Experimental signature of phonon-mediated spin relaxation

T. Meunier, I. T. Vink, L. H. Willems van Beveren, K-J. Tielrooij, R. Hanson, F. H. L. Koppens, W. Wegscheider, L. P. Kouwenhoven, and L. M. K. Vandersypen

1Kavli Institute of Nanoscience, Delft University of Technology, PO Box 5046, 2600 GA Delft, The Netherlands
2Institut für Angewandte und Experimentelle Physik, Universität Regensburg, Regensburg, Germany

(Dated: March 23, 2022)

We observe an experimental signature of the role of the phonons in spin relaxation between triplet and singlet states in a two-electron quantum dot. Using both the external magnetic field and the electrostatic confinement potential, we change the singlet-triplet energy splitting from 1.3 meV to zero and observe that the spin relaxation time depends non-monotonously on the energy splitting. A simple theoretical model is derived to capture the underlying physical mechanism. The present experiment confirms that spin-flip energy is dissipated in the phonon bath.

PACS numbers: 03.65.w, 03.67.Mn, 42.50.Dv

Relaxation properties of a quantum system are strongly affected by the reservoir where energy is dissipated. This has been seen clearly for electron spins embedded in nanostructures. Spin relaxation times \( T_1 \) up to a few ns have been observed for free electrons in a two-dimensional electron gas (2DEG) where energy is easily given to the motion \( \hbar \omega \). In quantum dots, the discrete orbital energy level spectrum imposes other energy transfer mechanisms. Near zero magnetic field, the electron spin can directly flip-flop with the surrounding nuclear spins, inducing short \( T_1 \)'s of the order of \( \mu s \). When a small magnetic field is applied, direct spin exchange with nuclei is suppressed. Lattice vibrations, i.e. phonons, are expected to become the dominant reservoir in which spin-flip energy can be dissipated. This dissipative mechanism is inefficient and very long spin relaxation times follow \( T_1 \)'s of the order of \( \mu s \).

For spin relaxation involving phonons, two physical processes are important. The coupling between electron orbitals and phonons is responsible for dissipation of energy and the spin-orbit interaction provides the essential mixing between different spin states so the spin states are coupled through electron-phonon interaction \( \hbar i \varepsilon _{\text{ph}} \). Energy conservation requires that the phonon energy corresponds to the energy separation between the excited and the ground spin state. Changing the energy separation affects the efficiency of the electron spin relaxation in two ways. First, since the phonon density of states increases with energy, the relaxation rate is expected to increase with energy as well. Furthermore, since the electron-phonon interaction is highly dependent on the matching between the size of the dot and the phonon wavelength \( \hbar / \varepsilon _{\text{ph}} \), we expect a suppression of relaxation for very large and for very small phonon wavelengths in comparison to the dot size. Mapping the relaxation time \( T_1 \) as a function of the energy splitting between the two spin states will provide insight in both the electron-phonon interaction and the spin-orbit coupling as well as an understanding of the limitations on \( T_1 \). This is of particular relevance in the context of both spintronic and spin-based quantum information processing devices \[12\].

Here, we study the spin relaxation time from triplet to singlet states for different energy separations in a single quantum dot containing two electrons. Singlet and triplet states have respectively two electrons in the lowest orbital and one electron each in the lowest and in the first excited orbital. In the experiment, the energy splitting \( \Delta E_{ST} \) between these two-electron spin states could be tuned from 0.9 meV to zero with a perpendicular magnetic field and from 0.9 meV to 1.3 meV by deforming the dot potential \[13\].

All the experiments are performed in a dilution refrig-
ical potentials are moved back above the Fermi energy and an electron in the ground state (initialization). During the pulse, the singlet and triplet electrochemical potentials are below the Fermi energy and a second electron tunnels into the dot. Due to the difference in tunnel rates, most likely a triplet state will be formed. We allow relaxation to occur during a waiting time that we vary. After the pulse, both electrochemical potentials are moved back above the Fermi energy and a second electron tunnels into the dot. We extract experimentally the energy splitting $\Delta E_{S,T}$ [6], making $T_1$ close to the degeneracy point, we use a tunnel-rate selective read-out procedure (TRRO) (see Fig. 2). The measured spin relaxation time $T_1$ as a function of $B$ is presented in Fig. 3. The shape of the $T_1$ dependence on magnetic field exhibits a striking non-monotonous behavior. From $0.4$ T to $\sim 2$ T, corresponding to a decrease in the energy splitting from $0.8$ meV to $0.2$ meV, the relaxation time first decreases, reaching a minimum of $180 \, \mu s$. In between $2$ T and the degeneracy point ($2.82$ T), $T_1$ increases whereas the energy splitting continues to decrease.

As a complementary study, we change $\Delta E_{S,T}$ in a different way by controlling the electrostatic potential of the dot via the voltage $V_T$ applied to gate ‘T’ and again look at $T_1$. The dependence of $\Delta E_{S,T}$ on $V_T$ is presented in Fig. 3(b). With this second experimental knob, $\Delta E_{S,T}$ can be varied from $0.8$ meV to $1.3$ meV. We interpret the change in the observed energy splitting as a consequence of a change in the dot ellipticity. A more positive $V_T$ implies a more circular dot and a larger energy splitting. We observe that $T_1$ further increases with $\Delta E_{S,T}$ as $V_T$ is varied at $B = 0$ T (see the inset of Fig. 3). The maximum energy splitting reached at $-530$ mV, $1.3$ meV, corresponds to a maximum of $T_1 = 2.3$ ms. With both experimental knobs, we observe that when $\Delta E_{S,T}$ is constant, $T_1$ is constant too (respectively for $V_T < -650$ mV and $B < 0.4$ T). These observations clearly indicate that the most important parameter for the variation in the triplet-singlet relaxation time is their energy separation.

The observed minimum in $T_1$ is precisely what one would expect for energy relaxation mediated by the electron-phonon interaction [8, 11]. Indeed, the energy splitting $\Delta E_{S,T}$ determines the relevant acoustic phonon energy (acoustic phonons are the only available phonons for the explored energy range). At $B \sim 2$ T, $\Delta E_{S,T} \sim 0.3$ meV, the associated half-wavelength, approximately $30$ nm (the group velocity for acoustic phonons $c_2 \sim 4000$ m/s), is comparable to the expected size of the dot and therefore the coupling of the electrons in the dot to phonons is strongest. For energy separations smaller (larger) than $0.3$ meV, the phonon wavelength is larger (smaller) than the size of the dot, the coupling to the orbitals becomes smaller and $T_1$ increases. Taken together, all these observations strongly suggest that the phonon bath is the dominant reservoir for dissipating the spin-flip energy during relaxation.

In order to get more insight in the role of the phonon wavelength, we present a simplified model of the energy relaxation process between triplet and singlet as a function of their energy splitting $\Delta E_{S,T}$. From Fermi’s golden

![FIG. 2: (a) Voltage pulses applied to gate ‘P’ for the relaxation measurement. The starting point is a dot with one electron in the ground state (initialization). During the pulse, the singlet and triplet electrochemical potentials are below the Fermi energy and a second electron tunnels into the dot. (b) Schematic of the $\Delta I_{QPC}$ induced by the voltage pulse on gate ‘P’ if the state is singlet, a step from a slow tunneling event is added to the QPC response just after the read-out pulse. If the state was triplet, the tunneling event is too fast to be observed. (c) After averaging over many single traces, a dip is observed and its amplitude is proportional to the probability of having singlet present in the dot. (d) Relaxation curve obtained for $B = 1.02$ T constructed by plotting the dip amplitude of the averaged traces at a pre-defined time after the read-out pulse. The relaxation time, $T_1 = 0.79 \pm 0.05$ ms, is extracted from an exponential fit to the data (all the data are taken with a $100$ kHz low-pass filter). Inset: curve resulting from the averaging over 500 individual traces for the longest waiting time (20 ms) and for the shortest waiting time (300 ms), offset by 100 ms and 0.2 nA for clarity.](image-url)
rule, the relaxation rate between the triplet and the singlet states with energy separation $\Delta E_{S,T}$ is proportional to their coupling strength through electron-phonon interaction and to the phonon density of states at the energy $\Delta E_{S,T}$ \cite{11}. To obtain a simple analytical expression, we assume that the only effect of the perpendicular magnetic field, the Coulomb interaction between electrons and the modification of the potential landscape is to change the energy splitting. Especially, their effects on the spatial distribution of the wavefunctions are neglected and we neglect the Zeeman energy. Furthermore, we restrict the state space of the analysis to $|T_\pm\rangle$, $|T_+\rangle$, $|T_0\rangle$ and $|S\rangle$, both the orbital part (assuming Fock-Darwin states) and the spin part are present. Finally, we also neglect higher order (e.g. two-phonon) processes, which are important at small magnetic field \cite{22}.

In contrast to the one electron case \cite{7, 9}, the spin-orbit interaction admixes directly the first excited states $|T_\pm\rangle$ with the ground state $|S\rangle$. Due to the selection rules of the spin-orbit interaction, it does not affect $|T_0\rangle$ in lowest order \cite{22}. As a consequence, the spin relaxation time of $|T_0\rangle$ can be much longer than $|T_\pm\rangle$ \cite{24}. However, we do not observe any signature of a slowly relaxing component in the experiment \cite{25}. Since the spin-orbit coupling strength $M_{SO}$ is small in comparison with $\Delta E_{S,T}$ (in the range accessed in the experiment), we can approximate the new eigenstates of the system as:

$$|S'\rangle = |S\rangle - \frac{M_{SO}}{\Delta E_{S,T}} (|T_+\rangle + |T_-\rangle)$$

$$|T'_\pm\rangle = |T_\pm\rangle + \frac{M_{SO}}{\Delta E_{S,T}} |S\rangle$$

In general, $M_{SO}$ is dependent on the magnetic field, but to simplify the discussion, we neglect this dependence \cite{21, 22}. Since the electron-phonon interaction preserves the spin, the coupling between $|T'_\pm\rangle$ and $|S'\rangle$ has the following form:

$$\langle T'_\pm | H_{e,p} | S' \rangle = \frac{M_{SO}}{\Delta E_{S,T}} (\langle S | H_{e,p} | S \rangle - \langle T_\pm | H_{e,p} | T_\pm \rangle)$$

where $H_{e,p} \sim e^{iqr_1} + e^{iqr_2}$ is the interaction Hamiltonian between electrons and phonons, $\mathbf{q}$ the phonon wavevector and $\mathbf{r}_1$ the positions of the electrons. One can then interpret the coupling between $|T'_\pm\rangle$ and $|S'\rangle$ as the difference of the electron-phonon interaction strength for the corresponding unperturbed states $|T_\pm\rangle$ and $|S\rangle$. If the phonon wavelength is much larger than the dot size, the coupling to the phonons is the same for both states and the two terms will cancel. If the phonon wavelength is much shorter than the dot size, the coupling is small for each state separately.

To provide a quantitative comparison to the data, we need to model the electron-phonon interaction. Following \cite{8, 11}, we assume bulk-like 3D phonons. For the energy separations discussed in our experiment, only acoustic phonons are relevant. The Hamiltonian $H_{e,p}$ has then the following expression:

$$H_{e,p} = \sum_{j,\mathbf{q}} \frac{F_{\mathbf{q}}(q_z)}{2\rho q e_j / \hbar} (e^{iq_1 r_1} + e^{iq_2 r_2})(e^{i\beta_j \mathbf{q}} - iq \Xi_j \mathbf{q})$$
where \((q,j)\) denotes an acoustic phonon with wave vector \(q = (q_x,q_y)\), \(j\) the phonon branch index and \(\rho = 5300 \text{ kg/m}^3\) is the density of lattice atoms. The factor \(F_z(q_j)\) depends on the quantum well geometry and is assumed to be 1 in our model [11]. The speed of sound for longitudinal and transverse phonons are respectively \(c_l = 4730 \text{ m/s}\) and \(c_t = 3350 \text{ m/s}\). We consider both piezo-electric and deformation potential types of electron-phonon interaction. In the considered crystal, the deformation potential interaction is non-zero only for longitudinal phonons (with a coupling strength \(\Xi = 6.7 \text{ eV}\)). In contrast, all phonon polarizations \(j\) are important for piezo-electric coupling. The coupling strength depends on \(\theta\), defined as the angle between the wavevector and the growth axis, and varies for different polarizations as \(e\beta_j q = A_j(\theta) e\beta\) where \(e\beta = 1.4 \times 10^9 \text{ eV/m}\) [20]. Due to the different dependence on \(q\) for both mechanisms (\(\sqrt{q}\) for deformation potential interaction, \(1/\sqrt{q}\) for piezo-electric interaction), the piezo-electric (the deformation potential) coupling between electrons and phonons is dominant for energy separations below (above) 0.6 meV. From direct application of Fermi’s golden rule, we derive the following analytical expression for the spin relaxation rate 1/\(T_1\) :

\[
1/T_1 = \frac{M_{SO}^2 \alpha^2}{2 \pi^2 \hbar^2 c^2} \int_0^{\pi/2} d\theta \sin^5 \theta e^{\frac{-\Delta E^2 S,T \sin^2 \theta}{2k^2 l^2}} + \sum_j \frac{e^2 \beta_j \Delta E^2 S,T}{c^2} \int_0^{\pi/2} d\theta |A_j(\theta)|^2 \sin^5 \theta e^{\frac{-\Delta E^2 S,T \sin^2 \theta}{2k^2 l^2}}
\]

where \(\alpha\) is the dot radius (in our model \(\alpha\) is independent of \(\Delta E_{S,T}\) and is estimated to be 23nm, from the measured single particle level spacing). This simple model reproduces the most important feature in the measurements, which is that the coupling to the phonons vanishes for large and small energy separations and is strongest when the phonon wavelength matches the dot size (see Fig. 1).

The spin-orbit strength \(M_{SO}\) appears in the expression of 1/\(T_1\) only as a scaling factor. With a value \(M_{SO} = 0.4 \text{ eV}\) (corresponding to a spin-orbit length equal to \(\hbar/2\alpha m^* M_{SO} \approx 50 \mu\text{m}\)), the model reproduces the peak amplitude of the data quite well (Fig. 1 solid line). However, this value for \(M_{SO}\) is about six times smaller than the values reported in [27, 28] (the dotted line in Fig. 4 corresponds to the relaxation rate using this value of \(M_{SO}\) in the model). The discrepancy could be the result of the exclusion of higher orbitals and the magnetic field dependence of \(M_{SO}\) in our model [21, 22]. Again, we emphasize that both curves have a maximum corresponding to a phonon wavelength matching the dot size.

For single electron spin states, comparable variations of \(T_1\) with the energy splitting are expected although direct spin-orbit coupling between Zeeman sublevels of the same orbital is zero. To maximize the relaxation time of electron spin qubits, one needs then to choose an energy separation between the spin states such that the corresponding phonon wavelength is different from the dot size. To complete our study of spin relaxation, it will be interesting to rotate the sample with respect to the magnetic field since the spin-orbit coupling strength depends on the angle between the crystallographic axis and the magnetic field [3, 4, 24].

We thank V. Golovach and D. Loss for drawing our attention to the role of the phonon wavelength in spin relaxation and for useful discussions; R. Schouten, B. van der Enden and W. den Braver for technical assistance. Supported by the Dutch Organization for Fundamental Research on Matter (FOM), the Netherlands Organization for Scientific Research (NWO), the DARPA QUIST program and a E.U. Marie-Curie fellowship (T.M.).

\[\text{[1]}\ Y. Ohno et al., Phys. Rev. Lett. 83, 4196 (1999).
\[\text{[2]}\ A. C. Johnson et al., Nature 435, 925 (2005).
\[\text{[3]}\ T. Fujisawa et al., Nature 419, 278 (2002).
\[\text{[4]}\ J.M. Elzerman et al., Nature 430, 431 (2004).
\[\text{[5]}\ M. Kroutvar et al., Nature 432, 81 (2004).
\[\text{[6]}\ A.V. Khaetskii and Y.V. Nazarov, Phys. Rev. B 61, 12639 (2000).
\[\text{[7]}\ V.N. Golovach, A.V. Khaetskii, D. Loss, Phys. Rev. Lett. 93, 016601 (2004).
\[\text{[8]}\ D. V. Bulaev and D. Loss, Phys. Rev. B 71, 205324 (2005).
\[\text{[9]}\ T. Fujisawa et al., Science 282, 932 (1998).
\[\text{[10]}\ U. Bockelmann, Phys. Rev. B 50, 17271 (1994).
\[\text{[11]}\ D. Loss and D.P. DiVincenzo, Phys. Rev. A 57, 120 (1998).
\[\text{[12]}\ L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, Rep. Prog. Phys. 64 (6), 701 (2001).
\[\text{[13]}\ Jordan Kyriakidis et al., Phys. Rev. B 66, 035320 (2002).
\[\text{[14]}\ M. Ciorga et al., Phys. Rev. B 61, R16315 (2000).
\[\text{[15]}\ J. M. Elzerman et al., Appl. Phys. Lett. 84, 4617 (2004).
\[\text{[16]}\ R. Schleser et al., Appl. Phys. Lett. 85, 2005 (2004).
\[\text{[17]}\ L.M.K. Vandersypen et al., Appl. Phys. Lett. 85, 4394 (2004).
\[\text{[18]}\ M. Field et al., Phys. Rev. Lett. 70, 1311 (1993).
\[\text{[19]}\ D. G. Austing et al., Phys. Rev. B 60, 11514 (1999).
\[\text{[20]}\ V.N. Golovach, A.V. Khaetskii, D. Loss, preprint.
\[\text{[21]}\ P. San-Jose et al., Phys. Rev. Lett., 97, 076803, (2006).
\[\text{[22]}\ S. Dickmann and P. Hawrylak, JETP Lett., 77, 30 (2003).
\[\text{[23]}\ S. Sasaki et al., Phys. Rev. Lett. 95, 056803 (2005).
\[\text{[24]}\ By applying the complete relaxation measurement protocol presented in Fig. 2 (a), we observe experimentally a reduction of the read-out visibility (the contrast between the signal for short and long waiting time) when \(\Delta E_{S,T}\) is varied. Several reasons can be considered to explain the reduction of visibility observed in the experiment. At zero magnetic field, the difference between the singlet and triplet tunnel rates can be increased as the dot is somewhat elliptical. At relatively high magnetic fields, the dot loses its elliptical nature and then the measurement loses visibility. Another possible explanation is related to the...
slow spin-orbit decay from $|T_0\rangle$ to $|S\rangle$ in comparison with the longest waiting time we explored (20 ms). Therefore, the $T_0$ population does not contribute to the visibility unless $|T_0\rangle$ gets first mixed with $|T_{\pm}\rangle$. As $B$ increases the mixing between triplets gets suppressed and the visibility goes down.

[26] For longitudinal phonons, the piezo-electric constant $A_l(\theta) = 3\sqrt{2}/4 \sin^2(\theta) \cos \theta$. For the two transverse polarizations, $A_{l1}(\theta) = \sqrt{2}/4 \sin 2\theta$ and $A_{l2}(\theta) = \sqrt{2}/4 (3\cos^2 \theta - 1) \sin \theta$.

[27] D. M. Zumbuhl et al., Phys. Rev. Lett. 89, 276803 (2002)
[28] S. Amasha et al., cond-mat/0607110
[29] V.I. Fal’ko, B.L. Altshuler, and O. Tsyuplyatyev, Phys. Rev. Lett. 95, 076603 (2005)