Suppression of nuclear spin fluctuations in an ensemble of (In,Ga)As/GaAs quantum dots excited with a GHz-pulsed laser

E. Evers,1,* N. E. Kopteva,1 I. A. Yugova,2 D. R. Yakovlev,1,3 M. Bayer,1,3 and A. Greilich1

1Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany
2Physical Faculty of St. Petersburg State University, 198504 St. Petersburg, Russia
3Ioffe Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

(Dated: November 16, 2020)

The coherent electron spin dynamics of an ensemble of singly charged (In,Ga)As/GaAs quantum dots in a transverse magnetic field is driven by periodic optical excitation at 1 GHz repetition frequency. Despite the strong inhomogeneity of electron g-factor, the spectral spread of optical transitions, and the broad distribution of nuclear spin fluctuations, we are able to push the whole ensemble of excited spins into a single Larmor precession mode that is commensururate with the laser repetition frequency. Furthermore, we demonstrate that an optical detuning of the pump pulses from the probed optical transitions induces a directed dynamic nuclear polarization and leads to a discretization of the total magnetic field acting on the electron ensemble. Finally, we show that the highly periodic optical excitation can be used as universal tool for strongly reducing the nuclear spin fluctuations and preparation of a robust nuclear environment for subsequent manipulation of the electron spins, also at varying operation frequencies.

The last decade has been marked by unprecedented progress in the development of quantum technologies. This is confirmed by the development and first implementation of quantum communication1 and quantum computing2. At the heart of these technologies are solid state quantum bits (qubits) and their entanglement3. As the race for the best qubit candidate is still ongoing, it becomes clear that there will be no monolithic solution, but rather a hybrid solution combining different excitations, each exploiting its own best property while contributing to the common goal of the targeted quantum technology.

One of the possible hybrid qubit realizations is the spin of an electron confined in a semiconductor quantum dot (QD), which is interacting with the surrounding nuclear spins4. The prominent advantage of QDs is their strong optical dipole moment, which allows efficient coupling of photons to the confined electron spins, according to optical selection rules. The electron spin is coupled to the nuclear spins of the QD crystal lattice by the hyperfine interaction4, which could allow one to design schemes where the angular momentum of the photon is transferred to the nuclear spins using the electron spin as auxiliary state. The advantage of this approach is that the electron spin coherence is limited to several microseconds at low temperatures5, but the nuclear spin coherence can last milliseconds6, allowing in particular the implementation of quantum repeater schemes7.

The idea to transfer the electron spin state to the surrounding nuclear spins is aggravated by the intrinsic nuclear spin fluctuations8. A way to reduce these fluctuations was first elaborated theoretically8 and later demonstrated in a series of experiments9–13. Furthermore, every QD in the ensemble contains about 105 nuclear spins, so that one expects nuclear-spin fluctuations in the Overhauser field (δB_M)4,15 acting on the electron spins in the QDs due to the hyperfine interaction, see sketches in Fig. 1a. Due to the variation of the constituent material, the inhomogeneity is also present in the electron g-factors in the ensemble, whose dispersion is shown by the black line in Fig. 1b. The combination of these effects manifests itself as a fast dephas-
To study the coherent spin dynamics in the QD ensemble, we use time-resolved Faraday rotation (FR). Exemplary traces for pulsed excitation with repetition frequencies of 75.76 MHz (red) and 1 GHz (black), corresponding to repetition periods of $T_R = 13.2$ ns and $T_R = 1$ ns, respectively, are shown in Fig. 1c for $B_x = 1.28$ T. As one can see, for the case of $T_R = 13.2$ ns the signal decays within $T^*_2 = 1.2$ ns, while there is no observable spin decay for 1 ns pulse separation. Here, an assessment of the temporal dynamics is impossible for times exceeding 1 ns, therefore we apply an adapted extended pump-probe method. The spin dynamics is shown in Fig. 1d, demonstrating electron spin dephasing on a timescale of $T^*_2 = 17$ ns. In this case, pump and probe pulses are picked by electro-optical modulators and hit the sample in bunches, with a controlled delay time between the pump and probe pulse combinations, see the inset in Fig. 1d.

To explain the observed difference in $T^*_2$ for both repetition frequencies, we first consider the case of 13.2 ns repetition period. The FR signal exhibits a pronounced rise of the electron spin polarization before each pump pulse arrival (0 ns or 13.2 ns delay) which mirrors the decay thereafter (effect of SML). Both the decay and the rise of the signal are caused by the superposition of multiple precession modes which leads to destructive signal interference between the pump pulses. At a delay of a multiple integer of $T_R$, constructive interference occurs for particular modes with discrete electron spin precession frequencies $\omega$. Generally, $\omega = g\mu_B B_x/\hbar$ in the external magnetic field $B_x$, where $\mu_B$ is the Bohr magneton and $\hbar$ is the reduced Planck constant. The frequencies of the constructively interfering precession modes satisfy the phase synchronization condition (PSC) $\omega = K\omega_R$, where $K$ is an integer.
where \( \omega_R = 2\pi/T_R \) is the repetition rate of the laser pulses and \( K \) is an integer characterizing each contributing mode. As discussed in Refs. [20, 21], the number of PSC precession modes, \( M \), within the inhomogeneous ensemble is given by: (1) the \( g \)-factor spread of the optically excited electron spins (\( \Delta g \)), (2) the nuclear spin fluctuations (\( \delta B_N \)), (3) the external magnetic field (\( B_x \)), and (4) the laser repetition period \( T_R \).

The black solid line in Fig. 1b demonstrates the dependence of the electron \( g \)-factor on the optical excitation energy, following roughly a linear dependence with a slope of \( \Delta g/\Delta E = -1.75 \text{eV}^{-1} \) [22 and 23]. Using the laser energy and the spectral pulse width, this dependence allows us to determine the average \( g \)-factor at the probe energy of 1.3867 eV to be \( |g| = 0.57 \) with a spread of \( \Delta g = 0.004^{24} \). The nuclear field fluctuations are known to be \( \delta B_N = 7.5 \text{mT} \) for this sample\textsuperscript{20}. Therefore, the number of contributing PSC modes at \( B_x = 1.28 \text{T} \) is dominated by the \( g \)-factor spread which covers \( M = 8 \) modes for \( T_R = 13.2 \text{ns} \), as shown by the gray-shaded area in Fig. 1c\textsuperscript{25}.

The number of modes \( M \) is derived here for a width of the Gaussian precession frequency distribution taken as six times its half width at half maximum (HWHM), to account for 99.7% of the spins:

\[
M = 6\Delta \omega T_R/2\pi, \tag{1}
\]

where

\[
\Delta \omega = \mu_B \sqrt{(\Delta g B_x)^2 + (g\delta B_N)^2}/\hbar. \tag{2}
\]

The nuclear fluctuation field \( \delta B_N \) shown by the orange-shaded area is dominant only at small magnetic fields.\textsuperscript{20} For pulsed excitation by the black trace in Fig. 1c, the Overhauser field adds to the external \( B_x \) for electron spins which do not satisfy the PSC, driving their frequency.\textsuperscript{26} The situation for 1GHz laser repetition rate is shown by the black trace in Fig. 1c. For pulsed excitation with \( T_R = 1 \text{ns} \), the separation between neighboring PSC modes is \( B_0 = \hbar \omega_R/(g\mu_B) = 128 \text{mT} \), which is much larger than the \( \delta B_N \) of the nuclear spin fluctuations (7.5 mT). Moreover, the \( g \)-factor spread is also not sufficient to allow for more than one mode within the 128 mT range at a field of \( B_x = 1.28 \text{T} \) (see the black lines in Fig. 1c). As a result, the signal shows a single, slowly decaying oscillation instead of a multi-mode signal with fast dephasing. Hence, the pump-probe signal between two pump pulses for 1GHz excitation can be evaluated using a single cosine function with a frequency \( \omega \):

\[
S(t) = S_1 \cos(\omega t). \tag{3}
\]

\( S \) is the signal amplitude, \( S_1 = S_0 \exp(-t/T_Z^*) \) where \( S_0 \) is the electron spin polarization created by the pump, \( t \) is the pump-probe time delay and \( T_Z^* \) is the electron spin dephasing time related to the single-mode frequency bandwidth.

**Influence of nuclear spins**

As the next step, due to the time-resolution limitations set by the electronics in the extended pump-probe scheme, we use the common pump-probe protocol and fit Eq. (3) to the FR data taken for different external magnetic fields (\( B_x \)) for 1GHz excitation. \( S_1 \) is considered to be time independent here as \( T_2^* \gg T_R = 1 \text{ns} \). The oscillation frequency should depend linearly on the external magnetic field, as shown in Fig. 2a by the red line. The data of the Larmor frequency evaluated by Eq. (3) are shown by the black dots in Fig. 2a, and demonstrate a non-linear step-like dependence of \( \omega \), normalized by the laser repetition rate \( \omega_R \).

As one can see in Fig. 2a, the electron spin precession frequency shows small deviations from the linear dependence in small magnetic fields (\( B_x < 0.5 \text{T} \)). Increasing the magnetic field leads to the appearance of pronounced plateaus in the frequency dependence. The positions of the plateaus are fixed by the PSC on integer numbers of full spin revolutions during the laser repetition period \( T_R \) or \( \omega = K \omega_R \). The center of each plateau corresponds to \( B_x = KB_0 \) (the upper axis in Fig. 2a). The origin of this dependence is related to a nuclear magnetic field (\( B_N \)) resulting from the Stark field. This field is perpendicular to the external field.\textsuperscript{27,28} One can see in Fig. 2b that the maximal amplitude of \( B_N \) reaches 50 mT and can be oriented parallel or antiparallel to the external field.

Such plateaus in the dependence of the electron spin precession frequency on the external magnetic field were observed earlier for electron spins localized on Fluorine donors in ZnSe epilayers.\textsuperscript{26} One can explain them in terms of a dynamic nuclear polarization in the following way: the non-resonant optical excitation of the trion resonance leads to the appearance of an effective magnetic field along the light propagation direction - the optical Stark field. This field is perpendicular to \( B_x \), the electron spin precesses about the total magnetic field which is tilted relative to the \( x \)-axis. This leads to the appearance of a sizable component of electron spin polarization along the \( x \)-axis (\( S_x \)), which efficiently polarizes the nuclear spins along the external magnetic field \( B_x \). The nuclear polarization plays the role of the additional field described in the previous paragraph – the Overhauser field, which acts back on the electron spins.\textsuperscript{29,30}

In our experiments, we use a negative optical detuning, where the energy of the probe at the trion resonance is higher than the pump excitation energy.\textsuperscript{23,27} For a negative optical detuning in combination with the negative sign of the electron \( g \)-factor in the (In,Ga)As QDs, the Overhauser field adds to the external \( B_x \) for electron spins which do not satisfy the PSC, driving their fre-
frequency to the PSC-consistent value, i.e. a laser period-commensurate value. This leads to the plateau-like behavior seen in Fig. 2a. The Overhauser field $B_N$ reaches the maximal amplitude of about 50 mT when the external field is slightly larger than $B_x = 0.5KB_0$ (see Fig. 2b). Its amplitude decreases with increasing $B_x$ and becomes zero at $B_x = KB_0$, the center of the plateau. A further increase of $B_x$ changes the direction of $B_N$. Here, it reaches the maximal negative amplitude slightly below $B_x = 0.5KB_0$.

Figure 2c demonstrates the value of $S_1$ in Eq. (3) determined from the fits to the data as function of $B_x$, demonstrating a strong modulation. The magnetic field positions of the peaks correspond to integer spin precession periods within $T_R$, i.e. to fulfilled PSC. This allows us to assume that the $T_2^*$ time should be similarly modulated, as the amplitude $S_0$ in Eq. (3) is expected to stay constant across the plateau, due to the constant values of the Larmor frequencies. To understand this behaviour we use the theory presented in Ref. [28], which relates the extension of the spin dephasing time at the plateau centers to the feedback strength between the electron and nuclear systems, and to the reduction of the nuclear spin fluctuations (see Supplementary Note 5 for more details).

Figure 2d demonstrates the simulation of the frequency behavior (normalized by the laser repetition frequency $\omega_R$) as a function of the external magnetic field $B_x$. The bottom scale gives the applied field, while the top one is normalized by the mode separation $B_0$. One finds fully developed plateaus around the modes 5 and 6. Figure 2e shows the Overhauser field $B_N$ building up in the QD system as function of $B_x$. The parameters of the modeling are given in the Supplementary Note 5.

The build-up mechanism of the dynamic nuclear polarization is strongly influenced by the nuclear spin fluctuations. As suggested in Ref. [26], for the electron spins satisfying the PSC the strong feedback should lead to a reduction of the nuclear spin fluctuations and, as a result, the spin dephasing time of the ensemble $T_2^*$ should be prolonged. As soon as $B_x$ differs from $B_x = KB_0$,
the nuclear fluctuations recover due to the reduced feedback strength (see the gray curves in Fig. 2e and the Supplementary Note 5). The dynamical nuclear polarization process looses its efficiency, even though the $x$-component of the electron spin polarization is largest for $B_x = 0.5K_0B_0$. At this field ($B_x = 0.5K_0B_0$), the amplitude of $B_N$ becomes redirected within a relatively narrow magnetic field interval.

The magnetic field variation of the spin dephasing time $T_2^*$ calculated by Eqs. (S1)-(S6), see Supplementary Note 5, is demonstrated in Fig. 2f. Depending on the magnetic field, this time becomes strongly modulated due to the periodic changes of the amplitude of the nuclear fluctuations. For the parameters used in our modeling, we expect a prolongation of the $T_2^*$ time by a factor of 2.5.

The process of reduction of the nuclear field fluctuations at the center of the plateaus without build-up of nuclear polarization can be qualitatively understood in a similar way as the process of coherent population trapping suggested for a single QD. As a reminder, once the difference of the photon energies of two linearly polarized continuous wave (CW) lasers $\Omega_1$ and $\Omega_2$ is equal to the Zeeman splitting of the ground state electron spin ($\uparrow$ and $\downarrow$), the system goes into a coherent dark state without the possibility of photon scattering into the excited trion state, see Fig. 2g. Due to the nuclear spin fluctuations, the electron Zeeman splitting varies, moving the system out of the dark state. This leads to enhanced driving of one of the two optical transitions that causes scattering of photons and pulls the Zeeman splitting back to that of the dark state by changing the nuclear spin orientation in the surrounding. Such locking into the dark state induces the reduced variance of the Overhauser field.

In the case of pulsed excitation, as we use in our demonstration, the situation is different, see Fig. 2h. In the transverse magnetic field, the pulsed circular excitation leads to creation of a coherent superposition of the ground state spin states (shown by the multiple lines for the ensemble). This superposition precesses in the magnetic field at the Larmor frequency $\omega = g\mu_B B_x/h$. When this frequency is commensurate to the laser repetition frequency $\omega_R = 2\pi K/T_R$, the efficiency of spin polarization is strongly enhanced. Once the electron spin oscillates at one of these frequencies, it can also be seen as locked in a coherent dark state, as the Pauli principle forbids further excitation of the spin by circularly polarized pulses. If the nuclear field fluctuations bring the Zeeman splitting (or the Larmor frequency) out of the resonance condition, the interaction with the nuclear surrounding pulls the frequency back to the dark state, leading similarly to a reduction of the variance of the Overhauser field. In comparison to CW lasers, the pulsed excitation allows us to excite a spectrally broad distribution of QD transitions and can be seen as universal tool without strict requirement concerning the excitation laser energies for spectrally different QDs.

Two-laser protocol

The relaxation dynamics of the contributing electron and nuclear spins differ by several orders of magnitude. The spin lifetime ($T_1$) of the resident electrons in the studied QDs was previously measured, reaching 1.7 $\mu$s, while the lifetime of the nuclear spins for this sample ranges from several seconds under laser illumination up to hours in darkness. We want to make use of this difference and implement a protocol that suppresses the nuclear fluctuations by the 1 GHz excitation and subsequently allows us to manipulate the electron spins with an arbitrary laser source in the pre-prepared nuclear environment. The Supplementary Note 2 shows such an implementation of the suggested alternating driving by the two available lasers. As this, however, takes a long time for the experiments (about 3 hours for one temporal trace), we present here an alternative realization of this idea.

Figure 3a demonstrates a scheme, in which both lasers (75.76 MHz and 1 GHz) are applied simultaneously to the same QD ensemble. In this case we measure a pump-probe trace using the 75.76 MHz laser while pump pulses of the 1 GHz laser simultaneously excite the same ensemble, without any synchronisation between the lasers. Figure 3c demonstrates a comparison between the case when...
only the 75.76 MHz laser is applied (black) and the situ-
ation with both lasers (red). As one can see, in the latter
case the dephasing of the ensemble is strongly reduced, 
which is a direct demonstration of a strong reduction of 
the frequency spread compared with the pure 75.76 MHz case. 
As the emissions of the two lasers are not synchro-
nized to each other, the Faraday rotation measured by 
the probe pulses only stems from the electron spins ori-
tented by the pump pulses with the repetition period of 
75.76 MHz. There is still some minor mode-locking sig-
nal, as seen by the weak signal increase before the pump 
at 13.2 ns, which might arise from the QDs not excited 
by the GHz laser. The sketch in Fig. 3b demonstrates 
the calculated mode distributions for both cases, with 
the corresponding changes in the frequency distributions 
of the ensemble given by the spreads $\Delta g$ and $\delta B_N$, 
the colors correspond to the traces in Fig. 3c.

Using the two-laser approach we measure the mag-
netic field dependence of $T^*_2$ for the two cases, using 
only the 75.76 MHz laser and using both lasers applied. 
Figure 3d demonstrates such a measurement, where the 
pump-probe traces are measured at the center of plateaus 
for a set of magnetic fields. For the analysis of these 
traces we took into account that the signal of a single 
mode oscillation for the two-laser approach can interfere 
with the multi-mode signal from the 75.76 MHz laser 
applied alone (see Supplementary Note 3 for a detailed 
trace analysis).

We characterize the dephasing behaviour of the ex-
tracted signal using the form:

$$T^*_2 = \frac{\hbar}{\mu_B \sqrt{(\Delta g B)^2 + (g \delta B_N)^2}}.$$  

(4)

This leads to the following fit values: (i) 75.76 MHz 
only, $\Delta g = 4 \times 10^{-3}$, $\delta B_N = 7.5$ mT and (ii) both lasers, 
$\Delta g = 4.2 \times 10^{-4}$, $\delta B_N = 1.3$ mT. The reduction of 
the g-factor dispersion by one order of magnitude can 
be explained by the reduction of the frequency spread 
to a single mode, which is also additionally reduced 
in width by the NIFF. The reduction of the nuclear 
fluctuations $\delta B_N$ for the whole QD ensemble can be 
extracted from the width extrapolated to $B_x = 0$ and 
gives a factor of 5.8, which is comparable to the value of 
12 achieved in optimal conditions for a single QD, using 
the coherent population trapping technique\textsuperscript{12}. Note, 
that our experiment demonstrates a higher reduction 
than the factor 2.5 suggested by our model calculation, 
see Fig. 2f, which, taking into account the simplicity of 
the model, is a good estimate.

Conclusions and outlook

The 1 GHz laser repetition frequency used in this 
study allows us to explore the electron-nuclear spin 
dynamics for a pure single-mode Larmor spin precession 
in the inhomogeneous ensemble of (In,Ga)As/GaAs 
QDs. This is the first experimental realization of such a 
situation, which allows us to demonstrate the discretization 
of the total magnetic field acting on the electron 
spins. Furthermore, we confirm that at the center of the 
frequency plateaus, the nuclear spin fluctuations become 
reduced without build-up of a dynamic nuclear polariza-
tion, a situation comparable to the coherent population 
trapping experiments performed on single quantum dots. 
The pulsed excitation relaxes the requirement of a strictly 
accurate spectral tuning of the lasers (as 
required for a single QD) and makes this technique more 
universal. Additionally, the suggested two-laser protocol 
opens up a promising way to establish a reduced nuclear 
spin fluctuation surrounding using a high repetition laser 
oscillator, while the lower repetition laser can be used 
for readout and manipulation of a large ensemble of spins.

Methods

Sample. The (In,Ga)As/GaAs QD ensemble was 
grown by molecular beam epitaxy on a (100)-oriented 
GaAs substrate. Adjacent sheets in the 20 QD layers 
are separated by 60 nm wide GaAs barriers. Resident 
electrons are provided by a $\delta$ doping layer of Silicon 
placed 16 nm above each layer. The sample is thermally 
annealed at a temperature of 945 °C for 30 seconds to 
homogenize the QD size distribution and to shift the 
average transition energy to 1.39 eV.

Setup. The electron spin polarization is measured at a 
sample temperature of $T = 5.3$ K using pump-probe spec-
troscopy in an external magnetic field $B_z$ applied perpen-
dicular to the light propagation (Voigt geometry). Two 
lasers are used. The first one is a Ti:Sapphire laser with 
a pulse duration of 2 ps, a spectral full width at half max-
imum (FWHM) of 0.9 meV at, and a pulse repetition fre-
quency of 75.76 MHz (repetition period of 13.2 ns). 
The second laser is a Ti:Sapphire laser with a pulse duration 
of 150 fs and a repetition frequency of 1 GHz (repetition 
period of 1 ns). The pulses of the 1 GHz laser are spec-
trally shaped using two sets of holographic gratings and 
slits (one set for pump and one for probe) to reach about 
1.5 nm FWHM (duration of 1.5 ps). The gratings enable 
us to introduce an energy detuning between the pump 
and probe beams. Both lasers are not synchronized or 
phase-locked.

The time-resolved measurements are enabled by me-
dechanical delay lines. To reduce the impact of scattered 
light, a double modulation scheme is used. The pump is 
helicity-modulated between left- and right-circular polar-
ization using a photo-elastic modulator with a frequency 
of 84 kHz. The probe is intensity modulated with a fre-
quency of 100 kHz while being vertically polarized. 
The signal is measured by a lock-in amplifier using the dif-
ference frequency of 16 kHz as a reference. The pump 
beams of both lasers are sent through the same lens and are fo-
cused to a spot diameter of 50 µm. The probe beams are 
focused to 40 µm spots. In this way, approximately 
$5 \times 10^5$ QDs are excited at the same time. The Far-
day rotation of the probe beam is proportional to the 
electron spin projection along the light propagation di-
rection and is measured using an optical bridge consisting
of a Wollaston prism to separate the linear polarizations and Si-based balanced photo diodes.

For the extended version of the pump-probe experiment, we use the 1 GHz laser, where the pump and probe pulses are picked by electro-optical modulators (EOM), hitting the sample in bunches. The pump and probe bunches are separated by an electronically controlled delay. As the devices used here are not fast enough to have a high extinction ratio between neighboring pulses within a nanosecond, the rising and falling edges of the bunches have varying pulse amplitudes within about 6 ns. This time sets a limit on the time resolution of the extended pump-probe and makes it not usable for decay times shorter than 6 ns. The pump and probe pulses stay synchronized to each other and the varying phase of the EOMs relative to the laser repetition frequency mainly add an additional exponential decay proportional to the falling edge of the EOMs used to select the pulse bunches. Here we use 130 pump pulses and six probe pulses for the corresponding bunches. This pump-probe sequence is repeated with a period of 516 ns.

**Data Availability**

The data that support the findings of this study are available from the corresponding author upon reasonable request.
Zhukov, E. A. et al. Spin inertia of resident and photoexcited carriers in singly charged quantum dots. *Phys. Rev. B* **98**, 121304 (2018).

**Acknowledgements**
We are grateful to V.L. Korenev for valuable discussions. We acknowledge the financial support by the Deutsche Forschungsgemeinschaft in the frame of the International Collaborative Research Center TRR 160 (Project A1) and the Russian Foundation for Basic Research (Grant No. 19-52-12059). A.G. acknowledges support by the BMBF-project Q.Link.X (Contract No. 16KIS0857). We acknowledge the supply of the quantum dot sample by D. Reuter and A. D. Wieck. The AFM figure was provided by Claudia Bock, Ruhr-Universität Bochum.

**Author Contributions**
E.E. and A.G. conceived the experiment. E.E. and N.E.K. carried out the experiment and took the experimental data. E.E., N.E.K., and A.G. analyzed the experimental data. N.E.K. and I.A.Yu. conceived the theoretical model. E.E., N.E.K., I.A.Yu., D.R.Ya., M.B., and A.G. wrote the manuscript.

**Competing Interests**
The authors declare no competing interests.
Supplementary materials: Suppression of nuclear spin fluctuations in an ensemble of (In,Ga)As/GaAs quantum dots excited with a GHz-pulsed laser

E. Evers,1,* N. E. Kopteva,1 I. A. Yugova,2 D. R. Yakovlev,1,3 M. Bayer,1,3 and A. Greilich1

1 Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany
2 Physical Faculty of St. Petersburg State University, 198504 St. Petersburg, Russia
3 Ioffe Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

I. SUPPLEMENTARY NOTE 1: EXTENDED PUMP-PROBE

As discussed in the main text and in the Supplementary Note 5, the feedback strength of the electron-nuclear interaction is maximum at the center of the precession frequency plateaus, which should lead to reduced nuclear spin fluctuations and, therefore, to an extension of the spin dephasing time $T_2^\ast$. In contrast, in between the modes at the jumps between plateaus, the feedback strength is strongly reduced, which should lead to a strong influence of the fluctuating nuclear field and a reduced spin dephasing time. To support this observation, we measured the $T_2^\ast$ using an extended pump-probe scheme. In this case we use only the 1 GHz laser, where the pump and probe pulses are picked by electro-optical modulators and hit the sample by a trains of pulses. The pump and probe trains are separated by an electronically-controlled delay.

The extended dynamics are shown in Supplementary Fig. 1 for an external field corresponding to $10 B_0$ (on a mode, panel a) and to $10.5 B_0$ (in between modes, panel b). Note the different time scales in panels a and b. On the mode, the electron spin polarization exhibits a $T_2^\ast$ time of 17 ns, which coincides also well with the time presented in the main text in Fig. 3d for the 1 T case with both lasers applied. In between the modes, the amplitude is strongly reduced (about ten times) and is close to the noise level, which hints towards a much smaller $T_2^\ast$ time, as the accumulated spin amplitude depends directly on it.

II. SUPPLEMENTARY NOTE 2: ALTERNATIVE DRIVING AND READOUT

Supplementary Figure 2 shows the pump-probe signal for the following measurement: the magnetic field is fixed close to the center of a plateau at 2.3 T.

The 1 GHz laser is switched on for 15 s, then switched off and within 0.3 s the sample is kept in darkness. Then the pump and probe from the 75.76 MHz laser are switched on and the measurement for 10 steps (with 100 ns of lock-in integration for each step) of the delay line is done, which takes about 1.5 s. Then the laser is switched off, and after 0.3 s of darkness the 1 GHz laser is switched back on for 15 s. This cycle is repeated for the full pump-probe picture. An elongated dephasing time ($T_2^\ast = 5.6$ ns) is observed. However, accumulation of this time trace took about 3 hours. Note, that the dephasing time is slightly shorter than with the 1 GHz laser switched on all the time ($T_2^\ast = 8$ ns), but much longer than without it ($T_2^\ast = 1.2$ ns), see the main text to Fig. 3d. The timescale of the 1 GHz-laser influence discovered in our experiment (several seconds) is puzzling if compared with the several milliseconds of fluctuation reduction timescales in the Ref. [2]. However, it is well supported by the Ref. [3], where the variation of $T_2^\ast$ is happening on the timescale of nuclear spin diffusion, which is on the order of several seconds under laser illumination in our case. These statements require additional investigations. Making the measurement interval

Supplementary Figure 1. Long time dynamics of the electron spin polarization compared between the magnetic field on a mode a, and in between modes b. On a mode, the electron spin polarization is amplified and decays within $T_2^\ast = 17$ ns. In between modes the amplitude is tenfold lower and decays fast below the noise limit. Note that panels a and b cover different time scales. $E_{Pu} = 1.3878$ eV, $E_{Pr} = 1.3864$ eV.
Supplementary Figure 2. Alternating switching between the 1 GHz and 75.76 MHz lasers, the 1 GHz laser is used to polarize the system at one mode, while the 75.76 MHz laser is blocked. The measurement is done by the 75.76 MHz laser, while the 1 GHz laser is blocked. All lasers are degenerate at $E_{P_{\text{pump}}}/\nu_r = 1.3867$ eV.

for the 75.76 MHz longer or the polarization time for the 1 GHz laser shorter resulted in a reduction of the signal. Such a measurement demonstrates the implementation of the possibility to use the 1 GHz laser for a preparation of the nuclear system for a subsequent measurement with another laser system for about 1 s.

III. SUPPLEMENTARY NOTE 3:
DECOMPOSITION OF THE TIME-TRACES FOR TWO-LASERS PROTOCOL

The supplementary Figure 3 demonstrates pump-probe traces measured at the plateau centers for different magnetic fields. The dephasing times of the extended single-mode time dynamics are extracted from the signals taking into account that both lasers can contribute to the signal, see the exemplary decomposition of the signal for $B_x = 3.08$ T in Supplementary Fig. 3b. One can clearly separate three contributions: the non-oscillating trion decay, which disappears within 0.4 ns; the multimode component with a Gaussian decay originating from the QDs only excited by the 75.76 MHz laser (red); and the slowly decaying single mode component created by the common action of both lasers (black). The dephasing times for this last component are presented in the main text in Fig. 3d.

Additionally, it is clearly seen that above $B_x = 5$ T an additional beating structure starts to appear, signaling that the spread of the frequencies for the 1 GHz laser is increased above the mode separation, see Supplementary Note 4 for more details.

IV. SUPPLEMENTARY NOTE 4:
SINGLE-MODE AND PLATEAU-SIZE LIMITS

Here we discuss the factors limiting the single-mode condition and the maximal length of the plateaus for the QDs studied in this paper. At high repetition frequencies (small repetition periods) the limitation is given by the trion recombination time, $\tau_r$. As is known from Ref. [4], the degree of the electron spin polarization under pulsed excitation depends on the recombination of the trion state. For $T_R < \tau_r$, the efficiency of the spin initialization becomes strongly reduced. Therefore, one can set the qualitative limit for $T_R$ at $3\tau_r$ at which the trion decays by about 95% on average. In the studied sample $\tau_r = 0.4$ ns, so that $3\tau_r = 1.2$ ns. For $T_R = 1$ ns we are close to this condition. This limitation can be reduced, by placing the QDs into an optical microcavity and using the Purcell effect to enhance the spontaneous emission and shorten $\tau_r$.

Furthermore, in the case of $\Delta g = 0$, the limitation for the low repetition frequency side is set by the nuclear fluctuations $\delta B_N$. The mode distance in magnetic field is defined as $B_0 = h\omega_R/(g\mu_B)$, with $\omega_R = 2\pi/T_R$. The nuclear field fluctuation is defined as the HWHM. So to be able to reach the single mode regime, one has to overcome at least $3\delta B_N$ (a half of $B_0$). Therefore, $T_R < 2\pi h/(g\mu_B 3\delta B_N) = 5.6$ ns. It is useful to remind here, that the fluctuating nuclear field is proportional to $1/\sqrt{N}$, with $N$ being the number of the nuclear spins in the QD. This means that for bigger QDs, the fluctuating field is reduced and the limit set on $T_R$ can be relaxed, a slower repetition laser can be used. An additional effect of the bigger QD size is the acceleration of the trion recombination ($\tau_r$ becomes smaller), which is preferential for smaller $T_R$. Bringing both limits together, for a robust single-mode regime with highest electron spin po-
larization, the repetition period of the laser should be in the range 1.2 ns < T_R < 5.6 ns.

In the previous paragraph we have assumed that Δg = 0. If it is nonzero, this parameter sets an additional limitation on the applied external magnetic field, as the spread of frequencies is linearly increasing with field strength. For Δg = 0.004 and T_R = 1 ns, M = 2 starts to become possible at B_x > 5.9 T, see Eqs. (1) and (2) of the main text. This conclusion is supported by the observation of a beating in the trace in Supplementary Fig. 3a, recorded for B_x = 6.16 T. Note that once the number of possible modes M is increased above 2, the dephasing is defined by Δg. This is the case for the 75.76 MHz laser, presented by the black circles in Fig. 3d. However, if the mode separation is only allowing one mode, the spread Δg is effectively reduced by the effect of NIFF. Furthermore, Δg can be selected by the spectral width of the laser, as described in the experimental details, or by choosing another growth technique for the QDs. For example, the Δg can be strongly reduced using QDs grown by infilling of droplet-etched nanoholes or many-electron GaAs/(Al,Ga)As QDs, which avoids the use of Indium atoms that affects the g-factor values strongly.

Finally, for any given T_R the maximal length of the plateaus, limited from top by the distance between the modes B_0 = 2πℏ/(gμ_B T_R), is determined by the maximal Overhauser field B_{N,max}, see Supplementary Fig. 4 and the following Supplementary Note 5. It is dependent on the types of constituting nuclear species, the leakage factor f_N, and the electron spin polarization ⟨S_x⟩ along the direction of B_x. The ⟨S_x⟩ in turn depends on the optical detuning between the pump and probe pulses Δ, the optical pump power Θ, the electron spin coherence time T_2, and T_R⁹, as demonstrated in Supplementary Fig. 4.

Supplementary Figure 4a shows a color map calculated for the optical pulse power of Θ = π in dependence on the optical detuning and the ratio T_2/T_R. In calculations the spin coherence time was assumed homogeneous, so the resident electron spin lifetime is equal to the spin coherence time. In our case T_R = 1 ns and the coherence time of the electron spins T_2 for these dots is about 3 μs, which makes the relation of T_2/T_R = 200 a secure underestimation. The dependence of ⟨S_x⟩ for several detunings is demonstrated additionally in the Supplementary Fig. 4b. The amplitude saturates at T_2/T_R close to 200 and the maximal amplitude is reached at a detuning of |Δ| = 0.6. These dependencies show that variation of the detuning has a strong impact on the value of ⟨S_x⟩, and therefore on the maximal Overhauser field B_{N,max}. So, the maximal value of B_{N,max} will be reached at |Δ| = 0.6. Finally, the Supplementary Fig. 4c demonstrates the power dependence of ⟨S_x⟩ for |Δ| = 0.6. The maximum of the nuclear polarization should be observed for Θ = π. This panel also shows that the maximal ⟨S_x⟩ is strongly influenced by the spin coherence time T_2. For strongly reduced T_2 values, the nuclear field is decreasing and so should the plateau length.

V. SUPPLEMENTARY NOTE 5: SIMULATION OF THE PLATEAU BEHAVIOR

The feedback strength in the electron-nuclear spin system can be described by the parameter λ₁⁰:

$$\lambda = \frac{T_{1e} + T_d}{T_{1e} T_d} \left(1 - \frac{A f_N Q_{av} \delta \langle S_x \rangle}{\hbar} \right),$$  
(31)

where T_{1e} is the nuclear spin polarization time via the electron spin polarization. T_d is the nuclear spin-lattice relaxation time. A is the average hyperfine constant. f_N is the leakage factor. Q_{av} = \sum_{i=3}^S A_i (I_i + 1) n_{QD,i}/3 is the factor dependent on the nuclear spin (I_i) averaged over all nuclei species with spin (I_i) and fraction n_{QD,i} for elementary cell with 2 atoms. Q_{av} = 20 for (In,Ga)As
QDs with 35% of In concentration. $\omega_N$ is the Larmor precession frequency in the Overhauser field.

One can use following equation for $T_{1e}$ Ref. [11]:

$$T_{1e} = \left( \frac{\hbar N}{A} \right)^2 \frac{1 + \omega_N^2 + \delta_C^2}{2F \tau_C}. \quad (S2)$$

$\tau_C$ is the correlation time in the electron-nuclear spin system. The factor $F$ represents the average fraction of time during which the dot is occupied. We assume that $F = 1$ for a resident electron.

One can write the second term of Eq. (S1) analytically using the terms $L$ and $M$ introduced in Ref. [9]:

$$\frac{\delta S_z}{\delta \omega_N} = S_I \left( \frac{1 + LM}{} \cos(\omega_N T_R) - \frac{LM}{1 + LM - (L + M) \cos(\omega_N T_R)} \right)^2, \quad (S3)$$

$$S_I = \frac{T_2(1 - Q^2)K}{4} \left[ 1 - \exp(-T_R/T_2/4) \right]. \quad (S4)$$

The functions $K$ and $Q$ are also described in Ref. [9].

According to the supplementary material of Ref. [12] the spin dephasing time is defined by the characteristic square of the nuclear spin fluctuations $\langle \delta I^2 \rangle$:

$$\langle \delta I^2 \rangle = \frac{I(f + 1)N|\lambda_0|}{|\lambda|^2}, \quad (S5)$$

where $\lambda_0 = \lambda(\omega_N = 0)$ is given by the nuclear depolarization time via the electron spin. The spin dephasing time ($T^*_2$) can be estimated using the approach from Refs. [11 and 13]:

$$T^*_2 = \frac{\hbar N}{A \sqrt{|\langle \delta I^2 \rangle |}}. \quad (S6)$$

The simulations in the main text are done using the equations given in Refs. [9 and 12] with the following parameters: $\Delta = -0.55$, $N = 5.5 \times 10^5$, the electron spin coherence time $T_2 = 2\mu$s, $\tau_C = 10\, \text{ns}$, $T_d = 2\, \text{min}$, $\Theta = \pi$, the electron $g$-factor $g = -0.57$, $Q_{av} = 20$, $f_{N0} = 0.86$, $A = 49.2\, \text{meV}$.

---

* e-mail: eiko.evers@tu-dortmund.de

1. Belykh, V. V. et al. Extended pump-probe faraday rotation spectroscopy of the submicrosecond electron spin dynamics in n-type GaAs. Phys. Rev. B 94, 241202 (2016).

2. Ethier-Majcher, G. et al. Improving a solid-state qubit through an engineered mesoscopic environment. Phys. Rev. Lett. 119, 130503 (2017).

3. Onur, A. R. & van der Wal, C. H. Two-laser dynamic nuclear polarization with semiconductor electrons: Feedback, suppressed fluctuations, and bistability near two-photon resonance. Phys. Rev. B 98, 165304 (2018).

4. Greilich, A. et al. Optical control of spin coherence in singly charged (In, Ga)As/GaAs quantum dots. Phys. Rev. Lett. 96, 227401 (2006).

5. Vuckovic, J., Fattal, D., Santori, C., Solomon, G. S. & Yamamoto, Y. Enhanced single-photon emission from a quantum dot in a micropost microcavity. Applied Physics Letters 82, 3596–3598 (2003).

6. Greilich, A. et al. Tailored quantum dots for entangled photon pair creation. Phys. Rev. B 73, 045323 (2006).

7. Lobl, M. C. et al. Correlations between optical properties and voronoi-cell area of quantum dots. Phys. Rev. B 100, 155402 (2019).

8. Markmann, S., Reichl, C., Wegscheider, W. & Salis, G. Universal nuclear focusing of confined electron spins. Nature Communications 10, 1097 (2019).

9. Kopteva, N. E. et al. Theoretical modeling of the nuclear-field induced tuning of the electron spin precession for localized spins. Physica Status Solidi (b) 256, 1800534 (2019).

10. Korenev, V. L. Multiple stable states of a periodically driven electron spin in a quantum dot using circularly polarized light. Phys. Rev. B 83, 235429 (2011).

11. Glazov, M. M. Electron and Nuclear Spin Dynamics in Semiconductor Nanostructures (Oxford University Press, Oxford, 2018).

12. Zhukov, E. A. et al. Discretization of the total magnetic field by the nuclear spin bath in fluorine-doped ZnSe. Nature Communications 9, 1941 (2018).

13. Merkulov, I. A., Efros, Al. L. & Rosen, M. Electron spin relaxation by nuclei in semiconductor quantum dots. Phys. Rev. B 65, 205309 (2002).