Direct Measurement of the Hole-Nuclear Spin Interaction in Single InP/GaInP Quantum Dots Using Photoluminescence Spectroscopy

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We measure the hyperfine interaction of the valence band hole with nuclear spins in single InP/GaInP semiconductor quantum dots. Detection of photoluminescence (PL) of both "bright" and "dark" excitons enables direct measurement of the Overhauser shift of states with the same electron but opposite hole spin projections. We find that the hole hyperfine constant is $\approx 11\%$ of that of the electron and has the opposite sign. By measuring the degree of circular polarization of the PL, an upper limit to the contribution of the heavy-light hole mixing to the measured value of the hole hyperfine constant is deduced. Our results imply that environment-independent hole spins are not realizable in III-V semiconductor, a result important for solid-state quantum information processing using hole spin qubits.

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The spin of a conduction band electron confined in a semiconductor quantum dot (QD) has been actively investigated for realization of a solid-state quantum bit (qubit) [1]. However, the hyperfine interaction with fluctuating nuclear spins leads to fast decoherence of the electron spin on the nanosecond scale [2,3]. For that reason, recently valence band holes have been considered as an attractive alternative. Unlike electrons having s-type atomic wave functions, the hole wave function is constructed from p orbitals with zero density at the nuclear site, leading to vanishing contact hyperfine interaction. This has led to the widely accepted conclusion that the spin of a localized hole is well isolated from its environment. In agreement with this expectation, slow hole spin relaxation up to 1 ms [4,5] and long-lived spin coherence up to 1 μs [6] have been demonstrated for InGaAs dots. However, recent theoretical studies have predicted that the hole hyperfine interaction can be as large as 10% of that of the electron [7,8]. Experimental evidence for nonvanishing hole-nuclear coupling has been demonstrated by Eble *et al.* [9] who used it to explain the hole spin dynamics in an ensemble of p-doped dots. Despite close similarities to the system studied in Ref. [6], a much shorter hole spin dephasing time of 14 ns has been reported in Ref. [9]. To remove this discrepancy and enable well-founded understanding of hole spin coherence effects, a direct measurement of the hole-nuclear interaction is required: this is provided by the present work.

In this work we directly measure hole Overhauser shifts in individual self-assembled InP/GaInP dots, which allows us to deduce the magnitude and sign of the hole hyperfine interaction constant. We use nonresonant laser excitation and a pump-probe method to achieve two crucial ingredients of this measurement: (i) nuclear spin polarization on the dot variable in a wide range by altering the polarization of the high power pump and (ii) detection of both "bright" and "dark" excitons in photoluminescence (PL) excited by a low intensity probe. This technique enables the measurement of energy shifts of the four excitonic states with all possible electron and heavy-hole spin projections at different magnitudes of optically induced nuclear spin polarization [10,11]. This allows simultaneous detection of the electron and hole Overhauser shifts, and as a result the ratio of the hyperfine constants of the heavy-hole (C) and the electron (A) can be measured. We find that, in good agreement with recent calculations for the hole-nuclear spin coupling induced by the dipole-dipole interaction [8], this ratio is negative and on average $C/A \approx -0.11$.

Our observations show that environment-independent hole spins are not realizable in III-V semiconductors. Robust QD-based hole spin qubits are only likely to be achievable in dots with negligible heavy-light hole mixing. For such dots hole-nuclear coupling has an Ising form with no spin flip-flops allowed [8], in contrast to the non-Ising hyperfine coupling in the case of nonzero mixing, which results in strong shortening of the hole spin coherence [6,7,9]. On the other hand, we find that when nuclear spins are polarized, QD holes can experience effective nuclear magnetic fields on the order of 100 mT, which needs to be taken into account when interpreting observations related to hole spin coherence [6,9].

Our experiments were performed on an undoped InP/GaInP QD sample without electric gates [11–14]. PL of neutral QDs was measured at $T = 4.2$ K, in external magnetic field B_z normal to the sample surface. QD PL at $\sim$1.84 eV was excited nonresonantly with a laser at $E_{\mathrm{exc}} = 1.88$ eV below the GaInP barrier band gap and analyzed with a 1 m double spectrometer and a CCD.

In a neutral dot electrons $\uparrow$ ($\downarrow$) with spin $s_z^e = \pm 1/2$ and heavy holes $\Uparrow$ ($\Downarrow$) with momentum $j_z^h = \pm 3/2$ parallel (antiparallel) to the growth axis Oz can form either optically forbidden ("dark") excitons $|\Uparrow\uparrow\rangle$ ($|\Downarrow\downarrow\rangle$) with spin projection $J_z = +2(-2)$ or "bright" excitons $|\Uparrow\downarrow\rangle$ ($|\Downarrow\uparrow\rangle$) with $J_z = +1(-1)$ optically allowed in $\sigma^+(\sigma^-)$ polarization. QD axis misorientation or symmetry reduction leads to weak mixing of bright and dark states: as a result, the latter are observed in PL [13,15]. This is demonstrated in Fig. 1(a) where PL spectra of QD1 measured at low excitation power $P_{\rm exc} = 200$ nW in magnetic field $B_z = 6$ T are shown for different magnitudes of nuclear spin polarization $\langle I_z\rangle$ (explained below). The dependence of PL energies of all 4 exciton states measured at different fields B_z is shown by the symbols in Fig. 1(b). Their fitting shown by the full lines allows the electron and hole g factors to be determined: $g_z^e = 1.65$, $g_z^h = 2.7$, respectively, in QD1 (see appendix in Ref. [13] for more details on QD characterization).

Nonzero average nuclear spin polarization $\langle I_z\rangle$ along the Oz axis acts as an additional magnetic field on the electron and hole spins. It is convenient to introduce the hole pseudospin $S_z^h = \pm 1/2$ corresponding to the $\Uparrow$ ($\Downarrow$) heavy-hole state. Coupling of the electron to the nuclei is described by the hyperfine constant A, whereas for the heavy hole the dipole-dipole interaction with nuclei [7,8] is described using a constant C expressed in terms of the normalized heavy-hole hyperfine constant γ as $C = \gamma A$. The expression for the exciton energy taking into account the shift due to nonzero average nuclear spin polarization can be written as

$$E[S_z^h, s_z^e] = E^{\rm QD} + E^0[S_z^h, s_z^e] + (s_z^e + \gamma S_z^h)A\langle I_z\rangle, \quad (1)$$

where the quantum dot band gap $E^{\rm QD}$ and shift $E^0[S_z^h, s_z^e]$ determined by the Zeeman and exchange energy [15] do

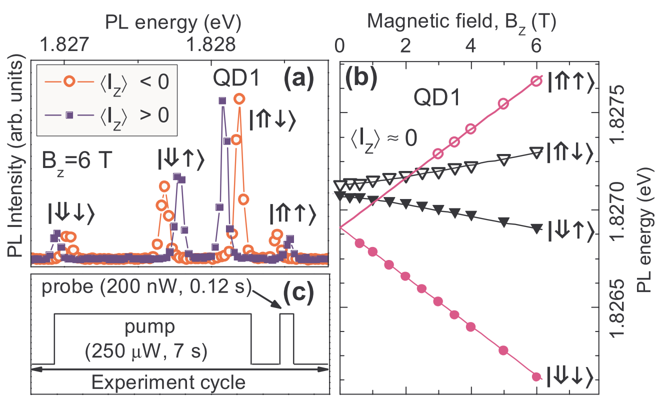

FIG. 1 (color online). (a) Exciton PL spectra in a neutral quantum dot at $B_z = 6.0$ T. Heavy holes $\Uparrow$ ($\Downarrow$) and electrons $\uparrow$ ($\downarrow$) with spin parallel (antiparallel) to the external field form optically allowed ($|\Uparrow\downarrow\rangle$, $|\Downarrow\uparrow\rangle$) and dark ($|\Uparrow\uparrow\rangle$, $|\Downarrow\downarrow\rangle$) excitons. The presented spectra correspond to different magnitudes of nuclear spin polarization $\langle I_z\rangle < 0$ (○) and $\langle I_z\rangle > 0$ (■). (b) Magnetic field dependence of exciton PL energies (symbols) at zero nuclear polarization $\langle I_z\rangle \approx 0$. A diamagnetic shift $\kappa = 5.8\ \mu$eV/T^2 is subtracted for clarity. Lines show fitting. (c) Timing diagram of the pump-probe experiment cycle. Nuclear spin $\langle I_z\rangle$ is initialized by a high power pump laser pulse, while a low power probe pulse is used to measure PL of both bright and dark excitons [as in (a)].

not depend on nuclear polarization. We note that Eq. (1) is strictly valid only for "pure" electron and heavy-hole spin states with possible deviations arising mainly from heavy-light hole mixing and leading to renormalization of γ (to be discussed in detail below). For description of the experimental results we will use the parameter γ^* in order to distinguish the hyperfine constant observed experimentally from the pure heavy-hole hyperfine constant γ.

Since mixing of dark and bright excitonic states is weak, the oscillator strength of the dark states is small, leading to their saturation at high powers. As a result, all four exciton states can be observed in PL only at low excitation power $P_{\rm exc} \lesssim 200$ nW. However, at this low power, optically induced nuclear spin polarization is small and weakly depends on polarization of photoexcitation [11], and thus the shifts of the hole spin states due to the interaction with nuclei cannot be measured accurately. To avoid this problem, we use a pump-probe technique [14] with the experiment cycle shown in Fig. 1(c). Nuclear spin polarization is prepared with a long (7 s) high power $P_{\rm exc} = 250\ \mu$W pump pulse. Following this, the sample is excited with a low power $P_{\rm exc} = 200$ nW probe pulse, during which the PL spectrum is measured. The duration of this pulse is short enough (0.12 s) to avoid the effect of excitation on nuclear polarization. This cycle is repeated several times to increase signal to noise ratio in PL spectra.

The direct and simultaneous measurement of the hole and electron energy shifts due to the hyperfine interaction is carried out by detecting the probe spectra recorded at different magnitudes of $\langle I_z\rangle$ prepared by the pump. For this, the linearly polarized pump laser first passes through a half-wave plate followed by a quarter-wave plate. In order to change $\langle I_z\rangle$, the half-wave plate is rotated to a new angle θ, leading to a change in the polarization of the pump, in turn producing a change in spin polarization of the photoexcited electrons in the dot. For each θ, $\langle I_z\rangle$ reaches the steady-state value proportional to the electron spin polarization. As a result $\langle I_z\rangle$ changes periodically as a function of θ. This is demonstrated in Fig. 1(a) where the probe spectra measured for σ^+ ($\langle I_z\rangle < 0$) and σ^- ($\langle I_z\rangle > 0$) polarized pump are shown: as expected when $\langle I_z\rangle$ changes, the exciton states with electron spin $\uparrow$ and $\downarrow$ shift in opposite directions.

As follows from Eq. (1), changes in nuclear polarization $\langle I_z\rangle$ will result in (i) changes in the energy splitting between $|\Downarrow\uparrow\rangle$ and $|\Downarrow\downarrow\rangle$ states $\Delta E[\Downarrow\uparrow, \Downarrow\downarrow] = E[\Downarrow\uparrow] - E[\Downarrow\downarrow] \propto A\langle I_z\rangle$ determined only by the electron-nuclear spin interaction and (ii) modification in the energy splitting between $|\Uparrow\uparrow\rangle$ and $|\Downarrow\uparrow\rangle$ states $\Delta E[\Uparrow\uparrow, \Downarrow\uparrow] = E[\Uparrow\uparrow] - E[\Downarrow\uparrow] \propto \gamma A\langle I_z\rangle = C\langle I_z\rangle$ due to the hole-nuclear spin interaction. The dependence of these two splittings on the angle of the half-wave plate θ (and consequently on the value of $\langle I_z\rangle$ induced by the pump) is shown in Figs. 2(a) and 2(b). It can be seen that the electron spin splitting [Fig. 2(b)] smoothly changes by almost 200 μeV when the pump polarization is varied from σ^+ to σ^-. At the same time a much weaker

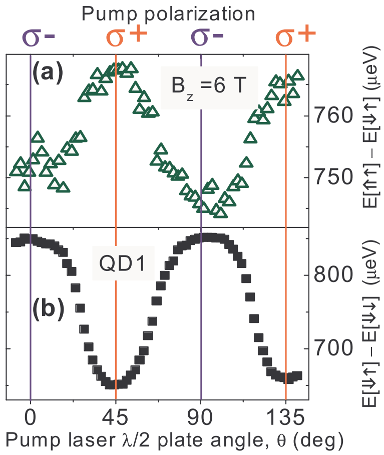

FIG. 2 (color online). Measurement of the electron- and hole-nuclear interaction in a neutral dot QD1 at $B_z = 6$ T. The angle θ of the $\lambda/2$ plate is varied to change polarization of the pump laser resulting in a change of nuclear spin polarization $\langle I_z \rangle$. Variation of the splitting between $|\Downarrow\uparrow\rangle$ and $|\Downarrow\downarrow\rangle$ excitons with θ [shown in (b)] is a result of the electron hyperfine interaction and reflects variation of $\langle I_z \rangle$ induced by the pump. The smaller change of the splitting between $|\Uparrow\uparrow\rangle$ and $|\Downarrow\uparrow\rangle$ excitons shown in (a) corresponds to variation of the hole spin splitting and is direct evidence for nonzero hole-nuclear spin interaction.

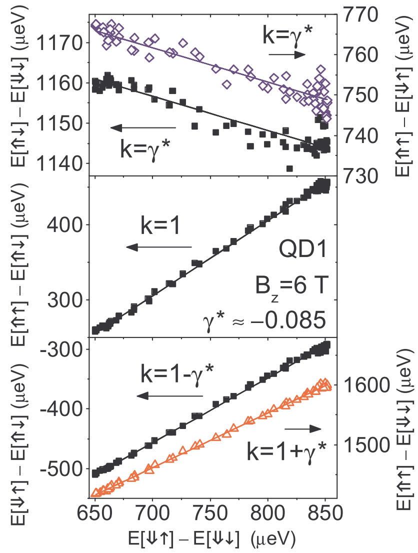

FIG. 3 (color online). Comparison of the experimental results for QD1 with the model for the electron- and hole-nuclear spin interaction Eqs. (1) and (2). Symbols show experimental dependences of splittings between different pairs of exciton states on the splitting of $|\Downarrow\uparrow\rangle$ and $|\Downarrow\downarrow\rangle$ states (i.e., the electron spin splitting). Straight lines with corresponding coefficients k from Eq. (2) show fitting with $\gamma^* \approx -0.085$.

change of the hole spin splitting in antiphase with the electron spin splitting can be seen in Fig. 2(a) providing direct evidence for nonzero hole hyperfine interaction.

For direct comparison of experiment with Eq. (1) we present the data in a slightly different way. We first note that according to Eq. (1) the energy splitting of any two states is a linear function of the splitting of any other two states. Choosing $\Delta E[\Downarrow\uparrow, \Downarrow\downarrow]$ as reference we can write for all other splittings:

$$\Delta E[\Downarrow\uparrow, \Uparrow\downarrow] \propto (1-\gamma)\Delta E[\Downarrow\uparrow, \Downarrow\downarrow]$$
$$\Delta E[\Uparrow\uparrow, \Downarrow\downarrow] \propto (1+\gamma)\Delta E[\Downarrow\uparrow, \Downarrow\downarrow]$$
$$\Delta E[\Uparrow\uparrow, \Uparrow\downarrow] \propto \Delta E[\Downarrow\uparrow, \Downarrow\downarrow] \qquad \Delta E[\Uparrow\uparrow, \Downarrow\uparrow] \propto \gamma\Delta E[\Downarrow\uparrow, \Downarrow\downarrow]$$
$$\Delta E[\Uparrow\downarrow, \Downarrow\downarrow] \propto \gamma\Delta E[\Downarrow\uparrow, \Downarrow\downarrow]. \tag{2}$$

Experimental dependences of these splittings on $\Delta E[\Downarrow\uparrow, \Downarrow\downarrow]$ are shown in Fig. 3. The solid lines show linear fitting with slope coefficients k determined by Eqs. (1) and (2). From this fitting we obtain $\gamma^* = -0.085 \pm 0.015$ for QD1. As seen, the model involving only one parameter γ^* describing the hole-nuclear spin interaction gives good agreement with the experiment: the deviation is within $\approx \pm 5$ μeV mainly determined by the accuracy of the measurement of the PL energies.

We have performed similar experiments on another 5 neutral dots from the same sample. 90% confidence probability estimates of γ^* obtained from the fitting using Eqs. (1) and (2) are given in Table I. As seen, the values of γ^* coincide within experimental error for all dots, with an average value of $\bar{\gamma}^* \approx -0.105 \pm 0.008$.

We will now discuss possible deviations from the model describing pure electron and heavy-hole states [Eq. (1)] and their consequences for the interpretation of the results presented above.

(i) Bright excitons exhibit fine structure splitting (FSS) δ_b at $B_z = 0$ and have zero electron spin projections along the Oz axis [15]. Magnetic field B_z partly restores electron spin projections: at high field $[\delta_b^2/(\mu_B g_z^e B_z)^2 \ll 1]$ they become $s_z^e \approx \pm 1/2[1 - (1/2)\delta_b^2/(\mu_B g_z^e B_z)^2]$ for $|\Uparrow\downarrow\rangle$ and $|\Downarrow\uparrow\rangle$ bright excitons. For dark excitons FSS is much smaller [13] and so $s_z^e \approx \pm 1/2$. This difference will result in violation of the model described by Eq. (1). In particular, the proportionality coefficients in Eq. (2) will deviate by $\approx (1/4)\delta_b^2/(\mu_B g_z^e B_z)^2$. However, at high magnetic field $B_z = 6$ T the largest correction for the studied dots (for QD1) is $\approx 6 \times 10^{-3}$. This is smaller than the uncertainty in measurements of γ and thus can be neglected.

(ii) Another source of electron spin projection uncertainty is mixing of the dark and bright states which we use to detect the dark excitons. The magnitude of this mixing can be estimated from the ratio of the maximum PL

TABLE I. Experimentally measured hole hyperfine constants γ^* and circular polarization degrees ρ_c of bright exciton PL for different neutral QDs at $B_z = 6$ T.

QD	γ^*	ρ_c	QD	γ^*	ρ_c
QD1	-0.085 ± 0.015	0.82	QD4	-0.117 ± 0.033	0.88
QD2	-0.110 ± 0.016	0.85	QD5	-0.111 ± 0.026	0.96
QD3	-0.106 ± 0.017	0.84	QD6	-0.117 ± 0.020	0.92

intensities of dark and bright states: the maximum intensity is proportional to the oscillator strengths which for dark states is determined by the admixture of the bright states [11]. For all dots this mixing is <0.01, negligible compared with our accuracy in determining γ [16].

(iii) Finally, mixing of heavy holes with $j_z^h = \pm 3/2$ and light holes with $j_z^h = \pm 1/2$ must be taken into account. In the simplest case it leads to hole spin states of the form $|j_z^h = \pm 3/2\rangle + \beta|j_z^h = \mp 1/2\rangle$ with $|\beta| \ll 1$ [17,18]. It has been shown that the hyperfine constant for the light-hole interaction with nuclear spins polarized along Oz is 3 times smaller than that for the heavy hole [7]. Thus in the case of mixed hole states the hole hyperfine constant will read as $\gamma^* = \frac{C}{A}\frac{1-\beta^2/3}{1+\beta^2} = \gamma\frac{1-\beta^2/3}{1+\beta^2}$. The mixing parameter β can be estimated from the circular polarization degree of PL resulting from recombination of $|j_z^h = \pm 3/2\rangle + \beta|j_z^h = \mp 1/2\rangle$ hole: $\rho_c = (I^{\sigma^\pm} - I^{\sigma^\mp})/(I^{\sigma^+} + I^{\sigma^-})$, where $I^{\sigma^\pm}$ is the PL intensity in $\sigma^\pm$ polarizations. In terms of β, $\rho_c = (1 - \beta^2/3)/(1 + \beta^2/3)$ with $\rho_c = 1$ for pure heavy holes [17]. Thus the corrected value of γ for heavy-hole states is expressed as

$$\gamma = \gamma^*(2 - \rho_c)/\rho_c, \tag{3}$$

where γ^* is the value measured experimentally ($\gamma^* < \gamma$). ρ_c values measured for the studied quantum dots at $B_z = 6$ T (averaged for $|\Uparrow\downarrow\rangle$ and $|\Downarrow\uparrow\rangle$ bright excitons) are shown in Table I. Using Eq. (3) for each dot we find that the pure heavy-hole hyperfine interaction $\gamma > -0.145$ with 90% confidence probability. This gives the lower limit for the magnitude of γ, as the magnitude of heavy-light hole mixing deduced from PL polarization may be overestimated due to imperfections of polarization optics and imperfect shapes of the subwavelength apertures used to select single QDs. We note that this lower limit does not differ significantly from the average value of uncorrected $\bar{\gamma}^* \approx -0.105 \pm 0.008$ for the dots that we have measured, and thus for pure heavy holes we can use an estimate $\gamma \approx \bar{\gamma}^*$.

γ is an average for interaction with P and In nuclei. However, the contribution of the spin 1/2 phosphorus nuclei into the total Overhauser shift is less than 10% [19] as the In nuclei possess spin 9/2. Since we observe nuclear polarization degree up to 50%, the contribution of the In nuclei is dominant, and as a result the estimated value of γ corresponds mainly to the hyperfine interaction with In. Using the value of the electron hyperfine constant in InP $A_{\rm In} = 47$ μeV [19] and the estimate $\gamma \approx \bar{\gamma}^*$, we obtain for the heavy-hole hyperfine constant $C_{\rm In} \approx \gamma A_{\rm In} \approx -5$ μeV. The hyperfine coupling with In nuclei in different III-V compounds (e.g., InP and InSb) is similar [19,20], and thus this estimate of $C_{\rm In}$ is applicable to the widely studied InGaAs QDs. For the InP dots it is possible to estimate the effective magnetic field corresponding to fully polarized nuclei: using experimentally measured g factors we obtain $B_{N,\max}^e \approx 2.4$ T for electrons and $B_{N,\max}^h \approx 0.16$ T for heavy holes.

In conclusion, we have employed PL spectroscopy of neutral excitons in single InP/GaInP quantum dots to measure the magnitude of the hole-nuclear spin interaction. For the spin 9/2 indium nuclei we deduce the hole hyperfine constant $C_{\rm In} \approx -5$ μeV in good agreement with theoretical predictions. By measuring the degree of circular polarization of PL, we obtain an estimate of the magnitude of heavy-light hole mixing and consequently deduce the hyperfine interaction for the pure heavy hole relative to that of the electron as $-0.15 \lesssim \gamma \lesssim -0.10$. We conclude that the hole spins in semiconductor quantum dots are sensitive to the presence of the nuclear spin bath. According to recent theoretical studies this suggests that only structures with negligible heavy-light hole mixing may be suitable for realization of long coherence hole spin qubits.

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Note added.—After completion of this work, we became aware of related experiments by Fallahi *et al.* [21].

[1] D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
[2] A. V. Khaetskii, D. Loss, and L. Glazman, Phys. Rev. Lett. **88**, 186802 (2002).
[3] R. Hanson *et al.*, Rev. Mod. Phys. **79**, 1217 (2007).
[4] B. D. Gerardot *et al.*, Nature (London) **451**, 441 (2008).
[5] D. Heiss *et al.*, Phys. Rev. B **76**, 241306 (2007).
[6] D. Brunner *et al.*, Science **325**, 70 (2009).
[7] C. Testelin, F. Bernardot, B. Eble, and M. Chamarro, Phys. Rev. B **79**, 195440 (2009).
[8] J. Fischer, W. A. Coish, D. V. Bulaev, and D. Loss, Phys. Rev. B **78**, 155329 (2008).
[9] B. Eble *et al.*, Phys. Rev. Lett. **102**, 146601 (2009).
[10] D. Gammon *et al.*, Phys. Rev. Lett. **86**, 5176 (2001).
[11] E. A. Chekhovich *et al.*, arXiv:1007.1241.
[12] J. Skiba-Szymanska *et al.*, Phys. Rev. B **77**, 165338 (2008).
[13] E. A. Chekhovich *et al.*, Phys. Rev. B **81**, 245308 (2010).
[14] E. A. Chekhovich *et al.*, Phys. Rev. Lett. **104**, 066804 (2010).
[15] M. Bayer *et al.*, Phys. Rev. B **65**, 195315 (2002).
[16] In order to further verify this point, we measured γ for two of the studied dots with the sample growth axis tilted by $\theta = 8°$ from the field direction. We found a significant increase of the dark-bright exciton mixing up to ≈ 0.05. However, at $\theta = 8°$ we found $\gamma = -0.091 \pm 0.004$ for QD1 and $\gamma = -0.116 \pm 0.014$ for QD2 in good agreement with $\gamma = -0.085 \pm 0.015$ and $\gamma = -0.110 \pm 0.016$, respectively, measured without the tilt.
[17] Y. Leger *et al.*, Phys. Rev. B **76**, 045331 (2007).
[18] D. N. Krizhanovskii *et al.*, Phys. Rev. B **72**, 161312(R) (2005).
[19] B. Gotschy *et al.*, Solid State Commun. **71**, 629 (1989).
[20] M. Gueron, Phys. Rev. **135**, A200 (1964).
[21] P. Fallahi *et al.*, Phys. Rev. Lett. **105**, 257402 (2010).