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Probing Charge and Spin Excitations in Quantum Dots and Molecules

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Introduction

We have investigated the potential to optically generate individual *electrons* in self-assembled quantum dots and program their spin orientation via the polarization of the laser excitation source. The results of these measurements demonstrate that spin relaxation in QD nanostructures is strongly suppressed when compared with higher dimensional systems and, moreover, show that spin relaxation is driven by spin-orbit coupling of the Zeeman levels mediated by single phonon scattering processes.

Quantum Dot Spin Memory Devices

Figure 1 shows the schematic structure and operating principles of the devices investigated. A single layer of self assembled InGaAs QDs is embedded in the intrinsic region of a p-*i*-Schottky photodiode. In the charge storage condition (Fig 1a), a negative potential is applied to the semitransparent Schottky gate with respect to the p-contact leading to a large static electric field along the growth axis of the dots. Single electron hole pairs are generated by selective optical excitation of the QD ground states. The spin orientation of these excitons is determined via the optical polarization due to the QD transition selection rules. Excitation with circularly polarized light with σ^+ helicity creates excitons with total angular momentum $J_X=S_e+J_h=+1$ ($S_e=-1/2$, $J_h=+3/2$) whereas σ^- excitation generates $J_X=-1$ ($S_e=+1/2$, $J_h=-3/2$). Whilst holes readily escape from the QDs by tunneling into the p-contact, electron escape is prevented due to the much smaller tunneling probability through the AlGaAs barrier. This process leaves single electrons with spin orientation $S_e=-1/2$ (+1/2) in the dots following σ+ (σ-) excitation. The distribution of electron spins in the QD sub-ensemble is tested after a storage time Δt by forward biasing the Schottky junction. Holes then drift into the negatively charged dots and generate a time delayed electroluminescence (EL) signal with a spectral distribution that directly reflects the spectral distribution of charges and polarization corresponding to their spin orientation.

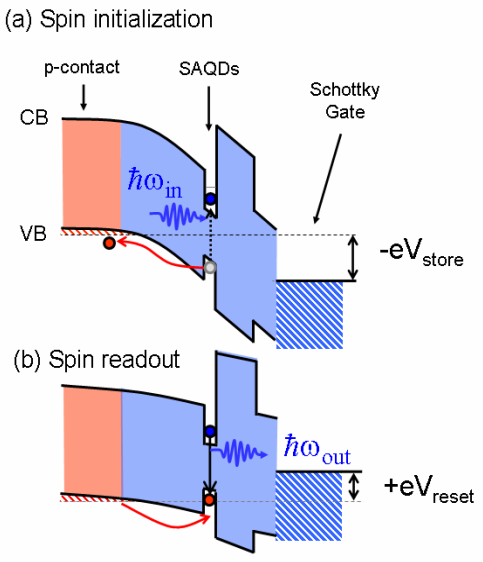

Figure 1 - Operating principle of the spin memory devices investigated.

As presented in figure 2, by performing measurements as a function of Δt we have directly probed the electron spin relaxation dynamics and their dependence on lattice temperature and magnetic field. The spin relaxation time (T_1) is found to be extremely long (e.g. >25ms at T=1K, B=4T) decreasing with

magnetic field according to a clear B^{-5} power law. Furthermore, T_1 is found to reduce linearly with lattice temperature and be very strongly sensitive to the motional quantisation (s-p shell splitting). Taken together, these observations firmly establish that spin-flip scattering in QDs is mediated by spin-orbit coupling at low temperatures and is strongly suppressed when compared to nanostructures with higher dimensionality.

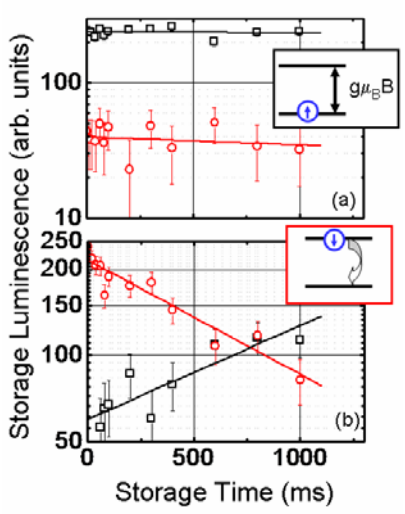

Figure 2 – Circular polarisation of storage luminescence following selective excitation of spins in lower (a) or upper (b) Zeeman levels. Following excitation of spin down electrons a clear temporal dynamic is observed from which we extract the spin relaxation time.

Quantum Dot Molecules

In the second part of the talk, our focus will shift to recent investigations of isolated QD-molecules (QDMs), consisting of two layers of vertically aligned QDs separated by a 10nm thick GaAs spacer. The QDMs are embedded at the centre of the intrinsic region of an n-type Schottky photodiode that enables us to tune the static electric field (*F*) oriented along the QDM growth axis via the potential applied to the Schottky gate. Electric field dependent photoluminescence was performed on single QDMs at low excitation powers in the single exciton regime. Two distinct exciton branches are resolved which show a clear anti-crossing as *F* is tuned.[2] Quantitative comparison of these observations with theory shows that the two states involved have spatially *direct* and *indirect* character, respectively, and can be field tuned into resonance due to their strongly dissimilar DC Stark shifts. At the resonance the electron component of the exciton wavefunction hybridizes, giving rise to a quantum coupling energy in the excitonic spectrum of $\Delta E \sim 2\text{-}3$meV, in good accord with the predictions of our calculations. The nature of the ground state in the QDM changes from direct to indirect character as the resonance is traversed. Quenching of the anti-bonding state with decreasing temperature indicates efficient population relaxation to the molecular ground state over the timescale of the excitonic radiative lifetime. Measurements as a function of excitation power enable us to identify few-exciton states in the QD-molecules. Similar anti-crossings are detected in the biexciton emission spectrum, demonstrating the potential to manipulate the coherent quantum coupling between few exciton states using an externally applied gate potential.

[1] M. Kroutvar, Y. Ducommun, D. Heiss, D. Schuh, M. Bichler, G. Abstreiter and J. J. Finley. Nature **432**, (2004)

[2] H. J. Krenner, E. C. Clark, A. Kress, D. Schuh, M. Bichler, G. Abstreiter and J. J. Finley. PRL **94**, 057402 (2005)