Observation of self-diffraction and refractive-index gratings induced in a glass by mutually coherent optical fields E_ω and $E_{2\omega}$

M. K. Balakirev and V. A. Smirnov
Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

(Submitted 24 February 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **61**, No. 7, 537–540 (10 April 1995)

A self-diffraction and the formation of refractive-index gratings have been observed experimentally in a glass subjected to mutually coherent optical fields at the fundamental frequency and second harmonic of a laser. The properties of these gratings have been studied as a function of the intensity and polarization of the fields and the phase shift between them. These properties can be explained satisfactorily by a model of a coherent photovoltaic effect. © *1995 American Institute of Physics.*

Illumination of an isotropic medium by mutually coherent optical fields with a nonzero average cubic field may induce a direct current in the medium.[1–4] This is the coherent photovoltaic effect. The spatial separation of charges caused by the coherent photovoltaic current **J** in the case of an inhomogeneous medium (or in the case of nonuniform illumination) should generate an electric field $\mathbf{E}_0$ and, correspondingly, a reversible optical anisotropy of the medium. An optical anisotropy of this type would presumably[5] cause an effective second-order polarizability $\chi^{(2)}$, which arises in glasses upon sufficiently prolonged illumination[6–8] (although there are other possible causes of a $\chi^{(2)}$; Ref. 9). However, an electric field should generate an anisotropy of the refractive index in an isotropic medium also, because of the quadratic Kerr effect. An effective diffraction grating may form in the glass, because of the spatial periodicity of the field $\mathbf{E}_0$ in the beam-intersection region. A "self-diffraction" of the beams illuminating a medium should thus be seen when the optical anisotropy is generated. One should also see a specific scattering of light over the lifetime of the grating. Such effects have not been discussed in the literature of which we are aware. Experiments with diffraction gratings make possible an independent study of the properties and nature of the optical anisotropy that arises. In particular, it becomes possible to establish the relationship with the models developed in Refs. 1–4.

In this letter we are reporting first results on the observation of self-diffraction and a study of certain properties of photoinduced diffraction gratings in a glass. We focus on the behavior of the diffraction efficiency as a function of the relative orientation of the polarizations of the optical fields, the phase shift between these fields, $\Delta\phi$, their intensities, and the polarization of the diffracting beam.

The test sample in our experiments was a type PM-40 glass plate (type K-8 glass). In the experiments, two parallel beams, at the fundamental frequency and the second

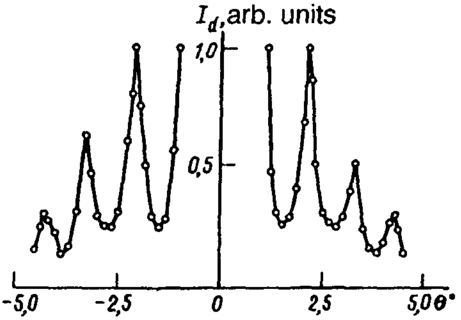

FIG. 1. Angular distribution of the intensity of the diffracted fundamental-frequency light in the case $\theta_0 = 1°$.

harmonic of a neodymium laser, were focused by a lens into the sample in such a way that the beam waists lay in the region in which the beams intersected. The angle between the beams, θ_0, was varied by displacing the beams incident on the lens. The beam polarizations $\mathbf{e}_1$ ($\lambda_1 = 1.079\ \mu$m) and $\mathbf{e}_2$ ($\lambda_2 = 0.539\ \mu$m) were varied with respect to each other and also with respect to the plane in which the beams were brought together, with the help of some half-wave plates. A phase-shifting element after the doubler made it possible to continuously vary the phase between the fields at a maximum rate of π rad/s. The length of the fundamental-frequency pulses was 15 ns, the maximum energy in the pulse was 18 mJ, the maximum intensity at the waist was $\sim 10^9$ W/cm^2, and the repetition frequency was 12.5 Hz. The transmitted and diffracted light was detected by a photomultiplier in the far zone. The sensitivity of the measurement system was $\sim 10^{-9}$ J/pulse at the wavelength 1.079 μm in a 300-MHz band.

The measurement cycle consisted of two parts. The sample was first illuminated with both beams, with the selected orientations of $\mathbf{e}_1$ and $\mathbf{e}_2$ (a grating was written). During this writing process, we detected primarily the intensity of the diffracted light of the fundamental frequency at its maximum. An upper limit (50 min) was set on the writing time by saturation of the intensity of the light being diffracted. In the next step (readout), one of the beams was blocked, and the diffraction of the other beam was detected. The polarization of the beam incident on the sample was either held the same or rotated $\pi/2$ around its axis.

The experimental results can be summarized as follows.

1. During the writing we observe a diffraction of the writing beams which intensifies as time elapses. At small angles θ_0 the angular distribution of the diffraction is similar to that of diffraction by an aperture (Fig. 1). As θ_0 is increased, the diffraction efficiency for the fundamental frequency increases by nearly two orders of magnitude near a certain angle $\theta_0 = \theta_B$ ($\theta_B \simeq 4.8°$ in the experiments; Fig. 2) and then begins to decrease again. At $\theta_0 = \theta_B$, the diffraction efficiency $\eta = I_d/I_0$ is on the order of 10^{-3} (here I_0 and I_d are the intensities of the transmitted and diffracted beams, respectively).

2. Changing the phase difference between the fields when we go from one writing cycle to another alters neither the intensity nor pattern of the diffraction. However, if the

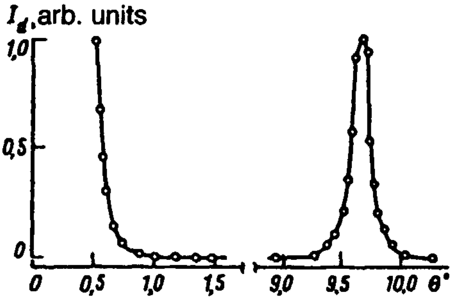

FIG. 2. Angular distribution of the intensity of the fundamental-frequency light diffracted in the case $\theta_0 = 4.8°$.

phase difference is modulated at the maximum frequency during the writing, the diffraction efficiency drops to zero. Observation during a very slow change in phase reveals a periodic modulation of the intensity of the diffracted beam.

3. The characteristic dark lifetime of the gratings is of the order of 40 min.

4. The diffraction efficiency depends strongly on the relative orientation of $\mathbf{e}_1$ and $\mathbf{e}_2$ of the writing beams and also on the orientation $\mathbf{e}_{1S}$ of the readout beam. If $\mathbf{e}_1$ and $\mathbf{e}_2$ lie in the plane in which the beams converge ($\mathbf{n}$ is the normal to this plane), and if the polarizations of the readout and writing beams are identical, then η is at a maximum. At $\theta_0 = \theta_B$ it reaches a value of 3×10^{-3}. If, on the other hand, the readout is carried out in the orientation $\mathbf{e}_{1S} \| \mathbf{n}$, the value of η decreases by a factor of 300. In the case of writing with mutually orthogonal field polarizations, under the conditions $\mathbf{e}_1 \| \mathbf{n}$ and $\mathbf{e}_2 \perp \mathbf{n}$, and if the readout is carried out with a beam with $\mathbf{e}_{1S} \perp \mathbf{n}$, the efficiency η decreases by nearly an order of magnitude ($\eta = 2.5 \times 10^{-4}$). In the case $\mathbf{e}_{1S} \| \mathbf{n}$, in contrast, the efficiency η decreases again by a factor of 60. Finally, for a writing orientation $\mathbf{e}_2 \| \mathbf{e}_1 \| \mathbf{n}$ the diffraction level is low (right at the resolution of our apparatus).

5. Figure 3 shows the intensity of the diffracted beam as a function of the total

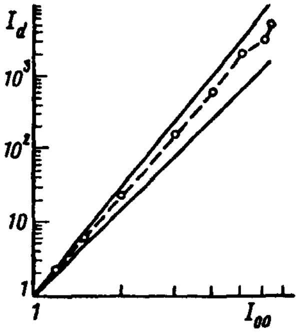

FIG. 3.

intensity of the fundamental-frequency beam incident on the doubler. We see that the dependence is approximately a power law with a power between 4 and 5.

All the results reported here, except for the kinetics of the writing and relaxation of the diffraction gratings, agree satisfactorily with a model for the coherent photovoltaic effect,[2-4] if one assumes that a long-lived field $\mathbf{E}_0$ proportional to the coherent photovoltaic current $\mathbf{J}$ arises in the sample as a result of charge separation:

$$\mathbf{E}_0 \sim \mathbf{J} = E_1^2 E_2 (\sigma_1 \mathbf{e}_1 (\mathbf{e}_1 \mathbf{e}_2) + \sigma_2 \mathbf{e}_2) \cos(2\mathbf{k}_1 \mathbf{r} + \mathbf{k}_2 \mathbf{r} + \Delta\phi). \tag{1}$$

The subscripts 1 and 2 specify the fundamental frequency and the second harmonic, $\mathbf{k}_{1,2}$ are the wave vectors of the waves in the medium, $E_{1,2}$ are the beam envelopes, and $\sigma_{1,2}$ are the conductivities.

The spatial structure of the altered refractive index, Δn, is proportional to the square of the field, $\mathbf{E}_0$. It contains a periodic component (the grating) and a smoothly varying component (the aperture). Each of these components is modulated spatially by the square of the field amplitude: $(E_2 E_1^2)^2$. Because of dispersion of the refractive index ($n_1 = 1.506$, $n_2 = 1.518$), the front of the grating "unrolls" until it is nearly perpendicular to $\mathbf{k}$ at small values of the angle θ_0. Accordingly, the aperture component dominates the diffraction. With increasing θ_0, the grating unrolls in such a way that Bragg diffraction conditions are satisfied for the angle $\theta_0 = \theta_B$. The calculated value $\theta_B = 4.8°$ agrees satisfactorily with experiment.

The polarization dependence can be explained as follows. In the case of writing with $\mathbf{e}_2 \| \mathbf{n}$, the coherent photovoltaic current flows in a direction parallel to the planes of the grating, and charge separation occurs at the periphery of the region in which the beams intersect; i.e., there is no clearly expressed diffraction grating. If the writing is carried out with $\mathbf{e}_1 \| \mathbf{n}$ and $\mathbf{e}_2 \perp \mathbf{n}$, the contribution of the first term in (1) drops out of the picture completely, and the efficiency of the diffraction grating decreases substantially. During readout, the beam which is the "extraordinary" beam with respect to the system, i.e., $\mathbf{e}_{1s} \| \mathbf{E}_0$, is evidently diffracted most efficiently.

It follows from (1) that a change $\Delta\phi$ simply shifts the phase of the diffraction grating in space. Accordingly, for any constant value of $\Delta\phi$ the diffraction efficiency is the same. In contrast, if $\Delta\phi$ is modulated in time with a period much shorter than the anisotropy buildup time, there is an averaging of $\mathbf{E}_0$, and a diffraction grating does not form. This is what was observed experimentally.

In the model which we have adopted, we evidently have $I_d \sim \Delta n I_0 \sim (E_1^2 E_2)^2 I_0 \sim E_2^2 I_0^3$. Experimentally, we find $E_2 \sim I_0$, so we must have $I_d \sim I_0^5$. This relation is close to that which is observed (Fig. 3).

We believe that the coherent photovoltaic effect is responsible for the effects observed in this study. Unfortunately, we do not have an unambiguous explanation of the mechanisms for the writing and relaxation of the diffraction grating, which are associated with the kinetics of the space charge in the glass. These topics require further research.

We wish to thank M. V. Éntin for useful discussions.

This study was supported by the Russian Fund for Fundamental Research (Grant 93-02-15717).

[1] N. B. Baranova and B. Ya. Zel'dovich, JETP Lett. **45**, 717 (1987).
[2] M. V. Éntin, Fiz. Tekh. Poluprovodn. **23**, 1066 (1987) [Sov. Phys. Semicond. **23**, 664 (1987)].
[3] É. M. Baskin and M. V. Éntin, JETP Lett. **48**, 601 (1988).
[4] V. B. Sulimov, Zh. Éksp. Teor. Fiz. **101**, 1749 (1992) [Sov. Phys. JETP **74**, 932 (1992)].
[5] E. M. Dianov, P. G. Kazanskiĭ, and D. Yu. Stepanov, Kvant. Élektron. (Moscow) **16**, 887 (1989) [Sov. J. Quantum Electron. **19**, 575 (1989)].
[6] R. N. Stolen and H. Tom, Opt. Lett. **12**, 585 (1987).
[7] V. M. Chyurikov, Yu. E. Kapitzky, V. M. Lukyanov, and B. Ja. Zeldovich, Sov. Lightwave Commun. **1**, 389 (1991).
[8] B. Ya. Zel'dovich, Yu. E. Kapitskiĭ, and V. M. Churikov, JETP Lett. **53**, 78 (1991).
[9] E. M. Dianov, A. M. Prokhorov, V. O. Sokolov, and V. B. Sulimov, JETP Lett. **50**, 13 (1989).

Translated by D. Parsons