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Ripples in amorphous chalcogenide films under homogeneous laser illumination

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ABSTRACT

Under homogeneous illumination of thin chalcogenide glass films by polarized light at a wavelength near the band gap ripples form, with a period of the order of 10–15 μm, directed normal to the light polarization. The formation of the ripples cannot be explained by interference phenomena, which predict the ripple periods of the order of light wavelength. Our experimental and theoretical studies of the ripple formation in 1 μm thick $As_{10}Se_{90}$ chalcogenide films show that the profile variation occurs due to lateral mass transport accelerated by light. The ripple formation is caused by competition between capillary forces and steady state electrostatic forces induced by redistribution of electrons and holes generated by light. Under these driving forces, each harmonic of the film roughness spectrum should exponentially grow or flatten, depending on its frequency. The average period of the ripples corresponds to those harmonics in the roughness spectra, which grow with maximum rate. Light-induced diffusion coefficients have been estimated from the kinetics of the ripple formation.

1. Introduction

Chalcogenide glasses (ChGs) are well-known materials for applications in micro-optics, integrated optics, and IR spectroscopy [1–3]. ChGs exhibit a number of remarkable structural and optical changes when exposed to light or electron beams [4–7]. Among these phenomena, there is an increasing interest in photo-induced mass transport (MT) in thin ChG films [8–12], which is used for dry patterning of the films by laser or electron beams and the fabrication of various optical components by a one-step technique [13,14]. In recent years, the mechanisms and kinetics of the MT were studied both theoretically and experimentally [15–17], however, many details of the process remain unclear.

This paper addresses one of the unsolved mysteries, namely, the formation of ripples on the surface of ChG films under homogeneous illumination by light at the band gap wavelength. The ripples lead to morphological instability of the films that affect their properties necessary for technical applications.

Previously, ripples were observed on metal, semiconductor, and dielectric surfaces after nano-, pico- and femtosecond pulsed laser irradiation [18–21]. They had periods of the order of the laser wavelengths and the mechanisms of their formation were explained by the interference of the incident laser and a scattered surface optical wave.

The ripples observed on the surface of ChG films (As_xSe_{1-x} [22] and As_2S_3 [23]) under cw illumination by unfocused band gap light have spatial periods of 10–20 μm depending on the exposure time and the film thickness. The nature of these large ripples, as well as the mechanism of their formation has not been properly explained previously.

We study here the kinetics and mechanisms of the ripple formation in $As_{10}Se_{90}$ ChG films which show the fastest MT kinetics compared to other film compositions [22].

2. Experimental

$As_{10}Se_{90}$ films of 1 μm thick were prepared by in vacuum (10^{-5} Torr) thermal deposition on microscope glass slides. The films were illuminated for various times by a polarized diode laser ($\lambda = 660$ nm) passing through a lens with focal distance of 23 cm. The films were located near the focal plane of the lens, so that the illuminated area was about 2×10^3 μm^2. The laser power densities varied from 10^4 to 10^5 W/cm^2 and time of illumination varied from 5 min to 1 h, depending on the power density. The surface profiles formed after illumination were photographed on an optical

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microscope and scanned by atomic force microscope (AFM).

3. Results

The main results consist of the following.

(1) Inside the illuminated spot we observe ripples directed perpendicular to the light polarization (Fig. 1).
(2) Initial ripple periods seen in optical microscope were about 5–12 μm; the period and the height of the ripples grew with exposure time until the ripple depth reached the substrate.

(iii) AFM studies of initial stages of the ripple formation show that rough wrinkles consist of smaller scale roughness with characteristic distance about 2 μm (Fig. 2a), and this distance increases with time due to coarsening of the profile (Fig. 2b). Considering the surface profile as a Fourier integral of harmonics with various wavenumbers, q, i.e.

$$z(x) = \int_0^\infty h(q)\cos qx \; dq, \tag{1}$$

the coarsening occurs as a result of mass redistribution with the disappearance of harmonics with higher q and the growth of harmonics with smaller q (Fig. 2c). As MT induced by light is anisotropic and occurs mainly in the direction of the light polarization, the ripples are directed perpendicular to the polarization.

4. Discussion

We suggest that formation of ripples under homogeneous illumination is initiated by random surface roughness of the films. Due to roughness, the film thickness varies locally, and this variation leads to the emergence of driving forces of lateral mass transfer induced by band gap light illumination. As the random roughness can be represented by Eq. (1) as a set of harmonics with various wavenumbers, q, we first analyze how homogeneous illumination affects a *sinusoidal profile*.

If the amplitude of the harmonic with wavenumber q is $h(t)$ after exposure time t, the film thickness varies with coordinate x (perpendicular to the polarization) as

$$H(x, t) = H_0 + h(t)\cos qx \tag{2}$$

where H_0 is the average film thickness.

$As_{10}Se_{90}$ is a photoconductive p-type semiconductor, in which

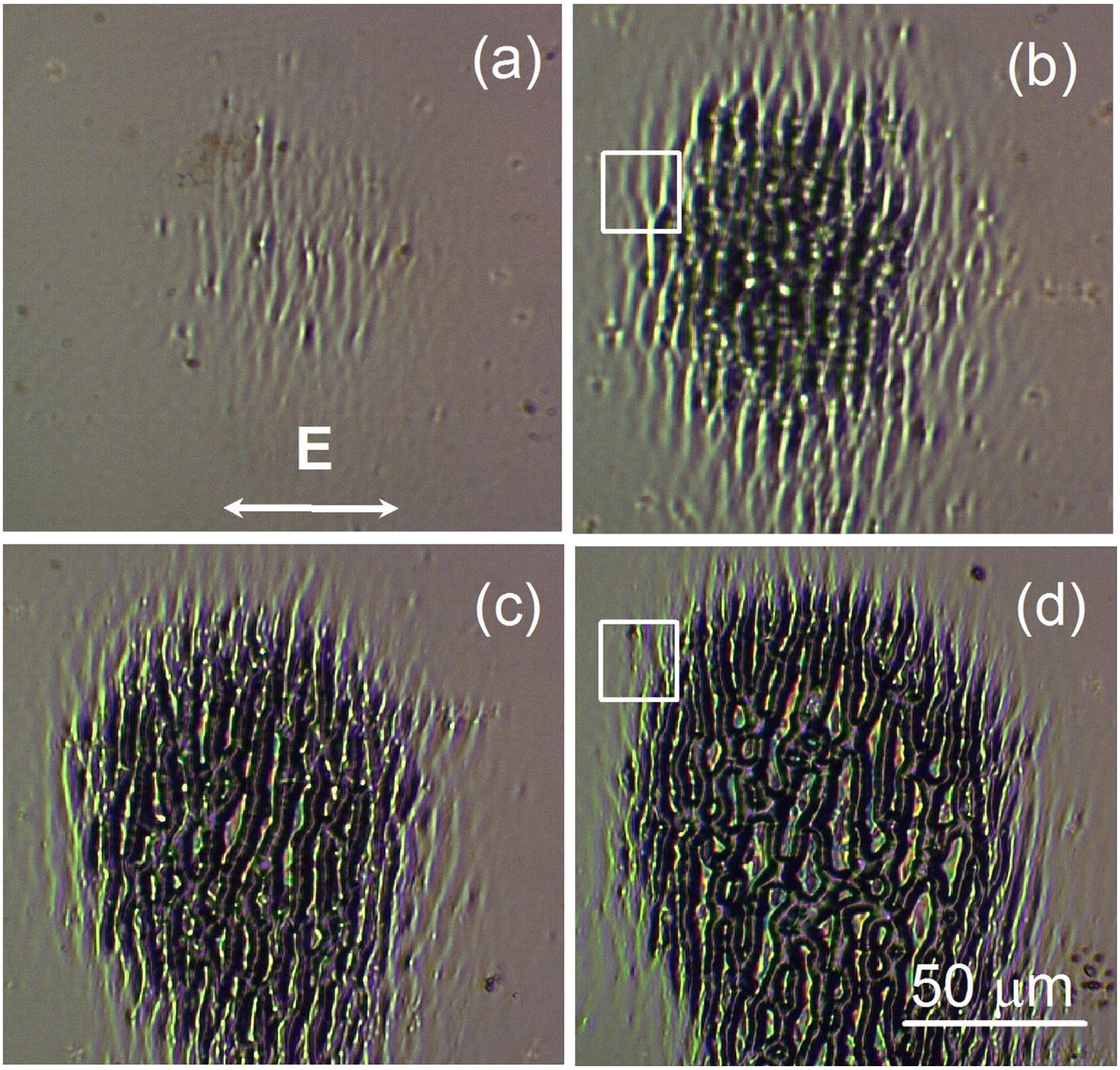

Fig. 1. Typical optical images of ripples produced in 1 μm thick $As_{10}Se_{90}$ films. Laser power was 30 mW, exposure times: (a) – 5 min, (b) – 11 min, (c) – 16 min, (d) – 27 min. Light polarization was horizontal [see arrow in (a)].

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