L.6: Nonlocal probing of amplitude mode dynamics in $EuTe_4$ using time-resolved X-ray diffraction

$EuTe_4$ is a quasi-two-dimensional metallic compound that shows semiconducting behaviour (~ 0.2 eV bandgap) in the charge-density-wave (CDW) state. In the CDW state below T_{CDW} ~726 K, amplitude mode emerges from the collective excitation of frozen lattice distortions. Here, we present the nonlocal probing of amplitude mode dynamics far beyond the optically excited depth of ~150 nm in the CDW state of ~10 μm thick single-crystal $EuTe_4$ using time-resolved X-ray diffraction (TXRD) in the transmission geometry. 'Nonlocal' refers to the effect of laser pumping propagation far beyond the photoexcited region.

The experiment was performed using a 1 kHz, 50 fs, ~ 5.7 mJ Ti:sapphire laser system. A beam splitter reflects ~10% energy, which acts as a pump pulse to excite the sample. The remaining (90%) beam was focused on a moving Cu wire target to create ultra-short laser plasma copper K_α X-ray source, which acts as a probe pulse. These X-rays were focused on the sample (spot size ~ 330 μm) using a polycapillary lens. Both the pump and probe pulses collinearly overlap at the sample. The $EuTe_4$ sample was a single crystal of dimension ~ 0.7 mm (W) × 1.0 mm (H) × 10 μm (T). A combination of a DRZ phosphor screen and an EMCCD camera was used to detect the XRD from $EuTe_4$ sample in transmission geometry. The TXRD pattern of $\boldsymbol{Q}_{CDW}$ = (2,2,0)−$\boldsymbol{q}_{CDW}$ peak ($\boldsymbol{q}_{CDW}$ = (0,0.643,0) r.l.u.) was recorded with 300 fs temporal resolution, which is limited by the pulse duration of the probe. To detect low-intensity signals of CDW peak, the path between the X-ray source and DRZ phosphor was shielded by a 30 mm thick lead sheet. A small shift in the diffracted angle of the (2,0,0) Bragg peak due to heat deposition by the pump beam was detected by an X-ray CCD.

Figure L.6.1(a) shows the underdamped oscillations of the CDW peak intensity for 2 mJ/cm^2 fluence. The oscillations in the CDW peak intensity are fitted with the dominant frequency (black line), corresponding to the peak observed in a Fast Fourier Transform (FFT) of the data (Fig. L.6.1(b)). The observed oscillations are found to occur at the amplitude mode frequency. Figure L.6.1(c) shows the decrease in oscillation frequency with increasing excitation fluence, which is rationalized by sample heating by comparing with polarized Raman scattering measurements performed at different temperatures. The temperature rise on laser excitation is measured from the peak shift of (2,0,0) Bragg peak, as shown in Figure L.6.1(d). The constant peak shift after the first delay point indicates the rise in the equilibrium sample temperature. The underdamped oscillation amplitude is at odds with the observed overdamped nature of the amplitude mode measured using meV resolution inelastic X-ray scattering and polarized Raman scattering.

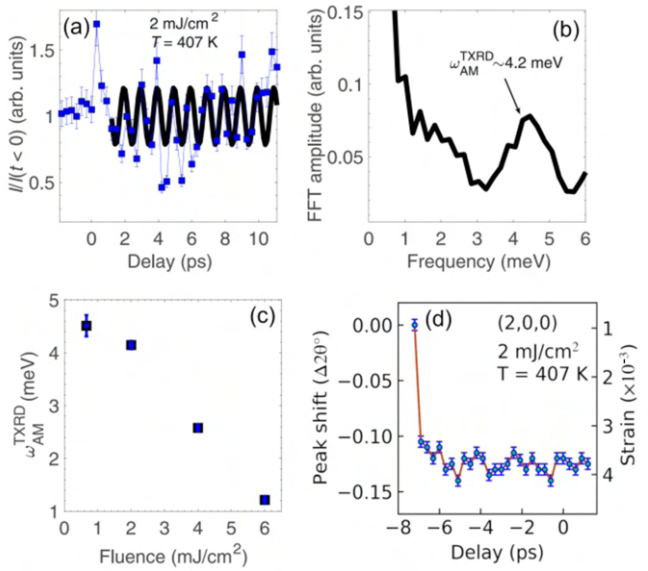

Fig. L.6.1: (a) TXRD intensity (I/I(t<0)) at $\boldsymbol{Q}_{CDW}$ (blue square markers) for 2 mJ/cm^2 fluence. The solid black line shows the fit with the dominant oscillation frequency, (b) FFT amplitude versus frequency of intensity reported in (a), (c) variation in oscillation frequency with excitation fluence, and (d) peak shift (strain) of the (2,0,0) Bragg peak as a function of delay for 2 mJ/cm^2 fluence. The rise in the equilibrium sample temperature is indicated in the panel.

We propose the nonlocal behaviour and underdamped nature of the oscillation amplitude are possibly due to the generation of either of the two quasi-particles, viz., phonon-polariton or exciton-polariton, which propagates with light-like speed beyond the optically excited depth. The observations cannot conclusively confirm or rule out one, but it is proposed that the measured oscillations are likely from propagating phonon-polariton.

In conclusion, the observed nonlocal behaviour and underdamped oscillations in the TXRD signal of the CDW peak can be rationalized by coupling of the polar amplitude mode with incident IR photons to form a phonon-polariton quasiparticle. The decrease in oscillation frequency with increasing excitation fluence is rationalized by the sample heating. The observed nonlocal behaviour of amplitude mode provides an opportunity to engineer material properties at a significantly faster time scale with optical pulses. For more details, please refer to: *R. Rathore, H. Singhal, V. Dwij*, M. K. Gupta*, A. Pathak*, J. A. Chakera, R. Mittal*, A. P. Roy*, A. Babu*, R. Kulkarni*, A. Thamizhavel*, A. H. Said*, and D. Bansal*, Ultrafast Science - A Science Partner Journal (AAAS), 3, 041 (2023).*

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