Femtosecond lattice and spin dynamics in topological phase-change materials

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ABSTRACT

Now, phase-change materials (PCMs) stand at a turning point, at which new physical effects, such as topological properties, have been introduced theoretically and experimentally. In addition, > 1 GHz memory operation has been demonstrated at telecommunication wavelengths, making PCMs broader interests in new research fields. Here, I overview recent experimental results on femtosecond lattice and spin dynamics of phase-change materials far from equilibrium, based on time-resolved reflectivity and time-resolved magneto-optical Kerr-rotation techniques. These results are mainly observed in interfacial phase-change memory (iPCM) structures, in which GeTe and Sb_2Te_3 layers are designed to be separated. All results are compared with those obtained in conventional $Ge_2Sb_2Te_5$ alloys. Thus, we will provide a possible new step to ultrafast optical and spin switching devices, which can be operated at > 1 THz frequency range.

Key words: Phase-change material, Coherent Phonon, Femtosecond, Spin.

1. INTRODUCTION

Ultrafast lattice and spin dynamics are fundamental physical phenomena [1-3], which would determine the switching speed of phase-change materials, such as $Ge_2Sb_2Te_5$ (GST225). Investigating spin dynamics, in particular, would open new technological fields for spin devises based on phase-change materials [4,5]. However, little is know about lattice and spin dynamics at ultrafast time region, less than picoseconds. Interfacial phase-change memory (iPCM) is one of the best candidates for realizing ultrafast lattice and spin switching, based on possible dominant atomic rearrangement around Ge atoms near the interfaces [6-8]. As a ultrafast spectroscopic technique, coherent phonon spectroscopy (CPS) is a powerful tool for exploring non-equilibrium lattice dynamics [9-14], while time-resolved magneto-optical Kerr-rotation (MOKE) technique, is useful for monitoring transient spin excitation and relaxation [2,15]. Here we discuss femtosecond structural transformation of iPCM films far from equilibrium monitored by coherent phonon spectroscopy under high-density electronic excitation. We also demonstrate that transient magnetization (< 1 ps) by femtosecond laser pulse is possible to occur in iPCM structure as well as in conventional GST225 alloys.

2. EXPERIMENTS

The sample used were $Ge_2Sb_2Te_5$ (GST225) alloy film and iPCM thin films, the later of which consist of the alternation of GeTe and Sb_2Te_3 (or Ge-Sb-Te) layers on Si (100) substrate grown by using a helicon-wave RF magnetron–sputtering. GST225 alloy films were deposited on Si (100) substrate as a reference sample. To measure time-resolved reflectivity change of the sample as a function of the time delay, 40 fs-width optical pulses (a central wavelength of 800 nm) from a Ti:sapphire regenerative amplifier system (100 kHz repetition rate) were utilized. A pair of the pump-pulses was generated through a Michelson-type interferometer, in which the separation time (Δt) of the double pump-pulses was precisely controlled by moving one arm of the mirrors. To explore ultrafast spin dynamics in iPCM and GST225 thin films, on the other hand, we carried out the pump-probe Kerr-rotation spectroscopy with femtosecond pulse laser, whose pulse width of 20 fs and a central wavelength of 830 nm at 80 MHz repetition rate. By rotating the quarter wave-plate inserted on the pump beam pass, the polarization of the pump pulse was varied between the linear *s*-polarized and circular-polarized states, so that one can selectively excite coherent spin via nonlinear magneto-optical effects. All the measurements were carried out at room temperature.

3. RESULTS & DISCUSSION

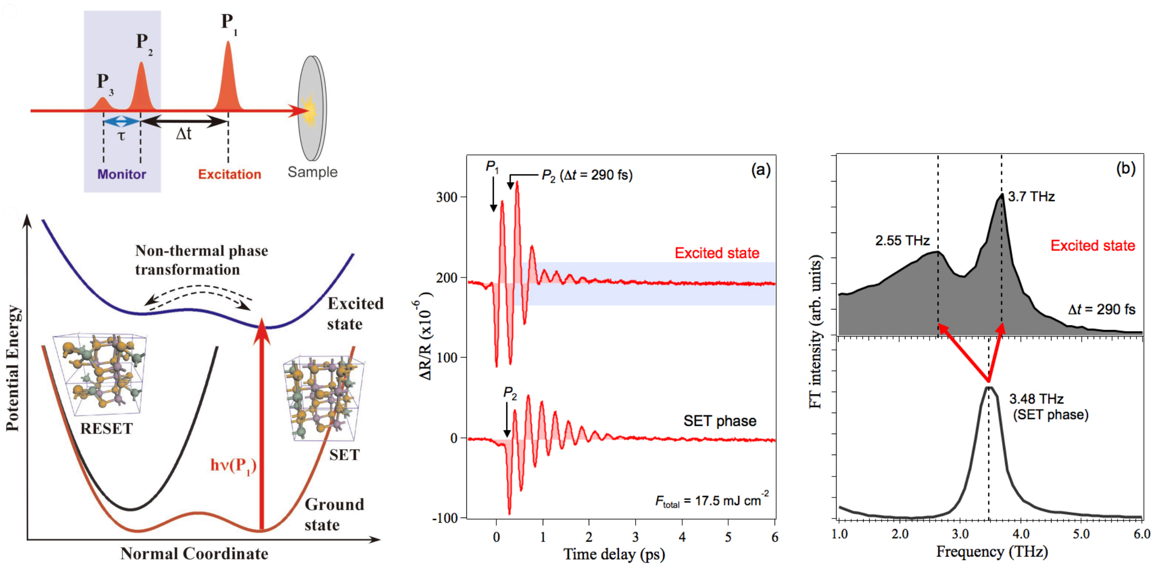

Figure 1. The left figure represents experimental pump-pump-probe sequence (top) and a possible non-thermal phase transformation in the excited state (bottom). (a) The reflectivity signal observed in the SET phase of a iPCM film before the irradiation by the stronger P_1 pulse (10.6 mJ/cm^2)(see the bottom trace obtained by the weaker P_2 pulse at 6.9 mJ/cm^2) and that after the irradiation by the P_1 pulse (see the top trace obtained with the total fluence of 17.5 mJ/cm^2). The separation times (Δt) was set to 290 fs. The rectangular light-blue region represents the coherent oscillation used for FT. (b) The corresponding FT spectra. In the initial SET phase the peak frequency of the phonon mode is 3.48 THz. In the excited state this peak splits into double peaks at 2.55 and 3.7 THz.

Figure 1 shows the results of coherent phonon spectroscopy for the SET phase of iPCM film under high-density electronic excitation with a characteristic separation time of Δt = 290 fs. The coherent phonon oscillation could be enhanced by the second pump-pulse (P_2), after that strongly damped oscillation was monitored, as highlighted by the rectangle region (Fig. 1a). The Fourier transformed (FT) spectra in the excited state shows an appearance of double peak structure (Fig. 1b), demonstrating ultrafast local lattice modification of iPCM. The observation of the double-peak FT spectra can be interpreted to two local structures, possibly three-fold and four-fold Ge atomic coordinations [8, 16, 17]. The ultrafast phase transformation, observed within one picosecond, suggests this process is triggered by electronic excitation. Such a non-thermal nature of the phase transformation has also been demonstrated in GST225 and related phase-change materials by using time-resolved x-ray or electron diffraction measurements by several different groups [18-21]. Note that our observation of the femtosecond to picosecond local lattice dynamics is observed under the condition that the state goes back to the original SET phase, which is different from those observed by the irradiation with femtosecond laser pulses above the threshold [18, 20]. The total pump fluence more than 17.5 mJ/cm^2, however, was required to induce the double-peak FT spectra for the SET phase of iPCM, implying the observed structure would be an intermediate state.

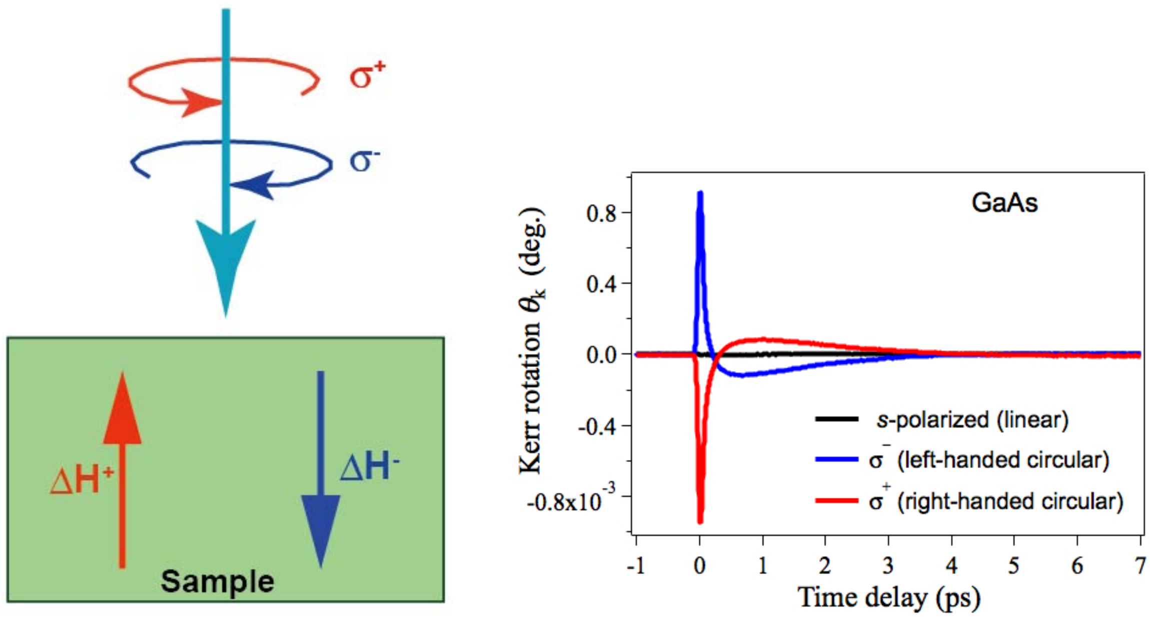

Figure 2. Left: Schematic of the generation of transient magnetic field $\Delta\mathbf{H}^{\pm}$ by the irradiation of the circular-polarized light. Here, σ^{+} corresponds to the right-handed circular and σ^{-} represents the left-handed circular polarization. Right: The time-resolved MOKE signal observed in a GaAs wafer at room temperature for the three different polarization states of the pump-pulse.

We have constructed the time-resolved pump-probe Kerr-rotation spectroscopy system and measured the spin dynamics in GaAs wafer for the system's checking purpose, as shown in Fig. 2. Here, the left-handed circular polarized pump light induces the positive MOKE signal, while the right-handed circular polarized pump shows the negative MOKE signal, indicating they originate from the inverse Faraday effect (IFE) [2, 22], where high-intensity laser irradiation induces a static magnetization ***M***(0),

$$\boldsymbol{M}\left(0;\omega,-\omega\right)=\frac{\chi}{16\pi}\left[\boldsymbol{E}(\omega)\times\boldsymbol{E}^{*}(\omega)\right], \qquad (1)$$

where ω denotes optical frequency, χ is the magneto-optical susceptibility, $\boldsymbol{E}$ is the electric field of the laser pulse. The induced magnetization $\boldsymbol{M}$ in Eq. (1) has generally zero frequency because of optical rectification, but in reality, it has non-zero frequency components, since the ultrashort laser pulse has broad spectra; the shorter the pulse width, the broader the spectral width. As expected from the selection rule of the IFE, the linear *s*-polarized pump light did not generate the MOKE signal in Fig. 2. However, this is not the case for the phase-change materials, like iPCM films, in which linear *s*-polarized pump light could even induce the magnetization [23]. By rotating the quarter waveplate from 0 to 360 degrees, we could map the Kerr rotation signal as a function of the polarization state, from which it was possible to compare the spin dynamics in iPCM with those in conventional GST225 alloy films.

4. CONCLUSION

We have observed femtosecond structural transformation of phase-change materials far from equilibrium monitored by coherent phonons under high-density electronic excitation. We have also mention the results on ultrafast spin dynamics observed in iPCM films as well as in conventional GST225 alloy films.

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