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Probing Lattice Dynamics in Quantum-Confined Materials on Ultrafast Timescales

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Experimental measurement of lattice dynamics in few nanometer semiconductor nanocrystals present difficulty owing to low frequency phonon modes and the possibility of rapid dynamics. We utilize recently developed femtosecond stimulated Raman spectroscopy in order to characterize longitudinal optical (LO) phonon production and dissipation throughout the process of confinement-enhanced, ultrafast intraband carrier relaxation. Prior to photoexcitation, CdSe nanocrystals produce a stimulated Raman spectral shape that resembles the spontaneous Raman spectrum. Upon excitation, we observe a decrease in stimulated Raman amplitude and note a size-independent LO phonon formation time. Mode softening is observed as is evidence of phonon down-conversion processes. Furthermore, spectrally and temporally resolved photoluminescence suggest evidence of acoustic phonon dissipation times that follow diffusive transport, which we can manipulate.