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State-to-state femtosecond relaxation dynamics of excitons in semiconductor quantum dots. PATANJALI KAMBHAMPATI, SAMUEL SEWALL, RYAN COONEY, KEVIN ANDERSON, EVA DIAS, McGill University — Size dependent exciton relaxation dynamics are measured in colloidal CdSe quantum dots using exciton selective femtosecond spectroscopy. Preparation of the initial excitonic state allows evaluation of state-to-state exciton dynamics. These methods reveal the electron and hole relaxation dynamics from a specified initial state to a specified final state, with a precision of 10 femtoseconds. These state selective, size dependent experiments confirm previously observed confinement induced femtosecond Auger channels for electrons with increased precision. This increased precision allows for unambiguous, quantitative evaluation of size dependent transition matrix elements. These experiments furthermore show that the hole relaxation rate increases for smaller quantum dots, contradicting expected relaxation mechanisms for holes. We propose a new confinement enhanced non-adiabatic pathway for hole relaxation in colloidal quantum dots, overcoming the predicted phonon bottleneck for holes. Finally, these experiments show exciton state specific biexcitonic interactions.

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