Coherent Quantum Control of Biexciton in a Single Quantum Dot

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We demonstrate the direct two-photon coherent control of the biexciton state in a single self assembled QD achieved by the variation of the relative phase in a sequence of two optical pulses. The excitation photon energy is tuned to the half-energy between the groundstate and the biexciton state, consisting of two electron-hole pairs in the lowest single-particle energy shells. Utilization of II-VI QDs, where the difference in binding energies of exciton and biexciton is as large as 20 meV, allows us to exclude single photon processes by choosing the pulses as short as 1 ps. At the same hand interference in the biexciton population can be monitored by the spontaneous emission of the biexciton itself as well as by the subsequent decay of the single exciton left behind in the biexciton recombination.

The CdSe/ZnSe QD samples are grown by molecular beam epitaxy. The pure CdSe core has a height and lateral size of about 2 nm and 5-10 nm, respectively. Under resonant single pulse excitation we observe Stokes and anti-Stokes features placed symmetrically with respect to the excitation laser (~10 meV). The yield of the lines as a function of excitation energy and power as well as the polarization properties demonstrate unambiguously that the signal originates from cascaded biexciton (XX) and exciton (X) emission, subsequent to resonant two-photon excitation, in one and the same QD.

Under excitation with a sequence of two-linearly co-polarized pulses, both X and XX lines oscillate in phase when the change of the delay occurs on the wavelength scale (<500 nm). When the pulses totally overlap, the interferograms reflect the optical interference between the two pulses and, accordingly, all lines oscillate with the laser frequency. At a delay larger than 2 ps, where the pulses are well separated in time, X and XX oscillate with twice the laser frequency. Oscillations at these time delays with a frequency corresponding to the energy between the ground and biexciton states result from the interference of the biexciton wave functions, i.e. coherent quantum control. Dephasing destroys the interference. We find however that interference contrast decays non-exponentially on a 10-ps time scale. The reason for this is a dynamical inhomogeneous broadening of the energy levels caused by long-term fluctuations in the QD environment. Data both in spectral and time domain reveal a specific situation, where exciton and biexciton energies fluctuate largely in parallel, giving rise to a narrow biexciton PL. The intrinsic decoherence is slower than the decay of the interference contrast, enabling hence coherent manipulation on longer time scales with appropriate single pulse sequences.