Coherent Phonons: Electronic Softening or Anharmonicity?

In the pump-probe experiments of Hase *et al.* [1], the A_{1g} zone-center phonon of bismuth was coherently excited by the optical generation of a dense electron-hole plasma in a 100 nm film of the semimetal. The resulting oscillations of the reflectivity were analyzed, and the oscillation frequency was shown to be time dependent. This time dependence was ascribed to anharmonicity of the phonon, with the frequency $\omega = \omega_0 + \gamma a^2$, where a is the amplitude of coherent phonon motion. We show here that the expected time dependence of the plasma density [2,3] is likely to be an important effect and that the observed variations in the frequency of reflectivity oscillations can be very well understood in terms of it alone, without any explicit dependence of the phonon frequency on phonon amplitude. Hence, while true anharmonicity (i.e., an explicit dependence of the phonon frequency on the phonon amplitude) is undoubtedly present to some extent, these experiments cannot distinguish it from the expected effects of a time-dependent electron-hole plasma density and further study is required to establish the physical mechanism for the observed time-varying period of the reflectivity oscillations.

A similar time dependence of the probe reflectivity oscillation frequency was observed earlier in tellurium by Hunsche *et al.* [2]. In that case, density functional theory calculations [3] demonstrated that amplitude dependence of the phonon frequency was negligible, and simulations of optical reflectivity showed that the time variation of the coherent phonon frequency was determined by the decay of the plasma density at the surface due to diffusion of the plasma into the bulk.

We have simulated the pump-probe reflectivity oscillations in bismuth using a simple phenomenological model [3] of the optical reflectivity in the presence of diffusion and recombination of the electron-hole plasma. No explicit dependence of the phonon frequency on phonon amplitude is introduced, but the phonon is softened by the presence of the electron-hole plasma; fitting to the low-fluence data of Ref. [1], we take $(1/2\pi)\partial\omega/\partial n = -0.35 \times 10^{-21}$ THz cm^{-3}, where n is the plasma density. The phonon lifetime is 2.5 ps for a pump fluence of 1.9 mJ/cm^2 and is reduced to 1 ps for a pump fluence of7.6 mJ/cm^2 [1,4]. We allow for nonlinear pump absorption at high fluence, reducing the linear absorption efficiency of the 7.6 mJ/cm^2 pulse by a factor of 0.65, in keeping with the observed initial frequency [1].

Assuming an ambipolar diffusion constant $D = 5$ cm^2/s (corresponding to a room-temperature mobility of 200 cm^2 V^{-1}s^{-1} [5]) and an electron-hole recombination time of 5 ps gives excellent agreement with the experimental data in Fig. 4 of Ref. [1], as shown in Fig. 1. In particular, the full range of data points for the 7.6 mJ/cm^2 pulse fluence, which show a nonlinearity not predicted in the anharmonicity model of Ref. [1], are found to be in excellent agreement with these simulations based on plasma diffusion and recombination. The principal effect causing the difference in shape between the curves for 1.9 and 7.6 mJ/cm^2 is the difference between the phonon lifetimes: the phonon amplitude decays more rapidly for the 7.6 mJ/cm^2 pulse. The approximately linear relation between amplitude squared and frequency for the 1.9 mJ/cm^2 pulse is largely coincidental, due to the particular phonon decay rate in that case.

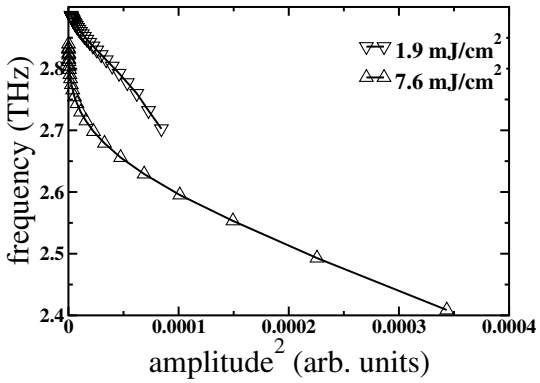

FIG. 1. Variation of the frequency of reflectivity oscillations versus the square of the oscillation amplitude during the first 5 ps after the pump excitation. The symbols indicate values at consecutive periods of the oscillations, starting from high amplitude values.

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