Optical bistability and critical slowing in the amorphous semiconductor GeS_2

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An optical bistability has been observed in monolithic glassy semiconductors of the Ge–S system during subband excitation ($h\nu < E_g$). The bistability is accompanied by an abrupt change in the refractive index. The process by which the transmission switches in the region of the hysteresis loop exhibits a critical slowing with a time scale $\sim$ 1–10 s. The effect is attributed to a photoinduced cooperative interaction of "native defects" of the amorphous semiconductor, which results in an abrupt structural change in the material.

Glassy chalcogenide semiconductors exhibit several photoinduced effects, in particular, a photoinduced anisotropy, a photoinduced gyrotropy, and a photoinduced light scattering. These effects are substantially larger during subband excitation of bulk samples[2] ($h\nu < E_g$) than during interband excitation of film samples[1] ($h\nu > E_g$). Hajto and Janossy[3] have reported observing an optical bistability of the transmission in free-standing $GeSe_2$ films at a wavelength corresponding to weak absorption. Hajto and Janossy[3] suggested a purely electronic mechanism, a purely thermal mechanism, and a photostructural mechanism for the optical bistability.

In the present letter we are reporting the observation of an optical bistability in bulk glassy chalcogenide semiconductors of the Ge–S system during subband excitation ($h\nu < E_g$). We also report the observation of a giant critical slowing of the switching dynamics near the hysteresis loop. In contrast with Ref. 3, this study demonstrates directly that the optical bistability is accompanied by an abrupt change in the refractive index and by the formation of a waveguide channel in the interior of the amorphous semiconductor. Studies of the switching dynamics (of the critical slowing) show that photostructural conversions (which are possible in an amorphous semiconductor) play a major role in the mechanism for the optical bistability. They are responsible for the giant critical slowing times.

The optical bistability in amorphous semiconductors is a timely topic because this is a nonequilibrium phase transition in an open system, and it allows one to study general trends in the development of disordered nonlinear systems.[4] On the other hand, bistable optical devices have important technical applications.[5] There is particular interest in the switching dynamics, since it is determined by the mechanism and the time scales for the formation of "condensation centers" (structural elements) which lead to the phase transition.[6]

We studied monolithic glass samples prepared by cooling a melt. The samples had two polished faces, the distance between which was varied from 0.1 to 1.0 cm. We used

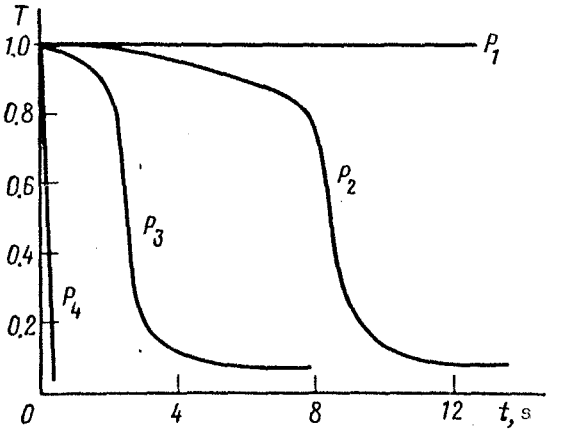

FIG. 1. Kinetics of the transmission at various power densities of the incident light. $P_1 = 50$ W/cm^2; $P_2 = 55$ W/cm^2; $P_3 = 57$ W/cm^2; $P_4 = 70$ W/cm^2. The kinetics was detected with the help of a photodetector with an entrance window 1 cm in diameter, at a point 10 cm from the sample. The 0.5-cm-thick sample had the composition GeS_2.

the light from an Ar^+ laser ($h\nu_1 = 2.41$ eV; $h\nu_2 = 2.54$ eV). This light satisfied the condition $h\nu < E_g$ (for GeS_2, $E_g = 2.7$ eV; Ref. 7). The light was focused onto the entrance face of the sample with a long-focal-length lens, into a spot 100 μm in diameter. The absorption coefficient was ~ 10 cm^{-1}. The light transmitted through the sample was detected by means of a photodetector and a storage oscilloscope. Figure 1 shows the kinetics of the light transmission for four values of the power density P of the light incident on the sample ($h\nu_1 = 2.41$ eV). These power densities were held constant in time. At low power densities (P_1), there is essentially no change in the intensity of the transmitted light. As the power density is raised (P_2), we see an abrupt change in the transmission. When P is raised further, the switching occurs in progressively shorter times (P_3). At very high values of P, the surface of the sample suffers damage, involving an evaporation of material.

The transmission switching at intermediate values of P (55 W/cm$^2 < P <$ 65 W/cm^2) turned out to be reversible. In other words, when the light was turned off and then on again, the transmission underwent a repeated switching. The kinetics of the switching was reproducible well at $P = P_3$ but only in general features at $P = P_2$ (Fig. 1).

This switching of the intensity of the transmitted light was accompanied by an abrupt increase in the diameter of the light beam transmitted through the sample and also a decrease in the intensity of the scattered light. Both of these effects were also reversible (Fig. 2). In this case the optical bistability is evidently accompanied by an abrupt change in the refractive index, i.e., a photorefraction, since the change in the beam paths could in fact be seen visually. A waveguide channel formed in the interior of the sample in the volume through which the laser beam passed. This channel could lead to a decrease in the intensity of the scattered light. That a waveguide channel

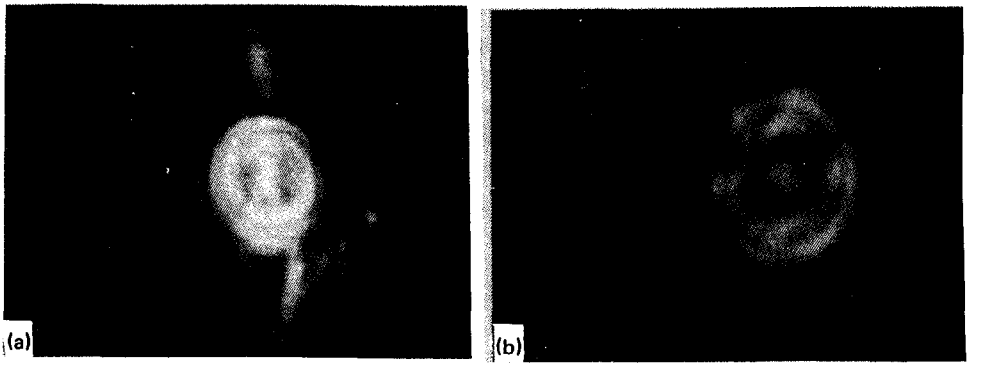

FIG. 2. Image of a light beam 0.5 cm in diameter which has been transmitted through a sample. This image was formed on a screen put in the position of the photodetector. a—Before; b—after the switching.

formed is confirmed by the circumstance that the image in Fig. 2b is ring-shaped.

When we used the other line of the Ar^+ laser ($h\nu_2 = 2.54$ eV), we did not observe this switching.

The observation of the switching and the critical slowing suggested that there should be hysteresis on a plot of $I_{out} = f(P_{in})$, where P_{in} is the power density of the light incident on the sample, and I_{out} is the intensity of the light transmitted through the sample and incident on the photodetector. This hysteresis was indeed observed (Fig. 3). The power P_{in} was swept smoothly with the help of a quarter-wave plate and a Glan prism at a rate of 0.01 mW/min in the region of the hysteresis loop. In other words, the sweep rate was almost two orders of magnitude lower than that in Ref. 3. We believe that this low rate is necessary because of the unusually long switching times.

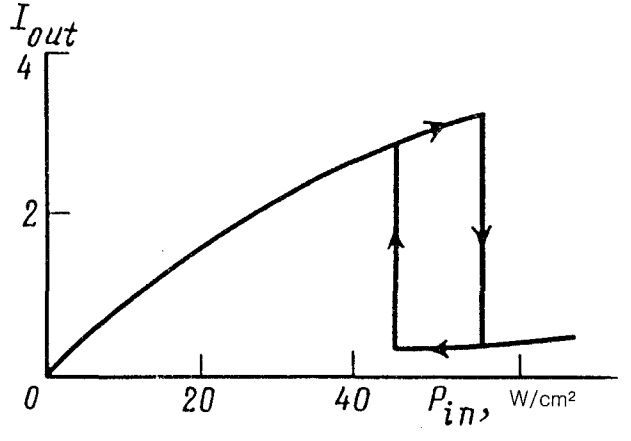

FIG. 3. Hysteresis in the plot of $I_{out} = f(P_{in})$.

In certain cases, we observed oscillations in the transmission in the hysteresis-loop region. We observed both regular oscillations, with a typical period ~ 1 s, and some random oscillations. The switching thresholds in the case of the optical bistability and the nature of the oscillations depended on the polarization of the incident light. We recall that a polarization dependence of photoinduced effects in chalcogenide glasses is a property of these materials.[1,2]

These results show that in this particular case of an optical bistability in an amorphous semiconductor the nonlinear mechanism for the interaction of the light with the material cannot be purely thermal or purely electronic. The reason is that in the former case the interaction time constant is on the order of 1 ms, and in the latter it is even shorter. The observed critical slowing times indicate that we should look for some other mechanism, with an interaction time constant of 1–10 s. This other mechanism might involve photostructural conversions, which are extremely characteristic of amorphous semiconductors.[1,2] These conversions are particularly effectively during subband excitation ($h\nu < E_g$), during which the light interacts with native defects of the amorphous semiconductor, i.e, structural elements which are inherent in specifically the amorphous state and which determine its existence.[8]

The positive feedback required for the occurrence of an optical bistability results from a cooperative interaction of these native defects, which make up 1–10% of the total number of atoms.[8,9] It is natural to suggest that in this situation the photoinduced creation (or reorientation) of one defect creates conditions which are more favorable for the creation (or reorientation) of other, neighboring defects, with the result that the structure of the amorphous material becomes more porous (or more ordered).

The effects described here obviously occur during a heating of the samples. This circumstance might suggest that the optical bistability results from the purely thermal mechanism which has recently been studied in crystalline semiconductors.[10,11] However, the experiments described here as well as the data of Ref. 3 show that the most characteristic features of the optical bistability observed in amorphous semiconductors can be explained only on the basis of a metastability (or bistability) of the structure of the amorphous state (atomic configurations).[8,9]

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