Anomalous behavior of the coefficient of second-harmonic conversion during the readout and writing of $\chi^{(2)}$ gratings

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An abrupt jump in the efficiency of second-harmonic conversion as a function of the intensity of the incident IR light has been observed during the readout and writing of $\chi^{(2)}$ gratings in several multicomponent glasses. A possible mechanism for the effect is discussed.

Induced second-harmonic generation in optical fibers was discovered in 1986 by Osterberg and Margulis.[1] Lawandy and Selker[2] showed that this effect can also occur in the bulk germanium silicate preforms from which the fiber is made. Numerous studies (e.g., Ref. 3) have shown that the simultaneous application of optical fields $\mathbf{E}_\omega$ and $\mathbf{E}_{2\omega}$ to a centrally symmetric medium (a fiber or glass) lowers the symmetry, probably because a grating of a static field E_{st} is induced. As was shown in Ref. 4, we have in this case $\mathbf{E}_{\text{st}} \sim \langle \mathbf{E}(\mathbf{E}\cdot\mathbf{E})\rangle$, where $\mathbf{E}$ is the sum of the real fields $\mathbf{E}_\omega$ and $\mathbf{E}_{2\omega}$. As this field acts on the third-order nonlinear susceptibility $\chi^{(3)}$, it creates an effective $\chi^{(2)}$ grating, which automatically ensures a matching of the interaction. In addition, Lawandy and Selker[2] found that the intensity of the ω light must exceed a certain threshold in order to write an effective $\chi^{(2)}$ grating.

The nature of the induced second-harmonic generation has remained unclear. Furthermore, Dianov *et al.*[5] showed that the microscopic mechanism for the writing of the $\chi^{(2)}$ grating may differ from one type of glass to another. In order to reach an understanding of the nature of this effect, we believe it is important to determine how the intensity of the IR ω light affects the writing and the readout, since the ω light participates in both processes. In particular, the effect of the power of the incident IR light on the second-harmonic signal was studied in Ref. 6, among other places, for glasses doped with microscopic $\text{CdS}_x\text{Se}_{x-1}$ crystals and in Ref. 7 for germanium silicate fibers. In those experiments the dependence of the signal on the incident IR light was found to be quadratic. This result implies that the susceptibility $\chi^{(2)}$ induced in the glass is independent of the intensity of the readout IR light. In the present letter we discuss a similar experiment on the lead silicate glass ZhS-4, in which the mechanism for the effect appears to be slightly different.

We used a Nd:YAG laser with active Q switching and active mode locking. Half the average power was in 100-ps pulses. This light was partially doubled by KTP crystal, sent through a linear polarizer, and focused into the sample of ZhS-4 glass by a microscope objective. The maximum average power of the light transmitted through the glass was 900 mW. We first measured the conversion efficiency $\beta = I_{2\omega}/I_\omega^2$ as a function of the intensity of the IR readout light, I_ω; this intensity was varied with the

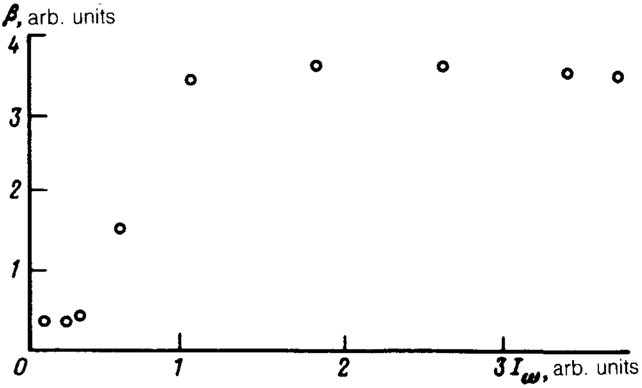

FIG. 1. Efficiency of the conversion into the second harmonic in ZhS-4 glass versus the intensity of the ω readout wave.

help of a $\lambda/2$ plate in front of the polarizer. For this purpose we wrote a grating to the point of saturation, using a fairly strong pump, so that the green signal was easily visible under natural room lighting. The result of the these measurements is shown in Fig. 1. The efficiency of the conversion into the second harmonic was constant when the IR light was fairly intense, but below a certain I_ω we observed an unexpected and sharp decrease in β. As I_ω was reduced further, the efficiency β became constant again, but at a level about an order of magnitude lower. This effect was unrelated to an erasure of the $\chi^{(2)}$ grating which had been observed previously,[8] since the perturbation of the grating during the readout was very brief. We checked to see whether the $\beta(I_\omega)$ dependence was highly accurately reversible as I_ω was varied from high values to low ones and back. We also carefully checked the linearity of the photodetector (an FÉU-127V photomultiplier) in the working interval. We observed the same dependence of the conversion coefficient, $\beta(I_\omega)$, in several other lead silicate glasses and also in an Al_2O_3- and CaO-based glass. The sharp decrease in the conversion efficiency was observed in all glasses in approximately the same value of I_ω, ~5 GW/cm^2 according to our estimates. The magnitude of the jump depended on the type of glass; it amounted to one or two orders of magnitude under given writing conditions.

Figure 2 shows a plot of $\zeta = I_{2\omega}/(I_\omega^w)^2$ versus I_ω^w, where I_ω^w is the intensity of the ω writing wave. The readout was carried out with the help of fixed IR light, the same for all points. The seed second harmonic was also fixed at 0.5 mW. The abrupt change in the grating efficiency $\zeta(I_\omega^w)$ is evidently due to the effects described above (Fig. 1). It may be analogous to the threshold observed in Ref. 2.

We believe that this result can be interpreted in the following way. As was mentioned earlier, we are probably dealing with second-harmonic generation in an induced electric field, so we have $\chi^{(2)}=\chi^{(3)}E_{\rm st}$, where the static field is $\mathbf{E}_{\rm st}\sim(\mathbf{E}\cdot\mathbf{E})\mathbf{E}\sim(\mathbf{E}_\omega^*)^2E_{2\omega}\exp(i\Delta kz)+$ c.c. In this model, the static field depends on

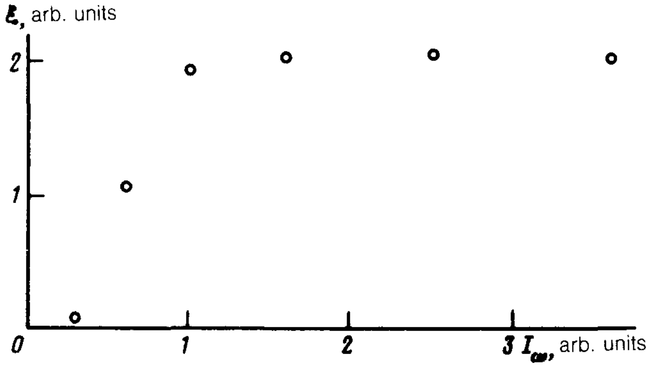

FIG. 2. Efficiency of the writing of the grating, $\zeta = I_{2\omega}/(I_\omega{}^{w})^2$, versus the intensity of the ω writing wave.

the writing parameters and does not vary during the readout, aside from a slight erasure. It is thus our suggestion that the observed behavior of the second-harmonic conversion efficiency may be caused by an increase in the nonlinear susceptibility $\chi^{(3)}(0=-2\omega+\omega+\omega)$ of the glass when the intensity of the IR light exceeds a certain critical level. This effect may be due to a sharp increase in the density of electrons in the tail of the conduction band or a redistribution of these electrons among excited levels in the band gap during the passage of the intense light pulse. We believe that the following mechanism of induced second-harmonic generation may be operating. The electrons are excited from impurities into the conduction band in a multiphoton and/or multistep process. The intense scattering then causes these electrons to lose energy, and the electrons go to capture centers, whose concentration is very high ($\sim 10^{20}$ cm^{-3}) in amorphous materials. The emission of electrons from capture centers may be caused by either IR photons or second-harmonic photons, depending on the depth of the capture level. The electrons scattered into the conduction band drift under the influence of the static polarization $\langle \chi_a^{(3)}(\mathbf{E}\cdot\mathbf{E})\mathbf{E}\rangle$, where $\chi_a^{(3)}$ is the third-order susceptibility $\chi^{(3)}(0=-2\omega+\omega+\omega)$, which has increased in an anomalous fashion because of the high electron density near the conduction band. The electrons which have moved to the periphery of the light beam are captured by capture centers and may remain there for a very long time (on the order of several days at room temperature in the absence of light). We believe that this model can explain the appearance in the glass of the strong internal field $E_{\rm st}$, required for effective second-harmonic generation, and also many aspects of this effect, including the dependence of the conversion efficiency on the pump intensity. It is quite likely that the mechanism proposed here is not the only one, but the main point which we wish to make is that this is a possible change in the nonlinear properties of the medium in the field of an intense light wave which affects the writing and readout of gratings of the quadratic polarizability.

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