Coupling of ultrafast laser energy to coherent phonons in bismuth

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Energy coupling to coherent phonons in Bi during femtosecond laser–bismuth interaction is investigated using a double-pulse femtosecond pulse train generated from a temporal pulse shaper. It is found that the increase of bismuth temperature is dependent on the separation time between the two laser pulses. Using a numerical fitting, which considers the effect of convolution between the incident pulses and the material response, the measured temperature increases using different pulse-to-pulse separations allow quantitative determination of the amount of laser energy coupled from excited electrons to coherent phonon vibration. © *2007 American Institute of Physics*.
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Since the early 1990's, many studies have been triggered by the observation of coherent phonons generated in semimetals (Bi, Sb) and semiconductors (Te, Ti_2O_3).[1–3] The generation of coherent phonon is attributed to a mechanism called displacive excitation of coherent phonon,[4–6] which was shown to be a special case of impulsive stimulated Raman scattering for absorbing materials.[7–9] During the laser interaction with materials, the laser energy is initially coupled to electrons, the excited electrons then transfer partial energy to lattice through electron-phonon coupling.[10] In materials possessing A_1 phonon modes, the excited electrons will alter the equilibrium position of ions, inducing the ions to coherently oscillate around the new equilibrium position.[4] Such coherent phonon vibration can be detected by a time-resolved reflectivity measurement. The large reflectivity change $\Delta R/R$ $(10^{-3}–10^{-2})$ of coherent phonons generated by amplified ultrafast laser pulses provides a convenient way for real-time investigation of phonon dynamics, such as phonon dephasing due to energy relaxation,[11] phonon softening induced by excited-free electron diffusion and phonon anharmonicity,[12–18] and atomic displacement.[19]

Time-resolved reflectivity measurements can be affected by many processes: electron excitation and diffusion, electron-hole recombination, electron-phonon coupling, and coherent phonon generation. All of these processes except diffusion will increase the temperature of lattice at surface. In this work, we perform a double-pulse pump-probe reflectivity experiments to investigate energy coupling during ultrafast generation of coherent phonons. Double-pulse trains are synthesized using a temporal pulse shaper which produces desired pulse separation and energy of each pulse. By analyzing the reflectivity signal and evaluating the lattice temperature rise under different pulse separation conditions, we quantitatively determine, for the first time the amount of energy coupled from the excited electrons to coherent phonon vibration.

The bismuth film is evaporated on a polished silicon substrate. The thickness of the bismuth film is around 100 nm, which is much thicker than the absorption depth (~15 nm) at the 800 and 400 nm laser wavelengths used in the experiments. We use a standard pump-probe setup to measure the time-resolved reflectivity. The laser pulses are produced by a Spectra Physics regenerative amplifier system, which outputs 100 fs full width at half maximum pulses with energy up to 1 mJ/pulse at the center wavelength of 800 nm and a repetition rate of 1 kHz. The pump beam is sent into a pulse shaper, which generates pulse trains by modulating the frequency components of the femtosecond laser pulses.[20] The output beam from the pulse shaper passes through a second harmonic generation crystal to generate the pulse train at a wavelength of 400 nm. A bandpass filter for wavelength of 400±20 nm is used to block the residue of 800 nm laser beam. The collinear 400 nm pump and 800 nm probe beams are focused on the sample surface by a 100 mm lens to diameters of 44 and 20 μm, respectively. A 100× charge coupled device imaging system is used to monitor the sample surface. The reflected beam is measured by a balanced detector and a lock-in amplifier.

Since the detected signal in a pump-probe measurement is a result of material response convolved with the cross correlation between pump and probe beams,[21] the effect of convolution must be considered when the material response time is comparable to the laser pulsewidth. In our case, the periodicity of A_1 mode coherent phonon in bismuth is around 0.35 ps, the experimental reflectivity is therefore analyzed using a numerical procedure considering the effect of convolution. The change in reflectivity caused by coherent phonon R_p is expressed as[16]

$$R_p = A_p e^{-t/t_p} \cos((\Omega + \beta t)t + \varphi), \tag{1}$$

where A_p, t_p, Ω, β, φ are coherent phonon amplitude, dephasing time, angular frequency, chirping coefficient (change of phonon frequency with time due to the decay of excited carriers), and initial phase, respectively. The change of reflectivity due to lattice heating R_l is approximated as

$$R_l = A_l(1 - e^{-t/t_l}), \tag{2}$$

where A_l and t_l are the maximum of lattice heating and its time constant, respectively. The response of electrons R_e is modeled as

$$R_e = A_{e1}e^{-t/t_{e1}} + A_{e2}e^{-t/t_{e2}}, \tag{3}$$

where A_{e1}, A_{e2}, and t_{e1}, t_{e2}, are the peak amplitudes and decay times, respectively. It was noted that two exponential terms fitted the experimental data better than one term approximation did. The total response of the system is a con-

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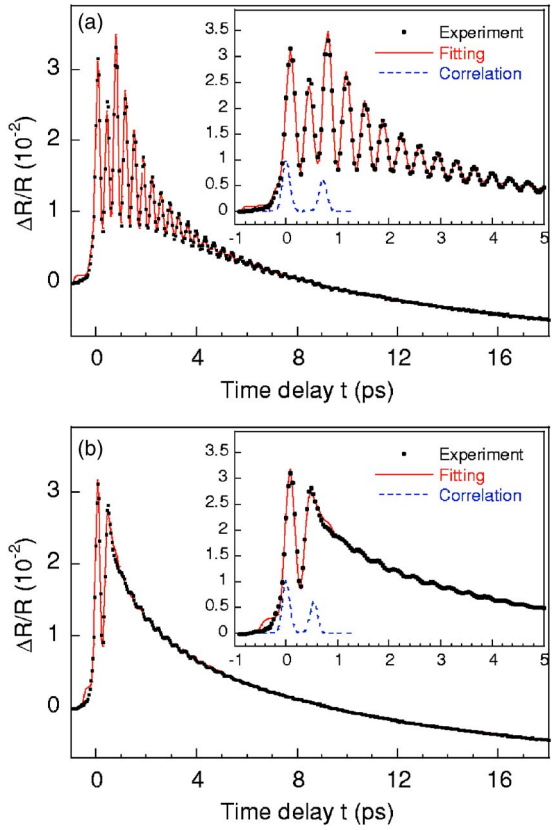

FIG. 1. (Color online) Time-resolved reflectivity of Bi film pumped by a double-pulse train with different separation times of (a) 0.711 ps and (b) 0.535 ps.

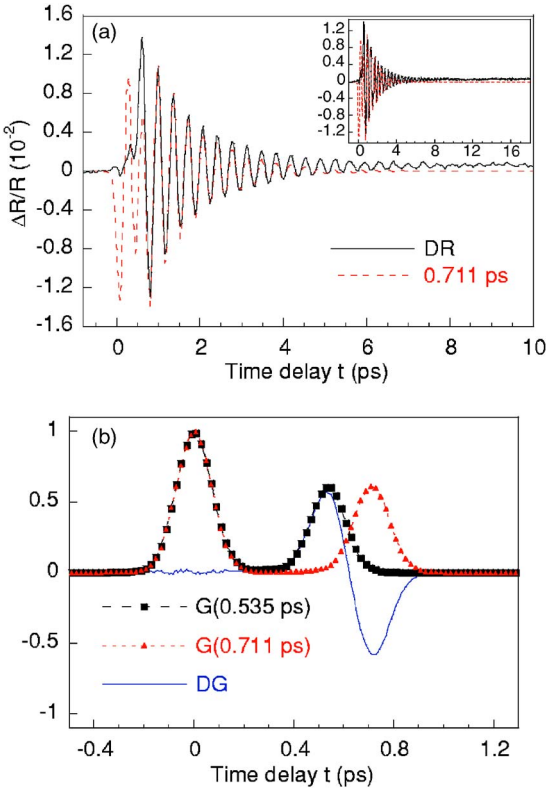

FIG. 2. (Color online) Time dependence of (a) difference in reflectivity change $\mathrm{DR}_{(t)}=\Delta R/R_{(\Delta t=0.535\ \mathrm{ps})}-\Delta R/R_{(\Delta t=0.711\ \mathrm{ps})}$ (black) and $-\Delta R/R_{(t,\Delta t=0.711\ \mathrm{ps})}$ due to the coherent phonon only (red), and (b) difference in cross correlation $\mathrm{DG}_{(t)}=G_{(\Delta t=0.535\ \mathrm{ps})}-G_{(\Delta t=0.711\ \mathrm{ps})}$. The cross correlations $G_{(\Delta t=0.535\ \mathrm{ps})}$ and $G_{(\Delta t=0.711\ \mathrm{ps})}$ are also shown in (b).

volution between $R=(R_p+R_l+R_e)U(t)$ and the pulse input (which can be double pulses, as described below), where $U(t)$ is a unit step function.

Double pulses have been used to control coherent phonon vibration.[22] In this work, we control the separation time and energy of each pulse to enhance or *completely* cancel phonon vibrations. The difference in lattice temperature rise with and without coherent phonon vibration is then analyzed to determine energy coupling to coherent phonons. Figure 1 shows measured time-resolved reflectivity change $\Delta R/R$ pumped by a double-pulse train with (a) $\Delta t=0.711$ ps which is intended to enhance phonon oscillation, and (b) $\Delta t=0.535$ ps which is intended to cancel phonon oscillation. The ratio of the intensities of the two pulses is 1.00:0.61, which was found from experiments to completely cancel phonon oscillation. The 400 nm double-pulse pump has a total fluence $F=0.69\ \mathrm{mJ/cm^2}$, and the 800 nm probe pulse has a fluence $F=0.03\ \mathrm{mJ/cm^2}$. The inset shows a shorter time period as well as the cross correlation between the pump and the probe beams. It is seen that with a separated time $\Delta t=0.711$ ps, the third peak of coherent phonon oscillation corresponds to the end of the second laser pulse, and its amplitude is enhanced. With a separated time $\Delta t=0.535$ ps, phonon vibration after the second peak is canceled completely. Note that at longer time when the system is approaching electron-lattice equilibrium, the reflectivity is less than the value before $t=0$ due to lattice heating. This negative temperature dependence of reflectivity is verified by the measurement of temperature dependence of reflectivity. The amplitude of phonon vibration is apparently stronger than those reported in literature (e.g., Ref. 15) which were all obtained using 800 nm pump beam. Our own experiments also confirmed that the phonon amplitude excited by 800 nm pump beam is much weaker. Figure 1 also shows that results of numerical fitting using Eqs. (1)–(3) are in excellent agreement with the experimental data, which allow quantitative analysis which is to be described below. The parameters fitted from the experimental data are summarized in Table I.

Figure 2(a) shows the difference in reflectivity change, $\mathrm{DR}=\Delta R/R_{(\Delta t=0.535\ \mathrm{ps})}-\Delta R/R_{(\Delta t=0.711\ \mathrm{ps})}$, and the extracted reflectivity change $-(\Delta R/R)_p$ caused by coherent phonon only for $\Delta t=0.711$ ps. The inset in Fig. 2(a) shows the result for a longer time, up to 18 ps. Figure 2(b) shows the first few picoseconds of DR and the difference in the cross correlation measurements $\mathrm{DG}=G_{(\Delta t=0.535\ \mathrm{ps})}-G_{(\Delta t=0.711\ \mathrm{ps})}$, where

TABLE I. Parameters fitted from the experimental data.

	A_p (10^{-3})	t_p (ps)	$\Omega/2\pi$ (THz)	β (rad/ps^2)	φ (deg)	A_{e1} (10^{-3})	t_{e1} (ps)	A_{e1} (10^{-3})	t_{e2} (ps)	A_l (10^{-3})	t_l (ps)
Cancellation	5.74	1.17	2.76	0.155	−87.7	1.98	0.42	2.26	3.42	−3.57	55
Enhancement	6.05	1.14	2.72	0.153	−94.3	1.86	0.46	1.91	2.94	−2.50	21

$G_{(\Delta t=0.535\ \mathrm{ps})}$ and $G_{(\Delta t=0.711\ \mathrm{ps})}$ are cross correlations measured at pulse separation of 0.535 and 0.711 ps, respectively, which are also shown in Fig. 2(b). The difference in reflectivity change DR can be considered as the signal generated by a virtual double-pulse train represented by DG. At time $t<0.25$ ps, both DR and DG are around 0 as they are determined by the difference in the first pulse in the double-pulse train, which are identical for the two cases at $t<0.25$ ps. At time $t>0.8$ ps, around the end of the virtual double-pulse train, DR is dominated by a oscillatory component, which is almost the same as $-\Delta R/R_{(\Delta t=0.711\ \mathrm{ps})}$ since the phonon oscillation for $\Delta t=0.535$ ps is completely canceled. The most relevant feature for our analysis in DR is that, at time $t>8$ ps, the oscillation is attenuated significantly, and DR approaches a value of 6.5×10^{-4}, which reflects lattice heating caused by enhanced coherent phonon vibration by the second pulse for $\Delta t=0.711$ ps.

The positive value of DR shows that more energy is extracted from excited carriers by coherent phonon vibration and is then transferred to lattice heating. This also indicates that energy transfer from coherent phonon to the lattice heating is a faster process compared with lattice heating by excited carriers directly due to the fast dephasing time of coherent phonons. The value of DR can be analyzed to determine quantitatively the energy coupling from the excited carriers to coherent phonons. The coherent phonon vibration can be seen as harmonic oscillation with energy proportional to the square amplitude of reflectivity change $(\Delta R/R)_p^2$. For $\Delta t=0.711$ ps, the amplitude of the first peak of reflectivity change caused by the coherent phonon is -1.33×10^{-2} at $t=0.08$ ps [note Fig. 2(a) shows $-(\Delta R/R)_p$]. From numerical fitting, the third negative peak of phonon vibration would have decayed to an amplitude of -5.91×10^{-3} at $t=0.80$ ps if there were no second pumping pulse. With the second pumping pulse, Fig. 2(a) shows that the amplitude of the third negative peak at $t=0.80$ ps is -1.38×10^{-2}, so the contribution from the second pumping pulse in the third peak is 82% $(1-0.591^2/1.38^2)$.

In order to relate coherent phonon vibration to lattice heating, the static temperature dependence of reflectivity change was measured independently by heating the sample with a patch heater and recording the reflectivity change. It is found that when the temperature increase is less than 95 °C, the reflectivity change decreases linearly with temperature as $d(\Delta R/R)/dT=-8\times10^{-5}/°\mathrm{C}$. As shown in Fig. 1, values of $\Delta R/R$ for both cases of $\Delta t=0.535$ and 0.711 ps are less than 6×10^{-3}, implying that the temperature increases are less than 75 °C, within the linear range of reflectivity change. The difference of the temperature (reflectivity) increase at $t>8$ ps in Fig. 2(a), which is caused by the difference between with and without coherent phonon vibration at the end of second pumping pulse, can be calculated from $\mathrm{DR}/(d(\Delta R/R)/dT)$. The energy needed for generating such a difference in temperature increase is $\Delta F=\rho c d\mathrm{DR}/(d(\Delta R/R)/dT)$, where ρ, c, d are density, heat capacity, and absorption depth of bismuth for the wavelength of the pumping laser, respectively. Using $d(\Delta R/R)/dT=-8\times10^{-5}/\mathrm{K}$ and $d=15$ nm, the laser energy ΔF is found to be 0.014 $\mathrm{mJ/cm^2}$. As mentioned before, 82% of the total energy of the coherent phonon at $t=0.80$ ps is from the second pulse which has a laser fluence of 0.26 $\mathrm{mJ/cm^2}$, therefore the energy coupled to coherent phonon vibration accounts for about 4.4% $(0.82\times0.014/0.26)$ of the energy of the second laser pulses.

In summary, we reported time-resolved reflectivity of bismuth pumped by a double-pulse train. The experimental data were analyzed by a numerical fitting which considered the effect of convolution on the experimental data. It was found that energy can be coupled from excited carriers to lattice heating by the generation of coherent phonon vibration. The increase in lattice temperature shortly after the laser pulses depends on the separation time between the two pumping pulses. The difference of temperature increase with enhanced and canceled phonon vibration was used to quantitatively estimate energy coupling from excited carriers to coherent phonons.

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