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LASER-INDUCED TRANSIENT GRATINGS: THE METHOD AND ITS APPLICATIONS

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Abstract - Transient grating techniques are shown to be a very well adapted method to follow the response of a material to short laser excitation pulses. Besides its enhanced sensitivity this technique is suited for the study of processes requiring a spatially modulated excitation of the medium. The choice of experimental conditions will select the response mechanisms and generate different types of gratings. Each type will be illustrated by an example.

INTRODUCTION

The development of the transient grating technique followed the capabilities of laser sources: shorter pulses give the possibility to record faster transients of a system. The grating is produced by crossing two laser beams in the material under investigation: their interference realises a spatially modulated intensity (and/or polarization) distribution and the coupling of the light field with the medium creates in turn a spatial modulation of the optical properties of the medium, i.e. a grating. The system then exhibits a transient behaviour and relaxes with one or more characteristic times. A third laser (probe) beam is diffracted on the grating and the temporal evolution of the signal gives informations on the relaxation time(s).

The mechanisms involved in the grating formation and decay can be selected by a proper choice of experimental conditions. As the field of applications is very wide and has already been intensively explored /1/, we shall outline here some specific features of the method. We will not consider the following situations more easily interpreted within other theoretical frames: i) zero or near zero time delay between pump and probe pulses and ii) two pump pulses of different frequency (moving gratings /2/), situations more often and conveniently analyzed by four-wave mixing, iii) time delay between the two pump pulses more often defined as photon echoes.

BASIC PRINCIPLES

The geometry used for the production of a grating is shown in Fig. 1. Two laser beams with wave vectors \( k_1 \) and \( k_2 \), obtained by splitting a single beam, intersect at an angle \( \theta \) and create an interference pattern.
The grating wave-vector \( \mathbf{q} \) is such that \( \mathbf{k}_1 - \mathbf{k}_2 = \pm \mathbf{q} \) and the spatial period \( \Lambda = 2\pi/q \) is related to the excitation light wavelength in the material \( \varepsilon \) by \( \Lambda = \lambda/(2 \sin \theta/2) \). The response of the material is expressed by a modification of the complex refractive index \( \tilde{n} = n + ik \). \( \Delta n \) and \( \Delta k \) will be the modulation depths of the refractive index \( n \) and of the absorption constant \( K \). \( \Delta K = 0 \) defines a pure phase grating and \( \Delta n = 0 \) a pure absorbing grating.

The detection of the grating is performed by diffracting a third laser beam. When the temporal evolution is not too rapid, fast detectors can be used to directly time resolve the signal generated by a continuous or quasi-continuous beam. When higher time resolution is required a sampling method is used: the grating is probed by a retarded third laser pulse, the delay of which with respect to the pump pulses is conveniently realized by means of an optical delay line /3/. The geometrical arrangement must take into account the thickness \( d \) of the grating: for a thin grating which roughly satisfies the condition \( \lambda d \ll \Lambda^2 \), several diffraction orders are observed whereas for a thick grating only one diffracted wave-vector \( \mathbf{k}_d \) respects the Bragg condition \( \mathbf{k}_d = \mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k} \), where \( \mathbf{k} \) is the probe wave-vector. Fig. 1 shows a possible geometry for a thick grating with a probe pulse of wavelength longer than the pump one. General diffraction theories using a plane-wave approximation give the full expressions of the diffraction efficiency \( \eta \) in both the cases of thin and thick gratings /1,4/. The conditions of validity of this approximation are fulfilled if the laser beams are not too much focused and if the medium is only slightly absorbing. In the limit where the variations of the complex index of refraction are very small, i.e. when \( 2\pi|\Delta n|/\lambda \ll 1 \), a condition which is almost always verified, both the diffraction efficiency for a thick grating in the Bragg condition and the intensity diffracted in the first order from a thin grating are proportional to \( |\Delta n|^2 \).

The last step consists in relating \( \Delta n \) to the parameters of the system which of course has to be done for each case studied.

**ADVANTAGES OF THE METHOD**

As this technique is experimentally more demanding than for example a pump-probe scheme, it is necessary to emphasize its main advantages. First, as the diffracted signal is emitted in a direction well separated from the transmitted pump and probe beams (see Fig. 1), the signal is collected against zero background giving a good sensitivity. Second, the spatially periodic arrangement of the diffracting sources realises constructive addition of the diffracted signal in selected directions (forced scattering), with a corresponding enhanced intensity and increased signal.

Third, the spatial modulation of the excitation is certainly the most specific feature of the technique, which offers its best applications when this spatial modulation is required in order to follow excitation mobility for example. So, transport phenomena will find there a particularly well suited method.

**THE DIFFERENT TYPES OF GRATINGS**

The mechanisms involved in a grating formation depend, of course, upon the pump light properties. The choice of the excitation wavelength allows to select resonant, strong interactions leading to the creation, for example, of an electronic or vibrational excited state grating which may decay towards intermediate excited states forming thereby a secondary grating. With a proper choice of the probe wavelength it is possible to follow the rise and decay of the intermediate excited states. Finally it will only remain a heat deposition in the fringe pattern giving rise to a temperature grating. Then the heat will diffuse accompanied by density variations.

When the excitation beams are both polarized perpendicular to the plane of incidence the light intensity is modulated but its polarization remains parallel to the incident ones. This situation is reverse when one of the two polarizations is set in the plane of incidence: this gives no intensity modulation but a spatially periodic rotation of the polarization. This geometry may be interesting for dichroic media and more generally for an anisotropic property study (see first example).
The pulse duration $t_p$ is a fundamental parameter for two reasons. The first is that relaxation processes of characteristic time shorter than $t_p$ are not observable as their decay will follow the excitation pulse temporal envelope. The second, more specific, concerns the excitation of an oscillatory response of the system. A good example is given by the impulsive generation of hypersound waves of wavelength $\Lambda$ which is possible if $t_p$ is sufficiently short (see third example).

Finally as the types of gratings appear to be as numerous as the physical mechanisms creating them, it seems to be judicious to introduce a classification according to their wave-vector $q$ which is a specific parameter of the method. So, we will separate $q$-independent gratings for which the grating spacing is not a relevant parameter of the temporal evolution, from $q$-dependent ones where on the contrary the value given to $\Lambda$ by the experimental geometry is fundamental. In this second category, a further distinction can be made between oscillatory and non-oscillatory behaviors as described below.

1. $q$-independent grating behavior

If the excitations decay at the place they have been created, the time evolution of the signal is independent of $q$ and the increased sensitivity of the method to measure various excited state life times or relaxation times is used. An example (after Ref. /5/) Fig. 2a shows the signal diffracted by a dye solution (9-aminoacridine in ethylene-glycol) when the excitation light pulses have the same polarization, the probe light being polarized parallel or perpendicular to the pump light.

In Fig. 2b, the two excitation pulses have crossed polarizations. The dye molecules with a transition moment parallel to the excitation field are preferentially excited. These gratings read out by a polarized light pulse are seen to decay with a time simply related to the orientational correlation time $\tau_{or}$ and excited singlet life-time $\tau_{ex}$.

2. $q$-dependent, non-oscillatory grating behavior

In this case, the decay time of the grating depends upon the fringe spacing. This occurs when there is a migration of the excitations from a region of high density (bright fringes) to a region of lower density (dark fringes). The technique has been applied to heat diffusion, mass diffusion, carrier diffusion in semiconductors, charge and energy transfer ... Fig. 3, after Ref. /6/, illustrates such an application for singlet exciton transport in anthracene single crystals. A one-dimensional diffusion equation for the density of excitations with life-time $\tau$ and diffusion constant $D$ leads to a grating decay time $\tau_q$ easily related to $\tau$ and $D$ by $1/\tau_q = 1/\tau + \varepsilon Dq^2$.

Fig. 3 shows a plot of $K = 2/\tau_q$ versus $\varepsilon^2(q = \varepsilon \omega << 1)$ giving $\tau$ and $D$.

3. $q$-dependent, oscillatory grating behavior

The present case is a very particular one as it concerns the formation of hypersound waves of fixed wavelength equal to the grating spacing $\Lambda$, i.e. determined by the angle $\theta$. The mechanism invoked in this case is either stimulated Brillouin scattering or thermal expansion following
a fast heat deposition in the fringes of an absorbing sample [7]. The propagation produces an oscillation of the density and thereby of the index of refraction. The second mechanism gives a much higher diffraction efficiency of the form:
\[ n = A(1 - \exp(-\alpha t) \cos \frac{2\pi t}{T})^2 \]
where \( \alpha \) is the acoustic attenuation and \( T \) the acoustic period. \( n(t) \) gives a and \( T \) i.e. \( v \) the speed of sound. The method has been used for the study of acoustic properties of a polymer material undergoing a glass-rubber transition as shown in Fig. 4 [8]. Similarly, impulsive stimulated Brillouin scattering has been used for the generation of transverse acoustic waves and applied to investigations of structural phase transition in KD\reflectbox{P} [9].

**SUMMARY**

This rapid survey of the transient grating techniques shows that it is very sensitive for the detection of small variations of the optical properties of a material (\( \Delta n = 10^{-6} \) is easily detectable), with a wide field of application. Moreover it becomes very well adapted when a spatial mobility of the induced modification of the medium is expected.

**References**


8. R. Casalsegnio, M. Pierre, Following paper