Ultrafast dynamics of coherent optical phonons in $GeTe/Sb_2Te_3$ superlattices: Thermal conductivity and coherent control

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ABSTRACT

We report on the evaluation of lattice thermal conductivity of $GeTe/Sb_2Te_3$ superlattice (SL) by using a coherent phonon spectroscopy at various lattice temperatures. The time-resolved transient reflectivity obtained in amorphous and crystalline $GeTe/Sb_2Te_3$ SL films exhibits the coherent A_1 optical modes at terahertz (THz) frequencies with picoseconds dephasing time. The relaxation time and frequency of the coherent A_1 modes are used to compute the lattice thermal conductivity based on the Debye theory, including scattering by grain boundary and point defect, umklapp process, and phonon resonant scattering. The results indicate that the thermal conductivity in the amorphous SL film is less temperature dependent, due to the dominant phonon-defect scattering, while in the crystalline SL it is temperature dependent because of the main contributions from umklapp and phonon resonant scatterings. We argue the higher thermal conductivity in the $GeTe/Sb_2Te_3$ SL films than that in the $Ge_2Sb_2Te_5$ alloy films implies that the phase change in $GeTe/Sb_2Te_3$ SL is not purely promoted by thermal process, i.e., lattice heating, but rather by nonthermal process, i.e., coherent lattice excitation, because the thermal process generally requires lower thermal conductivity.

Keywords: Femtosecond, phase change, thermal conductivity, superlattice

1. INTRODUCTION

1.1 Phase change materials

Phase change data storage technology offers high-speed, rewritable, and reliable nonvolatile solid-state memory, which may overcome current generation of Si-based memory technologies. In the phase change memory (PCM) materials, the switching between a high resistance amorphous and low resistance crystalline phases can be operated by the irradiation of laser pulses [Fig. 1(a)].

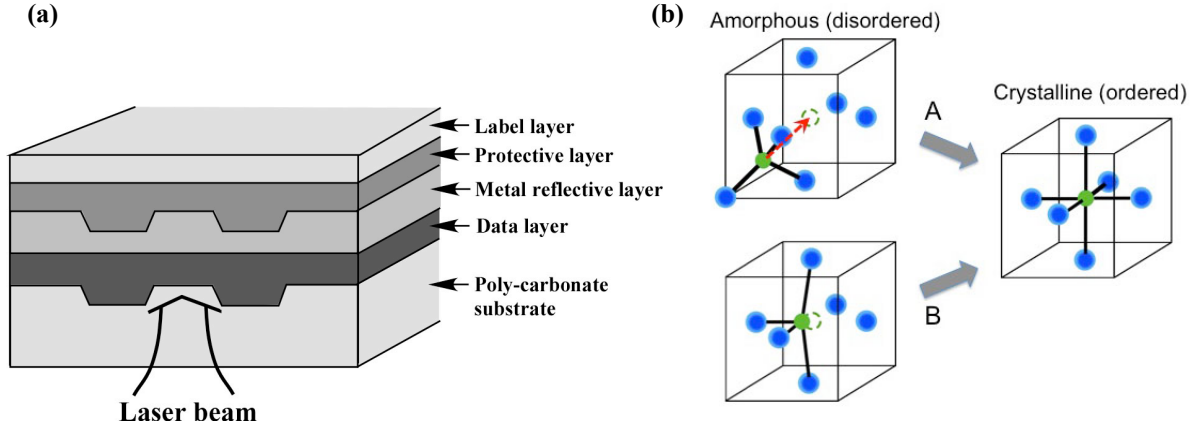

Figure 1. (a) Schematic view of DVD disks. In DVD players LD is tightly focused into the data layer, which corresponds to the phase change material. (b) Possible two models for the fast phase change in $Ge_2Sb_2Te_5$. The process A is based on the umbrella-flip model[1], while B represents a new model based on the resonant bonding[2,3].

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One of the most common and reliable materials for the modern optical recording is $Ge_2Sb_2Te_5$ (GST), in which the phase transition between the crystalline and amorphous phases serves rewritable recording [Fig. 1(b)] [1-4]. Recently, extensive theoretical investigations on the mechanism of the phase change in GST have been made using molecular dynamics simulations[5,6]. In addition to the theoretical works, experimental studies using extended x-ray absorption fine structure (XAFS) [7], time-resolved x-ray absorption near-edge structure (XANES) [1] and Raman scattering measurements[8] have also examined the structure of local atomic arrangements in GST materials.

One of the advantages of using GST films as the optical recording media is its high speed switching characteristics, whose time scale is less than 1 nanosecond[5-7]. In the last decade, however, most of the literatures have studied nanosecond dynamics of the phase change in GST materials using nanosecond and picosecond laser (or electrical) pulses[9]. Hence thermal properties of GST materials have been found to govern the phase change characteristics in GST materials when it is promoted by laser heating. There, thermal conductivity is important to engineer the performance of the phase change[10], such that lower thermal conductivity enables one to realize low power operation of the switching, where focused laser irradiation causes lattice heating[11,12]. In the present manuscript, we investigate the thermal conductivity of $GeTe/Sb_2Te_3$ SL using coherent phonon spectroscopy, and found that thicker GeTe layer induces additional scattering events, which reduced thermal conductivity.

2. EXPERIMENTAL TECHNIQUES

2.1 Coherent phonon spectroscopy

Coherent phonon spectroscopy (CPS) [13-16] has recently been applied to GST materials of alloy[17] and superlatticed films[18], and Sb_2Te_3 films[19]. In those study, the observed local phonon modes in the amorphous GST films were found to be strongly damped modes, with its relaxation time of less than a few picoseconds due to the scattering by lattice defects[17,18]. Wang *et al.* have recently proposed to use CPS as a powerful method to obtain lattice thermal conductivity, where the relaxation time and frequency of the coherent optical modes were used to compute the conductivity based on the Debye theory[20]. In the present study, a reflection-type pump-probe measurements using a mode-locked Ti:sapphire laser (pulse width = 20 fs and a central wavelength = 850 nm) was employed at the temperature range of 5 – 300 K. The average power of the pump and probe beams were fixed at 120 and 3 mW, respectively, from which we estimated the pump fluence to be 284 μJ/cm^2 at 120 mW. The excitation of the $GeTe/Sb_2Te_3$ SL films with the 850 nm (= 1.46 eV) laser pulse generates nonequilibrium carriers across the narrow band gap of ≈ 0.5 - 0.7 eV. The transient reflectivity (TR) change ($\Delta R/R$) was measured as a function of the time delay between the pump and probe pulses (Fig. 2).

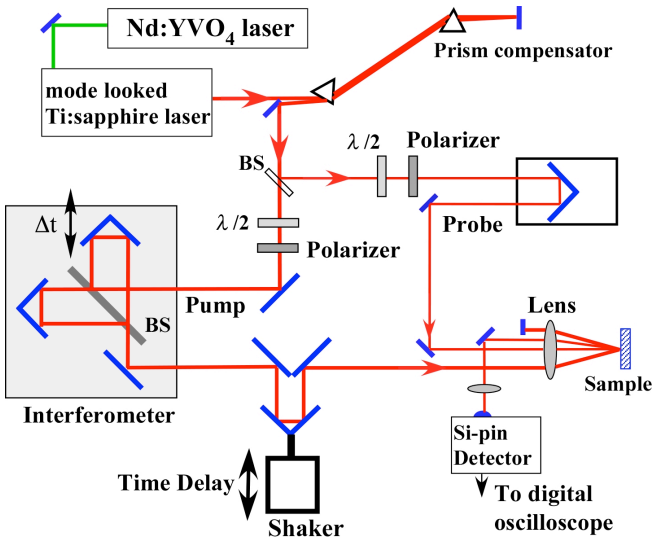

Figure 2. Optical layout of the transient reflectivity change measurement with a mode-locked Ti: sapphire laser. The Michelson interferometer is utilized when controlling the amplitude of coherent phonons by varying the separation time of the double pulses.

2.2 Sample preparation

We have chosen $GeTe/Sb_2Te_3$ SL as a sample after the proposal of a class of superlattice-like $GeTe/Sb_2Te_3$[21]. Significantly lower SET and RESET programming current for the SL cells has been discovered[21] and thus $GeTe/Sb_2Te_3$ SL will be a potential candidate for the future PRAM devises. The samples used in the present study were thin films of $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ and $[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$ SLs fabricated using a helicon-wave RF magnetron sputtering machine on Si (100) substrate[22]. The annealing of the as-grown SL films at 503 K (230 ºC) for ten minutes changed the SL films from amorphous (*a*-) into crystalline (*c*-) states[18]. The TEM measurements confirmed that the *c*-$GeTe/Sb_2Te_3$ SL films have layered structures with clear interfaces.

3. EXPERIMENTAL RESULTS

3.1 Time resolved transient reflectivity

Figures 3(a) and (b) show the time-resolved TR signal ($\Delta R/R$) observed at 5 – 300 K in $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ SL films with the amorphous and crystalline phases, respectively. After the transient electronic response due to the excitation of nonequilibrium carriers at the time delay zero, coherent oscillations with several picoseconds relaxation time appear. Fourier transformed (FT) spectra are obtained from the time-domain data, in which two broad peaks are observed at ≈ 5.1 THz and ≈ 3.78 THz in amorphous film, while a sharp peak at 3.68 THz is observed in crystalline film at 300 K[18]. These peaks in the amorphous film were assigned to the A_1 optical modes due to the local $GeTe_4$ unit (3.78 THz peak)[17,18], and that due to the local pyramidal $SbTe_3$ unit (5.1 THz peak)[8,18]. The red shift of the local A_1 mode frequency in the crystalline phase (3.78 THz → 3.68 THz) has been attributed to the local structural change from tetrahedral $GeTe_4$ into octahedral $GeTe_6$ species[18]. It is to be noted that the zone-folding modes of the acoustic dispersion[23] cannot be detected in our measurements because the SL period ($d \approx 5$Å) of $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ film is an order of the lattice constant.

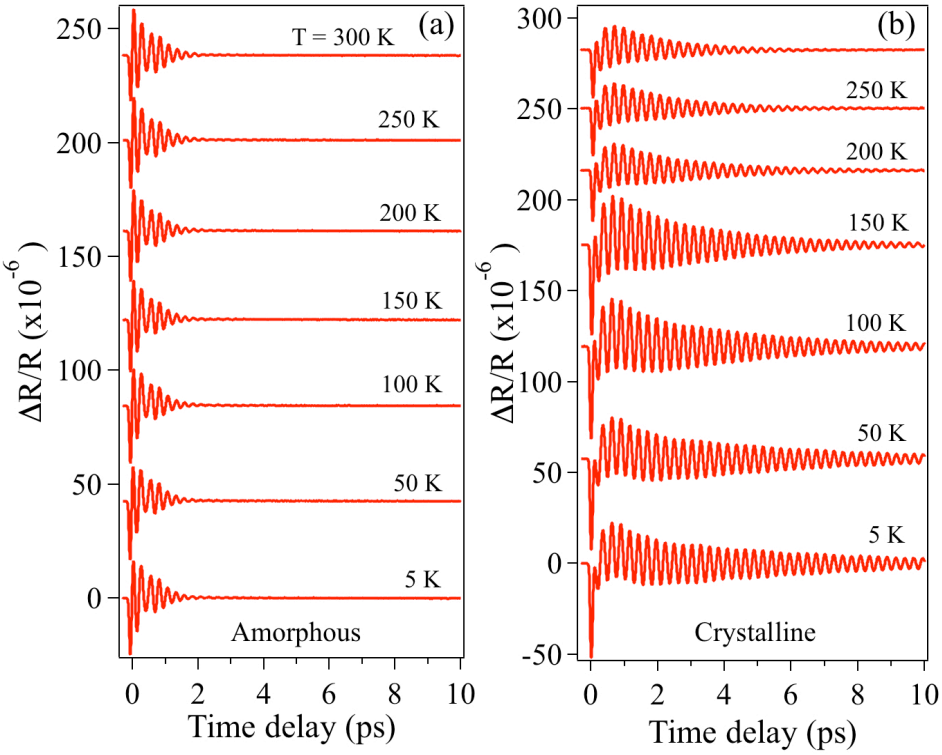

Figure 3. (a) Transient reflectivity trace for *a*–$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ film recorded by use of the single pump-pulse at 286 μJ/cm^2. (b) Transient reflectivity trace for *c*–$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ film with the same fluence.

3.2 Phonon decay dynamics at various temperatures

Figure 4 shows the decay rate of the coherent A_1 mode as a function of the lattice temperature obtained by fitting the time-domain data with damped harmonic oscillations for $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ and $[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$. In the two samples, the decay rate of the crystalline phase increases with increasing the temperature, while that in the amorphous phase is almost constant when the temperature is varied.

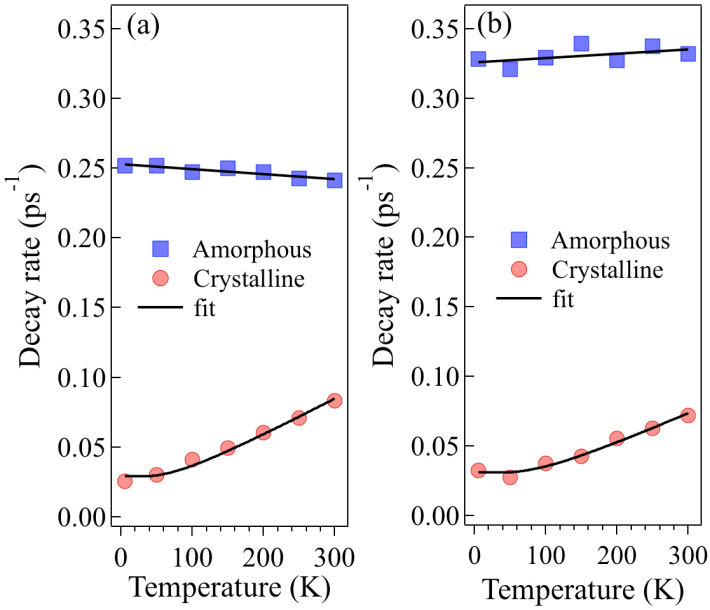

Figure 4. (a) The decay rate of the coherent A_1 mode, which is localized in GeTe layer, in amorphous and crystalline $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ as the function of the lattice temperatures. (b) Same as (a) in $[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$ SL. The fit curves are the linear function for the amorphous and the anharmonic decay model for the crystalline films.

The behavior of the decay rate in the crystalline phase is well explained by the anharmonic decay model[24,25], in which the optical phonon decays into the two acoustic phonons under the conservation of energy and the momentum; the acoustic phonons with half the frequency of the optical mode ($\hbar\Omega/2$) and with opposite wavevectors[24,25],

$$\Gamma(T) = \Gamma_0\left[1+\frac{2}{\exp\left(\frac{\hbar\Omega/2}{k_BT}\right)-1}\right] \quad (1)$$

Here Γ_0 is the effective anharmonicity as the fitting parameter and k_B the Boltzmann constant. Γ_0 is determined to be 0.029 ps^{-1} for $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ and 0.031 ps^{-1} for $[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$. The good agreement of the time domain data with the anharmonic decay model indicates that the damping of the coherent A_1 mode in crystalline SL films is due to anharmonic phonon-phonon coupling (population decay). The damping in the amorphous phase, on the other hands, does not depend on the temperature and therefore would be dominated by phonon-defect scattering (pure dephasing), whose rate is proportional to the density of lattice defects (vacancy)[26]. Note that the anharmonic phonon-phonon coupling would partly contribute to the damping of the A_1 mode also in the amorphous phase, however, the almost flat temperature dependence of the decay rate in Fig. 4 suggests that the anharmonic phonon decay path is blocked by the

randomly distributed vacancies (or voids). In Fig. 4, it is also found that the decay rate is larger in $[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$ than that in $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ SLs, indicating that thicker GeTe layer induces additional scattering events.

4. ANALYSIS AND DISCUSSIONS

4.1 Evaluation of thermal conductivity

To investigate the effect of the scattering by these local phonon modes on lattice thermal conductivity, the parameters of the coherent A_1 mode (the frequency and the decay rate) are used to compute the lattice thermal conductivity based on the Debye theory, combined with the resonant scattering model[27,28]. Lattice thermal conductivity is expressed as,

$$\kappa(T) = \frac{1}{3} C_V \upsilon^2 \tau_c \tag{2}$$

$$= \frac{k_B}{2\pi^2 \upsilon}\left(\frac{k_B T}{\hbar}\right)^3 \int_0^{\Theta_D/T} \frac{x^4 e^x}{\tau_c^{-1}\left(e^x - 1\right)} dx. \tag{3}$$

where $x = \hbar\omega / k_B T$, C_V is the lattice specific heat, υ the sound velocity, Θ_D the Debye temperature, ω the phonon frequency, τ_c the acoustic phonon relaxation time, whose inverse can be given by contributions from various scattering mechanisms[27,28]:

$$\tau_c^{-1} = \frac{\upsilon}{L} + A\omega^4 + B\omega^2 T e^{-\Theta_D/3T} + \frac{C\omega^2}{\left(\Omega^2 - \omega^2\right)^2}. \tag{4}$$

where L, A, B, and C characterize grain boundary, phonon-defect scattering, phonon-phonon umklapp scattering, and phonon resonant scattering, respectively. Ω is the optical phonon frequency observed in the CPS and the last term in Eq. (4) represents the resonant scattering between the localized optical modes and acoustic phonon modes. From the low temperature limit of the decay rate of the coherent A_1 modes (0.253 ps^{-1} for *a*-$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ and 0.026 ps^{-1} for *c*-$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$), we can estimate the ratio of the phonon-defect scattering rate in the amorphous to the crystalline A_a/A_c to be ≈ 10 for the $[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ SL film. The same ratio of $B_a/B_c = C_a/C_c = 10$ has been applied in the simulation. In the same way, $A_a/A_c = B_a/B_c = C_a/C_c = 13$ has been applied to $[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$ SL film. We take the resonant phonon frequency (Ω) at 300 K from the FT spectra[18] obtained by the time domain data in Fig. 3. The magnitudes of all the parameters (L, A, B, and C) are determined as listed in Table I to give the experimental value of κ for *a*-$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ SL ($\kappa \approx 0.33$ $Wm^{-1}K^{-1}$ at 300 K)[29].

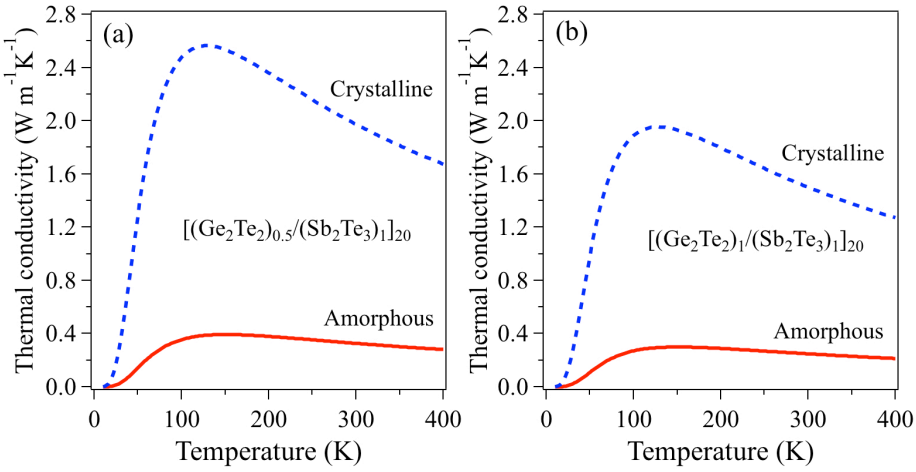

Figure 5. (a) Lattice thermal conductivity of *a*-$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ SL film as a function of lattice temperature calculated by Eqs. (3) and (4). (b) Same as (a) of *a*-$[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$ SL film.

Table 1. Parameters used in Eqs. (3) and (4). For *a*–$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ and *a*–$[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$ SLs C_1 and C_2 represent the resonant scattering coefficient due to the A_1 local modes at 3.78 THz and 5.1 THz, respectively, while for the *c*–$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$ and *c*–$[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$ SLs C_1 represents that at 3.68 THz. [a]from Ref. [30] and [b]from Ref. [10].

Samples	Θ_D (K)	υ (m/s)	L (nm)	A (s^3)	B ($s \cdot K^{-1}$)	C_1 (s^{-3})	C_2 (s^{-3})
a–$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$	250[a]	2250[b]	10.0	40.0×10^{-43}	40.0×10^{-18}	20.0×10^{38}	20.0×10^{38}
c–$[(Ge_2Te_2)_{0.5}/(Sb_2Te_3)_1]_{20}$	300[a]	3190[b]	100.0	4.0×10^{-43}	4.0×10^{-18}	2.0×10^{38}	–
a–$[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$	250[a]	2250[b]	7.6	53.0×10^{-43}	53.0×10^{-18}	26.0×10^{38}	26.0×10^{38}
c–$[(Ge_2Te_2)_1/(Sb_2Te_3)_1]_{20}$	300[a]	3190[b]	100.0	4.0×10^{-43}	4.0×10^{-18}	2.0×10^{38}	–

As shown in Fig. 5, comparing the thermal conductivity obtained for the SL films in different phases, we found that the thermal conductivity in *a*-$GeTe/Sb_2Te_3$ SL films is less temperature dependent, being due to dominant contribution from the phonon-defect scattering. On the contrary, in the crystalline phase thermal conductivity is strongly temperature dependent[31], being attributed to significant contribution from umklapp and phonon resonant scatterings, both of which are related to the phonon dispersion curves and therefore they are significantly temperature dependent[24]. We note further that the thermal conductivity obtained in the SL films is high compared to the conventional GST alloy films; $\kappa \approx 0.2$ $Wm^{-1}K^{-1}$ for the amorphous and $\kappa \approx 0.4$ $Wm^{-1}K^{-1}$ for the crystalline (cubic) phases[9]. The higher thermal conductivity while the lower operation current found in $GeTe/Sb_2Te_3$ SL films, suggests that the phase change in the SL films under the irradiation of ultrashort laser pulses would not be promoted by thermal process, but rather governed by nonthermal process, which has recently been experimentally observed in sub-picosecond time scale by using femtosecond pump-pulse pair excitation[32]. In other words, the high thermal conductivity in $GeTe/Sb_2Te_3$ SL films would enable us to coherently control the phase changes in sub-picosecond time scale because such the nonthermal process should require thermal flow from the excited area to prevent from unnecessary thermal heating.

5. CONCLUSIONS

In conclusion, our results on ultrafast coherent phonon spectroscopy have illustrated temperature dependence of lattice thermal conductivity in $GeTe/Sb_2Te_3$ SL films. These data show that the Debye model, including scatterings by grain boundary and point defect, umklapp process, and phonon resonant scattering, well reproduces the experimental value of thermal conductivity measured by using thermo-reflectance. The thermal conductivity in the *a*–SL films is less temperature dependent, due to the dominant phonon-defect scattering, while in the *c*–SL films it is strongly temperature dependent because of the main contributions from umklapp and phonon resonant scattering. The results on the higher thermal conductivity found in $GeTe/Sb_2Te_3$ SL films support the nonthermal nature of the fast phase change observed in sub-picosecond time scale, where the coherent control of the A_1 mode governed the manipulation of the phase switching between the amorphous and crystalline phases.

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