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Hot carrier transport limits the displacive excitation of coherent phonons in bismuth

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ABSTRACT

We performed femtosecond transient reflectivity measurements on epitaxially grown bismuth (Bi) films in the weak photoexcitation regime. Single crystalline ultrathin Bi films down to a thickness of 7 nm enabled us to determine a clear correspondence between the amplitude of the coherent A_{1g} phonon and the photoexcitation level. We were able to empirically measure the effective hot carrier penetration length that determines the excited carrier density governing the magnitude of the coherent A_{1g} phonon in Bi. Our findings suggest that the transport behavior of hot carriers is to be taken into consideration in order to provide insights into the mechanism for the displacive excitation of coherent phonons.

Femtosecond optical excitation of carriers in solids is usually characterized by the fast redistribution of the excess of absorbed energy within the electron gas, followed by a slower electron-lattice equilibration process that occurs on the picosecond timescale. Depending on the electronic and structural properties of the material under study and the photoexcitation level, the lattice potential can suffer very different degrees of alteration.[1–9] Bismuth (Bi) is a Peierls-distorted charge density wave material.[10–15] The crystal structure of Bi can be described by a rhombohedral unit cell that contains a pair of atoms with alternating interatomic distances as we move along the ⟨111⟩-Peierls distortion coordinate.[16] This periodic lattice distortion (PLD) or pair formation is concomitant with the modulation of the electron density, which stabilizes the electron gas through the opening of a small energy gap that confers on Bi semimetal properties. In such a strongly correlated electron-lattice system, femtosecond optical excitation has a profound effect on the lattice cohesive forces responsible for the formation of PLD and launches the fully symmetric A_{1g} phonon mode. Different theories have been developed to explain the generation mechanism of coherent A_{1g} phonons in semimetals and semiconductors, highlighting the challenges faced when describing the driving mechanism for such coherent lattice vibration.[17–26]

Most femtosecond transient reflectivity (fs-TR) studies of Bi have focused on the investigation of the dynamics of coherent A_{1g} phonons in either bulk single crystalline samples or relatively thick Bi films.[27–31] Some exceptions are the recent works by Ishioka *et al.*,[32] He *et al.*,[33] and Shin *et al.*[34] Ishioka *et al.*[32] were able to observe coherent A_{1g} phonon oscillations in *in situ* grown Bi films on Si(111) with thicknesses down to three bilayers. They attributed the loss of signal in thinner specimens to transformation from the rhombohedral to the black-phosphorus-like structure. He *et al.*[33] arrived to similar observations by studying films as thin as 4 nm. Shin *et al.*[34] performed a detailed fs-TR investigation of Bi films with thicknesses down to 25 nm deposited on glass and Si substrates. They studied the effects of carrier confinement on the frequency and dephasing time of coherent A_{1g} phonons. They concluded that confinement yields a significantly higher density

of excited carriers, which results in an increased softening of the phonon frequencies and faster dephasing.[34]

In this Letter, we perform single-color fs-TR measurements to monitor the dynamics of coherent A_{1g} phonons as a function of Bi film thickness in the low photoexcitation regime to investigate the effects of confinement on the amplitude of the coherent A_{1g} phonon (A_{ph})—a parameter that has not been previously analyzed owing to its dependence on the crystalline quality. High quality epitaxially grown ultrathin Bi films enabled the observation of the behavior of A_{ph} with decreasing film thickness down to 7 nm. Our analysis of fs-TR data reveals that hot carrier transport plays an important role in defining the effective excited carrier density that governs the displacive excitation of coherent A_{1g} phonons.[17]

(111)-oriented single crystalline Bi films were grown by molecular beam epitaxy on (001)-oriented NaCl single crystalline substrates at room temperature.[35] Optically grade NaCl(001) crystals were cleaned *ex situ* by etching with de-ionized water for a few seconds followed by a short rinse with propanol to displace the water. Any remaining propanol was removed by compressed air. This recipe results in atomically flat surfaces which, after transfer to the ultrahigh vacuum chamber, exhibits the clear (1 × 1) reconstruction of the bare NaCl(001) surface in low energy electron diffraction. Bismuth (99.999%) was deposited at room temperature from a commercial e-beam source at a deposition rate of 5 nm/min using a graphite crucible liner. The absolute coverage was calibrated *ex situ* with atomic force microscopy. Bi grows as epitaxial film with (111) orientation with two by 90° rotated domains with a typical domains size of 100–200 nm. The films are continuous and smooth.[35] These optically transparent and thermally and electrically insulating NaCl substrates helped us to minimize coupling effects between the Bi films and the substrates as well as light absorption by the latter. The nominal thicknesses of Bi films were monitored during the deposition through the application of a quartz microbalance, which was calibrated by *ex situ* atomic force microscopy. Bi films with nominal thicknesses in the range of 10–200 nm were obtained. The effective thicknesses of the crystalline Bi films were calculated by correcting these nominal values for the known formation of a ∼3 nm thick bismuth oxide (Bi_2O_3) layer following exposure to ambient conditions.

We employed a mode-locked Ti:Sapphire oscillator as the pulsed light source for our single-color fs-TR measurements. Our laser system delivers 30 fs pulses with a central wavelength of 800 nm at a repetition rate of 80 MHz. The pump induced transient reflectivity changes were monitored by three times weaker probe pulses at varying time delays t through the implementation of an optical delay stage. The pump beam intensity was modulated at 2 kHz by means of a mechanical chopper to record $\Delta R(t)/R_0$ as a function of t. Both the pump and the probe beams were focused almost collinearly onto the sample. The pump and probe beams were set with orthogonal polarizations to further reduce the amount of scattered pump light reaching the silicon photodiode detector. The reflectivity signal was finally transduced into voltage and monitored with a digital lock-in amplifier.

The magnitude of the transient reflectivity signal is often expressed in the generalized form[17,23]

$$\frac{R(t)-R_0}{R_0}=\frac{\Delta R(t)}{R_0}\approx\frac{1}{R_0}\left[\sum_i\left(\frac{\partial R}{\partial q_i}\right)_Q q_i(t)+\left(\frac{\partial R}{\partial Q}\right)_q Q(t)\right], \quad (1)$$

where $R(t)$ is the probe beam reflectivity measured in the presence of the pump excitation at a given time delay t, R_0 is the reflectivity of the probe beam in the absence of photoexcitation, q_i represents electronic and lattice degrees of freedom different from Q such as the photoexcited carrier density (or electronic temperature) and lattice temperature, and Q is a nuclear displacement coordinate of interest, i.e., the coherent A_{1g} mode coordinate. In the low-excitation regime, $(\partial R/\partial q_i)$ and $(\partial R/\partial Q)$ can be assumed to be constants, and therefore, $\Delta R/R_0$ is expected to increase linearly with the incident pump fluence F since both the number of photoexcited carriers and coherent phonons are proportional to F.

Figure 1 shows raw $\Delta R(t)/R_0$ time traces originated from films with effective Bi thicknesses of (7 ± 1), (17 ± 1), (47 ± 3), (97 ± 7), (197 ± 15) nm and under a constant $F = 0.13$ mJ/cm^2. All traces show a nearly instantaneous increase in the reflectivity owing to photoexcitation of carriers followed by a subsequent gradual decrease with

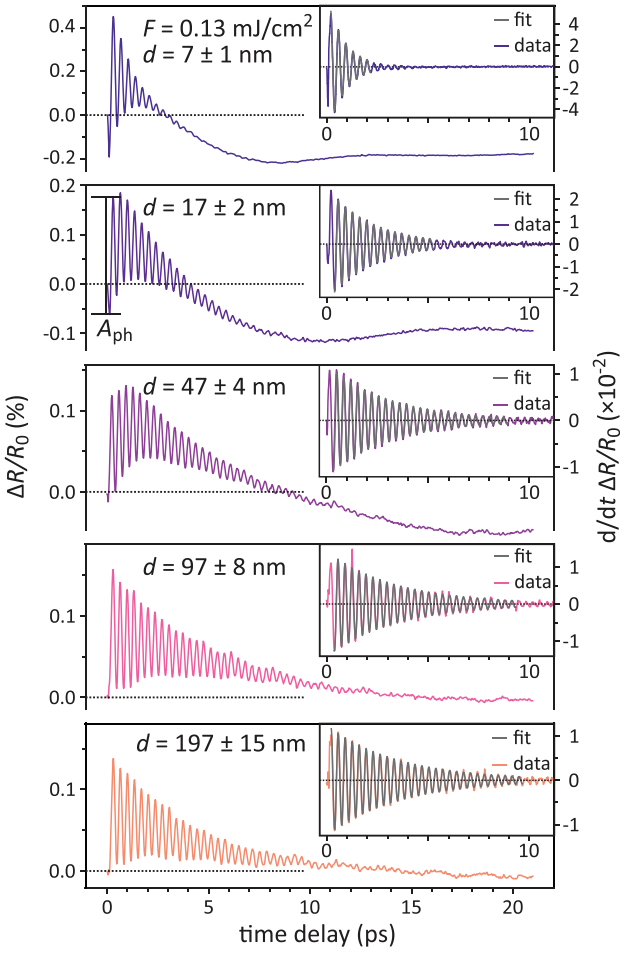

FIG. 1. Transient reflectivity signal $\Delta R/R_0$ for 7, 17, 47, 97, and 197 nm thick bismuth films. All are excited with a fluence of $F = 0.13$ mJ/cm^2. The oscillation amplitude A_{ph}^{init} is the difference between the minimum and maximum signal in the first oscillation. The inset shows the numerical derivative of the signal $\frac{d}{dt}(\Delta R/R_0)$ with the fit in black. The fit is used to extract f_0 and A_{ph}^{fit}.

increasing t. The latter effect is caused by the relaxation of hot carriers via electron–phonon scattering. The pronounced oscillatory component of $\Delta R(t)/R_0$, superimposed on the gradually decaying electronic background arises from the last term in Eq. (1) and, therefore, is due to changes in the dielectric function of the material introduced by the coherent A_{1g} phonon, which has been routinely observed in high-quality crystalline Bi samples.[27–32,34] The insets correspond to the time derivative of $\Delta R(t)/R_0$, i.e., $d/dt(\Delta R(t)/R_0)$. These were taken to remove the slowly varying electronic background and simplify the analysis of the coherent A_{1g} phonon dynamics. We determined the initial amplitude A_{ph}^{init}, of the coherent A_{1g} phonon from our raw $\Delta R(t)/R_0$ data as shown in Fig. 1. We found these values to be in good agreement with those, A_{ph}^{fit}, obtained from fitting with an expression

$$d/dt\left(\Delta R(t)/R_0\right) \approx 2\pi f_0 A_{ph}^{fit} \exp(-t/\tau)\sin[2\pi(f_0+\beta t)t+\varphi]. \quad (2)$$

In accord with the displacive excitation of a coherent phonon mechanism,[17] A_{ph} should be proportional to an effective excited carrier density, n_{eff}, which we can define as

$$n_{\mathrm{eff}} \propto F \cdot Abs/d_{\mathrm{eff}}, \quad (3)$$

where Abs is the absorbed fraction and d_{eff} is the effective depth that determines the excited volume—a value that is often calculated assuming that d_{eff} equals the optical absorption depth of the pump, d_{op}, which, for this case, would correspond to $d_{\mathrm{op},\lambda=800\,\mathrm{nm}} \cong 16\,\mathrm{nm}$.[15,28,31] The value of Abs has been determined implementing the general transfer-matrix method for multilayer thin film systems[36,37] for each sample accounting for the thicknesses of Bi_2O_3 and Bi layers and the interfaces air-Bi_2O_3, Bi_2O_3-Bi, and Bi-NaCl.[38–40] The values of A_{ph} and Abs for each film are presented in Table I.

By combining Eq. (3) and the fact that $A_{ph} \propto n_{\mathrm{eff}}$,[17] we obtain $A_{ph}/(F \cdot Abs) \propto 1/d_{\mathrm{eff}}$. Since transient reflectivity measurements essentially probe the dielectric changes of Bi at the Bi/Bi_2O_3 interface, for thick Bi samples, $d_{\mathrm{eff}} = \ell$, where ℓ is a constant that we left broadly defined for now. Hence, $A_{ph}/(F \cdot Abs) \propto 1/\ell$ (regime 1) becomes a constant irrespective of the film thickness, d. On the other hand, in the limit of ultrathin specimens or efficient confinement of hot carriers, the effective penetration depth becomes $d_{\mathrm{eff}} = d$, and therefore, $A_{ph}/(F \cdot Abs) \propto 1/d$ (regime 2), which obeys a linear regression with origin at (0, 0) when $d \to \infty$.

Figure 2(a) shows a plot of $A_{ph}/(F \cdot Abs)$ as a function of the inverse of the film thickness, $1/d$. As can be seen, the three thickest specimens fall within regime 1, whereas the two thinnest films are in the confined regime 2. The intersection between these regimes renders

TABLE I. Absorbed fraction Abs, measured A_{ph}^{init} and fitted A_{ph}^{fit} phonon amplitudes, initial frequency f_0, and initial chirp β as a function of film thickness d.

d (nm)	Abs	$2A_{ph}^{init}$ ($\times 10^{-3}$)	$2A_{ph}^{fit}$ ($\times 10^{-3}$)	f_0 (THz)	β (THz ps^{-1})
7	0.35	6.4	8.1	2.58	0.102
17	0.40	2.7	2.9	2.82	0.010
47	0.36	1.4	1.3	2.92	0.001
97	0.39	1.6	1.6	2.90	0.002
197	0.40	1.4	1.4	2.90	0.002

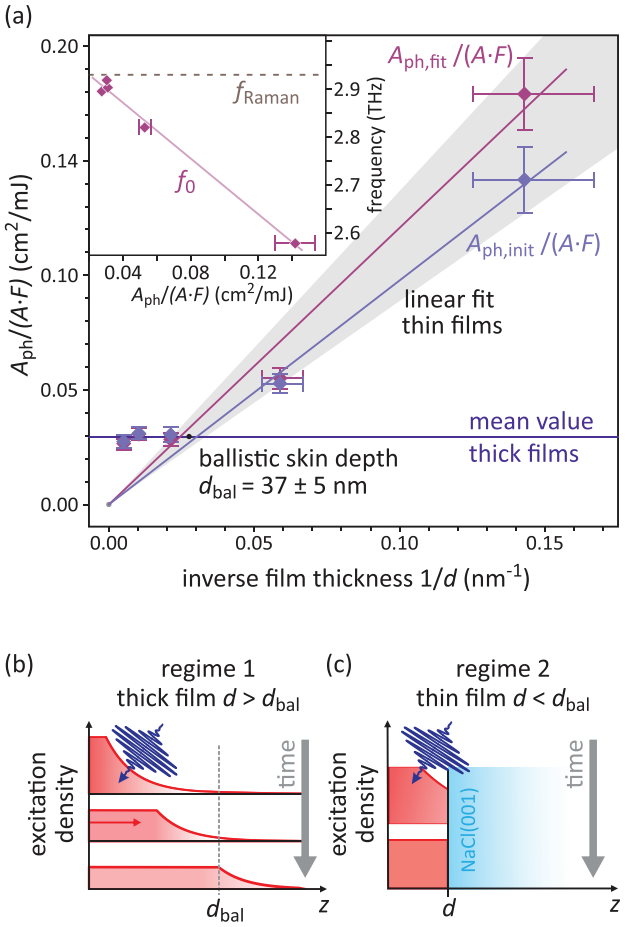

FIG. 2. (a) Phonon amplitude A_{ph} normalized by absorbed fraction Abs and incident fluence F, whereas A_{ph}^{init} and A_{ph}^{fit} are the measured and fitted results, respectively. The ballistic skin depth d_{bal} is marked at the mean value of the intersections of the linear fits for thin films and the mean normalized. Amplitude for thick films. The gray area marks the possible statistical error of d_{bal} times $\sqrt{2}$. The inset shows the initial frequency f_0 plotted vs $A_{ph}/(Abs \cdot F)$ and the Raman measured phonon frequency f_{Raman} for the thickest sample. (b) Sketch of the excitation mechanism for thick films. The gray arrow marks an arbitrary timescale starting from the excitation of carriers and ending with the emergence of the A_{1g} mode. (c) Sketch of the excitation mechanism for thin films, analogous to the one for the thick film.

a value of $1/d_{\mathrm{bal}} \cong 0.29\,\mathrm{nm}^{-1}$; and $d_{\mathrm{bal}} \cong 35\,\mathrm{nm}$ is herein referred to as the effective penetration length of hot electrons, which was found to be approximately a factor of 2 larger than d_{op}. This observation implies that the initial amplitude of the coherent A_{1g} phonon is not given by the very initial distribution of hot carriers and that their transport behavior, as schematically illustrated in Fig. 2(b), must be taken into account. Moreover, the linear behavior observed for f_0 as a function of $A_{ph}/(F \cdot Abs)$ [the inset in Fig. 2(a)] indicates that the degree of phonon softening can be employed as another reference parameter.[34]

Photocurrent measurements of attenuation lengths of hot carriers in gold (Au) and other metals date back to the 1960s.[41–43] More recent time-resolved fs-TR in Au films at room temperature[44–48] indicate the existence of ballistic energy transport by hot carriers with a mean free

path Λ comparable to that of electrons at equilibrium conditions, $\Lambda_{Au} \approx 100$ nm.[46,48] These experimental observations were explained by Suárez *et al.*[45] in the frame of the Fermi-liquid theory[49] within the relaxation time approximation,[50] which also accounted for the subsequent creation of quasiparticles (hot carriers) owing to particle–particle scattering. A larger penetration depth of $\cong 26$ nm and a small diffusivity constant, $D \cong 2.3\,\mathrm{cm^2\,s^{-1}}$ have been determined by Johnson *et al.*[15] in their depth- and time-resolved x-ray diffraction study of a bulk crystal of Bi, who proposed a two-fluid diffusive model to track the spatiotemporal evolution of the energy carried by electrons and holes.

To this end, we enter a rather complex out-of-equilibrium many-body problem, in which ballistic and diffusive transport of hot carriers leads to a value of $d_{bal} > d_{op}$. As a semimetal with a small number of conduction carriers, Bi is known to present a huge Λ_{Bi}, which can be as large as a couple of micrometers at room temperature.[51] This can be explained by the much smaller effective mass and lower density of conduction electrons[52,53] as compared to Au. The values of Λ are anticipated to depend on the crystalline properties of the specimen. For instance, polycrystalline samples composed of tiny crystallites will limit Λ through the scattering of carriers at grain boundaries. However, the latter argument does not apply to our (111)-oriented crystalline Bi films, which present $d_{bal} \ll \Lambda_{Bi}$.

According to the Fermi-liquid theory, the electron–electron equilibration rate for a hot carrier with excess energy E, $k_{ee} \sim a(E - E_F)^2 + bT_e^2$,[49] where T_e is the electronic temperature of the Fermi bath, will experience a higher scattering rate and, therefore, presents smaller Λ and D.[15] Although Landau's theory of Fermi liquids may agree qualitatively with our observation, direct time-resolved techniques capable of monitoring the lifetime of hot electrons[54,55] as a function of film thickness will be necessary to shine some light on this topic. We presented a rather simple and robust analysis method to determine the effective hot carrier density responsible for driving the coherent excitation of A_{1g} phonons in Bi, which will be relevant for many other material systems.

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The authors declare no competing financial interest.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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