Time-Resolved Optical-Pump/Terahertz-Probe Spectroscopy of the Charge-Density-Wave Phase Modes in Blue Bronze

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Charge-density waves (CDW) are elementary excitations in low-dimensional organic/inorganic crystals spontaneously coupling conduction electrons and lattice. They manifest in a periodic charge-density modulation and the formation of an electronic gap at the Fermi edge [1]. Besides excitation across this gap (which typically occurs in the mid-IR), the CDW possess low-lying modes [2], which usually separate into a Raman-active amplitude- and an IR-active phase-channel. The latter appear as "phase-phonon" bands (including a low-energy "phason" near ω=0 [3]) in THz conductivity spectra. While the amplitude-phonons have been studied by others via Raman probing [4], we investigate the phase-phonons via their non-equilibrium response after pulsed optical excitation for the case of the prototypical quasi-1D CDW system blue bronze ($K_{0.3}MoO_3$).

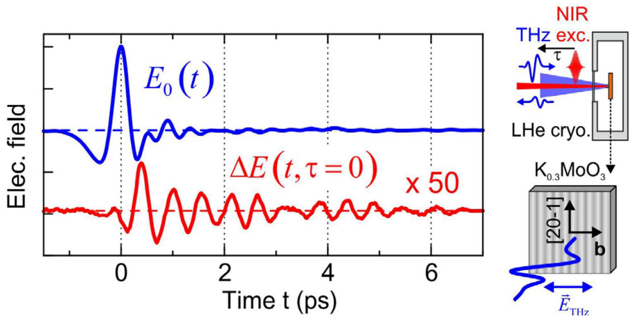

Fig.1. An example of a pump-induced differential THz field ΔE(t,τ=0) for T=50 K ($h\upsilon_{ex}$=1.6 eV, F_{ex} =550 μJcm^{-2}) measured in reflection, with a detection bandwidth of 0.5-2.8 THz. From 2D maps of such data, we derive the transient complex conductivity Δσ(ω,τ) which reveals a photoinduced blue-shift of the phase-phonon bands and a dynamic broadening.

From the transient THz response of the system (see Fig. 1) and a comparison with the response expected on the basis of the time-dependent Ginzburg-Landau framework (TDGL) previously introduced for the amplitude-phonons [4], we find that we can reproduce the data, if we generalize the TDGL model to account for screening effects and impurity interactions. It turns out that such interactions specifically affect the phase-channel, and remain nearly hidden if only the amplitude-channel is investigated. This can be understood qualitatively because the phase-modes involve the translation of the CDW condensate, hence are more directly related to the charge transport and should be more sensitive to effects such as impurities, intra- and inter-chain interactions. Moreover, a comparison of the predictions of the phenomenological TDGL and quantum-mechanical models [2] reveals *qualitative* differences. Further experimental studies of the phase-channel and theory development are required to reach a complete, unified description of CDW physics.

References

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