curve of the emulsion, can be used as a guideline in choosing optimal exposure energies. An analytical description of the characteristic curve is given by C. M. Vest [Ref. 19, p.45-53]. Note, however, that in the presence of the energy fluctuations mentioned above, a high degree of uncertainty remains whether the exposure energies are within the linear part of a determined characteristic curve. For all exposures performed in this work, the ratio between object and reference beam is kept to \( \sim 1:1 \). Therefore, a maximal diffraction efficiency is obtained in principle. However, due to a polarization ratio of
the laser beam of ~50:1 and different frequency response of the involved optics the intensity modulation of the recorded “grating structures” in the emulsion will be reduced. By considering a typical characteristic curve for the emulsion exhibiting, we assume a linear response on medium exposure energies. Typical commercially available Silver Halide emulsion in the blue region of the light spectrum are [Ref. 19, p. 47].

<table>
<thead>
<tr>
<th>Emulsion type</th>
<th>Resolution (lines/mm)</th>
<th>Sensitivity (µJ/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kodak 125</td>
<td>1250</td>
<td>2 µJ/cm² @ 441.6nm</td>
</tr>
<tr>
<td>AGFA GEVAERT 8E56</td>
<td>5000</td>
<td>15 µJ/cm² @ 476nm</td>
</tr>
<tr>
<td>AGFA GEVAERT 10E56</td>
<td>2800</td>
<td>1.4 µJ/cm² @ 476nm</td>
</tr>
</tbody>
</table>

Other holographic emulsions have been considered, for example the photoresist material [Ref. Syms] or photothermal emulsions. However, these emulsion require higher exposure energies than available from the used lasers. Additionally, high energies would saturate the molecular transitions of the target species in the flame. In this work the AGFA GEVAERT 8E56 (4 x 5 inch) was chosen as a holographic emulsion, since it is readily applicable in the laboratory and exhibits a high spatial resolution. The plates are developed in three steps. The holograms are developed with Kodak D-19 developer, in standard solution, until a Optical Density (OD) of 2-3 was obtained. The plates are subsequently immersed in a stop-bath, a mild solution of Ascorbic-acid, for two minutes. The plates are fixed for three minutes in the fix-bath Ilford G 350 After rinsing in running de-mineralized water for 4-5 minutes the plates are bleached in a GP 431 bleach bath (AGFA GEVAERT standard recipe for transmission holograms) and then re-immersed in the water bath for about ten minutes. In all process steps a soft manual agitation was applied.
CARS measurements of temperature

It has been pointed out that two combustion-related parameters primarily determines the refractive index: the species concentration and the temperature of the flame. The real and the imaginary parts of the refractive index are interdependent, since the derivation of the complex refractive index is a natural consequence of fundamental Electromagnetic Theory. Measuring each part of the complex refractive index, would result in a system of two equations with two unknowns. Having the one unknown, each equation must result in the other unknown. In this way, a correlation between the two independent measurements can be made. It is therefore desirable to have an independent and reliable measurement of at least one of these two parameters.

Coherent Anti-Stokes Raman Spectroscopy (CARS)

![Figure 12: Folded BOXCARS phase matching geometry applied on the NH₃-O₂ diffusion flame stabilized over the W.-P. burner.](image)

It was chosen to utilize the transportable CARS system, developed her at Paul Scherrer Institute [Ref. 5], in order to obtain Temperature profiles over the reaction zone of the NH₃-O₂ flame. The CARS measurements were performed by B. Hemmerling and A. Stampanoni. For the measurements the USED BOXCARS phase matching geometry is applied. For this configuration of the CARS system, temperatures are determined with
an accuracy of ±60 K at temperatures of ~2000 K [Ref. 5]. At higher temperatures the accuracy is expected to increase.

On the other hand measurements in a NH$_3$-O$_2$ flame pose significant difficulties. The high temperatures and the low concentrations of N$_2$ in combination with the high non-resonant background, see Figure 13, resulted in an reduced accuracy of ±100 K. Due to the low N$_2$ concentrations, see numerical simulations in Appendix B, the standard single shot measuring mode could not be applied. Additionally, N$_2$ was available only in the proximity of the reaction zone. Consequently, a complete temperature profile could not be achieved without seeding N$_2$ in all three slots. That would of course change the flame conditions and is avoided. The flame stability ensures that averaging can be performed.

Figure 13: 200 pulse average N$_2$ spectrum at a height of x = 3.5 mm above the burner surface and at y = 4.2 mm relative to the symmetry axis of the W.-P. burner. The solid line shows the measured spectrum, while the stippled line shows the simulation performed to obtain the temperature (courtesy B. Hemmerling and A. Stampanoni).
The measurements presented here are averages over 200 shots. The burner is positioned on a translation stage and measurements at different positions can be performed. The probe volume has a longitudinal length of \( \sim 10 \) mm and a diameter of \( \sim 200 \) µm. The burner is aligned so that the propagation axis of the CARS probe beams is parallel to the symmetry axis of the W.-P. burner. Taking into account that the length of the flame is \( L = 42 \) mm, the spatial resolution of the temperature measurements is determined by the diameter of the probe volume. Since the temperature is not expected to change rapidly as a function of the height above the burner surface, the point-wise measurements were performed at a height of \( x = 3.5 \) mm above the burner surface. In Figure 13 a representative example of a N\(_2\) spectrum is presented. This spectrum resulted in the maximum temperature, \( T = 2827 \pm 100 \) K.

**Figure 14:** Temperature profile at a height of \( x = 3.5 \) mm above the burner surface. The solid line shows the 5\(^{th}\) order polynomial fit to the point-wise measurements. The error bars indicate the \( \pm 100 \) K accuracy of the measurement.

In Figure 14 the obtained temperature profile at a height of \( x = 3.5 \) mm above the burner surface is shown. The positions of the point-wise measurements in relation to the symmetry axis of the W.-P. burner were obtained by calibrating the translation stage to
the wall separating the oxidizer and the fuel slots.

A fifth order polynomial fit to the point-wise measurements is used to estimate the temperatures at positions where the species concentration will be calculated from the experimental results.

The holographic set up

In order to record a wavefront that can be successfully reconstructed, two coherent wavefronts have to interfere on the hologram plate. There are many different configurations for a holographic set up. For a detailed analysis please refer to Refs. 17, 19 and 39. Since we are interested in producing double exposure transmission holograms by using two different frequencies, several configurations for Holographic Interferometry will be discussed in the following. Furthermore, an analytical description will be given for the set up utilized in this work.

One reference arm set up

Figure 15 shows the simplest holographic setup. It consists of a beam splitter separating the laser beam into two parts. They propagate through collimating optics and produce two plane waves that interfere at an angle on the holographic plate. By passing two pulses of different frequencies through the system sequentially, a double exposure recording is obtained. This configuration corresponds to the derivation described in the theoretical part of this work. Referring to equation (12), we recall that the interferogram which is reconstructed from the hologram will be a function of the read-out beam propagation vector $\mathbf{k}_{RO}$. By re-illuminating the hologram with the read-out beam, two wavefronts will re-emerge from the hologram plates at angles that satisfy the Bragg condition. It is obvious that as long as the Bragg condition is satisfied, the two beams
will have a fixed angle to each other and the interferogram will be defined by the constant $K_{\text{int}}$. Therefore, the fringe distance in the interferogram will be fixed in space.

Figure 15: One reference arm holographic set up.

The locked fringes in space represent a disadvantage of the one reference configuration. In fact, the accuracy of the optical phase determination from the interferograms is limited since it is often difficult to locate the center of the fringes to better than 10% of their spacing. Note, that one spacing corresponds to a wavelength difference. In addition ambiguities arise if only a few number of fringes are available that are not equally spaced. Further errors are introduced by non-linear interpolation that are often required to determine fractional fringe orders. Higher accuracy can be obtained by analyzing a digitized interferogram by dedicated software that processes the fringes of the interferograms and evaluates their center lines. However, it is preferable to use techniques which permit direct measurements of the optical phase difference by electronic means as described in the next chapter.
Two reference arm set up

Figure 16: Two reference beam holographic setup. In order to address the object beam of each recording with a different reference beam, two mechanical shutters are used, switching ON/OFF the two reference beams.

In order to circumvent the inaccuracy of fringe interpretation, a two reference arm configuration has been suggested [Ref. 19, Ref. 40, 41, 42]. In this case each exposure has a different reference incidence angle. During reconstruction, the doubly exposed hologram is illuminated with two read out beams. Each beam satisfies the Bragg condition for the two holograms stored on the medium. The reconstruction yields, therefore, four wavefronts.

Only two of the four wavefronts will interfere in space [Ref. 40]. So far no improvement is achieved over the one reference arm set up. However, if one of the two reference beams is manipulated, for example by changing the angle of intersection, the corresponding wavefront will be affected. The change is reflected in the vector $K_{int}$ and
we think of the fringes as being ‘alive’. The dynamic effect of the fringes can be utilized to gain the full 2D phase information that is hidden in the interferometric comparison of the two wavefronts. An analytical description of how to take advantage of these ‘live’ fringes is described in Ref. 41.

In the experiments performed during this work several factors did not permit the utilization of the two reference arm holographic set up. First the time delay between the two exposure pulses was kept small, a few nanoseconds, in order to avoid spurious fringes arriving from flame instabilities. To perform a double exposure hologram on this time scale by using a two reference arm configuration would involve very fast switching units with high energy losses. In both laser configurations the laser has output energies of 2-3 mJ/pulse prohibiting excessive energy loses (see below). Since this work is focused on studying the behavior of the real and imaginary refractive index, the one reference arm set up is used. As a consequence, a limited accuracy of the fringe shift evaluation is expected. On the other, this set up allows the registration of the full 2D species distribution of the target species. Moreover, the maximum absolute fringe shift can be readily obtained and contains the information of the maximum species concentration.

*Image plane holography*

According to the theory, the read-out beam is a critical parameter for the correct reconstruction of the hologram. A perfect reconstruction of the two wavefronts requires a read-out beam which is identical to the original reference beam. There is a way to circumvent this difficulty. By using a relay lens the wavefront under study is projected on, or in the proximity of the hologram plane. This projection of the wavefront on the hologram plane has given the name to the technique, since it is rather the image of the wavefront that is recorded than the wavefront itself.

There are several advantages in using image plane holography [Ref. 43, 44, 45, 46].
The biggest advantage is the fact that the holograms can be read-out with white light. The explanation for the white light read-out was given by L. Rosen [Ref. 44] and by G.B. Brandt [Ref. 45]. They showed that the smear introduced by the many different reconstruction angles provided by the broadband illumination is a function of the object to hologram distance. By projecting the image on, or in the proximity of the hologram plane, minimal smear is achieved, since the object-hologram distance is nearly zero.

**Figure 17:** Set up for recording image plane holograms. The set up is in essence the same as in Figure E4 where the relay lens is added in the object arm. The 1:1 imaging is shown with the projection and inversion of the flame.

Stenson [Ref. 46] reported that the image sharpness is apparently unaffected by the emulsion thickness and other defects in the hologram plate. Since image plane holography obeys the general laws of holographic recording and reconstruction, multiple exposures can also be recorded in the usual manner. Indeed, in the referenced works the successful recording and reconstruction of 3D objects was illustrated.
Commonly, Argon-ion lasers are used for holography. These lasers are usually considered as point sources. However, the utilization of extended sources has also been demonstrated [Ref. 44]. The pulsed Lambda Physik 3002 laser system used in this work does not provide a gaussian shaped beam and cannot be considered, therefore, as a point source. This drawback is circumvented by using image plane holography that allows the use of extended light sources. In the case of the applied double exposure holographic interferometry, the utilization of image plane holography displays an additional advantage. The two registered wavefronts will interfere on the hologram plane making the later recording of the interferograms by a camera straightforward. Furthermore, the fringe spacing of the interferograms is constant, independently of the read-out angle. The only problem of the method is the requirement of a clear hologram surface, since dust particulates could disturb the read-out.

In Figure 17, a set up utilizing a relay lens in order to record image plane holograms is shown. The system consists of a beam splitter that creates the object and reference beam, collimating optics to create the two plane wavefronts and the relay lens. The lens is positioned such that 1:1 imaging is achieved for the configuration, i.e. a 2f:2f projection is used. The situation is indicated in the Figure by the projection of a flame. It is also shown that the flame is inverted following the 1:1 imaging.

Optimizing the projection for the case of a flame

The burner is now placed into the object arm of the set up. As explained previously, the burner is placed with the slots parallel to the propagation axis (z-axis) of the probe beam. The wavefronts will thus experience a 2D refractive index distribution that is a function of the two other directions (x, y). The refractive index distribution is a function of the combustion parameters (density and temperature) and the resonant refractive index for a wavefront exhibiting a frequency in the proximity of a molecular transition
(see theoretical section). Obviously, the 2D refractive index field exhibits steep gradients that deviate the vector field of the wave from its straight path. It has been shown analytically that the effects of refraction have to be considered for a successful interpretation of double exposure holograms and that the position of the burner in respect to the relay lens is of importance [Ref. 16. Ref. 26]. Briefly, it can be shown that errors are minimized when projecting the probe wavefront at a plane located at the 2/3’s of the flame-wavefront interaction length, even in the case of a three dimensional refractive index field. In other words, that position of the object plane the fringe count may be considered in the straight line approximation, as if no refraction occurred.

Figure 18: One ray passing through the flame zone with no flame on, one ray passing through the flame zone with the flame on. Choice of object plane of the relay lens in the flame at 2/3L.

Figure 18 illustrates the correct projection of the wavefront for minimizing the refractive index measurement errors. Typically, double exposure holograms are used to register the total refractive index field. The first recording is performed with the burner Off, and the wavefront records the ambient refractive index. Subsequently the burner is
turned On and the second wavefront records the refractive index due to the flame. Obviously, the two beams pass through the same optics. Thus any distortion due to the optical system will be interferometrically canceled. The resulting interferogram contains the information of the optical path difference between the two wavefronts due to the refractive index of the flame only. When the flame is on, the individual rays follow a parabolic trajectory due to the refractive index gradients, Figure 18.

Figure 18 also demonstrates that an appropriate imaging of the wavefront minimizes the error for the correct calculation of the refractive index. It is seen that the two individual rays that interfere on the hologram plane do not originate from the same point. Therefore an error in the refractive index measurement is introduced. The error is minimized by a correct projection assuring that the interfering rays originate from the same point. For an analytical description of this error analysis the reader is referred to Ref. 16.

Figure 19: Illustrating the effect of refraction on the plane wavefront as observed on the hologram plane after passing through the flame. On the left, the wavefront is recorded without the relay lens, on the right the lens is introduced at 2/3L.
For the illustration two images are presented in Figure 19. On the left side, the wavefront is recorded on the hologram plane without using the relay lens. The effect of the refractive index gradients on the wavefront is clearly discernible. Rays cross each other and the spatial distribution of the wavefront is destroyed. On the right side, the relay lens is introduced. Due to the 1:1 projection the recorded image is of course inverted by the 1:1 imaging, but here it is presented in normal view. The plane wavefront is almost perfectly reconstructed. Some residual intensity fluctuations can be detected just above the top of the burner where the flame front is located.

Setting up the experiment

The configuration sketched in Figure 17 is set up on a vibration isolated table. The laser is mounted on another table to avoid disturbances of the experiment by vibrations originating from the dye pumps. Interferometric studies of the long term stability indicate a slight movement of the monitor fringes due to the air circulation in the room. Since pulsed holography is used in this work and the delay between the two exposure pulses is short, these disturbances are of no concern. The polarization ratio of the laser is 50:1, thus quite low for holographic experiments. On the other hand, the resonant effect is polarization independent. Additionally, the angle of 30 degrees between object and reference beam suppresses the horizontally polarization grating. Indeed, when reading out the image planes holograms, no spurious images could be detected. Therefore, no attempts to improve the polarization ratio have been envisaged. The laser pulse has to propagate 3m before entering the holographic system. The pulse is vertically polarized to the input plane defined by the object and reference beam propagation axis. The distortion of the pulse shape observed in the far field is insignificant. The double exposure ensures that this type of phase distortions as well as the distortions arising from the steering optics or the bulk refractive index of the flame
are interferometrically subtracted during the hologram read-out. Thus no high quality optics are required. Moreover, as mentioned above for the image plane configuration, extended sources can be used and no spatial filtering of the beams is required. The two beams pass through two x20 beam expanders. In order to reassure that the emerging beams are plane parallel waves, the following qualitative procedure is used. The second lens operates as an aperture and limits the beams to a diameter of ~50 mm. The first lens of the telescope is aligned until the far-field beam is of the same size. As indicated in Figure 16, the hologram plate is placed perpendicular to the incoming object beam. This ensures that the projected wavefront from the 2/3’s of the wavefront-flame interaction length is projected on the hologram plate. The relay lens used for the 1:1 projection is a 2 inch diameter UV lens with f = 200 cm. The position of the lens and the burner/flame is optimized until the distortion due to refraction is corrected as illustrated in Figure 19. In order to calibrate the flame height and width, calibration markers spaced by 0.1 inch for both axes (x and y-axis) are placed in the object beam. 2D images on the hologram plane are recorded by using a square quartz cell filled with a Rhodamine dye solution. For details please refer to the chapter concerning the 2D absorption measurements. A special construction reassured that the Rhodamine dye cell is quickly and accurately placed in the position of the hologram plate. The CCD camera used for the absorption measurements is placed between the two arms of the holographic set up, at an angle of 10 degrees to the dye cell. Taking advantage of the calibration markers and operating the camera lens with a small aperture, i.e. long Depth Of Field (DOF), sharp absorption images are obtained. By locating the camera behind the dye cell, background signals on the CCD chip due to luminescence of the flame or residual UV radiation are avoided. The reconstruction of the holograms can be performed with white light and there are no strict requirements on repositioning the plates in respect to the reference beam [Ref. 44]. The holographic plates are therefore mounted on a 4x5 inch glass plate equipped with a manual shutter. In this way transport of the plates is straightforward. The plate holder is positioned behind a Ø = 10 cm size
mechanical shutter that is electrically addressed and limits the exposures of the plates to
the surrounding light.

For NH exposures the Excimer laser is run with a pulse repetition rate of 1 Hz. First the
manual shutter is removed, and the mechanical shutter is triggered open between two
shots of the Excimer, such that only one shot exposed the hologram. For OH exposures,
the Nd:YAG laser is externally triggered with the Flash-lamps in free running mode
while the Q-switch is off. Single-pulsing the Q-switch, after opening the two
mechanical shutters, ensures the single pulse exposure.

![Interferogram of a double exposure hologram.](image)

**Figure 20:** Interferogram of a double exposure hologram.

First exposure with the flame OFF and second exposure with the flame ON.

To control the ability of the set up to record double exposure holograms of good
contrast and diffraction efficiency, ‘flame On/Off’ holograms are recorded. In Figure 20
a typical example of a such a recording is presented. These experiments are performed
by using the laser in the NH mode. The time separation between the two recording
shots is several minutes, the time the flame takes to thermally stabilize with its
surroundings. The holograms show good fringe contrast and a high diffraction
efficiency, indicating that the path difference of the object and reference beams is well
within the coherence length of the laser and that the optical set up is interferometrically stable. These preliminary holograms are also used to define the range of exposure energies [\mu J/cm^2] that can be used to produce holograms of a good quality. For both frequency regions, NH and OH, the emulsion produces good holograms for energies between 10-80 \mu J/cm^2. This energy is the sum of the intensity of object and reference beams for one exposure. The OD is controlled by the development time.

Next, the flame stability is holographically investigated. With the laser operating in the NH mode, several double exposure holograms are performed with the flame on during both exposures. Flame stability is of importance for the subsequent experiments which involve a frequency shift between exposures. In particular a good guess is required for the maximum delay between the two pulses that yield a cancellation of the bulk refractive index. The flame must not change or move between the two pulses.

![Figure 21: Illustrating the effect of flame instabilities on the recording of a double exposure hologram. On the left side a slight movement of the flame has created several random fringes. On the right side a successful recording of a stable flame is achieved during the two exposures.](image)

In Figure 21 the impact of the flame instabilities is shown. On the left side a slight movement of the flame has created several random fringes. On the right side a successful double exposure hologram read-out is shown. A small deviation can be
observed around the flame structure that is due to a small lateral movement of the flame. The latter hologram has been obtained by applying a delay of 500 ms. However, most holograms, even with shorter time delays, contained random fringes. Such measurements are not sufficient for quantitative nor qualitative measurements of species concentrations.

For the NH mode, the laser can be triggered with a maximal repetition rate of 500 Hz. For the OH mode there is a maximum repetition rate of 10 Hz. Thus, for a successful double exposure hologram the flame has to be stable for more than 100 ms! A further restriction on the minimum delay time is given. The Lambda Physik laser is controlled by a computer to perform the movement from one frequency to another. The minimum time for such a frequency change is 300 ms, basically due to the handshake between the laser control unit and the computer. Thus even higher constraints on the flame stability are required. Typical flame instabilities observed with the Mach-Zehnder interferometer exclude a double exposure with the same laser and a delay of 300ms. Other schemes to create the two frequencies exhibiting time separations below the characteristic time scales of the flame fluctuations have to be found. One solution involves the application of a non-linear process called Stimulated Brillouin Scattering.
Stimulated Brillouin Scattering

Theory of Stimulated Brillouin Scattering

The presence of a time varying electric field in a liquid (or crystal or even a gas) gives rise to a time-varying electrostrictive strain and is thus capable of driving acoustic waves in the medium. The acoustic wave modulates the optical dielectric constant and causes an exchange of energy between electromagnetic waves whose frequencies differ by an amount equal to the acoustical frequency. The effect is analogous to stimulated Raman scattering with the acoustic waves playing the role of the molecular vibrations [Ref. 47, p. 475-476]. Following the classical treatment of Brillouin scattering as presented by A. Yariv [Ref. 47] it can be shown that the maximum gain of the SBS process is obtained for the case of backward scattering. Two basic equations govern the closed system of the two electric fields and the acoustic field: \( \omega_s = \omega_2 - \omega_1 \) and \( \vec{k}_s = \vec{k}_2 - \vec{k}_1 \), where \( \omega_2 \) is the angular frequency of the incident field, \( \omega_s \) that of the driven acoustic wave and \( \omega_1 \) that of the resulting field. \( \vec{k}_1 \) represent in the same manner the corresponding propagation vectors.

The exponential gain of the driven acoustic wave and of the resulting electric field can be shown to increase with the acoustic frequency. Since \( \omega_s << \omega_2 \), we have that \( \omega_2 \approx \omega_1 \) and in an isotropic medium \( k_2 \approx k_1 \). The vector relationship \( \vec{k}_s = \vec{k}_2 - \vec{k}_1 \) becomes identical to the Bragg scattering. The resulting forward acoustic wave will have a frequency of

\[
\omega_{s,\text{max}} = 2\omega_2 \frac{v_{\text{sound}} n}{c} \quad \text{or} \quad v_{s,\text{max}} = 2v_2 \frac{v_{\text{sound}} n}{c}
\]

where \( v_{\text{sound}} \) is the sound velocity in the medium and \( n \) is the refractive index of the medium. When the exponential growth constant is positive, excited acoustic waves propagating along \( k_s \) and zero-field optical waves at \( \omega_1 \) travelling along \( k_1 \) will experience amplification. The stationary Brillouin gain \( g \) is given by [Ref. 48]

\[
g = \frac{\omega_2^2 v_c^2 \tau_B}{c^3 v_{\text{sound}} n \rho_0}
\]
where \( \omega_2 \) is the incident light frequency, \( \gamma_e \) the electristrictive coefficient and \( \tau_B \) the phonon lifetime, \( c \) and \( v_{\text{sound}} \) the velocities of light and sound respectively, and \( \rho_0 \) the density of the Brillouin scattering medium.

Phase conjugation is defined as the process in which the phase of the output wave is complex conjugate to the phase of the input wave [Ref. 49, p. 251-252]. In other words, the process reverses the phase of the input wave. If the phase-conjugated output propagates in the backward direction with respect to the corresponding input wave, it can be used to correct aberrations due to phase distortion experienced by the input wave.

As illustrated in Figure 22, the input beam passing through a medium suffers wave distortion. Unlike an ordinary mirror, the phase conjugate mirror reverse the wavefront distortion of the input beam upon reflection. Then, as the phase-conjugated wave reflects back through the medium again, the wavefront distortion is completely removed (as long as diffraction is negligible).
Figure 23: Set up for investigating the properties of the SBS wave. A beams passes through a beam splitter BS1 and its profile can be monitored (a). The beam passes through a randomizing medium (R), i.e. a flame. The sampled image (b) displays a distorted beam. The beam is focused into the Brillouin cell and a counter propagating beam is generated which still possesses the distortions (c). However, the beam will identically follow the incident beam. When this backward going wave passes through the medium (R) the second time, the original distortions are completely removed which can be verified at (d).

To generate intensive phase-conjugated beams by stimulated Brillouin scattering (SBS), the incident beam is usually focused into a medium to increase the local light intensity. For a liquid media a quartz cell containing the appropriate fluid can be used. In Figure 23 a typical set up for investigating the properties of the SBS wave is presented.

Brillouin scattering is considered as a technique that can create a secondary laser beam with the same spatial and temporal characteristics of the original beam. The frequency shift introduced by the interaction with the acoustic wave can be chosen such that the original and phase conjugate beam address different frequencies on the real part of the resonant refractive index by correctly selecting the SBS medium. Since SBS is an instantaneous process, the temporal separation of the two beams will be much shorter than the time constants of the flame fluctuations. In fact, stimulated Brillouin scattering
is an attractive research field due to the interesting properties of the phase conjugate wave [Refs. 50, 51, 52, 53, 54, 55]. For a successful implementation of the SBS process in order to create the second wave required for the RHI experiments, several parameters have to be optimized. First, the Brillouin medium must be carefully chosen for a large gain (electrostrictive coefficient) to obtain a strong acoustic wave which in turn will strongly diffract the driving light wave. The reflectivity of the SBS medium increases with the intensity of the incident light pulse. The temporal reflectivity changes too, due to the changing input power and the time constant of the sound wave. With increasing pulse intensity the temporal shape of the phase-conjugate pulse approaches more and more the shape of the incident pulse. But even at very high powers an initial delay can be observed. Reduced phonon lifetime results in an increase of the threshold energy and a reduced slope of the energy reflectivity as a function of the input pulse energy [Ref. 48]. Additionally, the sound velocity must be sufficiently high, since this parameter determines the frequency shift introduced between the original and the phase-conjugate wave. In addition, the temporal and spatial characteristics of the driving wave are of importance. Tests have verified the polarization-state independence of the SBS process [Ref. 50]. Thus, no special consideration have to be taken into account concerning the polarization of the laser beams used in this work. Furthermore, the laser linewidth is of importance for the threshold intensity of the SBS process and for the reflectivity that can be achieved. The narrower the bandwidth, the lower the threshold intensity and the higher the reflectivity [Ref. 51]. The spatial quality of the beam driving the SBS is also of importance. For example, the reflectivity is almost doubled when improving the spatial quality of the laser from 50xD.L. to 20xD.L., where D.L. is the diffraction limit of the beam [Ref. 51]. Since SBS is a scalar process, the intensity of the original beam must therefore be optimized in the cell containing the Brillouin medium. To obtain low SBS threshold and high reflectivity, the coherence length should exceed the longitudinal sound wave extension so that an efficient coherent wave interaction is possible. Only in cases where the reflection takes place in a sufficiently small volume,
are the phases expected to be correlated and to yield a perfect phase-conjugation. Thus the focusing lens and its position must be carefully chosen by considering the distribution of the reflectivity in the focal range in the medium as a function of the applied energy at a given pulse length. The sound wave is built mainly in front of the incident laser beam and depending on the focal length of the entrance lens, the sound wave may be sharply concentrated in the cell or distributed over the whole cell length. Thus the sound wave extension may considerably exceed the Rayleigh length of the incident laser beam [Ref. 48].

The gain length, the efficient length where the SBS process takes place, is defined by the shortest of (i) the SBS medium length, i.e. the cell length, (ii) ~5 times the Rayleigh range or (iii) ~3 times the coherence length [Ref. 51]. The cell size, in relation to the depth of focus (DOF) is of importance. It has been experimentally shown that high phase conjugate fidelity is obtained for a gain length that is at least three times the DOF of the input beam [Ref. 50]. Furthermore, the good phase reproduction was independent of the f-number of the entrance lens. Also, high phase fidelity will be independent of the laser bandwidth provided the coherence length is much longer than the DOF [Ref. 50]. A minimum SBS threshold is obtained if the interaction region of the process is equal to or greater than three times the DOF [Ref. 50].

Implementation of the SBS process

As mentioned above, the SBS medium must have a large sound velocity to induce a large frequency shift, and a high gain to obtain a high reflectivity for the phase-conjugate beam. In the Table 3 presented below, the properties of some SBS mediums are presented. The sound velocity is calculated by $v_{\text{sound}} = \left( \gamma / \rho C_T \right)^{1/2}$, where $\gamma$ denotes the specific heat ratio and $C_T$ the compressibility of the medium. The values used in these calculations can be found in the Handbook of American Institute of Physics, 2nd edition, McGraw-Hill, 1963.
Table 3: The sound velocity, gain coefficient and induced frequency shift for the SBS media investigated in this work.

For this experimental work, n-hexane is found to be an appropriate SBS medium. n-hexane creates a frequency shift on the order of the FWHM widths of the NH and OH transitions for the flame conditions used in these experiments and exhibits a high gain. Furthermore, it is a liquid easing its application in an experiment. A quartz cell of diameter $\theta = 19$ mm and a cell length of 10 cm is used. Following the work of Menzel [Ref. 48], a $f = 200$ mm lens focuses the laser beam and the focal point is positioned $\sim 10$ mm in front of the end surface of the cell.

As mentioned above, the Lambda Physik 3002 laser does not produce a typical Gaussian beam. The only data available on the laser beam is its beam spot, $d = 2.5$ mm, and its typical divergence $\theta_L = 2.5$ mrad. The deviation from a diffraction limited Gaussian beam is given by the ratio, $m$, between the divergence of the laser and the far-field angle of a TEM$_{00}$ beam, $\theta_0 = \lambda/\pi w_0 n$. $w_0$ is the beam waist radius as defined by $w_0 = d/2$. Thus $m$, the deviation from the diffraction limited beam can be estimated as $m \times$ diffraction - limited $= \frac{\theta_L}{\theta_G}$.

For a diffraction limited beam of diameter $d$ and wavelength $\lambda$, focused by a lens of a focal length $f$, the Rayleigh range, $z_R$, is defined by:

$$z_R (D.L.) = \frac{\lambda f^2}{\pi d^2}.$$
Following Reference 51, it is assumed that a beam of a divergence of m x diffraction-limit will show a Rayleigh length similar to a diffraction limit beam of 1/m x the diameter of the diffraction-limited beam, i.e.

\[ z_R(m \times D.L.) = \frac{m^2 \lambda f^2}{\pi d^2}. \]

The Depth of Focus (DOF) can be estimated as \( \text{DOF} = \frac{0.32 \pi d_f^2}{\lambda} \), where \( d_f \) is the diameter of the focused beam. \( d_f \) is given by Selt’s formula [Ref. 55]

\[ d_f = d \left[ \left( 1 - \frac{s}{f} \right)^2 + \left( \frac{z_R}{f} \right)^2 \right]^{-\frac{1}{2}}, \]

where \( d \) is the beam waist and \( s \) is the distance of the position of the beam waist, i.e. the center of the laser cavity, to the lens.

<table>
<thead>
<tr>
<th>NH mode @ ( \lambda = 335.925\text{nm} )</th>
<th>OH mode @ ( \lambda = 308.396\text{nm} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 x coherence length</td>
<td>75 cm</td>
</tr>
<tr>
<td>5 x Rayleigh range</td>
<td>40.4 cm</td>
</tr>
<tr>
<td>3 x DOF</td>
<td>3.32 cm</td>
</tr>
<tr>
<td>SBS medium length (cell)</td>
<td>10 cm</td>
</tr>
</tbody>
</table>

Table 4: The different parameters for the two modes of operation of the Lambda Physik laser used to characterize the SBS process.

An estimate of the relevant parameters for the Lambda Physik laser operating in the NH and the OH mode is summarized in Table 4. With these parameters the gain length in our experiments is equal to the cell length. Since the cell length is ~ 3 times the DOF, a good phase-conjugate fidelity is expected, independent of the f-number of the entrance lens, as well as minimum SBS threshold.

A similar set up to that illustrated in Figure 22 is used to investigate the phase conjugate mirror applied in these experiments. In extension, a simple Michelson interferometer has been established. The interference fringes display a higher contrast as compared to the original beam. This is attributed to the filtering of the beam during the SBS process. Broadband emission of the laser (Amplified Stimulated Emission) is
Resonant Holographic Interferometry on NH and OH

highly suppressed by the SBS process due to its broadband nature and its relatively low intensity. The coherence length is similar to the one of the original beam suggesting that the SBS process produces indeed a replica of the original beam.

By simply filling the cell with n-hexane, a satisfactory SBS signal was achieved displaying high pulse to pulse stability. However, optical breakdown occasionally occurred even at low incident power due to impurities present in the medium. Even though the n-hexane is of Uvasol quality, dust and other particulates cannot be excluded. Instead of using complicated purification schemes [Ref. 52], the following procedure has been applied. A glass syringe is filled with n-hexane and filtered by utilizing a 0.2 μm pore size Millipore filter (PTFE). The cell is flushed several times with filtered n-hexane before filling it. This method proves to be a very fast, convenient and inexpensive way to clean the phase-conjugating medium.

As expected from the theoretical considerations above, the use of different lenses does not show any influence on the quality of the SBS beam when examined with the Michelson interferometer. The best reflectivity is obtained with a f = 200 mm lens. The reflectivity versus laser bandwidth and input intensity has been investigated. When operating the laser without the intracavity etalon (δλ = 0.2 cm⁻¹) a reflection efficiency of the SBS mirror of approximately 20% is achieved with a weak dependence on the input intensity. By using the intracavity etalon, the reflectivity improved to approximately 50%. Driving the SBS mirror with an intensity of 1.5 mJ/pulse, 350-400 μJ/pulse were available for the experiments. Unfortunately, such energies impede a two reference arm set up, since the intensity distribution for the expanded beams is insufficient to expose the holograms. Consequently, the one reference arm holographic set up has to be applied.

To determine the frequency shift obtained by the SBS set up, the following scheme is used. The laser, working on the NH mode, is first tuned to a resonant transition of NH. The Rhodamine dye cell is positioned on the hologram plane. Subsequently, the laser is
manually scanned over the molecular transition by moving the grating and etalon, while the 2D absorption image is viewed on a TV screen, tuned for highest contrast. In a next step, the frequencies are determined where no absorption could be observed. The two positions are referred to as the blue and the red by considering their position relative to the center wavelength. Control of the etalon position is achieved by the laser unit. Note that one etalon driver step represents a wavelength change of $-7 \times 10^{-5}$ nm ($0.0062$ cm$^{-1}$ / 186 KHz) for the NH mode, and $-7.5 \times 10^{-5}$ nm ($0.0079$ cm$^{-1}$ / 236 KHz), for the OH mode, respectively. Such a resolution is sufficiently accurate for the wavelength measurements. The absolute wavelength is determined by using the equation controlling the etalon driving motor in the motherboard of the laser. As the points of zero-absorption must be visually detected, an ambiguity in finding the correct position remains. Therefore, the process has to be repeated several times in order to get some statistics on the error of the blue and red wavelength position. The two positions are detected with an uncertainty of ±2 etalon steps. The center wavelength is determined from these two wavelength measurements with an accuracy of better than 0.00015 nm as indicated below:

<table>
<thead>
<tr>
<th>Mode</th>
<th>Q$_{5}(5)$</th>
<th>335.9255 nm</th>
<th>±0.00014 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>29768.5078 cm$^{-1}$</td>
<td>±0.0124 cm$^{-1}$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mode</th>
<th>R$_{2}(17)$</th>
<th>307.8966 nm</th>
<th>±0.00015 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>32425.7895 cm$^{-1}$</td>
<td>±0.0158 cm$^{-1}$</td>
<td></td>
</tr>
</tbody>
</table>

This procedure is repeated for the original and the phase-conjugate beam. A frequency difference for the center wavelength $0.284 \pm 0.0248$ cm$^{-1}$ or $8.51 \pm 0.74$ GHz is obtained. The shift is slightly lower than the theoretical value calculated for n-hexane. However, the value is within the uncertainty limits of the measuring procedure.

The same set up for SBS is used for the OH mode of the laser. Unfortunately, the implementation of SBS has not been successful in this case. Even though a sufficient reflectivity is measured, the expanded phase-conjugate beam is displaying significant intensity fluctuations from pulse to pulse that impede any 2D measurements.
Additionally, optical breakdown occurs in the SBS medium that is caused by the high peak intensity values rather than impurities. In both modes of laser operation the SBS is pumped with ~1.5 mJ/pulse. The pulse duration of the Nd:YAG laser is 6ns as compared to the 25 ns width of the Excimer laser. Thus, the peak intensity is increased by a factor of four. Changing the focusing lens in front of the SBS medium eliminates the optical breakdown. However, the strong intensity fluctuations are not removed. Two possible explanations are given for this effect. For the NH mode, the UV light is created directly from the dye laser (PTP dye). On the other hand, for the OH mode the UV is generated by frequency doubling in a KDP crystal. Since frequency doubling is quadratically dependent on intensity small fluctuations will be amplified. The nonlinear SBS process will further amplify these fluctuations and result in intense pulse to pulse variations. Furthermore, the frequency doubling process tends to shorten the pulse duration and to sharpen its leading edge. In fact, it has been experimentally verified that the shot-to-shot SBS conjugated fidelity is sensitive to the slope of the rising edge of the input pulse [Ref. 56]. For a pulse exhibiting a rise time that is short compared to the acoustic lifetime, the SBS gain may exceed threshold over a relative large cell volume near the focus before the pump wave is significantly reduced by SBS extraction. This leads to the amplification of noise modes, a process that is in direct competition with the phase-conjugation by SBS. Unfortunately, a gradually increasing slope of the temporal pulse profile that is favorable for the generation of the phase-conjugate beam is not provided by the OH laser mode. This was experimentally confirmed by the driving the SBS process with the fundamental radiation of the Dye laser, i.e. no frequency doubling. The expanded beam showed again the random intensity fluctuations. Attempts to find different SBS media have been made. In particular, the use of gases exhibiting high optical breakdown intensities seemed promising. SF$_6$, CO$_2$ and H$_2$ are investigated in a high pressure cell. For the SF$_6$, the reflectivity has been found to be a few percent only. As a consequence no further investigations were carried out. This is in accordance to the fact that the gain coefficient of SF$_6$ is an order of
magnitude smaller than for n-hexane. For CO\textsubscript{2} a high reflectivity (~50\%) is achieved but the frequency shift created by the SBS process amounts to only ~1.6 GHz due to the low sound velocity (267.8 m/s @ 25 bar pressure). H\textsubscript{2} is expected to be a good SBS medium, since the sound velocity in H\textsubscript{2} is similar to the velocity in liquids. However, the dominating process occurring in H\textsubscript{2} is rather Raman shifting than SBS. Though the 1\textsuperscript{st} Raman shift should diminish at a pressure of 4-5 bar it was still evident at high pressures up to 25 bar. The intense wave created by the 1\textsuperscript{st} Raman shift is also counter propagating, just as the SBS wave, and would mask a weaker phase-conjugate signal. The low SBS efficiency in H\textsubscript{2} is probably due to the small electrostrictive coefficient. No further trials were attempted to achieve SBS with the available laser in the OH mode.
Absorption measurements

The 2D absorption measurements are performed on the center frequency of a transition line. Errors in adjusting the laser on the center of a line within the accuracy described above (±0.00015 nm) are found to be negligible. All images are recorded at the position of the hologram plane in order that the recorded wavefronts are directly comparable to the exposure wavefronts. The laser is first tuned to the center and a single shot image is recorded on the CCD camera and stored in a computer for further processing. Then the laser is tuned off any resonance and a second single shot image is recorded on the CCD camera. The two images are normalized in areas where no absorption took place. Finally, the resulting absorption image is deduced by a pixel-wise division of the ON resonance image by the OFF resonance image. For a successful implementation of this scheme it is necessary that the intensities of the wavefronts, after passing through the flame, are correctly recorded. The linear response of the measuring technique to intensity variations must be controlled. Therefore, a square cell with quartz windows is positioned exactly at the position of the hologram plate. This cell is filled with a Rhodamine dye solution of a density assuring that the UV radiation is absorbed within the first mm of the solution. The fluorescence of the dye can then be detected by a regular CCD camera (SONY CCTV camera, WV-BP310 and Nikkor lens f = 105 mm), instead of using a UV sensitive camera. Due to the linearity of the fluorescence response of the dye to the incoming intensity, this technique has been often utilized for absorption measurements [Ref. 57, 58]. The linearity of the CCD camera is controlled by using Normal Density (ND) filters and a light bulb illuminating a white sheet of paper. The ND filters (ND = 0.025-2.00) are inserted in front of the camera that imaged the sheet. Several images are recorded with each ND filter. A linear response is found within the error of the specified ND filters. Note, that the automatic gain control of the CCD camera is switched off to inhibit intensity balancing. Additionally, standard single line absorption measurements are performed with the set up shown in Figure 10 at
selected positions in the flame by scanning over a specific transition. The peak absorption is then compared with the 2D absorption at the same location.

Figure 24: 2D absorption measurements. Top left: 2D single shot recording of the wavefront by tuning the laser ON resonance. Top right: 2D single shot recording of the wavefront by tuning the laser OFF resonance. Bottom left: the normalized 2D absorption image. Bottom right: a profile of the normalized absorption at a height above the burner of $y = 2.54$ mm. On the bottom of the absorption profile the two walls separating the middle NH$_3$ slot from the adjacent O$_2$ slots are depicted.

In order to ensure that the intensity used for the line absorption measurements is the same as for the 2D absorption images and for the RHI experiments, the expanded beam
is used by introducing an aperture of ~1mm², see Figure 10. The signal from the two
photo-diodes are directed to two gated Boxcar integrators and the normalization of the
absorption signal is performed by a computer. Zero transmittance is determined by
simply blocking the light after the flame. Hence, the maximum absorption is directly
stored. As previously explained, utilization of narrow band lasers permits a direct
measurement of the absorption, i.e. no deconvolution with the laser bandwidth is
necessary. Typical single line absorption scans are found later in the text. There is an
excellent agreement of these two measurements techniques within an uncertainty limit
of ±5%. Figure 24 represents a typical 2D absorption measurement. The first two
measurements shown in the top row are the 2D images ON resonance and OFF
resonance. Below left, the normalized image is depicted. The normalized value in areas
where no absorption takes place is set to 100 and corresponds to a 100% transmission.
Below right, a profile of the 2D absorption measurement is shown at a pixel position
corresponding to a height above the burner surface of \( y = 2.54 \) mm. The axis gives the
position of the absorption profile relative to the symmetry axis of the burner. The
calibration markers on the images are used to perform the distance normalization (from
pixels to mm). The two walls separating the center fuel slot from the adjacent oxidizer
slots are graphically depicted at the bottom of the absorption profile.
Implementation of two color PC RHI on NH

[Graph with axes labeled and data points]

Figure 25: Absorption scan over the spectral range of the 0-0 electronic band of NH. The set up shown in Figure 10 is used.

As discussed above, the intensity of the probe waves used for addressing the specific transition must be carefully chosen in order to avoid saturation effects. Since the coherence dephasing rate, $\gamma_{12}$, and the population decay rate, $\Gamma_0$, is not available from the literature, similar collisional relaxation rates are assumed as for OH. By using $\gamma_{12} = \Gamma_0 = 1.13 \times 10^{10} \text{s}^{-1}$ [Ref. 4] and estimating the transition dipole moment $\mu_{12} = 0.55 \text{D}$, we obtain an estimate on the saturation intensity of $I_{\text{sat}} = 0.56 \text{ MW/cm}^2\text{cm}^{-1}$. Considering a pulse duration of ~25 ns and a laser width of 0.04 cm$^{-1}$, we can estimate the intensities of the probe beams. For a typical operation, energies of ~20-30 μJ/cm$^2$ for the expanded probe beams are measured by using a high resolution pulsed Joule meter. The resulting intensities of the probe waves are $I_{\text{probe}} = 20-30 \text{ KW/cm}^2\text{cm}^{-1}$, well below the saturation intensity.

It is important that the selected transition line is spectrally isolated from other transitions. When examining the real part of the complex refractive index, see Figure 8, it is observed that for a Voigt profile the wings extend far out of the resonance domain.
Therefore, the transition line must be chosen such that the wings of nearby transitions have declined sufficiently. In Figure 25 an absorption scan over the congested 0-0 electronic band of NH is shown. A part of the spectrum is listed on the table below.

<table>
<thead>
<tr>
<th>Transition</th>
<th>Center frequency (cm⁻¹)</th>
<th>Center wavelength (nm)</th>
<th>Δν (cm⁻¹)</th>
<th>Δν/Δν_D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Q₃(4)</td>
<td>29770.6781</td>
<td>335.90098</td>
<td>2.1703</td>
<td>7.23</td>
</tr>
<tr>
<td>R₁(1)</td>
<td>29770.6222</td>
<td>335.90161</td>
<td>2.1144</td>
<td>7.05</td>
</tr>
<tr>
<td>Q₃(5)</td>
<td><strong>29768.5078</strong></td>
<td><strong>335.92547</strong></td>
<td>1.0776</td>
<td>3.59</td>
</tr>
<tr>
<td>Q₂(2)</td>
<td>29767.4302</td>
<td>335.93763</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The Q₃(5) transition has a separation to the nearest neighbor Q₂(2) of 3-4 times its FWHM. This line seems to be the best choice since it exhibits, for these flame conditions, a peak absorption of approximately 50% (about 25% absorption at the exposure wavelengths). The center wavelength is defined with the procedure described above to be λ₀=335.92547±0.00014 nm. The absorption scans indicate a Voigt width of 0.3 cm⁻¹ giving an α parameter of α = 0.203 (T=2335K).

Having selected the appropriate transition line we proceed in the fine alignment of the holographic set up. In Figure 26 the final set up as used for the recording of the two color resonant holographic is presented. The first beam splitter reflects 50% of the incoming pulse towards the SBS cell. The counter propagating phase conjugate pulse is transmitted by to the same beam splitter and 50% of its intensity is available for the experiment. The reflected 50% is directed towards the laser. To avoid amplification of this counter propagating pulse in the dye laser an optical delay is introduced between laser and SBS cell. The time separation ensures that the counter propagating pulse does not return while the pumping process of the laser is in progress. Moreover, the optical delay excludes any interference of the original and the phase conjugate beam on the hologram plane. Such an interference can result in spurious holograms disturbing the correct read-out of the holograms. However, the interference is advantageously used to align the two beams prior to exposure as will be shown for the OH experiments [Ref. 59]. For the NH experiments, this adjustment technique can not be not used due to the
optical delay that prohibits the interference of the two probe beams on the hologram plane. The position of the SBS cell yields a sufficient time delay between original and phase-conjugate beam of 25-30ns, a time much shorter than the relevant flame constants. Obviously, the original beam is more intense. Therefore, it is attenuated to the intensity level of the phase-conjugate beam by means of a beam splitter positioned between the two 50/50 beam splitters depicted in Figure 26. The reflection of this beam splitter was directed to a high resolution etalon (FSR=0.2 cm⁻¹) monitoring the single mode operation of the laser during scanning, see Figure 27.

**Figure 26:** Holographic set up for performing two-color Phase conjugate Resonant holographic recordings.

Theory and absorption scans show that the target species concentration in this flame gives raise to a Fringe Shift of a few present. If the holographic set up is configured in a way that interferograms of infinite fringes are recorded, i.e. the two read out waves are parallel to each other, a problem concerning the fringe interpretation would arise. In this configuration the fringe shift is detected as a reduction of the interferogram intensity by
a few present in positions were the probe wavefronts interact with the target species. However, this change can also occur by intensity attenuation due to absorption that reduces the visibility of the interferogram. An ambiguity arises on the cause of the intensity reduction, absorption or a fringe shift. The system is therefore aligned such that finite fringes are introduced in the read-out interferograms. This is achieved by introducing a small angle between the original and the phase-conjugate beam. By positioning the two 50/50 beam splitters correctly, a vertical angle in respect to the input plane of the set up is achieved. The bigger the angle the more fringes appear as is expected for an interferometer. Any phase difference introduced by the real part of the resonant refractive index of the two exposure wavefronts will result in a deviation from the parallel equidistant finite fringes.

![Figure 27](image.jpg)

**Figure 27:** Fringes indicating the single frequency operation of the laser, monitored by an external etalon of FSR = 0.2 cm⁻¹.

In order to obtain a high visibility (V≈1) at locations on the holograms where the wavefronts interact with the target species, the wavelengths of the two probe beams are carefully adjusted symmetrically to the center frequency of the transition (see above).
An asymmetric tuning of the two wavelengths in respect to the center line would result in an unequal absorption. As a consequence, areas of the interferogram where absorption takes place would appear as reduced fringe contrast and obscure the measurement. The 2D absorption images for each of the two wavefronts are recorded during alignment of the flame in respect to the two probe wavefronts. The 2D absorption images are recorded while turning the burner until the width of the absorption profile is minimized and the absorption maximized, indicating that the two probe wavefronts pass parallel to the flame front.

Off resonant double exposure image plane hologram far from the $Q_d(5)$ electronic transition of NH.

$\lambda_0 = 335.9255$ nm

$\lambda_1 = 335.9224$ nm, $\lambda_2 = 335.9192$ nm

On resonant double exposure image plane hologram in the proximity of the $Q_d(5)$ electronic transition of NH.

$\lambda_0 = 335.9255$ nm

$\lambda_1 = 335.9239$ nm, $\lambda_2 = 335.9271$ nm

**Figure 28:** Eliminating the bulk refractive index. On the left image the finite reference fringes created by a slight angle between the two probe wavefronts are seen. On the left side the deviation of the finite reference fringes due to the interaction of the two probe wavefronts with the target species is shown.

The ability of the PC RHI to eliminate the bulk refractive index is controlled in the following manner. By tuning the two probe waves off any resonance, a single shot, two-
color experiment is performed. In Figure 28 the resulting interferogram is shown. Finite fringes of excellent contrast are seen with no deviation in positions where the two wavefronts interact with the flame front. The small declination of the reference fringes indicates an imperfect projection of the wavefronts by the relay lens. Comparison with the flame On/Off interferograms, Figure 20, indicates, that indeed the bulk refractive index, though orders of magnitude stronger than the resonant refractive index, is interferometrically completely canceled. The interferogram shown on the right side of Figure 28 corresponds to the situation where the frequencies of the two probe wavefronts are adjusted symmetrically to the center frequency of the $Q_3(5)$ transition of NH. The deviation of the fringes in positions where the two probe waves interact with the target species is clearly seen. For comparison on the relative position of the fringes shifts, refer to Figure 30 where a normalized 2D absorption image is shown.

In summary, the experiments confirm the theoretical predictions that the holographic set up is able to record fringe deviations due to the resonant refractive index of a target molecule. A trade off between fringe resolution and spatial resolution has to be made in order to apply the technique to flame diagnostics. Experimentally, fewer fringes are introduced by decreasing the angle between the two probe waves.

A series of PCRHI experiments are performed in order to investigate the applicability of the PC RHI technique for quantitative measurements of species concentration in a flame. Experiments are performed in such a way that the results of the RHI measurements are comparable with the 2D absorption measurements. The comparison with an established technique will indicate the applicability of the technique.

First, a 2D absorption measurement is performed with the phase-conjugate wave blocked. Subsequently, the dye cell is substituted with a hologram plate. The laser is tuned such that the frequencies of the two probe wavefronts are symmetrically adjusted to the center frequency. A single shot, two color exposure is performed. Finally, original laser beam is readjusted to the peak absorption and the procedure is repeated. In this
way four holograms are recorded and six 2D absorption images. By following this procedure, we ensure that the flame conditions are the same for all experiments but at the same time, each measurement is new in terms of determining the exact laser frequency. In Figure 29 two of the four recorded holograms are presented. No degradation of the fringe visibility in areas of the interaction with the target species is observed. This indicates a correct positioning of the wave frequencies in respect to the center frequency. Additionally, by comparing the two holograms a small change in the position of the finite fringes is observed. The effect is due to a slow beam walking originating from the laser itself or from temperature dilatation of the optical table.

![Image of interferograms](image-url)

**Figure 29:** Two examples of the interferograms recorded in the proximity of the \( Q_3(5) \) transition line of NH by PC RHI.

To quantify the fringe shifts, the interferograms are recorded and stored in the computer as 512x512 pixel images. Careful illumination assures that the images are not saturated in intensity (gray scale recording in the range from black to white [0..255]). These digitized images are processed with Adobe Photoshop V 3.1 providing the pixel position and the gray scale value. The fringes show an excellent modulation with a clear minimum and maximum. The fringe position is always measured on the top side of a fringe as the first pixel with a zero gray-scale value in a top-down direction. The difference in pixels for one fringe spacing, i.e. one wavelength difference, is determined
Resonant Holographic Interferometry on NH and OH

by subtracting the pixel value of two adjacent fringes and denoted by $I_{sp}$. The fringe shift due to resonance is measured by averaging two pixel positions on the left and right side of the shifted fringe and subtracting the pixel value for its maximum, $I_{sh}$. The fringe shift in fractional wavelength units is then given by $I_{sh}/I_{sp}$. The positions of reference fringes are normalized in height by using the calibration markers. As an error in determining the Fringe Shift position a worst case calculation is given taking an uncertainty of one pixel in determining the first zero-value pixel. This error is then introduced for the calculation of the uncertainty in the species concentration.

Figure 30: A typical example of the six (6) normalized 2D absorption images performed on the center line of the $Q_3(5)$ transition of NH. To the right, a profile of the 2D absorption image at a specific height above the burner ($x = 1.25$ mm) is shown. On the bottom of the absorption profile the two walls separating the center NH$_3$ slot from the adjacent O$_2$ slots are depicted.

For the absorption images the On resonance images are normalized with the Off resonance images and calibrated by using the position markers. Profiles are obtained at a position that corresponds to the location of the fringe shift measurements of the four interferograms. In Figure 30 a representative example of the six absorption images is shown in addition to a absorption profile at $x = 1.25$ mm above the burner surface. The absorption profile in terms of pixels is rescaled to a real position (mm) relative to the
symmetry axis of the burner, i.e. the zero point of the y-axis, using the markers at the top of the absorption images. The standard deviation of the six measurements indicates the error of the measurement. This procedure is used for both flame sheets. The data for fringe shift and absorption is then used in the simulation program computing the Voigt profile, equation (23) and (24). For the calculations a fixed value of the temperature is used, \( T = 2335 \, \text{K} \), and the program is run with the species concentration as a variable. In Table 5, the results as derived from the simulation program for one position in the flame \((x = 1.25 \, \text{mm})\) is summarized.

<table>
<thead>
<tr>
<th>(N_{\text{tot}})</th>
<th>(\alpha_0)</th>
<th>(1-\exp[-\alpha_0 L])</th>
<th>(n(v_1))</th>
<th>(n(v_2))</th>
<th>Phase shift (\Phi_1)</th>
<th>Phase shift (\Phi_2)</th>
<th>Fr.Sh. ((\Phi_1 - \Phi_2)/2\pi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(5.99 \times 10^{20})</td>
<td>14.24</td>
<td>0.45</td>
<td>1.0000002304</td>
<td>0.9999997696</td>
<td>0.18097</td>
<td>-0.18097</td>
<td>0.0576</td>
</tr>
<tr>
<td>(10.51 \times 10^{20})</td>
<td>24.96</td>
<td>0.65</td>
<td>1.0000004042</td>
<td>0.9999995958</td>
<td>0.3175</td>
<td>-0.3175</td>
<td>0.1011</td>
</tr>
</tbody>
</table>

Table 5: Computation results for the experimental data at position \(x = 1.25 \, \text{mm}\) above the burner surface. The two highlighted cells contain the experimentally determined absorption and fringe shift used to compute the other values.

In Figure 31 the NH concentrations in units of particles per million (ppm) are plotted as a function of the height above the burner for illustrative reasons. The error bars are derived from the measurement errors. An error of \(\pm 1\) pixel in determining the measuring positions results in an species concentration error of \(\pm 15\%\). Alternatively, an error of \(\pm 5\%\) in the absorption measurements results in a species concentration error of \(\pm 15\%\). The concentration determined by PC RHI and 2D absorption measurements differ by a factor 2. Furthermore, the absorption that corresponds to the concentration predicted by the experimentally determined fringe shifts is too high. On the other hand, the fringe shift corresponding to the measured absorption is of the order of 5-6\% of a fringe, thus we once again have a discrepancy of a factor of 2, see also Table 5 for the numerical details.
Figure 31: Presentation of the experimental data from PC RHI and 2D absorption measurements. Concentrations derived from fringes shifts and absorption are presented as a function of the height above the burner for an assumed temperature of 2335 K. The two dotted lines correspond to the absorption measurements and the two solid lines to the PC RHI measurements. Bars denote a 15% error for absorption and for RHI.

Since the formulation of the complex refractive index is derived from fundamental electromagnetic theory, the discrepancy is most probably due to an experimental error. The 2D absorption images are compared with standard line-of-sight measurements at different positions in the flame and a excellent correspondence is found, see the chapter concerning the absorption measurements. Hence, the discrepancy that is observed has to origin from the RHI measurements. An inaccurate tuning of the exposure frequencies in respect to the center frequency results in a decrease of the signal since the probed refractive index is decreased. Also, the influence due to the wings of adjacent transitions decreases the signal since their contributions will have an opposite sign for the two exposure frequencies. The detection of the maximum fringe shift has been investigated as well. Even a crude inspection of the interferogram clearly shows a fringe shift of 10 % rather than 5-6%. Up to now no obvious explanation can be given for the
discrepancy between the theoretically predicted and measured fringe shift. The conclusion, that some other process might influence the PC RHI technique has to be drawn. First, recall equation (23) assuming two parallel interfering rays parallel, apart from an small tilt to produce the finite fringes. A more careful study of the optical path of the two beams is therefore required. Second, the influence of the refractive index gradients giving rise to lensing effects must be investigated. Third, the influence of heat dissipation created by the high absorption of the first pulse on the refractive index has to be analysed. Such a variation in refractive index might be experienced by the second pulse and distort the measurement of the fringe shift. The analysis of these three processes can be found in the chapter following the presentation of the experiments performed on the second species of investigation: the OH radical.
Implementation of two color RHI on OH.

Finding an appropriate transition for the case of the OH radical is a slightly easier task than for the NH radical. The spectrum is less congested and sufficiently isolated transitions are found. The one dimensional simulations indicate that the species concentration for OH are expected to be two orders of magnitude higher than the measured NH concentrations, see Appendix B.

![Absorption scan over the R2(17) transition of OH. The set up shown in Figure 10 is used.](image)

Figure 32: Absorption scan over the R2(17) transition of OH. The set up shown in Figure 10 is used.

The problem is primarily to find an isolated transition with an absorption on the order of 50%. Several absorption scans have been performed and the R2(17) transition has been found to meet the requirements. The smallest distance to an adjacent transitions is $\Delta \tilde{\nu} = 3.153$ cm$^{-1}$.

<table>
<thead>
<tr>
<th>Transition</th>
<th>Center frequency (cm$^{-1}$)</th>
<th>Center Wavelength (nm)</th>
<th>$\Delta \tilde{\nu}$ (cm$^{-1}$)</th>
<th>$\Delta \tilde{\nu}/\Delta \nu_D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>R1(18)</td>
<td>32475.279</td>
<td>307.9265</td>
<td>3.153</td>
<td>9.8</td>
</tr>
<tr>
<td>R2(17)</td>
<td>32478.432</td>
<td>307.8966</td>
<td></td>
<td></td>
</tr>
<tr>
<td>R2(3)</td>
<td>32489.479</td>
<td>307.3919</td>
<td>11.058</td>
<td>34.5</td>
</tr>
</tbody>
</table>

The relatively large distances to the neighboring transitions eliminates the influence on the measurements of the resonant refractive index.
With $\gamma_{12} = \Gamma_0 = 1.13 \times 10^{10} \text{s}^{-1}$ [Ref. 60] and $\mu_{12} = 0.245 \text{D}$ an estimate on $I_{\text{sat}} = 1.4 \text{MW/cm}^2\text{cm}^{-1}$ is obtained, in agreement with experimental results [Ref. 3]. For typical operation conditions the expanded probe beams have energies of $\sim 20-30 \mu\text{J/cm}^2$. With a pulse duration of $\sim 6\text{ns}$ and a laser width of $0.04 \text{cm}^{-1}$ the intensity of the probe waves is $I_{\text{probe}} = 83-125 \text{KW/cm}^2\text{cm}^{-1}$ well below the saturation level.

As mentioned above, it is not possible to apply SBS on the UV pulses from the laser operating in the OH mode. Therefore, a second laser is employed. Unfortunately, the available laser system produced an output exhibiting a broad laser pulse width of about $0.2 \text{ cm}^{-1}$. Such band widths decrease the RHI signal significantly, as mentioned in the chapter on laser considerations. Assuming a gaussian intensity distribution for employing the narrow band laser ($0.04 \text{ cm}^{-1}$) and the broad band laser ($0.2 \text{ cm}^{-1}$) the estimated signal reduction is less than 15%. Note however, the signal can be rescaled if the spectral intensity distribution is known.

More specifically, a Nd:YAG (Continuum NY81, Santa Clara, USA) pumped dye laser (Continuum ND60) is used in order to generate the second pulse. Initial double exposure holographic experiments have shown that the laser produces reproducible phasefronts, in contrast to the alternative laser system available (Nd:YAG pumped Quanta-Ray PDL-3 pulsed dye laser). Even though the PDL-3 laser would be the first choice by exhibiting a bandwidth on the order of $0.07 \text{ cm}^{-1}$, its phase reproducibility has been poor causing random fringes on the interferograms that varied from pulse to pulse. The specifications of the Continuum laser is summarized below.

<table>
<thead>
<tr>
<th>Dye (nm)</th>
<th>Doubling</th>
<th>Pump laser</th>
<th>Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>OH</td>
<td>Rhodamine 101</td>
<td>KDP</td>
<td>Nd:YAG @532nm</td>
</tr>
<tr>
<td></td>
<td>614-672</td>
<td>307-336</td>
<td>$\sim 275 \text{ mJ/pulse}$, $6 \text{ ns pulse duration}$, $\Delta \tilde{\nu} = 0.2 \text{ cm}^{-1}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>$\sim 3-4 \text{ mJ/pulse}$</td>
</tr>
</tbody>
</table>

Fixed Rep. rate: 20 Hz

It is essential for the experiments to synchronize the firing of the two pump lasers. This proved difficult as the two pump lasers run at different fixed repetition rates of 10 and
20 Hz respectively. A configuration of two delay generators (Stanford Instruments DG 535) is applied to trigger individually the flash lamps and the trigger for the Q-switches. With a third delay generator the delay between the two pulses is controlled.

In order to control the wavelength of the two laser systems, a part of the fundamental radiations is directed to a high resolution wave meter (Burleigh Instruments). The Continuum laser exhibits a wavelength jitter of $\pm 0.05\text{cm}^{-1}$ (~$0.5 \times 10^{-3}\text{nm}$) which is not removable, even by a careful alignment of the laser cavity. The wavelength jitter of the Lambda Physik laser is an order of magnitude lower, indicating the good operation of the intracavity etalon and the stability of the laser cavity.

![Diagram](image.png)

**Figure 33:** Holographic set-up for performing two-laser, two-color Resonant holographic recordings. Here the first beams splitter is removed and the second laser beam is introduced similar to the phase-conjugate beam.

Apart from using a second laser instead of the phase-conjugate beam the optical set up for the OH experiments remains in essence the same. The first beam splitter is removed and the second laser is introduced as shown in Figure 33. The two lasers used in the experiment are investigated in respect to their divergence. Using SBS to produce the
second pulse, this control can be omitted due to the phase-conjugate nature of the pulse. As previously stated, two pulses of the same frequency impinging simultaneously on a screen or a dye cell will interfere due to the high coherence of the two pulses. By careful alignment, the divergence of the two lasers is studied by inspection of the interference fringes. In such a way the lasers are optimized for similar divergence. The Figure 34 shows the two laser interference fringes for an optimized alignment. Note that the visibility of the fringes is quite low. The parallelity of the fringes as shown in Figure 34 indicate that, within the measurement accuracy, the two lasers have the same divergence.

![Image of interference fringes](image)

**Figure 34**: Interference fringes of two laser pulses on the dye cell.

These interference fringes are also used to align the optical system. As mentioned in the chapter on the NH measurements, an angle is introduced between the two probe beams in order to obtain finite fringes in the read-out interferograms. The same procedure is applied here. The observation of the interference fringes on a TV screen speeds up the aligning procedure dramatically.

Again, the frequencies of the two lasers have to be tuned symmetrically in respect to the center frequency of the $R_2(17)$ transition line of OH. Since the two frequencies are not locked by the fixed shift due to the SBS process, as for the NH experiments, the wave
meter is used for calibration of the two lasers. As mentioned above, the fundamental radiation of the two lasers is continuously monitored with the wave meter. The narrow band Lambda Physik laser is tuned to the center frequency in the same manner as for the NH experiments. The blue and red frequencies are found by scanning off the transition until no absorption is observed. The line center is then given by the mean value of the two frequencies. The Continuum laser is now tuned by utilizing the wave meter reading for the narrow band laser. The two lasers are subsequently positioned on the blue HWHM point and the red HWHM point of the R$_2$(17) transition, respectively. For a Temperature of $T = 2790$K, the theoretical value of the Voigt FWHM is $\Delta \tilde{\nu} = 0.32$cm$^{-1}$ ($\Delta \lambda = 0.003$nm). Thus, the two exposure wavelengths become:

| $\lambda_1$ | $307.8966 + 0.003/2 = 307.898$ nm | $32478.432 - 0.32/2 = 32478.272$ cm$^{-1}$ |
| $\lambda_2$ | $307.8966 - 0.003/2 = 307.895$ nm | $32478.432 + 0.32/2 = 32478.592$ cm$^{-1}$ |

**Figure 35:** Two examples of the interferograms recorded in the proximity of the R$_2$(17) transition line of OH by RHI by applying two lasers. $\lambda_0 = 307.8966$ nm and $\lambda_1 = 307.898$ nm, $\lambda_2 = 307.895$ nm.
The single shot experiments are performed by single triggering the Q-switches of the lasers by the delay generator. A delay of 25ns is chosen, which is equal to the time separation of the original and phase-conjugate beam in the NH experiments. The exposure intensities are controlled by changing the input pump intensities of the two lasers, rather than turning the angle of the doubling crystal. This method yields even intensity profiles of the probe beams and ensures a correct operation of the doubling crystal. Note that the spectral intensity of the Continuum laser beam is lower due to its broader bandwidth.

For the calculations, the temperature derived from the CARS measurements, $T = 2790$ K, is used and the program is run with the species concentration as a variable. In table 6 the results derived from the simulation program at one position in the flame ($x = 3.4$ mm) is listed. The same procedure is used for measuring the fringe shifts and the absorption as for the NH experiments.

Figure 36: Example of the six (6) normalized 2D absorption images performed on the center of the R$_2$(17) transition of OH. To the right a profile of the 2D absorption is given at a height of $x = 3.4$mm above the burner. On the bottom of the absorption profile the two walls separating the center NH$_3$ slot from the adjacent O$_2$ slots are depicted.
Table 6: Computation results for the experimental data at position x = 3.4 mm above the burner surface. The two highlighted cells contain the experimentally determined absorption and fringe shift that is used to compute the other values.

<table>
<thead>
<tr>
<th>N_{tot}</th>
<th>\alpha_0</th>
<th>1-\exp[-\alpha_0L]</th>
<th>n(v_1)</th>
<th>n(v_2)</th>
<th>\Phi_1</th>
<th>\Phi_2</th>
<th>Fringe Shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>m^{-3}</td>
<td>m^{-1}</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>81.7 \times 10^{20}</td>
<td>14.78</td>
<td>0.46</td>
<td>1.000000215</td>
<td>0.9999997850</td>
<td>0.18427</td>
<td>-0.18427</td>
<td>0.0586</td>
</tr>
<tr>
<td>166.3 \times 10^{20}</td>
<td>30.09</td>
<td>0.72</td>
<td>1.0000004376</td>
<td>0.9999995624</td>
<td>0.3751</td>
<td>-0.3751</td>
<td>0.1194</td>
</tr>
</tbody>
</table>

**Figure 37:** Presentation of the experimental data from RHI and 2D absorption measurements. Concentrations derived from fringes shifts and absorption are presented as a function of the height above the burner for an assumed temperature of 2790 K. The two dotted lines correspond to the absorption measurements and the two solid lines to the RHI measurements. Bars denote a 15% error for absorption and for RHI.

In Figure 37 all the results obtained from the evaluation of the three first reference fringes are plotted as a function of the height above the burner in units of parts per million (ppm). Note that the fringe shifts are rescaled assuming a gaussian intensity...
distribution for the broadband laser. In Figure 35 it is observed that the two flame sheets display a different response in contrary to the interferograms recorded for NH. The fringe shifts on the right flame sheet are about the half in compare to the fringe shifts on the left flame sheet. The different species concentration in the two flame sheets could be due to an asymmetrical gas flow within the three burner slots. The absorption measurements also indicate a small deviation. However, this deviation in absorption does not rationalize the fringe shift difference in the interferograms. On the other hand, a misalignment of the burner in respect to the incoming probe wavefronts could be considered in conjunction to the divergence of the beams. In fact, a high divergence of the two laser beams could result in probing the one of the two flame sheets parallel and the other at a slight angle.

As for the NH experiments, we find that a direct comparison of absorption and RHI is not feasible. The absorption measurements indicate lower species concentrations in respect to the concentrations obtained by two color RHI. Measured fringe shifts for the left flame sheet are between 19% and 33% of a fringe. That would require an absorbed intensity between 80 and 95 percent on the center of the transition!

Figure 38: Interferogram recorded in the proximity of the \( R_2(17) \) transition line of OH by RHI by applying two lasers, during the preliminary studies.

\[
\lambda_0 = 307.8966 \text{ nm and } \lambda_1 = 307.898 \text{ nm, } \lambda_2 = 307.895 \text{ nm}
\]
In Figure 38, a typical interferogram is shown recorded during the preliminary stages of the OH experiments. In this interferogram it is seen that similar fringe shifts are obtained for both flame sheets. The quality of the finite fringes is not sufficient for a digital read-out of the fringe shifts. The interferogram indicate a fringe shift between 30-35 %. The findings indicate that for the series of holograms used for evaluation, it is rather the high value fringe shifts that should be considered.

The large scattering of the measured fringe shifts data obtained by the four RHI experiments is of another concern. As shown in Figure 37, the four fringe shifts measured at a height position about $x = 10$ mm are:

<table>
<thead>
<tr>
<th>Hologram</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Height (mm)</td>
<td>10.16</td>
<td>10.21</td>
<td>10.16</td>
<td>9.9</td>
</tr>
<tr>
<td>Fringe Shift (%)</td>
<td>23.36</td>
<td>19.85</td>
<td>14.93</td>
<td>12.77</td>
</tr>
</tbody>
</table>

This scatter could be attributed to the frequency jitter of the Continuum laser. As mentioned above this laser displays a pulse to pulse frequency variation of $\pm 0.05$ cm$^{-1}$. By inspecting carefully the recorded interferograms, Figure 35, a slight decrease of the contrast appears in areas where the probe wavefronts interact with the OH target species. The visibility of the fringes decreases if the intensity of the two interfering wavefronts is not equal. Indeed, the 1$^{st}$ hologram exhibiting the largest fringe shift, appears to have a better contrast than the 4$^{th}$ hologram displaying the smallest fringe shift. By assuming a stable narrow band laser, a frequency shift of the Continuum laser towards the center of the line by the measured average jitter of 0.05 cm$^{-1}$ would induce a fringe shift decrease of $\sim 15\%$ and an absorption increase by $\sim 30\%$. Although these values cannot explain the substantial fringe shifts that have been determined experimentally, they clearly indicate the drawback of utilizing the two laser system RHI set up. The fact that the two frequencies are not locked in respect to each other by SBS leads to difficulties in correctly interpreting the interferograms. However, sufficient fringe shifts are achieved even with such large frequency deviations. This can be understood if the form of the real part of the complex refractive index as a function of
frequency is considered, see for example Figure 8. The flat top of the curve indicates that variations in frequency will not influence the induced fringe shift significantly. Another drawback of the two laser system is stated in the following. When utilizing the SBS technique, a phase-conjugated beam created for the second exposure wavefront. The phase-conjugation ensures a perfect match of the two probe wavefronts. Consequently, small pulse to pulse deviations occurring in the phasefronts are subtracted automatically during interferometric reconstruction. Unfortunately such a interferometric correction is not achieved for the two laser configuration. On the contrary, the pulse to pulse deviations of the laser beams lead to erroneous fringe shifts. Unfortunately, the obtained data is not sufficient for reliable quantitative OH species measurements. As for NH, a discrepancy is observed between the 2D absorption results and the two color RHI measurements. A discussion of these findings is given in the next chapter.
Discrepancy between RHI and 2D absorption measurements

In formulating the equations describing the two experimental techniques applied in this work, equation (23) and (24), it has been pointed out that two parameters are of importance, i.e. the species concentration and the temperature that is included in the partition function of the target species. By performing two individual measurements of the target species, absorption and RHI, these two parameters can be computed. It is important to mention that for a given temperature, the two experimental techniques should result in the same species concentration. The experiments presented in this work showed that this is clearly not the case. In the chapter describing the experiments performed on the NH molecule, three possible explanations for the discrepancy of the species predictions derived from the two techniques are considered:

- The influence of the finite fringes used in the set up on the fringe shifts.
- The change of the optical path length due to ray bending effects induced by the refractive index gradients in the flame.
- Heat release to the surroundings due to the high absorption.

To address the possibilities above, the first two points have been investigated theoretically, where the heat release effects have been investigated experimentally.

Fringe Shift and Finite Fringe spacing

To derive equation (24), which is describing the resulting fringe shift on the interferogram, it is assumed that the two probe waves, each with a different frequency, follow the same path through the flame. Additionally, the refraction of the beams due to the refractive index gradients present in flames is neglected and a straight line propagation is assumed.

As explained in detail in the experimental implementation of the RHI technique, finite fringes are introduced on the interferograms in order to ease the interpretation of the experiments. This is achieved by introducing an angle between the original and the
phase-conjugate beam in vertical direction to the input plane of the set up by correctly positioning the two 50/50 beam splitters. As expected for an interferometer, an increase in angle yields more fringes. The parallel and equidistant finite fringes are distorted by any phase differences between the two exposure wavefronts. This effect is observable for all the experimental results obtained in this work. Furthermore, the measurements indicate, that the refractive index variation is more substantial along the y-axis (perpendicular to the flame front) than along the x-axis (along the flame front), see for example Figures 31 and 37. This situation justifies the use of the term *stratified medium* as introduced in the theoretical part of this work.

As mentioned above, the angle between the two beams for finite fringe generation is aligned vertically in respect to the input plane of the set up. Therefore, the finite fringes will appear perpendicular to the flame front. Obviously, such an angle between the two probe wavefronts will result in different paths through the flame. In the following, it is assumed that the refractive index is only a function of y-axis, neglecting the small variations along the x-axis. The resonant refractive index is computed by taking into account the molecular constants of the $Q_3(5)$ transition of NH, see Figure 8 and related text. For all the calculations, we assume that the resonant refractive index extends over a length along the z-axis of $L = 0.042 \text{ m}$. By introducing an angle $\theta$ between the two beams, one trace through the flame is increased by $L / \cos(\theta)$. Note, that this angle is difficult to measure geometrically. On the other hand, the finite fringe spacing can be calculated with a high accuracy by using the calibration markers seen on all images is this work. The angle $\theta$ yields a fringe spacing of $d = \lambda_0 / \sin(\theta/2)$. The phase shift experienced by the tilted beam along $L / \cos(\theta)$ can then be calculated. Subsequently, the resulting fringe shift is computed for the two interfering beams as a function of the finite fringe spacing. The deviation from the fringe shift as calculated by equation (24) that assumes an infinite fringe spacing. In Figure 39 this deviation is shown. Additionally, the angle between the two probe wavefronts is also calculated and shown in the same Figure.
The angle introduced in the optical system will have an effect on the detected fringe shift only when the resulting finite fringes are in the micrometer range, see Figure 39. In the region of interest, i.e. finite fringes of the order of millimeters, the error is negligible small. Consequently, the discrepancy between the target species concentration derived from RHI and the absorption measurements cannot be attributed to the introduction of finite fringes in the optical set up.

![Figure 39](image)

**Figure 39**: The resulting error on RHI fringe shift measurements by introducing an angle between the probe beams to produce the finite fringes. The error is presented as percent deviation from the theoretical value assuming that both beams followed the same path. The angle between the two probe beams as a function of finite fringe spacing is also displayed.

*On the propagation of individual rays through a 2D flame*

From the theoretical consideration it is seen that a wavefront can be considered as an ensemble of individual rays. Thus, instead of investigating the complete wavefront propagation through a phase object, individual rays can be studied separately. The results are then combined to gain information on the entire wavefront propagation.
Taking into account the preceding concept, a model is constructed to describe the propagation of an individual ray through a phase object, which is here a 2D diffusion NH$_3$-O$_2$ flame stabilized on a Wolfhard-Parker slot burner. A detailed description of the burner is found in Appendix B. Since the flame is symmetric in respect to the burner axis, it is sufficient to model one flame sheet only. A description of the 2D refractive index distribution is given in the next section, followed by the numerical method developed in this work to simulate the propagation of the rays through the flame.

**Position relative to the symmetry axis of the W.-P. burner (mm)**

*Figure 40:* The absorption profiles of NH (solid) and OH (dotted) across the flame front at a height of $x=3.5$ mm. Additionally single point CARS temperature measurements are shown. On the bottom of the figure the two walls separating the center NH$_3$ slot from the adjacent O$_2$ slots are depicted.

In the chapter on the Theory it has been shown that the refractive index experienced by the probe wavefront along the path through the flame is given by $n'(v) = [n_0 + n(v)] - iK(v)$. $n_0$ is the bulk refractive index given by the Gladstone-Dale formula, equation (16), and $n(v) - iK(v)$ is the complex refractive index for a frequency of the probe wavefront in the proximity of a molecular transition as given by equations (23) and (24). The two contributions to the refractive index extend over the length of the burner slot, $L = 0.042$. 
m. Outside this area an ambient refractive index is assumed. By analyzing the normalized 2D absorption profiles, see Figure 40, the spatial distribution of the individual species in the flame is accurately obtained. Additionally, the point wise CARS measurements yield temperature profiles in respect to the symmetry axis.

As the light wave passes through the flame, it experiences, apart from the bulk, a refractive index due to resonance. Considering the species and temperature distribution, we assume substantial gradients perpendicular to the flame front, along the y-axis. Temperature and species variations as a function of height are insignificant for the simulations and are, therefore, neglected. We follow the formulation of the Theory chapter by assuming a wave propagation along the z-axis and a refractive index variation along the y-axis only. This situation corresponds to a *stratified medium*. The temperature and the species concentration profiles are closely approximated by using a Gauss function. Air is assumed to constitute the bulk refractive index which is dependent on temperature only. Consequently the refractive index decreases across the flame from the ambient refractive index \( n_a \) to a minimum at the position of maximum

---

**Figure 41**: Plots of the two refractive index contributions used for the simulation. The values defining these two plots can be found in Table 7.
temperature, see Figure 41. Here only the part of the temperature dependent refractive index distribution that takes part in the simulation is shown.

<table>
<thead>
<tr>
<th>Q3(5) transition in the (0,0) vibrational band of the A^3Π-X^3Σ^- electronic system of NH</th>
<th>λ₀ = 335.9255 10⁻⁹ nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>λₙₐₜ = 335.92373 10⁻⁹ nm</td>
<td>nₙₐₜ = -291.422999 10⁻⁹</td>
</tr>
<tr>
<td></td>
<td>κₙₐₜ = 248.390008 10⁻⁹</td>
</tr>
<tr>
<td></td>
<td>αₙₐₜ = 9.2918256</td>
</tr>
<tr>
<td>λₚₒₛ = 335.92721 10⁻⁹ nm</td>
<td>nₚₒₛ = 291.422999 10⁻⁹</td>
</tr>
<tr>
<td></td>
<td>κₚₒₛ = 248.390008 10⁻⁹</td>
</tr>
<tr>
<td></td>
<td>αₚₒₛ = 9.2918256</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Table 7: Values used in the computation of the fringe shift for the different refractive index contributions. The Q₃(5) transition of NH is chosen. The values for calculating the complex refractive index are the same as for Figure 8. The two wavelengths are selected such that a maximum refractive index difference is obtained, i.e. at the HWHM points of the Q₃(5) transition. Additionally shown are the absorption coefficients used in the calculation of the beam attenuation.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature profile</td>
</tr>
<tr>
<td>----------------------</td>
</tr>
<tr>
<td>nₐ = 1.00029</td>
</tr>
<tr>
<td>Resonant profile</td>
</tr>
</tbody>
</table>

In Table 7 the values of refractive index used in these computations for NH can be found. In Figure 41 the plot of the refractive index distribution due to resonance is superimposed. This refractive index distribution n(ν) is calculated as described in Appendix A by considering the probe frequency for the given species. Furthermore, the concentration and the CARS temperature at the maximum position of the concentration profile is used to calculate the resonant refractive index. This provides the maximum resonant refractive index that is subsequently multiplied by a Gauss curve normalized to 1(one) in order to obtain the spatial distribution of the refractive index. In principle,
\( n(v) \) should be calculated as a function of the species concentration distribution and the temperature distribution. Such a complex calculation is not necessary for an estimate of the ray bending effects. Therefore, we obtain for the total refractive index a functional dependence of

\[
\begin{align*}
n = n_0 + \Delta n_T \exp \left( - \frac{y - y_{Te}}{\Delta T} \right) + n_0 + \Delta n_0 \exp \left( - \frac{y - y_e}{\Delta n} \right)
\end{align*}
\]

The same procedure is used for the imaginary part of the refractive index, where the absorption coefficient \( \alpha(v) \) is calculated for the probe frequency. These values used can also be found in Table 7.

We recollect now that the propagation of a ray through a \textit{stratified medium} is given by equations (7) and (8)

\[
\frac{y'' dy}{1 + y'^2} = \frac{dn}{n(y)} \quad (7)
\]

\[
1 + y'^2 = \left( \frac{n}{n_e} \right)^2 \quad (8)
\]

with the boundary conditions of the ray trajectory given as: \( y(z, y_e) \) at \( z = 0 \): \( y(0, y_e) = y_e, y'(0, y_e) = 0, y''(0, y_e) = 0 \) and \( n(y_e) = n_e \). The subscript \( e \) denotes the values at the entry position. Equation (7) can be written in a more analytical way as:

\[
\frac{\partial y^2(z)}{\partial z^2} - \frac{1}{n(y(z))} \frac{\partial n(y(z))}{\partial z} \left( 1 + \frac{\partial y(z)}{\partial z} \right) = 0 \quad (26)
\]

The optical length of the ray which joins points \( P_1 \) and \( P_2 \) is given by equation (4)

\[
[P_1P_2] = \int_{P_1}^{P_2} nds = \Phi(P_1) - \Phi(P_2)
\]

Given equation (8) and the fact that \( ds = \sqrt{1 + \left( \frac{dy}{dz} \right)^2} \) \( dz \), the integral can be substituted with a differential equation that follows the development of the optical path length of the ray

\[
\frac{\partial \Phi(y(z))}{\partial z} - n_e \left( 1 + \frac{\partial y(z)}{\partial z} \right) = 0 \quad (27)
\]
What remains to be calculated is the attenuation of the beam due to the absorption taking place along the ray. Lambert Beer's law gives \( I(z) = I_0 \int \exp[-\alpha(z)z] ds \), thus the normalized attenuated intensity \( 1 - \text{absorbed light intensity} \) is given by \( \text{ABS}(z) = I(z) / I_0 \). In the computation this attenuation is described in a similar way as for the optical pathlength

\[
\frac{\partial \text{ABS}(y(z))}{\partial z} + \alpha(z) \sqrt{1 + \left( \frac{\partial y(z)}{\partial z} \right)^2} = 0
\]  

(28)

Here \( \text{ABS}(y(z)) \) is the normalized attenuated intensity and \( \alpha(z) \) the absorption coefficient as a function of \( y(z) \). For the two last differential equations, additional initial conditions are required: \( \Phi(0)=0, \Phi'(0)=n_e \), \( \text{ABS}(0)=1 \), \( \text{ABS}'(0)= \alpha_e \).

The three equation (26), (27) and (28) are solved numerically as a system of three coupled differential equations. The program provides all three values \( y(z), \Phi(z) \) and \( \text{ABS}(z) \) as a function of \( z \), the propagation axis for \( z = [0..L] \). The numerical method used is a seventh-eighth order continuous Runge-Kutta method, a Numerical Solution of Ordinary Differential as applied in the mathematical program MAPLE (dverk78) [Ref. 61].

In order to verify the correct computation of the applied method, the numerical results are compared with an analytical solution for a refractive index distribution that in the first case is linear, \( n(y) = n_0 + (n_\delta - n_0)y/\delta \), and in the second exponential, \( n(y) = n_0 + (n_\delta - n_0)\left[1 - \exp\left(-y/\delta\right)\right] \) [Ref. 16, p. 9-13]. For both cases the numerical solution agrees down to the 13\(^{th}\) decimal with the analytical. Note that the computation is performed by using a 20 digits precision.

Thus, the verified model is applied to gain information on the influence of the refractive index gradients on the total optical path length difference of the probe beams. Therefore, the total fringe shift is calculated as the difference between the optical path of two individual rays exhibiting one of the exposure frequencies. For the computations, frequencies resulting in maximum resonant refractive index are chosen,
i.e. the two HWHM points of the transition line. Notice in Figure 41 that only the refractive index due to one exposure frequency is shown. The frequency of the second HWHM point of the transition yields a negative refractive index distribution. For the calculation of the total fringe shift as a function of y the difference of the two distributions is taken into account.

![Graph showing fringe shift distributions](image)

**Figure 42:** Comparison of the fringe shift distribution in the straight line approximation with that obtained by a model simulation that includes resonant refractive index variations along the path of the two probe wavefronts. In addition, the residuals are depicted on the figure.

The model is now compared with the simple situation of a beam passing in a straight line through a constant refractive index field. In particular, the two fringe shift distributions across the flame are compared. In Figure 42 the distributions are shown. In addition, the residuals are depicted on the Figure. Only the gradients due to the resonant refractive index are taken into consideration, i.e. the bulk refractive index containing the temperature dependence is omitted. We observe that at the center of the refractive index distribution the same fringe shifts values are obtained, since the ray in this
position follows a straight line due to the two opposing gradients. It is interesting to observe the fact that the gradients narrow the fringe shift distribution. However, since the evaluation of the interferograms is performed by determining the maximum fringe deviation only, see the experimental analysis, the influence of these gradients can be neglected.

A further refinement of the model is attempted by including the temperature, first term in Equation (25), and shown in Figure 43. The resulting fringe shift distribution is plotted in Figure 43 in addition to the results from the simpler simulations in Figure 42. Additionally, residuals due to the temperature effect are included, multiplied by a factor 100.

![Fringe Shift for the three models: straight line approximation, refraction of the probe beams without the influence of the temperature and refraction with the influence of the temperature. The residuals due to the temperature are also plotted on a magnified scale (x100).](image)

**Figure 43:** Fringe Shift for the three models: straight line approximation, refraction of the probe beams without the influence of the temperature and refraction with the influence of the temperature. The residuals due to the temperature are also plotted on a magnified scale (x100).

Again, we conclude from the modeling that the fringe shift variations introduced by refraction of the probe beams are negligible. In fact, the simulations yield a maximum
fringe shift due to refraction on the order of a few percent of the fringe shift due to the RHI process.

The fringe shifts for this analysis have been calculated exactly at the exit of the flame, i.e. at the position \( z = L \). In principle, the propagation through the whole optical system should be calculated. By applying holographic interferometry however, the influence of the optical system is insignificant for the following reasons. Distortions due to the different optical elements are subtracted in the same way as the bulk refractive index, since the two waves exhibit essentially the same frequency and are, therefore, distorted in the same way.

We examine now the projection of the two distorted wavefronts after exiting the flame region. We recall that the projection lens, introduced between the flame and the hologram plane, is optimized in space in order to minimize the effects of refraction on the propagation of the individual rays by placing the object plane of the lens at the 2/3’s of the flame length \( L \). The effect of the correct imaging with the lens is detailed in the experimental chapter. The lens is in essence an absolute instrument, since it images stigmatically a three-dimensional domain [Ref. 1, p. 143-1147]. For such an absolute instrument, Maxwell stated that “the optical length of any curve in the object space is equal to the optical length of its image”. Even though this theorem was derived for a homogeneous object and image space, more rigorous proofs were given later. Thus “a perfect imaging between two homogeneous spaces of equal refractive indices is always trivial in the sense that it produces an image which is congruent with the object”. It is therefore clear that it is sufficient to calculate the individual distorted wavefronts at the exit point of the flame and merely project them on the hologram plane.

Similarly, the influence of refraction on the absorption of the propagating wavefronts is investigated. As mentioned before the ABS(\( z \)) is the normalized absorbance as a function of the distance in the flame and is derived directly from the system of three coupled differential equations. In Figure 44 the attenuation of a beam is calculated by
using the values indicated in Table 7. In the Figure the attenuation of the straight line approximation is compared with that of the varying refractive index distribution including bulk temperature and the resonance effect. The residuals are also plotted.

**Figure 44:** Normalized absorbed intensity with and without refraction effects (due to temperature and resonance). The computation data can be found in Table 7. The difference illustrates the effect of refraction on the species concentration obtained from absorption measurements.

Following conclusions are drawn from the calculations. Utilization of 2D absorption measurements have the inherent problem that the spatial distribution will be narrowed in the same way as the fringe shift distribution shown on Figure 42. To derive the real spatial distribution of the target species a correction would be required. On the other hand, the maximum absorbed intensity, yielding the position of maximum species concentration, is correctly predicted, just as for the prediction of the Fringe Shift. Since this work is only focused on the position of maximum species concentration, the 2D absorption measurements can still provide the required quantitative information.
Conclusively, we summarize that the effects of refraction can be neglected on the correct evaluation of the interferograms resulting from the RHI measurements as well as the 2D absorption measurements.

*Heat release to the surroundings due to the high absorption.*

The experiments have shown that the absorption for the frequencies used in the RHI experiments were quite high, on the order of 25-30%. The absorbed energy deposited in the molecules will dissipate in the form of heat to the surroundings by collisional redistribution and quenching. This effect can occur on time scales comparable to the time separation between the first and second exposure pulse. It is therefore speculated that the heat dissipation from the first exposure pulse is affecting the refractive index probed by the second pulse. Since the heat released will be confined to the proximity of the resonant species, the second pulse will experience the refractive index due to temperature, the resonant refractive index specified by its frequency and finally an additional thermal refractive index due to the heat dissipation. Note that this additional refractive index will not be interferometrically subtracted since it is only experienced by the second exposure pulse alone.

In order to investigate the influence of this effect, the following procedure has been used. Off resonant, two laser, two color interferograms are recorded while a third laser pumps a transition of a target species in the time span between the two pulses. If the pumping of the transition induces a heat release of sufficient magnitude to distort the refractive index that is probed by the second probe wavefront, a deviation of the finite fringes would appear similar to the resonant recordings. Two broadband lasers are used. The Continuum laser, described previously and the Quantel laser, that has a similar features as the Continuum. Both lasers are operated in the OH mode. Since the two probe pulses are not probing the resonant refractive index the necessity of high spectral resolution is relaxed. The set up as shown in Figure 33 is used without major changes.
The pump pulse is obtained by the Excimer pumped Lambda laser, running in the NH mode. The output is formed into a light sheet of a 2mm thickness that propagates parallel to the burner surface and perpendicular to the flame, i.e., parallel to the y-axis. The Excimer laser is synchronized with the two probe lasers by applying three delay generators (Stanford Instruments DG 535). It is triggered between the first and the second exposure pulse. The time separation between the two exposure pulses is set to 50ns in order to permit various time delays for the NH pumping pulse. Since we are interested on the heat dissipation induced by absorption, the laser is tuned to the center frequency of an NH transition. As for the NH experiments, the Q_3(5) transition is used at \( \lambda_0 = 335.9255 \text{nm} \). Several exposures are performed while varying the time delay of the excitation pulse. Unfortunately, no apparent fringe deviation is detected. In order to enhance the heat dissipation effect, the laser is tuned to the band head of the 0-0 band, \( \lambda_0 = 336.090 \text{ nm} \), where the absorption is nearly 100% for the flame conditions. The same experiments are performed, but once again, no fringe shifts are detected.

In conclusion, the heat release taking place within the first 50ns yields an insignificant contribution to the refractive index. Note that for this investigation the NH species have been excited perpendicular to the probe beam as opposed to the regular RHI configuration. However heat dissipation induces a non resonant contribution to the refractive index independent of the measuring technique used to probe it.

In summary we conclude, that the discrepancy of the quantitative results obtained by RHI and 2D absorption cannot be rationalized by our investigations. In fact, simulations show clearly that the fringe shifts measurements are not significantly sensitive to the refractive index gradients appearing in the flame. Furthermore, the heat release induced in the flame due to substantial absorption does not produce a sufficient increase in fringe shift. More theoretical and experimental work is required to shed more light on the involved processes.
Conclusions and Outlook

An experimental investigation of Resonant Holographic Interferometry has been performed. It has been shown that RHI is a promising technique that can measure important trace species in flame environments. Taking advantage of the dispersive form of the real part of the refractive index near molecular transitions, a molecule can be unambiguously addressed. In particular, it is concluded that the RHI technique is able to yield quantitative measurements of species concentrations. The interferometric process ensures that disturbances such as scattering on particles, high background luminosity or low quality optics are successfully canceled. Image plane holography is applied by using a 1:1 imaging step at the 2/3’s of the flame-probe wave interaction path. In this way, all influences of the pressure and temperature gradients on the result are avoided. Additionally, the hologram read-out can be performed with white light sources.

The experimental set up at the Paul Scherrer Institute, is based on a single narrow band laser. The second color necessary for the RHI experiments is generated by optical phase-conjugation (PC) from Stimulated Brillouin Scattering (SBS) in a cell. Implementation of SBS provides a second, phase-conjugate and frequency shifted laser beam. The frequency shift of 8.5-9 GHz matches favorably the linewidth of many molecular transitions at typical flame temperatures. Therefore, the two-color PC RHI technique can be performed by addressing near resonant frequencies yielding a maximum refractive index difference.

The SBS process is experimentally and theoretically investigated. The Brillouin medium is filtered through a 0.2 μm pore filter in order to avoid optical breakdown by interception of the focused input pulse on particles in the cell. This proves to be a cheap and reliable method to filter the liquid Brillouin medium in contrast to the usually applied purification schemes by vacuum distillation. The characteristics of the SBS driving pulse are investigated. It is shown that by decreasing the laser bandwidth the
reflectivity increases and the threshold energy decreases. Good phase-conjugate fidelity is also demonstrated. The experimental parameters confirm theoretical and experimental assumptions indicating that good phase-conjugate fidelity is obtained for a SBS gain length of at least three times the depth of field of the input beam. Application of SBS on the applied Nd:YAG pumped laser pulses is not feasible. This is rationalized by the fast rising edge of the input pulse. When the slope of the leading edge of the pulse is short in comparison to the acoustic lifetime, amplification of noise modes compete with the SBS gain process and random phase jumps appear in the phase-conjugated beam. Therefore, even though high reflectivities have been achieved, the reflected beam is not applicable to RHI measurements. In order to successfully introduce PC RHI, a near diffraction limited, narrowband laser exhibiting a smooth and relatively long temporal intensity distribution must be chosen.

Experimental results show that the PC RHI has several advantages over a two laser system configuration. First, the two probe frequencies are fixed in respect to each other by the SBS process. The error in probing the correct frequencies due to frequency jitter is therefore decreased. Furthermore, the phase-conjugate nature of the pulse ensures that small phase disturbances are canceled during the interferometric read out. Additionally, alignment constraints and costs are dramatically reduced, since the method requires one laser system only. Such advantages are of importance for the application of the technique in a combustion diagnostic laboratory.

A burner with a 2D flame structure is chosen and a straightforward interpretation of the recorded interferograms is achieved. By applying RHI and 2D absorption measurements the basic features of the applied model of the complex refractive index are verified. Numerical simulations of the NH$_3$-O$_2$ chemistry applied on an opposed jet burner indicate species concentration of $\sim 10^{21}$ and $\sim 10^{23}$ for NH and OH, respectively. The absolute concentration values cannot be directly compared, due to the different flow conditions and uncertainties on the applied kinetic model. The maximum species concentrations for NH and OH measured by the two applied techniques are slightly
lower. On the other hand, the expected species concentration difference between NH and OH is successfully verified experimentally.

A numerical model of the complex refractive index is developed by applying a numerical computation of the Voigt profiles. This model is extended to calculate the fringe shift for a two-color RHI experiment and the intensity attenuation. Two combustion related parameters are the unknowns in the model. The local temperature and the species concentration. CARS measurements are performed in parallel and provide absolute temperatures in the flame. By considering these temperatures, RHI and absorption can be directly compared. The absolute species concentration measurements obtained by RHI and absorption disagree. This discrepancy appears to be species concentration dependent. For both target species, the measurements are performed on transitions with an absorption coefficient of \( \sim 14-15 \text{m}^{-1} \). This value is measured for both species at the center frequency by absorption. The NH measurements are performed on a transition line with a species concentration of 150-350 ppm, resulting in a shift of 10-12% of a fringe. On the other hand, the OH measurements are performed on a transition with a species concentration of 4000-12000 ppm, resulting in a shift of 20-30% of a fringe. Therefore, for a given absorption coefficient, a higher species concentration will result in an increased fringe shift.

Three possible problems are investigated. The refraction of the waves by the gradients of the refractive index, the finite fringe scheme applied and heat dissipation effects. A model is developed describing the parabolic trajectory of a light ray through the 2D refractive index medium represented by the \( \text{NH}_3-\text{O}_2 \) flame. Numerical calculations are performed by using values derived from the experiments. It is concluded that ray bending has no effect on the absolute species concentration measurements and can therefore be neglected. A vertical angle in respect to the input plane of the set up is introduced between the original and the phase-conjugate beam. Geometrical considerations show that a measurable contribution to the fringe shift would occur only at unreasonably large angles. Finally, experiments are performed by recording off
resonant RHI holograms while pumping a NH transition in the time span between the two exposure pulses. The absorbed energy will be subsequently released to the surrounding medium. If this heat release contributed to the refractive index an additional fringe shift would be detected. No effect could be observed. Conclusively, the investigations are not providing a reliable answer to the discrepancy detected between the two applied techniques.

**Figure 45:** Experimental set up for performing simultaneously 2D absorption and two reference arm PC RHI. Each reference beam has a different frequency. The two cameras record the wave impinging on the dye cells.

In Figure 45, a scheme for combining phase stepping techniques with simultaneous absorption measurements is shown. For this configuration of PC RHI two references are
applied, as developed by Metrolaser [Ref. 62]. In the set up a correcting holographic optical element is recorded (CHOE) prior to the resonant double exposure hologram. Between the two exposures a small tilt angle is introduced. The CHOE provides two exact replicas of the original reference beams used for recording the RHI data hologram. The CHOE is sandwiched with the resonant data hologram. During reconstruction, the perfect replicas of the reference beams emerge that subsequently reconstruct the data hologram, even with a different wavelength. A perfect cancellation of aberrations is achieved. By manipulating the relative position of data hologram and CHOE, live fringes appear that can be converted to phase information. The two beam splitters placed before and after the flame will record the probe wavefront. Normalization will yield the absorbed 2D profile in a similar way as in the experiments performed for this work. Care has to be taken to select only one of the probe waves to perform the absorption measurements. This can be achieved by using different polarizations for the two probe waves and a polarizer in front of the two cameras.

In the future, a more thorough investigation on the RHI technique must be conducted. Choosing a \( \text{H}_2-\text{O}_2-\text{air} \) flame will permit simulation of kinetic models matching the flow configuration of the Wolfhard-Parker burner. Additionally PLIF can be favorably applied on the Wolfhard-Parker burner and a direct comparison of the three measuring techniques can be performed.

It is worth noting that the spatial resolution of the RHI technique is not limited by the spatial resolution of the camera, as opposed to PLIF. The minimum resolution of the hologram defines the resolution of the measurement. Thus, for the emulsions used in this work, a spatial resolution of less than 1 \( \mu \text{m} \) can be achieved. Real time resonant holographic interferometry can be readily applied using bacteriorhodopsin thin films [Ref. 63]. The measuring repetition rate can be increased theoretically up to full video frame rate of 30Hz. Sensitizing bacteriorhodopsin for UV radiation can be achieved also, though till date not realized [Ref. 64].
The measured species concentration for OH and NH indicate that the upper level of this technique has not been reached. For OH concentration of up to $\sim15000$ ppm have been measured. OH transitions with lower oscillator strengths can be found, thus increasing the measuring range. The minimum detectable species concentration depends on the fringe interpretation technique applied. In the present experimental set up the lowest detectable concentration would be on the order of 100 ppm yielding a fringe shift of $\sim3\%$. However, the error would increase dramatically. In order to increase the sensitivity, the absolute phase rather than the fringe shift must be measured. Utilizing fast phase measurement techniques with resolution of up to $1/100^{th}$ or $1/1000^{th}$ of a fringe [Ref. 41] the sensitivity for the investigated molecules can be of the order of 30 ppm or 3 ppm, respectively. Such sensitivities are not directly comparable to PLIF measurements exhibiting detectable concentrations in the sub-ppm range. On the other hand, the high spatial resolution in combination with fast phase measurements techniques could provide a good alternative to PLIF. In particular, the verification of numerical simulations could become the niche market of RHI.
Appendix A

Derivation of the complex refractive index

In the following we follow directly the derivation of the complex refractive index as shown by Anne P. Thorne in Reference 65.

A system of an assembly of bound electrons driven by an electric field, can be considered as a damped, forced oscillation. For simplicity, we consider oscillations in the x-direction only, produced by a plane-polarized electromagnetic wave traveling in the z-direction. The solution of Maxwell’s equations for the electric vector $E$ in a medium of permittivity $\varepsilon$ is

$$E(z) = E_0(0)\exp[i\omega(t - \frac{z}{v})],$$

where the phase velocity $v$ is given by $v = \frac{1}{\sqrt{\varepsilon\mu_0}} = \frac{c}{\sqrt{\frac{\varepsilon}{\varepsilon_0}}} = \frac{c}{n}.$

$\varepsilon_0$ and $\mu_0$ are the permittivity and permeability of vacuum and $n'$ is the complex refractive index, which can be written as

$$n' = \sqrt{\frac{\varepsilon}{\varepsilon_0}} = n - ik$$

leading to $\frac{z}{v} = \frac{n'}{c}z = \frac{n}{c}z - i\frac{k}{c}z$. Therefore

$$E(z) = E_0 \exp[i\omega(t - \frac{n'z}{c} + i\frac{kz}{c})] = E_0 \exp[-\omega\frac{kz}{c}] \exp[i\omega(t - \frac{n'z}{c})].$$

Thus $k$ and $n$ determine the absorption and the phase velocity respectively, $k$ can be related to the more familiar absorption coefficient $\alpha$, by finding the power attenuation due to the layer $z$:

$$I(z) \propto |E(z)|^2 = E_0^2(0) \exp[-2\left(\frac{\omega k}{c}\right)z],$$

with $2\frac{\omega k}{c} = \alpha$, $\alpha$ being the absorption coefficient. Both $n$ and $k$ can be determined in terms of the properties of an oscillator, by solving the equation of motion for the bound electron driven by the force $-eE(t)$ at some fixed value of $z$. The equation is

$$\ddot{x} + \gamma \dot{x} + \omega_0 x = \frac{c}{m_e} E_0 \exp[i\omega t],$$

where $\omega_0$ is the resonant frequency of the oscillator, $m_e$
the mass of the electron and $\gamma$ covers all forms of damping. The equation has the known solution

$$x = x_0 \exp[i\omega t] \text{ with } x_0 = \frac{-e/m_e E_0}{(\omega_0^2 - \omega^2) + i\omega \gamma}.$$ 

The connection to permittivity is made via susceptibility. The instantaneous value of the dipole formed by an electron displaced a distance $x$ is $-e x = -e x_0 \exp[i\omega t]$. If there are $N$ oscillators per unit volume the susceptibility $\chi$, defined as polarization per unit field, is

$$\chi = -\frac{Ne_0 \exp[i\omega t]}{E} = \frac{N e^2/m_e}{(\omega_0^2 - \omega^2) + i\omega \gamma}.$$ 

The standard relation between permittivity and susceptibility, $\varepsilon = \varepsilon_0 + \chi$, leads to

$$\frac{\varepsilon}{\varepsilon_0} = 1 + \frac{Ne^2}{2\varepsilon_0 m_e (\omega_0^2 - \omega^2) + i\omega \gamma} = (n')^2 = (n - i\kappa)^2$$

so

$$\left(n^2 + \kappa^2\right) - i2n\kappa = \left(1 + \frac{Ne^2}{\varepsilon_0 m_e (\omega_0^2 - \omega^2)^2 + \omega^2 \gamma^2}\right) - i\left(\frac{Ne^2}{\varepsilon_0 m_e (\omega_0^2 - \omega^2)^2 + \omega^2 \gamma^2}\frac{\omega \gamma}{\omega_0^2 - \omega^2}\right)$$

By equating the imaginary parts, for $n = 1$ we get:

$$\kappa = \frac{Ne^2}{2\varepsilon_0 m_e (\omega_0^2 - \omega^2)^2 + \omega^2 \gamma^2} \frac{\omega \gamma}{\omega_0^2 - \omega^2}$$

For the real parts, we can put $n = 1 + \delta$ and neglect the second-order term $\delta^2$, giving

$$1 + 2\delta + \kappa^2 = 1 + \frac{Ne^2}{2\varepsilon_0 m_e (\omega_0^2 - \omega^2)^2 + \omega^2 \gamma^2} \frac{\omega_0^2 - \omega^2}{\omega_0^2 - \omega^2}$$

The second term on the right-hand side is equal to $2\delta$ if $\kappa^2$ is also a second-order term.

That this is indeed the case, except in the middle of an absorption line, can be seen by comparing this provisional value of $\delta$ with $\kappa$ in the previous equation. Provided that $(\omega_0^2 - \omega^2) > \omega$, $\delta > \kappa$. Therefore
\[ \delta = n - 1 = \frac{N\varepsilon^2}{2\varepsilon_0 m_e} \frac{\omega_0^2 - \omega^2}{\left(\omega_0^2 - \omega^2\right)^2 + \omega^2 \gamma^2} \]

This relation may not be valid in the middle of a strong absorption line - where a measurement of refractive index is somewhat impractical anyway.

With some more approximations, \( \omega_0^2 - \omega^2 = 2\omega_0 (\omega_0 - \omega) \) and \( \omega \gamma = \omega_0 \gamma \), we get

\[ \kappa = \frac{N\varepsilon^2}{4\varepsilon_0 m_e \omega_0} \frac{\gamma / 2}{(\omega_0 - \omega)^2 + (\gamma / 2)^2}, \quad \text{and} \quad a = \frac{N\varepsilon^2}{2\varepsilon_0 m_e c} \frac{\gamma / 2}{(\omega_0 - \omega)^2 + (\gamma / 2)^2} \]

using the previous relationship between absorption and extinction coefficient, and

\[ n - 1 = \frac{N\varepsilon^2}{4\varepsilon_0 m_e \omega_0} \frac{\omega_0 - \omega}{(\omega_0 - \omega)^2 + (\gamma / 2)^2} \]

We can choose now the frequency, \( \nu \), as a variable instead of the angular frequency. We therefore have:

\[ \kappa(\nu) = \frac{N\varepsilon^2}{4(2\pi)^2 \varepsilon_0 m_e \nu_0} \frac{\Delta v_L / 2}{(\nu - \nu_0)^2 + (\Delta v_L / 2)^2} \]

\[ n(\nu) - 1 = \frac{N\varepsilon^2}{4(2\pi)^2 \varepsilon_0 m_e \nu_0} \frac{\nu_0 - \nu}{(\nu - \nu_0)^2 + (\Delta v_L / 2)^2} \]

If there is thermal motion of the molecules of interest, the effective line center frequency is \( \omega_0' = \omega_0 \left(1 + \frac{u}{c}\right)\) or \( \nu_0' = \nu_0 \left(1 + \frac{u}{c}\right)\) for a molecule having a component of velocity \( u \) along the propagation axis of the light suffering dispersion. At thermal equilibrium, the molecules of a gas follow a Maxwellian velocity distribution. Thus at temperature \( T \), the number of molecules per unit volume having a velocity component \( (u, du) \) is

\[ N(u) du = \frac{N}{u_p \sqrt{\pi}} \exp \left[-\left(\frac{u}{u_p}\right)^2\right] du, \]
where \( N = \int N(u)du \) is the density of all molecules in the level, \( u_p = \sqrt{2k_B T/M} \) is the most probable velocity, \( M \) is the mass of the molecule and \( k_B \) is the Boltzmann's constant. The number of molecules with absorption frequencies shifted from \( v_0 \) into the interval \([v,v+dv]\) is therefore

\[
N(v)dv = \frac{Nc/v_0}{u_p\sqrt{\pi}} \exp \left[ -\left( \frac{c(v-v_0)}{u_p v_0} \right)^2 \right] dv.
\]

The absorbed intensity is proportional to the density \( N(v)dv \). Therefore the intensity profile of a Doppler broadened spectral line will have a gauss form [Ref. 66]

\[
I(v) = I_0 \exp \left[ -\left( \frac{c(v-v_0)}{u_p v_0} \right)^2 \right].
\]

The FWHM of this line can be calculated to \( \Delta v_D = \frac{2v_0}{c} \sqrt{\frac{2k_B T \ln 2}{M}} \) [Ref. 24, p. 87].

We see that we have to convolute the two profiles, the Lorentzian and the Gaussian over all the possible velocities of the molecules. Therefore:

\[
\kappa(v) = \frac{Nfe^2}{4(2\pi)^2 \epsilon_0 m_e \sqrt{2k_B \pi T}} \left[ \frac{M}{v_0} \right] \int_{-\infty}^{+\infty} \frac{\Delta v_L/2}{(v_0^* - v)^2 + \left( \Delta v_L/2 \right)^2} \exp \left[ -\frac{M}{2k_B T u^2} \right] du
\]

and

\[
n(v) - 1 = \frac{Nfe^2}{4(2\pi)^2 \epsilon_0 m_e \sqrt{2k_B \pi T}} \left[ \frac{M}{v_0} \right] \int_{-\infty}^{+\infty} \frac{v_0^* - v}{(v_0^* - v)^2 + \left( \Delta v_L/2 \right)^2} \exp \left[ -\frac{M}{2k_B T u^2} \right] du
\]

We use following substitutions to simplify the integrals:

\[
\alpha = \frac{\Delta v_L}{\Delta v_D} \sqrt{\ln 2} \text{ is the Voigt parameter, } \beta = 2 \frac{v-v_0}{\sqrt{\ln 2}} \text{ and } y = 2 \frac{v_0^* - v_0}{\Delta v_D} \sqrt{\ln 2}.
\]

\[
\frac{Nfe^2}{4(2\pi)^2 \epsilon_0 m_e \sqrt{2k_B \pi T}} \left[ \frac{M}{v_0} \right] = \frac{Nfe^2}{4(2\pi)^2 \epsilon_0 m_e \sqrt{\pi c \Delta v_D}} \frac{1}{\Delta v_D}.
\]

\[
\frac{(v_0 - v)}{(v_0^* - v)^2 + \left( \Delta v_L/2 \right)^2} = \frac{2\sqrt{\ln 2}}{\Delta v_D} \frac{(y-\beta)}{(y-\beta)^2 + \alpha^2}.
\]
\[ \left( \frac{\Delta v_L}{2} \right)^2 \left( v_0^* - v \right)^2 + \left( \frac{\Delta v_L}{2} \right)^2 = \frac{2 \sqrt{\ln 2}}{\Delta v_D} \frac{\alpha}{(y - \beta)^2 + \alpha^2}. \]

\[ v_0^* = v_0 \left( 1 + \frac{u}{c} \right) \Rightarrow u = \frac{v_0^* - v_0}{v_0} c. \]

We see that: \( y = \sqrt{\frac{M}{2k_B T}} u \) and \( du = \sqrt{\frac{2k_B T}{M}} dy \)

After some rearrangements we get that:

\[ n(v) = \frac{\sqrt{\pi}}{8\pi^2} \frac{N \epsilon_0^2 \alpha}{\varepsilon_0 m_e \Delta v_L v_0} \left[ \frac{1}{\pi} \int_{-\infty}^{\infty} e^{-y^2} \frac{y - \beta}{\alpha^2 + (y - \beta)^2} dy \right] \]

\[ \kappa(v) = \frac{\sqrt{\pi}}{8\pi^2} \frac{N \epsilon_0^2 \alpha}{\varepsilon_0 m_e \Delta v_L v_0} \left[ \frac{\alpha}{\pi} \int_{-\infty}^{\infty} e^{-y^2} \frac{1}{\alpha^2 + (y - \beta)^2} dy \right] \]

or

\[ n(v) = \frac{\sqrt{\pi}}{8\pi^2} \frac{N \epsilon_0^2 \alpha}{\varepsilon_0 m_e \Delta v_L v_0} \text{Im}\{W(z)\} dy \]

\[ \kappa(v) = \frac{\sqrt{\pi}}{8\pi^2} \frac{N \epsilon_0^2 \alpha}{\varepsilon_0 m_e \Delta v_L v_0} \text{Re}\{W(z)\} dy, \]

where \( W(z) = \exp(-z^2) \text{erfc}(-iz) \) is the Voigt function with \( z = \beta + i\alpha \) and

\[ \alpha = \frac{\Delta v_L}{\Delta v_D} \sqrt{\ln 2}, \quad \beta = 2 \frac{v - v_0}{\Delta v_D} \sqrt{\ln 2}. \]

The formulation derived corresponds to the complex refractive index presented by Blenstrup, Bershader and Langhoff in Reference 67.


Computation of the complex refractive index

For the computation of the Voigt function the rational approximation is the routine developed by Humliček is used, here presented in a pseudo FORTRAN code [Ref. 68].

\[
T = \begin{bmatrix}
0.314240376, & 0.947788391, & 1.59768264, & 2.27950708, & 3.02063703, & 3.8897249
\end{bmatrix}
\]
\[
C = \begin{bmatrix}
1.01172805, & -0.75197147, & 1.255772710^{-2}, & 1.00220082 \times 10^{-2}, & -2.42068135 \times 10^{-4}, & 5.00848061 \times 10^{-7}
\end{bmatrix}
\]
\[
S = \begin{bmatrix}
1.393237, & 0.231152406, & -0.155351466, & 6.21836624 \times 10^{-3}, & 9.19082986 \times 10^{-5}, & -6.27525958 \times 10^{-7}
\end{bmatrix}
\]

\[
WR = 0
\]
\[
WI = 0
\]
\[
y_1 = \alpha + 1.5
\]
\[
y_2 = y_1^2
\]

\[
\text{for } i \text{ from 1 to 6 do}
\]
\[
R = \text{beta} - T[i]
\]
\[
Dx = 1/(R^2 + y_2)
\]
\[
D_1 = y_1 \times Dx
\]
\[
D_2 = R \times Dx
\]
\[
R = \text{beta} + T[i]
\]
\[
Dx = 1/(R^2 + y_2)
\]
\[
D_3 = y_1 \times Dx
\]
\[
D_4 = R \times Dx
\]
\[
WR = WR + C[i] \times (D_1 + D_3) - S[i] \times (D_2 - D_4)
\]
\[
WI = WI + C[i] \times (D_2 + D_4) + S[i] \times (D_1 - D_3)
\]

\[
\text{end}
\]
\[
\beta = 2 \times \text{sqrt}(\ln(2)) \times (\text{niur} - \text{niu}) / D\text{n}\text{iud}
\]
\[
\alpha = \text{sqrt}(\ln(2)) \times D\text{n}\text{iul} / D\text{n}\text{iud}
\]
\[
Z = \beta + \alpha \times \text{I}.
\]

\( WR \) is the real part and \( WI \) the imaginary part of the refractive index, and \( \alpha \) and \( \beta \) are \( \alpha \) and \( \beta \) respectively. These two functions must be then multiplied with a constant

\[
K = \frac{\sqrt{\pi}}{8\pi^2} \frac{Nfe^2 \alpha}{\varepsilon_0 m \Delta \nu L \nu_0}.
\]
Following parameters are required in order to compute the complex refractive index. Spectroscopic constants for the molecule under investigation in order to calculate, the oscillator strength and the partition function for the studied transition of the target species. The target species concentration and the temperature in order to calculate the Doppler broadening, $\Delta v_D$, and the partition function. Finally the colissional broadening $\Delta v_L$. This can be theoretically calculated but not as easy as the Doppler broadening $\Delta v_D$. For a detailed analysis of the colissional broadening, the reader is referenced to A. Thorne [Ref. 65]. In this work another approach is taken. The combined Voigt width is measured by absorption scans over the line and the Doppler width is calculated for a Temperature predicted from the CARS measurements. An iterative loop in the program calculates the colissional broadening $\Delta v_L$.

List of spectroscopic parameters necessary to compute the Voigt function.

- Probed Transition => rotational quantum number and degeneracy factors $J_2$ and $J_1$, $g_2$ and $g_1$
- Einstein coefficient for spontaneous emission $A_{21}$ (s$^{-1}$)
- Vibrational quantum number $\nu$
- Molecular vibration frequency $\omega_e$ (cm$^{-1}$)
- Molecular rotation constant $B$ (cm$^{-1}$)
- Nuclear spin degeneracy $g_l$
- Electronic partition $Q_e$

In order to control the rational approximation suggested by Humlíček following procedure is used. The Voigt profile is calculated for an $\alpha$ parameter equal to zero ($\alpha = 0$), implying a zero Lorentzian width, $\Delta v_L = 0$. The parameters for the computation are: $Q_3(5)$ transition in the (0,0) vibronic band of the $A^3\Pi-X^3\Sigma^+$ electronic system of NH,
A_{21} = 2.5 \times 10^{-6} \text{ s}^{-1}, B = 16.667 \text{ cm}^{-1}, \omega_c = 3300 \text{ cm}^{-1}, f_j = 0.0423, T = 2335 \text{ K} \text{ and an assumed total NH concentration of } 10^{20} \text{ m}^{-3}. \text{ The Voigt FWHM is then } \Delta \nu_v = \Delta \nu_D = 0.266 \text{ cm}^{-1} (7.973 \text{ GHz}) \text{ for the temperature given above. The curve is then compared by an analytical gauss-formed curve:}

\kappa_G(v) = K \exp \left[ - \left( \sqrt{\ln 2 \frac{v-v_0}{\Delta \nu_D}} \right)^2 \right].

![Graph showing the extinction coefficient profile as a function of frequency.]

**Figure A1:** The extinction coefficient profile as a function of frequency, \( \kappa(v) \), with a gauss form (\( a = 0 \Rightarrow \Delta \nu_L = 0 \)) as calculated by a true gauss, \( \kappa_G(v) \), form and by the rational approximation as suggested by Humlíček, \( \kappa_{Huml}(v) \). The absolute difference between the two curves is also depicted (right y-axis).

As seen in Figure A1, the maximum absolute difference between the analytical gauss curve and the curve calculated with the rational approximation as suggested by Humlíček, is in the order of ~2 \( 10^{-6} \). Taking into account the magnitude of the experimental errors the rational approximation can adequately serve the purposes of this work.
Appendix B
The Wolfhard-Parker burner

In the following sketches the important dimensions of the W.-P. burner are presented. The burner is built following the design of Dr. Kermit C. Smyth, National Institute of Standards and Technology, Maryland USA, who kindly provided the construction drawings and gave fruitful advise in operating the W.-P. burner and stabilizing the flame.

![Diagram of the W.-P. burner dimensions]

**Figure B1:** The dimensions of the W.-P. number. All dimension in mm.

The burner parameters used in the experiments performed in this work are summarized below:

<table>
<thead>
<tr>
<th></th>
<th>Slot Area (cm²)</th>
<th>Input Flow (lt/min)</th>
<th>Flow velocity (cm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH₃</td>
<td>3.1</td>
<td>2.5</td>
<td>13.44</td>
</tr>
<tr>
<td>O₂</td>
<td>13.5</td>
<td>30</td>
<td>37.04</td>
</tr>
</tbody>
</table>

The velocity ratio of oxidizer to fuel is then \( \frac{V(O₂)}{V(NH₃)} = 2.75 \).
In order to obtain a stable flame a stable flow input is necessary. Small fluctuations could be observed on the flame when electronically controlled mass flow meters were used. They were attributed to the mass flow meters, since when the flows were controlled by two tube flow meters, a more stable flame was obtained. The Sho-Rate flow meters, Brooks Instrument, calibrated for NH$_3$ and O$_2$, are specified with an accuracy of ±5% and a repeatability of ±0.5% at an input pressure of 4bar absolute. During the experiments the setting on the flow meters were fixed using only the shut off valves to turn the flows On and Off.

![Figure B2: Stabilizing the flame, providing a flat velocity profile.](image)

A: 0.5mm steel nets, B: 0.5mm quartz beads.
C: 1mm quartz beads, D: Common steel-wool.

To assist the performed numerical simulation, a top-hat velocity distribution was desired. In order to achieve that, the flow must be sufficiently dispersed over the slot length. No velocimetry techniques were applied to measure the velocity profile of the three slots. The two flame fronts were observed and a uniform flame was sought in terms of height and form along the burner length. Several flow-dispersers were tried out: Honeycomb structures, honeycomb structures combined with small quartz beads of different diameters and more. The final choice that provided a stable flame and a flat
profile is presented in Figure B2. The first third of each slot is filled with common steel-wool. The two other thirds are filled with quartz beads of 1mm and 0.5mm diameter respectively. In order to stabilize the steel-wool and the quartz beads a fine 0.5mm steel net is suspended to the walls.

Below a presentation of graphically combined 2D absorption measurements of ON and NH is presented positioned over the burner surface indicated by the walls separating the three slots. Additionally the two axes (x, y) are presented following the nomenclature described in the chapter of fundamental theory. The z-axis is parallel to the slot symmetry axis, and perpendicular to the absorption profiles.

Figure B3: Graphical presentation of 2D flame stabilized over the Wolfhard-Parker burner.

Here, the absorption profiles of NH and OH, deriving from separate measurements, are plotted over the burner for illustrative reasons.
1D numerical simulation of a NH$_3$-O$_2$ diffusion flame

As guideline for the experiments, numerical simulations of the oxidation of NH$_3$ were performed by Dr. Jeremish Ching Lee of LVV/ETHZ. The simulations were performed using a detailed kinetic model of the chemistry of NH$_3$ oxidation [Ref. 69]. As a model an opposed-jet flame was used. Two tubes of a diameter of $\Phi = 1$ cm at a distance of 1 cm to each other provide the fuel (NH$_3$) and the oxidizer (O$_2$). The flame is stabilized between the two jets. Due to symmetry considerations, a 1D model can be applied [Ref. 70]. For the two charts presented below, the velocities of fuel and oxidizer were $V_{\text{fuel}} = V_{\text{oxid}} = 15\text{cm}\text{s}^{-1}$. In the first figure the mole fraction of the fuel and the oxidizer is shown together with the distribution of the N$_2$ product and the temperature.

![Graph](image)

**Figure B4:** Spatial distribution of fuel and oxidizer in units of absolute mole fraction. Also presented is the estimated temperature and the distribution of the N$_2$ product. The horizontal axis is the distance between the two input jets, in cm.
A few comments have to be made for these numerical results. First, the applied kinetics on ammonia oxidation are validated for low concentration of ammonia at temperatures below 1500K. Since the 1D opposed-jet solution for the pure NH$_3$-O$_2$ flame is based on this kinetics system, even if a good agreement with experiment is observed, it is not a valid comparison. The 1D opposed-jet solution for the pure NH$_3$-O$_2$ flame in the low strain rate limit can be used to get a good idea on the Temperature, OH, NH and N$_2$ concentrations. Also, one can plot out the solution in the mixture fraction coordinate, this way, a direct comparison with different types of diffusion flame can be compared. In this work the simulations are rather used to obtain the order of magnitude of the interesting parameters.

For CARS measurements adequate N$_2$, at least 10%, is required in order to obtain a good signal to noise ratio. Therefore, it is to expect that CARS measurements will not be possible for the whole temperature profile.
A high temperature is also calculated, $T_{\text{max}} = 2725\text{K}$. Such high temperatures are desirable for the resonant experiments, since the temperature is the parameter that primarily determines the FWHM of the transition lines to be probed. The positions where the concentrations of the two target species maximize are well separated. Additionally OH has a broader spatial distribution than NH. For the two species, NH and OH, a concentration of the order of $10^{21}\text{m}^{-3}$ and $10^{23}\text{m}^{-3}$ respectively is calculated. The OH concentration is therefore expected to be two orders of magnitude higher than for NH.

The kinetic model was also applied for a flow configuration matching the one used for the experiments presented here. It did not work at all with a full transient code; since at ignition the chemistry time scale is so much faster than the acoustic time that the code, based on the low Mach number expansion, simply failed. Additionally the full kinetics required an unreasonable amount of computer resources.
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Curriculum Vitae

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ABSOLUTE CONCENTRATION MEASUREMENTS USING DFWM AND MODELING OF OH AND S2 IN A FUEL-RICH H2/AIR/SO2 FLAME by Radi-PP, Mischler-B, Schlegel-A, Tzannis-AP, Beaud-P, Gerber-T to be published


Conference Contributions:

PHASE-CONJUGATE RESONANT HOLOGRAPHIC INTERFEROMETRY APPLIED TO NH AND OH CONCENTRATION MEASUREMENTS IN A 2D DIFFUSION FLAME
Tzannis-AP, Gerber-T
19th Task Leaders Meeting of the IEA Implementation Agreement “Energy Conservation and Emissions Reduction in Combustion”, 14th-17th September 1997, Capri-Naples, Italy (talk by GT)

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STIMULATED EMISSION PUMPING OF OH AND NH IN FLAMES BY USING TWO-COLOR RESONANT FOUR-WAVE MIXING
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STIMULATED EMISSION PUMPING OF OH AND NH IN FLAMES BY USING TWO-COLOR RESONANT FOUR-WAVE MIXING
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