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Coherent phonons in bismuth film observed by ultrafast electron diffraction

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The generation of coherent phonons in polycrystalline bismuth film excited with femtosecond laser pulse is observed by ultrafast time-resolved electron diffraction. The dynamics of the diffracted intensities from the (110), (202), and (024) lattice planes show pronounced oscillations at 130–150 GHz. The origin of these coherent acoustic phonons is discussed in view of optical phonon decay into two acoustic phonons. Different drop times in the intensity of the diffraction orders are observed and interpreted as anisotropy in the energy transfer rate of coherent optical phonons. *Copyright 2011 Author(s). This article is distributed under a Creative Commons Attribution 3.0 Unported License. [doi:10.1063/1.3574888]*

The interaction of femtosecond laser pulses with semimetals and semiconductors produces electronic excitations coupled with lattice vibrations through the deformation potential and stimulated Raman scattering.^{1,2} If the frequencies of Raman active phonons in the laser-excited material are smaller than the inverse duration of the laser pulse, then the laser-induced change in the equilibrium positions of the nuclei (displacive excitation) results in generation of coherent optical and acoustical phonons.³ Numerous studies were performed on the dynamics of coherent phonons in bismuth (Bi) by monitoring of the optical reflectivity modulation with femtosecond pump-probe techniques.^{4–10} Femtosecond time-resolved X-ray diffraction of lattice dynamics of high amplitude coherent phonons were made in Bi and provided important information on carrier diffusion, electron-hole interaction time, and density-dependent interatomic potential.^{11–13} Time-resolved electron diffraction was used to study the dynamics of coherent acoustic phonons in a single-crystalline Bi film (30–50 nm thickness) on a picosecond time scale.¹⁴ The intensity change of a single Bragg peak of the diffraction pattern was measured and three frequencies corresponding to coherent acoustic phonons were found to be 16, 21 and 29 GHz. These frequencies were tentatively assigned to a longitudinal and two shear acoustic waves, respectively.

We report on the characteristics of coherent phonons as observed in modulation of temporal behavior of the diffracted intensities from the (110), (202), and (024) lattice planes of polycrystalline free-standing 22 ± 2 nm thick Bi film. The sample was prepared by vapor-phase deposition and supported on a transmission electron-diffraction 3-mm grid. The temporal evolution of the electron diffraction pattern was monitored by ultrafast electron diffraction. An electron gun, described in our previous publications,^{15,16} was used to produce electron pulses by photoemission from silver thin film photocathode excited by frequency-tripled femtosecond (110 fs duration, 800 nm wavelength, at 1 kHz repetition rate) laser pulses. The gun operated at acceleration voltage 35 keV, producing electron pulses of ~ 1.5 ps duration at the photocathode excitation level used. The Bi film was exposed to the fundamental 800 nm wavelength at a pulse energy density ~ 2 mJ/cm² that corresponds to excited carrier's density $\sim 10^{21}$ cm⁻³.

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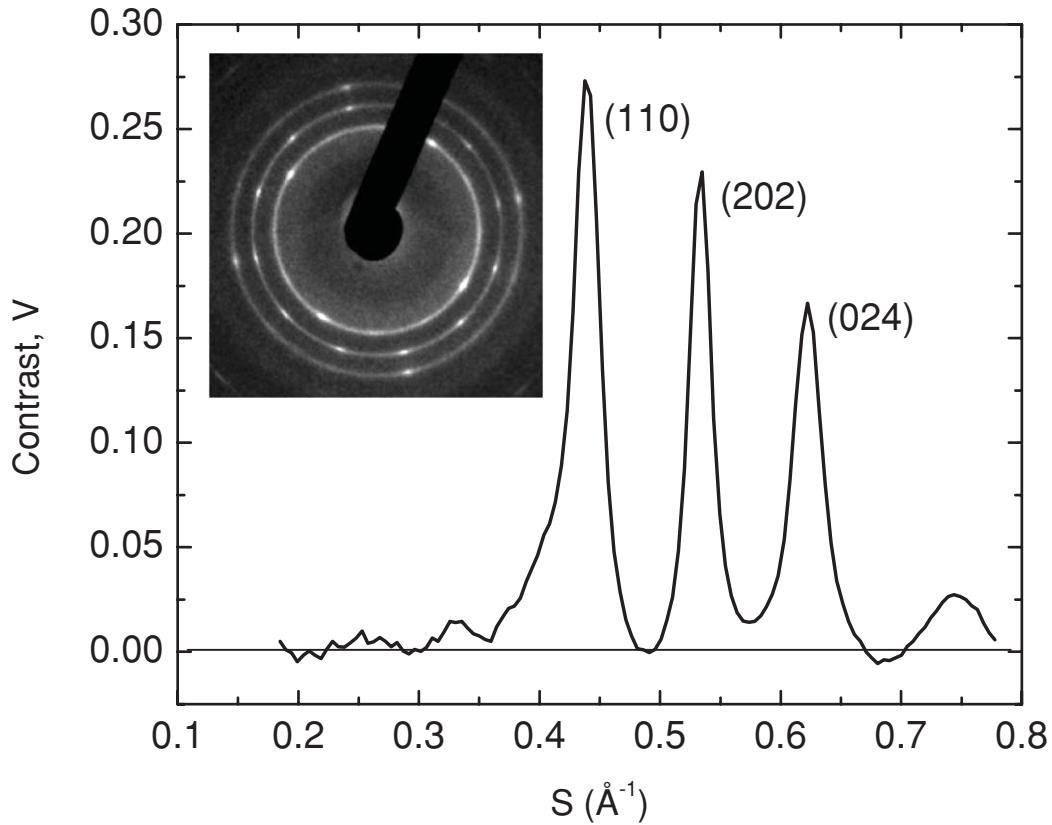


FIG. 1. Radial averaged profile of diffraction intensity of polycrystalline free-standing film of Bi as a function of scattering vector modulus S .

The electron diffraction pattern of the studied Bi film, without excitation, and its radial intensity distribution $I(r)$, determined as the contrast value $V(r) = \frac{I(r) - I_{bg}(r)}{I(r) + I_{bg}(r)}$, where $I_{bg}(r)$ is the background intensity distribution, are shown in Fig. 1. The intensity changes of the diffraction rings are given in terms of contrast value $V(r)$, which takes into account both the changes in diffraction peak and background intensities after the laser exposure. The diffraction patterns were captured in the delay-time range from -30 to 50 ps with delay step of 1.7 ps. Each time-resolved electron diffraction scan was acquired twice and the average value was used for analysis.

To analyze the data, the Boltzmann (sigmoidal) fit was used such that $I(\tau) = I_{min} + (I_{max} - I_{min}) / (1 + \exp(\frac{\tau - \tau_{infl}}{\tau_{rate}}))$ where I_{max} and I_{min} are the maximum and minimum intensities of Bragg diffraction peaks corresponding to that prior to laser exposure and that after sufficient time from exposure to allow the diffraction intensity to reach its steady value, τ_{infl} is the inflection point of the dependence of $I(\tau)$ on the delay time τ , and τ_{rate} is the rate (slope) of the process at this point. The dynamics of the intensity change $I(\tau)$ were characterized by the coordinate of the inflection point τ_{infl} and by the drop-off intensity time τ_{hkl} , which is determined as the full width at e^{-1} level of the maximum of the first derivative of the Boltzmann fit. The Fourier transform was used for frequency analysis.

The temporal development of the contrast values $V(r)$ of the diffraction orders as a function of delay time is shown in Fig. 2. It is seen from Fig. 2 that all three diffraction orders $I_{hkl}(\tau)$ follow the same temporal behavior, namely, the intensity decrease, which is accompanied with high frequency oscillations. According to the frequency analysis made with Fourier transforms shown in the insets of Fig. 2, the oscillations of all three lattice planes found in our experiment are localized in a frequency band centered at 130–150 GHz.

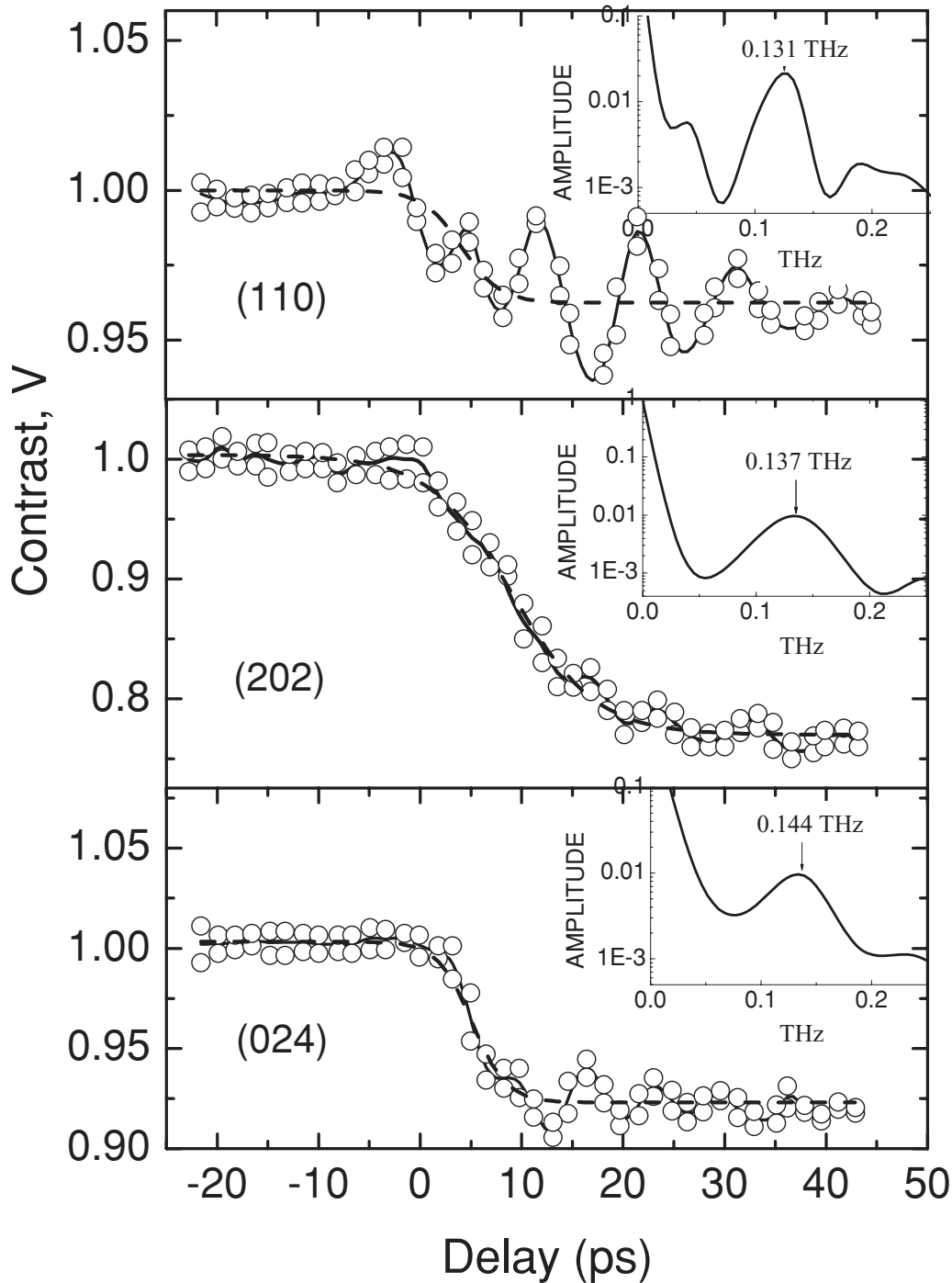


FIG. 2. Normalized contrast $V(r)$ of three diffraction orders as a function of the delay time. Dashed line is Boltzmann fit. Solid line is to guide the eyes only. Insets show the results of the Fourier transform.

Coherent lattice oscillations with a frequency of 124 GHz were previously observed in nickel under similar experimental conditions by measuring the shift in Bragg peak position.¹⁷ This frequency was identified as a breathing mode with vibration period corresponding to standing wave condition $T = \frac{2L}{v}$ for the sample used, where L is the film thickness, and v is the velocity of sound. This explanation is not applicable to the results in Fig. 2 because the highest oscillation frequency

of breathing mode T^{-1} for the 22 ± 2 nm thick Bi film cannot exceed 64 GHz (maximum velocity of sound $v = 2.57$ km/sec),¹⁸ which is approximately 2 times less than that found in the present experiments. The origin of the low-frequency bands in Bi was discussed previously.^{19,20} It was established that among the two main peaks of optical phonons, A_{1g} (2.78 THz) and E_g (2.2 THz), there are four additional peaks on the low-frequency side of the phonon spectrum, which were identified as acoustic phonons TA(X) at 0.68 THz, LA(X) at 1.02 THz, LA(Λ) at 1.42 THz, and LA(T) at 1.76 THz. Since an optical phonon can emit two acoustic phonons,²¹ different combinations of two or more phonons can give rise to a generation of new phonon frequencies like $A_{1g}(\Gamma) \rightarrow 2LA(\Lambda)$, $A_{1g}(\Gamma) \rightarrow LA(T) + LA(X)$, $2LA(X) - LA(\Lambda) \rightarrow TA(X)$, and so on. Therefore, one can find that the combination $A_{1g}(\Gamma) - E_g(\Gamma) - 2LA(X)$ yields the phonon frequency 134 GHz, which is in good agreement with the results of Fig. 2. However, the exact origin of this low frequency band is still not well determined because this low-frequency band remains in Raman spectra of nanocrystalline Bi even at low temperatures.¹⁹ The observed oscillations of coherent acoustic phonons decay approximately 40–50 ps after the femtosecond pulse excitation, which is four times longer than previously found for optical phonons in pump-probe reflectivity measurements.^{10,22,23} This behavior corresponds to the analysis according to which the single phonon decay rate is proportional to $(\hbar\omega_0)^{-2}$ (where ω_0 is a characteristic phonon frequency of the material),²² i.e. the high-frequency oscillations decay faster than the low-frequency ones.

Another interesting feature in Fig. 2, is the different drop time in the diffraction orders τ_{hkl} extracted from the Boltzmann fit, which are $\tau_{110} \sim 8.6$ ps, $\tau_{202} \sim 18$ ps, and $\tau_{024} \sim 7.8$ ps. In general, two mechanisms could contribute to the energy transfer τ_{e-ph} from electrons to the phonon subsystem resulting in lattice heating. The first one is direct-energy transfer from electrons to phonons through optical phonon decay. This can be evaluated from the following $\tau_{e-ph} \approx (\frac{\hbar\omega_D^2 T_L^2}{T_D^2 \varepsilon_F})$ ratio (here ω_D , T_D , T_L is the Debye frequency, the Debye temperature, and lattice temperature, respectively, ε_F is the Fermi level).²⁴ Accordingly, at our experimental conditions, the energy transfer rate τ_{e-ph} is 5–20 ps. The second mechanism affecting τ_{e-ph} is the electron-hole pair recombination accompanied by phonon emission. For Bi at room temperature this process can be as fast as a 2–3 ps.²⁵

An interesting behavior of the energy transfer process is its dependence on the lattice plane (hkl) of Bi. The anisotropy exhibits not only different drop time τ_{hkl} but also different time of the inflection points as well. Figure 2 shows that the maximum slope of the intensity change is achieved at ~ 5 ps for (110), at ~ 9 ps for (202), and at ~ 4 ps for (024) lattice planes. It is of interest to notice that the drop-off amplitude of the intensity of the diffraction orders does not follow a single Debye-Waller factor written for the harmonic approximation as $V_g = V_g^0 \exp(-Bs^2) = V_g^0 \exp[-B(T)(\frac{\sin\theta}{\lambda})^2]$ (here V_g^0 denotes the structure factor of a perfect crystal, θ is the Bragg scattering angle, λ is the electron wavelength, and $B(T)$ is the Debye-Waller factor). According to this expression, the decrease in the intensity of the diffracted beam is expected to be more pronounced for high-order diffraction, but this is not supported by our measurements shown in Fig. 2. There are two mechanisms, which can possibly affect the expected dynamics of the intensity decay. The first one is the anisotropy of $B(T)$ for bismuth caused by the different mean-square displacements of the atoms parallel $B_{\parallel}(T)$ and perpendicular $B_{\perp}(T)$ to the hexagonal c axis.²⁶ For Bi, $B_{\perp}(T) = 2B_{\parallel}(T)$ at $T = 293$ K and $B_{\perp}(T) = 8B_{\parallel}(T)$ at $T = 516$ K, therefore, the influence of anisotropy is very considerable. The second mechanism is the atomic motion of coherent optical phonons A_{1g} that takes place along the elongated body diagonal, whereas the atomic motion of E_g phonons occurs in the plane perpendicular to this diagonal.²⁷ The combined action of these two mechanisms on the differently oriented planes (110), (202), and (024) can result in significant change of the energy transfer rate of coherent optical phonons as well as of the amplitude of the diffraction intensity response.

In conclusion, we used ultrafast time-resolved electron diffraction to study femtosecond laser excitation of polycrystalline Bi film. Oscillations in diffracted intensity were found in the frequency range 130–150 GHz, which are attributed to generation of coherent acoustic phonons. The origin of these coherent acoustic phonons was discussed in view of optical phonon decay into two acoustic phonons. The possible combination of phonon frequencies leading to the observed oscillations was determined. The intensity change of the three diffraction orders (110), (202) and (024) do not follow a single Debye-Waller factor. The data obtained confirm the anisotropy of the energy transfer rate

in Bi, which could be relevant in design of nanoscale ultrafast devices. The results show the effect of structural anisotropy on the spatial-temporal transformation of the intensity distribution within the different orders of the electron diffraction pattern. Ultrafast time-resolved electron diffraction can be used to study the generation, propagation, and decay of coherent acoustic phonons and how they are affected by the anisotropic nature of the crystal structure. These studies are important to the understanding of thermal and elastic properties at the nanoscale.

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