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Resonantly enhanced X-ray impulsive vibrational spectroscopy in trigonal tellurium

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Laser control strategies enable perturbing selected degrees of freedom and driving matter in metastable states often inaccessible under equilibrium conditions. In the optical regime this is achieved through either non-linear phononics or impulsive vibrational spectroscopy (IVS), in which coherent phonons are launched upon ultrashort laser excitation. The impulsive vibration of the lattice causes macroscopic changes in the system's refraction index that can be detected using a second probe laser in a pump-probe fashion. Extending this scheme to the X-ray domain promises achieving element-selectivity in the generation process by tuning the pump pulses on-resonance with core electronic excitations.

In this contribution, we present our recent implementation of X-ray impulsive vibrational spectroscopy (XIVS) in trigonal tellurium performed at the FERMI free electron laser (FEL) in Trieste. By tuning the FEL pump photon energy either off- or on-resonant with the tellurium N_{4,5}-edge, we demonstrate a resonant enhancement of the coherent phonon amplitude by a factor of eight that is rationalized in terms of dispersive excitation of coherent phonons (DECP) mechanism. Ab initio simulations including ultrafast electron and coherent phonon dynamics in presence of electron-phonon interactions show an excellent agreement with our experiment, underscoring the key contribution of band-specific electron phonon coupling (EPC) terms.

These results demonstrate the possibility of harnessing EPC to enhance the light-induced activation of specific phonons, paving the way for element-selective coherent control strategies in functional materials.

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