

Generation of coherent acoustic phonons in lead free double perovskite $\text{Cs}_2\text{AgBiBr}_6$

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Halide double perovskite $\text{Cs}_2\text{AgBiBr}_6$ is a perspective material for photovoltaic and photoelectronic applications as a non-toxic and stable alternative for solution-grown lead-based perovskites. Phonons and electron-phonon interaction play an important role in its optical properties. $\text{Cs}_2\text{AgBiBr}_6$ possesses cubic crystal structure (space group #225) at room temperature and under 120 K it undergoes a phase transition to a tetragonal structure (space group #87).

Bulk crystal was studied using nonlinear optical pump-probe microscopy. In Figure 1 differential reflectivity transient obtained in the tetragonal phase is presented. At zero delay time between pump and probe, we observe the sharp change in reflectivity followed by exponential decay that is attributed to the dynamics of photoexcited carriers. After subtraction of the exponential signal, high-frequency oscillations are observed (see Fig.1 inset). The oscillations occur due to pump-induced excitation of the strain pulse and subsequent interference of the reflected probe beams from the crystal surface and the propagating strain pulse. Under excitation along [111] crystal direction in a cubic phase, we observe oscillations with only one frequency that correspond to the coherent longitudinal acoustic phonons (LA). For a lower symmetry tetragonal phase, we observe oscillations with two frequencies of 23 and 11 GHz that are attributed to LA and one of the transverse acoustic (TA) modes, respectively. With increasing the temperature close to phase transition at $T \sim 120$ K TA phonons frequency shift to lower energies and eventually disappear. A theoretical model based on anisotropic deformation in the tetragonal phase is used to explain the experimental results.

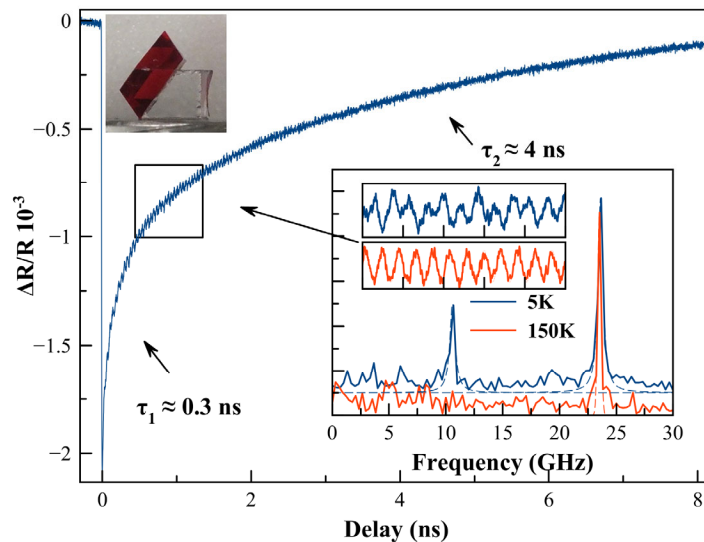


Figure 1. Pump-probe transients for bulk $\text{Cs}_2\text{AgBiBr}_6$ crystal in tetragonal phase (at 5K), along [111] optical direction.