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Sign of the dominant charge carriers in photorefractive crystals determined by a phase-locked holographic technique

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ABSTRACT

This work presents a holographic method based on active feedback techniques for determining the sign of the dominant charge carriers in photorefractive materials. A two-step procedure is proposed: first off a stationary phase-locked hologram is recorded; an electric field normal to the grating layers is then applied to the material, thus producing a running hologram. The sign of the charge carriers is determined by comparing the direction of the applied field with the direction of the hologram movement, which is known through the automatically attached light pattern. The method can be applied from highly photoconductive to highly insulating materials. Furthermore, no information on any material parameter is required. The method is validated by a set of holographic experiments using a $Bi_{12}TiO_{20}$ crystal that has electrons as the majority photocarriers.

1. Introduction

Photorefractive crystals are very well suited to record phase volume holograms by the exposition to a light interference pattern [1]. The record mechanism involves photoexcitation, migration and trapping of charge carriers (electrons and/or holes), resulting in a spatial redistribution of electric charge within the crystal volume. A space charge field arises, which in turn modulates the refractive index of the material through the linear electrooptic effect. An important feature of photorefractive materials is that they can be tailored to many different purposes, such as data storage, phase conjugation, optical filters, and signal processing, just to name a few [2]. This great variety of applications is only possible due to the large diversity of crystals developed during the last decades [3,4]. In addition, crystals have been doped with many types of impurities at different concentrations to provide adequate donor and acceptor centers of charge carriers; these impurities affect the photocarrier sign, among other properties.

The photocarrier sign is closely linked to the charge transport properties of the material and therefore of fundamental importance from the viewpoint of applications. Although usually unwanted, both electrons and holes may be photoexcited depending on the active donor centers. In any case, the sign of the dominant charge carriers depends not only on the host material and on doping, but also on the crystal's origin and on experimental conditions such as the illumination wavelength, light intensity, and grating period [3–5]. It is therefore very important to have access to reliable techniques for determining the photocarrier sign and the corresponding charge transport parameters; a fact that was recognized since the early days of the development of photorefractive materials. Some well known techniques that were adapted and applied to different crystals include: direct photoconductivity measurements [6], different approaches of energy transfer measurements based on holographic techniques [5,7–11], photoinduced Hall-current measurements [12,13], classical and holographic time-of-flight techniques [14,15], electron paramagnetic resonance spectroscopy [16], among others [17,18].

The method here presented is devoted to answer a simple question about the photoconductivity in a photorefractive material: are the dominant charge carriers electrons or holes (*n* or *p* photoconductivity)? In its present form, it does not provide any additional information on other charge transport parameters, such as the carriers mobility, the diffusion (or drift) length, or the density of photoactive centers. However, a combination of distinct properties make this new method very attractive: (1) wide range of application, covering from highly photoconductive to highly insulating materials, (2) the method can be applied to crystals subjected to the most common internal charge driving forces (diffusion and bulk photovoltaic), which include both local and non-local holograms, (3) no prior knowledge on any material parameter is required (such as the sign of the electrooptic coefficient or the

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polarity of the *c*-axis in photovoltaic crystals), and (4) no need of several measurements; in fact, a single measurement is enough to answer unambiguously the proposed question.

2. Theoretical principle of the method

Consider the interference of two coherent beams in a photorefractive crystal. The development of the hologram is sketched in Fig. 1. The light intensity (I) excites electrons into the conduction band (left column) or holes into the valence band (right column). The generation rate (G) of free charge carriers is in phase to the light pattern. Three different mechanisms can be involved in the movement of the charge carriers: diffusion, the bulk photovoltaic effect, and drift (when an external electric field is applied). For the sake of simplicity, we neglect the bulk photovoltaic effect in Fig. 1; the generalization including photovoltaic currents (which are common for non-centrosymmetric crystals) will be discussed later at the end of the paper. Assume that initially no external field is applied to the sample. The existence of a concentration gradient causes the photoexcited carriers to diffuse from the bright areas to the dark areas before they recombine with the photorefractive trap centers. Thus, the diffusion gives rise to an amplitude difference between the generation and recombination (R) rates, which means that positive and negative ionized immobile centers are distributed inside the crystal volume. The corresponding space charge density (ρ) is the origin of the internal space charge electric field that modulates the refractive index of the material through the Pockels effect. The vertical dashed lines in Fig. 1 draw attention to the phase shift between the charge modulation ρ and the light pattern I; it is 0 or π depending on whether electrons or holes diffuse. It is worth noting that this difference on the phase shifts leads to different energy redistribution between the recording beams in two beam coupling experiments, which can be used to identify the photocarrier sign [7]. However, this information by itself does not always allow determining the type of charge carrier; for example, additional experiments (such as ellipsometric measurements) are required to give the sign of the electro-optic coefficient in cubic crystals such as those of the sillenite family ($Bi_{12}(Ge,Si,Ti)O_{20}$) [5]. Back to Fig. 1, assume now that a steady electric field (E) is applied to the sample along the grating wave vector **K** (x-direction). The diffusion is still present, but due to the different signs of the charge carriers, the electric force shifts the carriers in opposite directions depending on whether they are electrons or holes; in the figure, electrons move to left and holes to the right. The amplitude of the applied field is supposed to be small enough so as to shift the recombination rate by a small amount (short drift length of the carriers). As a result, the space charge density shifts to the right (same direction as the electric field E) if the carriers are electrons, or to the left (opposite to E) if they are holes. Contrary to what we might think, it is interesting to note that the space charge density (and hence the hologram), moves in opposite direction to that of the mobile charges.

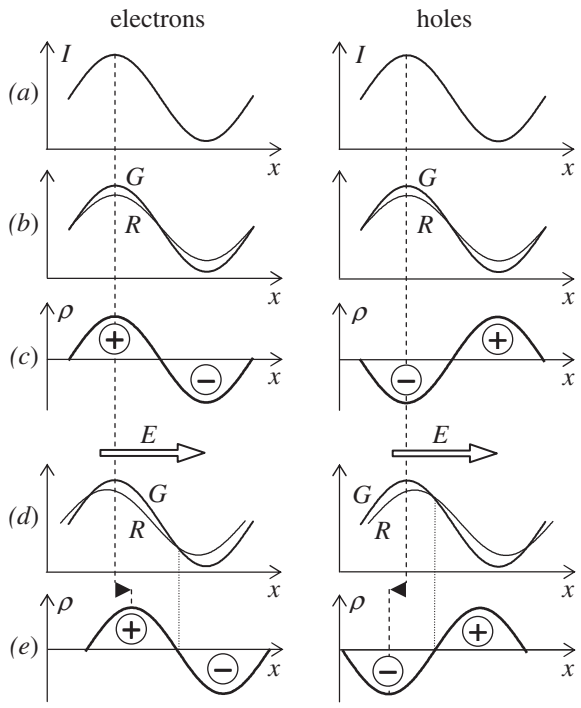

Fig. 1. Development of the photorefractive hologram. Charge carriers are assumed to be electrons in the left column and holes in the right one. The x coordinate is oriented along the direction of the grating wave vector. (a) Intensity (I) of the light interference pattern, (b) generation (G) and recombination (R) rates of free charge carriers under diffusion regime, (c) electric charge density (ρ) due to the ionized donor/acceptor photorefractive centers, (d) generation and recombination rates of free charge carriers under the influence of a steady electric field (E), and (e) the corresponding electric charge density. When compared to the diffusion regime, the charge density is shifted to the right (same direction of the electric field) if the free carriers are electrons and to the left in the case of holes.

3. Experimental implementation

In what follows, the experimental procedure on how to determine the direction of the hologram movement is described. The technique is based on the active phase control between the light pattern and the hologram being recorded. The general scheme of the interferometric setup used in the experiments is depicted in Fig. 2. The interference of two laser beams (R and S) generates a light pattern inside the crystal. A spatial modulation of the charge density grows up; it is accompanied by the internal space charge electric field and the refractive-index pattern (the hologram). Let us call x the spatial coordinate along the direction of the grating wave vector. Because the recording light fulfills the Bragg condition, part of the incident light is diffracted from one recording beam into the other one. Therefore, each beam in each one of the two output directions is the superposition of a transmitted and a diffracted wave. The phase shift φ between the interfering waves depends on the phase shift between the hologram and the light pattern (also called holographic phase shift), and on the beam coupling conditions between the R and S beams as they propagate

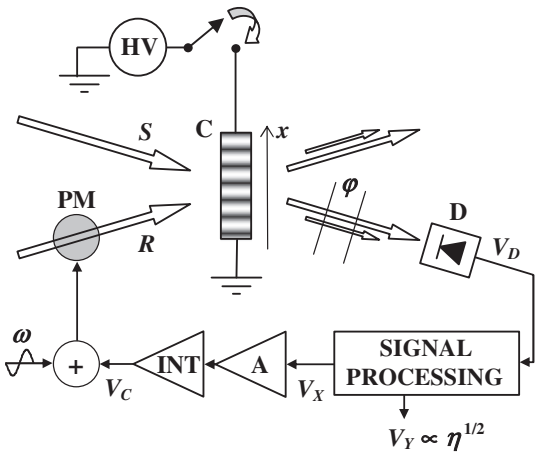

Fig. 2. General scheme of the holographic setup. HV: high voltage source, C: photorefractive crystal, R and S: input light beams, PM: phase modulator, φ: phase shift between the transmitted and diffracted beams, D: photodetector, V_D: photodetector's output voltage, V_X and V_Y: phase sensitive voltages generated by the signal processing circuit, A: amplifier, INT: integrator, V_C: control voltage fed back to the interferometer, ω: weak and fast sinusoidal phase modulation (dither signal), η: diffraction efficiency of the hologram.