Real Time Optical-electrical Characterization of PbTe Thin Film Material for Super-resolution Optical Memory[†]

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An experimental study was conducted in an effort to elucidate the origin of the nonlinear optical properties of PbTe thin film, which previously had been demonstrated to be potentially usable for super-resolution optical memory. Using PbTe thin film device designed expressly for the experiment, transient optical transmittance and reflectance were measured together with the electrical voltage across the PbTe during pulsed irradiation with a 658 nm laser of varying power. From the measured data, the absorption coefficient was derived as well as the carrier concentration as a function of laser power, demonstrating vividly that a decrease in the absorption coefficient with increasing laser power is directly related to the increase in photo-generated carriers. The observed correlation underlying the nonlinear optical properties of PbTe may be understood in light of saturable absorption resulting from band filling by carriers photo-generated under thermal influence and is a major cause of the material's super-resolution effect.

Keywords: optical data storage, super-resolution (SR), PbTe, nonlinear optical property, thermoelectric, absorption saturation, band filling, photo-induced carriers

1. INTRODUCTION

Progress in optical disk technology has reached a stage where the conventional means of boosting storage density, *viz.* use of a higher NA lens combined with a laser source of a shorter wavelength, can no longer meet the continuing demand for higher density information storage. Among promising solutions under development, super-resolution optical memory is regarded as the most attractive due to its capability of increasing memory density while still maintaining major advantages of the present optical disk technology.

In super-resolution (SR) optical memory technology, a thin film layer known as the SR layer is embedded into a removable optical media so as to dynamically control the intensity profile of a moving laser spot to yield an aperture-like effect. SR techniques using various materials have been studied^[1-5], but neither development of practically usable materials nor understanding of the SR mechanisms, even for relatively promising materials such as AgInSbTe and GeS-bTe has been achieved thus far.

In our recent study, PbTe thin film was proposed as a new class of SR material of which the thermoelectric nature of

the crystalline solid material may be utilized to yield a large SR readout signal and high durability^[6]. However, little understanding has been attained as of yet, regarding the mechanisms responsible for the observed SR effect or the origin of the observed nonlinear optical characteristics.

Herein, a brief report is forwarded of our attempt to obtain direct evidence confirming an underlying optical-electronic correlation behind the aforementioned findings. In doing so, we carry out a real time optical-electrical characterization using a thin film device having PbTe so as to determine the relationship between changes in optical properties and photo-generated carriers.

2. EXPERIMENTS

The laser-induced changes in optical and electrical properties were measured simultaneously using the experimental setup and sample device shown in Fig. 1 and Fig. 2, respectively. While applying a fixed probe voltage (5V) to a series circuit made up of the sample device and an oscilloscope, an area of the sample device was irradiated with a 658 nm pulsed laser beam focused through an objective lens of 0.6 NA to a $1/e^2$ spot diameter of around 1.2 µm. Laserinduced voltage wave forms were recorded together with optical reflectance and transmittance monitored by a probe

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Fig. 1. Schematic diagram of the experimental setup for real time optical-electrical characterization.



Fig. 2. Schematic diagram of cross section and optical image of the device layout.

laser beam of another wavelength (633 nm). The sample device was fabricated using an optically transparent Corning glass substrate coated with a ZnS-SiO₂ sputtered film of 150 nm thickness. PbTe thin film layer was deposited to 15 nm thickness thereon by sputtering. From an RBS analysis, PbTe film was found to have a composition of Pb 49at.%-Te 51at.%. The PbTe layer was patterned by way of a photoli-thography combined with a lift-off process. Subsequently, Au film (400 nm) was deposited and patterned similarly so that a $4 \,\mu\text{m} \times 4 \,\mu\text{m}$ active area of the resulting device was exposed to laser irradiation. Finally, a ZnS-SiO₂ film layer (150 nm) was formed to protect the exposed PbTe film area. A plane-view optical image of the sample device under laser irradiation is shown in Fig. 2 together with a schematic cross-section view.

3. RESULTS & DISCUSSION

By use of the experimental setup of Fig. 1 and the sample device of Fig. 2, real time optical-electrical characterizations were carried out, and the results are summarized in Fig. 3 and Fig. 4. Shown in Fig. 3 are the temporal variations of optical transmittance and reflectance as measured with a 633 nm probe laser during pulsed irradiation of 1 μ s duration with a 658 nm pumping laser of varying power from 1 mW to 5 mW. It is emphasized that the observed optical changes



Fig. 3. Transient optical reflectance and transmittance due to real time optical-electrical characterization of PbTe.



Fig. 4. Transient oscilloscope voltage wave forms due to real time optical-electrical characterization of PbTe.

are entirely solid state phenomena, as determined in our previous work^[6,7]. Figure 4 shows the voltage wave forms recorded with the oscilloscope during the aforementioned optical measurements. A rapid initial increase of the oscilloscope voltage indicates a corresponding increase in the electric current of the series circuit, caused by laser irradiation of the PbTe device. Notice a considerable delay in time to peak and decay time of each voltage wave form compared with the respective optical signal, which may be ascribed essentially to a RC delay in the electric circuit. To obtain the results summarized in Fig. 6, the data shown in Fig. 3 and Fig. 4 were processed further as follows. From maximum transmittance and the corresponding minimum reflectance determined from Fig. 3 for varying power of the pump laser, the absorption coefficient (α) was derived by way of regressive multi-layer optics calculations^[7]. From the electric current of the series circuit, as determined by dividing the maximum oscilloscope voltage value in each curve of Fig. 4 by its resistance (1 M Ω), the laser-induced carrier concentration (N) was



Fig. 5. Schematic diagram illustrating the device geometry along with a current path.

derived as described below following the manner of Haynes-Shockley's experiment^[8]

Figure 5 is a schematic diagram illustrating the device geometry along with the current path. Electrical current flowing between the Au electrodes across the PbTe film and the PbTe/Au contact may be expressed as

$$i = Ne\mu_{P+C}E_{P+C}A_P \tag{1}$$

where μ_{P+C} and E_{P+C} represent the carrier mobility and electric field across the PbTe load and the contact resistance, respectively. For simplicity, the cross-section area of the current flow was assumed to be the same in both the PbTe film (A_p) and the contact (A_c) . The carrier mobility may be approximated as equation (2) assuming a planar contact $(L_c \sim 0)$.

$$\mu_{P+C} \sim \frac{L_P^2}{t_{P+C}V_{P+C}}$$
(2)

where L_p is the PbTe load length, t_{P+C} is the time taken to travel across the PbTe load and the contact resistance, and V_{P+C} is the device voltage, given as $L_p E_{P+C}$. By combining (1) and (2), equation (3) is obtained to express the carrier concentration N in terms of all the measurable parameters.

$$N = \frac{t_{P+C}i}{eA_PL_P} \tag{3}$$

 t_{P+C} was determined from the time delay (9 µs) between the *time to peak* in Fig. 4 (~10 µs for every laser power ranging from 2 mW to 5 mW) and that in Fig. 3 (1 µs). With appropriate parameter values for the present experiment, equations (2) and (3) were found to yield μ_{P+C} of 0.0213 cm²s⁻¹V⁻¹ and *N* values of the order of 10¹⁸ cm⁻³. For comparison, a separate Hall measurement was carried out at room temperature using a PbTe thin film of 100 nm thickness on a glass substrate coated with a ZnS-SiO₂ layer of 150 nm thickness, yielding 71.5 cm²s⁻¹V⁻¹ for Hall mobility and 3.74x 10¹⁶ cm⁻³ for carrier concentration. These huge differences may be ascribed to various factors in the device such as contact resistance, confined geometry of the current path including film thickness, laser heating, and a large population of photo-



Fig. 6. Variation of changes in absorption coefficient ($\Delta \alpha$) and carrier concentration (ΔN) with pumping laser power, derived from the data shown in Fig. 3 and Fig. 4.



Fig. 7. Variation of changes in refractive index (Δn) and extinction coefficient (Δk) as function of changes in carrier concentration, derived from the data shown in Fig. 3 and Fig. 6.

generated carriers.

Figure 6 presents a critical summary of the present experiment. With regard to the origin of nonlinear optical characteristics of PbTe thin film, it is vividly demonstrated that the decrease in the absorption coefficient with increasing laser power is directly related to the increase in concentration of photo-generated carriers. This result may be understood in light of saturable absorption resulting from band filling by carriers photo-generated under thermal influence, as suggested in our latest study^[9]. Figure 7 shows another representation of Fig. 6, supplemented with refractive index (n) data obtained from regressive multi-layer optics calculations together with extinction coefficient (k) or, equivalently, absorption coefficient (α) data. As in the case of the extinction coefficient, the refractive index displays a decreasing tendency as photo-generated carriers increase. This is in sharp contrast to a proposed model emphasizing the role of bound electrons in modulation of the refractive index with laser power to produce super-resolution effects in Sb-Te based crystalline materials^[10].

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4. CONCLUSIONS

Real time optical-electrical characterization was carried out in an effort to attain better understanding of the origin of the nonlinear optical characteristics of PbTe thin films for super-resolution optical memory technology. From analyses of optical transmittance and reflectance, simultaneously measured with electrical voltage wave forms for varying laser power, it was vividly demonstrated that the decrease in the absorption coefficient with increasing laser power is directly related to the increase in photo-generated carriers. Accordingly, it is concluded that the laser-induced carriers may play a key role in the realization of nonlinear optical characteristics in PbTe thin films as well as their super-resolution effects. The observed optical-electronic correlation appears also to be responsible, at least in part, for the superresolution effects of other Te-based degenerate semiconductors such as GeSbTe and AgInSbTe materials. This issue has been investigated by means of the same experimental technique utilized in the present work and will be reported elsewhere.

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