Hyperfine-phonon spin relaxation in a single-electron GaAs quantum dot

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Understanding and control of the spin relaxation time $T_1$ is among the key challenges for spin based qubits [1–3]. A larger $T_1$ is generally favored, setting the fundamental upper limit to the qubit coherence and spin readout fidelity [4–6]. In GaAs quantum dots at low temperatures and high in-plane magnetic fields $B$, the spin relaxation relies on phonon emission and spin-orbit coupling [7–9]. The characteristic dependence $T_1 \propto B^{-5}$ [10] and pronounced $B$-field anisotropy [11] were already confirmed experimentally. However, it has also been predicted 15 years ago that at low enough fields, the spin-orbit interaction is replaced by the coupling to the nuclear spins, where the relaxation becomes isotropic, and the scaling changes to $T_1 \propto B^{-3}$ [12]. We establish these predictions experimentally, by measuring $T_1$ over an unprecedented range of magnetic fields – made possible by lower temperature [13–15] – and report a maximum $T_1 = 57 \pm 15$ s at the lowest fields, setting a new record for the electron spin lifetime in a nanostructure [5, 10, 16].

The decay of the energy stored in the qubit defines the relaxation time $T_1$. In qubits based on electronic spins, it corresponds to the relaxation of spin – a longstanding topic of research in semiconductors. The suppression of this process in a confined system compared to the bulk [7] makes quantum dot spin qubits a serious candidate for a quantum technology platform [1–3]. For spin qubits, the energy splitting is due to the Zeeman term of an applied magnetic field $B$. The requirement for a sizable splitting, necessary for many of the protocols to initialize, measure, or manipulate spin qubits [17–20], then imposes limitations on $T_1$, which in turn might influence these protocols in a profound way. This further motivates investigations of mechanisms and fundamental limits of the spin relaxation in quantum dots.

To understand this process in a GaAs quantum dot spin qubit, one needs to consider that it involves the dissipation of both energy and angular momentum, i.e. spin. The former proceeds by emission of a phonon. Considering, for simplicity, long-wavelength three-dimensional bulk phonons, one gets the spin relaxation rate $W \equiv T_1^{-1} \propto B^4 d^2$ for piezoelectric and $W \propto B^5 d^2$ for deformation potential phonons, where $d$ is the dipole moment matrix element between the initial and final state of the transition. For typical Zeeman energies, piezoelectric phonons dominate. Since the initial and final states are opposite in spin, a nonzero dipole element can only arise due to some spin-dependent interaction. In GaAs, the two most relevant ones are the spin-orbit and hyperfine interactions. Their essential difference here is their opposite behaviour under reversal of time. While the hyperfine (HF) interaction induces a $B$-independent moment, the time reversal symmetry of the spin-orbit interaction (SOI) results, through the Van-Vleck cancellation, in an additional magnetic field proportionality, $d^2 \propto B^2$. Putting these pieces together, the SOI, with $W \propto B^5$, will dominate at high fields, and HF, with $W \propto B^3$, at low. For the parameters of typical surface gate defined GaAs dots, the crossover is predicted at around $1–2$ Tesla.

Beyond field scaling, the SOI with competing Rashba and Dresselhaus terms results in a strong dependence of spin relaxation on the direction of the applied magnetic field in the plane of the 2D gas – the spin relaxation anisotropy [8,9,11]. The HF mechanism, on the other hand, is isotropic [12], even for a dot shape which breaks circular symmetry. These two hallmark features together – isotropic behavior and $B^3$ scaling – constitute a unique fingerprint of the HF relaxation mechanism. Note that this channel is fundamentally different from the elastic electron-nuclear spin flip-flop, which is strongly suppressed due to the pronounced mismatch of the electron and nuclear Zeeman energy for fields above a few mT [21].

Even though the HF assisted mechanism of spin relaxation was predicted early on [12], experimental observation has remained elusive so far for a number of reasons: rather low fields below 1 Tesla are required to reach the HF limit. For a spin doublet, only energy selective spin-readout is available, thus requiring rather low electron temperatures below 100 mK to keep the Zeeman splitting well above the thermal broadening. To check for the direction dependence of relaxation, suitable in situ control over the applied field direction is required, but this has only recently become available. Finally, very long $T_1$-times far exceeding 1 s are predicted at such low fields,
posing a formidable challenge on the long-term stability and control of a semiconductor nanostructure.

Here, we overcome these difficulties by employing a very stable 2D gas and implementing active feedback procedures to keep the energy levels aligned with sub-microvolt precision over days (see Supplementary Section S2). Specially developed Ag-epoxy filters [14] provide an electron temperature of $\sim 60 \text{ mK}$ – more than a factor of two lower than before [10]. We use a flexible gate layout (see Fig. 1a) to shape a nearly circularly symmetric dot and set up a cryogenic piezo-rotator to apply almost perfectly aligned in-plane fields (see Supplementary Section S1) up to 14 T with arbitrary angle $\phi$. When the sample is rotated by 90°, the states’ roles swap and the blue state becomes invariant, as expected, see Fig. 1b. Such striking observations, including further B-directions, can be modeled by an anisotropic harmonic oscillator [22, 23], which confirms the quantum dot main axes are well aligned with the $\hat{x}$- and $\hat{y}$-directions. This essential information about the dot orbitals makes possible a detailed understanding of all measurements, reproducing the measured $T_1$ quantitatively by numerics using a single set of parameters without phenomenological constants (see Methods for details).

With a full orbital model at hand, we now turn to spin relaxation measurements, done by cycling the dot through an ionization, charge and relax, and read-out configuration, as depicted in Fig. 2a. Averaging over many thousand cycles, we obtain the spin excited state
Spin relaxation rate $W$ (Hz) as a function of the waiting time $t_w$. Third, spin-charge conversion is used in “readout” to detect the spin state: the spin-down electron only will tunnel off the dot, which is detected by the charge sensor. The spin relaxation rate $W$ is extracted from the dependence of the probability $P_e$ to find the spin in the excited (down) state as a function of $t_w$, shown in b for a magnetic field of 4 T applied along different angles $\phi$ as indicated. Markers show measurements with statistical error bars, curves are fits to the formula $P_e(t_w) \propto \exp(-Wt_w - \exp(-\Gamma_{in}t_w))/(\Gamma_{in} - W)$, where the tunneling-in rate $\Gamma_{in}$ is determined independently (see Supplementary Section S3). $W$ is thus extracted as the only fit parameter.

probability $P_e$ as a function of the waiting time $t_w$ – the time the electron was given to relax into the spin ground state. A few examples are plotted over four orders of magnitude in $t_w$ on Fig. 2a at a magnetic field of 4 T. All such curves fit very well to the sum of two exponentials, from which we reliably extract the spin relaxation rate $W \equiv T_1^{-1}$ (see Supplementary Section S3 for more details). A pronounced dependence of $W$ on the direction of the magnetic field is observed, as displayed in Fig. 3a as a function of the field angle $\phi$. A modulation of $W$ by a factor of $\sim 16$ is found, with minimal relaxation rate along the $\hat{y}$-direction.

This pronounced anisotropy is rooted in a combination of the dot shape asymmetry and the interference of the Rashba and Dresselhaus SOI terms. The latter can qualitatively be understood from the dependence of the total effective spin-orbit magnetic field on the direction of the electron momentum (Supplementary Section S4). First derived for symmetric quantum dots [8], the spin relaxation anisotropy due to the dot shape asymmetry was also soon included in a theoretical generalization [9]. The shape-induced contribution to the anisotropy of $W$ is well known here from the orbital spectroscopy and found to be small. Thus, the anisotropy here is largely due to the SOI, and given the precisely measured orbital energies, it is possible to extract the SOI coupling strengths by fitting the model (see Methods for details). The best fit delivers a ratio $\alpha/\beta \sim 1.6$ and a spin-orbit length $l_{so} \approx 2.1 \mu m$ setting the overall strength of the SOI. These values are well in-line with previous reports for GaAs structures [10, 24, 25]. We note that $\alpha$ and $\beta$ are found to have the same sign for the 2D material used. Without knowledge of the orbital energies, the SOI parameters cannot be directly determined from $T_1$ [11, 19, 26].

An extremely long $T_1$ time can be achieved by reducing the magnetic field strength and orienting the magnetic field along the crystalline axis with minimal SOI field. Therefore, we carried out the same anisotropy measurements at 1.25 T. Indeed, $T_1$ times longer than 1 s are obtained. Interestingly, in contrast to the measurements at 4 T, around the $\hat{y}$-direction with minimal $W$, the measured spin relaxation rate $W$ (black markers) is seen to be almost a factor of three larger than the calculated SOI rate (red curve, Fig. 3b). This is far beyond the error bars, and indicates an additional spin relaxation channel beyond SOI-mediated phonon emission.
To substantiate this claim, we measure the field magnitude dependence of \( W \). In Fig. 4a we compare two sets, for the magnetic field along the \( \hat{x} \) and \( \hat{y} \)-direction, where the effects of the nuclei with respect to SOI are, respectively, maximal and minimal. The two curves indeed show pronounced differences. With the field along the \( \hat{x} \)-direction, the relaxation follows the \( B^5 \) scaling quite well over the entire range of the measured magnetic fields. Thus, for the \( \hat{x} \)-direction, the relaxation is dominated by the SOI for the full field range. In contrast, for fields along \( \hat{y} \), there is a clear crossover around 2 T with a change of the power law scaling from roughly \( B^3 \) at high fields to \( B^3 \) at low fields, corresponding to a crossover from SOI to HF dominated relaxation. We note in passing a \( T_1 \) time of \( 57 \pm 15 \, \text{s} \) for a magnetic field of 0.6-0.7 T along \( \hat{y} \). To our knowledge, this is the longest \( T_1 \) time reported to date in a nanoelectronic device \[5, 10, 10\].

Note that the simple observation of a change in the power law scaling of \( W \propto B^3 \) is not a sufficient as a proof of its HF origin. It could be that the phonons as an energy dissipation channel are replaced by another bath, e.g. charge noise or an ohmic bath also leading to a \( B^3 \) dependence \[27-29\]. The absence of deviations in the scaling of the \( B|\hat{x} \) data indicates that phonons are responsible for the energy dissipation throughout and the crossover in the \( \hat{y} \) data is not related to a specific value of \( W \). Also, if the SOI remained as the mixing mechanism and the dissipation channel instead were to change, then the spin relaxation anisotropy, quantified by the ratio \( W_X/W_Y \), would also prevail and remain large at low fields. However, as shown in Fig. 4b, the anisotropy is seen to decrease from about 16 at high fields down to about one at low fields well below 1 T. This behavior displays spin relaxation with equal speed in both principal directions, thus indicating isotropic relaxation at low fields. Together with the \( W \propto B^5 \) scaling, these observations constitute unequivocal demonstration of HF-mediated spin relaxation.

In conclusion, we have demonstrated a spin relaxation time of up to \( 57 \pm 15 \, \text{s} \) limited by HF-phonon spin relaxation in a single electron lateral GaAs quantum dot, exhibiting a \( \propto B^3 \) field scaling together with isotropic relaxation at low fields well below 1 T. At larger fields, the spin relaxation becomes strongly anisotropic, with \( W_X/W_Y \sim 16 \), and the \( B \)-field scaling follows a \( W \propto B^5 \) law. Using excited state spectroscopy, we determine the dot orbital energies, can extract the Rashba and linear Dresselhaus parameters from the \( B \)-field anisotropy of \( W \), and simulate the HF induced spin relaxation \( W \), in very good agreement with the experiment, all with a single consistent set of parameters. While ramping the magnetic field from 0.6 T to about 10 T, the spin relaxation rate changes by a striking 6 orders of magnitude. Yet this is very well captured by the theory throughout the entire range – putting the model to a very stringent test. With the SOI parameters at hand, it also becomes easy to max-

**FIG. 4.** Hyperfine induced spin relaxation. a Spin relaxation rate \( W \) for an in-plane magnetic field along the \( \hat{x} \)-direction (green, along \([110]\)) and the \( \hat{y} \)-direction (blue, along \([1\hat{T}0]\)) as a function of the magnetic field magnitude. The data are shown as triangles with error bars. Numerics considering various terms are shown as labeled. The pure \( B^5 \) scaling (red dash) and \( B^3 \) scaling (orange dash) are also given as a guide to the eye. b The relaxation anisotropy \( W_X/W_Y \) as a function of field magnitude. Experiment is shown as triangles with error bars, numerics with both SOI and HF as a solid curve, showing the transition to isotropic relaxation at low fields. Red dashed line is SOI theory only, orange dash at 0.1 magnitudes is SOI + HF, +HF = 10\) is the isotropic HF theory.

Because the dot orbitals are accurately determined, the HF contribution can quite precisely be calculated by numerics (see Methods). As shown in Fig. 3h, at \( B = 4 \) T the microscopic model predicts that the HF contribution (orange curve) is 1 to 2 orders of magnitude smaller than the one due to the SOI (red curve), and is therefore not observable experimentally. In comparison, at \( B = 1.25 \) T, as shown in Fig. 3h, the SOI model alone is unable to explain the data, but fits very well when the nuclei are included (purple curve), particularly now capturing the minimum close to the \( \hat{y} \)-direction very well. Backed by numerics, we thus conclude that this seemingly subtle feature in the angular modulation of \( W \) actually constitutes the first evidence of the HF relaxation mechanism.
imize the electric dipole spin resonance Rabi frequencies \[30\] in future experiments by selecting the strongest SOI direction, thus potentially facilitating a large improvement over previous experiments \[19, 31\].

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AUTHOR CONTRIBUTIONS

L.C.C, L.Y., P.S. and D.M.Z. designed the experiments, analysed the data and wrote the paper. L.C.C. and L.Y. processed the samples and performed the experiments. J.Z. and A.C.G. carried out the molecular beam epitaxy growth of the heterostructure. P.S. and D.L. developed and carried out the theoretical work and numerical modeling. All authors discussed the results and commented on the manuscript.

MATERIALS AND METHODS

Sample and measurement
The measurement was performed on a surface gate defined single electron quantum dot formed in a GaAs 2D electron gas. The device was fabricated on a GaAs crystal, grown along the [001] crystal direction, with a GaAs/AlGaAs single heterojunction located 110 nm below the surface with density \(2.6 \times 10^{13} \text{cm}^{-2}\) and mobility \(4 \times 10^{5} \text{cm}^{2}/\text{Vs}\). The layout of the surface gates (see Fig. 1a) is modified from that in \[10\], and allows effective control of the shape of the dot. Negative gate voltages were applied on the nanogates to locally deplete the 2D gas and form a quantum dot in the center of the device (blue ellipse in Fig. 1a) and the adjacent charge sensor quantum dot (black dashed circle). The main dot is tuned to the single electron regime and tunnel coupled only to its left lead.

The single electron quantum dot is capacitively coupled to the charge sensor, the conductance through which changes sharply by 50-100\% when adding or removing an electron to the main dot. Real time detection of the dot charge state was realized by monitoring sensor dot current with a measurement bandwidth of 30kHz obtained with a specially designed current preamplifier (Basel Electronics Lab) capable of handling capacitive input loads as appearing from the microwave filtering. The main dot is electrically extremely stable due to excellent 2D gas material and allows control of the dot energy levels using a level positioning algorithm (Supplementary Section S2) for an extended period of time, which is crucial for long spin relaxation measurements. This feedback technique was regularly carried out throughout the spin relaxation measurements to compensate drift of the dot energy level with respect to chemical potential of the lead. Additionally a feedback to compensate the drift of the sensor dot conductance was also performed regularly. Electron exchange processes with the reservoir \[13\] occurring during the charge and relax pulse step for long waiting times \(t_w\) are detected by continuously monitoring the dot charge state and are removed from the data sets. This becomes an important factor particularly at low fields.

Lots of efforts have gone into operating a low electron temperatures \[13, 15, 32, 35\]. The base temperature of the dilution refrigerator is \(T_{\text{base}} \approx 25 \text{mK}\) and the electron temperature is \(T_{\text{el}} \approx 60 \text{mK}\), measured by probing the Fermi-Dirac distribution of the coupled lead. By heating to 300 mK where \(T_{\text{el}} \approx T_{\text{base}}\), the Fermi-Dirac distribution method was also used to quantify the gate lever-arm. The sample was rotated (Attocube ANRv51 piezoelectric rotator) in a magnetic field up to 14 T applied in the plane of the 2D gas. The out-of-plane magnetic field is determined by standard Hall effect measurements using van der Pauw configurations (Supplementary Section S1). The maximal misalignment of the in-plane magnetic field is 1.3°, thus the effect of the out-of-plane component is negligible \[37\].

The numerical model
A microscopic model is used to describe the dot orbital spectroscopy and spin relaxation data. The implementation is based on an exact diagonalization of the electronic Hamiltonian which includes the kinetic energy with an anisotropic mass, a bi-quadratic (harmonic) confinement potential in the 2D plane, the Zeeman term, the linear and cubic spin-orbit terms, and the Fermi contact HF interaction with nuclear spins. This Hamiltonian is discretized in real space, typically on a grid of 100 by 100 points, with Dirichlet boundary conditions for the wavefunction. The resulting hermitian Hamiltonian matrix is diagonalized by the Arnoldi method using the ARPACK library, to obtain a few lowest eigenstates and the corresponding energies \[38\]. As an example, Fig. 1c,d (solid curves) shows the excitation energies calculated from such an exact spectrum as a function of the field. The spin relaxation rates are calculated by Fermi’s gold rule using the exact spectrum.

The rates denoted as “SOI” in the figures were obtained in the same way, but with the HF interaction excluded from the Hamiltonian. Similarly, the tag ”HF” means that the spin-orbit terms were excluded. The results from such a numerical procedure are expected to have a very high precision \[39, 10\], in the sense of convergence (numerical stability), and also compared to analytical results in cases where the latter are known. As an example, the energies of the Fock-Darwin spectrum for our parameters match the analytical formulas up to errors well below 1 µeV. The errors stemming from the numerical procedures themselves are therefore expected to be completely negligible compared to errors induced by uncertainties of the used parameters, the true confinement shape, or the departures from the assumed simple forms of the spin-orbit, electron-phonon and HF interactions. Whenever the Hamiltonian includes the HF interaction, the given relaxation rate is a geometric average of rates for 1000 configurations of static nuclear spins with random orientations (the approximation of unpolarized nuclei at infinite temperature). More details on the Hamiltonian
and the numerical methods used to solve it are given in the
Supplementary Material.

**Analytical results**

The following formulas reflect the main features of the relaxation
rate important in our experiments. The relaxation rate
due to transverse piezoelectric phonons and nuclear spins is

\[
\Gamma_{HF} \approx \frac{8(eh_{14})^2(I + 1)A^2}{315\pi\hbar^3 mp_e^2 N} \left( \frac{1}{E_2^2} + \frac{1}{E_0^2} \right) (g\mu_B B)^3. \tag{1}
\]

It is isotropic and proportional to \(B^3\). Replacing HF with
spin-orbit effects leads to

\[
\Gamma_{SOI} \approx \frac{(eh_{14})^2}{210\pi m^2 \hbar^2 r_s^2} \left( \frac{1}{E_2^2} + \frac{1}{E_0^2} \right) (g\mu_B B)^3 \times \cos^2 \xi \left(f_1 + f_2\right) + \sin^2 \xi \left(f_3 + f_4\right). \tag{2}
\]

The rate grows as \(B^3\) and is anisotropic, with the angular
dependence described by

\[
\begin{align*}
  \cos \varphi, \\
  \sin \varphi,
\end{align*}
\]

\[
\begin{align*}
  f_1 &= 1 + \sin 2\theta \sin 2\varphi, \\
  f_2 &= \sin 2\theta \sin 2\varphi + \sin 2\delta \sin 2\varphi + \cos 2\delta \cos 2\varphi, \\
  f_3 &= 2, \\
  f_4 &= 2 \sin 2\delta \sin 2\varphi, \\
  \epsilon &= (E_x^{-4} - E_y^{-4})/(E_x^{-4} + E_y^{-4}).
\end{align*}
\]

These formulas are derived in the Supplementary Material,
where their generalized forms, including the effects of finite
temperature, longitudinal phonons, and deformation electron-
phonon potential, are also given.

The parameters in these equations are (values given for
GaAs): piezoelectric potential \(h_{14} = 1.4 \times 10^6 \text{ V/m, nuclear}\)
spin \(I = 3/2,\) Fermi-contact interaction constant \(A = 45\)
\(\mu\text{eV, effective mass} m = 0.067 m_e \) with \(m_e\) the electron
mass in vacuum, material density \(\rho = 5300 \text{ kg/m}^3,\) transverse
acoustic phonon velocity \(c_l = 3350 \text{ m/s, Bohr magneton}\)
\(\mu_B = e/2m_e.\) The number of nuclei in the dot \(N \approx 8.3 \times 10^7,\)
the excitation energies \(E_x = 2.33 \text{ meV, } E_y = 2.61 \text{ meV,}\)
the g-factor \(g = -0.36,\) and the angle of the dot potential
axis with the [100] direction \(\delta \approx 50.6^\circ,\) were fitted from
spectral data such as in Fig. 1. The spin-orbit parameters
\(l_{so} = 2.1 \mu m \) and \(\vartheta = 31^\circ,\) defined by writing the
Rashba and Dresselhaus interaction strengths (see Supplementary
Eq. (11)) as \(\alpha = (h/2ml_{so}) \cos \vartheta,\) and \(\beta = (h/2ml_{so}) \sin \vartheta,\)
were fitted from the T1 data shown in Figs. 3 and 4. Fi-
nally, the magnetic field orientation is parameterized by writing
\(B = B[\cos \xi \cos \varphi, \cos \xi \sin \varphi, \sin \xi],\) referring to crystallo-
graphic coordinates.

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