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C. N. Afonso, J. Solis, F. Catalina, and C. Kalpouzos

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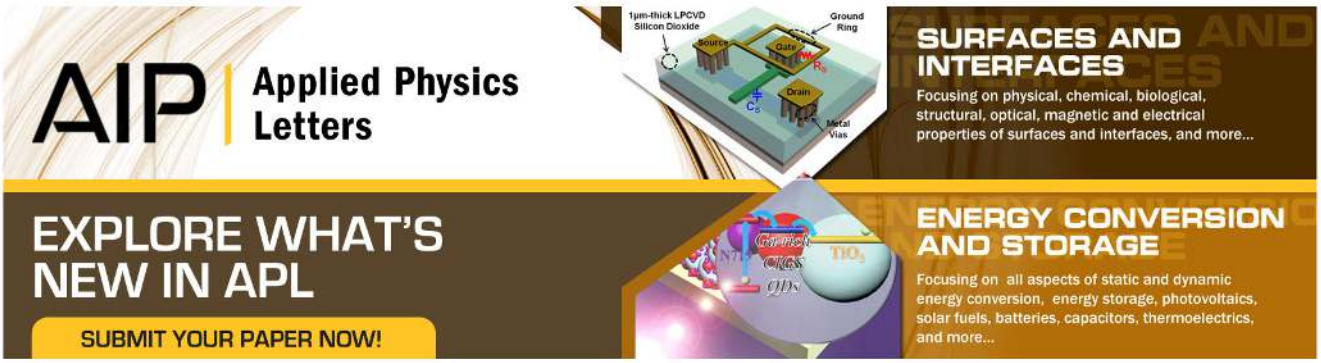


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# Ultrafast reversible phase change in GeSb films for erasable optical storage

C. N. Afonso, J. Solis, and F. Catalina
*Instituto de Optica, CSIC, Serrano 121, 28006 Madrid, Spain*

C. Kalpouzos
*FORTH-IESL, P.O. Box 1527, 7110-Heraklion, Crete, Greece*

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Amorphous-to-crystalline and crystalline-to-amorphous transformations are triggered in GeSb thin films by irradiation with femtosecond and picosecond laser pulses. Phase changes are accompanied with optical contrast and therefore the feasibility of phase-change optical recording at ultrafast rates is demonstrated for the first time. The phase reversal by ultrashort pulses seems to be related to the dependence of the degree of undercooling prior to solidification on the irradiation energy density.

Although magneto-optical recording has become the most mature technology for optical storage,[1,2] phase-change optical recording has several attributes which make it a very promising technology. It is an all-optical disk system which involves a simpler optical head, making the disks interchangeable with write-once optical disks. The phase-change optical contrast signal is very high and single-beam overwriting[3] is possible. The most significant drawback with phase-change materials is the demand for materials with high-speed crystallization which retain their long-term room-temperature stability. Crystallization has to be achieved with laser pulses of tenths of nanoseconds or even subnanosecond laser pulses.[2,4] Current trends in research of new media aiming to decrease the crystallization time have involved Sb alloys, i.e., GaSb,[4,5] InSb,[5,6] GeTeSb,[7] or InSbTe[8] films. However, fast crystallizing materials with crystallization times close to 10 ns have been confined to write-once applications.[5] There is therefore a real need to investigate phase transformations triggered by subnanosecond pulses. To our knowledge no experimental attempts have been done up to date to induce crystallization or amorphization in fast crystallizing materials by means of subnanosecond laser pulses.

The aim of this letter is to demonstrate for the first time that phase reversal (amorphization) can be achieved in fast crystallizing materials with femtosecond and picosecond laser pulses. It also aims to investigate the existence of subnanosecond triggered ultrafast crystallization phenomena.

The samples are either amorphous or polycrystalline $Ge_{0.1}Sb_{0.9}$ thin films on carbon-coated mica, which is a very suitable substrate to prepare samples for transmission electron microscopy (TEM) studies. The amorphous films [Fig. 1(a)] are obtained directly by dc magnetron sputtering in a multitarget system. The crystalline films are obtained by UV laser irradiation of the as-grown amorphous films with 12 ns (FWHM) pulses from an ArF excimer laser ($\lambda = 193$ nm) with an energy density of 56 mJ/cm$^2$. It is found that $Ge_{0.1}Sb_{0.9}$ films can be crystallized through a melting–rapid solidification process and the optical contrast between the amorphous and the crystalline phases is $\approx 15\%$–$20\%$. Further details of both film growth and crys-

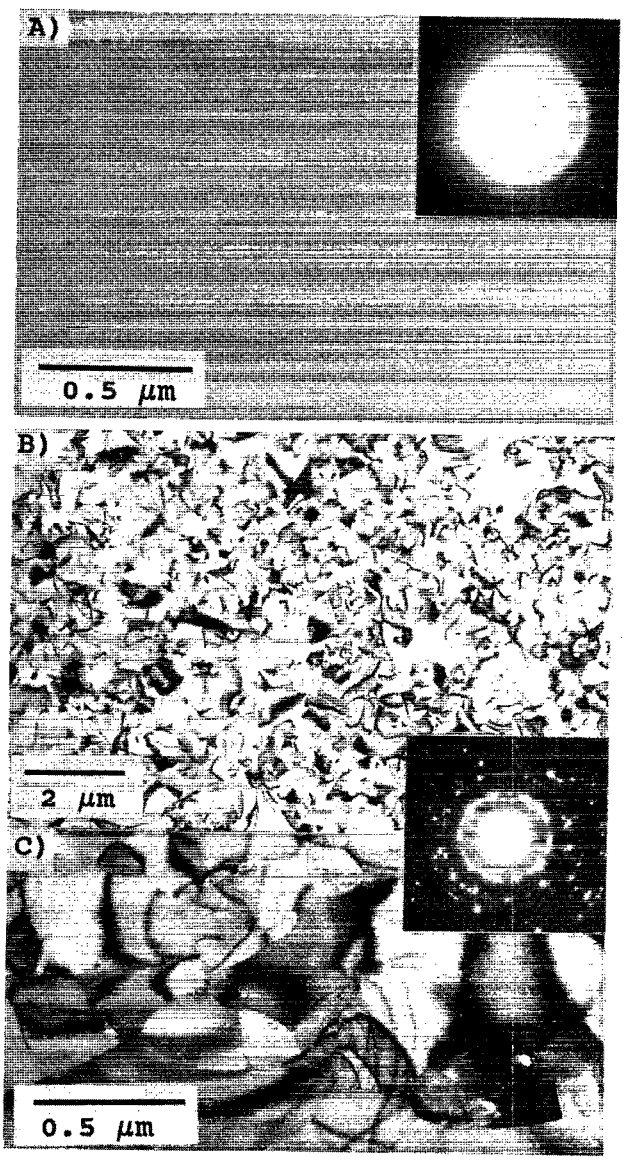


FIG. 1. TEM images and SAD patterns of the amorphous (as-grown) material (A) and the crystalline material (B,C) obtained after irradiating the as-grown material with a single 12 ns, 56 mJ/cm$^2$ pulse from an ArF excimer laser, with (C) a magnification of (B).

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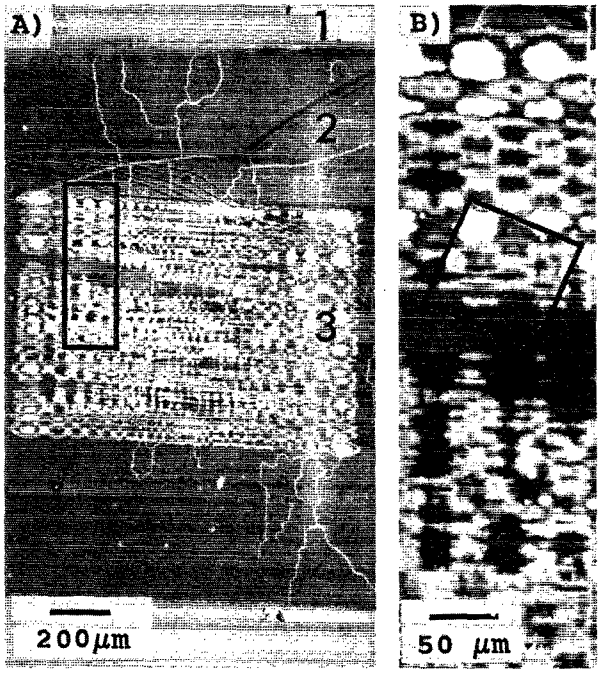


FIG. 2. Optical image (A) of the amorphous (as-grown) material (region 1), the crystalline material obtained after irradiation with a 12 ns, 56 mJ/cm$^2$ pulse (region 2), and the material obtained after irradiation with a 500 fs, 15 mJ/cm$^2$ pulse (region 3). The optical image shown in (B) corresponds to a magnification of the area in (A), which is within the rectangle.

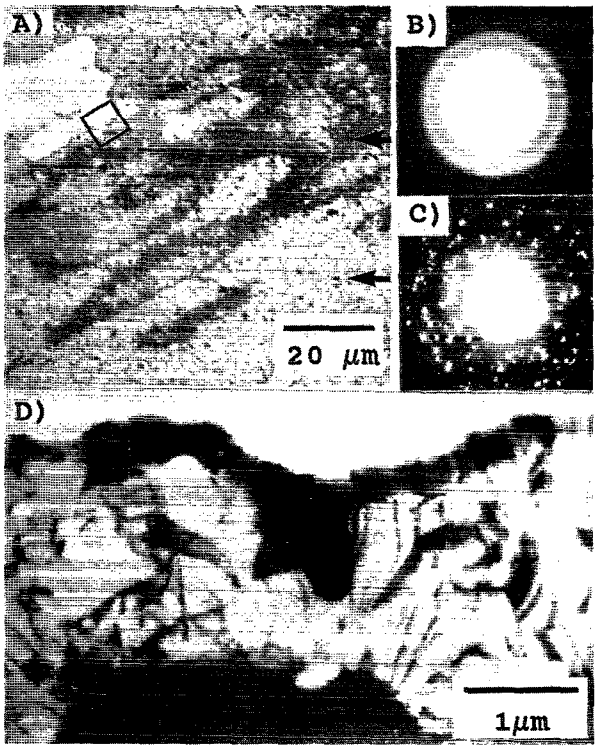


FIG. 3. TEM image (A) of the area included within the square in Fig. 2(B) and SAD patterns of dark (B) and bright (C) areas in (A). (D) is a high-magnification TEM image of the region within the square in (A).

tallization kinetics under nanosecond pulses can be found elsewhere.[9]

Both amorphous and crystallized films are then irradiated with 500 fs and 5 ps (FWHM) laser pulses at 248 nm. They are obtained from a frequency-doubled distributed feedback dye laser seed pulse in a two-pass amplification through a KrF amplifier cavity. In the case of the crystalline material, the ps/fs laser beam is located at the center of each nanosecond pulse-irradiated area, being partially focused to an area of $\approx 1$ mm$^2$. The average energy density is varied from 9 to 40 mJ/cm$^2$, and for values higher than 10 mJ/cm$^2$ the crystalline material is optically transformed, as can be seen in Fig. 2. The present area irradiated with fs pulses (region 3), bright, dark, and white (ablated) regions. This optical image clearly shows the existence of a quasi-periodic inhomogeneity in the spatial profile of the fs pulse (also ps pulse) due to the existence or diffraction effects in the KrF amplifier cavity.

The reflectivity and transmission of the fs (ps) irradiated areas are determined by scanning an optical probe (4 $\mu$m FWHM, $\lambda = 632.8$ nm) along them. The results show that the optical contrast (in Fig. 2) between the bright and dark regions in region 3 is similar to that between the amorphous as-deposited material (region 1) and the ns pulse crystallized material (region 2). This suggests that both amorphous and crystalline phases have been most probably formed during fs (and ps) pulse irradiation and proves that the crystallization process can be reversed with subnanosecond pulses at least from the point of view of optical contrast.

TEM images in Fig. 3 show that those areas which were bright in the optical images are now dark, except the ablated regions which are white in both images. The SAD patterns show that dark areas are amorphous whereas bright ones are crystalline in agreement with what was deduced from the optical contrast measurements. The border of the ablated regions (where the highest peak temperatures are reached) are crystalline [Fig. 2(D)].

The comparison of the optical micrographs obtained by increasing the average energy density when irradiating with fs pulses together with what is shown in Fig. 3(D) allow us to conclude that those crystalline regions inside the fs spot have been exposed to higher-energy densities than the amorphous regions. Therefore, these regions have melted as a consequence of the fs irradiation but have crystallized instead of reamorphizing upon solidification.

Figure 4 shows TEM images of both initially crystalline and initially amorphous materials after irradiation with a fs pulse. It can be seen that crystalline material can be directly obtained from an amorphous material under fs pulse irradiation and provides further demonstration that crystallization can be triggered by ultrashort laser pulses. No significant differences are observed between the crystals obtained in the two cases [Figs. 4(B) and 4(D)] and those obtained under ns laser pulse irradiation [Fig. 1(C)]. Although the results described here in detail were obtained with fs pulses, the same results and conclusions are achieved under irradiation with ps pulses.

Let us finally analyze the physical processes which



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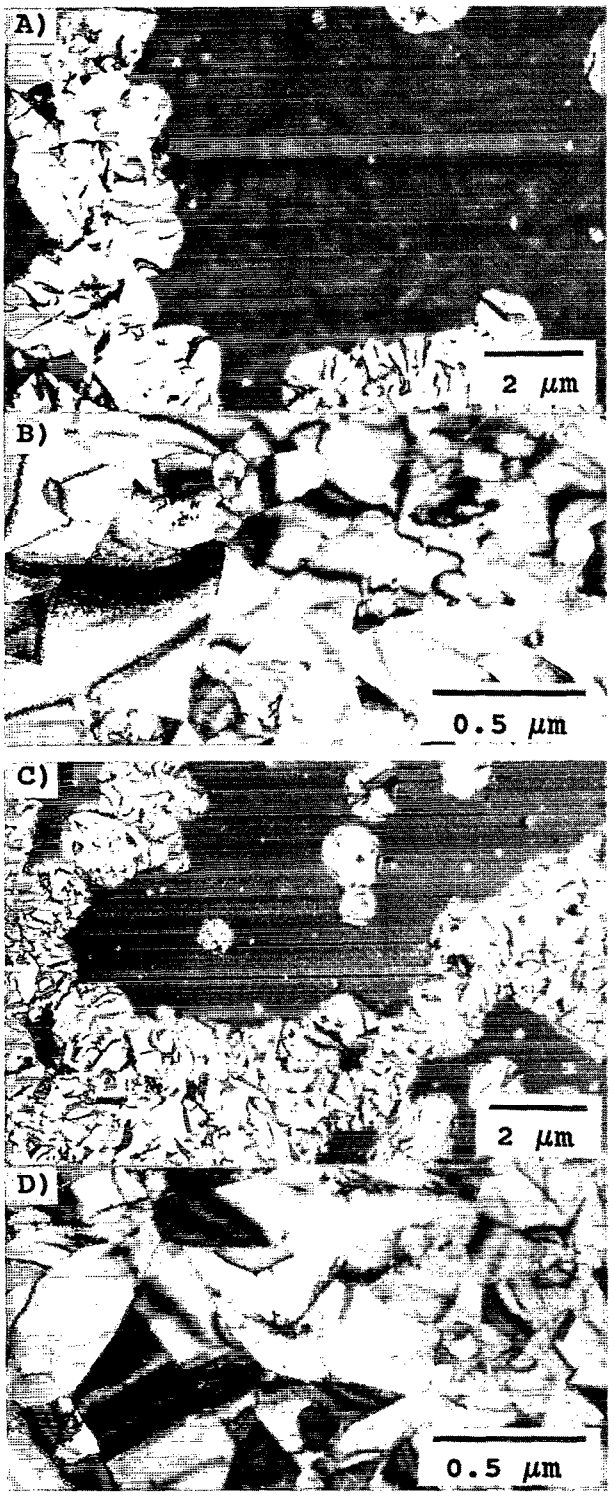


FIG. 4. TEM images obtained after irradiating the crystalline (A,B) and the amorphous (C,D) materials with a 500 fs pulse, with (B) and (D) high magnifications of (A) and (C).

may be responsible for this ultrafast triggered phase changes. The real-time reflectivity transients obtained during the nanosecond-induced crystallization of GeSb films[9] show that crystallization is produced at quenching rates $\approx 10^9$ K/s. The quenching rates expected[10] for irradiation with fs and ps are in both cases $\geqslant 10^{12}$ K/s and therefore the formation of amorphous phases may be expected. Nevertheless the undercooling achieved prior to solidification is another important factor involved in the formation of amorphous phases by rapid solidification processes.[11] Experimental evidence of the influence of the energy density on the undercooling achieved when irradiating thin-film materials on poor thermal conducting substrates has been already provided.[12,13] When the energy deposition is large enough, the substrate can be effectively heated. The thermal gradient along the surface normal is then reduced and solidification occurs at higher temperatures (undercooling decreases). A clear example of this reasoning is provided by the comparison of the crystalline microstructures induced by irradiation with fs [Fig. 4(D)] and ns [Fig. 1(C)] pulses on the amorphous as-deposited material. Both types of microstructures are essentially the same, indicating that solidification has occurred at similar temperatures in spite of the expected differences of cooling rate.

As a general conclusion, the results obtained show that irradiation of $Ge_{0.1}Sb_{0.9}$ thin films with 500 fs and 5 ps laser pulses yield the formation of either a high-reflectivity crystalline phase or a low-reflectivity amorphous phase depending on the pulse energy density and independent to the initial phase in the GeSb film. The physical explanation for this effect is the dependence of the degree of undercooling achieved prior to solidification on the irradiation energy density. The feasibility of phase-change reversible optical recording using subnanosecond pulses is therefore demonstrated.

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[1] E. M. Engler, Adv. Mater. **2**, 166 (1990).
[2] K. A. Rubin and M. Chen, Thin Solid Films **181**, 129 (1989).
[3] T. Nishida, M. Terao, Y. Migauchi, S. Horigome, T. Kaku, and N. Ohta, Appl. Phys. Lett. **50**, 667 (1987).
[4] D. J. Gravesteijn, H. M. van Tongeren, M. Sens, T. Bertens, and C. J. van der Poel, Appl. Opt. **26**, 4772 (1987).
[5] D. J. Gravesteijn, Appl. Opt. **27**, 736 (1988).
[6] Y. Gotto, K. Utsumi, A. Ushioda, I. Tsugma, and N. Koshino, Mater. Res. Soc. Symp. Proc. **74**, 251 (1987).
[7] M. Suzuki, K. Furuya, K. Nishimura, K. Mori, and I. Morimoto, SPIE **1316**, 374 (1990).
[8] Y. Sato, H. Minemura, Y. Maeda, I. Ikuta, A. Andoh, N. Tsuboi, and M. Nagi, SPIE **1316**, 267 (1990).
[9] J. Solis, J. F. Trull, and C. N. Afonso (unpublished).
[10] F. Spaepen, in *Ultrafast Phenomena V*, edited by G. R. Fleming and A. E. Siegman (Springer, Berlin, 1986), Vol. 46, p. 174.
[11] W. J. Boettinger and J. H. Perepezko, in *Proceedings of the Symposium on Rapidly Solidified Crystalline Alloys*, edited by S. K. Das, B. H. Kear, and C. M. Adam (Metallurgical Society of AIME, Warrendale, PA, 1986), p. 21.
[12] C. N. Afonso, F. Catalina, and J. Solis, Mater. Sci. Eng. B **7**, 169 (1990).
[13] C. N. Afonso, R. Serna, F. Catalina, N. Teixeira, M. F. da Silva, and J. C. Soares, Appl. Phys. A **52**, 69 (1991).



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