Induced $\chi^{(2)}$ gratings in glasses

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Second-harmonic generation in a glass has been observed. The angular selectivity of the $\chi^{(2)}$ grating has been measured. The decay and restoration of gratings have been observed. The conversion efficiency is 10^{-6} over a distance of 60 μm at an average optical power of 240 mW.

Osterberg and Margulis[1] have observed second-harmonic generation by optical fibers.[1] In a preliminary step in their experiments, light from a high-power picosecond Nd:YAG laser was passed through the fiber for several hours. Stolen and Tom[2] accelerated this preparation process by passing the second harmonic along with the fundamental frequency (simultaneously) through a fiber. Baranova and Zel'dovich[3] have proposed a phenomenological theory for the effect. Kapitsky and Zel'dovich measured the angular selectivity of such gratings in Ref. 4 and achieved phase conjugation at $\chi^{(2)}$ gratings in Ref. 5. Lawandy and Selker achieved second-harmonic generation in quartz from which optical fibers are fabricated.[6] The quartz itself has waveguiding properties.

Questions which have not been resolved are whether this induced second-harmonic generation is an exclusive property of amorphous quartz and whether waveguiding properties exist in the medium.

To observe the $\chi^{(2)}$ gratings in the glass, we used an Nd:YAG laser with active mode locking and active Q switching. The pulse length was 100 ps, the repetition frequency was 6 kHz, and there were about 30 pulses in a train. The IR light was partially converted into the second harmonic in a KTP crystal. Both beams were focused in the glass sample by a lens with a focal length of 9 mm. The recording step consisted of the simultaneous exposure of the sample to the IR and second-harmonic light for several minutes. The grating was read out by IR light; the second harmonic was removed by a filter for this purpose. The second-harmonic generation could be observed with the naked eye; the second-harmonic power was measured with a photomultiplier. In some preliminary experiments we used a PM-15 glass plate (type K-8 glass); the average power of the second-harmonic signal was 10^{-10}–10^{-9} W at an average IR power of 240 mW. Figure 1 shows the angular selectivity of the $\chi^{(2)}$ gratings in this glass. This curve is substantially broader than that in fibers (cf. Ref. 4), confirming that the mode composition of the fiber influences the structure of the $\chi^{(2)}$ gratings in fibers. As time elapses, the $\chi^{(2)}$ gratings in the glasses fade away. Figure 2 shows the time evolution of the power of the second-harmonic signal for two cases: with and without the IR light. The slower disappearance of the $\chi^{(2)}$ grating in the presence of the IR light is evidence of a partial supplemental writing of the grating, so the written grating and the interference pattern performing the writing are not out of phase.

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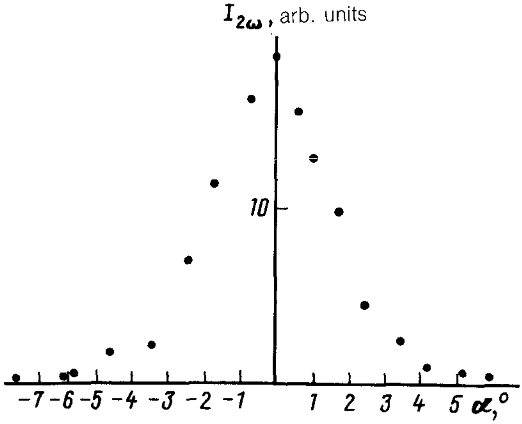

FIG. 1. Intensity of the second-harmonic signal as a function of the angle of incidence of the writing light on the grating.

We selected a glass sample in which the intensity of the second-harmonic signal was higher by a factor of 10^3 than in the first case, under the same conditions. This was type BS-7 glass from the All-Union State Standards 9411-81 set of filters. Figure 3 illustrates the disappearance of the grating in this glass: during exposure to the IR light, during exposure to the second-harmonic light, and without exposure to light. The intensity of the erasing light was the same as that during the writing. Interestingly, all the erasure curves, in both samples, have a fast stage (from $t = 0$ to point A) and a slow one (from point A to $t = \infty$). This effect is seen particularly clearly in logarithmic scale (not shown in Fig. 3). This behavior is characteristic of the relaxa-

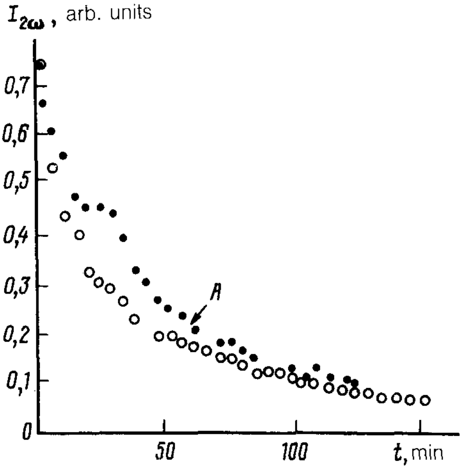

FIG. 2. Time evolution of the second-harmonic signal for K-8 glass. ●—In the presence of IR light; ○—in the absence of IR light.

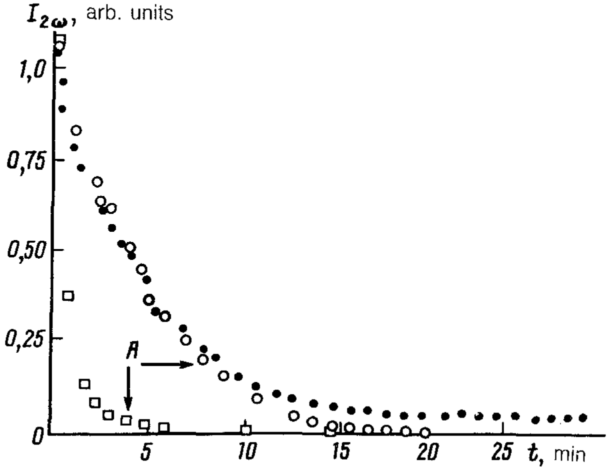

FIG. 3. Time evolution of the intensity of the second-harmonic signal for BS-7 glass. ●—In the presence of IR light; □—in the presence of second-harmonic light; ○—without exposure to light.

tion of a photoconductivity in an insulator containing attachment centers.[7] One might suggest that a spatial separation of charge is undergoing relaxation. We would then also find an explanation for the rapid disappearance of the grating during exposure to the second-harmonic light. The second-harmonic photons tear electrons away from the trapping centers. The electrons drift in the direction of the positive charge and recombine.

We thus believe that the following model can be used. A standing interference pattern resulting from an interference of the fields E_ω^2 and $E_{2\omega}$ exists in the medium. The medium is a wide-band insulator with impurity centers and attachment centers. We assume for definiteness that donor impurities are present. The valence electrons are then knocked away from the donors into the conduction band by two IR photons and a single second-harmonic photon. This ejection process depends on the phase relation between E_ω^2 and $E_{2\omega}$ (Ref. 8). The electrons which have been knocked out of the donors have a momentum in the direction of the nonzero field $\langle E^3 \rangle$ (Ref. 9). The electrons move until they lose their velocity as a result of collisions with lattice defects and reach trapping centers. A spatial separation of charge thus occurs giving rise to local static fields. The spatial structure of these fields is such that they automatically satisfy a synchronization condition. The second harmonic is generated at the $\chi^{(3)}$ nonlinearity of the following polarization:

$$P_i^{(3)}(2\omega) = \chi_{ijkl}^{(3)}(-2\omega, \omega, \omega, 0) E_j(\omega) E_k(\omega) E_l(0).$$

As was pointed out in Ref. 10, this mechanism requires the presence of a static field of 10^4–10^5 W/cm. Such fields can exist in insulators with a low photoconductivity. During exposure of $LiNbO_3$ to light in the blue-green region, for example, the photovoltaic voltage can reach such levels.[11]

A surprising result was that after the erasure of the $\chi^{(2)}$ grating by the second-harmonic light, or after erasure by IR light orthogonal to the writing light, it was possible to restore the grating and to read it without a second-harmonic seed. In other words, a rewriting occurred in the absence of a second-harmonic seed.

A grating was written by orthogonal fields E_ω and $E_{2\omega}$. In this case the efficiency of the second-harmonic generation was lower by a factor of 16 than in writing by parallel beams, if the readout was performed by an IR wave polarized parallel to the second-harmonic writing wave. It was smaller by a factor of $16\times9(\approx140)$ during readout by an IR wave with the perpendicular polarization. These results support the suggestions in Ref. 11.

In summary, it has been demonstrated experimentally that it is possible to write $\chi^{(2)}$ gratings in a glass at an efficiency which varies from one material to another by a factor $\sim10^3$.

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