

Ultrafast lattice-bonding dynamics in tellurium

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Abstract: A pump-probe technique measuring the dielectric function is presented and applied to the ultrafast dynamics of coherent phonons in Te. Oscillations in the bonding-antibonding splitting are revealed, allowing for THz modulation of a semiconductor-semimetal transition.

The study of ultrafast dynamics in semiconductors, in pump-probe reflection or transmission geometries, identifies materials that may be used as ultrafast switches, saturable absorbers, and other solid-state devices in next-generation telecommunications systems. In order to incorporate these materials into multilayer devices, the ultrafast dynamics of the spectral dielectric function must be measured to properly model device performance. We have developed a technique to measure the dielectric function of a material over a broad wavelength range (350 to 850 nm) with 50-fs time resolution. In this paper, we present a study of the ultrafast bonding dynamics of tellurium in the presence of symmetry-preserving coherent phonons. Coherent phonons in tellurium were first observed by Cheng and coworkers in 1990 [1], and a displacive excitation mechanism (DECP) was proposed [1,2]. In DECP, a pump pulse shorter than the phonon period instantaneously weakens the bonds and coherent oscillations ensue about newly established equilibrium lattice positions. In tellurium, this ultrafast lattice weakening increases the radius of the nearest-neighbor helices of which the lattice is composed and generates coherent lattice oscillations in the A1 phonon mode at >3.6 THz. Theoretical calculations show that lattice displacements along this phonon mode significantly alter the band structure [3]. For a large phonon amplitude, an indirect band-crossing transition occurs and tellurium becomes semimetallic. Because tellurium is uniaxial, two independent elements of the dielectric tensor must be measured to fully characterize the ultrafast response. Our femtosecond time-resolved ellipsometry technique involves measuring the absolute reflectivity of the white-light probe for multiple angles of incidence and numerically inverting the Fresnel formulae to obtain dielectric function values [4,5]. In the case of tellurium, four measurements must be made because each element of the dielectric tensor is complex. Each element describes the optical properties along certain directions in the crystal: the ordinary part corresponds to the ab-plane and the extraordinary part to the c-axis (helical axis). The symmetry-preserving phonon mode excited in Te is confined to the ab-plane, so we expect that the ordinary dielectric function will be most sensitive to these oscillations.

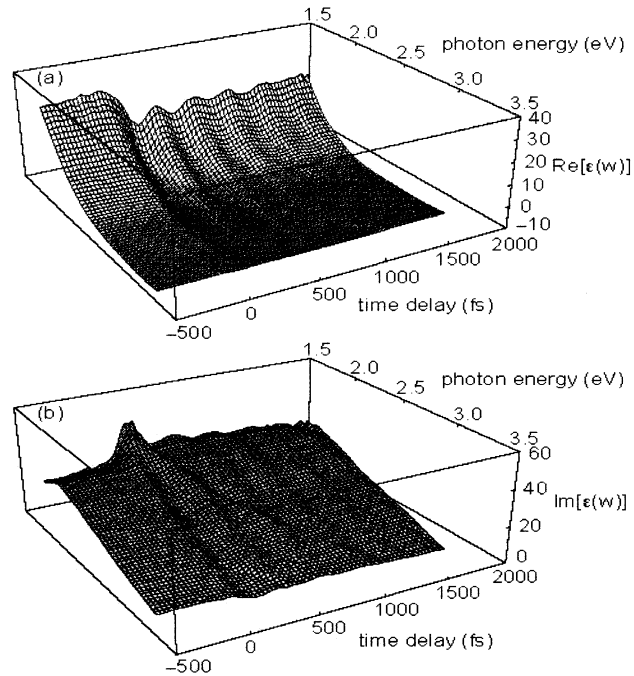


Figure 1 shows the dynamics of the ordinary dielectric function when the sample is irradiated by 50-fs, 800-nm pulses at a fluence of 120 J/m^2 . Before the pump arrives, our measurements of the dielectric function agree with literature values obtained by continuous wave ellipsometry [6]. After excitation, both the real and imaginary part of the ordinary dielectric function shift to lower photon energies and oscillate. The decaying offset from the initial values represents the relaxation of the equilibrium helical radius as electrons diffuse from the pumped region. Fourier transforms of the oscillatory signal show a redshift of the coherent phonon frequency with increasing excitation, in agreement with theoretical predictions [2,7] and previous reflectivity studies [8].

One important feature of the dielectric function is the photon energy where the real part crosses zero because it is a measure of the bonding-antibonding split in a material.

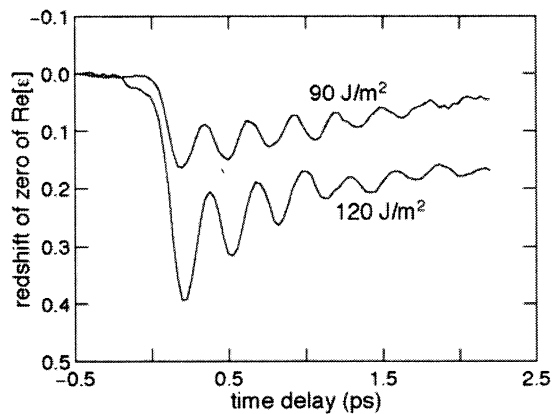


Figure 2 shows the change in this photon energy as a function of time delay for two pump excitations. As the excitation fluence is increased, the overall redshift of the zero and the amplitude of its oscillation

increases, indicating an increase in the shift of equilibrium lattice positions and in the amplitude of the coherent phonons. The decrease in the bonding-antibonding split reaches 0.4 eV for a pump fluence of 120 J/m², which exceeds the 0.3-eV band gap in Te. The magnitude of this decrease suggests that the band-crossing transition in tellurium can be modulated at THz frequencies.

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