Ultrafast Spectral Hole Burning Spectroscopy of Exciton Spin Relaxation in Quantum Dots

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The exciton spin relaxation within the radiative doublet of the exciton ground state in InAs/GaAs selfassembled quantum dots is studied via an ultrafast spectral hole burning technique. In the case of cross-polarized pump and probe pulses a spectral "anti-hole" emerges due to relaxation of the exciton spin. The measured relaxation time decreases rapidly from 1.15 ns at T = 5 K to 90 ps at 90 K, suggesting exciton-acoustic phonon interaction as the underlying spin relaxation mechanism.

Introduction

The carrier spin couples much weaker to the environment than the carrier wavefunction. Thus, spin states in semiconductor quantum dots (QDs) are a promising candidate for the implementation of future quantum logic devices. Indeed, very long (~ns) spin relaxation times have been found in III-V [1] – [3] and II-VI [4] self-assembled QDs. Studies of spin-polarized carriers confined in QDs have so far mainly been performed by time-resolved photoluminescence measurements on QD ensembles where spin populations are created using circularly polarized light excitation. In this contribution we report measurements of the spin relaxation in an ensemble of InAs/GaAs self-assembled QDs using an ultrafast spectral hole burning (SHB) technique [5].

Experimental

Sample Preparation

The investigated sample consists of 30 layers of InAs QDs with a dot density of approximately 2×10^{10} per cm^2 per layer. In order to be accessible to our Ti:sapphire laser system, the exciton transitions were shifted to higher energies by rapid thermal annealing. Excitonic ground state luminescence from the dots at T = 5 K is observed at 1.281 eV with an inhomogeneous broadening of the transitions of ~45 meV (FWHM).

Measurements

SHB measurements were performed using a modelocked Ti: sapphire laser that delivers 80-fs pulses with a center frequency of 1.285 eV and a spectral width of ~14 meV (FWHM). A grating pulse shaper was used to produce 1.4 meV (FWHM) broad pump pulses and the change in the transmission induced by the pump was measured with a weaker 80-fs probe pulse. The probe was spectrally dispersed with a monochromator which allowed the determination of the changes induced at photon energies different from that of the pump. A motorized translation stage controlled the delay between the pump and probe pulses. In addition, half-wave-plates were used to independently adjust the linear polarizations of both, pump and probe.

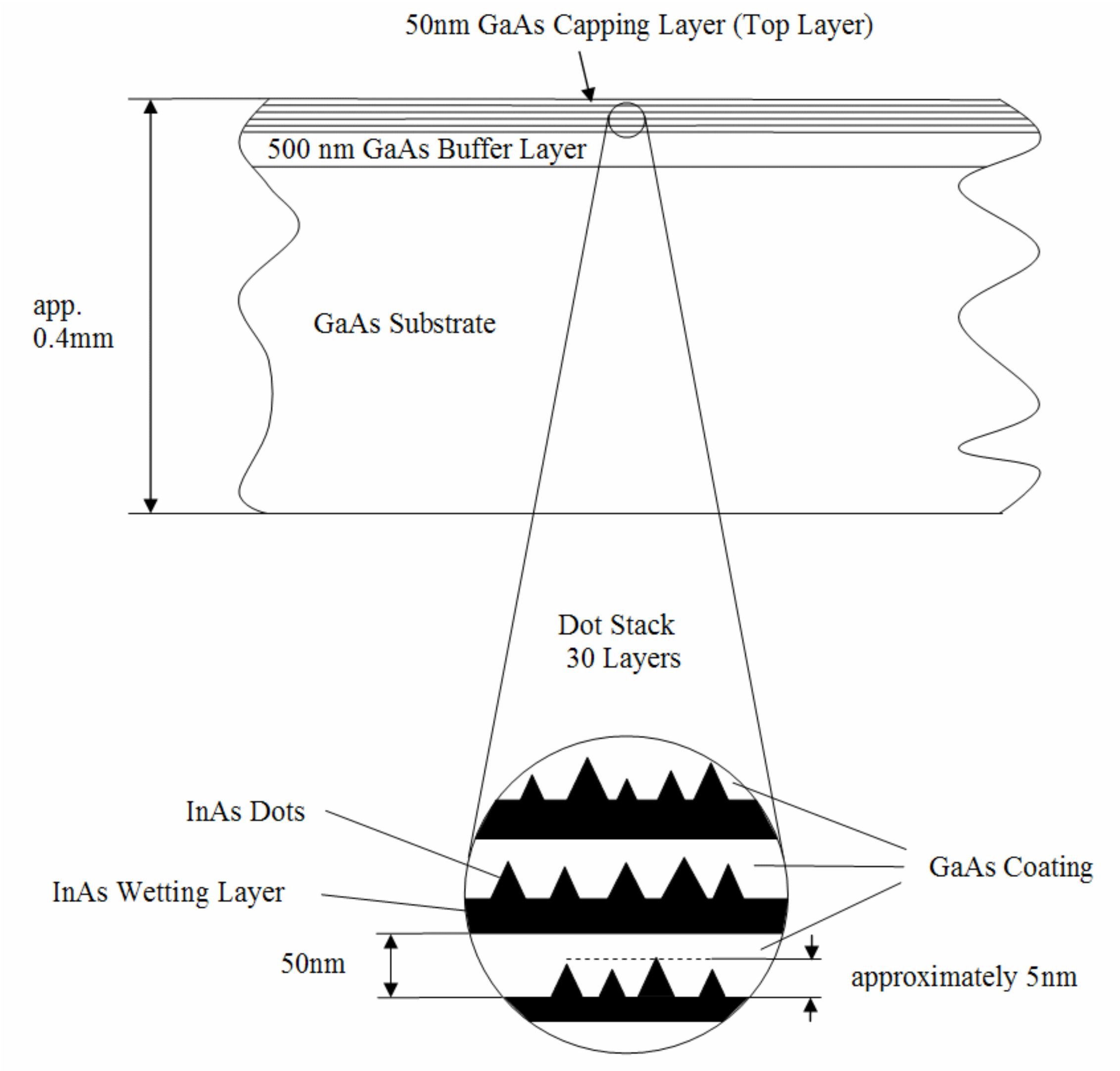

Fig. 1: Sample of self-assembled InAs / GaAs quantum dots

Figure 2 shows SHB signals recorded at T = 5 K. In these measurements a linear Πy-polarized pump pulse is tuned to the maximum of the excitonic ground state transition. The polarization of the broadband probe pulse is tuned either perpendicular (upper trace) or parallel (lower trace) to the pump and the differential transmission change of the probe is measured at a temporal delay of 10 ps after excitation. In the first case, the SHB signal shows enhanced transmission at the pump photon energy, corresponding to a bleaching of the |00> ground state population. In the case of parallel polarizations of the two pulses, we observe (i) enhanced transmission at the pump photon energy, and (ii) reduced transmission, i.e., an “antihole”, at energies below the pump photon energy [6]. The antihole arises from the population of the |01> exciton state which gives rise to absorption to the biexciton state |11> at an energy of ΔE_1 below the exciton peak. We determine a biexciton binding energy of 4.2 meV.

Results

The SHB signals strongly broaden at T = 90 K, and after deconvolution of the pump pulse we find a FWHM line width of 2.1 meV. Temperature dependent measurements show that the linewidth is determined by acoustic phonon-exciton interaction in the dots. The temporal evolution of the antihole at T = 90 K is shown in Fig. 2 (a). With increasing pump-probe delay, the ratio of cross-polarized signal to copolarized signal increases and reaches one for long delay times, as shown in the inset. It is obvious that the development of an antihole in the cross-polarized case is due to spin relaxation: As |01> excitons flip their spin, the |10> exciton state gets populated, giving rise to absorption to the biexciton state |11>. The spin relaxation rate r can be found from the equation $(A_{\parallel} - A_{\perp})/(A_{\parallel} + A_{\perp}) \sim \exp(-2\Gamma\tau)$, where $A_{\parallel}$ and $A_{\perp}$ denote the antihole amplitudes for co- and crosspolarized probe light, and τ is the pump-probe delay time. A fit yields $1/\Gamma$ = 90 ps. Figure 3 (b) presents the temperature dependence of the relaxation time. It increases rapidly from 90 ps to 1.15 ns as the temperature is reduced from 90 K to 5 K. At 5 K the spin relaxation time is more than two times longer than the exciton lifetime which remains approximately constant (~0.5 ns) over the investigated temperature range. The large change of the spin relaxation time suggests that phonon scattering related mechanisms become significant at high temperature. The observed temperature dependence is characterized by a small activation energy which strongly points to an acoustic phonon mediated spin flip process.

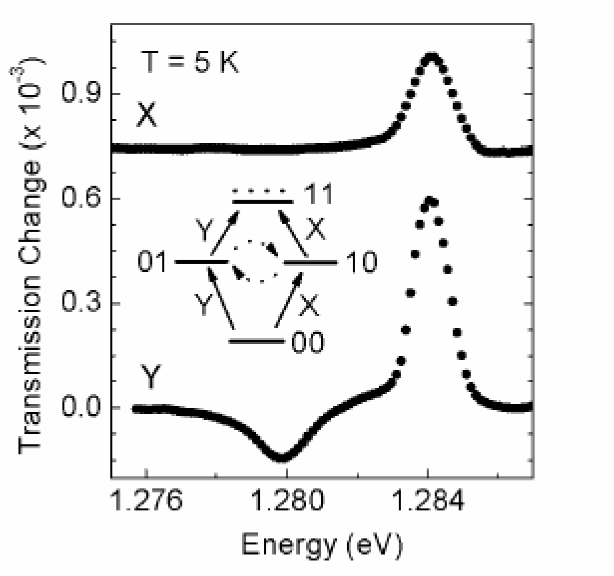

Fig. 2: SHB signals for Πx (upper trace) and Πy - polarized probe pulses. The polarization of the pump is in both cases Πy. Inset: Schematic drawing of the energy levels in a QD. |00> is the ground state; |10> and |01> correspond to the exciton states, which can be excited by Πx - and Πy - polarized light, respectively; and |11> denotes the biexciton state. The biexciton energy is less than twice the bare exciton energy, the difference between them being the biexciton binding energy ΔE.

Acknowledgements

This work was supported by the Austrian Fond zur Förderung der Wissenschaftlichen Forschung (SFBADLIS, SFB-IRON) and the ECNoE program SANDiE.

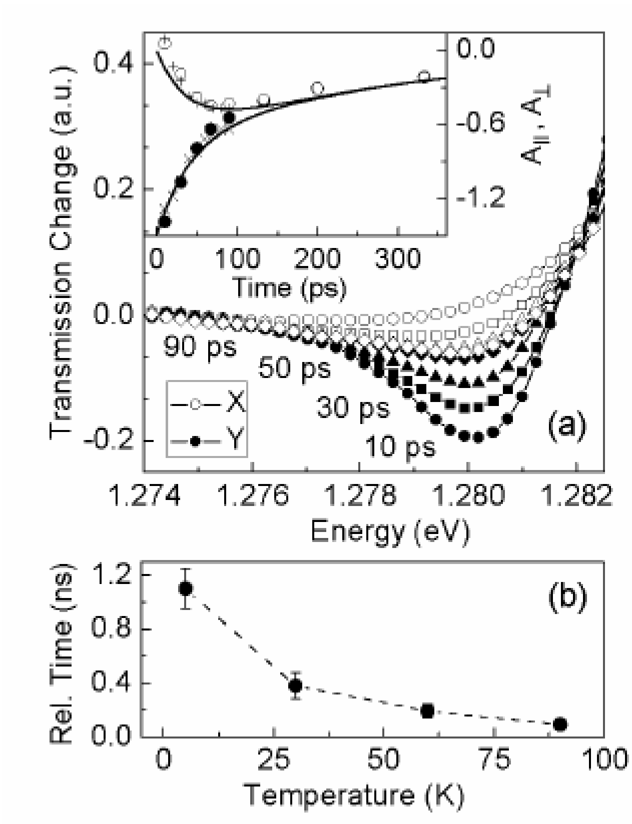

Fig. 3: (a) Temporal evolution of the antihole at T = 90 K. Inset: Antihole amplitudes for co- and cross-polarized pump and probe pulses (symbols: experimental data; lines: results from a simple rate equation model), (b) Temperature dependence of the spin relaxation time.

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