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Lattice and Carrier Dynamics in Quantum-Confined Materials on Ultrafast Timescales

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ABSTRACT: Excess carrier energy, which many aim to utilize for advanced energy conversion technologies, rapidly dissipates from electrons and holes in both bulk and quantum confined semiconductors despite expectation of slowed cooling in the latter. We aim to characterize dissipation channels available to carriers via time-resolved optical spectroscopies. We utilize both femtosecond stimulated Raman spectroscopy (FSRS) and time-resolved photoluminescence in order to gain insights as to rates and modes of dissipation. Using FSRS, we characterize longitudinal optical (LO) phonon production and dissipation throughout the process of confinement-enhanced, ultrafast intraband carrier relaxation. Upon excitation, we observe a decrease in stimulated Raman amplitude and note a size-independent LO phonon formation time. Mode softening is observed along with 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.

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