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Photoinduced surface deformations in ion-conducting Ag–As–S glasses.

II. Anisotropic deformation produced by large light spots

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Anisotropic surface deformations appear in AgAsS₂ chalcogenide films when exposed to linearly polarized 2.0 eV light spots with diameters of 5–200 μm . The anisotropic shape reflects the polarization direction. The shape also changes with the direction of light incidence. Illumination on the free surface of the films gives a craterlike deformation, and illumination through a substrate gives an anticrater deformation. In bulk samples, under any exposure conditions, an isotropic convex structure appears. The formation mechanisms of these patterns are discussed taking into account the photoinduced migration of Ag⁺ ions. © 2001 American Institute of Physics.
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I. INTRODUCTION

Photoinduced deformation phenomena are known to exist in a variety of solids.^{1–3} Some of them are caused by temperature rises induced with light absorption,¹ while in covalent chalcogenide glasses and other materials such as organic solids, some kinds of athermal processes appear.^{2,3} In these materials, deformation seems to occur through photo-electro-atomic coupling processes, while the detailed mechanisms remain to be studied. It should also be mentioned that some responses to light in biological systems may also be regarded as kinds of systematic photo-electro-atomic deformations.⁴ Accordingly, the physical understanding of photoinduced electro-atomic processes will be valuable in a wide range of scientific areas.

In a previous paper (Paper I), it has been demonstrated that, in Ag–As–S ion-conducting amorphous semiconductors, the isotropic volume expansion appears when the sample is exposed to light spots with diameters smaller than $\sim 5 \mu\text{m}$. The phenomenon has been understood by taking into account the photoinduced chemical modification (PCM),⁵ i.e., photoinduced Ag⁺-ion migration into illuminated regions gives the volume expansion.

However, the relationship between the expansion phenomenon described in Paper I and a patterning effect previously reported⁶ is not clear. In Ref. 6 it has been demonstrated that the *anisotropic* pattern accompanying fine fringes (with a periodicity of $\sim 1 \mu\text{m}$) and radiating streaks appears when Ag–As–S films are exposed to linearly polarized light beams with a spot size of $\sim 50 \mu\text{m}$ [see, Fig. 1(a)]. The appearance is very different from the isotropic expansion described in Paper I. Therefore, it is tempting to investigate what causes the difference. We will see in the present work that the light-spot diameter is responsible for the difference, and consider the reason. The present work will provide insights into the universal understanding of macroscopic pattern formations in ion-conducting amorphous semiconductors.

II. EXPERIMENT

The samples employed and evaluation procedures of photoinduced deformations were the same as those described in Paper I. In brief, the samples were photodoped AgAsS₂ films with a thickness of $\sim 1 \mu\text{m}$ and polished AgAsS₂ glass disks with a thickness of $\sim 0.2 \text{ mm}$. Photoinduced deformations were analyzed using a Nomarski-type interference optical microscope, an atomic force microscope (AFM), and an electron probe microanalyzer (EPMA).

In the present study, the spot diameter of He–Ne laser light ($\hbar\omega = 2.0 \text{ eV}$) at the sample surface was varied in a range of 2–200 μm using microscope objective lenses and single lenses with long focal distances. A preliminary experiment was also done using 2.6 eV laser light with a spot diameter of 40 μm . Here, the spot size means the full width at half maximum of the light spot with Gaussian shapes. These light beams were linearly polarized, and illumination was provided at room temperature, unless otherwise stated.

III. RESULTS

Figure 1(a) shows an AFM image of an AgAsS₂ film after an exposure to a linearly polarized 2.0 eV light beam with a spot diameter of $\sim 50 \mu\text{m}$ for 3 h. The light beam has irradiated the free surface of the film. Since a produced deformation extends more widely than the AFM scanning size of 75 μm , only a side-part view is shown here.

We can see in Fig. 1 two kinds of deformations, which are fine and gross. The appearance of fine deformations is consistent with that reported previously.⁶ That is, in the central part of the illuminated spot (black region) many small fringes exist, the direction of which is nearly parallel to the electric field (indicated by double arrow), and there exist streaks radiating from the central part to the direction nearly perpendicular to the electric field. In addition, Fig. 1 shows a gross anisotropic deformation, i.e., as shown by the cross-sectional view, a craterlike deformation exists in the direction along the radiating streaks. On the other hand, along the electric field, there exists only a hollow at the light spot. It is mentioned here that circularly polarized beams have pro-

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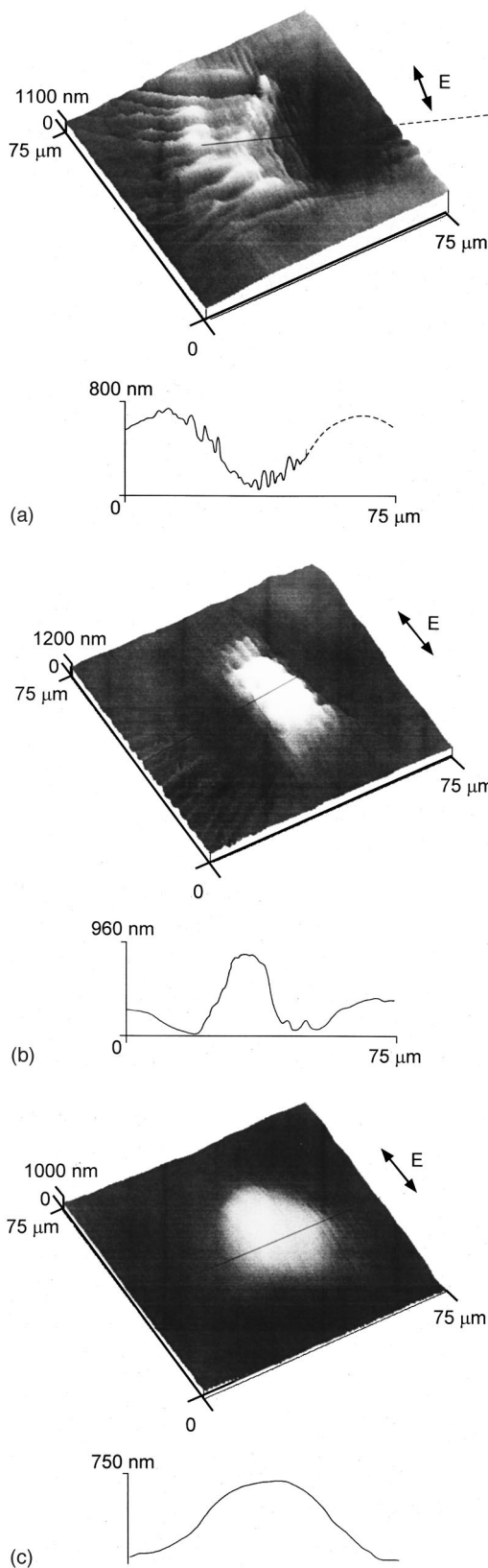


FIG. 1. AFM images of AgAsS_2 samples after an exposure to a linearly polarized 2.0 eV light beam with a spot diameter of $\sim 50 \mu\text{m}$ and an intensity of 2 mW for 3 h. The double arrow indicates the directions of the electric field. The lower figures show the cross-sectional views along the solid lines in the images. (a) A deformation in a 1- μm -thick film which is exposed to the light incident upon the sample surface. The illuminated center is located at the hollow part. (b) Similar to (a), except that the film is exposed through the substrate. (c) A deformation in a 0.2-mm-thick bulk sample.

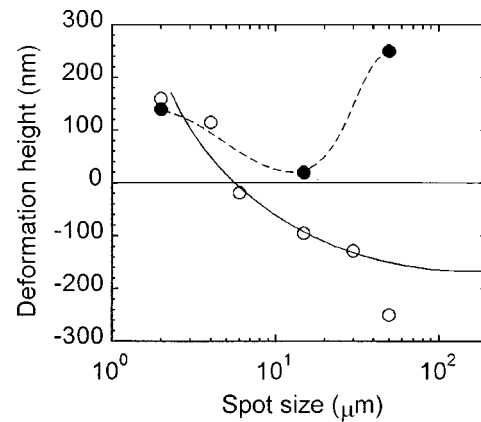


FIG. 2. The deformation height at the central part of the light spot in an AgAsS_2 film as a function of the light spot size. Illumination is provided from a 2 mW He-Ne laser. Open and closed circles are the data obtained when the light is incident on the sample surface and through the substrate.

duced isotropic craterlike patterns, which are less prominent in magnitude than those produced by linearly polarized light. At low temperatures of $\sim 80 \text{ K}$, no surface deformations appeared under similar exposure conditions, which may be consistent with a negligible diffusion coefficient of Ag^+ ions (see Fig. 5 in Paper I).

The deformation shown in Fig. 1(a) is distinctly different from that reported in Paper I in two respects. That is, the deformation shape is not isotropic and not convex. Here, in several experimental conditions, the size of light spots is substantially greater. Accordingly, in order to know the origin of the different shape, the dependence of the deformation upon the spot size should be investigated in detail.

Figure 2 shows the deformation height at the central part of light spots in a 1- μm -thick AgAsS_2 film as a function of the spot diameter (open circles). Light exposure has been provided until the deformation is mostly saturated, and the exposure time varies from 2 min for the 2 μm spot to 32 h for the 200 μm spot.

We see in Fig. 2 that, when the spot is small, the deformation is convex as described in Paper I, and it changes to concave at the spot diameter of $\sim 5 \mu\text{m}$. For wider deformations the height measurement using the AFM becomes less accurate due to the inclination of sample. However, Fig. 2 shows that concave patterns, being qualitatively the same as that shown in Fig. 1(a), appear up to 200 μm . Greater light spots may also give concave deformations, however, these cannot be investigated practically, because the exposure time needed becomes longer than ~ 2 days. Why does the characteristic change occur at $\sim 5 \mu\text{m}$?

What is more puzzling here is that the deformation shape depends upon the direction of light incidence. When a light spot of $\sim 50 \mu\text{m}$ diameter irradiates the film from the free surface, the concave pattern appears [Fig. 1(a)]. However, surprisingly, the same light beam irradiating the film from the substrate side produces an anisotropic *anticrater* structure (convex at the center) as shown in Fig. 1(b).

The deformation height at the center for the substrate-side illumination is shown in Fig. 2 by closed circles. When the spot size is $\sim 2 \mu\text{m}$, as described in Paper I, the defor-

mation is irrespective of the incident direction, and it is isotropic and convex. Upon substrate-side illumination with an $\sim 15 \mu\text{m}$ spot, the deformation becomes anisotropic and multicircular, and the height at the center is nearly zero. Then, in a spot of $\sim 50 \mu\text{m}$, the anticrater structure [Fig. 1(b)] appears.

Compositional analyses of the Ag–As–S films using the EPMA have demonstrated the following: The fine fringes possess an Ag compositional modification of $\sim 5 \text{ at. \%}$, which is consistent with the previous work.⁶ In contrast, the gross deformation brings about no appreciable Ag concentration changes.

Exposure experiments have also been performed for AgAsS₂ bulk samples with a thickness of $\sim 0.2 \text{ mm}$. Then, produced deformations were isotropic and convex as shown in Fig. 1(c), irrespective of the spot size. This expansion resembles that reported in Paper I in shape and magnitude, and accordingly, it can probably be ascribed to the PCM effect. That is, accumulated Ag⁺ ions through photo-electro-ionic interaction cause the volume expansion.

IV. DISCUSSION

In considering the mechanism of the anisotropic crater and anticrater formation in Ag–As–S films, we should underline the following two points. One is that photothermal effects such as thermal evaporation cannot be responsible, because the concave and convex structures are produced by the same light incident upon the film from the free surface and from the substrate side. In addition, the thermal diffusion of Ag cannot be a primary effect, since it cannot explain the anisotropy. The other is that the photo-electro accumulation of Ag⁺ ions from unilluminated to illuminated regions, which causes the convex deformation in the small-spot case (Paper I), cannot be an origin here, because corresponding Ag-content changes have not been detected.

In this context, we note that Bian *et al.* report a polarization-sensitive surface deformation in organic polymers when exposed to a polarized light beam.³ They propose a mechanism based on the interaction between the electric field of light and induced dipoles in the polymers. The interaction can generate forces, which seem to be essentially the same as that induced in the optical alignment of laser-trapped particles in fluids.⁷ However, the model cannot explain the contrasting deformations induced by the front- and backillumination. Accordingly, other possibilities should be examined.

At present, the most plausible motive force which can produce the anisotropic gross pattern in the film seems to be the one generated from the fine fringes (see Fig. 3). In the following, we will first discuss the possibility that the fringe produces expansive forces. Next, it will be suggested that the force direction can depend upon the direction of incident light, which is responsible for the crater and anticrater deformations.

The fringe can produce expansive forces for the following reason. As described in Sec. III, it has been demonstrated that the photoinduced fringe accompanies the Ag-content modulation of $\sim 5 \text{ at. \%}$.⁶ This Ag-content modification can

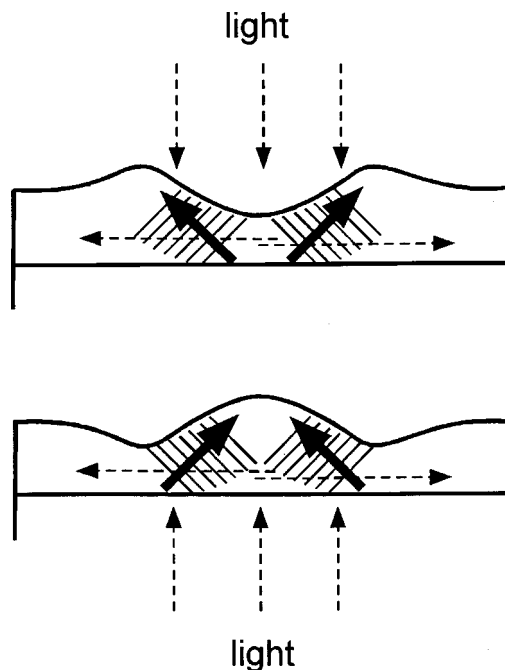


FIG. 3. A model for the deformations which depend on the directions of incident light. Dashed lines with arrows represent light propagation, slanted lines depict interference fringes, and thick arrows show the directions of material flows.

produce expansive forces, because Ag-accumulated phases will expand (Paper I), while Ag-depleted regions may hold mostly fixed volumes due to the smaller fluidity in amorphous networks in darker regions. Actually, we can see in Fig. 1(a) of Paper I that the volume ratio of the central expansion and the peripheral depression is 3/1, which suggests that the Ag-depleted region possesses smaller volume changes. Under these circumstances, an overall volume change in an illuminated region is expansive, which will produce expansive forces.

Here, the mechanism giving rise to the Ag-content modulation should be considered in detail. In the previous work,⁶ it has been proposed that the fringe accompanying the Ag-content modification appears through the following processes: Incident polarized light is scattered by inhomogeneities such as dust particles on sample surfaces and disordered glassy structures. The feature can be treated using the Mie scattering formulas, in which the scattering is stronger to the directions perpendicular to the electric field of light. Here, some scattered light will be converted to guided waves, since the sample consists of three-layer structures with air ($n = 1$)/film ($n = 3$)/substrate glass ($n = 1.5$). Then, interference fringes can be produced by incident and guided light, which will cause the Ag fringes through the PCM effect.⁵

We should note here that, as illustrated in Fig. 3, if the fringe is produced by the interference between the incident and the guided light, it inclines upward and downward depending upon the direction of incident light. These fringes will generate the expansive forces. Since a substrate exists, the upward and downward fringes will cause outward and inward forces (shown by the thick arrows in Fig. 3), respectively. If these forces can induce material flows, the illumi-

nated area will become thinner (crater) and thicker (anti-crater) for the front- and backilluminations.

The above-mentioned model is consistent with several observations. First, as shown in Fig. 1, the (anti)crater patterns can appear only in thin samples, since guided light cannot exist in bulk samples. Second, it has been demonstrated that 2.6 eV light does not produce the pattern. This is because the absorption coefficient for 2.6 eV light in the Ag-As-S film is 10^5 cm^{-1} ,⁸ so that the guided mode cannot practically propagate. Third, the (anti)crater deformations can be prominent only in the direction perpendicular to the electric field of light, since the propagation of guided waves follows the Mie scattering process. Fourth, it has been demonstrated that no patterns appear at 80 K for the light spot of 50 μm , which is simply because the Ag^+ diffusion is suppressed at low temperatures (see Paper I).

The spot-size dependence shown in Fig. 2 can also be understood. Here, the fringe pitch ($\sim 2 \mu\text{m}$) and the effective diffusion length [$\sim 1 \mu\text{m}$ (Paper I)] of Ag^+ ions, which seem to be comparable, probably play decisive roles in the deformation shapes. If the size of light spots is smaller than or comparable to these lengths, the fringe cannot be produced, and accordingly, the Ag^+ -ion accumulation appears as the volume expansion (Paper I). On the other hand, if the spot size is much larger, interference fringes of light can emerge in thin samples, which can produce the Ag-content modulation, resulting in the expansive forces.

Further studies such as quantitative estimations of the generated force are needed for understanding this patterning mechanism more deeply.

V. SUMMARY

In Paper I and the present one, it has been demonstrated that, in the ion-conducting amorphous semiconductor Ag-As-S, light illumination can produce surface deformations.

The shape changes with experimental conditions such as the sample thickness, the size of light beams, and the direction of light incidence.

Specifically, the present work demonstrates that linearly polarized 2.0 eV light beams with spot sizes of 5–200 μm can produce anisotropic deformations in AgAsS_2 films. The shape changes with the direction of light incidence. When the beam illuminates the film from the free surface and through a transparent substrate, the shape is crater- and anti-craterlike, respectively. The mechanisms producing these anisotropic deformations are not conclusive, while in essence the deformations seem to be caused by a photo-electro-ionic process. Namely, when the spot size is large, interference fringes of light can appear in thin samples, which can produce the Ag-content modulations, resulting in the anisotropic deformations.

Little, if anything, is known about the generation of such exotic surface deformations in solid films by light illumination alone. Further studies on the mechanisms and applications will be valuable.

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