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Title:Loosening quantum confinement: Observation of real conductivity caused by hole polarons in semiconductor nanocrystals smaller than the Bohr radius

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Abstract:We report on the gradual evolution of the conductivity of spherical CdTe nanocrystals of increasing size from the regime of strong quantum confinement with truly discrete energy levels to the regime of weak confinement with closely spaced hole states. We use the high-frequency (terahertz) real and imaginary conductivities of optically injected carriers in the nanocrystals to report on the degree of quantum confinement. For the smaller CdTe nanocrystals (3 nm < radius < 5 nm), the complex terahertz conductivity is purely imaginary. For nanocrystals with radii exceeding 5 nm, we observe the onset of real conductivity, which is attributed to the increasingly smaller separation between the hole states. Remarkably, this onset occurs for a nanocrystal radius significantly smaller than the bulk exciton Bohr radius a<inf>B</inf> ∼ 7 nm and cannot be explained by purely electronic transitions between hole states, as evidenced by tight-binding calculations. The real-valued conductivity observed in the larger nanocrystals can be explained by the emergence of mixed carrier-phonon, that is, polaron, states due to hole transitions that become resonant with, and couple strongly to, optical phonon modes for larger QDs. These polaron states possess larger oscillator strengths and broader absorption, and thereby give rise to enhanced real conductivity within the nanocrystals despite the confinement. © 2012 American Chemical Society.

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