

## Measurement of a Heavy-Hole Hyperfine Interaction in InGaAs Quantum Dots Using Resonance Fluorescence

P. Fallahi, S. T. Yilmaz, and A. Imamoglu

*Institute of Quantum Electronics, ETH Zurich, CH-8093 Zürich, Switzerland*

(Received 1 September 2010; published 14 December 2010)

We measure the strength and the sign of hyperfine interaction of a heavy hole with nuclear spins in single self-assembled quantum dots. Our experiments utilize the locking of a quantum dot resonance to an incident laser frequency to generate nuclear spin polarization. By monitoring the resulting Overhauser shift of optical transitions that are split either by electron or exciton Zeeman energy with respect to the locked transition using resonance fluorescence, we find that the ratio of the heavy-hole and electron hyperfine interactions is  $-0.09 \pm 0.02$  in three quantum dots. Since hyperfine interactions constitute the principal decoherence source for spin qubits, we expect our results to be important for efforts aimed at using heavy-hole spins in quantum information processing.

DOI: 10.1103/PhysRevLett.105.257402

PACS numbers: 78.67.Hc, 78.20.Ls, 78.47.J-, 78.55.Cr

Theoretical and experimental studies in the past decade have established that hyperfine interaction with the quantum dot (QD) nuclei constitutes the principal decoherence mechanism for electron spin qubits [1–6]. An interesting strategy to circumvent leakage of quantum information to the nuclear spin environment is to represent quantum information with the pseudospin of a QD heavy hole (HH) [7–9]: since HH states are formed mainly out of bonding  $p$  orbitals of the lattice atoms, it had been argued that the HH hyperfine interaction is negligible. Recently, it was shown theoretically that the hyperfine interaction of a HH with the nuclear spins in a QD, while being Ising-like, could in fact be comparable in strength to that of the electron [10,11]. While prior experiments have addressed hole-nuclei coupling in an ensemble of QDs [12,13], high resolution measurements of HH hyperfine interaction in single optically active QDs have remained unexplored.

In this Letter, we present resonance fluorescence (RF) measurements that directly reveal the relative strength of the HH hyperfine interaction in single-electron charged InGaAs QDs. To this end, we generate a precise amount of nuclear spin polarization in an external magnetic field by dragging the higher energy (blue) Zeeman shifted charged exciton (trion) transition using a nonperturbative laser field [14]. We then measure the resulting Overhauser shift of the QD transitions that are shifted either by the Zeeman energy of the exciton (the red trion transition) or the electron (the forbidden-diagonal transition) with respect to the blue trion resonance. The nuclear spin polarization induced energy shifts in these transitions are determined by the difference and the sum of the Overhauser field seen by the electron and the HH, respectively. Measuring the nuclear spin polarization induced shift of these two transitions allows us to directly determine the ratio of the HH to electron Overhauser shift to be  $\eta = -0.09 \pm 0.02$  in two QDs and  $\eta = -0.10 \pm 0.05$  in a third QD.

Our sample consists of charge-tunable InGaAs self-assembled QDs embedded in a Schottky-diode structure. Experiments are carried out at 4.2 K with an external field in the Faraday geometry; the applied gate voltage range ensures that the QD is single-electron charged. Figure 1 inset shows the relevant energy levels in this regime where the QD optical transitions couple the spin-up  $|\uparrow\rangle$  (spin-down  $|\downarrow\rangle$ ) ground state to an optically excited blue (red) trion state  $|\uparrow\uparrow\rangle$  ( $|\downarrow\downarrow\rangle$ ). The two ground and excited states are split by the Zeeman energies  $|g_e|\mu_B B$  and  $|g_h|\mu_B B$ . Because of heavy-light-hole mixing, the  $|\uparrow\rangle$  ( $|\downarrow\rangle$ ) state also couples to the other trion state  $|\uparrow\downarrow\rangle$  ( $|\downarrow\uparrow\rangle$ ), with an  $\sim 10^{-3}$  times smaller oscillator strength [15,16]; we refer to these as diagonal transitions.

Our experiments combine two recent advances in experimental QD spin physics: namely, the high-efficiency detection of RF [15,17,18] and the possibility to lock a QD resonance to an incident laser frequency via nuclear spin polarization [14]. Figure 1(a) shows the gate-voltage dependent RF signal from the blue trion transition at  $B = 4T$  as the pump laser is scanned from an initial blue detuning ( $\omega_p > \omega_{t-b}^0$ ) to a final red detuning ( $\omega_p < \omega_{t-b}^0$ ); here  $\omega_{t-b}^0$  is the trion resonance frequency in the absence of nuclear spin polarization and  $\omega_p$  is the pump laser frequency. Reflected photons from the linearly polarized excitation laser are blocked by polarization suppression [15]. The RF signal is strong at the edges of the plateau where cotunneling rate  $\kappa_{\uparrow\downarrow}$  is large, and disappears in the middle of the plateau due to spin pumping [19,20]. The characteristic extension of the peaks in the direction of the laser scan is due to dynamic nuclear spin polarization, i.e., the *resonance dragging* effect [14]. Experiments are performed at the gate voltage indicated by the dashed line where the dragging range is close to maximum. At this gate voltage the RF contrast is reduced  $\sim 4$  times due to spin pumping. Figure 1(b) shows laser scans obtained at a gate voltage denoted by the dashed line in Fig. 1(a) for two opposite scan

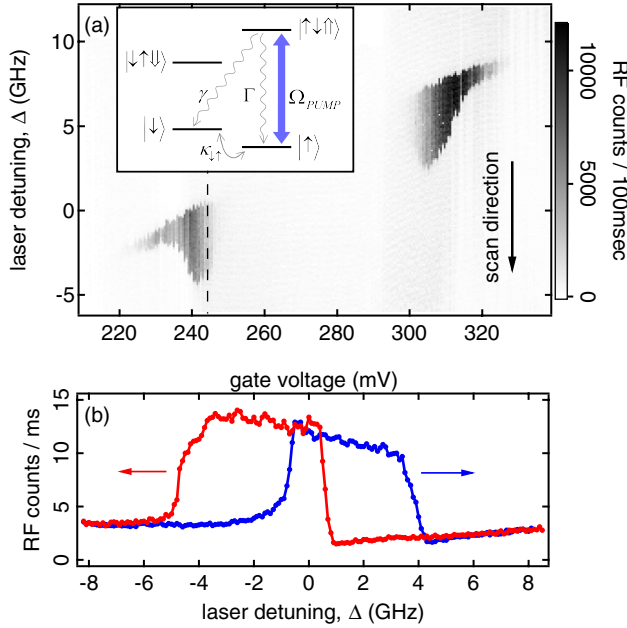


FIG. 1 (color online). (a) Resonance fluorescence (RF) signal from the blue trion transition versus gate voltage and pump laser detuning  $\Delta$  at  $B = 4T$  and  $P = P_{\text{sat}}/2$ . Remainder of the experiments are performed at the gate voltage indicated by the dashed line, where the signal is reduced  $\sim 4$  times due to spin pumping and a large line broadening due to dynamic nuclear spin polarization is observed. Inset: Energy level diagram for a quantum dot charged with a single electron. (b) Cross section of (a) across the dashed line, opposite scan directions indicated by the arrows. A total dragging range of  $\sim 8$  GHz is observed. Interference with the laser background is partly responsible for the change of RF counts along the dragging range.

directions: a total dragging range of  $\sim 8$  GHz is observed. We define the middle point between the onset of forward and backward dragging as  $\omega_p - \omega_{t-b}^0 = \Delta = 0$ . All other frequencies are measured relative to this point. In the remainder of the measurements we use the detuning  $\Delta$  of the pump laser that is slowly scanned across the blue trion transition, in either forward or backward direction, as a knob for adjusting the amount of nuclear spin polarization in the QD.

Since the electrons in the relevant optically excited states form a singlet, the nuclear-spin-polarization-induced change in the trion Zeeman splitting is given exclusively by the Overhauser shift of a single HH. To measure this HH Overhauser shift, we tune a strong pump laser to create a precise amount of nuclear spin polarization by dragging the blue trion transition from  $\omega_{t-b}^0$  to  $\omega_{t-b}^0 + \Delta$ . The pump laser which remains on resonance throughout the dragging range, at the same time leads to a substantial electron spin pumping into the  $|\downarrow\rangle$  state, causing a reduction in the strength of the RF signal. Subsequently, we scan a weak probe laser across the red trion and the diagonal resonances; once the probe laser is on resonance with either the red trion [Fig. 2(a)] or diagonal [Fig. 2(b)] [21] transition, it pumps the electron spin back to the  $|\uparrow\rangle$  state,

leading to a partial recovery of the RF signal. The top traces in Fig. 2(c) show the enhancement of the RF signal when the probe laser is on resonance with the red trion (left) or the diagonal (right) transitions, when the pump laser frequency was fixed at  $\Delta = 0.5$  GHz. When we scan the pump laser to a final detuning of  $\Delta = -2.5$  GHz, the resulting nuclear spin polarization modifies both the red trion and the diagonal transition resonance frequencies; the change in the red trion (diagonal) resonance is given by  $[-\delta E_e(\Delta) + \delta E_{\text{HH}}(\Delta)]/2$   $[[-\delta E_e(\Delta) - \delta E_{\text{HH}}(\Delta)]/2]$ , where  $\delta E_{\text{HH}}(\Delta)$  [ $\delta E_e(\Delta)$ ] denotes the  $\Delta$ -dependent Overhauser shift seen by a single QD HH (electron), i.e., the change in the Zeeman splitting of the HH (electron) in presence of nuclear spin polarization. Therefore, by measuring the shift in the corresponding resonances using the probe laser [Fig. 2(c), bottom trace], we determine the ratio  $\eta(\Delta) = \delta E_{\text{HH}}(\Delta)/\delta E_e(\Delta)$  of the HH and electron Overhauser shifts to be  $-0.09$ . This reveals that the HH and the electron hyperfine interactions have opposite signs.

The shift in the red trion and the diagonal transition frequencies are extracted by fitting a Lorentzian line shape to the resonantly enhanced RF signal [Fig. 2(c)]. To

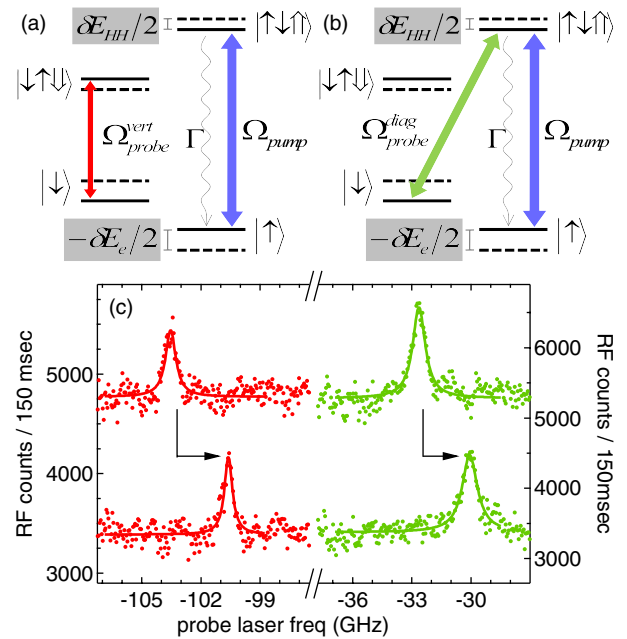


FIG. 2 (color online). (a),(b) Energy level diagrams showing the pump and probe lasers. The probe laser repumps the spin into the  $|\uparrow\rangle$  state by driving the red trion (a) or the diagonal (b) transition. Dashed (solid) lines indicate the ground and excited energy levels before (after) the polarization of nuclei by the pump laser.  $\delta E_{\text{HH}}$  ( $\delta E_e$ ) denotes the Overhauser shift seen by a single QD HH (electron). (c) RF signal recorded as the probe laser is scanned across the diagonal (right) or the red trion (left) transitions. An arbitrary offset is added to the top two scans for clarity. Prior to the probe laser scan the pump laser is scanned across the blue trion transition and stopped at a detuning of  $\Delta = 0.5$  GHz (top) or  $\Delta = -2.5$  GHz (bottom). Solid lines are Lorentzian fits. Peak positions are shifted due to nuclear spin polarization induced by the pump laser.

confirm that the ratio  $\eta(\Delta)$  is in fact independent of the amount of nuclear spin polarization, we repeat the experiment for a range of different  $\Delta$  values. Figure 3(a) shows a series of probe scans across the diagonal transition for different pump laser detunings. The extracted transition energies are plotted in Fig. 3(b) (green dots). The red dots in Fig. 3(b) show the shift of the red trion transition measured using the same technique. Dashed lines are linear fits to the extracted nuclear-spin-polarization-modified resonance frequencies.

The remarkable linear fit to the data in Fig. 3(b) not only shows that  $\eta(\Delta) = \eta$  as we anticipated, but also that both  $\delta E_{\text{HH}}$  and  $\delta E_e$  scale linearly with the pump laser detuning. This linear dependence allows us to simply determine the relative strength of the HH hyperfine interaction as  $\eta = -(\alpha - \beta)/(\alpha + \beta)$ , where  $\alpha$  ( $\beta$ ) is the slope of the red trion (diagonal) transition depicted in Fig. 3(b). Using the measured slope of  $-0.98 \pm 0.02$  for the red line and  $-0.82 \pm 0.02$  for the green line, we calculate  $\eta = -0.09 \pm 0.02$ .

An accurate determination of  $\eta$  requires that scanning the probe laser across the red trion or the diagonal transition does not modify the nuclear spin polarization that was created by the pump laser. Such a modification would occur if the probe laser itself caused dragging. If this were the

case, the pump laser would no longer be resonant with the blue trion transition, leading to a decrease in the (pump laser induced) RF signal that constitutes the background of the scans shown in Fig. 2(c) [22]. We choose the probe laser power for both the diagonal and the red trion transition such that there is no measurable change in the background as the probe laser is scanned across that transition. In addition, the linewidths of the recovered RF signal match the QD transition linewidth at  $B = 0T$ , in agreement with the absence of probe induced dragging. Furthermore, we repeated the measurement at various probe laser powers; only when the laser power is low enough does the slope become completely independent of laser power. Figure 4(a) shows the Overhauser shifts of the vertical and diagonal transitions, each for two different probe laser powers indicated on the graph. The measured slopes at different powers are indeed in excellent agreement. The oscillator strength of the diagonal transition is much weaker than that of the red trion transition, requiring higher probe laser powers to induce resonant spin pumping. By repeating the diagonal transition Overhauser shift measurements for two different gate voltages, 1 mV apart, around the gate voltage indicated by the dashed line on Fig. 1(b) [blue traces in Fig. 4(a)], we have also confirmed that the slopes do not depend on the gate voltage. The latter measurements also suggest that small changes in the charging environment and the resulting changes in the confined electron and HH wave functions do not alter the ratio  $\eta$ . The ratio  $\eta$  also shows no appreciable dependence on the strength of the external magnetic field [Fig. 4(a) inset].

Because of the large variation in HH  $g$  factor and the positively charged trion confinement energy, it is generally believed that the confined HH wave function could change substantially from one QD to another. To determine if these changes lead to a modification of HH hyperfine interaction, we have repeated our experiments on two other QDs. Figure 4(b) shows measurements on a second QD which yields  $\eta = -0.09 \pm 0.02$ . The ratio for a third QD, determined with a factor 2.5 lower accuracy, yielded  $\eta \sim -0.1$ . Remarkably, we find that the strength of the HH hyperfine interaction in these 3 QDs is almost identical, even though their HH longitudinal  $g$  factors vary substantially [Fig. 4(b)].

Despite the accurate measurement of the Overhauser shift of the HH and the electron that we have demonstrated, it is not straightforward to use our data to extract the actual HH hyperfine interaction constant with high accuracy due to differences in the confined electron and the HH envelope wave functions. The exact mechanism behind dragging of QD resonances is not well understood; however, it is safe to assume that the underlying nuclear spin polarization is mainly mediated by the electron [23]. The precise magnitude of the HH Overhauser shift is therefore influenced by the overlap between the electron and hole wave functions and their confinement length scales. Repeating the experiments on different QDs, with vastly different in-plane  $g$  factors, would yield further information about the sensitivity of  $\eta$  to the HH confinement.

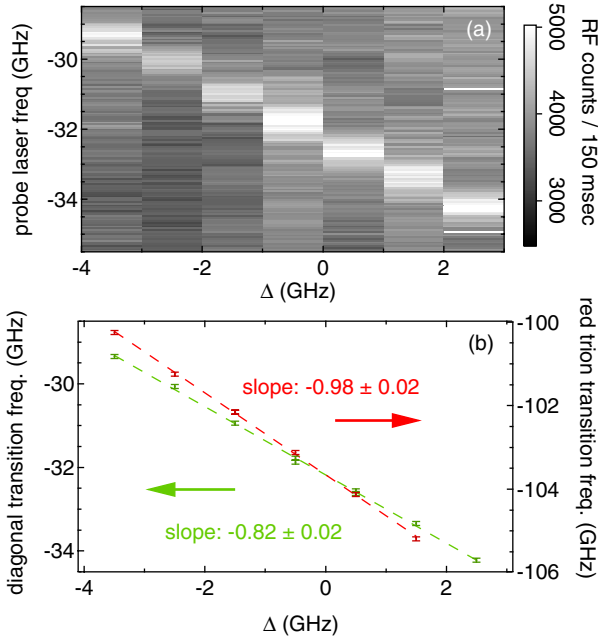


FIG. 3 (color online). (a) RF signal as a function of probe laser frequency scanned across the diagonal transition and pump laser detuning  $\Delta$ . The position of the peak corresponding to recovered RF counts shifts due to nuclear spin polarization induced by the pump laser. (b) (green points) The position of the peaks in (a) extracted from Lorentzian fits to the data. Red points correspond to peak positions extracted from similar scans where the probe laser is scanned across the vertical transition. Dashed lines are linear fits with slopes indicated in the figure. Arrows point to the relevant axis.

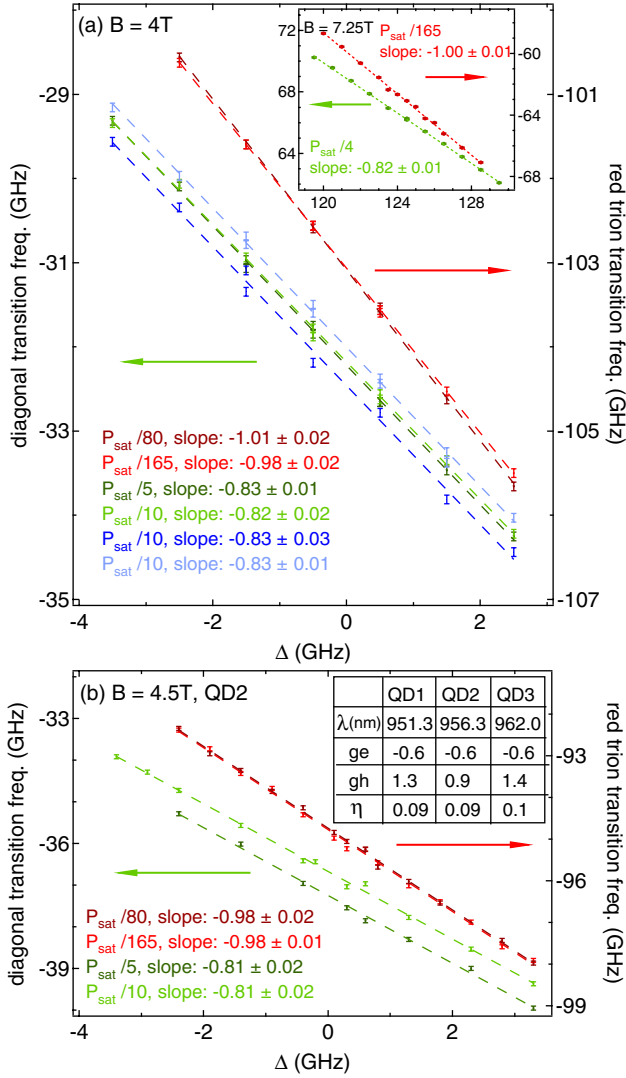


FIG. 4 (color online). (a) Transition energies of the red trion (red points) and the diagonal transition (green and blue points) versus pump laser detuning  $\Delta$  at  $B = 4T$ . Probe laser power and slopes of linear fits (dashed lines) are indicated in the figure. The dark and light blue points are measured with the same laser power at two different gate voltages, 1 mV apart, around the gate voltage indicated by the dashed line in Fig. 1(a). The dark and light green and red points are measured at two different laser powers at the gate voltage indicated by the dashed line in Fig. 1(a). The slopes do not change with changing probe laser power or gate voltage. Inset shows measurements at  $B = 7.25T$ : the axes are the same as those in the main figure. (b) Transition energies of the red trion (red points) and the diagonal transition (green points) versus  $\Delta$ , measured on a second quantum dot. The vertical shift between the two green lines is due to a shift in the gate voltage, caused by fluctuations in the quantum dot charge environment. The box compares the emission wavelength, electron (ge) and heavy-hole (gh)  $g$  factors and  $\eta$  for 3 different quantum dots.

The striking feature of our experiments is the (almost) perfect linear dependence of the Overhauser shift on the pump laser detuning. What is more remarkable is the fact that the slope of the red trion energy shift is  $\sim -1$ ,

indicating that the Overhauser shift of the blue trion transition satisfies  $[-\delta E_e(\Delta) + \delta E_{HH}(\Delta)]/2 = -\Delta + c$ , where  $c$  is a constant much smaller than the bare optical transition linewidth. However, theoretical models of dragging suggest that the amount of nuclear spin polarization has a nontrivial dependence on the laser detuning. Our experiments could help in identifying the mechanisms underlying the dragging of QD resonances [14,24].

In summary, we have developed a new measurement technique combining two recent advances in QD physics to determine the strength of HH hyperfine interaction. Our measurements on highly strained self-assembled QDs indicate a coupling strength that is  $\sim 2$  smaller than the theoretical prediction for strain-free GaAs QDs [10] and provide further support for efforts aimed at using confined HH spins as qubits in quantum information processing.

We thank Antonio Badolato for sample growth and Martin Kroner for useful discussions. S.T.Y. acknowledges financial support from the EU within the Marie-Curie Training Research Network EMALI. This work is supported by NCCR Quantum Photonics, research instrument of the Swiss National Science Foundation.

*Note added.*—After completion of this work, we became aware of related experiments by Chekhovich *et al.* [25].

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