Single-measurement Determination of Molar Fraction and Temperature of Binary Gas Mixtures from Combined Laser Induced Grating and Four-wave Mixing Signals

Jonas I. Hölzer[1], **Dimitrii N. Kozlov**[2], **Thomas Seeger**[1]

[1] Engineering Thermodynamics Siegen, University of Siegen, Paul-Bonatz-Str. 9-11, 57076 Siegen
{jonas.hoelzer, thomas.seeger}@uni-siegen.de

[2] A.M. Prokhorov General Physics Institute Russian Academy of Sciences, 119991, Moscow, Russia
dnk@kapella.gpi.ru

Abstract. *The laser induced grating (LIG) technique is a powerful, versatile, non-intrusive measurement technique that employs the generation of a density grating by application of the strong electric fields of a laser. Studying the resulting energy release in the time domain gives access to a manifold of thermodynamic, fluiddynamic and material properties. Typically, the determination of different properties is mutually exclusive i.e., all other properties need to be known for the accurate determination of an unknown. We demonstrate concurrent determination of temperature and concentration in CO_2-N_2 mixtures from the same measurement exploiting the occurrence of a four-wave mixing signal contribution to the LIG signal.*

Keywords. Laser induced grating spectroscopy, Gas phase diagnostics, time-domain measurements, four-wave mixing, multiparameter measurements

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1 Introduction

Crossing of short laser pulses in a medium leads to interference of the incident electromagnetic fields and generates a grating of alternating strong and weak electric field. The fringe spacing of this laser induced grating (LIG) is governed by the laser wavelength and the angle of incidence. The strong electric field at regions of constructive interference leads to a polarization of the medium. This generates two counter propagating density i.e. sound waves, which result in a modulation of the refractive index. This is known as an electrostrictive grating (LIEG). Therefore, observation of the intensity of a refracted probe beam over time gives access to the energy release and the sound velocity and thereby a multitude of thermodynamic, fluiddynamic and molecular properties like concentration and temperature [1–5]. If the laser energy coincides with a resonance in the medium a thermal grating appears due to absorption of pump radiation. A model for the intensity of a beam, diffracted of electrostrictive and thermal gratings can be found in [6]. Due to the interdependence of many parameters (e.g., the sound velocity depends on concentration and temperature) multi-parameter diagnostics is challenging. In this work we demonstrate the application of a four-wave mixing contribution to the LIG signal for simultaneous gas phase concentration and temperature determination.

2 Experimental section

A schematic representation of the experimental setup is shown in Fig. 1. The setup is designed to be robust and compact to be used in cramped test facilities. Therefore, only off-the-shelf lasers have been used. A flashlamp pumped Nd:YAG laser produces pulses at 1064 nm, 10 Hz and 42 mJ, which are used as the pump-beams. This output is guided through a half-wave plate to adjust polarization and is split in two by a beam-splitter. The forward scattered light from the half-wave plate is caught by a photodiode, which is used as a trigger for the signal detection. One of the pump beams is guided through a second half-wave plate for experiments with crossed pump-beam polarizations. The pump beams are overlapped in a heatable gas cell at an angle of about 2° using lenses with a focal length of 750 mm. The fringe spacing of the resulting LIG is about 31 µm. The probe beams is provided by a continuous wave (cw), frequency doubled, diode pumped solid state (DPSS) laser (532 nm, 50 mW), which is guided through a half-wave plate and overlapped at an opening angle of 0.5° from the centerline. The signal is separated from pump and probe by beam dumps and a laser line filter. Additional local filtering is done by focusing the beam into an optical fiber. The signal is detected by a photomultiplier tube that is connected to an oscilloscope. The neat gases CO_2, N_2 and CO_2-N_2 mixtures at 1 bar and 295 K and 375 K were probed. Typically, 500 single shots are recorded and averaged in the evaluation procedure. The detail on the evaluation procedure can be found in [7].

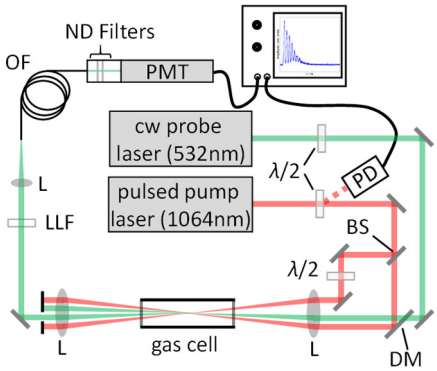

Figure 1. Schematic representation of the experimental setup: BS: Beam splitter, DM: Dichroic mirror, L: Lenses, LLF: Laser line filter, λ/2: half-wave plate, ND: Neutral density filter, OF: Optical fiber, PD: Photodiode, PMT: Photomultiplier tube.

3 Results and discussion

In LIEG measurements of CO_2 and N_2 are shown in Fig. 2. It can be clearly seen that the oscillation period of the signal amplitude is different for both gases. Furthermore, a strong peak appears in CO_2 at $t = 0$ ns that only has a small amplitude in N_2. Because of its dependence on the CO_2-N_2 mixture ratio the behavior of this peak may be used as a concentration probe, independent of sound velocity, which can then be applied for different parameters. To properly model the behavior of the signal shape and apply it for diagnostic purposes its nature needs to be elucidated. Due to its absence in nitrogen pump-laser straylight can be excluded and a resonant coherent effect like four-wave mixing is likely. Although a resonant (thermal) contribution to the LIG resulting from IR absorption can be excluded from fitting of the trace by the model from [6], there are very low cross-section resonances at the pump laser energy of 9398 cm^{-1}, which could promote a two-color four wave mixing (FWM) process.

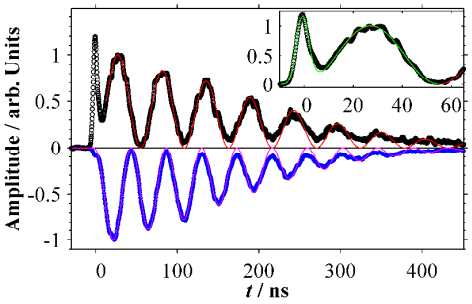

Figure 2. Amplitude of the diffracted beam of CO2 (black) and N2 (blue) as a function of time (500 averaged traces). The full traces have been fitted by the model function for LIGs from [1] (red and magenta respectively). The inset shows the first peak and the first LIG arc observed in CO2 which has been fitted by the model function to obtain the peak ratios (details in [7]) (green).

As additional proof for an FWM process the polarization dependence was probed as LIEG and FWM should exhibit different polarization dependences. The results of polarization dependent measurements are shown in Fig. 3. Since the density modulation comprising the LIG arises from interference of the pump-laser fields, crossed pump-beam polarizations (horizontal-vertical-vertical: HVV) do not lead to the formation of a LIG. Under these conditions the FWM signal amplitude is reduced, but not zero. Using vertical pump-beams and a horizontal probe beam (VVH), in contrast results in identical LIEG diffracted beam amplitude, but reduced FWM signal amplitude. Therefore, it can be safely assumed that the peak in fact arises from FWM. For quantification of the process an evaluation procedure has been developed that is in detail described elsewhere [7]. In brief the FWM peak and the first LIG arc are fitted by a Gaussian function and a squared sine function and their phase dependent crossterm respectively. The function yields the ratio of FWM and LIG in the form of the squared parameter R (displayed in Fig. 3).

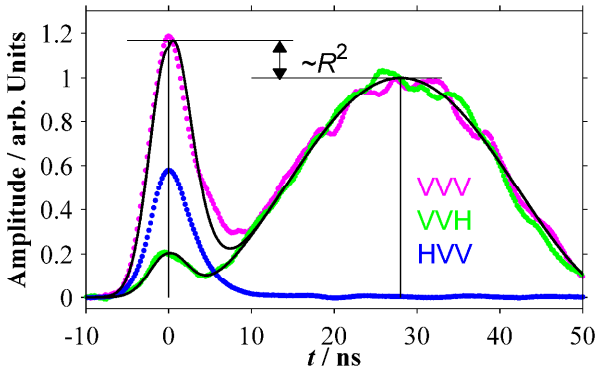

Figure 3. Plot of the amplitude of the diffracted beam as a function of time for three different polarization configurations: VVV: All-vertical, VVH: Vertical pump beams and horizontal probe beam, HVV: Horizontal pump-beam, vertical second pump and probe beams. The VVV and VVH traces have been fitted (black) by the model described in the text to obtain the parameter R^2 as a measure for the FWM- and LIG-diffracted beam amplitude ratio.

Because of its dependence on the CO_2-N_2 mixture ratio, the applicability of R as a probe for concentration was investigated. Fig. 4 shows the ratio of FWM and LIG signal amplitude R as a function of CO_2 molar fraction for 295 K and 375 K. The ratio increases with x from about 0.4 to 1.15 in a nonlinear fashion. This increase is weaker for higher temperatures. Using this curve as a calibration, R can be used as independent parameter for the determination of CO_2 concentrations in CO_2-N_2 mixtures. The concentration can then be used to determine the temperature from the sound velocity, which is encoded in the oscillation period. Due to the temperature dependence of R calibration curves for different temperatures need to be known and the temperature determination must be done in an iterative fashion. A physically derived model of the behavior including a detailed study on the temperature dependence of R can be found in [7].

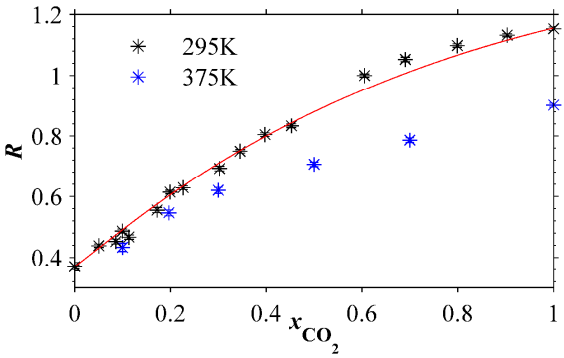

Figure 4. Plot of the parameter R (see text) as a function of CO_2 molar fraction in N_2 at 1 bar and 295 K (black) and 375 K (blue). The trace for 295 K has been fitted the model function from [7].

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