Ultra-Fast Coherent Exciton Dynamics in Core-Shell GaAs/AlGaAs Nanowires

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Entanglement and strong coupling are the basis for quantum computing. Current optical applications are based on the coupling of single quantum emitters. This approach, however, requires exact control over system parameters, initial states, and timing. A competing approach could be an open cavity quantum system in which many coupled quantum emitters reach a steady state, which is entangled. A plasmonic semiconductor nanowire structure using excitons and open cavity plasmons could serve as a many quantum emitter system.

To investigate the ultrafast coherent and lifetime dynamics of excitons in GaAs/AlGaAs/GaAs core-shell-cap semiconductor nanowires we performed power and excitation wavelength dependent three-beam heterodyne four-wave mixing (HFWM) measurements. The nanowires were removed from the GaAs substrate and transferred on a sapphire slide. A tuneable 150 fs Ti:sapphire laser was used as an excitation source. The generated HFWM signal is measured through balanced photodiode detection with a lock-in amplifier reducing the scattering background of the nanowires by several orders of magnitude. The measurements were performed at 14 K to mitigate scattering with optical phonons. Figure 1 shows the contour plot of the coherent HFWM amplitude of undoped GaAs(150nm)/$Al_{0.2}Ga_{0.8}As$(75nm)/GaAs(30nm) core-shell-cap nanowires as a function of the delay times τ_{12} between pulses $\mathbf{k}_1$ and $\mathbf{k}_2$ before the arrival of probe pulse $\mathbf{k}_3$. The generated FWM E-field amplitude is probed with a reference pulse $\mathbf{k}_{ref}$ as a function of delay τ_{3ref} in this photon echo configuration. The experiment shows distinct quantum beats along the τ_{12} axis as well as on the τ_{3ref} axis for positive delay indicating an in-built strain which splits the heavy- and light-hole exciton energies [1]. The measurements also reveal that the exciton transitions in the nanowires are predominantly homogeneously broadened with a fast exciton dephasing time T_2 of less than 1 ps. The fast dephasing is mainly attributed to a high scattering rate with electron hole pairs at an excitation wavelength of 820 nm as well as with impurities. The slight asymmetry of the photon echo signal with respect to negative and positive delays τ_{12} indicates the presence of weak inhomogeneous broadening which increases at lower excitation energy due to the involvement of acceptor bound exciton transitions. HFWM measurements in lifetime configuration ($\tau_{12} = 0$ and $\tau_{ref3} = 0$) as a function of delay time τ_{13} between pulses $\mathbf{k}_1$ and $\mathbf{k}_3$ reveal a triexponential decay of 3 ps, 40 ps and 1 ns. The different decay times are related to the initial dephasing, then fast capture of excitons at surface states or defects at the AlGaAs interface, and finally the radiative recombination, respectively. A many-level Optical Bloch equation model was used to examine the coherent effects observed in the GaAs/AlGaAs/GaAs core-shell-cap nanowires.

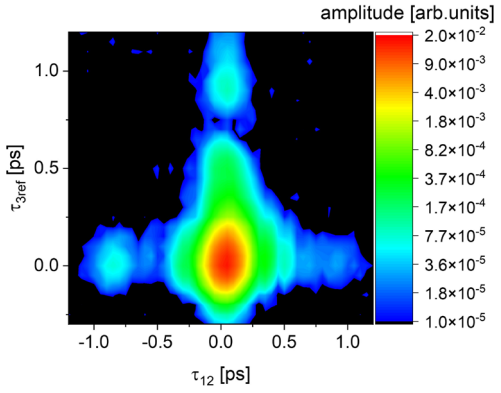

Fig.1. Contour plot of the heterodyne FWM amplitude of GaAs/AlGaAs/GaAs NWs resonantly excited at 820 nm at 14 K.

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References

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