Electronic Dynamics and Coherent Phonons in Gold-Bromide Perovskite

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Halide perovskites are polar crystalline materials with significant coupling between electronic and vibrational degrees of freedom, which leads to peculiar photo-induced non-equilibrium dynamic processes. For example, in the well-studied class of lead halide perovskites, a hot phonon bottleneck effect slows down the cooling of hot charge carriers. The toxicity and limited stability of lead-based perovskites make the case for exploring lead-free alternatives for opto-electronics. Here we study photo-induced processes in caesium gold bromide perovskite. Using femtosecond transient absorption and Raman spectroscopy inter-valence charge transfer states[1] and their coupling to the lattice is elucidated. Room-temperature pump-probe measurements reveal pronounced oscillations in the differential absorption lasting tens of picoseconds (Figure 1). Raman spectroscopy identifies LO phonon modes of the crystal that are coherently excited by the femtosecond optical excitation and modulate the spectrum. The amplitudes, frequencies and dephasing of the coherent oscillations are studied to understand the mechanism of generation and damping of coherent phonons. The slow dephasing of coherent lattice vibrations points towards a reduced anharmonicity of the LO phonons and hence a lower rate of heat dissipation into the other modes[2,3]. By deconvoluting the electronic and vibrational dynamics from the pump-probe spectra we reveal the potential of the material for optical manipulation of carrier cooling and lattice structural dynamics.

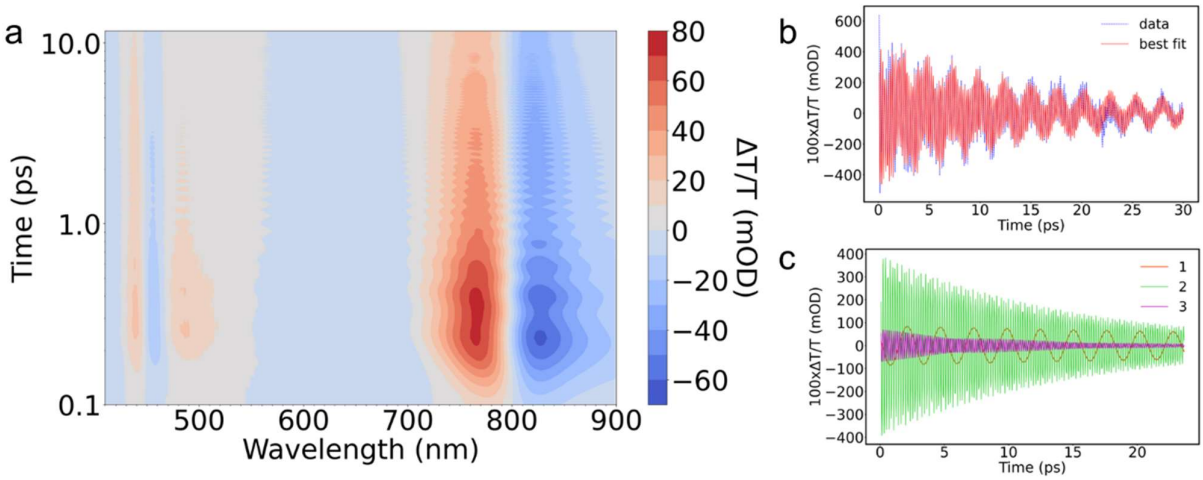

Figure 1: (a) Pump-Probe Spectra of Caesium gold bromide; (b) residual signal after subtracting the electronic relaxation at 790 nm demonstrating coherent nuclear dynamics; (c) deconvolution of the vibrational dynamics into components.

References

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