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Optics Communications 259 (2006) 545–552
OPTICS COMMUNICATIONS
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Theoretical consideration of pits recording and etching processes in chalcogenide vitreous semiconductors

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Received 10 May 2005; received in revised form 9 August 2005; accepted 12 September 2005

Abstract

We propose theoretical consideration, computer modeling and comparison with our recent experimental results for information pits recording and etching processes in chalcogenide vitreous semiconductors using Gaussian laser beam and selective etching. Our calculations demonstrate that photo-transformed region cross-section could be almost trapezoidal or parabolic depending on the photoresist material optical absorption, exposure, etchant selectivity and etching time. Thus our approach open possibilities how to select the necessary recording procedure and etching conditions in order to obtain pits with the optimum shape and sizes in $As_{40}S_{60}$ chalcogenide semiconductor. Obtained results quantitatively describe the characteristics of pits recorded by the Gaussian laser beam in thin film of $As_{40}S_{60}$.
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PACS: 78.20.Ci; 78.66.Jg

Keywords: Information pits; Optical disks; Photoresist; Selective etching

1. Introduction

Laser recording of profiled microstructures is widely used in modern manufacturing of optical compact disks (CD) [1–4] and originals of diffraction elements [5], synthesized holograms, etc. Numerous experimental [2–5] and theoretical investigations [6–8] devoted to the information recording and reading by optical methods have already been performed. Despite plethora of achievements [1,9–12], the great amount of different problems concerning recorded information quality and its density increase still remains unsolved. It was shown [2–4] that using the inorganic resist based on the ternary $As_{40}S_{60-x}Se_x$ chalcogenides allows to obtain the profiled structures with sizes much smaller than the diameter of the recording focused laser beam with wavelengths $\lambda = (436, \ldots, 532)$ nm. Optical properties and structure of thermally evaporated amorphous $As_{40}S_{60-x}Se_x$ thin films are studied in [13,14].

This paper is devoted to the consideration of the question how to record pits with the necessary depth profile in photosensitive materials by varying recording Gaussian beam energy density and exposure time, as well as the etchant selectivity and etching time. We used the following model of photoinduced structural transformations in the photoresist of As–S–Se type:

- Photons absorption with $\lambda \leqslant 630$ nm causes switching of the –As–S– bonds in amorphous film (see also p. 141 in [13]) and local polymerization of the nearest molecular group into the web of AsS_3 pyramids, i.e. *photo-structural transformations* appeared [9,10].
- Rather intensive focused laser light beam typical for CD recording causes significant thermal heating of the exposed region, which strongly influences on the photo-polymerization rate and even could cause polymerization itself, i.e. *thermo-structural transformations* appeared [9,11].

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doi:10.1016/j.optcom.2005.09.037

- Keeping in mind two aforementioned effects hereinafter the exposed region is considered as *photo–thermo-transformed region* with definite spatial distribution of the transformed material amount.
- In the course of illumination photosensitive material is quasi-equilibrium microscopic mixture of the *transformed* and *non-transformed* phases with relative fractions M and $(1-M)$, respectively. These phases have different photo-chemical properties and the optical absorption coefficients [4,12].

2. Fundamental equations

2.1. Modeling of photo-structural and thermo-structural transformations

When the focused laser beam illuminates the photosensitive layer, the optical absorption in the layer $0 < z < \ell$ is described by the Bouguer–Lambert–Beer law:

$$\frac{\partial I(x,y,z,t)}{\partial z} = -\alpha(x,y,z,t) I_0(x,y,t), \quad z \geqslant 0, \quad 0 \leqslant t \leqslant t_H. \tag{1}$$

Hereinafter α is optical absorption coefficient, $I_0(x,y,t) = I(x,y,0,t)$ is the incident intensity of the laser beam focused onto the surface $z = 0$, t_H is exposure time. In accordance with the adopted assumption [4,12], the absorption coefficient α is as follows:

$$\alpha(x,y,z,t) = \alpha_e \cdot M(x,y,z,t) + \alpha_0 (1 - M(x,y,z,t)). \tag{2}$$

Optical absorption coefficients α_e and α_0 corresponds to the transformed and non-transformed phases with relative fractions M and $(1-M)$. Hereinafter $0 < M(x,y,z,t) < 1$. It is obvious that α depends on the exposing beam wavelength λ, intensity, temperature and exposure time t_H owing to its dependence on M. The system (1) can be rewritten using the light transmission coefficient β, namely:

$$I(x,y,z,t) = \beta(M,z) \cdot I_0(x,y,t), \tag{3}$$

$$\beta(M,z) = \exp\left[-\alpha_0 \cdot z - (\alpha_e - \alpha_0) \cdot \int_0^z M(x,y,\xi,t)\, \mathrm{d}\xi\right]. \tag{4}$$

The total fraction of polymerized material M could be calculated using the *Kolmogorov–Avrami* nucleation model [15,1], in a self-consistent manner [8], namely:

$$M(x,y,z,t_H) \approx 1 - \exp\left(-\int_0^{t_H} \frac{\mathrm{d}\tau}{\tau_m} \cdot \frac{I(x,y,z,\tau)}{I_\mathrm{P}} \times \exp\left(-\frac{E_\mathrm{a}}{k_\mathrm{B} T(x,y,z,\tau)}\right)\right). \tag{5}$$

Here $\tau_m(\lambda)$ is the *nucleation specific time*, which depends on the specific bulk nucleation velocity (see [1, p. 84]), I_P has the meaning of the *intensity threshold*, any increase above which can provide registration of sizable amounts of polymerized material in the recording medium [9], E_a is the activation energy, $T(x,y,z,t)$ is the temperature distribution inside photoresist. The temperature distribution $T(x,y,z,t)$ is determined by the light absorption [1,8,9]. Dissipation of the thermal flux during the exposure time t_H could be neglected, if the characteristic penetration depth of the thermal flux $\chi_\mathrm{T} \sim \sqrt{t_H}$ is higher than the film thickness h. In this case, the stationary temperature distribution exists [8], namely:

$$T(x,y,z,t) = T_0 + \Delta T(x,y,z,t),$$
$$\Delta T(x,y,z,t) \approx \frac{I(x,y,z,t)}{I_\mathrm{P}} \frac{E_\mathrm{a}}{k_\mathrm{B}}. \tag{6}$$

Hereinafter let us consider the typical situation when the focused Gaussian laser beam with dispersion ρ_0 moves along Y-axis at the distance d with the constant velocity ϑ. Our numerical calculations [8] proved that the photo–thermo-transformed region cross-section $M_0(x,z) \equiv M(x,0,z,t_H)$ could be estimated by the iteration method starting from the point $z = 0$:

$$M_0(x,z=0) \approx 1 - \exp\left(-\frac{H(x)}{H_0} \exp\left(-\frac{E_\mathrm{a}}{k_\mathrm{B} T(x)}\right)\right),$$
$$H(x) = H_\mathrm{m} \exp\left(-\frac{x^2}{\rho_0^2}\right), \quad T(x) = T_0\left(1 + \frac{H(x)}{H_\mathrm{T}}\right). \tag{7}$$

Here ρ_0 is the recording beam dispersion, $H_\mathrm{m} \approx I_\mathrm{m} t_H \cdot (\sqrt{\pi}\rho_0/\vartheta)$ is the maximum exposure (I_m is the recording beam maximum intensity). Let us consider the characteristic exposures $H_0 = \tau_\mathrm{m} I_\mathrm{P}$ and $H_\mathrm{T} = I_\mathrm{P} t_H \cdot (k_\mathrm{B} T_0/E_\mathrm{a})$ as the exposure *photo-* and *thermo-threshold*. Note, that exposure H_T is inversely proportional to the activation energy, and the quantity $H(x,0)$ can be regarded as the *exposure distribution* at $z = 0$.

It is noteworthy, that at $H_\mathrm{T} \ll H_0$ significant thermal heating occurs even at small exposures H_m, and as a result visible structural transformations starts inside the exposed region (see the light-blue curve in Fig. 1). In the typical for the most recording materials case $H_\mathrm{T} \geqslant H_0$ only weak photo-structural transformations take place at small

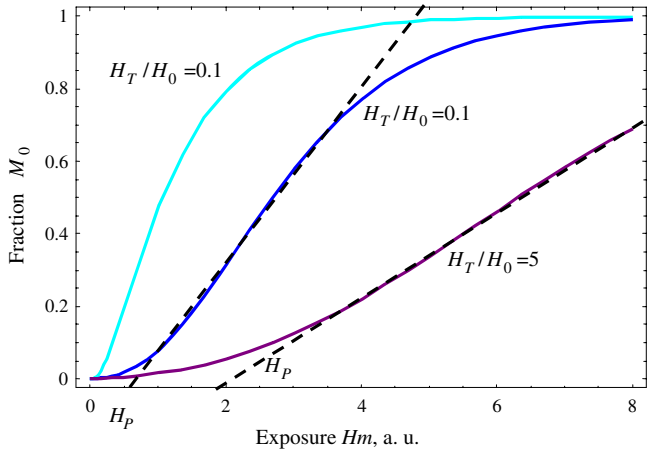

Fig. 1. Fraction $M_0(0,0,H_\mathrm{m})$ from (7) vs. the exposure H_m/H_0, at $E_\mathrm{a}/k_\mathrm{B}T_0 = 5$ and different H_T/H_0 ratios: 0.1 (light blue), 1 (dark-blue), 5 (violet). (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

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