Pauli blocking dynamics in optically excited quantum dots: A picosecond excitation-correlation spectroscopic study

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Pauli's exclusion principle is the cornerstone in the physics of the many fermions in finite systems. The phenomenon of Pauli blocking, i.e., the physical manifestation of the exclusion principle is of course responsible for stability of matter. It is also important in specific processes of interest in fields ranging from nuclear physics, cold atomic gases, to semiconductor devices. We have studied Pauli blocking through the time-resolved dynamics of carrier redistribution between the discrete energy states in the self-assembled InAs/GaAs QDs through a novel variant of the picosecond excitation-correlation (EC) spectroscopy [1,2].

A train of pulses of 100 fs duration were split into two beams of equal fluence. The arrival of the pulses onto the sample from one of the beams was controllably delayed with respect to the corresponding pulses from the other beam and the steady-state PL was measured as a function of this delay. Since the number of photons incident on the sample is independent of the delay, this technique specifically probes the time-resolved dynamics of the nonlinear PL. We have observed that while the ground state EC response is always snubbed when the two excitation beams are temporally nearly coincident, the excited state response can either be enhanced or reduced, depending on the excitation power. This is a direct consequence of the exclusion principle. The time evolution of this response was studied for the first three levels in a QD ensemble. The observations were quantitatively reproduced using a minimal theoretical model, which does not use the rate equations, but combines carrier loss kinetics with principle of detailed balance and the exclusion principle.

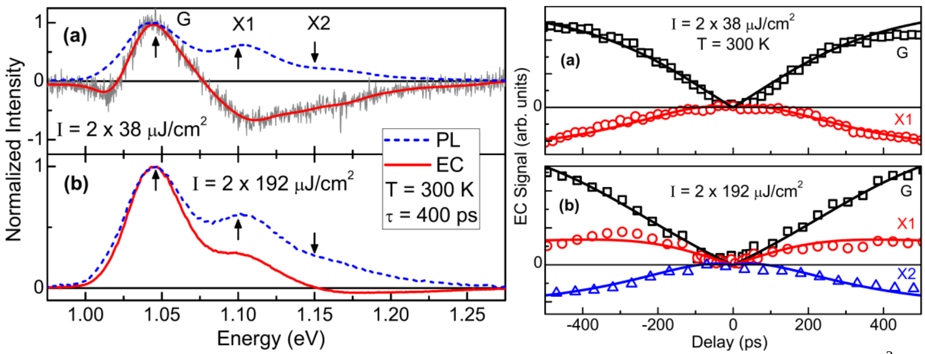

Fig: (Left) Normalized room temperature PL and EC spectra for the fluence I = 2 × 38 mJ/cm^2 (a) and 2 ×192 mJ/cm^2 (b). Note that the sign of the EC signal for the X1 state changes from negative to positive between (a) and (b). (Right) Delay dependence of the EC signal measured at energies indicated by arrows in Fig. (a) and (b) (Left). The solid lines are fits to the data by the model. Note that the definition of EC signal $EC(\tau, h\nu) = PL_{12}(\tau, h\nu) - PL_{12}(\tau=0, h\nu)$, where τ is the delay and $h\nu$ is the photon energy, is opposite to that in refs [1-2].

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