Spin and reoccupation noise beyond the fluctuation-dissipation theorem

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InGaAs quantum dots (QDs) hosting a single hole-spin are particularly promising solid-state candidates for spin-photon devices in view of quantum information processing [1]. They provide very long coherence times and a large spin-photon coupling strength, which can be further enhanced by embedding the QDs in a Bragg mirror cavity, enabling efficient spin detection and manipulation [2]. We investigate the spin and charge dynamics in such a coupled QD microcavity system under strong driving by a quasi-resonant light field addressing the relevant optical transition for spin-photon interfaces.

The spin dynamics of semiconductor systems can be optimally studied by spin noise (SN) spectroscopy, a quantum optical method that has been very successfully transferred to semiconductor physics during the last decade [3]. The fundamental principle of this method is to map stochastic spin fluctuations - referred to as spin noise - onto the polarization of a non-resonant weakly interacting probe laser via Faraday or Kerr rotation. The fluctuation-dissipation theorem states that the full dynamics of the spin system can then be derived from these fluctuations measured in thermal equilibrium. Driving the spin system by a strong resonant light field creates a highly non-equilibrium situation that requires a special theoretical analysis beyond the fluctuation-dissipation theorem. As will be shown below, the non-equilibrium SN spectroscopy is capable to deliver information not only about the spin dynamics in the ground and the excited state, but also about charge dynamics in the strongly driven QD.

The measurement setup (Fig. 1(a)) is a home-built low-temperature confocal microscope that enables photoluminescence (PL) characterization and SN spectroscopy on a specific QD via the same optical path. Figure 1(b) shows a typical PL spectrum of a single QD with the optical transition of the exciton (X^0) and the positively charged trion (X^{1+}) which indicates a resident hole in the QD. The noise of the hole spin can be sensed by tuning the probe laser over the X^{1+} transition which is quantified by the measured SN power shown as purple data points in Fig. 1(b). The distinct shape of the SN power curve with two sharp maxima around the resonance is characteristic for a SN measurement on a single homogeneously broadened QD transition. This is very special for a QD in an unbiased solid-state structure, as the transitions are usually inhomogeneously broadened due to charge fluctuations in the environment which also influence the spin dynamics [4]. To study dynamics the SN is analyzed in the frequency domain. A typical SN frequency spectrum is shown in the inset of Fig. 1(b). It consists of two Lorentzian contributions with a different width, which corresponds to a correlation rate, and a different area corresponding to the SN power. These two Lorentzian noise contributions α and β are analyzed separately regarding their SN power and correlation rate as a function of laser detuning in Fig. 2(a).

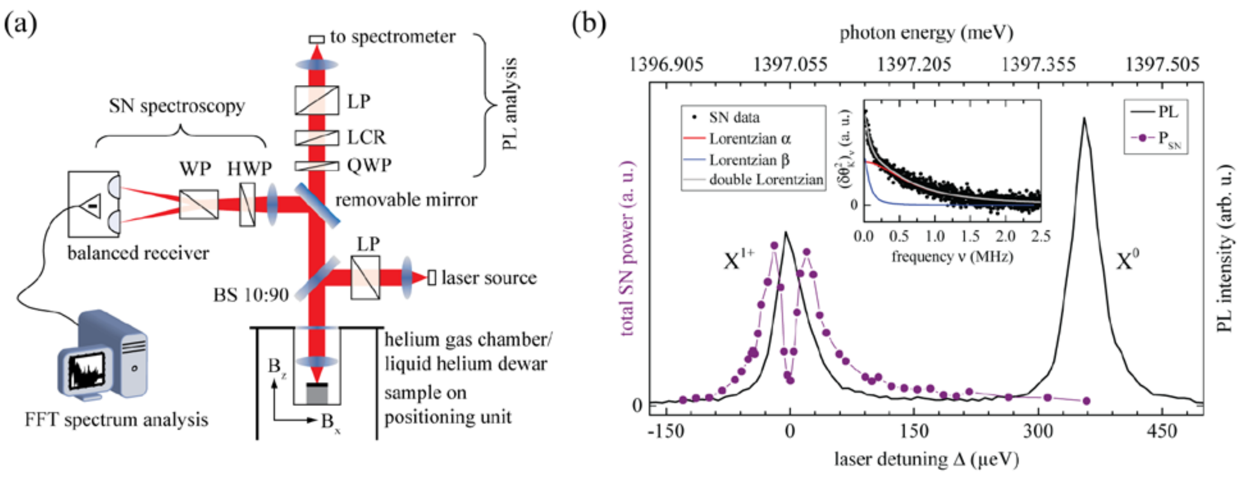

Fig. 1: (a) Experimental setup for PL analysis and SN spectroscopy on a single QD. (b) PL spectrum of a single QD showing the optical transition of the neutral exciton (X^0) and the positively charged trion (X^{1+}). The purple data points depict the SN power measured as a function of probe laser detuning with respect to the trion resonance. The inset shows a typical SN frequency spectrum revealing two Lorentzian noise contributions.

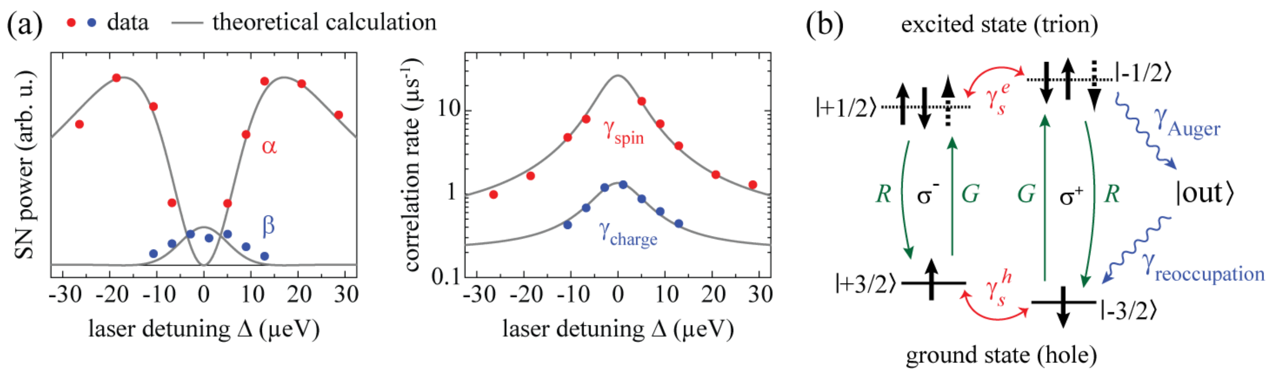

Fig. 2: (a) SN power and correlation rate of the two Lorentzian noise contributions α and β as a function of laser detuning. (b) Sketch of the QD states and the relevant transitions considered for the theoretical calculation.

The SN power spectra exhibit a differening dependence on the detuning which supports the identification of the underlying mechanism linked to the respective noise contribution by theoretical calculation. The calculations shown by the gray lines in Fig. 2(a) are in excellent agreement with the measured data. They are based on a four level system of the QD spin states as depicted in Fig. 2(b).

The SN power spectrum of the α contribution, which drops to zero at the resonance, can be assigned to the spin fluctuations. For large laser detunings, i.e. negligible optical excitation, the SN measurement yields the dynamics of the hole spin in the QD ground state with a long spin relaxation time of 2.5 µs deduced from the correlation rate. At small laser detunings absorption of the probe laser leads to generation of the trion, which is composed of a hole singlet state and an unpaired electron spin (cf. Fig. 2(b)). Hence, close to the resonance we measure the dynamics of the electron spin in the excited state which has a significantly shorter relaxation time of 30 ns. The correlation rate profile measured in Fig. 2(a) is obtained by the weighted sum of hole and electron spin relaxation rates and is proportional to the light absorption.

The second Lorentzian noise contribution β, which has a maximal SN power at the resonance, is found to belong to a different kind of noise linked to the charge state of the QD. The generation of a trion by light absorption is followed by recombination back to the QD ground state. The recombination energy is usually emitted radiatively, but it can also be transferred to the resident hole leading to the ejection of the hole from the QD (Auger process). The empty QD is reoccupied after a certain time via tunneling of a hole from an outer state in the solid-state environment (see Fig. 2(b)). The observation of this occupation noise in SN spectroscopy is possible due to a small magnetic field applied to sample which results in a finite Kerr rotation angle only in the case of an occupied QD. A magnetic field dependence of the β SN power proves the correct identification of this contribution [5]. The corresponding correlation rate yields the rate of the Auger process and increases proportional to the absorption as the probability for the Auger effect increases proportional to the population of the trion states. We extracted an Auger rate of 2.9 μs^{-1} which is in good agreement with the value found in [6] for similar QDs. For large laser detuning the correlation rate approaches the hole tunneling rate which yields an estimate of about 5 µs tunneling time for the reoccupation of the QD.

In summary, we have measured the non-equilibrium SN of a homogeneously broadened single QD in a microcavity. The measurements in combination with a theoretical analysis beyond the fluctuation-dissipation theorem reveal the spin dynamics in the ground and the excited state of the strongly driven artificial atom which is potentially useful for spin-photon interfacing. In addition, the measurements reveal a new noise contribution due to temporary escaping of the resident hole in the QD which may be parasitic for spin-photon interfacing. We expect that the reoccupation of the QD becomes slower with decreasing background doping densities leading to the conclusion that QDs in unbiased structures are disturbed by charge fluctuations at high background doping densities and by slow reoccupation at low background doping densities.

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