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Influence of an externally applied magnetic field on vectorial interaction in $LiNbO_3$:Fe crystals

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An experimental investigation of the influence of an externally applied magnetic field on the dynamic grating formation in iron-doped lithium niobate is carried out. The diffraction efficiency and the two-beam gain depends strongly on the applied magnetic field. We observe changes in the two-beam gain and the diffraction efficiency of up to 40 and 75 %, respectively. The magnitude depends on the direction of the magnetic field. The interaction is believed to occur due to the anomalously high mobility of the nonthermalized free electrons responsible for the photovoltaic current, which in the vectorial interaction scheme causes the grating formation. A phenomenological description of the photovoltaic current including the photo-Hall effect shows a linear dependence on the magnetic field, which does not explain the obtained experimental results.

I. INTRODUCTION

Magnetic interaction in photorefractive media is a fairly recent field of research. Early studies of the effect of magnetotransport in these materials[1] have shown that the influence scales with the product of the free-carrier mobility and the magnitude of the externally applied magnetic field: the larger the product, the stronger the influence of the magnetic field on the induced refractive index change. With moderate magnetic fields ($\simeq 1$ T) a very weak effect is expected for most photorefractive crystals, since the mobility of the charge carriers is low. For the widely used sillenite crystals BSO, BGO, and BTO, recent measurements[2] have shown a mobility of 2–5 $cm^2/(Vs)$. Some experimental work has been published very recently[3] on diluted magnetic semiconductors examining Faraday effect and Voigt effect. To enhance these effects, the measurements were made at low temperatures (50 K) and large applied magnetic fields (6 T).

In this work, an experimental investigation of magnetic field influence on grating formation and energy coupling in ferroelectric crystals of $LiNbO_3$:Fe at room temperature is presented. In contrast to the previous experimental work[3] this effect occurs due to the magnetic field interaction with nonthermalized electrons. These possess anomalously high mobility and are mainly responsible for the photoconductivity of these crystals.[4] When electrons are photoexcited, they acquire an excess of momentum (large kinetic energy) and during the nonthermalized period of time (10^{-13}–10^{-12} s) they contribute to the bulk photovoltaic effect. Measurements of the influence of the magnetic field on photovoltaic currents in $LiNbO_3$:Fe (Ref. 5) and in other ferro- and piezoelectrics[6] have shown that anomalously high Hall photocurrents, which are directionally dependent on the magnetic field ($B = 0.35$ T), can be found. At room temperature, this effect was attributed to the Lorentz force acting on the nonthermalized electrons. From these measurements the photo-Hall mobility of nonthermalized electrons in $LiNbO_3$:Fe was estimated[5] to be $\mu_{Hp} \simeq 10^3$ $cm^2/(Vs)$, which is at least 3 orders of magnitude larger than the mobility of the thermalized electrons, i.e., electrons which have lost the excess momentum by "dressing."

The capability of coupling energy between two coherent laser beams, often referred to as two-beam gain, is one of the most fundamental characteristics of photorefractive materials. In the experiments presented here, we investigate the influence of an externally applied magnetic field on the grating formation and energy coupling in a vectorial holographic recording scheme.[7,8] This scheme is chosen since the grating formation is caused solely by spatially oscillating photovoltaic currents, and hence will exhibit the strongest magnetic interaction. The present measurements are made at room temperature and with a moderate applied magnetic field of 0.5 T. The dynamics of the grating formation and energy coupling with and without the applied dc magnetic field is measured. The applied magnetic field is found to strongly reduce the maximum diffraction efficiency and energy coupling as compared with the measurements without such a field. Moreover, the measured energy coupling is dependent on the direction of the magnetic field. In the following the experimental method and results are described.

II. EXPERIMENTAL METHOD

The experimental setup used for the beam coupling measurement is shown schematically in Fig. 1(a). A linearly polarized beam is incident at an angle Θ in a plane perpendicular to the polar c axis. On entering the crystal the beam divides into the two eigenmodes of the crystal, i.e., an ordinary and an extraordinary wave, I_o and I_e, respectively. A more general setup with angularly separated beams is shown in Fig. 1(b). The two eigenwaves excite a spatially oscillating current transverse to the c axis.[7,8] Using a phenomenological theory,[9] the spatially oscillating part of the induced photovoltaic current for a crystal of $LiNbO_3$ belonging to the $3m$ point group and with a magnetic field of magnitude B

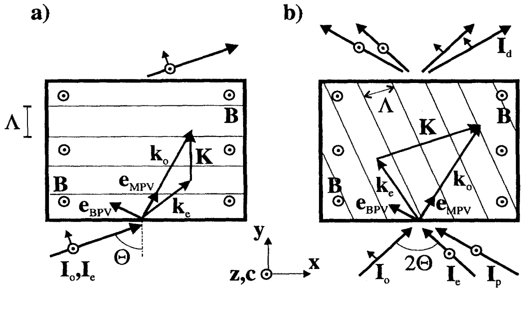

FIG. 1. Experimental setup: (a) Single incident beam vectorial interaction used for the beam coupling measurement. (b) General vectorial interaction with angularly separated incident beams. I_p and I_d are the HeNe-probe and diffracted beam. Unit vectors describing the direction of the components of the spatially oscillating current are shown.

applied parallel to the c axis is given by

$$\mathbf{j}^{PV}(\mathbf{r})=\frac{MI_T}{2}\mathbf{s}e^{i\mathbf{K}\cdot\mathbf{r}}+\text{c.c.},$$

where

$$\mathbf{s}=(\beta_{15}^L-i\beta_{12}^C)\hat{\mathbf{e}}_{\text{BPV}}+(S_3^L+iS_0^C)B\hat{\mathbf{e}}_{\text{MPV}}. \tag{1}$$

Here the indices L and C denotes the linear (real) and circular (imaginary) part of the bulk and magnetophotovoltaic tensors, β and S, of rank 3 and 4, respectively. For a crystal belonging to the $3m$ point group, S_L and S_C has 8 and 5 independent components, respectively. The components in Eq. (1) are $S_3^L=S_{1323}^L=-S_{2313}^L$ and $S_0^C=S_{131}^C=S_{232}^C$, where the subscripts 1, 2, and 3 refer to the principal crystallographic directions. The tensor S_C is antisymmetric in the last two indices and can therefore be written as a third-rank tensor. The optical modulation coefficient is $M=2\sqrt{I_oI_e}/I_T$, where $I_T=I_o+I_e$ is the total recording intensity. The part of the current stemming from the bulk photovoltaic effect is in the direction of the unit vector $\hat{\mathbf{e}}_{\text{BPV}}$ and is parallel to the state of polarization of the ordinary wave. The direction of the magnetophotovoltaic part is described by $\hat{\mathbf{e}}_{\text{MPV}}$, which is orthogonal to both $\hat{\mathbf{e}}_{\text{BPV}}$ and the applied magnetic field. Hence, it can be interpreted as a Hall photocurrent. The unit vectors describing the current directions are shown in Fig. 1.

The spatially oscillating current forms a periodically varying space-charge electric field, which in turn modulates the refractive index via the linear electro-optic (Pockels) effect. The induced refractive index grating has a wave vector given by $\mathbf{K}=\mathbf{k}_o-\mathbf{k}_e$, which in the configuration shown in Fig. 1(a) is parallel to the y axis. Only the component of the current parallel to $\mathbf{K}$ contributes to this modulation. Thus the induced space-charge field is proportional to $\mathbf{s}\cdot\hat{\mathbf{e}}_K$, where $\hat{\mathbf{e}}_K$ is a unit vector parallel to $\mathbf{K}$. Direct current measurements[10] have shown that $\beta_{12}^C \gg \beta_{15}^L$ in LiNbO$_3$ crystals. A similar relation is assumed to hold for the corresponding components of the magnetophotovoltaic tensor, S_0^C and S_3^L, respectively, assuming that these components are proportional to the photo-Hall mobility.[5] Hence, the largest grating component is associated with the circular part of the current. As seen from Eq. (1), this component is shifted by $\pi/2$ (nonlocal response) with respect to the linear component and gives rise to energy coupling between the two beams.[8] The intensity coupling coefficient is proportional to the imaginary part of $\mathbf{s}\cdot\hat{\mathbf{e}}_K$, while the diffraction efficiency of the induced refractive index grating is proportional to the modulus squared of $\mathbf{s}\cdot\hat{\mathbf{e}}_K$. Both these quantities can be shown to be effectively linear in B. Hence, the theory based on Eq. (1) predicts a linear variation in B of both the energy coupling and diffraction efficiency.

The actual measurements are performed using an expanded and linearly polarized beam from an argon-ion laser operating at 514.5 nm. The intensity ratio or modulation between the ordinary and extraordinary beam is controlled by varying the angle of polarization with a half-wave plate. The total incident intensity is 360 mW/cm^2, and a modulation of 0.31, with the extraordinary beam being the weaker of the two. It is this beam that increases in energy. The coupling is monitored by measuring the intensity in each of the two polarization states, corresponding to the eigenmodes of the crystal. Prior to each measurement, the crystal was thermally erased by heating it to a temperature of 250 °C for 5 h. In the experiments a crystal of LiNbO$_3$:Fe with a dopant concentration of 0.03 mol. % is used. The dimensions of the crystal are 10x10x10 mm^3. The absorption coefficient at the wavelength used was measured to be 1.0 cm^{-1} for the ordinary wave and 0.72 cm^{-1} for the extraordinary component. The faces perpendicular to the y axis of the crystal are antireflection coated for the actual wavelength. The magnetic field is applied by placing the crystal in a 15 mm long air gap between two rare-earth doped permanent magnets of 25.4 mm diameter. The magnets are connected via an iron yoke to ensure uniformity of the flux lines. Measurements of the magnetic-flux density showed an almost homogeneous field over the air gap with a magnitude of $B=(0.50\pm0.04)$ T.

III. RESULTS AND DISCUSSION

The dynamics of grating formation and energy coupling are shown in Figs. 2 and 3. The normalized change in the measured extraordinary intensity is plotted as a function of time in Fig. 2. This is done for the applied magnetic field in the two directions parallel to the c axis and without an applied magnetic field. The coupling is seen to depend strongly on the applied magnetic field. In all three situations the direction of energy transfer is from the ordinary wave to the extraordinary wave, which is an inherent property of LiNbO$_3$:Fe crystals.[8] Without the applied magnetic field, a maximum energy coupling of 5.5 % was obtained. With the magnetic field in either direction, the coupling was reduced. With $\mathbf{B}=(0,0,B)$, the maximum energy coupling was 3.8 % and for $\mathbf{B}=(0,0,-B)$, a maximum value of 3.3 % was measured. The sign of the magnetic field is chosen arbitrarily. Rotating the crystal 180° about the y axis yields the same results as those shown in Fig. 2. Both the effective electro-optic and the photovoltaic constants change sign with this operation, resulting in no change in the direction of energy coupling.

Similar measurements have been performed regarding the diffraction efficiency of the recorded grating, for which the experimental setup is shown in Fig. 1(b). This is achieved

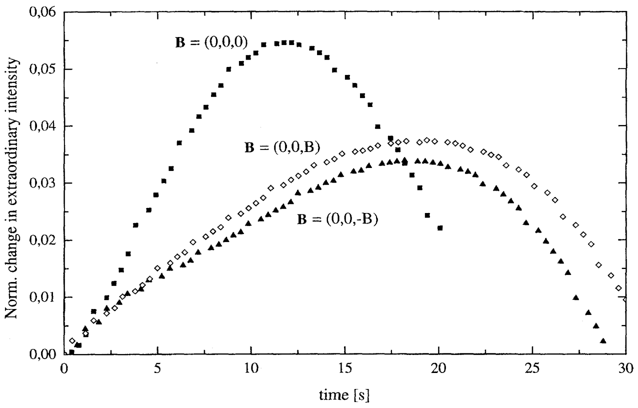

FIG. 2. Normalized change in the measured intensity of the extraordinary beam as a function of recording time, with applied magnetic field as parameter. The magnitude of the magnetic field is $B=0.5$ T. Same external angle of incidence for the two writing beams, 41.1°. Total intensity is 360 mW/cm^2 and a modulation of 0.31.

using an extraordinarily polarized and Bragg-matched HeNe-probe beam at 632.8 nm. In this setup the ordinary and extraordinary writing beams were symmetrically incident at an external angular separation of $2\Theta = 68.6°$, yielding a grating period of 0.45 μm. The magnitude of the magnetic field was 0.4 T. The results are shown in Fig. 3. Here, the measured diffraction efficiency is reduced for both directions of the applied magnetic field. The largest reduction from 4.0 % with no magnetic field to 1.0 % is found for $\mathbf{B}=(0,0,-B)$, which was also the case for the energy coupling.

From the previous discussion, the magnetic field influence on the spatially oscillating current given in Eq. (1) gives an expected linear dependence on the magnetic field, for both the energy coupling and the diffraction efficiency. Therefore the change in these quantities, when applying a magnetic field, is an uneven function of B. This is in contrast to the experimental results which show a reduction for both directions of the magnetic field $\pm B$. Thus, an additional mechanism independent of the sign of the magnetic field must be present in order to cause the reduction. Such an effect could be taken into account by including the second-order term in the expansion of the magnetophotovoltaic current in powers of the magnetic field.[4] In the derivation of Eq. (1) only the linear term, representing the photo-Hall effect, is included. A possible explanation may be found in the magnetic field dependence of the photoconductivity. Measurements[4] have shown a decrease in the photoconductivity of LiNbO$_3$:Fe crystals independent of the direction of the magnetic field. However, this would increase the space-charge field and yield an increase in both the energy coupling and the diffraction efficiency.

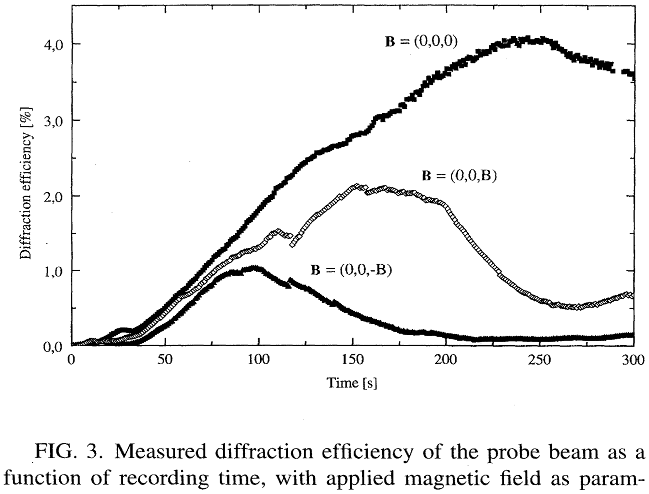

FIG. 3. Measured diffraction efficiency of the probe beam as a function of recording time, with applied magnetic field as parameter. The magnitude of the magnetic field is $B=0.4$ T. Symmetrically incident writing beams at an external half-angle of 34.3°. Total intensity is 390 mW/cm^2 and a modulation of 0.43.

Since the magnetic field is applied perpendicular to the direction of propagation of both interacting waves, the Faraday effect, i.e., the rotation of the state of polarization of the interacting waves in the presence of a magnetic field, is believed to have no influence on the measurements. This is confirmed by a measurement of the intensity of a single transmitted extraordinary beam, which showed no change with and without the applied magnetic field.

Based on physical reasoning, it is anticipated that the influence of the magnetic field becomes possible provided that the mean free path of the nonequilibrium electron is comparable with the grating period. This implies that if the grating spacing is much larger, no effect is to be expected. In the experiments presented, we have used gratings with periods of 5.3 and 0.45 μm, which both are longer than the mean free path in LiNbO$_3$:Fe, determined in independent measurements[9] to be $l_0 \simeq 10^{2}-10^{3}$ Å . In the configurations used here, the obtainable grating periods range from 0.25 to 5.5 μm. The smallest period of 0.25 μm can be obtained by using a reflection grating configuration, where an enhancement of the effect could be expected. However, it is not possible to give any quantitative conclusion on the mean free path of the nonthermalized electrons from these measurements.

IV. CONCLUSION

In conclusion, experimental measurements of the influence of an externally applied magnetic field on the holographic grating formation in LiNbO$_3$:Fe are presented. The diffraction efficiency and energy coupling are seen to be significantly changed by the presence of the magnetic field. Moreover, the observed change is dependent on the direction of the magnetic field. This was to be expected since the photovoltaic effect is of a tensorial nature. The physics of the effect is believed to be based on the interaction of the magnetic field with the nonthermalized electrons, which has a very high mobility. These are responsible for the photovoltaic current, which in the used vectorial interaction scheme, is solely responsible for the grating formation. A strong effect is observed for grating periods slightly longer than the mean free path of these electrons.

Using a phenomenological theory for the photovoltaic current that includes the photo-Hall effect, we expect a linear dependence on the magnetic field. The experimental results reveal a more complicated dependence, calling for an improved description. We believe this can be done by including second-order terms in the expansion of the magnetophotovoltaic current in powers of the magnetic field. This will be the subject of the future work, which will be based on more elaborate measurements. It is expected that the study of magnetophotovoltaics will give a better understanding of the fundamental transport processes.

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