Z-Scan Study of Nonlinear Optical Coupling of PtO_x and $\text{Ge}_2\text{Sb}_2\text{Te}_5$ of Near-Field Optical Recording Structure

Yuan Hsing FU¹, Yan Lan LU¹, Pei Hsin CHANG^{1,2}, Wei-Chih HSU⁴, Song-Yeu TSAI⁴ and Din Ping TSAI^{1,2,3*}

¹Department of Physics and Center for Nanostorage Research, National Taiwan University, Taipei 106, Taiwan

²Institute of Electro-optical Science and Technology, National Taiwan Normal University, Taipei 116, Taiwan

³Research Center for Applied Sciences, Academia Sinica, Taipei 115, Taiwan

⁴Materials Research Laboratories, Industrial Technology Research Institute, Hsinchu 310, Taiwan

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The strong local nonlinear optical coupling properties of a 5 nm PtO_x thin film sandwiched by two $\text{Ge}_2\text{Sb}_2\text{Te}_5$ phase-change structures of a new type super-resolution near-field optical disk are investigated and observed by Z-scan experiments.

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1. Introduction

The field of near-field optical recording using a near-field optical active layer is currently relevant to ultrahigh-density optical disk recording. The use of a near-field optical disk with a near-field optical active layer has the advantages of simpler recording head design, less mechanical damage, and higher recording speed over that of the near-field optical recording method using an optical fiber probe and a near-field optical pickup head. The as-deposited Sb thin film was first introduced by Tominaga *et al.* as a near-field active layer in 1998,¹⁾ and subsequently the metallic oxide active layers, such as AgO_x , WO_x , PdO_x , PtO_x , and ZnO_x , were used to obtain high carrier-to-noise ratio (CNR) signals.^{2–5)} Nonlinear optical properties were observed in research on AgO_x using near-field optical microscopy (NSOM) and the Z-scan method.^{6,7)} The enhancement in local optical intensity in a near field and the nonlinearity of such enhancement, which is related to a localized surface plasmon (LSP) have been studied and calculated using the finite differential time domain (FDTD) method.⁸⁾ Recently, the enhancement of signals in terms of CNR induced by elliptical bubble formation with PtO_x and AgInSbTe has been reported.⁹⁾ The decomposition and optical thermal properties of the PtO_x layer have been investigated.^{10,11)} However, the effect of the coupling between the PtO_x and phase-change recording layers has not yet been studied or discussed in detail. In this study, the nonlinear optical coupling properties of PtO_x layer and phase-change layers are investigated by the Z-scan method with a nanosecond pulse laser to prevent the complication of thermal accumulation. The experimental results for a sandwiched $\text{Ge}_2\text{Sb}_2\text{Te}_5$ phase-change layer, various sandwiched PtO_x [$\text{O}_2/(\text{Ar} + \text{O}_2) = 0.5, 0.2, \text{ and } 0.05$] metallic oxide layers, and a PtO_x metallic oxide layer sandwiched by two sandwiched $\text{Ge}_2\text{Sb}_2\text{Te}_5$ phase-change layers are studied and compared.

2. Experimental Methods

2.1 Sample preparation

The structures of three different samples are shown in Figs. 1(a)–1(c). A ZnS-SiO_2 thin film is employed as protective and spacer layers for all the samples. The

$\text{Ge}_2\text{Sb}_2\text{Te}_5$ phase-change recording layers in Figs. 1(a) and 1(c) are consist of as-deposited thin films. A PtO_x thin film layer is deposited by sputtering with a pure Pt target in a gas mixture of Ar and O_2 . The ratio of Pt to O_2 can be controlled by adjusting the partial pressure of O_2 . Figures 1(a) and 1(b) show the sandwiched samples of the 5 nm $\text{Ge}_2\text{Sb}_2\text{Te}_5$ phase-change layer and PtO_x thin film, respectively. Figure 1(c) shows a PtO_x metallic oxide layer sandwiched by two sandwiched $\text{Ge}_2\text{Sb}_2\text{Te}_5$ phase-change layers.⁹⁾

2.2 Optical measurement

The schematic of the experimental setup of the Z-scan system is shown in Fig. 2. The light source is a Q-switched Nd:YAG semiconductor pulse laser with a wavelength of 532 nm, a pulse width of 0.71 ns, and a repetition rate of 15.79 kHz. To study the nonlinear optical properties in a microscopic region, a Mistutoyo long-working-distance ($\text{WD} = 13 \text{ mm}$), high-numerical-aperture ($\text{NA} = 0.55$) objective lens is used to focus the laser beam. The transmission signals of the open and closed apertures can be detected by the D2 and D3 photodetectors, respectively. The reflective signal detected by the D4 photodetector could be used to determine the readout mechanism of our samples.¹²⁾ An adjustable attenuator is used to control incident laser power from 10 to 100 μW , and the incident laser power at the sample surface is monitored using an optical power meter (D1). In the Z-scan system, the sample scanning in the focal region is precisely positioned using a motorized stage along the optical axial direction (Z-axis). The sample is placed with the thin film layer facing the collecting lens. The positive scanning direction is used to move the sample from the focusing objective side toward the collection lens in 1 μm steps. Optical transmittance and reflectance can be simultaneously obtained in this experimental setup. The results of the measurements can provide the coefficient of absorption and the nonlinear refractive index of a sample simultaneously.¹³⁾

3. Results and Discussion

Figure 3(a) show the Z-scan experimental results of transmittance and reflectance at various incident laser powers of the sandwiched 5 nm $\text{Ge}_2\text{Sb}_2\text{Te}_5$ phase-change layer shown in Fig. 1(a). Figure 3(b) shows the normalized transmittance and reflectance at a focal point at different

*E-mail address: dptsai@phys.ntu.edu.tw

Fig. 1. Three different sample structures: (a) sandwiched Ge₂Sb₂Te₅ phase-change layer; (b) sandwiched PtO_x metallic oxide layer; and (c) PtO_x metallic oxide layer triple-sandwiched by two sandwiched Ge₂Sb₂Te₅ phase-change layers. The Ge₂Sb₂Te₅ phase-change layer of (a) and (c) is composed of an as-deposited thin film.

Fig. 2. Schematic of experimental setup of Z-scan measurements.

input powers. The normalized transmittance shown in Fig. 3(b) is the measured transmittance at the focal point divided by the background transmittance out of the focal point (T_p/T_0). The normalized reflectance is the measured reflectance at the focal position of the thin film layer divided by the reflectance that occurs at the air-glass interface (R_p/R_0). Position 0 shown in Fig. 3(a) is set to be at the focal position on the thin film layer. For the reflectance results shown in Fig. 3(a), the two peaks at the positions of 25 μ m and 125 μ m resulted from the reflections that occurred at the interfaces of the film-glass and air-glass, respectively. The distance between the two peaks confirms the thickness of the glass substrate, i.e., 0.12 mm. The normalized reflectance shown in Fig. 3(b) is lower than 1, which is lower than that of the interface of air-glass. Figure 3(a) shows that the reflectance dips at position 0 were possibly caused by the change of the as-deposited Ge₂Sb₂Te₅ thin film into an

amorphous state.¹⁴⁾ On the other hand, the peak transmittances shown in Fig. 3(a) indicate a gradual increase to saturation from 130 to 160% in the background transmittance shown in Fig. 3(b). In the mean time, the widths of the peaks shown in Fig. 3(a) broaden with the increase in incident average power. These results clearly indicate the area of the change from the as-deposited state into the amorphous state of Ge₂Sb₂Te₅ at the focus spot gets larger. The Z-scan transmission peaks shown in the Fig. 3(a) demonstrate the convolution of the laser focusing spot and the state-changed area of the sandwiched Ge₂Sb₂Te₅ thin film. Note that, in Fig. 3(b), the reflectance at the focusing position indicates a three-state change from 10 to 100 μ W.

For the 5 nm PtO_x [fabricated at O₂/(Ar + O₂) partial pressure of 0.5] sandwiched sample shown in Fig. 1(b), typical Z-scan experimental results are shown in Fig. 4(a). The transmittance peaks shown in Fig. 4(a) indicate a

Fig. 3. Results of Z-scan experiments of sandwiched $\text{Ge}_3\text{Sb}_2\text{Te}_5$ phase change layer as shown in Fig. 1(a). (a) shows plots of the transmittance and reflectance at each position for five different incident laser power. (b) shows plots of normalized transmittances and reflectances at the transmittance peaks and reflectance dip at input laser average powers from 10 to 100 μW .

Fig. 4. Results of Z-scan experiments of sandwiched PtO_x metallic oxide layer as shown in Fig. 1(b). (a) shows plots of the transmittance and reflectance at each position for five different incident laser power. (b) shows plots of normalized transmittances and reflectances at the transmittance peaks and reflectance dip at input laser average powers from 10 to 100 μW .

gradual saturated increase in background transmittance from approximately 100 to 140% as shown in Fig. 4(b). The widths of the transmittance peaks shown in Fig. 4(a) are in contrast with that of Fig. 3(a), where no significant broadening has been found for the increase in the incident laser power. This is due to the different mechanisms of the optical interactions of the sandwiched thin film. The peak reflectances shown in Fig. 4(a) can be three fold higher than that of the glass–air interface. Normalized transmittances and reflectances are plotted in Fig. 4(b) similarly to those shown in Fig. 3(b). The results show that only a two-state change occurred in the three PtO_x sandwiched samples at different $\text{O}_2/(\text{Ar} + \text{O}_2)$ partial pressure values, i.e., 0.05, 0.2, and 0.5, during their fabrication. In the low-incident-laser-power region, from 10 to 30 μW , as shown in Fig. 4(b), the

reflectances of the dips shown in Fig. 4(a) are higher than those at the air–glass interface. This possibly resulted from the decomposition of the PtO_x thin film into Pt nanoparticles, and the aggregation of Pt nano particles at the center of the focusing laser spot. When the incident laser power is above 40 μW as shown in Fig. 4(b), normalized reflectance approaches a value below 1.0. It is reported that a possibly hollow-ring structure formed in the PtO_x thin film under exposure to a focused laser similarly to that which occurred in the sandwiched AgO_x thin film.¹⁵⁾ A related study performed by Liu *et al.*¹¹⁾ indicates that local bubbles are produced and the optical interactions of these bubbles possibly assist in the enhancement in CNR for nanomarks. However, note that the differences caused by various partial pressures of $\text{O}_2/(\text{Ar} + \text{O}_2)$ during the fabrication of the PtO_x

Fig. 5. Results of Z-scan experiments of triple-sandwiched 5 nm PtO_x thin film as shown in Fig. 1(c). (a) shows plots of the transmittance and reflectance at each position for five different incident laser power. (b) shows plots of the plot of normalized transmittances and reflectances at the transmittance peaks and reflectance dip at input laser average powers from 10 to 100 μW.

thin film indicate the possibility of controlling the reflectance and transmittance at laser powers below 40 μW.

For the 5 nm PtO_x thin film triple-sandwiched by two sandwiched 12 nm Ge₂Sb₂Te₅ phase-change layers as shown in Fig. 1(c), Fig. 5(a) shows that the widths of the transmittance peaks at the focus are smaller than the width of those shown in Figs. 3(a) and 4(a), but the peaks have clear shoulders at incident laser powers above 60 μW. These results indicate that the interaction area is narrowed down by the coupling of the multilayered structure. Note that extra reflectance peaks appeared exactly at the focusing dip positions shown in Fig. 5(a) at laser powers above 60 μW. Figure 5(b) shows the results of this sample similarly to those shown in Figs. 3(b) and 4(b). The results show a two-step change process similar to that in Fig. 4(b), but with the threshold of the incident laser power increasing from 40 to 50 μW. Normalized reflectance increases markedly with input laser power, in contrast to the results of the sandwiched PtO_x thin film alone. Figure 5(b) also shows that the slopes of the curves of normalized reflectance for different laser incident power are related to the partial pressure of O₂. The O₂/(Ar + O₂) partial pressure of 0.5 yields the highest slope among the three different cases. The enhancement of reflection signal may be caused by both the scattering of the Pt nanoparticles and the surface plasmon coupling between the Ge₂Sb₂Te₅ phase-change layers and the decomposed Pt from PtO_x layer.

4. Conclusions

Three samples, a sandwiched 5 nm Ge₂Sb₂Te₅ phase-change thin film, a sandwiched 5 nm PtO_x thin film and a 5 nm PtO_x thin film sandwiched by two sandwiched 12 nm Ge₂Sb₂Te₅ phase-change layers, were studied by Z-scan to determine the coupling effects between Ge₂Sb₂Te₅ and PtO_x thin films. The strong local nonlinear optical coupling properties of a 5 nm PtO_x thin film sandwiched by two sandwiched Ge₂Sb₂Te₅ phase-change structures were clearly observed in contrast with the results of the sandwiched 5 nm

Ge₂Sb₂Te₅ phase-change thin film and sandwiched 5 nm PtO_x thin film alone. The confinement of the coupling effect and local interactions of this triple-sandwiched structure for superresolution near-field optical disk applications were clearly found.

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