Misalignment effects in laser-induced grating experiments

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Abstract

Laser-induced grating spectroscopy (LIGS) is an experimental method, in which two pulsed and a continuous-wave laser beams have to be superimposed under well-defined angles to generate a coherent signal beam. In this Note, the possible effects of different forms of misalignment are examined. This includes the overlap of the pump lasers as well as the influence of the probe laser alignment on the temporal profile of the signal.

The laser-induced grating (LIG) method is a versatile analytical technique that allows the derivation of a multitude of thermodynamic parameters and thermophysical properties of a medium such as a gas or a liquid. For example, the time-resolved LIG signals carry in principle information about the temperature,[1, 2] the chemical composition,[3, 4] the speed of sound,[5, 6] the thermal diffusivity,[7, 8] and the viscosity.[9] However, careful alignment is crucial in order obtain reliable and reproducible results. In this Note, we analyze how an experimental misalignment can affect the LIG signal and what it means for the further processing and evaluation of the data. We focus on the effects of the collimation state of the beams. For an assessment of the influences of transverse pump beam displacements, see the work of Schlamp and coworkers.[10]

In a typical LIG experiment, two equally polarized pump laser beams from the same pulsed laser source (wavelength λ_{pump}) are crossed at an angle, θ_{pump}. Consequently, a spatially periodic light intensity distribution is generated in the form of a fringe pattern with a characteristic spacing

$$\Lambda = \frac{\lambda_{\text{pump}}}{2\sin(\theta_{\text{pump}}/2)} \tag{1}$$

LIGs are spatially periodic modulations of the complex refractive index evolving from the interaction of the radiation field of this pattern with the medium. The main grating formation mechanisms are electrostriction and thermalization of absorbed light energy due to collisions, which lead to electrostrictive and thermal LIGs, respectively. Electrostriction is the deformation of dielectric materials in the presence of an electric field. As a result, a medium becomes denser in regions of a high electric field strength. This means that the LIG interference pattern is converted into a local modulation of the density and hence a spatially periodic modulation of the refractive index. On the other hand, if the molecules in the medium can absorb photons of the pump radiation, i.e. when the laser wavelength is in resonance with an electronic or vibrational transition of the molecules, the interference pattern is converted into a population grating. Thermalization of the absorbed energy via collisions leads to a temperature and thus a density and refractive index modulation.

The induced density modulation causes two counter-propagating acoustic waves to be generated perpendicular to the planes of the fringes. The result is a standing acoustic wave that oscillates with a period of $T_a = \Lambda/c_s$, where c_s is the speed of sound of the medium. The oscillation decays exponentially as the sound waves leave the interference pattern and/or they are attenuated by the viscous sound attenuation of the medium. In the case of thermal LIGs, there are two further effects that need to be considered: the time scale of the collisional processes and the diffusion of the molecules. The collisions determine how the thermal LIG is formed and the diffusion governs its decay. A detailed description of the mechanisms can be found in the literature.[11-13]

In order to probe the dynamics of such transient LIGs, a third beam from a continuous wave laser (wavelength λ_{probe}) is overlapped with the pump beams fulfilling the Bragg condition,

$$\sin(\theta_{probe}) = \frac{\lambda_{probe}}{2\,\Lambda} \tag{2}$$

The Bragg diffracted intensity represents the LIG signal and can be detected with high temporal resolution using a photomultiplier tube or a fast photo diode.

A common procedure for aligning the pump beams in a LIG setup is to have them parallel, and then focus and cross them with the same lens. If the beams are well collimated before the lens, the intersection region can be considered as illustrated in Fig. 1a, as the beam waist of a focused Gaussian beam can be approximated as a plane and parallel wave front. The red lines indicate the interference pattern. The blue and green sinusoidal curves illustrate the intensity distributions along the vertical solid blue and the dashed green lines. It becomes clear that the grating spacing is independent of the location in the grating. In other words, a uniform grating is formed. In contrast, when the beams are not perfectly parallel and/or not collimated before the lens, the situation illustrated in Fig. 1b is likely. In the intersection region, the pump beams are either converging or diverging and, therefore, the wave fronts are no longer plane parallel. Since the normal to the wave fronts from the two beams now cross at an angle that depends on x and z, the fringe spacing becomes a function of position, which becomes clear from Eq. (1). When the probe laser beam is overlapped with such a grating, the detected LIG signal will vary depending on where inside the grating the Bragg condition is best fulfilled. For completeness, we note that a variation in z-direction can be considered additionally as the vertical lines crossing the fringes could be drawn such that they are normal with respect to the individual fringes. This would result in a slight curvature of these lines. Since this effect appears small compared to the variation of the grating spacing along the x-axis it is neglected in the following. A more detailed assessment of the effects in z- and x-direction will be looked at in a future project.

Misalignment effects in the case illustrated in Fig. 1a, i.e. assuming plane wave fronts, were studied by Schlamp et al.[10] They analyzed the consequences of transverse displacements of the pump beams. For this purpose, a theoretical framework was developed and good agreement between theory and experiment was found. In the present manuscript, we focus on the effects of the collimation state of the beams as shown in Fig. 1b.

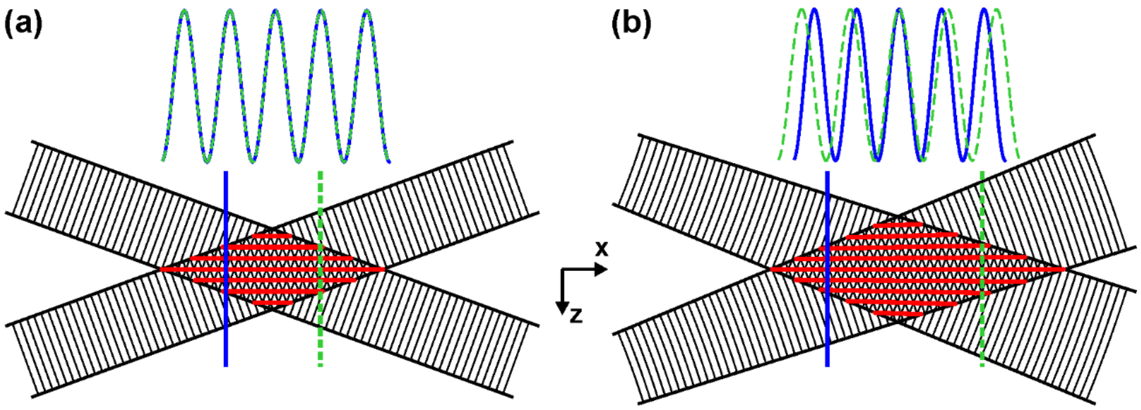

Fig. 1. Superposition of (a) two perfectly collimated and (b) two slightly diverging beams. The dashed green and solid blue sinusoidal lines indicate the intensity distributions along the corresponding vertical lines.

In order to highlight the effects, a series of time-resolved LIG signals is displayed in Fig. 2. The left diagram of panel (a) shows a simulated and thus artifact-free signal. The first part of the signal is dominated by the acoustic signal that manifests as a fast oscillation of the intensity. The latter part is governed by thermal diffusion. Therefore, it is straightforward to analyze the signal by determining the power spectrum using Fourier transformation (see right column diagrams of Fig. 2), and by fitting an exponential function to the decaying part. This rather simple procedure can yield information about the speed of sound and hence the temperature as well as the thermal diffusivity of the medium. The power spectrum shows a single pronounced peak indicating a uniform grating spacing. The signals in the other panels were recorded experimentally. Thermal LIGs were generated in a methane/nitrogen gas flow using mid-infrared pump radiation (3-4 ns, 5 mJ) at 3015 cm^{-1} to excite a vibrational Q-branch transition of methane. A cw laser (457 nm, 330 mW) served as probe laser. All laser beams were focused with the same 300-mm focal length lens and had a diameter of about 2 mm before the lens.

Figure 2b shows a LIG signal with only minor artifacts. There are two recurrences of the acoustic oscillations, which indicate a slight misalignment. The corresponding power spectrum, however, reveals a single peak. This indicates that the pump lasers are overlapped well to form a uniform LIG. Note that the probe laser in the experiment had a smaller diameter in the focal spot and hence did not probe the entire grating (assuming Gaussian beams and taking the above details of the laser beams into account, the beam waist in the focal spot of the IR beam is about six times larger than that of the probe beam). Consequently, acoustic waves from different regions of the interference pattern approach the probed volume and lead to the observed behavior. Nevertheless, it should be noted that deriving the oscillation frequency and decay time constant would yield reasonably good results.

This situation changes drastically in panels c) and d) of Fig. 2. The diagram to the left in Fig. 2c displays three signals generated with slightly different alignments for which the probe beam was moved from its center position by fractions of a millimeter (several times the grating spacing, $\Lambda \approx 31$ µm). In the blue and green curves there seems to be a beating between acoustic oscillations manifesting as recurring signal. The red curve shows an oscillation that slowly decays over an extended period of time. The power spectra of all of these signals (see panel c, right diagram) exhibit two or even more peaks. Different regions in the grating are characterized by a different grating spacing and hence by different oscillation frequencies of the acoustic signal. The frequency determined by the Fourier transform analysis is proportional to the speed of sound and hence it has a quadratic relationship with the temperature. Therefore, even a small error in the frequency can lead to large uncertainties in the derived parameters. Note that considering the situation illustrated in Fig 1b a broadened peak in the power spectrum would be expected as the grating spacing increases continuously. The experimental data, however, show distinct shoulders and sub-peaks. This is likely because the beams in the experiment are not perfectly Gaussian and consequently the fringe pattern does not have a perfectly continuous and regular shape; hence the observed signals.

For recording the two LIG signals shown in Fig. 2d, the probe laser beam was positioned at the edges of the interference pattern. In particular, the red signal reveals a kind of short delay before the strong oscillations start. The initial weak signal (shown enlarged) is likely due to the local acoustic wave while the stronger signals arise when the acoustic waves from other regions in the interference pattern arrive at the probed volume. Moreover, the stationary thermal grating, which normally governs the slowly decaying contribution to the signal, appears to be absent. This indicates that the local thermal grating is not very distinct and the thermal diffusivity leads to a rapid dilution and cooling of the hot molecules before they arrive at the probed location. The hypothesis that the signal is dominated by the acoustic contribution is also supported by the enhanced peaks, which appear at the doubled frequency in the power spectrum.[14]

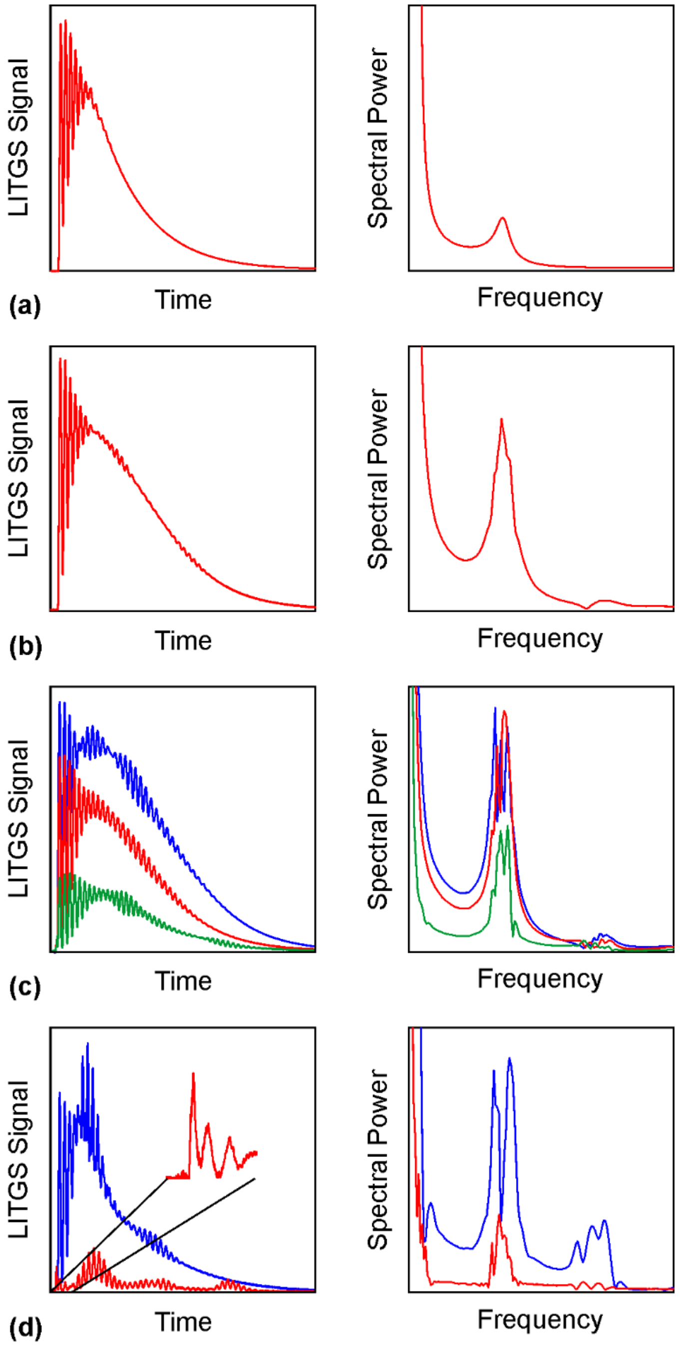

Fig. 2. Time-resolved LITGS signals (left column) over a period of 5 µs and their corresponding power spectra obtained by Fourier transformation (right column). (a) simulated signal, (b) slightly misaligned beams, (c) strongly misaligned beams, (d) probe beam crosses edge of LIG volume.

In conclusion, we have shown how minor and major misalignment in a laser-induced grating experiment can affect the signals and it was discussed what this means for the most common forms of data analysis. Common sources of artifacts in the signal are the crossing angle, the position, and the collimation of the pump beams. We focused on the latter as the collimation effects can significantly bias the parameters to be measured, while Schlamp et al.[10] found that transverse displacements of the pump beams have minor influence. However, such displacement effects may become crucial when the pump beams are not collimated in their intersection region. In the measurement volume, the beams should be quasi-collimated with a plane wave front. In the following, some recommendations are given for aligning a LITGS experiment:

- The beam profile of the pump beams should be well-defined, i.e. Gaussian. Depending on the profile provided by the laser source, it can be beneficial to cut the edges of the pump beams using an aperture in order to obtain an appropriate interference pattern. The resulting reduced signal intensity is normally outweighed by a significantly improved quality of the signal characteristics. However, producing a top-hat profile may result in high-order spatial modes, which could cause undesirable interference.
- When focused by the same lens, the pump beams should be parallel and collimated before the lens. This ensures that the beams cross each other with their minimal beam waist in the focal spot, where the wave front can be assumed plane and parallel.
- The path difference of the pump beams from the laser to the focusing lens should be identical. This enables the coherent superposition of the beams within the coherence length. This is particularly crucial when broad bandwidth lasers are used.[15, 16]
- The probe laser should cross the LIG fulfilling the first-order Bragg condition. This maximizes the signal intensity.

- Ideally, the probe laser should exhibit the same beam waist diameter in the measurement volume in order to obtain an optimal diffraction. A theoretical assessment of geometrical effects can be found in the work of Siegman.[17] When the pump and probe lasers have different wavelengths but should be focused by the same lens, chromatic aberration effects must be compensated by adjusting the collimation state.
- When the probe laser diameter is small compared to the interference pattern created by the pump beams, it should cross the LIG in the center rather than the edges in order to avoid artifacts in the signal traces.

Ongoing research in our lab includes the development of a comprehensive theoretical framework to predict misalignment effects and to identify them quantitatively in experimental LIGS signals.

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