P146

Journal of Non-Crystalline Solids 137&138 (1991) 981–984
North-Holland

JOURNAL OF
NON-CRYSTALLINE SOLIDS

ON THE KINETICS OF THE REORIENTATION OF LIGHT-INDUCED ANISOTROPY IN *a*-$GeSe_2$ FILMS

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We present experimental data on induced birefringence in *a*-$GeSe_2$ thin films with light pulses as short as *10ms*. In the case of tighter focusing of the laser beam, the presence of another process is indicated in the measured curves. We speculate that this process is photo-induced crystallization.

1. Introduction

In our recent paper[1] we described the kinetics of the reorientation of the anisotropic structure in amorphous $GeSe_2$ thin films. We explained the experimental results on the basis of the model of bistable centers with a wide distribution of relaxation times. We described experiments showing that one can detect photo-induced birefringence on a time scale shorter than *1* second.

In this paper we present the results of measurements with short light pulses (*10ms-400ms*) and tight focusing.

2. Experimental

For the measurements the two-beam (pump-and-probe) technique was used. The detailed description of our experimental setup is given in Ref.1 . A vacuum-evaporated, $7\,\mu m$ thick $GeSe_2$ film on glass substrate was investigated . The applied measuring method was the same as described in Ref.1. With a linearly polarized laser beam, birefringence was induced in the sample. By switching the polarization state of the beam between two orthogonal directions, keeping the intensity fixed, reorientation cycles were generated. A second laser beam of weak intensity measured the actual magnitude of the induced birefringence. The probe beam illuminated the sample only during the time of sampling the signal. During this time period (typically *30ms*) the pump beam was blocked with the help of an electromagnetic shutter in order to eliminate the disturbing background caused by the light scattered from the pump beam into the detector. This is especially important in the case of tight focusing. All measurements were performed at room temperature.

3. Results

The time dependence of the birefringence induced by linearly polarized light with periodically alternating polarization state, can be seen in Fig.1. The amplitude of the reorientation cycles shows a well observable change in the first period of irradiation but later on it becomes constant. The change in the amplitude of the cycles depends on the history of the sample. In the case of an *as-evaporated* sample, a decrease of the signal was detected.

3.1. Short time kinetics

After the stabilization of the amplitude of the cycles, we measured the change in the birefringence at the very beginning of the reorientation half cycles in the *10ms-400ms* time regime. The results are shown in

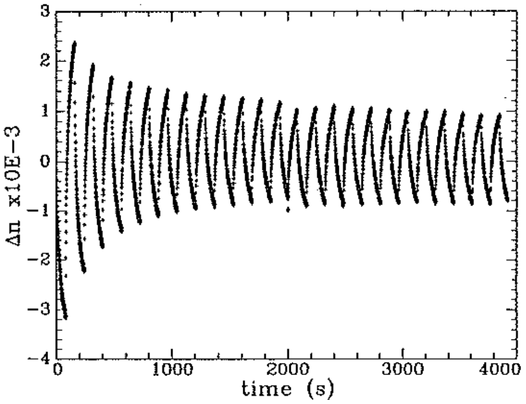

FIGURE 1

Time dependence of birefringence induced by polarized light with alternating polarization state.

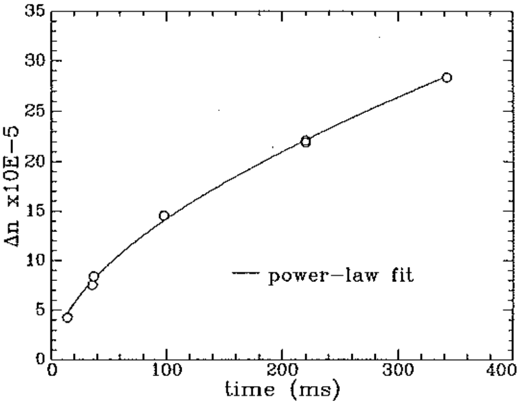

FIGURE 2

Short time behaviour of induced birefringence. The fitted power-law has an exponent of 0.51 .

Fig.2. During this measurement the pump beam intensity was *8mW* and it was focused onto a spot of *20 μm* radius. The measured curve can be well fitted with a power-law with a non-integer exponent; in the present case the exponent is *0.51.* In order to interpret the observed short time kinetics of the reorientation process we utilized the concept of anisotropic bistable centres. The detailed description of our model can be found in Ref.1. In our model we suppose that there are bistable centres with short relaxation time which give rise to the short time part of the signal. From the observed power-law time dependence, we concluded that the relaxation time distribution of the centres follows also a power-law dependence, at least in the short relaxation time limit. Converting the relaxation time distribution into distribution of energy barrier heights, one obtains an exponential distribution with the same characteristic width as in the case of the electron state distribution in the bandtails. From this fact we speculate that the bistable centres are realized by atomic configurations which create electronic states in the bandtails. In this respect our model is close to that proposed by Tanaka[4].

3.2. Structural transformation

In the case of tighter focusing, in certain parts of the sample, the reorientation cycles showed interesting change in their character (Fig.3). Initially, the reorientation cycles showed a behaviour similar to the one shown in Fig.1 (type I cycles). On continuing the irradiation, markedly different cycles developed (type II cycles). Type II cycles are characterized by: *a)* significantly smaller amplitude and *b)* opposite sign as compared to type I cycles. The phenomenon described above occurs with a variety of irradiation intensities *(3mW-12mW*). With decreasing intensity the rate of disappearence of the type I behaviour decreases. The transition from type I to type II could not be observed in the case of larger spot radius. However, the small spot radius is not a sufficient condition for the transformation; in certain parts of the sample we could observe the phenomenon,

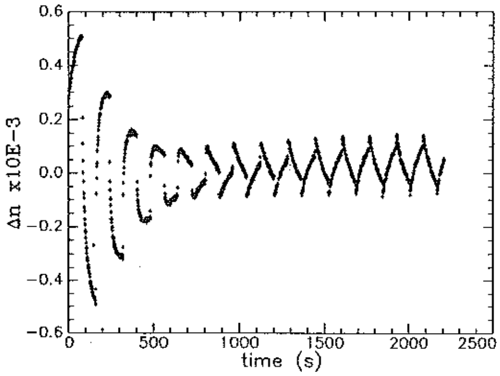

FIGURE 3
During the irradiation the character of the reorientation cycles changes drastically.

in other parts, we could not, though the focusing was the same. The experiments show that, in order to observe the change in the behaviour of the reorientation cycles, a small spot radius (tight focusing) rather then a high intensity is necessary. Thus we believe that it is the high intensity gradient which is crucial.

It is well known that, upon irradiation of an *as-evaporated* thin film, irreversible photostructural changes take place. These changes include changes in both the optical absorption edge and the density. These phenomena are known as photobleaching[2] and photodensification[2,3], respectively. During the light exposure, a density difference builds up between the exposed and unexposed parts of the sample, leading to a mechanical stress. Obviously, the higher the irradiating intensity gradient, (i.e. the smaller the spot radius), the higher the density gradient and the mechanical stress will be. This photoinduced stress, superimposed on the stress in the sample originating from the sample preparation procedure, might initiate the growth of polycrystalline regions in the sample. It is known that crystallization in $a\text{-}GeSe_2$ thin films is a rather complex process. The crystalline regions grow into the amorphous network in a fractal-like manner[5]. Therefore in this area there is a very large amorphous-crystalline boundary interface. While there is no structural freedom for photostructural changes in the crystalline phase[2], the bistable centres in the solid-solid interface might be responsible for the opposite sign of the reorientation behaviour.

4. Conclusion

We presented experimental data demonstrating that one can induce and detect birefringence with light pulses as short as *10ms*. The theoretical model will be presented elsewhere. With tighter focusing one can speed up the process. However, when the beam spot on the sample is too small another process, modifying the nature of the reorientation can occur. We speculated that this process is photo-induced crystallization.

This work was supported by OTKA Grant Number 1043.

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