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Infrared sensitive liquid crystal photorefractive hybrid cell with semiconductor substrates

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Abstract Refractive index grating recording is achieved in the infrared in a nematic liquid crystal placed between semiconductor CdTe substrates. Light-induced space-charge field created in photorefractive semiconductor substrates varies an alignment of liquid crystal molecules inhomogeneously in space forming a spatial modulation of the refractive index in liquid crystal. Two-beam coupling is studied in the hybrid cell, in which a gain factor $\Gamma = 16\ \text{cm}^{-1}$ is achieved in the layer of liquid crystal of the sandwich.

1 Introduction

When a photorefractive crystal is illuminated by an interference pattern from two crossing beams, free carriers are photogenerated from deep centers in the crystal. These carriers migrate through diffusion to the dark areas where they are captured by traps, creating a spatially modulated space-charge field. This space-charge field results in a refractive index modulation in electrooptic crystals through the Pockels effect. For pure diffusion, the resulting light-induced refractive index grating is $\pi/2$ shifted with respect to the interference fringes. Unidirectional energy transfer between the two recording beams, i.e., amplification of the one beam at the expense of another, appears as a result of diffraction from such grating [1].

It has recently been demonstrated that hybrid liquid crystal (LC) cells fabricated as a sandwich with LC between ferroelectric photorefractive crystalline windows may demonstrate efficient nonlinear coupling of the optical waves with coupling constants much larger than the coupling constants of inorganic photorefractives [2–5]. The space-charge field created in photorefractive substrates affects the alignment of the LC molecules via flexoelectric effect. The large gain factor has been achieved in such hybrid cells with ferroelectric crystal windows in the visible range of the spectrum [3–5].

Semiconductor crystals like GaAs, CdTe, and InP exhibit the photorefractive effect in the infrared. These materials are therefore of interest for applications with the semiconductor lasers emitting in the infrared and with optical fibers possessing the smallest dispersion and minimum losses in this region of spectrum. An important advantage of photorefractive semiconductors is their fast response time, which is much shorter than that of the wide-band-gap ferroelectric crystals. At the same time, semiconductors exhibit smaller coupling constants compared with ferroelectric photorefractives because of their much smaller electrooptic coefficients. These relatively small coupling constants impose some limitations for practical applications.

The LC refractive index modulation may be influenced by the space-charge field created in a photorefractive semiconductor with a low intensity infrared light in the same way as it happens with ferroelectric crystals. It should be emphasized that the relatively small electrooptic constants of semiconductors are not important because the space-charge field drives the nonlinearity of LC layer. This space-charge field is proportional to the so-called diffusion field. Thus, implementation of a hybrid cell with semiconductor windows may

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combine synergistically the high sensitivity of photorefractive inorganic semiconductors in the infrared and high nonlinearity of LCs, creating a new device with the enhanced response in the infrared (a spectral region that LCs are not typically sensitive).

In this work, two-beam coupling is demonstrated for a hybrid cell with nematic LC placed between CdTe windows. The dependence of the gain factor on grating spacing is studied at a wavelength of $\lambda = 1.064$ µm. A gain factor up to 16 cm^{-1} is achieved in the LC layer of the hybrid cell.

2 Cell fabricating and experimental set-up

The cell windows are fabricated from CdTe:Ge photorefractive crystals grown in Chernivtsy National University, Ukraine, by Zakharuk and Rarenko. The windows' dimensions are 7 mm × 4 mm × 1 mm along the $[1\bar{1}0]$, $[11\bar{2}]$, and [111] crystallographic axes, respectively. The largest faces (7 mm × 4 mm), parallel to (111) plane, serve as input/output windows of the LC cell shown in Fig. 1a. The nylon multipolymer Elvamide 8023R (DuPont, USA) is used for inducing nearly planar alignment of the liquid crystal. An anhydrous methanol solution of elvamide is prepared with weight concentration of 0.125% as described elsewhere [5]. The elvamide films are deposited on one side of photorefractive crystal substrates by spin-coating method (4000 rpm, 30 s), and subsequently baked (50 °C, 30 min) and rubbed along the longest dimension of the windows. The rubbing directions in sandwich cell are antiparallel as it is schematically shown in Fig. 1a by dashed arrows. To minimize the gain from photorefractive windows, the orientation of the CdTe crystals for both substrates in the cell is chosen in such a way that the two-beam coupling have different directions in both windows thus ensuring partial compensation of the coupling in the two photorefractive substrates. The LC layer thickness, $d \approx 10$ µm, is imposed by glass spherical spacers placed between the substrates.

Experimental set-up is shown schematically in Fig. 1b. The single-mode single-frequency diode-pumped Nd^{3+}: YAG laser emitting at 1.064 µm is used as a light source. The output laser radiation, which is polarized in the plane of drawing, is divided by a beam splitter into two beams I_{S0} and I_{P0} with the intensity ratio $\beta \approx 1:2.5$. Both beams have Gaussian intensity distribution with about 2 mm full-width at half maximum on the cell input and with total light power of the order of 200 mW. The beams impinge upon the sample at an angle $2\theta \approx 65°$, thus recording a grating inside photorefractive crystals with grating spacing $\Lambda \approx 1$ µm.

The temporal variations of the grating build-up are measured in the experiment. The phase electrooptic modulator EOM is used to control the grating recording and erasure. The packets of sinusoidal oscillations introduce high

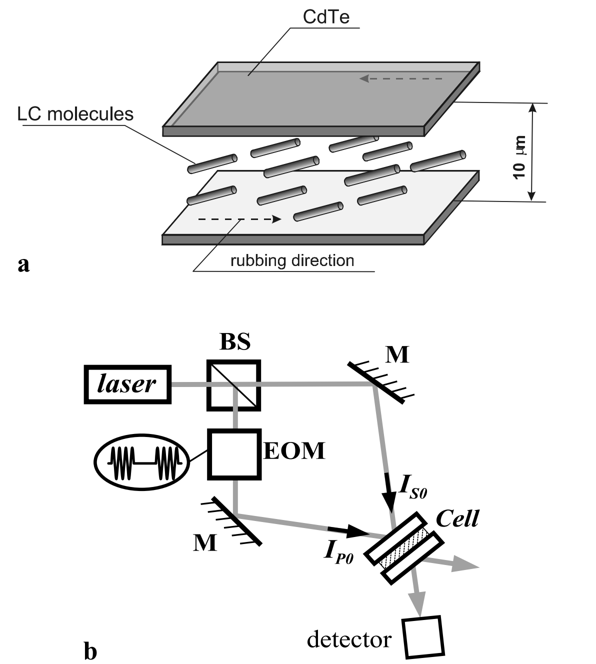

Fig. 1 (**a**) Schematic representation of the hybrid cell with LC placed between two CdTe windows; (**b**) experimental set-up for study of the two-beam coupling

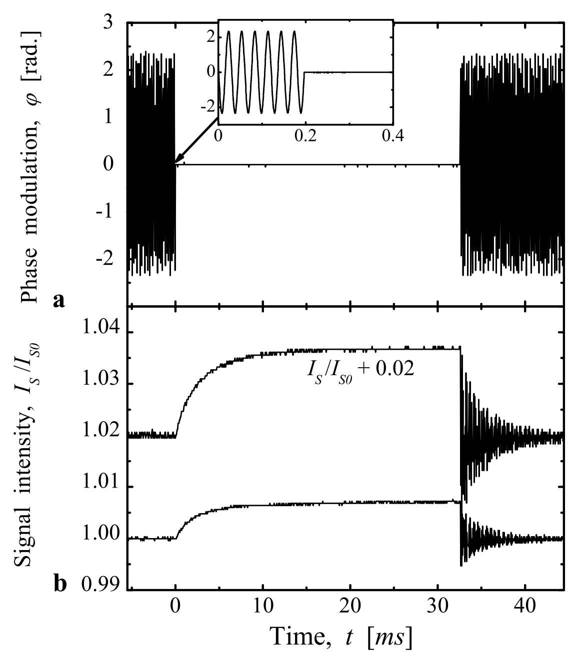

Fig. 2 (**a**) Phase modulation of the pump beam; (**b**) temporal behavior of the signal beam intensity for the empty cell (*lower trace*) and for the cell filled with LCs (*upper trace*)

frequency phase modulation $\varphi(t) = \Delta\varphi \sin(2\pi f t)$ in the pump beam, as it is shown in Fig. 2a. The frequency of the sinusoidal modulation $f = 32.5$ kHz is much higher than the reciprocal relaxation time of the CdTe windows $2\pi f \gg 1/\tau_{SC}$ for the intensities of the recording beams and spatial frequencies of the gratings used in our experiments. The duration of the pulse packets and the interval between them are chosen to be the same ($T_p/2$) and equal $T_p/2 = 32.5$ ms. This value is much larger than the relaxation time of the CdTe substrates that allows for complete grating erasure during the packets of fast sinusoidal oscillations and grating recording to the steady state between the packets. The amplitude of the phase modulation $\Delta\varphi \approx 2.4$ rad corresponds to the first zero of the zero order Bessel function $J_0(\Delta\varphi) \approx 0$. For the fast sinusoidal modulation with such an amplitude and averaging interval much longer than the reciprocal frequency of the oscillation, the averaged contrast of the recording fringes pattern is zero [6]. Thus, a complete erasure of the dynamic grating is ensured when the sinusoidal modulation is applied. Such modulation technique of the grating recording/erasure allows for more accurate measurement of the signal beam intensity change in two-beam coupling experiment as compared to traditional procedure of blocking/unblocking of the pump beam.

Environmental perturbations do not usually permit efficient recording of stationary holograms or dynamic holograms in materials with slow response times. Active stabilization techniques [7–9] have been used since the 1960s for compensation of such perturbations. In particular, self-stabilized recording of dynamic gratings in photorefractive crystals has been intensively studied [10, 11]. However, the response time of our cell τ_{SC} is few milliseconds, allowing the grating to self-adapt for all instabilities and no techniques for active or self-stabilization are necessary in our system.

3 Experimental results and discussion

First, the response of the empty cell is studied. The temporal variation of the signal beam intensity behind the crystal is shown in Fig. 2b, where the lower trace represents grating build-up cycle measured at grating spacing $\Lambda = 1$ μm. The beam power increases nearly exponentially with a time constant $\tau_{SC} \approx 3$ ms. The intensity increment is about $(I_S - I_{S0})/I_{S0} \approx 0.7\%$, where I_{S0} and I_S are the signal beam intensity behind the crystal with and without the fast phase modulation, respectively. Similar dependences are measured at different grating spacings and the intensity change I_S/I_{S0} is evaluated. The cell was then filled with the LC and the temporal variations of intensity are measured at the same grating spacings as for the empty cell. The upper trace in Fig. 2b represents the dependence measured for the cell with LC at $\Lambda = 1$ μm. The curve is displaced by 0.02 for better representation. It should be noted that the cell was filled with LC without changing its position in the experimental set-up; the measurements for the empty and filled cell are performed at exactly the same experimental conditions. The intensity for the filled cell grows in time with nearly the same rate as for the empty cell but the intensity increment increases up to $(I_S - I_{S0})/I_{S0} \approx 1.7\%$. Therefore, the space-charge grating recorded in CdTe windows gives rise to the refractive index modulation in the LC layer, which increases the overall nonlinearity of the hybrid device as compared with the empty cell. The closeness of the time constants of the beam coupling in the empty and filled cell demonstrates that it is the CdTe substrates that determine the response time of the cell. The response of LC is faster so that it follows the space-charge variation in CdTe with no additional delay.

We assume that the coupling constant for the cell filled with LC, Γd_F, is determined by two contributions to the two-beam coupling, contribution from the windows, Γd_W, and contribution from LC Γd_{LC}. The steady state intensity of the signal beam can be written as

$$I_S = I_{S0} \exp(\Gamma d_F) = I_{S0} \exp(\Gamma d_W + \Gamma d_{\mathrm{LC}}). \quad (1)$$

Therefore, for evaluation of the LC contribution to the two-beam coupling the coupling strength of the empty cell $\Gamma d_W = \ln(I_S/I_{S0})_W$ is subtracted from the coupling strength of the filled cell $\ln(I_S/I_{S0})_F$ measured under the same experimental conditions:

$$\Gamma d_{\mathrm{LC}} = \Gamma d_F - \Gamma d_W. \quad (2)$$

This contribution is shown in Fig. 3 as a function of grating spacing. The line is shown as a guide to the eye. The largest value $\Gamma d_{\mathrm{LC}} \approx 0.016$ is achieved at the smallest grating spacing $\Lambda = 0.8$ μm, which can be realized in the present cell with 1064 nm wavelength because of geometrical reason of beams intersection. The large space-charge field is reached in CdTe at this smallest period (over the range measured) and this field penetrates in the LC layer. One could expect a decrease of the LC coupling strength for smaller periods because the modulation of LC orientation diminishes with decrease of the grating period due to elastic forces.

The large space-charge field is created at smallest grating spacing in photorefractive semiconductors with a large

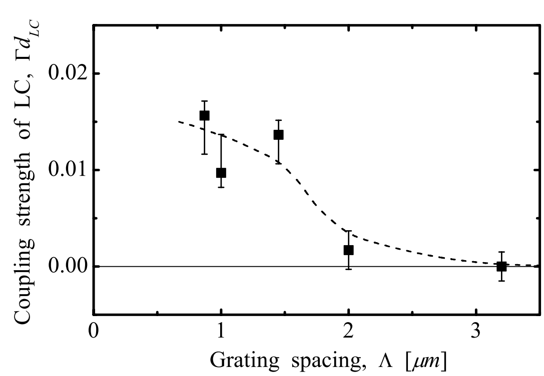

Fig. 3 Grating spacing dependence of the coupling strength of the LC layer

effective trap density when the space-charge is not limited by traps concentration [12, 13]. The space-charge field E_{SC} for transmission gratings in this case is nearly proportional to the diffusion field, E_D, which in turn is inversely proportional to the grating spacing: $E_{SC} \sim E_D \sim (2\pi/\Lambda)(k_B T/e)$, where k_B is the Boltzmann constant, T is the absolute temperature, and e is the electron charge. That is why the strength of the LC grating vanishes to zero at large grating spacing $\Lambda \geq 2$ μm where the space-charge field becomes small. The two-beam coupling gain factor $\Gamma = 16\ \text{cm}^{-1}$ corresponds to the maximum value $\Gamma d_{\text{LC}} \approx 0.016$ achieved with a 10 μm thick cell at the smallest grating spacing $\Lambda = 0.8$ μm. This gain factor is comparable with gain factors reported earlier for stimulated orientation light scattering in nematic LC [14] reached at much higher intensities of 1.064 μm radiation. It exceeds also 20 times the best known value for transmission gratings in all infrared sensitive semiconductors, which has been achieved in CdTe [13].

4 Conclusions

Our experiments prove that hybrids with photorefractive semiconductor substrates allow operation in the infrared. The large two-beam coupling gain factor is reached in the LC layer of the hybrid cell. Larger coupling constants have been reached in the visible range of spectrum for hybrid cells with ferroelectric windows [3, 5]. The reduced coupling constant Γ for semiconductor hybrids can be explained in part by the fact that the coupling strength $\Gamma = 4\pi\Delta n/\lambda$ is inversely proportional to the wavelength for a fixed refractive index change. That is why the response of any medium in the infrared is reduced when compared with response of the media with similar characteristics in the visible region of the spectrum. Another reason for reduced gain is related to nonoptimized pretilt and splay of the LC. It is known that pretilt of nematic LC is crucially important for the value of coupling constant of LC in the hybrid cell [4, 15]. The ferroelectric windows ensure larger pretilt than that obtained with isotropic materials, which semiconductors are, using identical fabrication of the cell [9]. It has been shown also that doping of LC with nanoparticles [16] can increase significantly the coupling constant of hybrids. Therefore, optimization of the fabrication techniques, e.g., rubbing procedure, which improves the pretilt of nematic LC between isotropic semiconductor windows, and dopant optimization should increase considerably the response of the CdTe hybrid. At the same time, it should be noted that the demonstration of operation of the semiconductor-LC hybrid cell in itself with the gain factor in the LC layer exceeding typical values for photorefractive semiconductors more than one order of magnitude can bring new optical nonlinear device with large coupling strength in the infrared, a region where LCs are typically not sensitive.

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