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Thin Solid Films 493 (2005) 207 – 211
thin solid films
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Nonlinear optical properties of As_2Se_3 thin films

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Received 14 April 2004; received in revised form 20 May 2005; accepted 8 August 2005
Available online 12 September 2005

Abstract

We propose As_2Se_3 thin film, which has large third nonlinear optical properties, as one of the candidates for super-resolution near-field structure (super-RENS) materials in the optical recording disks. As_2Se_3 thin film was deposited by thermal evaporation technique and surface roughness, linear/nonlinear optical properties were characterized. As_2Se_3 thin film shows large nonlinear optical characteristics ($\chi_R^{(3)}=7.42\times10^{-7}$ m^2/W, $\gamma=1.268\times10^{-5}$ m^2/W) and self-focusing phenomenon. Super-resolution effect was measured by a laser beam profiler. As a result, recording density can be enhanced by 2.3 times in case when 300 and 350 nm As_2Se_3 thin films are applied as a super-RENS.

PACS: 81.15.E; 42.70.L; 42.79.W; 78.20
Keywords: Evaporation; Selenides; Optical coating; Optical properties

1. Introduction

The third order nonlinear optical properties of chalcogenide glasses have received much attention with regard to their ability to be used in various optical systems for ultrafast optical applications [1]. Among the nonlinear optical properties, self-focusing effect has received significant interest for reducing the spot size of the laser beam beyond the diffraction limit. Therefore, chalcogenides which show self-focusing behavior might be a candidate for super-RENS in optical recording disks.

Tominaga et al. proposed Sb mask layer which is sandwiched between two dielectric layers as a super-RENS [2]. The near-field effect occurred from Sb mask layer reduced the spot size of the laser beam beyond the diffraction limit. To reach the ultrahigh recording density, researchers have investigated various super-RENS technique [3]. However, these super-resolution materials have some problems such as slow response time and low CNR (carrier to noise ratio), since the super-resolution phenomenon occurred by melting or chemical decomposition of a super-RENS materials [4]. Therefore, we propose that using nonlinear optical properties, namely self-focusing effect, of chalcogenide thin film can be a solution of aforementioned problems.

In this paper, As_2Se_3 film was introduced for super-RENS, since it has very large nonlinear susceptibility [5]. To investigate the nonlinear optical properties of As_2Se_3 film, we have performed z-scan measurements at wavelength of 633 nm. Although many important features of the third nonlinear optical properties of As_2Se_3 have been reported, self-focusing behavior and intensity profile of self-focused laser beam were not reported yet. Therefore, laser beam profiler was adapted to investigate the self-focusing behavior and observe the intensity variation of laser beam profile.

2. Experimental details

The bulk As_2Se_3 was prepared by introducing stoichiometric amount of arsenic (99.99%, Sterm chemicals, Japan)

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doi:10.1016/j.tsf.2005.08.007

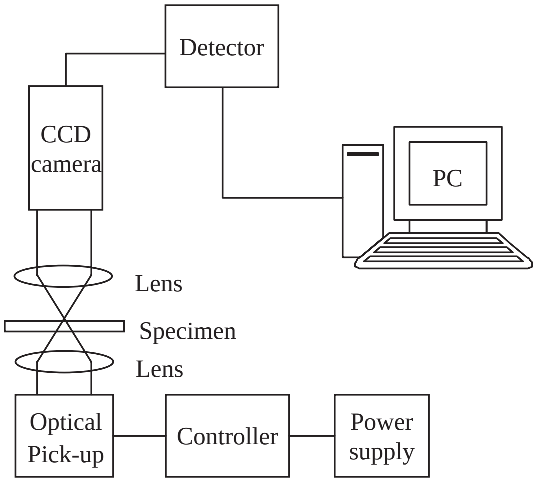

Fig. 1. Schematic drawing of the laser beam profiler.

and selenium (99.99%, High purity chemicals, Japan) into a quartz ampoule which was sealed under 1.3×10^{-3} Pa vacuum. The sealed ampoule was melted at 800 °C for 24 h using rocking furnace to homogenize the liquid. The ampoule was quenched in air and glass transition temperature (T_g) was determined by differential scanning calorimetry (DSC, STA-1500/DSC-SP, U.S.A.) with heating rate of 5 K/min. The chalcogenide glass was annealed at T_g for 15 min and used as a thermal evaporation source. As_2Se_3 film was prepared by thermal evaporation technique using W boat and the base pressure of the chamber was 6.6×10^{-4} Pa. The films were deposited on (100) Si wafer and alkali-borosilicate glass substrate at room temperature with deposition rate of 30 nm/min. The thickness of the As_2Se_3 thin films were 100, 200, 250, 300, 350, 400 and 500 nm, respectively.

X-ray diffraction (XRD, Rigaku, D-Max, Japan) was conducted with Cu K_α radiation to identify the structure of bulk As_2Se_3 and the film. The composition of thin films was confirmed by electron probe micro analyzer (JXA-8900R, JEOL, Japan). Surface roughness of the films was observed by atomic force microscope (AFM, SPM-9500J, Shimazu, Japan). AFM was measured with contact mode using SiN probe. The scanned surface and the scan rate were 1×1 μm^2 and 1 Hz, respectively. Optical transmittance of the film was measured by spectrophotometer (V-5700, JASCO, Japan) with a resolution of 1 nm. Refractive index (n) and extinction coefficient (k) of the films were determined by variable angle spectroscopic ellipsometer (VASE, VU-302 J.A.Woollam, U.S.A.).

Nonlinear optical properties were measured by standard z-scan set up which is described by Sheik Bahae et al. [6]. Continuous wave (CW) He–Ne laser which generates a wavelength of 633 nm and a power of 99.5 kW/cm^2 was used and a lens with a focal length of 1.7 mm led to 1.69×10^3 nm spot radius at the focal point. The z-scan measurements were carried out with scan distance 400×10^3 nm by 100 step. The z-scan experiment was conducted with open aperture mode and closed aperture mode. Then the self-focusing behavior of the film was explained by the numerical analysis.

The spot size of the laser beam was measured by laser beam profiler, which is depicted in Fig. 1. Laser beam profiler was set up to measure the spatial laser intensity distribution and equipped a charge coupled device (CCD) detector. Optical pick-up which was used for commercial optical disk drive (SH-W08A, Samsung, Korea) was adapted as a laser source. Optical pick-up was composed with lens with numerical aperture (N.A.) of 0.6 and laser diode which generates a wavelength of 650 nm and a power of 40 mW. Reference spot size of the laser beam was measured with bare glass substrate and the variation of the spot size was measured to optimize the film thickness of As_2Se_3. In order to determine the width of X-direction, Y-direction and average diameter of the laser beam spot, quantitative 90/10 knife-edge of the laser beam profiler was set as a base line.

3. Results and discussions

The structure of bulk As_2Se_3 and the film was investigated by XRD. Three humps were observed at 17.5°, 32.0° and 55° for the bulk As_2Se_3 and the film, which is typical XRD pattern for amorphous chalcogenides. From the result of DSC measurement with the heating rate of 5 K/min, T_g of bulk As_2Se_3 was about 176 °C.

Surface roughness of super-RENS layer is an important parameter in fabrication process of optical recording disks, since the surface roughness of bottom layer (super-RENS layer) can affect the roughness of other layer such as recording layer, protective layer and reflective layer. Root-mean-square (RMS) roughness was estimated by AFM.

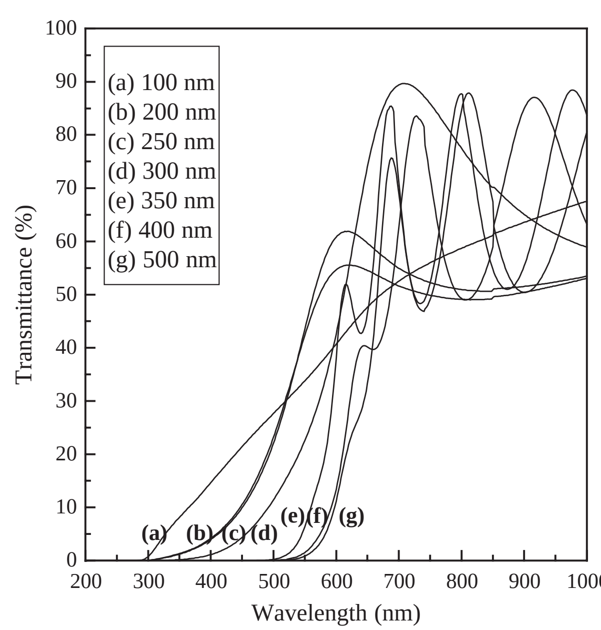

Fig. 2. Transmission spectra of As_2Se_3 with different thickness.