Ultrafast dynamics of tellurium under high intensity photoexcitation

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Introduction

Ultrashort pulses of light offer the opportunity to study both the electronic and structural dynamics of a solid on the femtosecond time scale. When an intense femtosecond laser pulse is incident on an absorbing material, the dielectric function determines the linear optical response. Coherent phonons in tellurium

Ultrasound phase transitions in solids do not always involve disorder. In tellurium, impulsive photo-excitation causes a lattice displacement toward a band-crossing transition. At the same time, coherent phonons are generated and the lattice maintains its symmetry. Below damage threshold, ultrashort-pulse excitation of tellurium weakens the bonds of the lattice without disordering. Instead, new equilibrium positions are established and the ions oscillate around them. Changes of the lattice directly affect the band structure and, in turn, ε(ω). By monitoring the oscillation of ε(ω), we infer information about the oscillatory changes of the band structure.

A wide variety of techniques are used to study phase transitions in solids: from single-wavelength to broadband probes, from IR to visible to x-ray. We choose a broadband probe to measure the response of the dielectric function from the near-IR to the near-UV to intense femtosecond-pulse excitation. Because the dielectric function is determined by the band structure and electron occupation, it is a probe of both the electronic configuration and lattice structure. We track major features (e.g., the E₁ and E₂ peaks) of the dielectric function using femtosecond ellipsometry.

Below damage threshold, the plot below shows the dynamics of Re[ε(ω)] for a pump fluence of 120 J/m². The shift of ε(ω) to lower photon energy indicates that the bonding-antibonding splitting decreases as the lattice distorts. The magnitude of this decrease exceeds the band gap for about 100 fs, indicating that the bands indirectly cross.

The anharmonic effect is less important that, in the case of tellurium, anharmonic effect shown in the top right graph. This indicates that, in the case of tellurium, anharmonicity is less important than softening in determining the phonon frequency. We performed a Short Term Fourier Transform (STFT) on our data to track the evolution of the local frequency over the entire signal. We find that the local frequency does not significantly change over the observed range of our data compared to the change in phonon frequency with excitation strength.

The plots show the error in Re[ε] and Im[ε] given a 1% accuracy in the measurement of p-polarized reflectivity at 45° and 78° angles of incidence. Solid lines indicate the dielectric properties of various solids from the near-IR to the near-UV (1.5 to 3.5 eV). The plots below show the dynamics of Re[ε(ω)] for a pump fluence of 120 J/m². The shift of ε(ω) to lower photon energy indicates that the bonding-antibonding splitting decreases as the lattice distorts. The magnitude of this decrease exceeds the band gap for about 100 fs, indicating that the bands indirectly cross.

By performing Fourier transforms on the oscillatory signal we observe that the phonon frequency decreases with increasing excitation. This observation suggests either weakening of the covalent bonds (softening) or that the large amplitude phonons experience anharmonic effect. We find that the local frequency does not significantly change over the observed range of our data compared to the change in phonon frequency with excitation strength. Our STFT spectra do not exhibit the anharmonic effect shown in the top right graph. This indicates that, in the case of tellurium, anharmonicity is less important than softening in determining the phonon frequency.

Coherent phonons in tellurium

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Time and frequency resolved dynamics

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