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Impact of coherent phonons on the linear and non linear time-resolved optical properties of Tellurium and Tungsten Ditelluride

The study of transient, far-from-equilibrium states in complex materials can offer profound insights into their microscopic properties, particularly when multiple observables are examined simultaneously. A central question in this field is whether atomic lattice displacements can be accurately tracked and controlled.

In this work, we investigate the ultrafast dynamics of Tellurium (Te) and Tungsten ditelluride (WTe_2) through time-resolved reflectivity (TR-R) and second-harmonic generation (TR-SHG) measurements, as a function of the pump fluence. This dual-probe approach, comprising both linear and nonlinear optical responses, provides a comprehensive framework for analyzing electronic and lattice dynamics in both crystals.

Tellurium has four zone-center optical phonons, three of which are Raman active. These include the non-degenerate A_1 mode, which is linked to symmetric intrachain dilation and compression along the chain axis, impacting bond lengths and angles. Our measurements specifically identify the A_1 mode in tellurium sample, where a marked reduction of the SHG signal after Te photoexcitation suggests a transient transition toward a more centrosymmetric phase.

We conducted TR-R and TR-SHG measurements on WTe_2, with the electric field oriented along two orthogonal in-plane axes of its orthorhombic unit cell. We identified two primary coherent phonon modes. One, having frequency of ~ 0.24 THz, consists in a uniform in-plane atomic shift and is termed shear mode, while the other, at ~ 2.4 THz, reflects atomic displacements depending on the specific atom.

A notable (~ 90°) phase shift in the shear mode, as induced by varying the pump fluence, is observed in both the TR-R and TR-SHG signals, and along the two crystallographic directions. This finding suggests that the phase shift of the coherent oscillation is induced by a true fluence-dependent modification of the initial phase of the atomic displacements, allowing for a precise control of the atom dynamics.

These findings provide new perspectives on the interactions between coherent phonons and the electronic and lattice properties of complex materials, emphasizing the importance of selecting appropriate experimental observables to extract quantitative information on material properties. Notably, we demonstrate that pump fluence can serve as an effective tool to significantly adjust and tune the initial oscillation phase of coherent displacements.

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