An experimental investigation on the origin of super-resolution effects of Te-based chalcogenide semiconducting thin films

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ABSTRACT

An experimental study was carried out to understand the origin of the super-resolution effects of three different chalcogenide thin film materials, namely $Ge_2Sb_2Te_5$, Ge-doped SbTe and PbTe. By way of a real time optical-electrical characterization involving measurement of optical reflectance/transmittance combined with measurement of electrical resistance during pulsed laser irradiation of varying power, a finding was made for all of three chalcogenide materials that decrease in absorption coefficient takes place with increasing pumping laser power, accompanied by decrease in electrical resistivity. It is suggested that the nonlinear optical effect responsible for the super-resolution capability of these chalcogenide materials may have an origin in common, presumably, saturable absorption due to band filling by thermally assisted photo-excited carriers, as proposed in our previous study for PbTe.

Key words: super-resolution (SR), PbTe, $Ge_2Sb_2Te_5$, Ge-doped SbTe, nonlinear optical effect, absorption saturation, band filling.

1. INTRODUCTION

The so-called super-resolution (SR) technique for optical discs is regarded very attractive because of its potential capability of achieving ultrahigh storage density while still using the present optical pickup system. In the SR technique, a thin film material layer deliberately embedded in an optical medium is made to control dynamically the intensity profile of an incident laser beam to produce an effectively reduced spot size for SR readout. Various materials have been studied to date [1-5] and SbTe-based chalcogenide materials such as AgInSbTe and Ge-doped SbTe have been demonstrated as most promising SR materials [4-6]. As for the causes of the SR effects of these materials, conventional melting phase change [1], ferroelectric phase transition [7] and thermoreflectance effect [8] have been proposed, yet no prevailing explanation is presently available.

In our recent study, PbTe was proposed as an SR material of which the thermoelectric nature of the crystalline solid state was presumed to produce a pronounced nonlinear optical effect that can be utilized to yield a large SR readout signal and a high cyclability as well.[9,10] Most recently, it was further suggested that the nonlinear optical effect of PbTe may be understood by taking into account the prevailing role of a thermal contribution within the framework of saturable absorption due to band filling by excited free carriers.[11] Since both PbTe and the aforementioned SbTe-based materials belong to the same class of Te-based degenerate semiconductors, we presume that the SR effects of these materials may be accountable on a common basis.

Herein, a report is forwarded of our investigation to examine the origin of the SR effects of these Te-based chalcogenide semiconducting thin films in light of a potentially common characteristic. A real time optical-electrical characterization was carried out by use of a micro-fabricated device having a chalcogenide thin film ($Ge_2Sb_2Te_5$, Ge-doped SbTe, or PbTe) in-between two lateral electrodes. Optical reflectance and transmittance were measured with a probe laser beam during pulsed irradiation by a pumping laser of varying power, along with a simultaneous measurement of change in device voltage across the irradiated region. From these measurements, relative changes in optical absorption coefficient as well as in electrical resistivity were determined and compared among different chalcogenide materials.

2. EXPERIMENTS

Laser-induced resistance changes were measured simultaneously with optical reflectance and transmittance by use of the experimental setup and the sample device illustrated in Fig. 1. The sample device was fabricated using an optically transparent Corning glass substrate coated with a $ZnS-SiO_2$ sputtered film of 150 nm thick. Each chalcogenide thin film of $Ge_2Sb_2Te_5$ (10nm), Ge-doped SbTe (10nm) and PbTe (15nm) was deposited thereon by sputtering and annealed at 250 ^oC for 5 min. in an RTA furnace (except for PbTe which is crystalline in the as-deposited state) to have the respective crystalline state. From either RBS or XRF analysis, each of these films was found to have the following composition: Ge 22.5at%-Sb 23.3at%-Te 54.2at%, Ge 5.3at%-Sb 75.8 at%-Te 18.9at% and Pb 49at%-Te 51at%, respectively. Subsequently, each chalcogenide film layer was patterned to have a bow tie shape with a minimum feature size of 4 μm wide by way of a photo-lithography combined with a lift-off process. A $ZnS-SiO_2$ (150nm) film was deposited and patterned similarly to form a coating layer for the chalcogenide film area of about 20 μm long around the spot for laser irradiation and the entire chalcogenide-free area as well. The rest area without the $ZnS-SiO_2$ coating was made to have a TiN film layer (150nm) by the subsequent patterning process.

With a fixed DC probe voltage (5V) over a circuit made up of the sample device, an oscilloscope with 1MΩ resistance and with a 120kΩ load resistor in parallel connection, the center of the bow tie area of each device was irradiated with a 658 nm pulsed pump laser beam of varying power, focused through an objective lens of 0.6 NA to have a $1/e^2$ spot diameter of around 1.2μm. Laser-induced voltage wave forms were recorded together with optical reflectance and transmittance, as monitored by a CW probe laser beam of another wavelength (633 nm).

3. RESULTS & DISCUSSION

Shown in Fig. 2 are the transient optical transmittance and reflectance signals for the three different chalcogenide materials, as measured with a probe laser during irradiation with a pumping laser of 1μs pulse duration and varying power from 1 mW to 4 mW. Among different materials, prominent similarities are found as follows: Firstly, reflectance/transmittance changes upon firing the pumping laser, yet restoring its initial level with ending the pulse regardless of laser power. Delayed recovery of transmittance/reflectance during post-pulse cooling period is an indication of a varying optical signal with thermal decay.[9] Secondly, significant increase in sample transmittance as well as decrease in reflectance takes place with increasing laser power. It is emphasized that melting of PbTe film is not supposed to take place in the range of laser powers and with the multilayer stack in use [9]. The possibility of melting may be also ruled out for the $Ge_2Sb_2Te_5$ and Ge-doped SbTe thin films as well, as the peak values of transmittance and reflectance appear to vary gradually rather than abruptly with laser power (compare with what is shown in Fig. 2 of reference [9]). From these, we tend to believe that the observed optical changes are entirely solid state phenomena of the same nature for all of three chalcogenide materials.

Shown in Fig. 3 are the real time variations in device resistance that were derived from the voltage waveforms at the oscilloscope via applying the conditions of a constant current and a voltage conservation to the primary series circuit. Once again, strong similarities are observed among different chalcogenide materials: device resistance decreases with increasing laser power. As compared with the optical signals shown in Fig. 2, rising and decaying portions of the device resistances are exceedingly prolonged for all the chalcogenide materials. This considerable difference in time response between the different types of signals may be ascribed to an RC delay in the electrical circuit and also possibly to the difference in sensitivity of the respective signal to carrier transport as well as in the system size contributing to the respective signal. The optical signals represent the response of the material region of a size comparable to a laser spot size whereas the electrical signals come from the whole device consisting of an irradiated portion and the rest coupled electrically. We have not attained yet a clear understanding of the detailed transient characteristics of the resistance signals but tend to believe that the peak values may be used as measures of sufficient significance to correlate with the peak values of the optical signals.

From Fig. 2, maximum transmittance and the corresponding minimum reflectance values were taken to obtain absorbance values for varying power of the pump laser and likewise the minimum resistance values from Fig. 3. For each material, absorbance values were converted to the reduced values by way of subtraction of and division by the reference value pertaining to the respective irradiation-free state, thereby obtaining the relative changes in absorption coefficient, $\Delta\alpha/\alpha_o$, ($\Delta\alpha = \alpha - \alpha_o$, α_o is the absorption coefficient in the absence of a pumping irradiation) as a function of pumping laser power. Similarly, relative changes in electrical resistivity, $\Delta\rho/\rho_o$, were derived from the device resistance values. The results are shown in Fig. 4(a) and Fig. 4(b) respectively where absorbed laser power is used as the abscissa instead of nominal laser power in order to account for varying degrees of optical absorption among different devices. Notice $\Delta\alpha/\alpha_o$ values decreasing with absorbed laser power in conjunction with the concomitantly decreasing $\Delta\rho/\rho_o$ values for all of three chalcogenide materials.

Differences among the chalcogenide films appear contrasting between Fig. 4(a) and Fig. 4(b). The rates of change in $\Delta\alpha/\alpha_o$ with absorbed power are not significantly different *i.e.* -4% for Ge-doped SbTe to -8% for PbTe at the absorbed power of around 1.5 mW, whereas the corresponding rates of change in $\Delta\rho/\rho_o$ are noticeably smaller for $Ge_2Sb_2Te_5$ and Ge-doped SbTe. Considering that electrical resistivity depends on carrier concentration and mobility but the latter tends to decrease with increasing temperature of a material [12], it follows that the decreasing trend of the relative changes in electrical resistivity with absorbed power may be entirely due to increase in carrier concentration [13]: $\Delta\rho/\rho_o$ depends on $-\Delta n/n$. Providing that the increase in carrier concentration may be somewhat comparable at the same absorbed power for these narrow band gap materials, $\Delta\rho/\rho_o$ would be very small for the material with a large carrier concentration which is conceivably the case for $Ge_2Sb_2Te_5$ and Ge-doped SbTe in view of their exceedingly lower device resistances than PbTe.

From the preceding results, it is evident that, for three chalcogenide materials considered, the nonlinear optical effects are essentially the same phenomena that involve reduction in absorption coefficient accompanied by reduction in electrical resistivity. A z-scan measurement of the nonlinear absorption coefficient of the PbTe thin film in conjunction with a real time electrical-optical characterization has recently led us to conclude that the nonlinear optical effect of PbTe responsible for its SR capability is thermally enhanced saturable absorption due to band filling by excited carriers.[11] We believe that the same explanation may be applicable to the nonlinear optical effect as observed for the $Ge_2Sb_2Te_5$ and Ge-doped SbTe in the present work and presumably their SR effects as well. It is still uncertain how the material parameters derived above may be used as a guide to assess the relative SR capability among different chalcogenide materials and this remains to be explored in a future work.

4. CONCLUSION

A successful development of an SR material is supposed to be based on a good understanding of the SR mechanism in order to design and control material properties as required. Nevertheless, understanding remains quite incomplete regarding the origin of the promising SR effects of chalcogenide phase change materials such as $Ge_2Sb_2Te_5$ and Ge-doped SbTe. In the present study, it was shown that these materials display nonlinear optical characteristics essentially identical to those of the crystalline chalcogenide PbTe that have been reported to be responsible for its capability of SR readout [9-11]: significant decrease in absorption coefficient was commonly found with increasing laser power, accompanied by reduction in electrical resistivity. From these findings, we propose that the SR effects of chalcogenide phase change materials may be largely, if not entirely, accountable by a mechanism suggested previously for PbTe emphasizing the role of free carriers [11], as compared with a proposal emphasizing the role of bound electrons during ferroelectric transition in Sb-Te based materials [7].

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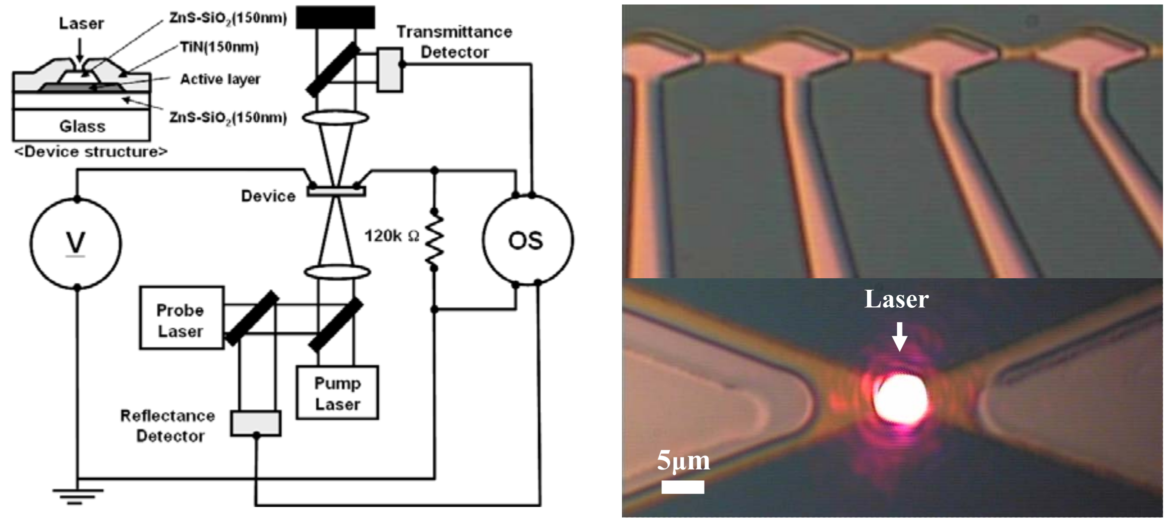
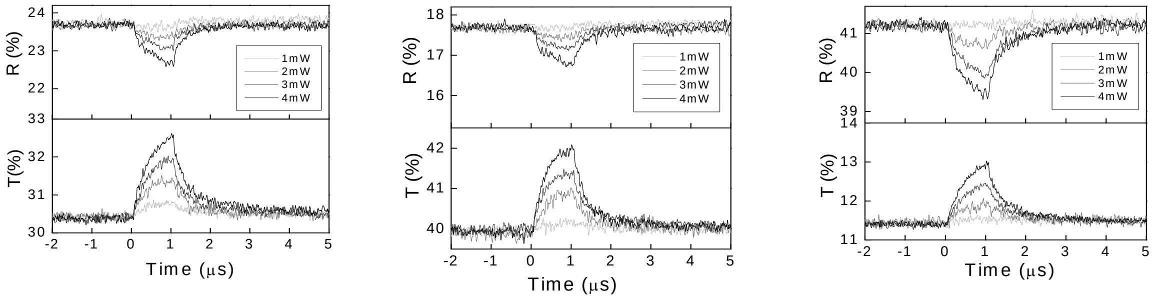


(a)

(b)

Fig.1: (a) Schematic diagram of the experimental setup for real time optical-electrical characterization and (b) optical image of the device layout.

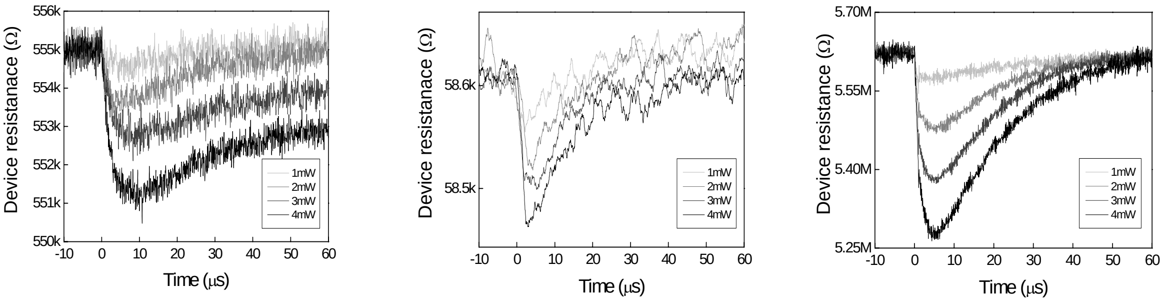


(a)

(b)

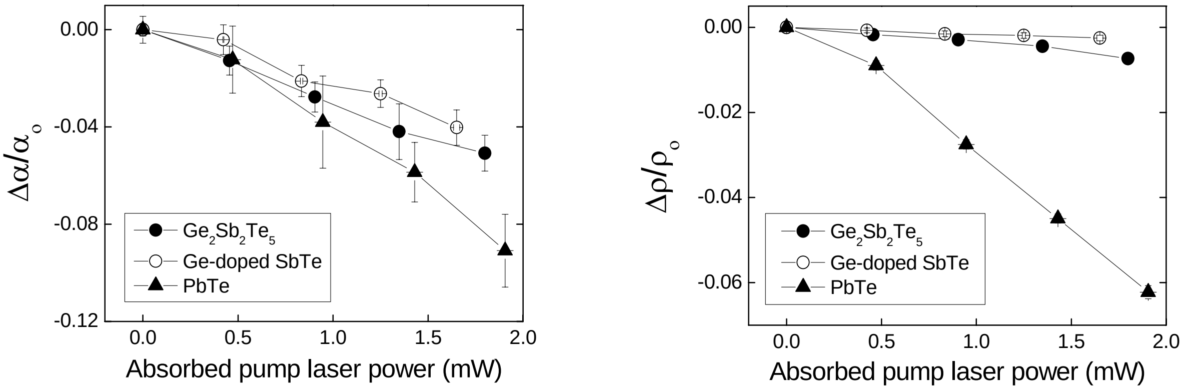
(c)

Fig.2: Transient optical reflectance and transmittance due to real time optical-electrical characterization of (a) $Ge_2Sb_2Te_5$, (b) Ge-doped SbTe, and (c) PbTe.



(a) (b) (c)

Fig.3: Transient device resistances derived from the voltage wave forms due to real time optical-electrical characterization of (a) $Ge_2Sb_2Te_5$, (b) Ge-doped SbTe, and (c) PbTe.



(a) (b)

Fig.4: Variation of the relative change in absorption coefficient (a) and in resistivity (b) with absorbed pump laser power, derived from the data shown in Fig. 2 and Fig. 3.

Biographyies

"Hyun Seok Lee" is a 2nd year Ph.D. candidate student at the Department of Materials Science and Engineering of Korea University in Seoul, Korea where he received a B. E. and a M. E. degree in 2002, 2004 respectively. He has been jointly working in KIST under the supervision of Dr. Byung-ki Cheong since his work for a master's degree on super-resolution optical memory.

"Byung-ki Cheong", Ph.D. has been a senior/principal researcher at Korea Institute of Science and technology (KIST) since 1994. His research area has been chalcogenide thin film materials for phase change optical recording, super-resolution optical storage and non-volatile phase change electrical memory. He received a B.E and a M.E. degree in Metallurgy from Seoul National University, Seoul, Korea and a Ph.D. degree in Materials Science and Engineering with a thesis on phase transformation in solids from Carnegie Mellon University, Pittsburgh, U.S.A. He spent two years as a postdoctoral research associate in Data Storage Systems Center (DSSC) of Carnegie Mellon University, doing research on thin film materials for magnetic and magneto-optical information storage until joining KIST.