Coherent phonons imprinted into reflectivity oscillations of laser-excited Bi through electron-phonon coupling

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Abstract. We show that the reflectivity of laser-excited solid relates to phonons, driven by thermal forces, through the electron-phonon coupling rate. Controlled excitation of phonons is available by the optimum combination of laser and material parameters.

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The experimental and theoretical studies of the reflectivity oscillations of the probe beam from the single-crystal of bismuth excited at the deposited energy density 2.7mJ/cm^2- 6.7mJ/cm^2 are presented. The reflectivity was measured by the pump-probe technique with the accuracy 10^{-5} and with time-resolution of 35 fs that allowed observing a novel feature - the initial sharp drop of the reflectivity. The reflectivity oscillates with the frequency of A_{1g} phonon in Bi in agreement with earlier findings [1,3-7].

Our analysis establishes the direct link between reflectivity oscillations and atomic vibrations through the electron-phonon coupling rate that is proportional to the phonon's amplitude. The major force driving atomic motion in a laser-excited solid is thermal force proportional to the temperature gradients. It is demonstrated that DECP [1] and strain-dependent polarisation [2-4] are lesser parts of the external field effect on a solid than the thermal force in opaque medium. The proposed theory explains all experimentally observed features of transient reflectivity without any ad hoc assumptions. The response of a medium on the laser action describes by the dielectric function that depends on atomic displacement, q, on electron density, n_e, and on the electron-phonon momentum exchange rate, ν_{e-ph}:

$$\varepsilon = \varepsilon_{r,p}(q) + \varepsilon_{r,m}(n_e, \nu_{e-ph}) + i\varepsilon_{1,m}(n_e, \nu_{e-ph}) \quad (1)$$

Reflectivity variations are calculated through the Fresnel formulae and the Drude-like dielectric function as follows:

$$\Delta R = A_1 \cdot \Delta\varepsilon_{r,p} + A_2 \cdot \Delta n_e / n_{0,e} + A_3 \cdot \Delta\nu_{e-ph} / \nu_{0,e-ph} \quad (2)$$

Here A_1, A_2, A_3 are constant coefficients expressed through known optical data for Bi [8-9]. A_1 and A_3 are negative for Bi, while A_2 is positive. The polarization-related term estimates in the Placzek approximation [2]. The absorbed energy density [10] comprises, $2A \cdot F(t_p)/l_s$= (0.48-1.19) x10^3J/cm^3 for the laser fluence of $F(t_p)$=(2.7-6.7) mJ/cm^2 (absorption, A=0.258, skin depth l_s=2.984x10^{-6} cm). The number density of electrons, excited to conduction band by the avalanche-like process, reaches to the end of the pulse $n_e(t_p) \approx 2AF_p(t)/\Delta \cdot l_s$ = (2-5) x 10^{22}cm^{-3}. Thus, $n_e \propto T_e$. The perturbation in electron-phonon collision rate is presented with the help of kinetic theory [11] as function of lattice temperature and phonon's amplitude**.** The time-dependent electron, T_e, and lattice temperature, T_L, in the skin-layer has been calculated using 2-temperature approximation [12] and material parameters from [8,9]. Time-dependent amplitude of atomic vibrations has been obtained as solution of thermal force-driven equation for damped (γ is damping) harmonic oscillations. Time-dependent reflectivity is determined by laser and laser-excited material parameters without ad hoc assumptions as the following:

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$$\Delta R = -A_{11} \cdot \left(I/10^{11} W/cm^2\right) + A_{22} \cdot \left(T_e(t)/T_{e,m}\right) - A_{33} \cdot \left(T_L(t)/T_{e,m}\right) - \\ -A_{44} \cdot \left\{\left(T_e + T_L\right)/T_{e,m}\right\} e^{-\gamma t} \cdot \cos\left\{\left(\omega_0^2 - \gamma^2\right)^{1/2} t - \varphi\right\} \tag{3}$$

The calculated $\Delta R/R$ function at 6.7 mJ/cm^2 is presented at Fig. 1in a good fit to the experiments.

Summary: We show that the reflectivity of laser-excited solid relates to phonons, driven by thermal forces, through the electron-phonon coupling rate. Controlled excitation of phonons is available by the optimum combination of laser and material parameters.

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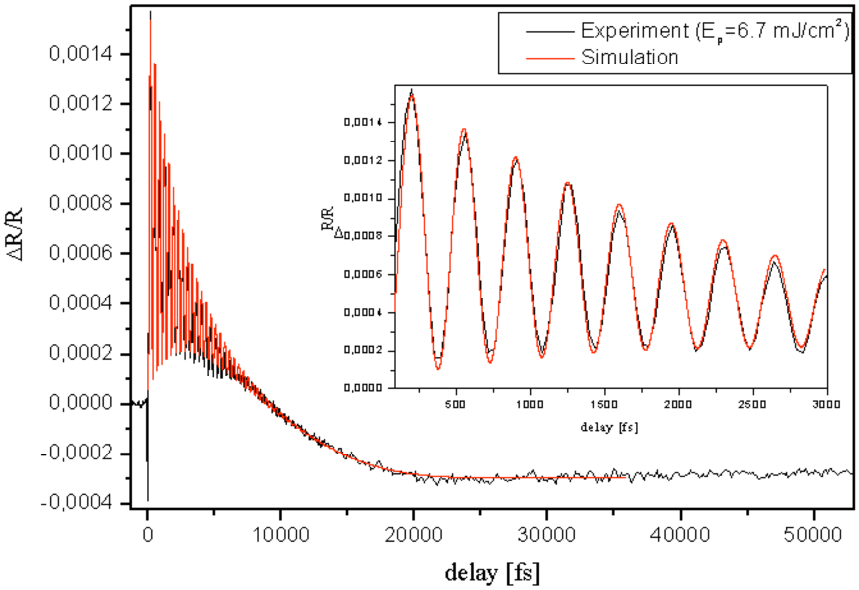

Fig.1The reflectivity variations as function of time-delay between pump and probe pulses