Knight Field Enabled Nuclear Spin Polarization in Single Quantum Dots

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We demonstrate dynamical nuclear spin polarization in the absence of an external magnetic field, by resonant circularly polarized optical excitation of a single electron or hole charged quantum dot. Optical pumping of the electron spin induces an effective inhomogeneous magnetic (Knight) field that determines the direction along which nuclear spins could polarize and enables nuclear-spin cooling by suppressing depolarization induced by nuclear dipole-dipole interactions. Our observations suggest a new mechanism for spin-polarization where spin exchange with an electron reservoir plays a crucial role. These experiments constitute a first step towards quantum measurement of the Overhauser field.

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Hyperfine interactions in quantum dots (QD) are qualitatively different than those in atoms: coupling of a single electron-spin to the otherwise well-isolated quantum system of nuclear spins in a QD gives rise to rich physical phenomena such as non-Markovian electron-spin decoherence \[ \alpha, \beta \]. It has also been proposed that the long-lived collective nuclear-spin excitations generated and probed by hyperfine interactions could have potential applications in quantum information processing \[ \alpha, \beta \]. Several groups have previously reported QD nuclear-spin cooling using external magnetic fields \[ \alpha, \beta \]. To achieve dynamical nuclear spin polarization (DNSP), it has generally been assumed that a small but nonzero external magnetic field is necessary.

Here, we use resonant circularly polarized optical excitation of a single electron or hole charged QD to demonstrate DNSP in the absence of an external magnetic field. We show that optical pumping of the electron spin induces an effective inhomogeneous magnetic (Knight) field that can be more than an order of magnitude larger than the characteristic nuclear dipolar fields, which in turn ensures that DNSP is not suppressed by the latter. Our experiments constitute a first step towards projective quantum measurements of the effective nuclear (Overhauser) field operator that could in turn suppress electron spin decoherence in QDs \[ \alpha, \beta \].

DNSP is investigated using single self-assembled QDs in gated structures that allow for deterministic charging of a QD \[ \alpha, \beta \] with a single excess electron or hole. The sample is grown by molecular beam epitaxy on a (001) semi-insulating GaAs substrate. The InAs quantum dots are grown 25 nm above a 40 nm heavily doped n\(^+\)-GaAs layer, followed by 30 nm GaAs and 29 periods of AlAs/GaAs (2/2 nm) superlattice barrier layer, and capped finally by 4 nm GaAs. A bias voltage is applied between the top Schottky and back ohmic contacts to control the charging state of the quantum dots. The density of quantum dots is below 0.1/\( \mu \)m\(^2\), allowing the addressing of a single quantum dot using a microphotoluminescence (\( \mu \)-PL) setup. In this letter, data based on two different QDs, labeled as QD-A and QD-B, is analyzed.

The standard \( \mu \)-PL setup is based on a combination of a solid immersion lens (Z\( r \)O\(_z\), refractive index \( n \approx 2.2 \)) in Weierstrass (or supersphere) configuration and an objective with a numerical aperture of 0.26. A longitudinal (\( z \)-axis) magnetic field ranging from \( B_{\text{ext}} = 0 \) to 20 mT is produced by Helmholtz coils positioned around the flow cryostat. The spectroscopy system consists of a 0.75 m monochromator and a liquid-nitrogen cooled CCD camera, providing a spectral resolution of \( \sim 30 \) \( \mu \)eV. By using a scanning Fabry-Perot interferometer of 15 GHz (62 \( \mu \)eV) free spectral range and a finesse \( >70 \), a spectral resolution \( <1 \) \( \mu \)eV is achieved.

The PL polarization and spin splitting are studied by resonantly exciting a single QD in one of its (discrete) excited (p-shell) states under external magnetic fields \( (B_{\text{ext}}) \) ranging from \( B_{\text{ext}} = 0 \) to 20 mT, applied along the crystal growth \( z \)-axis at \( T = 5 \) Kelvin. The PL spectral lines associated with different charging states of a single QD \[ \alpha, \beta \] can be identified from the PL intensity contour plot as a function of the bias voltage and emission energy (Fig. \[ 1 \]). The neutral exciton \( X^0 \) line exhibits a fine-structure splitting of \( \sim 20 \) \( \mu \)eV due to the anisotropic electron-hole exchange interaction. The negatively (positively) charged trion \( X^- \)\( (X^+) \) emission arising from optical excitation of a single electron (hole) charged QD is red (blue) shifted by \( \sim 5.5 \) meV (\( \sim 3.0 \) meV) with respect to the neutral exciton \( X^0 \) line.

The polarization for excitation and detection are denoted as \( (\sigma^x, \sigma^y) \), where \( \sigma^x \) and \( \sigma^y \) correspond to excitation and detection, respectively. The index \( \alpha \) or \( \beta \) assumes one of four values: linear polarization along the [110]\( |\sigma^y\rangle \), [001]\( |\sigma^z\rangle \) crystal axes or circular polarization \( \sigma^\pm \). The degree of circular polarization is defined as \( \rho^z \equiv (I^+ - I^-)/(I^+ + I^-) \), where \( I^\sigma \) denote the intensity of PL under the \( (\sigma^x, \sigma^y) \) configuration. The polarization characteristics of the system is calibrated by the Raman scattering by the longitudinal optical phonon \[ 1 \] of the GaAs substrate layer and the degree of polarization is found to be better than 98%.

Circularly polarized resonant p-shell pumping of a single electron (hole) charged QD \[ \alpha, \beta \] generates optically oriented trions with hole (electron) spin \( J_z = 3/2 \)
ized excitation. Under linearly polarized \((\sim)\) interactions \([14]\). The excitation is at 1.35615 eV, which corresponds to the p-shell resonance for \(X^0\); \(~\sim\) 40.4 meV above \(X^0\). (b) and (c) Degree of circular polarization \(\rho_c\) of PL for \(X^+\) (b) and \(X^-\) (c) under \((\sigma^+)\) excitation with energy \(\sim\) 35 and 40 meV above \(X^-\) and \(X^+\) lines, respectively. \(\rho_c\) depends strongly on bias voltage and weakly on pump power (not shown).

\((S_z = -1/2)\) or \(J_z = -3/2\) \((S_z = +1/2)\), under \(\sigma^+\) and \(\sigma^-\) pumping, respectively. The intra-dot excitation ensures maximal electron (hole) spin preservation during relaxation, which is confirmed by the high degree of circular polarization \(\rho_c\) of the \(X^+\) \((X^-)\) lines, ranging from \(~\sim\) 60% for QD-A (Fig. 1c) to \(~\sim\) 90% for QD-B (Fig. 2). The initial state of \(X^+\) trion is composed of two holes in a singlet-state: \(\sigma^+\) \((\sigma^-)\) polarized PL from this state indicates that the optically excited electron is in \(S_z = -1/2\) \((S_z = 1/2)\) state. For a \(X^-\) trion, the initial state is composed of an electron singlet and the PL polarization is determined uniquely by the polarization of the hole state: \(\sigma^+\) \((\sigma^-)\) polarized PL however indicates that the electron remaining in the QD after spontaneous emission is in \(S_z = 1/2\) \((S_z = -1/2)\) state. In both cases polarized PL implies a spin-polarized QD electron, which can in turn polarize nuclear spins via hyperfine interactions [14].

Figure 2a shows the spectra of \(X^-\) (central energy \(\sim\) 1.31634 eV) for QD-B obtained using a scanning Fabry-Perot interferometer under linearly and circularly polarized excitation. Under linearly polarized \((\sigma^+)\) laser excitation, no fine structure splitting is observed, confirming the diminished effect of anisotropic exchange interaction and absence of nuclear spin polarization. Under circularly-polarized \((\sigma^+)\) excitation, spin doublets with \(\sim\) 10 \(\mu\)eV splitting appear even in the absence of an externally applied magnetic field. The \(X^-\) PL peaks that are co-circular with the excitation laser have lower energies for both \(\sigma^+\) and \(\sigma^-\) excitation (Fig. 2a), indicating that the direction of the effective magnetic field responsible for the observed splitting can be changed by switching the electron-spin polarization from \(S_z = -1/2\) to \(S_z = 1/2\). For \(X^+\) PL (Fig. 2b), this energy sequence is reversed, indicating that the electron spin is polarized in opposite directions in the \(X^-\) and \(X^+\) trions, for a fixed circularly-polarized laser excitation [14].

Finally, measurements carried out while modulating the excitation polarization between \(\sigma^+\) and \(\sigma^-\) show that the magnitude of the spin-splitting reaches steady-state in about \(\sim 1\) sec: for faster modulation rates a sharp decrease in both the spin-splitting and the PL polarization for \(X^-\) is observed. Based on these observations, we conclude that the spin-splitting shown in Fig. 2a is a clear signature of DNSP.

Coupling of a single confined electron to \(N \sim 10^5\) nuclear spins in a QD is well described by the Fermi contact
Hyperfine interaction:

\[ \hat{H}_{hf} = \sum_i A_i \lvert \psi(R_i) \rvert^2 \hat{S} \cdot \hat{I} \]

\[ = \sum_i \hat{B}_e \hat{I}_z + \sum_i A_i \lvert \psi(R_i) \rvert^2 \left[ \hat{S}_+ \hat{I}_- + \hat{S}_- \hat{I}_+ \right] \quad (1) \]

where \( \lvert \psi(R_i) \rvert^2 \) denote the probability density of the electron at location \( R_i \) of the \( i^{th} \) nuclear spin and \( A_i \) is the corresponding hyperfine interaction constant. \( \hat{S} \) and \( \hat{I} \) are the electron and nuclear spin operators, respectively. When the electron is spin polarized via circularly polarized optical excitation under a vanishing external magnetic field, hyperfine interactions play a triple role: First, spin polarized confined electron leads to an inhomogeneous Knight field \( \hat{B}_e \) seen by each QD nucleus. It should be emphasized that \( \hat{B}_e \) is an operator that has a finite mean value \( \langle \hat{B}_e \rangle = B_e \) for a spin polarized electron. Second, the flip-flop term \( \propto \sum_i [\hat{S}_+ \hat{I}_- + \hat{S}_- \hat{I}_+] \) in Eq. (1) enables nuclear spin pumping along the direction determined by the electron spin, provided that electron is continuously spin-polarized by optical excitation and that \( B_e \) is larger than local nuclear dipolar fields. Third, the Overhauser field \( B_o \propto \sum_i A_i \lvert \psi(R_i) \rvert^2 \langle \hat{I}_z \rangle \) induced by the polarized nuclei on the QD electron results in a spin splitting in PL spectrum that can be detected by high-spectral-resolution optical spectroscopy as shown in Fig. 2.

It has been argued that an external magnetic field exceeding the local nuclear dipolar fields is necessary to ensure that spin non-preserving terms in nuclear dipole-dipole interactions are rendered ineffective in depolarizing nuclear spins [10]; this argument is correct only if the Knight field is vanishingly small. A careful analysis shows that, if the inhomogeneous nature of electron-nuclear coupling could be neglected, the expectation value of the DNSP generated Overhauser field would be expressed as [13, 17, 18, 19]:

\[ B_n = f \frac{\mathbf{B}^*(\mathbf{B}^* \cdot \langle \mathbf{S} \rangle)}{\mathbf{B}^* \cdot \mathbf{B}^* + B_L^2}, \quad (2) \]

where \( \mathbf{B}^* = B_e \mathbf{z} + \mathbf{B}_{ext} \) is the total effective magnetic field seen by the nuclei, \( \langle \mathbf{S} \rangle \) is the expectation value of the electron spin, \( B_L \) is the effective local field characterizing nuclear spin-spin interactions [10], and \( f \) is a proportionality constant. In the present experiments, similar values of the Overhauser field are observed for \( B_{ext} = 0 \), 20 mT (Fig. 2), and 200 mT (measured using a permanent magnet; not shown): the expectation value of the Knight field produced by a single spin-polarized electron appears to be strong enough to ensure \( B_e^2 \gg B_L^2 \) and enables significant DNSP without an external magnetic field.

Based on Eq. (2) it could be concluded that application of an external field that cancels the Knight field (i.e. \( \mathbf{B}^* = 0 \)) should result in the complete disappearance of DNSP. Figure 3 shows the dependence of the observed spin-splitting of \( X^- \) trion under conditions where the Zeeman splitting due to the external magnetic field (\( \lesssim 50 \) Gauss) is negligible: for this particular QD (A), applied gate voltage, and the excitation intensity, a dip in spin splitting at \( B_{ext} = -B_e \approx +6 \) Gauss is observed under \( \sigma^- \) pumping. Even at this field however, the spin-splitting is only reduced from \( \sim 16 \) µeV to \( \sim 12 \) µeV, indicating that the cancelation of the Knight field \( B_e \) by the external field is far from being complete. The minimum in spin-splitting is observed at \( B_{ext} \approx -6 \) Gauss when the polarization of the excitation laser is switched.
from $\sigma^-$ to $\sigma^+$. The observed minima, which gives the average value of the Knight field $B_k$, ranges from 6 Gauss to $\sim 30$ Gauss depending on the degree of PL polarization, pumping intensity and the QD that is studied. In the case of high $B_k$, the inhomogeneity of the Knight field, arising from the confined electron wave-function, makes it difficult to measure the value of $B_k$ accurately. This is due to the fact that for any value of $B_{ext}$ most of the QD nuclei experience a strong non-zero total magnetic field and therefore the reduction in overall DNSP remains small.

Remarkably, a dip in the degree of PL polarization is also observed for the same $B_{ext}$ (Fig. 3a): this is at first surprising since polarization of the $X^-$ trion line is solely determined by the hole-spin and a direct interaction between the heavy-hole and the nuclei is unlikely to be strong enough [21] to lead to the observed dependence. A possible explanation is based on anisotropic electron-hole exchange interaction: after the resonant excitation of the QD, the electron excited into a p-shell state of the conduction band is expected to tunnel out into the n-doped GaAs layer in sub-psec timescale [21]. After tunneling, the QD is neutral and the remaining electron-hole pair is subject to anisotropic electron-hole exchange interaction which rotates the electron-hole spin in a correlated manner, in a timescale given by $\sim 1/\omega_{ex} \sim 35$ psec (for both of the QDs studied here $\hbar \omega_{ex} \sim 20$ $\mu$eV). This coherent rotation is interrupted by re-injection of another electron from the n-doped GaAs layer into the QD s-shell to form an electron-singlet in $\tau_e \sim 5 - 20$ psec, as required by the charging condition. Because tunneling is a random process with an average waiting time $\tau_t$, the post-tunneling hole-spin state is partially randomized and leads to a finite PL polarization. The Overhauser-field competes with the exchange interaction: a reduction in DNSP will therefore lead to a reduction in $\rho_e$, as depicted in Fig. 3c.

The PL polarization in the presence of a nuclear Overhauser shift ($\Omega_{hf}$, $B_n$) and exchange interaction can be approximated as [22]:

$$\rho_e = \frac{1 + \Omega^2_{hf} \tau_t^2}{1 + (\Omega^2_{hf} + \omega^2_{ex})\tau_t^2},$$

provided other spin relaxation processes are neglected. Fitting the polarization $\rho_e(X^-)$ with the measured spin splitting in Fig. 3, taken for QD-A, $\tau_e = 30$ psec is obtained. The magnitude of $\tau_t$ [21] is consistent with the previously reported values for $\tau_t$ [20]. For QD-B (Fig. 3h), $\tau_t \sim 10$ psec is obtained for $\rho_e(X^-) \simeq 90\%$ using Eq. 3. Below saturation, a reduction in the excitation power results in a decrease in both spin-splitting and $\rho_e$: this observation corroborates the model described by Eq. 3.

The electron (spin) exchange with the n-doped GaAs layer also explains how QD electron-spin pumping is achieved in a negatively charged QD: irrespective of the pre-optical-excitation electron state, the sequential tunneling events ensure that the QD ends up in a trion state where the electrons form an s-shell singlet. Preservation of hole-spin in these QDs [23] then implies that the post-recombination electron is always projected into the same spin-state. Presence of a shorter barrier layer between the QD and the n-doped GaAs could ensure that hole-spin rotation due to exchange interaction is negligible: in this limit however, spin co-tunneling of the remaining QD electron could reduce the electron-spin polarization.

Some of the open questions that warrant further investigation include the reasons for relatively low level of DNSP where only $\sim 10\%$ of the QD nuclei appear to be polarized; differences in the magnitude of DNSP among the four different nuclear species present in self-assembled QDs; and the role of quadrupolar interactions enhanced by the strain [24]. By using differential transmission measurements [21], it should be possible to enhance the accuracy with which the Overhauser field can be measured by at least an order of magnitude.

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As shown in Fig. 1b, QD-A exhibits an asymmetric polarization under $\sigma^\pm$ pumping: the origin of this asymmetry is not clear. We observed no such asymmetry for QD-B.