Polarization freezing of $10^4$ optically-cooled nuclear spins by coupling to a single electron

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The nature of the nano-scale environment presents a major challenge for solid-state implementation of spin-based qubits. In this work, a single electron spin in an optically pumped nanometer-sized III-V semiconductor quantum dot is used to control a macroscopic nuclear spin of several thousand nuclei, freezing its decay and leading to spin life-times exceeding 100 seconds at low temperatures. Few-millisecond-fast optical initialization of the nuclear spin is followed by a slow decay exhibiting random telegraph signals at long delay.

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times, arising from low probability electron jumps out of the dot. The remarkably long spin life-time in a dot surrounded by a densely-packed nuclear spin environment arises from the Knight field created by the resident electron, which leads to suppression of nuclear spin depolarization.

Spin-based approaches are one of the promising routes for quantum information processing (QIP), for which an essential requirement will be long-lived spin memory and coherence relying on effective isolation of a spin-qubit from its magnetic environment. In this respect, good candidates for solid-state implementation of QIP are materials with well-isolated nuclear spins such as $^{29}$Si nuclei [1, 2] and $^{31}$P impurities [3, 4] in silicon, nitrogen nuclei in N@C$_{60}$ molecules [5] and $^{13}$C [6, 7, 8] in diamond. In III-V semiconductors, favored for fabrication of advanced quantum dot (QDs) nano-structures suitable for both electrical [9, 10, 11] and optical [12, 13, 14, 15] control of spin states, all atoms carry non-zero nuclear spin. This results in efficient spin decay via spin non-conserving nuclear dipole-dipole interactions introducing an uncontrollable dephasing in QD-based qubits [10, 11]. Dynamic nuclear polarization enabling control over the magnetic environment is a possible way to circumvent this problem [16, 17, 18, 19, 20, 21]. Here we show that in a III-V semiconductor QD a single electron stabilizes the optically excited nuclear spin system leading to very long spin life-times of several hundred seconds, limited only by the electron residence time on the dot.

Our approach makes use of optical manipulation of a single electron-charged InP self-assembled quantum dot embedded in bulk GaInP. Addressing individual well-isolated dots is crucial for these studies to avoid sample inhomogeneities [14, 22] and to operate with a precise knowledge of the Overhauser field ($B_N$) on the dot. A typical dot (4nm x 20nm x 20nm) contains about $10^4$ nuclei and one trapped electron originating from residual donors in the GaInP [23]. We employ the technique of optically induced dynamic nuclear polarization and photoluminescence (PL) measurements to monitor the temporal evolution of the Overhauser field at a temperature of 4.2K and magnetic field $B_z$ up to 8T. We find nuclear polarizations which persist for hundreds of seconds after the optical pumping has been turned off, with the decay at long times having a random telegraph character, which we attribute to random jumps of the trapped electron from
the dot. The trapped electron plays a crucial role: when the electron occupies the dot, its inhomogeneous Knight field freezes the nuclear spin decay mediated by the dipole-dipole interaction, leading to the suppression of spreading of the nuclear polarization from the dot into the surrounding semiconductor.

The dynamic nuclear polarization occurs under circularly polarized optical excitation of the dot leading, initially, to pumping of the electron spin [24]. The hyperfine interaction leads to spin flip-flops between the electron and a single nucleus [24]. Macroscopic nuclear spin, accumulated on the dot as a result of the dynamic nuclear polarization, acts back on the confined electron with an effective magnetic field \( B_N \) [24]. This results in the modification of the electron energy spectrum manifested in the splitting \( \Delta E = \mu_B [g_e B_z - g_e (B_z + B_N)] \) observed in the electron-hole recombination spectrum \([g_e(h)]\) is the electron (hole) g-factor]. The sign of \( B_N \) depends on the helicity of the circularly polarized excitation. In this work we measure \( \Delta E \) to determine \( B_N \), directly reflecting the degree of the nuclear spin polarization on the dot.

The probability for the spin transfer between the residual electron and the nuclear spin system,

\[
    w_s \propto \frac{|A_{hf}|^2}{(\Delta E_e^2 + \gamma^2/4)},
\]

depends on the major energy cost of the electron-nuclear spin flip-flop event [25] - the electron Zeeman splitting \( \Delta E_e = \mu_B g_e (B_z + B_N) \). \( A_{hf} \) is the hyperfine interaction constant and \( \gamma \) is the electron state broadening [24]. The resonant form of the rate \( w_s \) in Eq. 1 results in feedback between the nuclear spin polarization on the dot and the spin transfer rate. In particular, an abrupt build-up of nuclear spin on the dot under optical excitation, the so-called nuclear spin switch effect [26 21 23], occurs when \( \Delta E_e \approx 0 \) for \( B_N \approx -B_z \) leading to a sharp increase of \( w_s \).

Measurement of the nuclear spin life-time on the dot has been carried out with a pump-probe method based on single-dot PL detection. First, the dot is excited by the strong pump and then a PL spectrum excited by a weak delayed probe is measured. In order to identify an optimum duration and optical power of the probe, which does not perturb the system, the nuclear polarization rise time is firstly measured in the experiment sketched in Fig. 1A. First, a long \( \sigma \)-polarized erasing pulse is used to destroy any nuclear
polarization left from the previous excitation cycle. This is followed by a pump pulse with helicity opposite to that of the erase pulse. The nuclear polarization generated by the pump is determined from the spectral splitting $\Delta E$, measured during the last quarter of the pump pulse. The erase/pump cycle is repeated several times to improve the accuracy of the measured $\Delta E$ value. The polarization build-up is then obtained by plotting the nuclear field $B_N$ as a function of the pump pulse duration $t_{pump}$.

The nuclear spin pumping dynamics strongly depend on the polarization of the optical excitation. Fig.1B shows the dependence of $B_N$ on the optical pumping time in a charged QD at $B_z=0.32$ T, measured for both circular polarizations of the pump. For $\sigma^-$-polarized pump a slow change in $B_N$ is observed. In contrast, for the $\sigma^+$ pump $B_N$ abruptly reaches a constant value after $t_{pump} \approx 0.2$ s. This indicates the condition when $B_N$ compensates the external field $B_z$ and positive feedback of the spin pumping on the nuclear spin transfer takes place eventually leading to the nuclear spin switch [26, 21, 23]. In the $\sigma^+$ case the observed sharpness of the transition is limited by averaging of the PL signal during the detection time $t_{det} = t_{pump}/4$. In the case of $\sigma^-$-excitation negative feedback of the nuclear polarization on the spin transfer rate plays a major role, slowing down the spin pumping dynamics. In the limit of long $t_{pump}$ the nuclear field reaches a steady-state value, $B_N^{cw}$, strongly dependent on the polarization of incident light due to the feedback described by Eq.[1].

Such dependence (Fig.1C) persists in a wide range of magnetic fields $B_z < 3$T, with $B_N^{cw}$ growing with $B_z$ from $B_N^{cw} \approx 0.2$ T at $B_z=0$. It reaches its maximum $\approx 1.2$ T under $\sigma^+$ excitation at $B_z \approx 1.2$ T, when $B_z$ and $B_N$ compensate each other, thus leading to enhanced electron-to-nuclei spin transfer rate $w_s$. In high magnetic fields ($B_z > 3$ T) the difference in $B_N^{cw}$ as well as in the pumping dynamics for $\sigma^+$ and $\sigma^-$ excitation vanishes as the nuclear field becomes small compared to the external field and does not lead to any significant feedback on the spin transfer rate $w_s$ [23]. Due to a large electron Zeeman splitting $\Delta E_e$ in this regime, the nuclear spin pumping becomes less efficient and $B_N^{cw}$ decreases down to $\approx 0.6$ T for both polarizations of excitation.

Below we use the build-up time ($\tau_{build-up}$) required to reach 70% of the temporal dynamic range of $B_N$ as a measure of the nuclear spin rise time. The dependence of $\tau_{build-up}$ on $B_z$ for $\sigma^+$ polarized pump is shown by the triangles in Fig.2. The nuclear
spin build-up time increases from $\tau_{\text{build-up}} \approx 5$ ms at $B_z = 0$ to 2.5 s at $B_z = 2$ T. The very marked slowing down of the spin pumping dynamics occurs due to the increase of the electron Zeeman splitting in the external field $B_z$, as described above by Eq. 1. However, the saturation of $\tau_{\text{build-up}}$ at $B_z > 2$ T observed in Fig. 2 is not explained by Eq. 1. This discrepancy might be evidence for several other processes contributing to the dynamic nuclear polarization with strongly differing magnetic field dependences.

Understanding of the build-up dynamics enables us to measure the nuclear spin decay using the pump-probe techniques shown schematically in the inset in Fig. 3A. The nuclear spin polarization was induced by a circularly polarized pump pulse with duration $t_{\text{pump}} \approx 10 \tau_{\text{build-up}}$, long enough to reach the steady-state nuclear field $B_N^{\text{up}}$. Photo-excitation was then blocked by a mechanical shutter for a time $t_{\text{delay}}$. After the delay, the nuclear polarization was measured in PL excited with a short pulse (much shorter than $\tau_{\text{build-up}}$).

An example of the pump-probe curves measured in this way is shown in Fig. 3A. Here the dependences of $B_N$ on $t_{\text{delay}}$ for both $\sigma^+$ and $\sigma^-$ polarized pump pulses at $B_z = 8$ T are shown. For $t_{\text{delay}} \leq 100$ s no significant decay of the nuclear field $|B_N^{\text{pump}}| \approx 0.8$ T is observed. For longer delays, the nuclear polarization fluctuates from 0 to almost $B_N^{\text{pump}}$, significantly larger than the experimental error $\sim 0.15$ T. Such large fluctuations of $B_N$ with random telegraph character observed after long delays mean that the nuclear spin decay in the QD is a random process and $B_N$ is not a continuous function of $t_{\text{delay}}$. Mean-square fitting with a single exponential functions, averaging over the abrupt jumps, leads to an estimate for $\tau_{\text{dec}}$ of $\approx 250$ s (see solid lines in Fig. 3A). As shown in Fig. 2 $\tau_{\text{dec}}$ (squares) is weakly dependent on $B_z$ and exceeds 100 s. At $B_z = 0$ the longest delay time which can be measured in our pump-probe set-up is $\tau_{\text{delay}} \approx 30$ s (shown with a horizontal bar in Fig. 2), within which no detectable decrease of the nuclear polarization on the dot was found. We also have not observed any dependence on the pumping time of the nuclear depolarization time in the charged dots studied here, indicating the absence of nuclear spin diffusion.

We attribute the very long decay times we observe to the role of the Knight shift induced by the residual electron in the charged dot. The Knight field it creates on the dot can be measured directly from the magnetic field dependence of the Overhauser splitting $\Delta E_N = \mu_B g_e B_N$ at low $|B_z| < 20$ mT as shown in Fig. 3B. When an external magnetic field
compensates the inhomogeneous Knight field for a portion of the QD nuclear spins, the Zeeman splitting becomes zero for these nuclei. This enhances spin relaxation, observed as a partial decrease (<10%) of the Overhauser field. Excitation with the opposite circular polarization creates a Knight field of the opposite sign. The splitting $2B_e$ between the minima of $\Delta E_N$ curves for the two polarizations gives the Knight field $B_e \sim 3$ mT. The measured value of $B_e$ significantly exceeds the local field $B_L \sim 0.1$ mT [24] created by the nuclear dipole-dipole interaction. Thus we conclude that the electron spin generates an inhomogeneous Knight field leading to a strong inhomogeneity in the nuclear Zeeman splitting (see diagram in Fig.3C), which suppresses nuclear spin diffusion and relaxation due to the dipole-dipole interaction [27].

The abrupt step-like nature of the nuclear spin relaxation with the form of a random telegraph signal is very different from that observed in previously studied systems [28, 29, 30]. We observe large fluctuations $\Delta B_N$ comparable to the value of $B_N$ itself, which suggests that the nuclear spin relaxation is triggered by a discrete process, most probably electron hops out of the dot. In the absence of the electron, the stabilizing Knight field disappears enabling the nuclear spin relaxation due to the dipole-dipole interaction, leading in particular to nuclear spin diffusion [29]. Note, that a similar effect is expected if an additional (second) electron is captured by the dot, forming a spin singlet double-charge state. The proposed scenario explains the weak magnetic field dependence of $\tau_{\text{dec}}$, since the latter depends only on the average time for the dot recharging.

The spin decay times found in our sample are about $10^4$ times longer than in Schottky-gated structures [28]. This occurs since the electron and nuclear spins in QDs in our sample form a stable closed system, where the spin exchange with the electron sea in the contact [28] is eliminated. The observed $\tau_{\text{dec}}$ up to 500 s is considerably longer than $\tau_{\text{dec}} \leq 5$ s in neutral InGaAs dots placed in high magnetic fields, where the nuclear spin diffusion was found to be the main mechanism of the nuclear spin relaxation [29]. The lower limit of $\tau_{\text{dec}} \approx 30$ s observed in our work for $B=0$ is also more than one order of magnitude longer than in a neutral InGaAs dot measured at zero field in Ref. [28].

We also find that the Overhauser field produces a similar stabilizing effect on the electron spin at zero external field. A clear correlation between the electron spin-flip time
and nuclear spin polarization has been observed. This arises from the suppression of the electron spin-flip (caused by the hyperfine interaction) for non-zero Overhauser field on the dot, as can be expected from the strong dependence of the spin flip-flop rate $w_s$ on the electron Zeeman splitting $\Delta E_z$ in Eq. 1.

Our results demonstrate previously un-revealed dynamic properties of the strongly coupled spin-system of a single electron and an ensemble of nuclei in a semiconductor nano-structure. In the charged quantum dots a single electron and $\approx 10^4$ nuclei form an almost closed system with a conserved total spin. Optical control through the electron enables fast optical initialization and read-out, that combined with very long spin lifetimes, leads to opportunities for the use of this nano-system as a resource for storage of spin-encoded information.

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**Methods**

*QD sample:* The sample containing InP/GaInP quantum dots was grown on a GaAs substrate by metal organic vapor phase epitaxy (MOVPE). In order to access single quantum dots we deposited an opaque metal mask with 400 nm clear apertures. Although the sample was not doped intentionally, the background doping is sufficient for the majority of the dots to be negatively charged. The charge state of the dots used in this study was verified in several ways as explained in Ref. [23] in detail. The evidence of negative charging is obtained from polarization-resolved photoluminescence (PL) spectroscopy in external magnetic field both perpendicular and parallel to the sample surface. At zero magnetic field the PL spectrum of a negatively charged dot consists of a single line. Magnetic field perpendicular to the sample surface (Faraday geometry) splits this line into a circularly polarized doublet. For magnetic field parallel to the sample surface (Voigt geometry) two doublets with orthogonal linear polarization are observed. The energy splitting between these four lines is directly proportional to the magnitude of the field. Such a spectral
pattern in the Voigt geometry is characteristic of dots containing a single charge, and is different from that of neutral dots [23]. However, the sign of the charge in the dot cannot be determined in this way. In order to prove the negative sign of the charging we use PL spectroscopy under circularly polarized excitation. For close to resonance excitation we observe negative circular polarization (NCP): at high excitation rate PL of the dot is predominantly circularly polarized with helicity opposite to that of the exciting laser. This phenomenon is a fingerprint of negatively doped dots and is attributed to electron spin memory [22, 31]. Finally, the observed effect of the Knight field gives direct evidence that the dot is charged with a single electron.

**Experimental technique:** In all of the experiments the sample was kept in a low-pressure He atmosphere and was mounted on a metal plate in direct contact with liquid He at 4.2 K. Magnetic field perpendicular to the samples surface up to 8 T was provided by a superconducting magnet. In this work we studied quantum dots emitting at $\approx 1.85$ eV. We used semiconductor lasers emitting below the GaInP band-gap at $\approx 1.9$ eV for PL excitation as well as for nuclear spin pumping. The exciting laser was focused on the surface of the sample with an aspheric NA 0.65 lens, providing a focusing spot of $\approx 2 \mu m$. The same lens was used to collect PL, which was then dispersed by a 1 m double monochromator coupled with a CCD camera. The accuracy of PL peak spectral positions determined by the CCD pixel size was $\approx 30 \mu eV$. However, measurement of the center of mass for each peak provided much better precision, and was used to measure spectral splittings with accuracy of better than $1 \mu eV$. In pump-probe experiments mechanical shutters were used to modulate both lasers and PL signals, providing time resolution of $\approx 2$ ms.

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Figure 1: Nuclear spin build-up dynamics in a single negatively charged QD under circularly polarized optical excitation. (A) Temporal diagram of a measurement cycle for the nuclear spin build-up dynamics. A long circularly polarized erase pulse erases the polarization left from the previous cycle. Then a pump pulse of variable duration and of the opposite helicity initializes nuclear polarization on the dot. The dynamics of the Overhauser field build-up is obtained from the dependence of the spectral splitting in dot PL, measured during $t_{\text{det}}$, on $t_{\text{pump}}$. (B) Nuclear spin build-up dynamics under $\sigma^+$ (triangles) and $\sigma^-$ (circles) polarized excitation at $B_z=0.32$ T. (C) Absolute magnitude of the steady-state Overhauser field $|B_N|$ under $\sigma^+$ (triangles, $B_N<0$) and $\sigma^-$ (circles, $B_N>0$) polarized excitation.
Figure 2: Magnetic field dependence of the optically induced nuclear spin dynamics. Triangles show the spin build-up time under $\sigma^+$ polarized pumping. Squares show the nuclear spin decay time once the pumping has been turned off. At $B=0$ no decay of nuclear polarization is found for the longest measured waiting time of 30 s (shown by the horizontal bar). Nuclear spin build up is slowed down from $\tau_{\text{build-up}} \approx 5$ ms at $B=0$ to $\approx 2.5$ s at $B>3$ T. At the same time the nuclear spin decay time is almost independent of external field.
Figure 3: (A) Decay of nuclear spin polarization on the QD measured at $B_z=8$ T in a pump-probe experiment schematically shown in the inset. Squares and circles show data measured with $\sigma^+$ and $\sigma^-$ polarized pumps respectively. For delays $t_{\text{delay}}>100$ s large fluctuations of $B_N$ can be seen, demonstrating that nuclear spin decay is a random discrete process. The single exponential fit (shown with solid lines) gives an estimate of the nuclear spin decay time $\tau_{\text{dec}}\approx250$ s. (B) Schematic representation of the effect of a trapped electron on nuclear spin decay: inhomogeneous Knight field causes energy splitting mismatch between different nuclei, leading to suppression of nuclear spin diffusion out of the dot. (C) Magnetic field dependence of spectral splitting $\Delta E_N$ induced by nuclear polarization on the dot under $\sigma^+$ (squares) and $\sigma^-$ (circles) excitation. The observed minima of nuclear polarization correspond to a situation when the external field $B_z$ compensates electron Knight field $\pm B_e$ induced by $\sigma^\pm$ excitation, resulting in partial depolarization of nuclei. The value of $|B_e|\approx3$ mT is estimated from splitting between the minima.