Doctoral Thesis

Laser-induced gratings in the gas phase formation mechanisms and applications for diagnostics

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Laser-Induced Gratings in the Gas Phase: Formation
Mechanisms and Applications for Diagnostics

A dissertation submitted to the
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for the degree of
Doctor of Natural Sciences

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2003
To my parents and Bernd Hemmerling
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Abstract

The subject matter of this thesis is laser-induced gratings in the gas phase. Laser-induced gratings arise from the nonlinear interaction between a medium and the radiation from an interference structure generated by two crossed laser beams. This interaction leads to a periodic modulation of the refractive index, i.e. to the formation of a dynamic optical grating by various resonant or non resonant mechanisms. Such gratings can be detected by diffracting a probe beam. The dominant grating formation processes in the gas phase are electrostriction and collisional relaxation of the absorbed laser energy. These two mechanisms which lead to the formation of electrostrictive and thermal gratings, respectively cause a density modulation in the medium. By the temporal resolved detection of a cw probe beam scattered from the induced density variation, various thermodynamic properties of the medium can be determined such as the temperature, the viscosity, the thermal conductivity and, under favorable conditions, the composition. If a laser induced grating is generated in a flow, the scattered probe beam will be Doppler shifted by an amount which is proportional to the flow velocity. From the analysis of the temporal evolution of the scattered beam the flow velocity can be determined without seeding the flow.

In this thesis the potential of laser induced transient gratings for the diagnostics in the gas phase is demonstrated. The technique is applied for the first time to our knowledge for thermometry, velocimetry, concentration measurements of binary gas mixtures of known temperature and for imaging purposes. The induced density variation is calculated by solving the linearized hydrodynamic equations. The contributions from electrostriction, instantaneous and slow relaxation of the absorbed radiation energy into heat are given explicitly.

We used time resolved light scattering from laser induced electrostrictive and thermal gratings to measure nonintrusively temperatures of premixed CO/O₂ and methane/air flames in the pressure range between 1 and 25 bar and of air at atmospheric pressure up to 1400 K. Moreover, simultaneous, instantaneous, nonintrusive and remote measurements of flow velocity and temperature in a submerged air jet at atmospheric pressure are determined from the time resolved detection of the light scattered off laser-induced electrostrictive gratings. In addition, the gas composition of CH₄/N₂ and H₂/N₂ mixtures of known temperature has been determined by using electrostrictive laser induced gratings. Finally, the potential of laser-induced gratings for imaging purposes has been demonstrated to visualize a helium flow in air and to map the particles concentration in a sooty acetylene/air diffusion flame.
Zusammenfassung


Curriculum Vitae

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to her help, the yearly check of all journals took half the time it usually does). Also my thank
to her and my sister for their moral support.
1 Introduction

1.1 Motivation and outline

With the progress of technology laser sources have become extremely versatile and affordable. They have found a variety of application fields in medicine, industry and science, and - in particular - in the field of combustion diagnostic. With the concern of hydrocarbon fuel source availability and environmental considerations, combustion processes need to become more efficient and more clean. To test and validate combustor design and to verify predictive computational results, it is necessary to measure the temperature and chemical composition of the combustion products. However, there are some limitations in probing combustion processes to the extent which is required for both empirical and theoretical advances. These limitations arise for two primary reasons. First of all, the high temperatures and heat transfer rates render these systems difficult to probe physically with the desired space and time resolutions. In addition, high demands on the probe materials are imposed to endure the harsh environment. Furthermore, combustion processes are delicately stabilized and thus easily altered by physical intrusion. Optical techniques have been recognized by combustion researchers for their potential to overcome these limitations. Laser-based techniques allow the remote non-intrusive, in situ, spatially and temporally resolved measurement of important chemical parameters [Eck 1988]. However, laser techniques are not without disadvantages: A major limitation is the requirement of an optical access to the test volume. In addition, at present they require a fairly high level of operator skill. All of the instruments required, e.g. lasers, detectors, spectrographs, etc. are commercially available. Nevertheless there has been little commercial integration of the discrete items into complete “turnkey” instruments, at least in regard to techniques for species and temperature measurements. Furthermore, for the correct determination of temperature and concentration by laser spectroscopic techniques a deep understanding of all physical processes involved is essential. Consequently, an elaborated analysis of the signal has to be performed for the quantitative determination of the parameters of interest. The measured spectra need to be carefully modelled and this usually requires computationally intensive numerical programming. For these reasons the optical diagnostics techniques continue to be developed and improved in order to render them more versatile and user friendly.

The present thesis demonstrates the potential of laser induced electrostrictive and thermal gratings as a new diagnostic tool in the gas phase. In the following a list of the specific
diagnostics applications of the technique that have been accomplished for the first time to the author’s knowledge is given. The method is applied for thermometry in air at atmospheric pressure up to 1400 K and in premixed CH\textsubscript{4}/air and CO/O\textsubscript{2} flames at atmospheric and higher pressures (up to 25 bar). In addition laser induced gratings have been used for the simultaneous, instantaneous, nonintrusive and remote measurement of flow velocities and temperatures in air flows in the ranges 10-100 m/s and 295-600 K, respectively. Laser induced electrostrictive gratings have been applied for the determination of sound velocities in gases and to measure the composition of binary isothermal gas mixtures of CH\textsubscript{4}/N\textsubscript{2} and H\textsubscript{2}/N\textsubscript{2}. Furthermore, the potential for imaging application of this technique is demonstrated. By using a 2D setup the technique has been applied to visualize a helium stream in air and to map soot in a acetylene /air diffusion flame. Also, the influence of various parameters such as the alignment and spatial intensity profile of the beams, the pressure, the laser intensity on the generated electrostrictive and thermal grating signal is shown. In particular, the coherence length of the output of the multimode, pulsed laser source is measured by using electrostrictive laser-induced gratings. Furthermore the potential of laser induced gratings for the investigation of optical breakdown in gases is qualitatively shown. These investigations provide a test for the theoretical model and allow to select the best matching experimental configuration for the measurement of the desired parameters. With the exception of the setup used for imaging, the different experimental arrangements have been realized for the first time to the author’s knowledge to obtain the best results in the application of this novel diagnostic technique. In the theory section, the linearized hydrodynamic equations have been solved to obtain an expression for the laser induced density variation caused by electrostriction and thermalization of absorbed laser energy. The full calculation is presented in the appendix. The main results are published in Refs. [Hem 1993], [Sta 1995], [Sta 1998], [Hem 1999], [Hem 1998], [HeH 1998], [Hem 2000] and [Hem 2001], and will be described in detail in the following. In the summary section, the results are recapitulated and possible future work is outlined in this growing field of coherent laser diagnostics.

1.2 Laser techniques for combustion diagnostics

No single laser technique is capable to measure all parameters simultaneously; various approaches are complementary and several may have to be combined to characterize the medium to the desired extent. For example, Laser-Induced Fluorescence (LIF) has been widely used to measure minority species concentration and temperatures [Luq 1997], [Tam 1997]. LIF is the spontaneous emission of radiation from an upper energy level of an
atom or molecule, which has been excited by absorption of laser radiation. However, a molecule or atom in an excited state may not necessarily emit radiation; several other pathways of energy loss are available to compete with fluorescence. Some of these are dissociation, ionization, energy transfer to another molecule, energy transfer to other internal energy states within the same molecule, and chemical reactions. These processes, all competing with fluorescence, are termed quenching processes. They reduce the amount of the fluorescence signal and obscure its interpretation. Because the LIF signal is strongly affected by collisional quenching its application is limited to low pressure environments. Other methods as, e.g., Raman Scattering or Coherent Anti-Stokes Raman Scattering (CARS) require high species concentration and are therefore not very sensitive [Eck 1988]. In view of these facts a lot of efforts were undertaken to refine the known techniques, or to develop new ones, which would be capable to circumvent the limitations of the established ones. Sometimes the obstacles encountered in measuring some parameters by one specific technique turn out to be the starting point for the determination of other parameters and might lead to different, new techniques. An example are Degenerate Four-Wave Mixing (DFWM) experiments where the laser radiation is absorbed by the molecules (or atoms) of the medium under investigation and a spatially modulated population difference between excited and ground state arises [Vac 1998]. Part of the excitation energy is converted to heat by collisional relaxation leading to the spatial modulation in temperature and pressure and therefore to the formation of a grating in the refractive index. Such optical gratings are called thermal gratings and may give, especially at high pressures, an important contribution to the DFWM signal [Dre 1995], rendering its interpretation more difficult. On the other hand, thermal gratings have a potential for diagnostic applications in the gas phase. The laser-induced grating techniques have been applied to measure kinetic and thermodynamic properties such as quenching rates, sound velocity, temperature, heat conduction, viscosity, gas composition, flow velocity. For more details on some of these topics please see section 3 and references given therein. Like degenerate four-wave mixing, which exhibits a similar beam geometry, laser-induced gratings are coherent techniques [Eich 1986]. In coherent light scattering processes the generated signal is a strong laser like beam and can be collected as a whole at large distances from the measuring volume. This signal is proportional to the number of molecules squared.
1.3 Creation and detection of laser induced dynamic gratings

A laser-induced change of optical properties in materials is always caused by some material excitation, i.e. a deviation from thermal equilibrium, which couples to the complex refractive index. Light scattering from a laser-induced grating can be regarded as a two-step process, see Figure 1. First, irradiation of the sample causes some disturbances of the optical properties of matter by e.g. the opto-acoustic effect (writing of a dynamic grating). Afterwards light is coherently diffracted into a signal beam by these disturbances through the acousto-optic effect (reading of the grating).

![Diagram](image)

**Figure 1:** generation of a laser induced grating structure in a medium. Detection of the grating by diffraction of a laser beam.

In the following the formation mechanisms of a laser-induced grating will be described in more detail. The optical properties of matter, e.g. the refractive index and the absorption coefficient, become spatially modulated in the interference region of two intensive laser beams crossing at a small angle. The spatial modulation of the optical material properties acts as a diffraction grating. The interaction region of the beams defines the sample volume and thus the achievable spatial resolution. Permanent holographic gratings can be produced in this way by photographic processes. In contrast, dynamic or transient gratings disappear after the inducing light source (the laser) has been switched off. Laser-induced gratings can be detected by diffracting a third beam off the laser-induced variations of the optical properties of the material. In gaseous samples the most pronounced optical effects are variations of the density caused by collisional thermalization of absorbed laser energy and by electrostriction, which is the tendency of a material to become dense in presence of an electric field. Gratings generated by such mechanisms are called thermal and electrostrictive gratings, respectively and are
special kinds of laser-induced, dynamic gratings [Eich 1986]. From the temporally resolved
detection of the signal beam diffracted by the induced modulations of the material properties
the grating formation mechanisms as well as gas-kinetic and hydrodynamic properties of the
medium can be inferred.

1.4 Application of laser-induced dynamic gratings

Thermal gratings have found many applications for diagnostics in the gas phase. They have
been employed to investigate quenching processes [Hem 1996], [Fan 2000], [Hub 2000] and
for the measurement of species concentrations with low fluorescence quantum yields
[Hem 1996]. The transport properties of high-pressure fluids have been measured by Kimura
et al. [Kim 1995]. Latzel et al. [Lat 1997] have employed thermal gratings to determine sound
velocity and thermal conductivity of gaseous mixtures of ethylene and nitrogen-helium. In
addition, thermal gratings have been proposed as an alternative method to CARS
thermometry, which is a very reliable and versatile tool for measurements of temperatures
[Gre 1988]. CARS, however, exhibits a complex experimental setup and requires an
elaborated signal analysis. Using laser-induced thermal gratings, the temperature can be
determined by a simple frequency analysis of the oscillations of the grating diffraction
efficiency. Williams et al. [Wil 1994] have applied thermal gratings for the measurement of
temperature in an atmospheric pressure H₂/O₂ flame. Barker et al. [Bar 1999] have employed
thermal gratings for single shot temperature measurements in supersonic air flows. Since soot
particles absorb widely across the electromagnetic spectrum it is possible to generate thermal
gratings with almost any pulsed laser source. Starting from this idea, Brown et al. [Bro 1999]
recognized the potential of laser-induced thermal gratings to perform temperature
measurements in sooty flames.

At the same time, the availability of fixed-frequency laser sources favors the use of laser
induced electrostrictive gratings for several applications, e.g. to study acoustic modes and
their attenuation and coupling in condensed matter [Nel 1981], [Nel 1982], [Yan 1987]. For
diagnostic purposes in the gas phase electrostrictive laser-induced gratings have been applied
to image a stream of helium in air [Hem 1993], to determine sound velocity and acoustic
attenuation in gases [Cum 1994], [Sta 1995], to measure temperatures in air and flames
[Sta 1998], to determine the concentration of binary gas mixtures [Hem 1999], [Hem 1998],
[HeH 1998], and to measure flow velocities and temperatures simultaneously [Hem 2000].
2 Theory

2.1 Superposition of two laser beams

The experimental arrangement for the production of laser-induced gratings is conceptually simple although its realization is sometimes quite demanding. The basic setup is shown in Figure 2, [Eich 1986].

![Grating formation by interference of two light beams with wavelength $\lambda_{\text{exc}}$ and wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$.](image)

Two laser beams, in the following called excitation beams, with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ and electric field amplitudes $\mathbf{E}_1$ and $\mathbf{E}_2$ intersect at an angle $\theta$ in the medium. Their superposition generates an interference pattern characterized by the grating vector $\mathbf{q}$

$$\mathbf{q} = \pm (\mathbf{k}_1 - \mathbf{k}_2)$$  \hspace{1cm} (1)

The absolute value of the grating vector, $q = |\mathbf{q}|$, is related to the fringe spacing of the interference pattern according to

$$q = \frac{2\pi}{\Lambda},$$  \hspace{1cm} (2)
where
\[
\Lambda = \frac{\lambda_{\text{exc}}}{2 \sin \frac{\theta}{2}} \tag{3}
\]

Here \(\lambda_{\text{exc}}\) denotes the wavelength of the excitation laser. \(\Lambda\) is also called “grating wavelength”. The total electric-field amplitude distribution \(E(r, t)\) inside the interference region is the sum of the electric field amplitudes of the two excitation beams. The excitation beams have been assumed to be plane waves. If they have a finite cross section, the lateral extent of the interference region is limited. A calculation of this effect is given in Refs. [Lat 1997] and [Sie 1977].

2.2 Optical gratings

Laser-induced gratings are formed in a medium by various resonant and non-resonant mechanisms as a response to the spatially modulated light field that arises from the interference of two excitation beams. Their total electric field produces some material excitation, which then leads to a change of the optical properties. More precisely, the material excitation couples to the refractive index \(n\) and to the absorption coefficient \(\alpha\) which then exhibit a grating like modulation with amplitudes \(\Delta n(\lambda_{pr})\) and \(\Delta \alpha(\lambda_{pr})\), respectively [Eich 1986]. Both amplitudes are functions of the probing wavelength \(\lambda_{pr}\). Such laser-induced gratings can be probed by detecting the diffracted light from a third laser beam, often having a frequency different from that one of the excitation beams. The part of the probe beam which has been diffracted by the grating is called the signal beam. Gratings characterized by a large grating thickness compared to the fringe spacing, are called thick gratings, see Figure 3b. The Fourier transform spectrum of a thick grating is dominated by two well defined peaks along \(\pm q\) with negligible contributions in other directions. Therefore, thick gratings can be efficiently probed only if the Bragg condition
\[
\mathbf{k}_{pr} - \mathbf{k}_S = m \mathbf{q}, \quad m = \pm 1,2,\ldots \tag{4}
\]
is obeyed, where \(\mathbf{k}_{pr}\) and \(\mathbf{k}_S\) are the wave vectors of the probe and signal beam, respectively. On the contrary, the Fourier transform spectrum of a thin grating is not an isolated spike along one direction, say \(x\), but contains contribution along a direction orthogonal to \(x\) of the order of \(d^{-1}\) caused by the finite thickness of the sample.
Therefore, constructive interference is possible at arbitrary directions of the probing beam, [Eich 1986]. For the experiments described in this thesis only thick gratings and their first-order diffraction, i.e. \( m = \pm 1 \) will be considered. One can see from Figure 4 that the Bragg condition given in Eq (4) determines both the direction of the wave vector \( \mathbf{k}_{pr} \) and its absolute value, which is equal to the absolute value of \( \mathbf{k}_S \).

\[
\frac{I_S}{I_{pr}} = \eta = \left| \frac{\pi \Delta n d}{\lambda_{pr}} \right|^2 = \left( \frac{\pi \Delta n d}{\lambda_{pr}} \right)^2 + \left( \frac{\Delta \alpha d}{4} \right)^2 ,
\]

where \( I_S \) and \( I_{pr} \) are the intensities of signal beam and probe beam, respectively.
Eq. (5) for the diffraction efficiency $\eta$ of a laser-induced grating is valid for gratings with sufficiently small $|\Delta n|$ and low absorption in the material, i.e. $\alpha d \ll 1$. For beams of finite width, the intensities in Eq. (5) can be replaced by the respective ratio of the light fluxes. Very small refractive index changes $\Delta n$ and optical path changes $\Delta nd$ can be measured by diffraction. For example a diffraction efficiency of much less than $10^{-5}$ can be easily detected, corresponding to an optical path length change $|\Delta nd| \approx \lambda/1000$. The phase is thus measured with interferometric sensitivity.

In this work two mechanisms responsible for light-induced modulation of the material properties will be considered: electrostriction and thermalization of the absorbed laser radiation by quenching collisions. Scattering of light from fluctuations of the optical properties of a material caused by the presence of a radiation field are referred to stimulated or forced light scattering processes.

### 2.3 Electrostriction

The process of light scattering by sound waves is known as Brillouin scattering. The variation of pressure, associated with the acoustic waves (i.e. the adiabatic density fluctuation), causes a variation of the real part of refractive index and thus the sound waves act as a phase grating. Sound waves can be generated by e.g. electrostriction, which is the tendency of materials to become dense in the presence of an electric field. Electrostriction occurs at any frequency of the excitation radiation. The electric field of the interference structure of two crossed excitation beams polarizes the dielectric medium. The spatial in-homogeneity of the total electric field in the medium exerts a force on the atoms (or molecules) leading to a non-equilibrium velocity distribution that causes a motion of mass. If the wavelength of the incident light is larger than the nearest molecular resonance, the molecules are accelerated towards the regions of high laser intensity (this case is shown in Figure 5). Otherwise they are accelerated towards the regions of low laser intensity. If the medium is dense enough this velocity disturbance relaxes within a distance that is small compared to the fringe spacing of the interference structure causing a bulk compression and rarefaction of the medium. Therefore, acoustic waves with wavelength and direction that match the geometry of the interference structure are generated. They propagate in opposite directions, normal to the planes of the fringes. The counter-propagating acoustic waves form a standing acoustic wave and thereby a spatially periodic density grating that oscillates in time. The signal beam, obtained by scattering a probe beam off the grating, is modulated in time by the evolution of
the laser-induced perturbation. In Figure 5 the formation process of electrostrictive gratings is summarized.

![Figure 5: Density modulation induced by the electrostrictive force of the excitation field.](image)

In the presence of the electric field the molecules of the medium develop the induced dipole moment, which for isotropic molecules is given by

\[
p = \tilde{\alpha} E,
\]

(6)

where \( \tilde{\alpha} \) is the molecular polarizability.

The energy stored in the polarization of the molecule is

\[
U = \frac{1}{2} \int_0^E \tilde{\alpha} E' \, dE' = -\frac{1}{2} \tilde{\alpha} E^2
\]

(7)

The force acting on the polarized molecules in presence of this field is

\[
F = -\nabla U = \frac{1}{2} \tilde{\alpha} \nabla (E^2)
\]

(8)

The excess of pressure induced by the electric field, called electrostrictive pressure, is given by [Boy 1992]:

\[
p_{str} = -\frac{\varepsilon_0}{2} \rho (\partial \varepsilon / \partial \rho)_{\rho = \rho_0} E^2 = -\varepsilon_0 \frac{\gamma_c}{2} E^2
\]

(9)

where \( \gamma_c \equiv \rho (\partial \varepsilon / \partial \rho)_{\rho = \rho_0} \) is known as the electrostrictive constant.
2.4 Thermalization by collisional quenching

Consider again a medium located in the interaction region of two crossed excitation beams. If the frequency of the excitation beams is resonant with an atomic (or molecular) transition of the medium there will be a change of the population distribution among atomic (or molecular) levels away from a thermodynamic equilibrium. Population gratings, reflecting the intensity distribution of the interference pattern, are formed in ground and excited state, respectively. Subsequent thermalization of the absorbed laser energy by quenching collisions leads to temperature and hence to density variations within the medium. Such gratings are called thermal gratings. If the energy release is slow a stationary density variation is generated, leading to thermally stimulated Rayleigh scattering. In this process light is scattered by isobaric density fluctuations that are driven by the process of optical absorption. If the energy release is fast the induced temperature variations cause a thermal expansion that propagates with roughly the sound velocity. In this case sound waves and a stationary density modulation with equal amplitude are created. The acoustic waves formation process from fast relaxation of absorbed radiation energy leads to thermally stimulated Brillouin scattering. [Boy 1992]

2.5 Hydrodynamic equations

In general, any light-induced modulation of a material property with amplitude $\Delta X$ inside a medium will be accompanied by an optical grating with amplitude

$$\Delta n = \left( \frac{\partial n}{\partial X} \right) \Delta X \quad (10)$$

The light-induced modulation of the complex refractive index due to electrostriction and to thermalization of the absorbed laser radiation, is

$$\Delta n = \left( \frac{\partial n}{\partial N_{ij}} \right) \Delta N_{ij} + \left( \frac{\partial n}{\partial T} \right) \Delta T + \left( \frac{\partial n}{\partial \rho} \right) \Delta \rho \quad (11)$$

Here $\Delta N_{ij}$ is the deviation from the thermal equilibrium population difference between the two molecular levels $j$ and $i$ connected by the radiation of the excitation laser. The first term in Eq. (11) describes the influence of the population transfer induced by the radiation of the excitation laser on the complex refractive index. Once the absorbed energy is thermalized the complex refractive index is a function of local thermodynamic quantities. In a gas the thermooptic coefficient $\left( \frac{\partial n}{\partial T} \right)$ is usually small [Boy 1992], [Fab 1968] and can be neglected in comparison to the change of the complex refractive index caused by the density perturbation.
In the following the variation of the density from the initial undisturbed spatially homogeneous value $\rho_0$ will be denoted $\rho'$ (i.e. $\rho'=\rho-\rho_0$) and can be written as

$$\rho'(r,t) = \Delta \rho(t).f(r) \quad (12)$$

where $f(r)$ contains the spatial dependence of the density variation $\rho'$. The temperature variation $T'$ can be expressed analogously. For the experiments described in this study the condition of local thermal equilibrium in the medium is fulfilled, because the grating spacing is large compared to the mean free path, and the mean time between two collisions of a molecule is small compared to the pulse duration of the excitation laser and to the decay time of the density wave.

### 2.5.1 Pure electrostrictive gratings

Let us first consider the situation where the variation of density $\rho'$ is induced only by electrostriction. In this case the variation of the complex refractive index reduces to the last term of Eq. (11). The following linearized equations of fluid dynamics are used to calculate the variations $\rho'(r,t)$ and $T'(r,t)$ from their unperturbed values, (i.e. only terms linear in $\rho'$ and $T'$ are considered) [Boy 1992]:

$$-\frac{\partial^2 \rho'}{\partial t^2} + \frac{\gamma}{\gamma-1} \nabla^2 \rho' + \frac{\beta_p}{\rho_0} \nabla^2 T' + \frac{\eta_{visc}}{\rho_0} \frac{\partial}{\partial t} \left( \nabla^2 \rho' \right) = \frac{\varepsilon_0 \gamma_e}{2} \nabla^2 \mathbf{E}^2$$

$$\rho_0 c_v \frac{\partial T'}{\partial t} - c_v \frac{(\gamma-1)}{\beta_p} \frac{\partial \rho'}{\partial t} - \kappa \nabla^2 T' = 0 \quad (13)$$

Here $v_s = \sqrt{\left( \frac{\partial \rho}{\partial p} \right)_s}$ is the adiabatic sound velocity, $\beta_p = -\left( \frac{1}{\rho} \right) \left( \frac{\partial \rho}{\partial T} \right)_p$ is the thermal expansion coefficient at constant pressure, $\kappa$ is the thermal conductivity, $\eta_{visc}$ the viscosity, $c_p$ is the heat capacity at constant pressure, $\gamma = \frac{c_p}{c_v}$ is the heat capacity ratio, where $c_v$ is the heat capacity at constant volume.

In Eqs. (13) the continuity equation in its linearized form has been used to eliminate the flow velocity from the equation of momentum flow (upper equation) and from the energy transport equation (lower equation). The source term in the equation for momentum flow is the electrostrictive force, i.e. $\nabla^2 p_{str}$, while in absence of absorption the energy transport equation is homogeneous, i.e. its source term is zero.
The bar sign over $E^2$ in the source term of the momentum flow equation denotes a time average over one optical period, and $E(r,t)$ is the sum of the fields of the two excitation beams

$$E(r,t) = E_1(r,t) + E_1(r,t)$$  \hspace{1cm} (14)

The excitation fields are assumed to be polarized parallel to each other and are approximated by $\delta$-function pulses at $t=t_0$. This approximation is valid for a laser pulse duration $\tau$ that fulfills $\tau<<1/\omega'$, $1/\beta_1$, $1/\beta_2$, where $\omega'$ is the acoustic frequency, $\beta_1$ and $\beta_2$ are the decay rates, see definitions below. The induced density variation due to a Gaussian pulse of width $\tau$ is obtained by convoluting the solution of the $\delta$-function with the Gaussian pulse, see Appendix. It is the spatially modulated part of the total field $E^2(r,t)$ averaged over an optical period that leads to the formation of the optical grating [Eich 1986], i.e.:

$$\overline{E^2} = 2E^2 \cos(qx) \tau \delta(t - t_0).$$  \hspace{1cm} (15)

Here the x-axis has been chosen to be parallel to the grating vector $q$ and $q = |q|$, (see Eq. (1) for the definition of the grating vector), and $E$ denotes a real constant. Solving equation system (13) by taking into account only terms up to first order of of $\beta_1/\omega'$ and $\beta_2/\omega'$ the following expression for the induced density variation is obtained, see Appendix and [Hub 1995]:

$$\rho'(t,x) = \frac{\gamma_c E_0^2 \tau q^2}{2 \omega'} \cos(qx) \Theta(t - t_0) \left[ e^{-\beta_1(t-t_0)} \left[ L \sin(\omega'(t-t_0) + \phi) \right] + \xi e^{-[\beta_2(t-t_0)]} \right]$$  \hspace{1cm} (16)

with

$$\xi = \left( \frac{\beta_2(\gamma -1)}{\omega'} \right)$$

$$L = \left( 1 + \xi^2 \right)^{1/2}$$

$$\phi = \arctg (-\xi)$$  \hspace{1cm} (17)

where $\omega'$ is the acoustic frequency, $\beta_1$ and $\beta_2$ are the decay rates. In the approximation that takes into account only linear terms in $\beta_1/\omega'$ and $\beta_2/\omega'$, they can be written as
\[ \omega' = qv_s, \]
\[ \beta_1 = \frac{q^2}{2\rho_0} \left[ \eta_{\text{visc}} + \left( \gamma - 1 \right) \kappa \right], \]
\[ \beta_2 = \frac{\kappa q^2}{\rho_0 c_p}, \]

The solution \( \rho'(x, t) \) is a standing acoustic wave with its time dependence described by the first term in the curled brackets of Eq. (16). In addition to the standing acoustic wave a stationary density modulation with small amplitude is formed. Its time dependence is described by the second term in the curled brackets of Eq. (16). Viscosity and heat conduction damp the acoustic wave while the stationary wave is damped solely by heat conduction as it results from the expression for the decay rates \( \beta_1 \) and \( \beta_2 \). For the experiments considered in this thesis the ratio of the decay constant and the generated acoustic frequency \( \omega' \) is very small compared to 1, i.e. \( \xi' << 1 \). As an example, with a typical acoustic frequency of 50 MHz generated in air at 22°C, \( \xi = 6 \times 10^{-3} \). It follows that the stationary density modulation can be neglected, while the amplitude of the standing acoustic wave is \( L = 1 \) and its phase is \( \phi \approx 0 \).

The density related with the standing acoustic wave oscillates in time passing through its unperturbed value twice during each acoustic period \( T_a \), which is given by

\[ T_a = \frac{2\pi}{\omega'} = \frac{2\pi}{q v_s} = \frac{\Lambda}{v_s}. \]

The diffraction efficiency \( \eta \) of the grating is proportional to the squared amplitude \( \Delta \rho(t) \) of the induced density variation \( \rho'(r, t) \), see Eqs. (5), (11) and (12),

\[ \eta(t) \propto \left( \frac{\gamma \varepsilon_0 E^2 \pi q^2}{\omega'} \right)^2 e^{-2\beta_1 (t-t_0)} (1 - \cos(2\omega'(t-t_0))) \]

Therefore, the diffraction efficiency of a pure acoustic grating, which in this case results from electrostriction, oscillates at half the acoustic period, i.e.

\[ T_g = \frac{1}{2} T_a, \]

and decays with a time constant \( 2\beta_1 \). For air at ambient conditions \( 2\beta_1 \) is of the order of \( 10^6 \text{ s}^{-1} \) for a grating spacing of \( 1.5 \times 10^5 \text{ m}^{-1} \).

Note that linearity in \( \rho' \) has been assumed for the derivation of Eq. (16). In fact for the experiments described in this study the maximum value of \( \rho' \) is small enough to justify this
assumption. As an example, using Eq. (16) one obtains for the induced maximum density variation \( \rho'_{\text{max}} = 1 \times 10^{-4} \text{ kg/m}^3 \) which corresponds to a pressure disturbance of \( 6.2 \times 10^{-5} \text{ bar} \) for an electrostrictive grating generated in argon at 5 bar with excitation energies of roughly 90 mJ each at a wavelength of \( \lambda = 1064 \text{ nm} \) and pulse length of 5 ns, a beam diameter of 1 mm and an intersection angle of the excitation beams of about 1.5°.

### 2.5.2 Thermal gratings generated by absorption followed by fast thermalization

If the medium under investigation absorbs the excitation radiation, thermalization of the absorbed laser energy, as well as electrostriction, will cause a corresponding density variation. Thermal gratings are thus generated in addition to the electrostrictive gratings. In this case the variation of the complex refractive index is described by the first and the last term of Eq. (11). Release of absorbed laser radiation in form of heat by quenching collisions provides the source term in the energy transport equation and Eq. system (13) becomes

\[
\begin{align*}
- \frac{\partial^2 \rho'}{\partial t^2} &+ \frac{V^2}{\gamma} \nabla^2 \rho' + \frac{V_p^2}{\eta} \rho_0 \frac{\partial}{\partial t} \left( \nabla^2 T' \right) + \frac{\eta_{\text{visc}}}{\rho_0} \frac{\partial}{\partial t} \left( \nabla^2 \rho' \right) = \frac{\epsilon_0 E^2}{2} \nabla^2 E^2 \\
\rho_0 c_v \frac{\partial T'}{\partial t} - \frac{c_v}{\beta} \frac{\partial \rho'}{\partial t} - \kappa \nabla^2 T' = \epsilon_0 n c \alpha E^2
\end{align*}
\]

(13'),

where \( \alpha \) is the optical absorption coefficient, \( \zeta \) is the ratio of the instantaneous heat release to the absorbed radiation energy, \( n \) the refractive index, and \( c \) the speed of light. The energy conversion process has been assumed to be instantaneous.

Solving Eqs. (13') by taking into account only terms up to first order of \( \beta_1/\omega' \) and \( \beta_2/\omega' \) one obtains for the density variation, see Appendix and [Hub 2000]

\[
\rho'(t,x) = \frac{\epsilon_0 E^2 x^2}{2 \omega'} \cos(qx) \Theta(t-t_0) \left( P_{\text{electr}}(t) + P_{\text{therm}}(t) \right)
\]

(22)

\[
P_{\text{electr}}(t) = \gamma_{\text{e}} \left\{ e^{-\beta_1(t-t_0)} \left[ L \sin(\omega'(t-t_0)+\phi) \right] + \xi e^{-\beta_2(t-t_0)} \right\}
\]

(23)

\[
P_{\text{therm}}(t) = \gamma_{\text{s}} \left\{ e^{-\beta_1(t-t_0)} \left[ L' \sin(\omega'(t-t_0)+\phi') \right] - e^{-\beta_2(t-t_0)} \right\}
\]

(24)

with
\begin{align*}
L' &= \left(1 + \xi'^2\right)^{1/2}, \\
\phi' &= \text{arctg}\left(-\frac{1}{\xi'}\right), \\
\xi' &= \left(\frac{\beta_2 - \beta_1}{\omega'}\right)
\end{align*}

and \( L, \xi, \phi \) from Eqs. (17) above. \( \gamma_a \) is a dimensionless absorptive coupling constant defined by

\[ \gamma_a = \frac{2\alpha n c v \beta \rho}{c_q q} \]  

Note, that the decay constants \( \beta_1 \) and \( \beta_2 \), see Eq. (18), of the induced density variation \( \rho' \) are proportional to the square of the grating vector and inversely proportional to the equilibrium density. Therefore, laser-induced gratings generated at higher pressures or with large fringe spacing have a longer lifetime. Since electrostriction occurs at any frequency, the temporal evolution of the density variation \( \rho' \) comprises the contributions \( P_{\text{electr}} \) and \( P_{\text{therm}} \), which are caused by electrostriction and by collisional induced release of the absorbed laser energy in form of heat, respectively. Electrostriction generates mainly a standing acoustic wave and a stationary density wave with small amplitude. The diffraction efficiency of the electrostrictive grating oscillates at half the acoustic period.

Release of the absorbed laser radiation in form of heat likewise forms a standing acoustic wave and a stationary density modulation, see Eq. (24). Driven by the heat release the stationary density modulation, described by the last term in the curled bracket on the right hand side of Eq. (24), decays by heat conduction. Again, since for the experiments described in this work \( \omega' >> \beta_2 \), one can approximate the amplitude of the oscillating density by \( L' \approx 1 \) and its phase by \( \phi' = \pi/2 \). The standing acoustic wave induced by instantaneous heat release, i.e. the thermal acoustic wave, is thus \( \pi/2 \) phase shifted with respect to the standing acoustic wave induced by electrostriction. Instantaneous energy release causes, at the locations of high total field intensity, a temperature and pressure rise as in a constant volume process. With time, the acoustic energy leaves the volume leading to a pressure drop and hence to a density reduction. A standing acoustic wave and a stationary density modulation are formed with equal amplitude leading to a density \( \rho' \) which is modulated only below its undisturbed value. Analogously, the density change at the location of low field intensity will be modulated only above its equilibrium value. Thus the total density returns to its undisturbed value once each
acoustic period $T_a$. The diffraction efficiency is proportional to the squared density variation. It is a periodic function that can be decomposed into a superposition of a constant (non oscillating) term, two oscillating terms with frequency $\omega'$ and $2\omega'$, respectively. Since the term with frequency $\omega'$ is dominating, the reflectivity displays an oscillation at the acoustic frequency $\omega'$.

Now, depending on the ratio of $\gamma_a$ and $\gamma_e$, i.e. which mechanisms governs the grating formation, the temporal evolution of the reflectivity will oscillate at the acoustic frequency or at twice the acoustic frequency. If electrostriction and heat release contribute roughly in the same way to the grating formation, the resulting reflectivity will display an oscillation at twice the acoustic frequency with enhanced odd peaks.

2.5.3 Thermal grating by slow relaxation of the absorbed laser radiation

Assuming now that the absorbed energy is no longer instantaneously released into heat the source term in the energy transport equation, lower equation in system (13'), acquires a source term $K$, which can be written as

$$K = 2\varepsilon_\nu n c h \zeta \alpha T^2 \cos(qx) \Theta(t - t_0) e^{-\sigma(t-t_0)} ,$$

where $\sigma$ is a decay rate that can be expressed as $\sigma = h + v$ with $h$ is the decay rate of the heat release from collisional de-excitation transitions between molecular internal energy levels. The term $v$ is the molecular diffusion rate that accounts for the “wash-out” effect of molecular diffusion, which diminishes the spatial modulation of the population grating formed in the ground as well as the excited state. $\Theta(t)$ is the Heavy-side unit step function indicating the start time for the grating formation, and $\zeta$ the portion of excitation energy that is released as heat. The energy conversion process occurs here in a single step characterized by a single decay rate $\sigma$. A calculation of multiple relaxation processes with the corresponding decay rate is given in [Hub 2000]. Considering only terms up to first order in $\beta_1/\omega'$ and $\beta_2/\omega'$, the contribution to the density from heat release can be written as, see Appendix:

$$\rho'_{therm}(t,x) = \frac{1}{2} \frac{\varepsilon_\nu n c h_3 \zeta}{\omega^3} v_5 \Theta(t - t_0) E^2 T \cos(qx) \left\{ e^{-\beta_1(t-t_0)} L^* \sin[\omega'(t-t_0) + \phi^*] \right\}$$

$$\left\{ \begin{array}{c}
-\frac{\omega'}{\beta_2 - \sigma} e^{-\sigma(t-t_0)} \\
\left(1 - \frac{2\beta_2 \sigma}{\omega^2} + \frac{\sigma^2}{\omega^2} e^{-\beta_2(t-t_0)} \right)
\end{array} \right\}$$

with
Depending on the sign and magnitude of the heat release rate $\sigma$ (i.e. if an exo- or endothermic process is considered) the induced density variation exhibit a completely different temporal behaviour. If the absorbed energy is released into heat instantaneously, i.e. $\sigma \gg \omega'$, Eq. (28) becomes

$$L^* = \left( \frac{\sigma(\beta_1 - \beta_2)}{\omega'^2} + 1 \right)^2 + \left( \frac{2\beta_1 - \beta_2 - \sigma}{\omega'} \right)^2 \right)^{1/2}$$

$$\phi^* = \arctan \left( \frac{\left( \frac{2\beta_1 - \beta_2 - \sigma}{\omega'} \right)}{\left( \frac{\sigma(\beta_1 - \beta_2)}{\omega'^2} + 1 \right)} \right)$$

(29)

As expected, this result is equivalent to the one obtained for instantaneous heat release, see Eqs. 22 and 24.

If the process of collisional relaxation of absorbed energy is slow compared to the acoustic oscillation period $2\pi/\omega'$, i.e. if $\sigma \ll \omega'$, Eq. (28) becomes

$$\rho_{\text{therm}}'(t,x) = \frac{\varepsilon_0\gamma_a h q^2 \Theta(t-t_0) E^2 \tau \cos(qx) \left\{ \cos(\omega'(t-t_0)) - e^{-\beta_2(t-t_0)} \right\}}{2\omega'}$$

(30)

When the heat is slowly released, the pressure rise is continuously drained by acoustic waves. The process is nearly isobaric and favors the formation of a stationary density modulation, whereas the development of the standing acoustic wave is hindered by the destructive interference of the sound waves generated at different times. Note that, in order to correctly describe the diffraction efficiency of thermal gratings, one has to include the contribution to the complex refractive index modulation due to the induced population modulation in addition to the induced density modulation, see Eqs. (5) and (11).

For the derivation of the laser-induced variation of density and temperature the condition of local thermal equilibrium has been assumed. If this condition is no longer fulfilled, more complex gas kinetic models have to be employed [Han 2000], [Gri 2000].
3 Results

3.1 Choice of the laser sources and of the experimental parameters

One of the findings of this thesis concerns the choice of the most appropriate optical sources for the application of laser induced gratings in the diagnostics of the gas phase. To set up the grating a strong pulsed laser, and a cw or a pulsed laser to read out the grating are required. Multimode pulsed Nd: YAG lasers, with ns pulse length are good candidates because of their compactness, ease to use, relative low costs and high intensity output. Of course care must be taken when setting up an experiment because their characteristics will affect the measurement in some way. First of all one has to be aware that multimode lasers have a coherence time that is shorter then their pulse length. Therefore the optical path length of the excitation beams must be carefully adjusted to obtain good interference and thus a high diffraction efficiency of the laser induced grating. The influence of this feature upon a generated signal is presented and discussed in the following section 3.1.1. Additionally, the relative high output intensity of these pulsed laser must be controlled since the nature of the induced grating depends on it. High laser intensities can lead to physical and chemical changes of the sample rendering the measure no longer non-invasive. Furthermore damage of the used optics can occur using high laser intensity. The effects of the pump intensity upon the generated signal are discussed in the following section 3.1.2.

Reading out the grating with a cw laser allows to record the grating scattering efficiency over the whole range of its lifetime in a single shots measurement. This is realized by the temporally resolved detection of the signal by fast digitizers, which presently are available at moderate costs. Further advantages which make the cw laser the source of choice for the measurements presented in this thesis are shown and discussed in section 3.1.3.

Understanding how the geometrical alignment and the size of the beams affects the signal through sound propagation out of the measuring volume is a further result of this work which is presented in section 3.1.4. The best experimental configurations for the different application of the laser-induced technique in diagnostics presented in chapter 3.3 have been chosen based on these findings.
3.1.1 Coherence effects

In general, since the coherence time is inversely proportional to the bandwidth, multimode lasers have a short coherence time. For example a Nd: YAG multimode pulsed laser with a bandwidth of ca. 30 GHz has a coherence time of about 30 ps. If the duration of the excitation pulses is much longer than the coherence time of the excitation laser one has to account for the time dependence of the excitation fields to correctly describe the diffraction efficiency $\eta$ of a grating generated by partially coherent excitation beams, i.e. when the time delay $\tau_d$ between them is not zero. The diffraction efficiency of an electrostrictive, laser-induced grating depends on the temporal coherence function of the source, which generates the grating. For the excitation laser used in this work a Gaussian frequency spectrum has been assumed. In this case the diffraction efficiency of the grating can be written as [Sta 1995]:

$$\eta(\tau_d) \approx A \left[ e^{-\left( \frac{\tau_d}{\tau_c} \right)^2} B(t) + \frac{\tau_c}{\tau} C(t) \right]$$

where $\tau_c$ the coherence time of the excitation laser is assumed to be small compared to the pulse width $\tau$. $A$ is a constant that include the grating vector, the intensities of the pump beams, the electrostrictive constant and the grating oscillation frequency. $t$ is the time between grating formation and read out, i.e. the time between excitation and probe beams. $B(t)$ and $C(t)$ are functions that oscillate and decay with increasing time $t$. If $|\tau_d| >> \tau_c$, i.e. when the grating is generated with fully incoherent excitation beams, the first term in the squared brackets of Eq. (32) can be neglected. The diffraction efficiency $\eta$ is reduced approximately by a factor $\tau/\tau_c$, when using temporally incoherent excitation beams instead of temporally coherent ($\tau_d=0$) excitation beams, in the case where the grating oscillation period is large compared to the pulse duration $\tau$, see for details [Sta 1995].

The experimental configuration depicted in Figure 6 is used to investigate the influence of the temporal coherence of the excitation laser on the generated electrostrictive grating signal (see Ref [Sta 1995]).
Figure 6: Experimental setup for the coherence measurements; BS1, BS2 beam splitters (R=70%, R=50%). P1, P2 and P3 polarizers, PMT photomultiplier tube.

To setup the gratings a frequency doubled multimode Nd: YAG (Yttrium Aluminium Garnet, Y₃Al₅O₁₂) laser (Continuum NY81-10) with a pulse width of about 5 ns (full width at half-maximum, FWHM), a repetition rate of 10 Hz, an output energy in the range of 70 mJ/pulse (oscillator output) to 160 mJ/pulse (amplifier) and a beam diameter of 6 mm is employed. The IR spectral bandwidth of its radiation is 1 cm⁻¹ (FWHM). The output of this laser, to which in the following it will be referred to as YAG I, is split by a 50% beamsplitter, BS1, into two excitation beams EB1 and EB2. A variable path length is obtained by directing EB2 through an optical delay line formed by two prisms before it is steered by a second 50% beamsplitter BS2 into a gas cell, where it crosses EB1 at an angle θ=3°. The frequency doubled output of a second Nd:YAG laser (Continuum NY81-20), in the following referred to as YAG II, provides the probe beam, which is diffracted off the grating into the signal beam. YAG II has a pulse width of roughly 8 ns (FWHM), a repetition rate of 20 Hz, a diameter of about 6 mm, and an output energy of approximately 80 mJ/pulse. The probe beam is perpendicularly polarized to the excitation beams and is aligned to be counter-propagating to EB1. Its k-vector lies in the plane defined by the excitation beams EB1 and EB2. The polarizers P1 and P2 (extinction coefficient of 10⁻⁵) in the path of EB1 and EB2, respectively, serve to prevent back-traveling light from YAG I laser to damage laser YAG II and vice versa. Satisfying the Bragg condition, the signal beam is parallel and counter-propagating to EB2. Half of the
signal passes the beamsplitter BS2 and is directed into a photomultiplier tube. A polarizer P3 with extinction coefficient of $10^{-5}$ in the signal path efficiently suppresses stray light from the BS2. Spatial filtering is achieved by coupling the signal into a 200 µm diameter optical fiber of 15 m length. The two lasers are electronically synchronized and the time delay between excitation beams and probe beam can be varied by a digital delay generator (Stanford Research Systems DG535). The data acquisition and the controlling of the delay between the two lasers are computer controlled. This setup has also been used for the measurement of the temporal evolution of the electrostrictive signal. Usually ten scans over the lifetime range were recorded and each data point was averaged over 100 laser shots. Depending on the lifetime of the generated grating and on the scanning time step chosen such measurement required between ca. 20 minutes and 1 hour. The data files contain thousand up to five thousand data points.

Figure 7 shows a measurement in ambient air of the signal intensity versus the time delay between the two excitation beams. The data are fitted by a sum of a Gaussian function and a constant (solid line). In addition, the data are fitted by the sum of a symmetric exponential function and a constant (dashed line) as one would expect for a Lorentzian frequency spectrum. However, this latter model fails to correctly describe the measurements, especially in the wings where a stronger decrease of the signal intensity with increasing delay time is observed. The assumption of a Gaussian frequency spectrum is therefore justified. The wings of the coherence function display a constant contribution. The Gaussian fit to the data yields a FWHM of $18 \pm 0.5$ ps, which corresponds to a coherence time $\tau_c$ of $20.1 \pm 0.5$ ps. The coherence length and the bandwidth obtained are 6 mm and 1.1 cm$^{-1}$, respectively. The bandwidth of the Nd:YAG laser without any narrowing elements is specified to be 1 cm$^{-1}$ [Con 1990]. For a delay time of 95-101 ns between excitation and probe pulses the signal intensity at $\tau_d=65$ ps is measured to be smaller by a factor of $(3.0 \pm 0.2) \times 10^2$ compared to the signal intensity at $\tau_d=0$. This matches fairly well the calculated value $\tau / \tau_c=4.0 \times 10^2$ and is an additional proof, that in the experiments described here, the assumption of a Gaussian temporal photon statistics for the intensity of the excitation laser is justified.
Figure 7: Signal intensity of an electrostrictive laser-induced grating generated in ambient air vs. time delay $\tau_d$ between the excitation beams. The solid line is a fit by using the sum of a Gaussian function and a constant. The dotted line is a fit by a Lorentzian function.

For a pulsed multimode laser the coherence time is much smaller than the pulse duration and, therefore, in order to obtain high grating diffraction efficiency it is crucial to reduce the optical path difference of the excitation beams to a small fraction of the coherence length of the laser source. This can be achieved by an optical delay line in the path of one (or both) of the excitation beams. On the other hand, the measurement of the grating efficiency as a function of the delay time between the excitation beams allows to determine the temporal coherence function of the excitation laser.
3.1.2 Optical breakdown in gases

The grating diffraction efficiency is proportional to the intensity of the excitation source. Using high intensity excitation beams leads on one side to an increased signal, on the other side it may change the properties of the sample and it influences the formation mechanisms of the induced grating.

Figure 8: Temporal behavior of a laser induced grating generated in air at atmospheric pressure and ambient temperature. The excitation beams have a wavelength $\lambda_{\text{exc}}=532$ nm, a diameter of roughly 6 mm and are focused by a 750 mm focal length lens to intersect at an angle of 1.6°. The probe beam arises from an cw Ar$^+$ laser at $\lambda_{\text{pr}}=514.5$ nm. The phase matching condition (Bragg condition) of the three beam is achieved in a 3D forward geometry. a: signal obtained with roughly 90 mJ/pulse excitation laser intensity; b signal obtained with ca 380 mJ/pulse excitation laser intensity.
The traces in Figure 8 show the temporal behavior of a laser-induced grating generated with excitation beams arising from the frequency doubled output of a Nd: YAG laser at the intensity of roughly 90 mJ/pulse and 380 mJ/pulse, respectively. Both measurements are obtained in 1 bar argon at room temperature using the experimental setup and beam geometry as described for the temperature measurements, see section 3.3.1. Approaching the threshold for optical breakdown the temporal signature of the grating reflectivity changes and its oscillation frequency is reduced to half of its initial value, indicating a release of heat in the medium. The energy deposition in the medium by absorption is excluded since the excitation wavelength $\lambda_{\text{exc}}=532$ nm is not resonant with any transition of argon. In fact, at high laser intensities the mechanisms of energy deposition in the gas occurs in two steps. Initially, electrons may be generated in the gas by multiphoton ionization. Electron avalanche ionization can develop if the initial electrons can gain energy from the laser field, since they can attain enough energy to ionize an atom in collision. Repetition of the process can lead to a rapid multiplication of the number of electrons. From the requirement of energy and momentum conservation, an electron can absorb a photon only if it is colliding with an atom or ion. The electrons in the laser field will gain kinetic energy through electron-neutral inverse Bremsstrahlung collisions [Hug 1975], [Bek 1976]. (this is the inverse of the Bremsstrahlungs process in which high energy electrons, upon traversing a gas emit radiation as they slow down). The accelerated electrons will lose their energy by elastic and inelastic collisions with neutral atoms through excitation of external (i.e. translational) and internal (e.g. electronic) degrees of freedom of the atoms. In this way the radiation energy is “absorbed” by the gas, which is locally heated resulting in the formation of a thermal grating.

Figure 8a shows the situation where a pure electrostrictive grating oscillating at twice the acoustic frequency is generated while in Figure 8b the transition to a thermal grating oscillating at the acoustic frequency is well visible. No attempt was made to quantify this effect. Nevertheless laser-induced gratings show a potential to investigate the behavior of gases at the threshold of optical breakdown.

3.1.3 The read-out beam source

The choice of the pump beam source depending on its characteristics and their influence on the generated grating has been described so far. This section is dedicated to the reasons that makes an Ar+-laser the most suitable read-out beam source even better than the pulsed one used in the previous section. The great advantage of probing the grating with a cw-laser combined with the time resolved acquisition is that the temporal behavior of the transient grating over the whole range of its life time can be detected by a single-shot measurement.
Using a pulsed probe beam to read out the grating, as described in Ref [Sta 1995], one has to electronically delay the probe beam with respect to the grating beams in order to scan over the whole range of the grating lifetime. In this way every measured point of the recorded temporal evolution of the signal arises from a different pulse of the excitation laser and of the probe laser. Thus no single shot measurement of the signal temporal evolution over whole grating lifetime range is possible. Furthermore, to perform a measurement over the lifetime range of the grating can be very time consuming and variations of the laser characteristics, occurring during this time, will eventually also affect the signal rendering its interpretation more difficult. Note that the signal obtained with an intense pulsed probe beam (some MW per pulse) is much stronger than using a cw readout laser (about 1 W). This signal is nearly a single photon event. Despite the high number of photons per pulse no statistical advantage results from using a pulsed probe beam because there is no correlation between the measured data points over the whole range of the lifetime, since they are measured with different excitation and different probe pulses.

The modulation depth of the signal oscillations is larger when using a probe beam from a cw-laser. In fact, the measured signal is the convolution of the temporal response of the grating reflectivity with the temporal shape of the probe beam and the time response function of the detection system. For a cw read out of the grating the convolution simplifies to the temporal response of the grating reflectivity convoluted with the time response of the detection system.

Figure 9a shows the temporal evolution of the signal obtained by diffracting a pulsed probe beam off a laser-induced electrostrictive grating generated in argon at 5 bar and ambient temperature. Every measured data point is averaged over 100 laser pulses. For the measurement shown in Figure 9b a cw probe beam has been employed to perform a measurement of the temporal evolution of a laser-induced electrostrictive grating. This signal has also been generated in Ar at 5 bar and ambient temperature and has been averaged over 10 single shots. Both electrostrictive gratings were generated with unfocused excitation beams at 532 nm intersecting at an angle of about 3°. The excitation beams and the probe beam were arranged in a planar backward geometry. Comparison of the signals depicted in Figure 9a and in Figure 9b shows that the modulation depth of the oscillation of the signal is more pronounced for a cw read-out of the grating as expected.
Figure 9: Temporal behavior of an electrostrictive laser-induced grating generated in argon at 5 bar and ambient temperature. A planar geometry with unfocused beams and an excitation beams intersection angle of about 3° has been used. a: probe beam from a pulsed Nd: YAG laser, every point is averaged over 100 laser shots. b: probe beam from an Ar+ laser, signal is averaged over 10 laser shots.

In the following the wavelength issue of the pump beam source laser is discussed. With the exception of some measurements, the fundamental wavelength of the Nd: YAG laser for the generation of the grating has been used, i.e. $\lambda_{\text{exc}}=1064$ nm. This is more convenient since the photomultiplier tube is not sensitive to infrared radiation, and thus stray light from the excitation beams can be efficiently suppressed without spectral filtering the signal beam. In addition, as one can see from Eq. (3), the grating spacing is proportional to the wavelength of the excitation beams. In this way the wash out of the interference structure occurs later for longer wavelengths of the excitation beams resulting in a longer lifetime of the grating. The scattering efficiency of the grating increases with decreasing ratio $\lambda_{\text{pr}}/\lambda_{\text{exc}}$ because the probe beam must cross the overlap region of the excitation beams at a smaller angle to satisfy the Bragg condition. The smaller the angle between the probe beam and the bisectrix of the angle
between the excitation beams, the larger is the probed portion of the overlap volume of the excitation beams. Since the signal intensity is proportional to the square of the number of molecules, a larger probed volume, like a higher density, leads to larger signals. Using two different wavelengths for the excitation beams and the read out beam allows the collection of the whole signal even in a planar phase-matching beam geometry. In fact the direction of the signal beam does not coincide with the direction of any of the two excitation beams as it happens when using probe and excitation beams of the same wavelength.

### 3.1.4 Sound propagation effects made visible by the alignment of the beams and by their intensity profiles

Since the laser induced grating diffraction efficiency depends on the intensity of the pump beams, one would try to increase the signal intensity by focusing the beams. However, this is not unproblematic and some experimental arrangement are required in order to really attain the wished result. Another important feature of any optical diagnostics approach is its spatial resolution, i.e. how small is the achievable probed volume. This is important in the case of inhomogeneous media or in presence of turbulences. The spatial resolution of the laser-induced grating method is determined by the size of the interaction volume of excitation and probe beams [Sie 1977]. Focusing the beams reduces the interaction volume and may increase the signal intensity, but also increases the decay of the oscillating signal amplitude because of the propagation of the sound wave packet out of the probed volume. This effect might dominate over the usual decay by viscosity and heat conduction. A reduction of the lifetime of the standing acoustic wave leads to a smaller number of visible oscillations of the grating reflectivity. Therefore, the determination of the investigated medium properties from the oscillation period of the standing acoustic wave will be less accurate. In the following these effects are shown and discussed in detail.

Consider for the moment the spatial intensity profile of the excitation beams. Figure 10 shows the temporal evolution of two electrostrictive laser-induced grating signals obtained with different intensity cross sections of the excitation beams.
Figure 10: Temporal behavior of an electrostrictive laser-induced grating in air at room temperature and atmospheric pressure, generated with $\lambda_{\text{exc}}=1064$ nm and $\lambda_{\text{pr}}=514.5$ nm. The excitation beams are crossed by using a 750 mm focal length lens at an angle $\theta=1.5^\circ$. a: the intensity profile of the excitation beams is Gaussian with a diameter of 6 mm ($1/e^2$ FWHM); b: signal generated with astigmatic excitation beams.

Both signals are obtained in air at 1 bar and ambient temperature using excitation beams arising from the fundamental output of the Nd: YAG laser at 1064 nm and a probe beam from a cw Ar$^+$ laser at 514.5 nm. The pump beams are crossed at an angle $\theta=1.5^\circ$ by a 750 mm focal length lens and the phase matching condition of the three beams is realized in a planar backward geometry. Figure 10a shows the temporal evolution of a grating generated by using excitation beams with a Gaussian intensity profile of about 6 mm diameter ($1/e^2$ of the maximal intensity). The measurement depicted in Figure 10b is obtained using astigmatic excitation beams having a strip shaped intensity profile. To obtain such an intensity cross...
section the Nd: YAG beam pass through the Galilean cylindrical telescope before being split into two excitation beams. In this way two astigmatic excitation beams with strip shaped (very tight ellipses) intensity profile about 6 mm high and 1 mm wide are obtained. In the following the ellipses will be referred as stripes, for simplicity. Due to diffraction the heights of the stripes will be focused tighter by the 750 mm focal length lens than their widths resulting in a strip-shaped focus approximately 1 mm wide and 160 µm high.

The orientation of the strip-shaped excitation beams with respect to their plane of incidence is chosen such that the direction of the larger extension of the overlap volume coincides with the propagation direction of the induced sound waves. The signal in Figure 10b displays a remarkable number of additional oscillations and a longer grating lifetime compared to the signal in Figure 10a obtained without shaping the excitation beams intensity cross section.

Essentially two factors limit the lifetime of a laser-induced grating: the propagation of the sound wave packet out of the interaction region, see Eq. (33), and the damping of the acoustic waves due to viscosity and heat conduction, see Eq. (18). The intensity cross section of the astigmatic excitation beams in the focal region is a “strip” of about 1 mm in the direction of propagation of the sound waves. Using $v = 347$ m/s for the sound velocity in air at 1 bar and room temperature, the sound waves would require about 3 µs to completely leave the interaction volume. On the other hand the grating lifetime is also limited by the damping of the acoustic wave which is characterized by a decay constant corresponding to a decay time of the order of 1 µs, see Eq. (18), for these experimental conditions. Thus the damping of the acoustic wave dominates the decay of the generated grating in this case.

The situation is different for excitation beams having a circular intensity profile of about 6 mm diameter, which are focused by a 750 mm focal length lens. The sound waves propagate completely out of the overlap volume, exhibiting a dimension of about 170 µm, after roughly 500 ns, rendering the damping of the acoustic wave unimportant. Therefore, the propagation of the acoustic waves out of the interaction region is the dominant factor limiting the grating lifetime. Using astigmatic excitation beams one observes a larger number of oscillations of the generated standing acoustic wave because the propagation of the sound waves out of the overlap region takes longer compared to the situation where the grating is generated by excitation beams with a circular intensity cross section. Cumings et al. [Cum 1995] provided a thorough analysis of the temporally resolved grating signal, including finite-beam-size effects. However, the resulting expressions are very intricate to provide direct insight. It is often more convenient to account for the faster decay of the standing acoustic
wave by phenomenologically including an additional factor, $f_d$, in the temporal evolution of the diffraction efficiency.

$$f_d = \exp\left(-\frac{4v^2t^2\cos^2\theta/2}{w^2}\right) \quad (33)$$

where $w$ denotes the beam waist (diameter) of the excitation beams. Latzel et al. [Lat 1997] obtained Eq. (33) from the convolution of two counter-propagating Gaussian acoustic wave packets of standard width $\sigma = w/(2\cos\theta/2)$. In summary, using astigmatic excitation beams allow to achieve a trade-off between having a good spatial resolution, high intensity signal and reducing the effect of propagation of the sound waves out of the measuring volume.

In the following other situations where the effect of sound propagation may as well become visible are described. A propagating sound wave causes a non-oscillating contribution in the laser-induced grating signal intensity, which grows and decays in time underneath the oscillatory part. The temporal growth in time of this contribution leads to a reflectivity, which is less and less modulated. Comparison between the measurements obtained in Ar at 5 and 1 bar shown in Figure 11a and Figure 11b, respectively, clearly illustrates this behavior.

Even though the dissipation of the acoustic wave is smaller at 5 bar, the number of visible peaks is roughly the same as in the 1 bar sample. The non-oscillating part of the reflectivity, however, decays much slower at 5 bar. Another parameter influencing the modulation depth of the signal is the accuracy of the geometrical alignment of the three beams. If the three beams are perfectly aligned to satisfy the Bragg condition, the probe beam scatters mainly from the center of the overlap volume of the three beams where the standing acoustic wave is present. A slight misalignment of the beams however, results in poor interference of the counter-propagating acoustic waves and therefore to the presence of additional propagating acoustic waves in the center of the overlap region.
Figure 11: Temporal behavior of an electrostrictive grating generated in argon at room temperature a: at 5 bar and b: at 1 bar. Both measurements are performed using a 3D-forward phase matching geometry with excitation beams at 532 nm of 6 mm diameter focused by a 400 mm focal length lens at an angle of about 3° and a probe beam from a cw Ar+ laser at 514.5 nm. Both signals are averaged over 100 laser shots.

As a consequence, the signal generated in this region displays a slowly varying background resulting from the scattering of the probe beam off the propagating sound wave concurrently with the oscillating signal from a standing acoustic wave. This effect is visible when comparing the signal shown in Figure 12a, obtained with carefully aligned beams, and the signal displayed in Figure 12b obtained with excitation beams crossing before their focal point while keeping the position of the probe beam unchanged.
Figure 12: Electrostrictive grating signals obtained with a: carefully aligned beams and 
b: with slightly misaligned beams, respectively. Both signals are generated in air at 
atmospheric pressure and ambient temperature. The beam geometry is planar with an 
intersection angle of the excitation beams of 1.5°. The wavelength of the excitation 
beams is 1064 nm. The probe beam arises from a cw Ar⁺ laser at 514.5 nm. All the 
beams have a circular intensity cross section with approximately the same diameter. 
Both signals were averaged over 100 laser shots. 

In both measurements the signal arising from the center of the overlap region is recorded 
with a diaphragm aperture of about 4 mm. The beam geometry is planar with an intersection 
angle of the excitation beams of 1.5°, an excitation wavelength at 1064 nm, a probe beam 
from a cw Ar⁺ laser at 514.5 nm and a circular intensity cross section of approximately the 
same diameter for all the beams. Both signals were averaged over 100 laser shots and were 
generated in air at atmospheric pressure and ambient temperature. 
In the following the influence of the size of the probe beam on the occurrence of a 
contribution due to sound propagation in the temporal evolution of the signal is described.
The signal oscillations mirror the oscillations of the standing acoustic wave resulting from the superposition of the counter-propagating sound waves. With time the sound wave packets propagate out of the overlap volume in directions perpendicular to the grating planes. Thus, the resulting standing acoustic wave will be more and more confined in the center of the overlap volume before it disappears completely when the acoustic waves no longer overlap. Afterwards in the periphery of the overlap region, only propagating sound waves will be present. One can distinguish between the two situations sketched in Figure 13, see Ref. [Sie 1977]: the probe beam waist $w_{pr}$ is smaller (a), larger (b), than the spot size of the overlap region $w_{exc}$.

Figure 13: Scattering behavior in the two limits of a: $w_{pr} \ll w_{exc}$, or large walk-off effects, and b: $w_{pr} \gg w_{exc}$, or point scattering behavior.

Consider only the case shown in Figure 13a, when $w_{pr} \ll w_{exc}$, which corresponds to the situation, when astigmatic excitation beams are used. Explicitly, the waist of the focused astigmatic excitation beams in the overlap region is ca. 1 mm in the direction of the propagation of the sound waves while the Ar$^+$ beam’s waist is about 100 µm As depicted in Figure 13a, the scattered beam is much wider than the original probe beam in the plane
containing all three beams. Therefore, the signal comprises contributions generated at the center and at the periphery of the overlap region, respectively. For a selective observation of these contributions an iris diaphragm with a variable aperture was placed in front of the photo-multiplier tube. Its position could be translated in height and in width perpendicularly to the direction of propagation of the signal beam. For the measurement shown in Figure 14a the iris diaphragm with an aperture of about 4 mm was placed at the center of the scattered beam.

![Graph](image1.png)

**Figure 14:** Temporal behavior of a laser induced electrostrictive grating generated in air at ambient conditions. Signal arising from a: the center of the overlap volume and b: 3 mm apart from the center. The measurements were performed using excitation beams from a Nd:YAG laser at 1064 nm intersecting at an angle of about 1.5° and a probe beam from an Ar+ laser at 514.5 nm arranged in a planar backward beam geometry. Every measured point is averaged over 100 laser shots.

Afterwards the diaphragm was translated 3 mm to the left (with respect to the propagation direction of the signal beam) keeping the height and all other experimental parameter
unchanged. The result of this measurement is shown in Figure 14b. Besides a reduction of the signal intensity a contribution from the propagating wave is visible as a slowly varying background leading to a reduction of modulation of the oscillation.

In summary, the possible causes of the presence of a slowly varying background in the temporal evolution of the diffraction efficiency of the grating are a misalignment of the beams or the small dimensions of their interaction volume. Tight focusing of the beams combined with high pressure of the gas render this effect even more visible.

### 3.2 Signal dependence upon pressure and beam intensity

The measurements presented in this section serve on one side to verify the theoretical model and on the other side to observe where it fails to describe the experimental findings and thus how it needs to be improved in a future work.

The pressure dependence of the signal originating from an electrostrictive laser induced grating is given by the electrostrictive constant $\gamma_e$. The ratio of the signal intensity to the probe beam intensity, i.e. the reflectivity, is proportional to the square of $\gamma_e$, see e.g. Eq. 20. From its definition $\gamma_e$ is proportional to the density which scales with the pressure for ideal gases. Figure 15 shows a double logarithmic plot of the electrostrictive laser-induced grating signal intensity versus the air pressure. The fit to the data gives a linear relation with a slope of about 2.

![Figure 15: Signal intensity of an electrostrictive laser-induced grating versus the air pressure](image)

In addition, the dependence of the signal intensity of an electrostrictive laser-induced grating on excitation and probe beam intensity is measured. A variable attenuator (Newport 935-10)
is used to control the intensity of the beams. These attenuators are based on the principle of Fresnel reflection from four uncoated, counter-rotating UV fused silica wedged plates (Newport catalog). Before beginning a measurement it has been verified that the attenuator does not introduce any beam steering effects. The dependence of the signal intensity upon the laser intensity is investigated in air for the range of 15-90 MW/cm$^2$. All three input beams originate from a pulsed Nd: YAG at 532 nm and are arranged in a planar backward geometry in a slightly modified setup as that one used for the coherence measurement described in section 3.1.1. Figure 16 shows a double-logarithmic plot of the electrostrictive laser-induced grating signal intensity versus the laser intensity. A linear fit to the data results in a slope of 3.1, as expected.

![Double-logarithmic plot of electrostrictive grating signal intensity versus laser intensity](image)

Figure 16: Electrostrictive grating signal dependence upon the intensity of the excitation laser. Measurements are performed in air at ambient conditions with $\lambda_{exc}=532$ nm.

The damping of the acoustic wave is described by Eq. 18. Since the viscosity and the heat conduction of gases depend only slightly on the pressure [Ency 1976], the decay time of the grating diffraction efficiency is proportional to the density, and therefore to the pressure. The measurements shown in Figure 17 are performed in argon at pressures of 1 bar (Figure 17a) and 5 bar (Figure 17b) respectively. The experimental setup used for these measurements is identical to the one used for the investigation of the coherence effects, see section 3.1.1. A gas cell built up by standard vacuum supplies is used. It is cylindrical shaped with a variable length and it can be filled to pressures of up to 5 bar. Two fused silica windows allow the optical access from both ends of the cell. The decay time of the diffraction efficiency of the electrostrictive grating signals is obtained by applying an exponential fit to each data set. The
ratio of the decay time for the two measurements is found to be $4.65\pm0.6$, which is in good agreement with the theoretical value of 5.

![Graph showing the temporal behavior of the diffraction efficiency of an electrostrictive grating generated at 1 bar and 5 bar argon at ambient temperature with excitation and probe beams arising from two pulsed frequency doubled Nd: YAG lasers. The phase matching is obtained by a planar backward geometry.](image)

Figure 17: Temporal behavior of the diffraction efficiency of an electrostrictive grating generated a: in 1 bar, and b: 5 bar argon at ambient temperature with excitation and probe beams arising from two pulsed frequency doubled Nd: YAG lasers. The phase matching is obtained by a planar backward geometry.

For a laser-induced thermal grating the calculation of the signal intensity dependence upon the pressure is not straightforward. In comparison to electrostrictive gratings, the generation process is significantly more complex. Figure 18 shows the temporal evolution of the diffraction efficiency of a thermal grating obtained using the excitation wavelength of 1064 nm. Within the bandwidth of the laser, this wavelength is resonant with four transitions in the hot-bands of H$_2$O [HITDATA]. The measurements are performed in the high-pressure...
burner, described in section 3.3.1, in the post flame region of a premixed methane/air flat flame at 5 bar (lower trace) and 10 bar (upper trace).

Figure 18: Temporal behavior of a thermal grating generated in a premixed methane/air flame at 5 (lower trace) and 10 bar (upper trace). A planar backward beam geometry (crossing angle $\theta=2.86^{\circ}$, focusing lens $f=400$ mm) with astigmatic excitation beams at $\lambda_{\text{exc}}=1064$ nm and $\lambda_{\text{pr}}=514.5$ nm is used.

The intensity ratio of the two signals is about 5.7, which is higher than the squared ratio of the pressures. For the calculation of the intensity many parameters are required, e.g. the variation of the local composition with pressure, the kind of species acting as quencher and the speed of the quenching process. Here the effect is shown qualitatively, the quantitative rationalization is an object for future work. Note, that the decay of both signals is similar. At higher temperatures ($\approx 2400$ K) the increased sound velocity ($\approx 990$ m/s) renders the walk off of the sound waves the dominant effect and, therefore, the same decay time for the two gratings results.
3.3 Diagnostics by transient laser-induced gratings

3.3.1 Temperature measurements

One of the most established techniques for the determination of temperatures for diagnostics purposes is Coherent anti Stokes Raman Scattering (CARS) see Ref. [Eck 1988]. Under the numerous advantages of CARS thermometry the two most important are its accuracy, ± 15 K in single shot measurements of flame temperatures e.g. [Pal 1991], and its applicability in harsh environments (such as those characterized by high temperature, high pressure and high turbulence). On the other side the experimental equipment required, i.e. two lasers (narrowband and broadband laser), a spectrometer, a diode array camera, and the complex data analysis correlated with this technique, has fostered the development of alternative diagnostics approaches.

In the following the potential of laser-induced gratings for the determination of temperature in the gas phase and in flames is demonstrated. The results have been published in Ref. [Sta 1998], where the authors reported for the first time to their knowledge the application of this technique for thermometry in gases up to 1400 K and in flames at atmospheric pressures. According to theory the oscillation period of the diffraction efficiency of a laser induced grating is given by

$$T_g = \frac{\Lambda}{f v_s}$$

(34)

where \(f=1\) for a thermal grating and \(f=2\) for an electrostrictive grating.

If the fringe spacing \(\Lambda\) of the grating is known, one can deduce the adiabatic sound velocity in the medium from the oscillation period of the grating diffraction efficiency. For an ideal gas the sound velocity is given by

$$v_s = \sqrt{\frac{\gamma RT}{M}}$$

(35)

where \(T\) denotes the temperature, \(R\) the molar gas constant, \(M\) the molar mass, and \(\gamma\) the specific heat ratio. For gas mixtures \(M\) and \(\gamma\) are given by

$$M = \sum_{i=1}^{s} x_i M_i$$

(36)

$$\gamma = \frac{\sum_{i=1}^{s} x_i M_i c_p}{\sum_{i=1}^{s} x_i M_i c_v}$$

(37)
where $x_i$ denotes the mole fraction, and $s$ the number of different compounds. By measuring the sound velocity one can either determine the temperature, if the gas composition is known, or, under favorable conditions, deduce the concentration of the components in a binary mixture if the temperature is known, see chapter 3.3.2. The experimental configuration used for the temperature measurements is depicted in Figure 19, see Ref. [Sta 1998].

The two excitation beams are obtained from the fundamental output of a pulsed Nd: YAG laser (Continuum, NY81-10). The pulse length is about 5 ns, the repetition rate 10 Hz and the spectral bandwidth of the radiation is 1 cm$^{-1}$ (FWHM). The output energy of the oscillator is about 140 mJ/pulse, which can be increased to 320 mJ/pulse after the amplifier. The output radiation of the Nd: YAG passes through a Galilean telescope consisting of two cylindrical lenses and is subsequently split by a 50% beamsplitter into the two excitation beams. The
circular intensity cross section (diameter ~ 6 mm) of the excitation laser beam becomes elliptical shaped (6 mm high, 1 mm wide) after the cylindrical telescope. The reason why it is advantageous to use astigmatic excitation beams is explained in section 3.1.4. In order to obtain equal optical path lengths for the excitation beams they both pass delay lines before intersecting at an angle \( \theta = 1.5^\circ \) by a 750 mm focal length lens (L1). The laser-induced grating is read out by the beam of a cw Ar\(^+\) laser (Spectra Physics 587 Z-LOK), operated in single line modus at 514.5 nm with a power of approximately 1.7 W. As shown in Figure 19b, a 3D backward geometry was used to adjust the Bragg angle. The readout beam is focused by a second lens, L2, with focal length of 750 mm and directed into the interaction region of both excitation beams. The signal beam leaves the interaction region counter-propagating to the direction of the excitation beams, is collimated by lens L1 and directed over a path of 5 m onto the aperture of a photo-multiplier tube (Philips XP2020). The time resolved acquisition of the signal is performed by a digitizer (Tektronix RTD 720) with a full bandwidth of 500 MHz and a 2 GHz sampling rate. The time response of the detection system is given by the bandwidth of the digitizer, and the rise plus decay time of the photo-multiplier of about 3 ns each.

The determination of the spatial resolution of the beam geometry of the setup used is described here below. Compared to air, the grating diffraction efficiency in helium is smaller by a factor of about 66. A helium flow was translated across the grating volume along the direction of the bisector of the two excitation beams. The helium emerged in ambient air from the circular outlet (diameter 5 cm) of a flat flame burner (McKenna). In Figure 20 the total grating signal is plotted versus the relative distance between helium flow and grating volume. The zero point of the translation was chosen arbitrarily in such a way that the grating volume is entirely within the helium flow. The solid line depicted in Figure 20 is a fit to the data points by a cumulative normal distribution (error function) resulting in a standard deviation \( \sigma = 4.1 \pm 0.7 \) mm. Therefore 95% of the signal is generated in a volume extending \( 4\sigma = 16.4 \pm 2.8 \) mm in the direction of the bisector of the two excitation beams. This value can be considered an upper limit of the longitudinal size of the probe volume. Certainly there is no sharp boundary of the helium flow in air but no attempt was undertaken to account for such edge effects and their influence on the determined spatial resolution.
Figure 20: Determination of the spatial resolution of the laser-induced grating technique used for temperature measurements. The total electrostrictive signal is plotted versus the relative distance between helium flow and grating volume. The data are fitted by an error function (solid line) with a standard deviation $\sigma=4.1$ mm. The excitation wavelength is 1064 nm and the crossing angle is 1.5°.

To evaluate the error of the temperatures measured by electrostrictive laser-induced gratings, measurements in the center of an open tube furnace operated up to a maximum temperature of 1370 K have been performed. The length of the isothermal zone ($\sigma=5$ K) exceeds 10 cm at all temperatures. The temperature reading of the furnace was measured by a thermocouple located in the center of the heated zone. Laser-induced electrostrictive gratings measurements are carried out in air at different furnace temperatures. Since at high temperatures the decreased density causes a reduction of the signal intensity, measurements at temperatures above 600 K were performed with and without amplifier of the Nd: YAG laser. Beside a better signal-to-noise ratio at higher output powers of the Nd: YAG laser no systematic discrepancy for the determined oscillation periods is observed.

The largest error in the determination of the sound velocity results from the measurements of the angle $\theta$ between the excitation beams. Its determination, obtained by measuring the distances for the geometry of the two crossed excitation beams, has an estimated accuracy of 0.05°. This corresponds to an accuracy of 2% in the determination of the sound velocity. Alternatively, to avoid the determination of the fringe spacing $\Lambda$ by a geometric measurement of the angle $\theta$, a reference measurement at room temperature in ambient air has been carried out. By using Eqs. (34) and (35), the measured temperature is given by

$$T = T_R \left( \frac{T_{g,R}}{T_{g,T}} \right)^2 \left( \frac{\gamma_R}{\gamma_T} \right) \left( \frac{M_T}{M_R} \right) \quad (38)$$
Here the subscripts, R and T refer to reference and temperature measurement, respectively. Changes in the gas composition caused by thermo-diffusion can be neglected and, therefore, the ratio of the relative molecular masses is assumed to be unity. With increasing temperature the specific heat ratio $\gamma$ decreases due to excitation of internal degrees of freedom of the molecules like rotation or vibration. For almost all gas molecules the rotational degrees of freedom is fully excited at room temperature (e.g. rotational temperature of O$_2$ and N$_2$ is 2.1 K and 2.9 K, respectively). The vibration temperatures of O$_2$ and N$_2$ are 2239 K and 3352 K, respectively. At temperatures up to 1400 K, the excitation of vibrational degrees of freedom gives an important contribution to the heat capacity. However, due to the high frequency of the generated sound waves, about 10 MHz, one has to consider dispersion caused by the different relaxation times of the internal degrees of freedom. While the rotational and translational relaxation frequencies lie well above the generated sound wave frequency, the vibrational relaxation frequency is some 100 kHz, depending on the humidity of air, see [Har 1999] and references therein. Therefore, the vibrational contribution to the specific heat can be safely neglected and the specific heat ratio for O$_2$ and N$_2$ can be approximated by 7/5 for the investigated temperature range.

In Figure 21 an example of the temporal evolution of the grating diffraction efficiency of an electrostrictive measured in air at atmospheric pressure for two different temperatures is shown. Both traces are normalized to their respective maximum. At higher temperature the lower density causes a stronger damping of the acoustic wave. This results in a faster decay of the grating diffraction efficiency. The higher sound velocity at higher temperature results in a shorter oscillation period of the grating diffraction efficiency.
Figure 21: Temporal evolution of the grating signal in ambient air at temperatures of 297 K (upper trace) and 978 K (lower trace). The signals are averaged over 100 shots.

In Figure 22 the temperatures determined by laser-induced electrostrictive gratings are plotted versus the temperature readings of the furnace. For clarity, a broken line indicating equal temperatures reading for each method is also shown. Each measured value is an average over 100 laser shots. Typically, the standard deviation of the measured oscillation period is 0.5%. With increasing temperature a decreased gas density, together with increased values of thermal conductivity and viscosity, leads to a reduction in signal strength. Additionally, since the damping of the acoustic wave is stronger, a smaller number of visible peaks in the temporal evolution of the laser-induced electrostrictive grating signal is observed. The standard deviation for the determined oscillation period is therefore larger and thereby an increased error for the temperature measurement results.
Figure 22: Temperature measurement in air at atmospheric pressure by laser induced electrostrictive versus temperature measurement by a thermocouple inside the furnace. The signal is averaged over 100 shots. The dotted line corresponds to an equality of the temperature measurements.

There is agreement within the error bars between the temperature determined by the laser-induced gratings method and the thermocouple readings. To estimate the statistical error of single-shot temperature measurements by the laser-induced electrostrictive gratings method, a number of single-shot measurements have been carried out at a fixed furnace temperature of 1370 K (reading of the thermocouple). A number of 100 single-shot measurements is considered to be a good basis for a statistical analysis. The measured temperatures were compiled in a normalized histogram with a temperature interval of 10 K. Together with the histogram of the temperatures, a fit to the data with a Gaussian probability function is shown in Figure 23. From the measurements a mean temperature of 1470 K with a standard deviation of 72 K is deduced. The width of the distribution essentially reflects the uncertainty of the measurement. The consistency with the Gaussian distribution implies that random processes are responsible for the observed fluctuation in the determined temperatures. Because of the limited photons flux these fluctuations may arise from the photoelectron statistics and the quantization noise of the digitizer.
Figure 23: Single shot temperature measurements by laser-induced electrostrictive gratings in air at a furnace temperature $T_f=1376$ K. The temperatures are compiled in a normalized histogram with a temperature interval of 10 K. The dotted line corresponds to a Gaussian probability function with an average temperature and standard deviation of 1370 and 72 K, respectively, fitted to the data.

The potential of the laser-induced grating technique for diagnostics was tested in stoichiometric flames at atmospheric pressures stabilized on a flat flame burner (McKenna). The diameter of the water-cooled, sintered stainless steel flame holder is 5 cm. In order to isolate the flame from the ambient air the flame holder is surrounded by a $N_2$-floated guard ring. Mass flow controllers, assuring stable operating conditions, were employed to adjust the equivalence ratio. Depending on the topic of investigation different fuels and oxidizers are used. Figure 24 shows the temporal evolution of the grating diffraction efficiency obtained in the post-flame region of a laminar premixed stoichiometric methane/air flame. The measured volume is located about 2 mm above the burner surface and the Nd: YAG laser is operated without amplifier.
Figure 24: Temporal behavior of a thermal grating obtained in a stoichiometric CH₄/air flame. The height above the burner surface is 2 mm. The signal is averaged over 100 laser shots. From the measured oscillation period of the signal a temperature of 2080 ± 245 K is determined.

Although the signal intensity was averaged over 100 laser pulses, its signature still exhibits distortions because of the limited photon flux. The determined oscillation period of the grating diffraction efficiency is 43.3±2.5 ns. The small peak at the beginning of the signal does not fit into the determined period and can also not be attributed to scattered light from the Nd: YAG laser. Using Eq. (34) with f=2 for an electrostrictive grating, taking into account the gas composition known from equilibrium calculations and the measured oscillation period one obtains for the temperature roughly 580 K. However, 2065±50 K is an established temperature value for a stoichiometric methane/air flame at atmospheric pressure [Law 1990]. This strong discrepancy together with the appearance of the small peak at the beginning of the signal suggests that the flame absorbed the excitation radiation. In this case a thermal grating is formed and one has to take f=1 in Eq. (34). A flame temperature of 2080±245 K is obtained which is a more realistic value. For measurements in a stoichiometric hydrogen/air flame and taking f=1 in Eq. (34) also results in a realistic value for the determined flame temperature. Therefore the most probable species absorbing the fundamental wavelength of the Nd: YAG laser are H₂O and OH. A search through the database of HITRAN [HITDATA] containing a file with transition of water vapor at 1500 K, delivered four absorption lines of H₂O within the spectral bandwidth of the Nd: YAG laser. The final proof for the correctness of the assumption of a signal from a thermal grating generated by H₂O hot band absorption lines is supported by the measurement depicted in Figure 25, showing a signal obtained in a premixed laminar CO/O₂ flame stabilized on a flat flame burner.
Figure 25: Temporal evolution of an electrostrictive laser-induced grating generated in a stoichiometric CO/O\textsubscript{2} flame 2 mm above the burner surface. The signal is averaged over 100 shots. From the measured oscillation period and the gas composition a temperature of 2215 ± 240 K is determined.

Every data point is averaged over 100 laser pulses. The measurement volume is 2 mm above the burner surface. To perform this measurement the amplifier of the Nd: YAG laser is used in order to obtain enough signal intensity. However, the signal is very low and dominated by single-photon events. The determined oscillation period was 23±1.3 ns. The local gas composition in the measurement volume is estimated from a one-dimensional freely propagating flame calculation. This modeling provides the major species mole fractions of \( x_{\text{CO}_2} = 0.39, \ x_{\text{CO}} = 0.37 \) and \( x_{\text{O}_2} = 0.15 \). From the oscillation period of the signal and the gas composition a temperature of 2215±240 K is calculated using \( f = 2 \) in Eq. (34). This result agrees fairly well with the adiabatic temperature of 2460 K obtained from the model calculation. However, heat conduction from the flame to the burner surface leading to a lower adiabatic flame temperature, has not been taken into account in the calculation. Thus the good agreement between the measured and calculated temperature allows to conclude that the nature of signal recorded in a CO/O\textsubscript{2} flame is purely electrostrictive. Note that it is not clear a priori which is the nature of the generated grating. On the other side the large difference in the determined temperatures when using different values for \( f \), together with the possibility to verify if the excitation wavelength is resonant with a molecular transition of the sample or not, will finally consent to make the right selection.

Finally, the method was applied for thermometry in a high-pressure burner, which was constructed at PSI following drawings of O.N.E.R.A. France [Arn 1997]. The flames were
stabilized in a pressure range between 1 and 25 bar on a water cooled sinter plate of 20 mm diameter. Gas velocity and mixture composition are controlled by mass flow controllers, and the possible mixing ratios $\lambda$ (air to fuel ratio) were between 0.5 and 1.3. Optical access to the cylindrical vessel is obtained by two pairs of suprasil/w windows (20 mm diameter and 10 mm thick) orthogonal to each other. To validate the temperature measurements by the method of laser-induced gratings nitrogen-Coherent Anti-Stokes Raman (nitrogen CARS) thermometry is performed simultaneously in the post-flame zone of premixed methane/air flames using the mobile CARS system developed at the Paul Scherrer Institute. Up to pressures of 10 bar a fairly good agreement between the temperatures obtained by CARS and by the laser-induced grating method is observed. This is shown in Figure 26 where temperatures obtained by laser-induced gratings recorded at different days together with a single CARS temperature are plotted vs. the pressure. At higher pressures the flame in the high-pressure burner becomes unstable (turbulent) and the temperatures determined with the two methods differ strongly.

![Figure 26: Temperatures measurements in the post-flame gases of a premixed methane/air flame by laser-induced thermal gratings, recorded at different days and by CARS (open circles) vs. pressure.](image)

The reason of this discrepancy together with the bad reproducibility of the temperatures obtained by laser induced gratings at different days, are at present unclear and needs to be investigated in more detail in the future. Beam steering resulting in a shot-to-shot variation of the intersection angle of the excitation beams at higher pressures may be an explanation for the discrepancy between the two temperatures. Moreover the small diameter of the flame cone
in addition to the limited spatial resolution of the laser-induced grating may have affected the temperature evaluation.

### 3.3.2 Concentration measurements

Laser-induced electrostrictive gratings can also be employed to determine the concentration of binary mixtures of known temperature. There are two possible strategies to perform such a measurement. Since every individual gas has a specific electrostrictive constant the total signal intensity obtained in a mixture will depend on the relative concentration of the components. The inconvenience of this approach, as for every absolute intensity measurement, is that the intensity fluctuations of both excitation and read-out lasers affect the signal intensity. To account for these intensity fluctuations it is necessary to simultaneously record the intensity of the three beams during the whole measurement period and correct the signal by these reference measurements. Furthermore, the temporal evolution of the signal has to be modeled carefully to include the effect of viscosity and heat conduction. On the other hand, the sound velocity and thereby the oscillation period of the electrostrictive grating diffraction efficiency varies by changing the gas composition. The determination of the oscillation period of an electrostrictive laser induced grating allows to determine the sound velocity of the binary mixture, see Eq. (35). For a binary mixture Eq. (36) simplifies to

\[ M = x_1 M_1 + (1 - x_1) M_2 \quad (36') \]

where \( x_1 \) is the concentration of the one species. Using Eq. (35) it is possible to determine the concentration \( x_1 \) if the temperature of the mixture is known. The concentration of the other species is equal to \((1-x_1)\).

To evaluate the performance of this technique, measurements in binary mixtures in a cell at room temperature are carried out with the same experimental setup used for thermometry. As model for commonly used fuel/oxidant combinations, methane/nitrogen and hydrogen/nitrogen are studied. Since the electrostrictive response in oxygen and nitrogen is very similar, nitrogen has been preferred in all experiments for safety reasons.

In Figure 27 the oscillation period of the electrostrictive grating diffraction efficiency, which is inversely proportional to the sound velocity, is plotted versus the methane mole fraction in nitrogen.
Figure 27: Oscillation period of the diffraction efficiency of an electrostrictive laser-induced grating measured at 1 bar total pressure and room temperature in a methane/nitrogen mixture. The dashed line is the theoretical curve based on a calculation using Eqs. (34) to (36'). The solid line is a polynomial fit to the measured oscillation periods.

The determined oscillation period is obtained from the measurement of the temporal evolution of the electrostrictive grating diffraction efficiency. Every measurement is an average over 100 laser shots. Increasing the methane mole fraction results in a decrease of the determined oscillation period, which corresponds to an increased sound velocity, until the sound velocity in pure methane is obtained. The dashed line in Figure 27 is calculated using the Eqs. (34), (35), and (36’) and employing the oscillation period measured in pure nitrogen as a reference. The effective values of molar mass and of molar heat capacities are calculated from the corresponding values of the gas mixture components. As shown in Figure 27, good agreement between calculation and measurement is found, especially for a low mole fraction of fuel in N₂. The small systematic difference that becomes more important with increasing methane mole fraction is probably caused by the dispersion of sound velocity that takes place in methane. The solid line in Figure 27 is a polynomial fit to the measured oscillation periods. The slope of the curve increases with increasing methane mole fraction. At a value \( x_{\text{CH}_4} = 0.5 \) the slope is 13.5 ns/(mole fraction). The standard deviation of the measured oscillation period is about 0.5%. This uncertainty limits the detection sensitivity for the variation of the gas composition in a 1:1 mixture of methane and nitrogen to 2%. The detection sensitivity will increase for larger sound velocity difference of the mixture components. As an example Figure 28 shows a plot of the measured oscillation period of the electrostrictive grating diffraction efficiency versus the mole fraction of hydrogen in nitrogen. At \( x_{\text{H}_2} = 0.5 \) mole fraction, the slope is 40 ns/(mole fraction). This translates to a
minimum detectable concentration variation of the gas composition value of 0.6% for a 1:1 mixture of hydrogen and nitrogen. In almost pure hydrogen the detection limit for nitrogen is about 0.1%.

Figure 28: Oscillation period of the diffraction efficiency of an electrostrictive laser-induced grating measured at 1 bar total pressure and room temperature in a hydrogen/nitrogen mixture. The dashed line is the theoretical curve based on a calculation using Eqs. 34 to 36'. The solid line is a polynomial fit to the measured oscillation periods.

The dashed line in Figure 28 is calculated analogously as described for the data in Figure 27. Again, good agreement between calculation and measurement is found, especially for low mole fractions of hydrogen in N₂. The small systematic difference that becomes more important with increasing methane mole fraction is probably caused by the dispersion of sound velocity that take place in hydrogen.

To assess the statistical error of a single shot determination of the oscillation period for concentration measurements, about 60 single-shot measurements in a mixture of 0.55 mole fraction of methane in nitrogen have been carried out. Figure 29 shows the oscillation periods compiled in a normalized histogram with a bin width of 0.1 ns. The fitted Gaussian probability density function (solid line) of the measured oscillation period has a well-defined maximum at 44.1 ns with a width (FWHM) of less than 0.2 ns.

By simultaneous measurements of the oscillation period and of the decay time τ, both concentration and temperature are eventually accessible at once. Figure 30 is a plot of the temperature vs. the mole fraction of methane in nitrogen for different values of sound velocities and of the parameter α, which is defined as α=ln(τ) . The contours of the sound velocity (dashed lines) are plotted for 300 m/sec, 400 m/s, 500 m/s and 600 m/s and are obtained using Eqs. (35), (36') and (37). The solid lines represent the contours of the parameter α=ln(τ) for the values between −1 and +1, where τ is the decay time in μs.
Figure 29: Single-shot measurements of the oscillation period of an electrostrictive laser induced grating diffraction efficiency in a mixture of 0.55 mole fraction of methane in nitrogen. The measured period is compiled in a normalized histogram with a bin width of 0.1 ns. The solid line is a Gaussian fit to the data centered at 44.1±0.2 ns.

Figure 30: Plot of the temperature vs. mole fraction of methane in nitrogen for constant sound velocities and logarithm of the grating decay time. The dashed lines are the contours of the sound velocities in m/s. The solid lines are the contours of the parameter $\alpha$ which is defined as $\alpha=\ln(\tau)$ where $\tau$ is the decay time in $\mu$s. For the calculation atmospheric pressure and a grating fringe spacing $\Lambda=30$ $\mu$m are assumed.

The temperature versus the mole fraction contours of parameter $\alpha$ are obtained by calculating the density, see Eq. 18, and the corresponding temperature assuming a pressure of 1 bar. For temperature values below 600 K the decay time remains nearly constant with concentration. Consider for example the line of parameter $\alpha=0$ that corresponds to a decay time of 1 $\mu$s.
this value of the decay time the possible temperatures are in the range between 430 K and 450 K. If in addition a sound velocity of 500 m/s is determined from the grating oscillations, then a range for the possible concentrations of methane in air between 0.79 and 0.83 can be deduced. Thus, for binary mixtures the method is applicable to determine (or to give a range of) the concentration and temperature simultaneously.

3.3.3 Flow velocity measurements

Flow velocity and temperature are key parameters in aerodynamic and reacting flows. While there is a variety of dedicated techniques available to determine one of these two parameters, there are few attempts to measure both simultaneously. Well established methods to measure flow velocity rely on seeding the flow with particles e.g. Laser Doppler Anemometry (LDA), Particle Imaging Velocimetry (PIV) and Doppler Global Velocimetry (DGV), which do not allow a non-intrusive measurement. Furthermore due to particle lag, these techniques fail to measure high-velocity flows and for the determination of flow velocity close to surfaces. Single-shot time domain coherent anti-Stokes Raman Scattering [Sche 2000] circumvents these problems and allows for simultaneous measurements of temperature and flow velocity. Due to the limitation by the Raman Coherence time, this technique is more suited for measurements in high velocity, low-pressure flows. For room temperature flows the pressure limit is about 0.01 bar. Schlamp et al. [Sch 1999] demonstrated the possibility to deduce flow velocities by analyzing the temporal shape of a signal originated from thermal grating generated with misaligned beams geometry. Walker et al. [Wal 1998] employed the Doppler shift experienced by a laser beam that reads out a thermal grating, in the frequency rather than in the time domain to determine flow velocities. Decay analysis of laser-induced electrostrictive gratings as well as time-of-flight approach have been proposed by Ribet et al. for flow velocity measurements [Rib. 2000].

In the following the results of Refs. [Koz 2000] and [Hem 2000] are presented, where it was demonstrated for the first time to authors knowledge that laser-induced electrostrictive gratings can be applied to measure flow velocities and temperature simultaneously. The principle of the method developed by the authors for this purpose is shown in Figure 31.
Figure 31: Principle of flow velocity measurement by laser induced electrostrictive gratings. Two signals generated from reading out the grating in opposite direction are Doppler shifted by $\Delta \omega = \pm q v_f$ due to the movement of the flow. From the heterodyne detection of the two signals the flow velocity is obtained.

It consists in local “seeding” of the jet with a laser-induced standing ultrasound wave (i.e. a laser induced grating) and the heterodyne detection of light scattered by this wave. If an electrostrictive grating is generated in a flow moving with velocity $v_f$, the induced density modulations are no longer at rest but will move along with the flow. A probe beam diffracted by such grating will be frequency shifted. This shift can be interpreted as a Doppler effect due to the movement of the grating. If the grating is read out in opposite direction to the flow, see upper left hand side of Figure 31, the frequency shift is $\Delta \omega = + q v_f$, where $q$ is the grating vector and $v_f$ is the flow velocity vector. For a read-out of the grating in opposite direction to the flow as shown in the lower right hand side of Figure 31 the frequency shift is $\Delta \omega = - q v_f$.

The frequency shift is given by the velocity component normal to the grating planes and it reaches its maximum value when the planes are perpendicular to the flow velocity vector. Another way to understand the principle of measuring flow velocity by electrostrictive laser induced gratings is the following. In the interference region of two crossed excitation beams, two sound waves of velocity $v_s$ propagating in opposite directions $\pm q$ and are generated due to electrostriction. Their superposition results in a standing acoustic wave that diffracts a probe beam of frequency $\omega_0$. The signal beam comprises two contributions arising from the scattering of the probe beam off each sound wave. Due to the Doppler effect each contribution to the signal is frequency shifted by an amount $\Delta \omega = \pm q v_s$, note that $q v_s$ is equal to the acoustic frequency $\omega'$. The beating of these two contributions results in an oscillation
of the signal at twice the acoustic frequency, $2q\nu_s$, i.e. $2\omega'$, in agreement with the theory. If
the grating is generated in a moving medium and it is read out in opposite direction by two
probe beams and the heterodyne detection of the two signals is performed as sketched in
Figure 31, the total signal comprises of four contributions. Heterodyne mixing of all
contributions results in signal characterized by a power spectrum containing the frequencies
$2\omega'$, $2q\nu_f$, $2\omega'\pm2q\nu_f$ in addition to the carrier frequency $\omega_0$ of the probe beam. Since
$q\nu_f/\omega'<<1$ the contributions of frequency $2\omega'\pm2q\nu_f$ can not be distinguished from those of
frequency $2\omega'$. The total signal will display a fast oscillation of frequency $2\omega'$ modulated
by a lower frequency $2q\nu_f$ due to the mixing of light of opposite Doppler shifted frequency.
The setup used to perform flow velocity measurements is obtained by a slightly modification
of that one used for the determination of temperatures, see Ref. [Hem 2000]. A scheme of the
modified setup is given in Figure 32.

Figure 32: Setup for the velocity measurements; BS, beamsplitter (50%); $L_1$, $L_2$ and $L_3$
lenses ($f = 1000$ mm); DL delay line; M, mirror; PMT, photomultiplier tube; DI,
digitizer; $S_1$, $S_2$, signal beams; $R_1$, $R_2$ read out beams.

Excitation and probe beams arise from the same lasers used for the temperature
measurements. To setup up the grating the fundamental output of the pulsed Nd: YAG is
directed through a 1000 mm focal length lens, $L_1$, and is split by a 50 % beamsplitter, BS,
into two excitation beams of roughly equal intensity. An adjustable delay line in the path of
one of the excitation beams serves to reduce the optical path difference to a small fraction of
the coherence length of the laser radiation. Both excitation beams intersect at the focal
distance of $L_1$, at an angle $\theta$–2.7°. The beam waists there, i.e. the diameter of the $1/e^2$
intensity contour $2w_0$, were determined to be about 300 $\mu$m. The resulting estimated effective grating thickness, which is approximately equal to the interaction length of the excitation beams, measures about 10 mm. A beam, $R_1$, from an Ar$^+$ laser at 514.5 nm is used to read out the grating. The probe beam $R_1$ is focused into the interaction volume of the excitation beams by a 1000 mm focal length lens, $L_2$. Phase matching of the three beams is obtained by a planar backward beam geometry. The signal beam $S_1$ leaves the interaction volume counter-propagating to the direction of the excitation beams. It is re-collimated by lens $L_3$ ($f=1000$ mm) and reflected back into itself by mirror $M$. After passing through the flow, the read-out beam $R_1$ is retro-reflected by mirror $M$ and forms a second read-out beam, $R_2$. This beam reads out the grating in exactly opposite direction to beam $R_1$ and generates the signal beam $S_2$. Both signal beams $S_1$ and $S_2$ leave the interaction volume co-propagating. However, signal $S_1$ is temporally delayed with respect to signal $S_2$ by $\Delta t=8$ ns, which is the time required to travel twice the distance between the interaction volume and the mirror $M$. Both signal beams are coherent to each other, have equal amplitudes, are spatially overlapped, and therefore fulfill the requirements for heterodyne detection. The signals are spatially filtered by an optical fiber and detected by a photomultiplier tube. The time resolved acquisition of the data is performed by a digitizer with full bandwidth of 500 MHz and 2 GHz sampling rate. All flow velocity measurements were performed in a submerged air jet at atmospheric pressure in the exit plane of a slot nozzle. The nozzle was manufactured from a copper tube 20 mm long with an inner diameter of 15 mm. By squeezing one end of the tube a slit-nozzle is obtained. Its length is about 20 mm, the height is 1.5 mm and the width is 22 mm. The nozzle cross section at its exit is $S_n\approx32.5$ mm$^2$. A resistance heater wrapped around the copper tube allows heating of the wall up to 1100 K. The temperature of the wall is controlled by a thermocouple.

In the experimental configuration for flow velocity measurements described above the two signal beams $S_1$ and $S_2$ were obtained by reading out the grating in opposite direction and thus their Doppler shift has the same size but the opposite sign. Assuming that the time delay $\Delta t$ between the two signal beams is much smaller than the oscillation period $T_g$ of the grating scattering efficiency, i.e. $\Delta t<<T_g$, one can deduce for the resulting signal impinging on the detector [Hem 2000]:

$$P_s(t) \approx [1 + m \cos(\Omega \Delta t + \psi)]\sin^2(\omega't)e^{(-2\beta t)}$$

(39)
Where $\Omega_M = 2qv_f = 2\frac{v_f}{v_{S_0}}\omega'_0$ is the modulation frequency of the signal due to the flow,

$$m = 2\frac{A_{si}^2 - A_{s2}^2}{(A_{si}^2 + A_{s2}^2)}$$

is the modulation coefficient of the beating, $A_{Si}$ is the amplitude of signal $S_i$, $\psi$ is a phase factor which takes into account the effective shift experienced by signal $S_1$ and read-out beam $R_2$ traveling on different optical paths to the back-reflecting mirror $M$. Subscript 0 refers to the reference condition at room temperature.

Measurements are carried out for air temperatures ranging from 295 K to 600 K and various flow rates $Q$ between 10 and 200 l/min. The average flow velocity $v_Q$ at the exit of the nozzle can be calculated from the flow rates using the equation of continuity.

A frame of reference is defined so that the origin of the laboratory fixed Cartesian coordinate system is at the focus of both excitation beams. The x-y-plane is defined by the two excitation beams, and the y-z-plane bisects their intersection angle. The nozzle was adjusted with its center coinciding with the x-axis, and its long side was aligned to be parallel to the y-axis at a distance of 1 mm (i.e. $x=-1.0$ mm), see Figure 33.

**Figure 33:** Slot nozzle used for flow velocity measurements by laser-induced electrostrictive gratings. It was manufactured by squeezing one end of a copper tube, (dimensions are in mm).

The upper trace in Figure 34a is the temporal evolution of a signal obtained in air at room temperature ($T_0=295K$) without flow. Each measured data point is averaged over 30 laser shots. The difference between measurements and best fit from Eq. (39) is plotted with an offset for clarity (lower trace). From the data in Figure 34a an angular frequency for the acoustic wave $\omega'_0 = 93.3 \pm 0.3$ MHz is obtained. Taking into account the sound velocity in air...
[Ency 1976] for the actual pressure and temperature and the determined acoustic oscillation period one can calculate the intersection angle of the two excitation beams $\theta = 2.63 \pm 0.01^\circ$, which corresponds to a fringe spacing $\Lambda$ of $23.19 \pm 0.07 \mu m$. The temporal evolution of the signal obtained in presence of a heated air flow is shown in Figure 34b (upper trace). The low-frequency oscillation due to the beating of the two signal contributions of opposite Doppler-shifts is clearly visible. Again the lower trace of Figure 34b represents the residual of measurement and a best fit from Eq. (35) to the data points.

Figure 34: Temporal evolution of the diffraction efficiency of laser-induced electrostrictive gratings at room temperature without flow (a) and at $T_{th}=380$ K and a flow rate of 80 l/min, (b), respectively. For both cases the bottom trace shows the residual of the measurement and a best fit from Eq. 39.

The flow temperature value of $T_{th}=380$ K was measured by a thermocouple and the averaged flow velocity of $v_0 = 52.8$ m/s has been calculated from the measured flow rate $Q=80$ l/min.
and $T_d$. From the best fit the angular frequencies for the acoustic wave of $\omega' = 107.3 \pm 0.3$ MHz and for the beating of $\Omega_M = 49 \pm 0.3$ MHz are deduced. Assuming that the air composition and the specific heat ratio $\gamma$ remain unchanged, a temperature $T = 388 \pm 3$ K can be calculated from the measurement of $\omega'$. The velocity of the flow $v_f = 89.8 \pm 0.6$ m/s can be obtained from the measured angular frequency $\Omega_M$.

Systematic measurements for various air flow rates in the range $Q = 20$ to 200 l/min show a linear relationship between the flow velocity $v_f$ determined from the beat angular frequency of the signal and the average flow velocity $v_Q$, $v_f = 1.5 * v_Q$. The measured flow velocity is higher than the average flow velocity because of the parabolic shaped velocity profile of the nozzle: high velocity of the flow in the center and almost no flow at the border. Thus measuring in the center of the nozzle a higher flow velocity is obtained.

Simultaneous measurements of flow velocity and temperature are carried out in an air flow at room temperature. From the data the averaged flow velocity determined from the flow rate was $v_Q = 41.1$ m/s. In Figure 35, about 300 temperatures and flow velocities are compiled in two normalized histograms with a bin width of 0.5 K and 1 m/s, respectively. A correlation coefficient $>0.99$ indicates that a Gaussian (solid line) describes the measured distribution quite well. The obtained average flow velocity was $59.0 \pm 2.5$ m/s and the average value for the temperature $296.7 \pm 1.8$ K. The spatial resolution of the beam geometry is 0.2 mm in z-direction 10 mm in y-direction. Therefore, the measurement volume fits into the nozzle cross section. However, the fairly low spatial resolution in y-direction (here ca 10 mm) results in a spatially averaged value of the extracted flow velocity. To increase the spatial resolution of the laser-induced electrostrictive grating method while keeping a high accuracy for the determined flow velocity the angle between the excitation beams has to be increased. This results in a large grating vector and therefore in a shorter lifetime of the grating, since the decay time of the diffraction efficiency of the grating is inversely proportional to the square of grating vector, see Eq. (18).
Figure 35: Normalized histogram of 300 single-shots simultaneous measurements of
temperature (a) and of flow velocity (b) in a submerged air jet at room temperature
(T=295 K) and a flow rate of 80 l/min.

Also, high temperature and turbulence shorten the grating lifetime, which poses a lower limit
to the measurable flow velocity by this method. Therefore, velocimetry by laser-induced
electrostrictive gratings will be applied with more advantage for flows exhibiting relatively
high density, low degree of turbulence and high speed. The high spatial resolution of the
laser induced electrostrictive grating technique in the z direction, roughly 0.2 mm, has been
used to measure the jet velocity profile across the nozzle, see Figure 36.
The measurements have been performed with a step of 0.1 mm at a distance x=1mm from the nozzle exit. The measured velocity distribution is nearly flat in proximity of the flow axis and varies noticeably when approaching the boundary regions of the jet. Averaging of the velocities over z within the range \(-0.75 \text{ mm} \leq z \leq 0.75 \text{ mm}\) yields \(v_Q=33.5 \text{ m/s}\), which agrees fairly well with the value \(v_Q=32.7 \text{ m/s}\) estimated from the measurements.

### 3.3.4 Imaging by laser-induced gratings

One way to map the combustion field is achieved by translating the measurement volume point by point over the region of interest. It is surely a demanding, though an acceptable procedure. Of course it is more convenient and sometime even necessary (if e.g. the combustion process under investigation is turbulent), to perform a measurement over the entire region of interest simultaneously. Imaging techniques can be incoherent, as e.g. laser induced fluorescence, or coherent as Degenerate four wave mixing. In this chapter the imaging potential of laser-induced gratings is demonstrated for the first time to the author’s knowledge, and the advantages of such approach are discussed.

Figure 37 displays the laser induced gratings setup used for imaging applications which is very similar to that one of Ref. [Rak 1990].
Figure 37: Experimental setup for imaging applications by laser induced gratings. BS1, BS2 beamsplitter (R=30%, 50%, respectively); P1, P2, polarizer; CT, cylinder telescope; HR high reflectivity mirror; CCD camera system.

The frequency doubled output of an injection seeded Nd: YAG (Continuum NY 81-20) laser running single-mode is used to generate all three beams. The IR spectral bandwidth of the Nd: YAG laser without any narrowing elements is 1 cm\(^{-1}\). Using the single-mode injection seeder reduces the bandwidth to 0.0045 cm\(^{-1}\) [Con 1990]. The resulting coherence length of 2 m allows the coherent superposition of the excitation beams without adjustment for equal optical paths length, which would be otherwise impossible in this setup. By switching the Nd: YAG laser from multi-mode to single-mode operation the electrostrictive signal intensity was found to increase by a factor of 180. The output of the laser is split by a 30% reflectivity beamsplitter BS1 into two excitation beams. The weaker excitation beam, referred in the following to as EB1, is directed into the interaction region by a 50% beamsplitter, where it crosses the stronger excitation beam, referred in the following to as EB2, at an angle $\theta$ of about 70 degrees. EB2 is formed into a light sheet about 500 $\mu$m thick and 6 mm high by using a Galilean telescope consisting of two cylindrical lenses. The probe beam is obtained by back reflection of the beam EB2. The back reflecting mirror HR is positioned in the focus of the telescope. For damage free operation the usable pulse energy is limited to 10 mJ. The stronger excitation beam passes a polarizer P1, which forms in combination with a $\lambda/4$ plate at the end of the beam path, an optical diode protecting the laser from the back reflected light that is used as probe beam. Passing the $\lambda/4$ plate twice, the back traveling wave, i.e. probe
beam, is perpendicularly polarized to the excitation beams. Optical dumps are used to block the weaker excitation beam after passing the interaction region and that part of it which passes through BS2. The two equally polarized excitation beams build up the grating in the medium under investigation and the probe beam is diffracted off the grating. Satisfying the Bragg condition, the signal beam is counter-propagating to the weaker excitation beam. Part of the signal passes beamsplitter BS2 and is detected by an intensified CCD camera (PI CSMA 130). The signal beam has the same polarization as the probe beam and it is thus perpendicularly polarized to the excitation beams. Therefore, stray light mainly arising from beamsplitter BS2, is efficiently suppressed by a polarizer P2 in the path of the signal beam. A diffraction efficiency $\eta=(1.5\pm0.2)\times10^{-9}$ is measured for the electrostrictive laser-induced grating generated in ambient air with a laser intensity of 90 MW/cm$^2$. Thus the intensity of the signal beam is large enough to be detected by an intensified CCD camera. Figure 38 shows a single shot image obtained in ambient air. Because the grating is formed in an isotropic medium no structure should be visible. The observable structure arises from in-homogeneities of the intensity profile of the Nd: YAG laser. Beside this coarse structure, which is caused by poor alignment of the laser, there are local shot to shot intensity fluctuations in the signal beam. By comparing several images subsequently recorded in air, the size of these intensity fluctuations is estimated to be about $\pm10\%$.

![Figure 38](image.png)

Figure 38: Single shot measurement of ambient air by laser induced electrostrictive gratings. The signal intensity variations present in this image arise from the inhomogeneities of the intensity profile of the laser beam. The diameter of the displayed spot is 6 mm, approximately.
The image is generated in the overlap region of a circular shaped excitation beam and the laser sheet formed by the probe beam and the other excitation beam. Therefore, the viewed area is an oval. With the CCD camera normal to the propagation direction of the circular excitation beam, the viewed area is compressed to a circular image. The compression factor is $\sin\theta$ for the axis lying in the plane stretched by the propagation directions of the two excitation beams. The distortion caused by this foreshortening has not been accounted for in this work. For better spatial resolution and less distortion of the image the use of relatively large crossing angles is therefore advantageous for imaging by this technique. Ideally, to avoid any distortion of the obtained image, the crossing angle $\theta$ of the excitation beams should be equal to 90°. This would lead though, to a very small grating spacing and to a correspondingly short lifetime of the grating (i.e., low signal intensity).

The electrostrictive signal obtained in helium is smaller by a factor of 66 compared with the one generated in air. Therefore, the signal obtained in a mixture of air and helium is dominated by the signal resulting from air. Figure 39 depicts a single-shot image of a helium flow in air. In the false color representation the areas of high helium concentration are displayed bright, while the atmospheric air regions are dark. Since the laser intensity is not homogeneous across the beam profile, the image has been corrected by dividing by a reference image obtained in air. Of course this correction will not account for the shot to shot laser intensity fluctuations since image and reference image result from two different shots.

The intensity values of the resulting image lie between zero and unity, corresponding to an air partial pressure lower than 30 mbar and 1 bar, respectively. The local fluctuations of the electrostrictive signal intensity caused by shot-to-shot fluctuation of the laser intensity correspond to an error of about 7% in the determination of the air partial pressure. Note that no care has been taken to establish a laminar helium flow. The observed structures may be caused by the turbulent streaming of helium as well as by the movements of the surrounding air.
Figure 39: Single-shot image of a helium jet in air by laser-induced electrostrictive gratings. The diameter of the nozzle is about 0.1 mm, and the helium pressure difference 0.1 bar. The areas of high helium concentration are displayed bright. The color scale is given in arbitrary units. The sketch on the right side represent the position of the nozzle and of the helium flow.

In addition, the technique has been applied to image sooty flames. For this purpose an acetylene/air diffusion flame is used. The fuel-air mixture fraction in the flame is set just below the point at which smoke would have started to be produced. Soot particles and soot precursors absorb widely across the electromagnetic spectrum. It is thus possible to generate very efficiently thermal gratings in sooty flames with almost any pulsed laser system. These gratings can be used to map soot particle concentrations, as it is proposed here, or to measure the flame temperature [Bro 1999]. The extinction losses of the incident laser radiation in the flame due to absorption or scattering is measured to be about 20 % over the intensity range from 15 MW/cm$^2$ to 90 MW/cm$^2$. Figure 40 shows a double logarithmic plot of the signal generated in the sooty flame versus the total laser intensity. The data are fitted by a linear regression with a slope of 4.37±0.1 (solid triangles). Furthermore a linear dependence of the signal generated in a sooty flame upon the probe beam intensity is measured (open squares in Figure 40). Soot particles in the flame absorb the incident laser radiation and heat to temperatures far above ambient. There are two mechanisms by which the soot particles lose their heat from the absorbed laser energy. Below 3300 K heat conduction is the dominant
loss process, while above 3300 K vaporization becomes the dominant loss process. Therefore, at high temperatures the absorbed laser energy is balanced by vaporization that causes shrinking of the soot particles.

The vapor streaming away from the soot particle consists mainly of $C_3$, $C_2$ and $C$. Since the wavelength of the probe beam at 532 nm is very close to a transition in $C_2$ $(d^3\Pi_g \leftarrow a^3\Pi_u, v' = 0, v'' = 0)$, the observed signal is comprised of a signal due to the solid soot particles and a signal generated in the $C_2$ vapor. The high order ($\geq 3$) signal dependence upon the excitation beams results from this complex signal formation process [Hem 2001].

The regions of high soot concentration in a laminar acetylene/air diffusion flame can easily be recognized by eye due to a bright, yellow emission. The signal formed in these areas is strong enough to be readily observed by the naked eye when directed on a piece of paper. For the grating generated in the sooty flame a diffraction efficiency of $\eta=(3.0\pm1.0)\times 10^{-4}$ was measured at a laser output intensity of 90 MW/cm$^2$. Figure 41 shows a single shot image of an axis symmetric acetylene/air diffusion flame. This image was also corrected by a reference image recorded in air. The regions with high particle concentration are displayed bright. The observed distribution reflects the shape of the fuel/air reaction zone. No attempt was made for a quantitative interpretation of this image.
Figure 41: Single-shot image of an axisymmetric acetylene/air diffusion flame. The flame is about 8 mm high and 6 mm wide. The color scale reflecting the particle concentration is given in arbitrary units.

Note that on the left side of the image the signal intensity is higher than on the right side since the 20% of probe beam is absorbed passing the flame.
4 Summary

One of the aims of this study is to understand the formation mechanisms of transient laser induced electrostrictive and thermal gratings generated in the gas phase. Laser-induced gratings arise from two crossed laser beams (excitation beams) in a medium. In their interference region the electric field envelope interacts with the matter leading to a spatial modulation of the optical properties and thus of the refractive index. This grating like structure can be detected by diffracting a third beam off the laser induced refractive index modulation. The characteristics of the diffraction efficiency of such a gratings depend on the mechanisms responsible for the grating formation. The calculation and the experiments presented in this work showed the following findings. Electrostriction generates mainly a standing acoustic wave in addition to a small amplitude stationary density wave. The density is modulated below and above its undisturbed value and the electrostrictive grating reflectivity, which is proportional to the squared density change, oscillates at twice the acoustic period. Release of the absorbed laser radiation in form of heat through collisions leads to the formation of thermal gratings. The time scale of the heat release process governs the characteristics of the temporal behavior of the induced thermal grating. Instantaneous heat release forms a standing acoustic wave and a stationary density modulation of equal amplitude. In this case, depending on the location, the density is modulated only below, or above its equilibrium value through which it passes once each acoustic period. The diffraction efficiency of a thermal grating, which is proportional to the squared density variation, displays an oscillation at the acoustic frequency. When the absorbed radiation energy is slowly released, the pressure rise is continuously drained by acoustic waves. The process is nearly isobaric and favors the formation of a stationary density modulation, whereas the growth of the standing acoustic wave is hindered by the destructive interference of the sound waves generated at different times. The standing acoustic wave is damped by viscosity and heat conduction while the stationary density wave decays by heat conduction alone.

In the experimental part the influence of various parameters on the generated electrostrictive or thermal grating such as beam size, reading out of the grating by a pulsed or cw probe beam, pressure, laser intensity, the coherence length of the excitation laser have been measured. These data provide on one side a test to the theoretical model and on the other side they consent to design the experimental configurations presented in this work. In addition the
possibility to apply laser-induced grating to investigate optical breakdown in gases has been demonstrated qualitatively.

The main goal of this work is to test the potential of electrostrictive and thermal laser induced transient grating as a diagnostic tool in the gas phase. In fact, the method offers a unique possibility to measure kinetic and thermodynamic properties of gases. The adiabatic sound velocity can be easily inferred from the frequency analysis of the oscillating part of the grating diffraction efficiency. By known composition of the medium the temperature can be deduced from the measured sound velocity. Due to their non-resonant nature electrostrictive gratings can be employed to determine temperatures in any gas in a single shot measurement.

The experimental setup for laser induced electrostrictive and thermal gratings is quite simple compared to other thermometry methods. Consequently, both techniques have a potential to become a valid alternative to classical laser based techniques for the measurement of temperature, especially in high-pressure environments where the damping of sound is low. One has to be aware, however, that the measured sound velocity and therefore the temperature, relies on the measurement of the intersection angle of the two excitation beams. Usually, this is accomplished by a reference measurement. In turbulent environments, however, shot-to-shot variations of the crossing angle can occur due to beam steering effects.

The capability of electrostrictive laser-induced gratings to measure the temperature of air from ambient up to 1400 K in an open furnace and of a stoichiometric CO/O\textsubscript{2} flame has been demonstrated. The obtained temperatures agree with thermocouple measurements within the estimated error bars. Thermal gratings have been used to measure temperatures in stoichiometric methane/air flames in the pressure range between 1 and 25 bar. CARS measurements have been performed simultaneously to validate laser-induced thermal grating thermometry. Up to 10 bar good agreement between the temperatures measured by CARS and by laser-induced gratings is obtained. The discrepancy occurring at higher pressures is probably due to beam-steering effects. Such issue should be the subject of future investigations. Systematic measurements of temperatures by laser-induced thermal grating and CARS are required in order to promote the laser-induced grating technique as a valid alternative to other established methods for the thermometry.

Furthermore, through the measurement of the sound velocity, laser-induced electrostrictive gratings have been applied to monitor the variation in concentration of binary gas mixtures. The advantage of a relative undemanding experimental setup is balanced by the drawback of the loss in species selectivity. The sensitivity is given by the ratio of the sound velocities of the mixture components. Furthermore the method has been applied to image a helium flow in
air without the need of particle seeding the flow as it is necessary in established non spectroscopic imaging techniques.

One of the most appealing features of this technique is to allow for the simultaneous, instantaneous, non intrusive measurement of temperature and flow velocities. This might be useful for the investigation of wind tunnels or in the premixing zone of large scale burners. In this work the technique has been used to measure simultaneously temperature in the range 295-600 K and flow velocity in the range 10-100 m/s in a submerged air jet. From the experiments described here one can conclude that the electrostrictive laser-induced grating technique is particularly suited for studying high speed flow of relatively high density, relatively low temperature, low degree of turbulence. The lower limit of the measurable flow velocities is due to the relatively short decay time of the laser-induced electrostrictive gratings at atmospheric pressure. In a future work the possibility to overcome this problem by the application of thermal gratings for velocimetry should be investigated.

Finally the imaging potential of the laser induced grating technique has been investigated. Using a planar backward geometry to produce the image a circular pump beam intersects the other pump beam and the probe beam which have been both shaped into a thin light sheet. Single shot images of a helium flow submerged in ambient air by electrostrictive gratings have been recorded showing that this technique can be applied for the remote, 2-dimensional diagnostics of gas mixing processes without the need of seeding the region of interest. In addition thermal gratings have been applied to map the particle concentration in a sooty flame. The oxidant-fuel reaction zone of is readily visualized by a single shot measurement by this technique. However in order to perform quantitative measurements, the mechanisms of interaction between the soot present in the flame and the strong laser radiation need to be investigated in a future work.
5 Appendix

5.1 Determination of the induced density and temperature variation

From the linearized hydrodynamic equations the laser induced variations of the density $\rho'$ and temperature $T'$ can be calculated [Boy 1992]:

$$
-\frac{\partial^2 \rho'}{\partial t^2} + \frac{\nu}{\gamma} \nabla^2 \rho' + \frac{\nu^2}{\rho_0} \frac{\partial}{\partial t} \left( \nabla^2 \rho' \right) + \frac{n_{\text{nucl}}}{\rho_0} \frac{\partial}{\partial t} \left( \nabla^2 \rho' \right) = \frac{\varepsilon_0 \nu}{\gamma} \nabla^2 \varepsilon^2 \tau (e^{i\omega t} - e^{-i\omega t}) \delta(t - t_0)
$$

(A1)

$$
\rho_0 c_v \frac{\partial T'}{\partial t} - \frac{c_v (\gamma - 1)}{\beta_p} \frac{\partial \rho'}{\partial t} - \kappa \nabla^2 T' = \varepsilon_0 n_{\text{nucl}} \varepsilon^2 (e^{i\omega t} - e^{-i\omega t}) s(t).
$$

Both source terms have the same spatial dependency, while the temporal dependence of the source term of the energy transport equation is described by the function $s(t)$, which will depend on the nature of the conversion process of the absorbed laser energy into heat.

Assuming instantaneous heat release $s(t)$ is equal to the product $\tau$ times a $\delta(t-t_0)$ function, which can be written as a Fourier integral

$$
\delta(t - t_0) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} e^{-i\omega(t-t_0)} d\omega
$$

(A2)

Since both source terms in system (A1) can be decomposed in a product of an exponential function of time and a exponential function of the spatial coordinate $x$, and the operators $\left( \frac{\partial}{\partial t} \right)^n$, $n=1,2$ and $\nabla^2$ map these functions into multiples of themselves, the “Lösungsansatz” for Eqs. (A1) will be of the form

$$
\rho'(x,t) = c_1 e^{i\omega t} e^{i\beta x} + c_2 e^{-i\omega t} e^{-i\beta x}
$$

(A3)

A standard way of solving a system of partial differential equations of third order as (A1) is the so called operator method, see e.g. [Bron 1987]. This method consists in transforming a differential equations system into an equations system by applying an integral transformation. The transformed equations system is in general straightforward to solve. Obviously the hurdle of finding the solution of the differential equations system is not simply removed, yet it appears when carrying out the back-transformation. Here the Fourier transformation of the time dependent part of the induced density and temperature is used and their spatial dependency is expressed in the real form.
\[ \rho'(x, t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \int \tilde{\rho}(\omega) e^{-i\omega(t-t_0)} d\omega \]  

\[ T'(x, t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \int \tilde{T}(\omega) e^{-i\omega(t-t_0)} d\omega \]  

(A4)

Using the variable \( t' = t - t_0 \) and inserting (A4) and (A2) into system (A1) gives

\[
\begin{align*}
\frac{1}{2\pi} \int \left[ -\omega^2 \tilde{\rho}(\omega) + \frac{\nu^2}{\gamma} q^2 \tilde{\rho}(\omega) + \frac{\nu^2 \beta \nu}{\gamma} q^2 \tilde{T}(\omega) - \frac{\eta \omega}{\rho} i \omega q^2 \tilde{\rho}(\omega) - \frac{\epsilon_0}{2} \gamma \varepsilon^2 q^2 \right] e^{-i\omega t'} 2\cos \omega x d\omega = 0
\end{align*}
\]

(A5)

Both equations are fulfilled for every \( \omega \), thus the expressions in the squared brackets of both Eqs. in (A5) must be equal to 0 and the following equations system is obtained

\[
\begin{align*}
-\omega^2 \tilde{\rho}(\omega) + \frac{\nu^2}{\gamma} q^2 \tilde{\rho}(\omega) + \frac{\nu^2 \beta \nu}{\gamma} q^2 \tilde{T}(\omega) - \frac{\eta \omega}{\rho} i \omega q^2 \tilde{\rho}(\omega) - \frac{\epsilon_0}{2} \gamma \varepsilon^2 q^2 &= 0 \\
-i \alpha \rho_0 \tilde{T}(\omega) + \frac{i \omega c_0 (\gamma - 1)}{\beta_p} \tilde{\rho}(\omega) + \kappa_\nu^2 \tilde{T}(\omega) - \epsilon_0 n c \alpha^2 \varepsilon^2 &= 0
\end{align*}
\]

(A6)

Solving (A5) for \( \tilde{\rho}(\omega) \) and \( \tilde{T}(\omega) \) gives

\[
\tilde{\rho}(\omega) = -\frac{\epsilon_0}{2} E^2 \tau_1 \frac{\gamma c \omega - i \gamma c \frac{\nu}{2} \Gamma_b + i \gamma c q^2 v^2}{\omega^3 + i(\Gamma_b + \frac{\nu}{2} \Gamma_b) \omega^2 - (q^2 v^2 + \frac{\nu}{2} \Gamma_b) \omega - \frac{i}{2} \Gamma_b q^2 v^2}
\]

(A7)

\[
\tilde{T}(\omega) = -\frac{\epsilon_0}{2} E^2 \tau_1 \frac{\gamma c \omega^2 - i \left[ \left( \frac{\nu}{2} \right) \Gamma_b + \frac{\nu}{2} \Gamma_b \right] \omega - \gamma c q^2 v^2 \xi}{\omega^3 + i(\Gamma_b + \frac{\nu}{2} \Gamma_b) \omega^2 - (q^2 v^2 + \frac{\nu}{2} \Gamma_b) \omega - \frac{i}{2} \Gamma_b q^2 v^2}
\]

where the Brillouin line width \( \Gamma_b = \frac{\eta_\nu q^2}{\rho_0} \), the Rayleigh line width \( \Gamma_R = \frac{2q^2}{\rho_0 c_p} \) and the absorptive coupling constant \( \gamma_a = \frac{2n c \alpha \beta}{c_\nu} \) have been introduced, see e.g. [Boy 1992]. Since for the experiments dealt with in this work the change of the refractive index caused by
temperature variation is small compared to that one caused by density variation, only the latter effect will be calculated in the following, i.e.:

\[
\rho'(t) = \frac{1}{2\pi} \left( \frac{1}{2} e_o E^2 \pi^2 \right) \int_{-\infty}^{\infty} \frac{\gamma_\epsilon \omega - i \gamma_\epsilon \frac{Z}{2} \Gamma_g + i \gamma_\epsilon \xi q \nu_\epsilon}{\omega^3 + i(\Gamma_g + \frac{Z}{2} \Gamma_g)\omega^2 - (\xi^2 q^2 \nu_\epsilon^2 + \frac{Z}{2} \Gamma_g \Gamma_g)\omega - \frac{Z}{2} \Gamma_g q^2 \nu_\epsilon^2} e^{-i\omega t} d\omega
\]  

(A8)

The denominator of (A8) is a third order polynomial in \( \omega \) that can be decomposed into \((\omega - \omega_1)(\omega - \omega_2)(\omega - \omega_3)\). Since its quadratic term and its constant term are pure imaginary, while the two odd terms in \( \omega \) are real, the roots of the polynomial will be of the form

\[
\begin{align*}
\omega_1 &= \omega' - i\beta_1 \\
\omega_2 &= -\omega' - i\beta_1 \\
\omega_3 &= -i\beta_3
\end{align*}
\]  

(A9)

The exponential term in (A8) diverges when both the imaginary part of \( \omega \) and \((t-t_0)\) are positive or when the imaginary part of \( \omega \) and \((t-t_0)\) are both negative, thus (A8) is evaluated by integrating over a circle of radius \( R \) in the upper half complex plane of \( \omega \) for \((t-t_0)<0\) and over a circle of radius \( R \) in the lower half complex plane of \( \omega \) for \((t-t_0)>0\).

\[
\rho(t) = \lim_{R \to \infty} \int_{-R}^{+R} \tilde{\rho}(\omega) e^{-i\omega t} d\omega = 2\pi \sum \text{residues in the circle}
\]

(A10)

Expressing \( \tilde{\rho}(\omega) \) as follow

\[
\tilde{\rho}(\omega) = \prod_{k=1,2,3} \frac{Z(\omega)}{(\omega - \omega_k)}
\]

(A11),

one can rewrite (A10)

\[
\rho(t) = \sum_{k=1}^{3} \prod_{i \neq k} \frac{Z(\omega_k)}{(\omega_k - \omega_k)}
\]

(A12)

Because all poles of the function \( \tilde{\rho}(\omega) \) lie in the lower plane, the integral (A8) is proportional to the heavyside function \( \theta(t-t_0) \). Evaluation of (A8) in the lower half complex plane yields

\[
\rho'(t) = \frac{e_o E^2 \pi^2}{2} \frac{\theta(t-t_0)}{(\omega')^2 + (\beta_1 - \beta_2)^2} (P_{\text{elec}} + P_{\text{throm}})
\]

(A13)

\[
P_{\text{elec}} = \gamma_\epsilon e^{-\beta(t-t_0)} \left[ \frac{\omega' + (\beta_1 - \beta_3)(\beta_1 - \frac{Z}{2} \Gamma_g)}{\omega'} \sin \omega'(t-t_0) - \frac{Z}{2} \Gamma_g - \beta_1 \right] \cos \omega'(t-t_0) - e^{-(\beta_1 - \beta_3)(t-t_0)}
\]

(A14)
\[
P_{\text{therm}} = \gamma_0 q v \left\{ e^{-\beta(t-t_0)} \left[ \frac{(\beta_1 - \beta_2)}{\omega} \sin \omega'(t-t_0) + \cos \omega'(t-t_0) \right] - e^{-\beta(t-t_0)} \right\}.
\]  

(A15)

Adding together the sine and cosine terms (A14) and (A15) transform into

\[
P_{\text{elec}} = \gamma_e \left\{ e^{-\beta(t-t_0)} L' \sin[\omega'(t-t_0) + \varphi'] - \left[ \frac{\gamma_R - \beta_2}{2} \right] e^{-\beta(t-t_0)} \right\}
\]

(A14')

\[
P_{\text{therm}} = \gamma_0 q v \left\{ e^{-\beta(t-t_0)} L^* \sin[\omega'(t-t_0) + \varphi^*] - e^{-\beta(t-t_0)} \right\}
\]

(A15')

with

\[
L' = \sqrt{\left( \frac{\gamma_R - \beta_2}{2} \right)^2 + \left[ \frac{(\beta_1 - \beta_2)}{\omega} \left( \frac{\gamma_R - \beta_2}{2} \right) \right]^2},
\]

\[
\varphi' = \arctan \left( \frac{\frac{\gamma_R - \beta_2}{2}}{1 + \frac{(\beta_1 - \beta_2)}{(\omega')^2}} \right)
\]

(A16)

\[
L^* = \sqrt{\left( \frac{(\beta_1 - \beta_2)}{(\omega')^2} \right)^2 + 1},
\]

\[
\varphi^* = \arctan \left( \frac{\omega'}{(\beta_1 - \beta_2)} \right)
\]

To calculate the parameters \( \omega', \beta_1 \) and \( \beta_2 \) in (A9) the following equation must be solved:

\[
\omega^3 + i(\Gamma_\gamma + \Gamma_R)\omega' - (q^2v^2 + \frac{\gamma_R}{2})\omega - \frac{i}{2} \Gamma_R q^2v^2 = 0
\]

(A17)

Approximate solutions of equation (A17) can be found by the following procedure: since the product \( q^2v^2 \gg \Gamma_\gamma \Gamma_R \) one approximate solution is obtained by solving

\[
\omega + \frac{i}{2} \Gamma_\gamma + O\left( \frac{\Gamma_R}{q^2v^2} \right) = 0
\]

(A18)

i.e.

\[
\omega = -\frac{i}{2} \Gamma_\gamma
\]

(A19)

Now Eq. (A17) can be expressed as
\[
\left(\omega + \frac{i}{2} \Gamma_R\right)\left(\omega^2 + a\omega + b\right) + O\left(\frac{\Gamma_{s,s}}{q_i^2\nu_i^2}\right) = 0
\]  
(A17')

Neglecting the terms of order \(\frac{\Gamma_{s,s}}{q_i^2\nu_i^2}\) and by performing a polynomial division the following expression results

\[
\left(\omega + \frac{i}{2} \Gamma_R\right)\left(\omega^2 + i\omega - \left(q_i^2\nu_i^2 + \frac{\Gamma_{s,s}}{2}(\Gamma_R - \Gamma)\right) + O\left(\frac{\Gamma_{s,s}}{q_i^2\nu_i^2}\right) = 0
\]  
(A20)

with

\[
\Gamma = \Gamma_R + \frac{\gamma - 1}{2} \Gamma_R
\]  
(A21)

The quadratic polynomial in \(\omega\) in (A20) has the following roots

\[
\omega_{1,2} = -\frac{i}{2} \Gamma \pm \sqrt{\Omega^2\nu_i^2\left[1 + O\left(\frac{\Gamma_{s,s}}{q_i^2\nu_i^2}\right)\right]}
\]  
(A22)

Comparing (A9) with the above obtained approximate solutions \(\omega_{1,2,3}\) yields

\[
\omega' = q\nu_s
\]

\[
\beta_1 = \Gamma_R + \frac{\gamma - 1}{2} \Gamma_R = \frac{1}{2} \rho_0 \left(\eta_m + \frac{(\gamma - 1)\kappa}{c_p}\right)
\]

\[
\beta_2 = \frac{\Gamma_R}{2}
\]

Dropping all nonlinear terms in \(\omega' / \beta_1\) the Eqs. (A14) and (A15) simplify into

\[
\rho'(t) = \frac{\rho'^2}{2} \theta(t-t_0)\left(P_{\text{elec}} + P_{\text{therm}}\right)
\]

\[
P_{\text{elec}}(t) = \gamma e^{-\beta_1(t-t_0)} \left\{ \sin\omega'(t-t_0) - \left[\frac{(\gamma - 1)\beta_2}{\omega'}\right] \left[\cos\omega'(t-t_0) - e^{-\beta_1\omega'(t-t_0)}\right] \right\}
\]

\[
P_{\text{therm}}(t) = \gamma e^{-\beta_2(t-t_0)} \left\{ e^{-\beta_1(t-t_0)} \left[\frac{(\beta_1 - \beta_2)}{\omega'}\right] \left[\sin\omega'(t-t_0) + \cos\omega'(t-t_0) - e^{-\beta_2(t-t_0)}\right] \right\}
\]

Inserting \(\omega' = q\nu_s\) into (A15') and adding the sinus and cosinus terms together the Equation (24) of the theory section is obtained.

Consider now the situation where the absorbed light energy is converted into heat no longer instantaneously and the energy conversion process that takes place is characterized by a single step relaxation with relaxation rate \(\sigma\). In this case the function \(s(t)\) in the source term of the energy transport equation can be written as
with $\sigma$ as defined in the theory section. Eq. system (A1) becomes

\[-\frac{\partial^2 \rho'}{\partial t^2} + \frac{\nu}{\gamma} \nabla^2 \rho' + \frac{v_0^3 \beta \rho_0}{\gamma} \nabla^2 T' + \frac{\eta_{\text{isc}}}{\rho_0} \frac{\partial}{\partial t} \left( \nabla^2 \rho' \right) = \frac{\varepsilon_0 \gamma_r E^2 \tau}{2} \nabla^2 (e^{i\omega_t} - e^{-i\omega_t}) \delta(t - t_0)\]

\[\rho_0^\prime \frac{\partial T'}{\partial t} - \frac{c_v (\gamma - 1)}{\rho_0^\prime} \frac{\partial \rho'}{\partial t} - \kappa \nabla^2 T' = \varepsilon_{\text{inc}} \beta \alpha \tau \varepsilon^2 (e^{i\omega_t} - e^{-i\omega_t}) \theta(t - t_0) e^{-\sigma(t-t_0)}\]

Expressing the time dependent part of the source term of the energy transport equation as the following Fourier integral

\[\theta(t-t_0) e^{-\sigma(t-t_0)} = \frac{1}{2 \pi} \int_{-\infty}^{+\infty} e^{-i\omega(t-t_0)} d\omega\]  

(A25)

and using the “Lösungsansatz”

\[\rho'(x,t) = 2 \cos qx \frac{1}{2 \pi} \int_{-\infty}^{+\infty} \tilde{\rho}(\omega) e^{-i\omega(t-t_0)} d\omega\]  

\[T'(x,t) = 2 \cos qx \frac{1}{2 \pi} \int_{-\infty}^{+\infty} \tilde{T}(\omega) e^{-i\omega(t-t_0)} d\omega\]  

(A26)

to solve (A1'), the following equations system is obtained

\[-\frac{\omega^2 \tilde{\rho}(\omega)}{(\sigma - i\omega)} + \frac{v_0^2}{\gamma (\sigma - i\omega)} \tilde{\rho}(\omega) + \frac{v_0^3 \beta \rho_0}{\gamma (\sigma - i\omega)} \tilde{T}(\omega) - \frac{\eta_{\text{isc}}}{\rho_0^\prime (\sigma - i\omega)} i\omega t \tilde{\rho}(\omega) - \frac{\varepsilon_0}{\gamma_r E^2 \pi_1^2} = 0\]

\[-i \omega t \tilde{\rho}(\omega) + \frac{i \omega t (\gamma - 1)}{\rho_0^\prime} \tilde{\rho}(\omega) + \kappa \tilde{T}(\omega) - \varepsilon_{\text{inc}} \alpha \tau \varepsilon^2 = 0\]  

(A27)

Solving for $\tilde{\rho}(\omega)$ yields

\[\tilde{\rho}(\omega) = -\frac{1}{2} \gamma_r E^2 \pi_1^2 \left( \frac{\gamma_r t^2}{(\sigma + \frac{\gamma_r}{2} \Gamma_k \omega - \gamma_r \frac{2 \Gamma_r}{2} + \gamma_r \frac{2 \Gamma_k}{2} \Gamma_r \omega) - \frac{i}{2} \Gamma_k \omega^2 - \frac{i}{2} \Gamma_k \omega^2} \right)\]  

(A28)

and finally

\[\rho'(t) = \frac{1}{2 \pi} \left( \frac{-i \left( \gamma_r t^2 + i \gamma_r \left( \frac{\sigma + \frac{\gamma_r}{2} \Gamma_k \omega - \gamma_r \frac{2 \Gamma_r}{2} + \gamma_r \frac{2 \Gamma_k}{2} \Gamma_r \omega - \frac{i}{2} \Gamma_k \omega^2} \right) e^{-i\omega t}}{\left( \sigma - i\omega \left( \omega^2 + i(\Gamma_b + \frac{\gamma_r}{2} \Gamma_k) \omega^2 - (q_1^2 \omega_1^2 + \frac{i}{2} \Gamma_k \omega^2) \right) - \frac{\varepsilon_0}{2} E^2 \pi_1^2 \right) d\omega} \right)\]  

(A29)

Eq. (A29) can be written as

\[s(t) \approx \theta(t-t_0) e^{-\sigma(t-t_0)}\]  

(A24)
\[
\rho'(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\gamma_1 \omega^2 + i \gamma_1 \sigma \left( \frac{\sigma + \frac{\gamma_1}{2} \Gamma_0}{\omega} \right) \omega - \gamma_1 \sigma \frac{\gamma_1 \Gamma_0}{2} + \gamma_1 h_{\xi} q_v}{(\omega - \omega_1)(\omega - \omega_2)(\omega - \omega_3)(\omega - \omega_4)} e^{-i\omega t} d\omega \tag{A30}
\]

with \( \omega_4 = -i\sigma \) and \( \omega_{1,2,3} \) as in (A9). To calculate (A22) the integral in the lower half complex plane where all poles are, i.e. summing over all residues. The result of this quite involving calculation is

\[
\rho'(t) = \frac{\varepsilon_0 E^2}{2} \theta(t-t_0)(P_{\text{elec}}(t) + P_{\text{therm}}(t)) \tag{A13''}
\]

with

\[
P_{\text{elec}}(t) = \frac{q^2}{(\omega')^2 + (\beta_1 - \beta_3)^2} \gamma_1 \varepsilon_0 e^{-\beta(t-t_0)} \left\{ \begin{array}{l}
\left[ \omega' + \frac{(\beta_2 - \beta_3)(\beta_1 - \frac{\gamma_1}{2} \Gamma_0)}{\omega'} \right] \sin \omega'(t-t_0) \\
- \left[ \frac{\gamma_1 \Gamma_0}{2} - \beta_2 \right] \cos \omega'(t-t_0) - e^{-i(\beta_2 - \beta_3)(t-t_0)} \end{array} \right. 
\tag{A14''}
\]

\[
P_{\text{therm}}(t) = \frac{\gamma_1 h_{\xi} q_v}{(\omega')^4 + \left( \frac{\beta_1 - \beta_2}{\omega'} \right)^2} \left[ 1 + \left( \frac{\beta_1 - \beta_3}{\omega'} \right)^2 \right] e^{-\beta(t-t_0)} \left\{ \begin{array}{l}
\left[ 1 + \left( \frac{\beta_1 - \beta_2}{\omega'} \right)^2 \right] \sin[\omega'(t-t_0)] \\
- \left[ 1 + \left( \frac{\beta_1 - \sigma}{\omega'} \right)^2 \right] e^{-\beta(t-t_0)} \cos[\omega'(t-t_0)] \\
- \left[ 2 \beta_3 - \sigma - \beta_2 \right] \sin[\omega'(t-t_0)] \\
\end{array} \right. 
\tag{A31}
\]

Note that the electrostrictive contribution remains the same as before, as expected. Finally the complete solution for the density change \( \rho'(x,t) \) is obtained by including the spatial dependence, i.e. \( \rho'(x,t) = 2\cos q\varphi \rho'(t) \). In the approximation that takes into account only linear terms in \( \beta_i/\omega' \) and adding together the sine and cosine terms, Eq. (A31) multiplied by Eq. (A13'') transforms into Eq. (28) of the theory section.
5.2 Density variation induced by a Gaussian temporal profile pulse

Assuming no radiation absorption and considering only linear terms of $\beta_i/\omega'$, the density variation induced by a $\delta(t)$ pulse is, see Eqs. (A12) and (A13)

$$\rho'_G(t) = \frac{\gamma_c}{2} \frac{E_0 E^2 \tau_i^2}{\omega'} \Theta(t) \left[ e^{-\beta_i(t)} [L \sin(\omega'(t)+\Phi)] + \xi e^{-[\beta_i(t)]} \right], \quad t_0=0 \quad (A32)$$

with

$$\xi = \left( \frac{\beta_2 (\gamma - 1)}{\omega'} \right)$$

$$L = \left(1 + \xi^2\right)^{1/2} \quad (A33)$$

$$\phi = \arctan(-\xi)$$

Since $\xi<<1$, $L \approx 1$ and $\phi=0$.

In the following a calculation for situation of a laser pulse with a Gaussian temporal profile is given. The source term in the momentum transfer equation, which can be expressed as a product of a function of time and a function of the spatial coordinate, is in this case

$$h(x,t) = \tilde{h}(x) \tilde{h}(t) = \nabla^2 \tilde{E}^2 = -E_0 \gamma E^2 \tau_i^2 \cos(qx) A^2(t) \quad (A34)$$

where $A(t)$ is the Gaussian temporal profile of the pulse

$$A(t) = \sqrt{2} \left(\frac{\tau}{\pi}\right)^{1/4} e^{\frac{1}{2} \left(\frac{t-t_c}{\tau}\right)^2} \quad (A35)$$

where $t_c$ is the position of the maximum of $A(t)$ and $\tau$ is its width and $\int_{-\infty}^{\infty} A^2(t) dt = 1$. The induced density variation is obtained by convoluting the induced density variation caused by a $\delta(t)$ shaped laser pulse with the time-dependent part of the new source term, i.e.

$$\rho'(x,t) = \frac{E_0 E^2 \tau_i^2}{\omega'} \cos(qx) [\rho'_G(t) * \tilde{h}(t)] \quad (A36)$$

where

$$[\rho'_G(t) * h(t)] = \int_{-\infty}^{\infty} \rho'_G(t-t') h(t') dt' \quad (A37)$$
Since $\xi << 1$, the term decaying with the constant $\beta_2$ is neglected in the following and (A37) becomes

$$[\rho'_G(t) * h(t)] = \gamma_\epsilon \sqrt{\frac{2E_0E^2}{\pi\omega'}} \left[ \int_{-\infty}^{+\infty} \theta(t-t') e^{-\beta_1(t-t')} \sin[\omega'(t-t')] e^{\left[ \frac{t-t'}{\tau} \right]^2} dt' \right]$$

(A38)

Expressing the sinus in polar coordinates the integral in (A38) develop into as a sum of two integrals $I_1$ and $I_2$

$$[\rho'_G(t) * h(t)] = I_1 - I_2 = \int_{t-t_0}^{+\infty} e^{\rho_1(t')} dt' - \int_{t-t_0}^{-\infty} e^{\rho_2(t')} dt'$$

(A39)

where $P_i(t')$ are two polynomials that read explicitly

$$P_{1,2} = \left[ \frac{t'-t_c}{\tau} \right] - (\beta_1 \pm i\omega')(t-t')$$

(A40)

After some calculation the following expression is obtained

$$[\rho'_G(t) * h(t)] = \gamma_\epsilon \frac{\epsilon E^2 q^2 \tau}{\omega'} \Im \left\{ e^\epsilon \left[ 1 + \operatorname{erf} \left[ \frac{1}{\tau} (t-b) \right] \right] \right\}$$

(A41)

with

$$b = t_c + \frac{\tau^2}{2} (\beta_1 - i\omega')$$

(A42)

$$c = \frac{\tau^2}{4} (\beta_1 - i\omega')^2 - (\beta_1 - i\omega')(t-t_c)$$

and with the error function $\operatorname{erf}(x)$ which is defined as

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-y^2} dy$$

(A43)

Thus the final expression for the induced density variation becomes

$$\rho'(x,t) = \gamma_\epsilon \frac{\epsilon E^2 q^2 \tau}{\omega'} \cos(qx) \Im \left\{ e^\epsilon \left[ 1 + \operatorname{erf} \left[ \frac{1}{\tau} (t-b) \right] \right] \right\}$$

(A44)
6 Bibliography


[HITDATA] HITRAN Database


