Current induced entanglement of nuclear spins in quantum dots

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We propose an entanglement mechanism of nuclear spins in quantum dots driven by the electric current. The current is accompanied by the spin \(I\) in quantum dots and gradually increases components of larger total spin of nuclei. This entangled state drastically enhances the spin relaxation rate of electrons, which can be detected by measuring a leakage current in the spin-blockade region. This mechanism is not relevant in optical experiments which examine the spin relaxation of single excitations.

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The spin relaxation time of electrons in quantum dots is an important issue. The time needs to be sufficiently long for the implementation of quantum computing devices utilizing electron spins in quantum dots. The relaxation time has been found to be quite long by optical experiments and transport experiments. In the former experiments, the transient measurements indicate a quenching of the spin relaxation on the exciton lifetime scale in self-assembled quantum dots. In the latter experiments, the tunneling current is measured through single or double quantum dots in spin-blockade situations, where electrons cannot be transported unless the spins are tipped in the dot. The relaxation time is as long as 200 s from a spin-triplet excited state to spin-singlet ground state in single quantum dots. In weakly coupled double quantum dots, the current suppression has been observed when two electrons occupy the lowest energy level in each dot with parallel spins: an electron tunneling from one dot to the other is forbidden by the Pauli exclusion principle.

In the present paper, we theoretically study a small leakage current in spin-blockade regions. The current is accompanied by the spin \(I\) in the quantum dots. As a spin-\(I\) mechanism, we investigate the hyperfine interaction between electrons and nuclear spins. Recent experimental results have implied its important role in the leakage current. We examine a formation of entangled states of nuclear spins driven by the electron transport. This mechanism is analogous to the current-induced dynamic nuclear polarization (DNP) in quantum Hall systems and transport experiments. The DNP is created by the electron scattering between spin-polarized regions, which leads to a hysteresis in the longitudinal resistance. In our case, the entanglement of nuclear spins drastically enhances the spin relaxation rate of electrons in quantum dots, which could be observed experimentally. This mechanism is not relevant in the optical experiments which examine the spin relaxation of single excitations. Hence, the role of the hyperfine interaction in the spin relaxation could be quite different in transport and optical experiments.

Model: To consider the spin blockade in quantum dots, we adopt a following model. A quantum dot is weakly coupled to external leads L, R through tunnel barriers. The Coulomb blockade restricts the number of electrons in the dot to be \(N_{el} \) or \(N_{el} + 1\). \(N_{el}\) electrons form a background of spin singlet. From lead L on the source side, an extra electron tunnels into the dot and occupies a single level with an envelope wavefunction \(|\phi\rangle\). The spin of the electron is either \((\mathbb{S}_z = 1)\) or \((\mathbb{S}_z = -1)\) with equal probability. We assume an easy-axis of electron spins in \(z\) direction for a while. The electron stays for a long time by the spin blockade because the spin relaxation time is much longer than the tunneling time. After the spin \(I\) is tipped in the dot, the electron immediately tunnels out to lead R on the drain side, and then the next electron is injected from lead L.

An electron occupying orbital \(|\phi\rangle\) interacts with \(N_{el}\) nuclear spins, \(I_{el}\), by the hyperfine contact interaction. \(N_{el}\) in GaAs quantum dots. We assume nuclear spins of \(1/2\) for simplicity. The Hamiltonian in the dot reads

\[
H_{hf} = \sum_{k=1}^{N_{el}} \gamma_k |I_{el} \rangle \langle I_{el} | I_{el} S_{I_{el}} I_{el} \rangle
\]

where \(\gamma\) is the volume of the crystal cell and \(I_{el} = \sum_{k=1}^{N_{el}} I_{el}\) is the total spin of nuclei. We have assumed that \(I_{el}\) is independent of the nuclear site, \(I_{el}\). This is a good approximation for a large part of nuclear spins in the quantum dot, except in the vicinity of nodes of \(|\phi\rangle\), since the distance between nuclei is much smaller than the size of the dot, or an extension of \(|\phi\rangle\).

Basic idea: The Hamiltonian indicates that N nuclear spins interact with a common \(|\phi\rangle\) of electron spin although there is no direct interaction between them (dipole-dipole interaction between nuclear spins is weak and neglected). This \(|\phi\rangle\) results in the entanglement among nuclear spins. To illustrate this, let us consider the simplest case of \(N_{el} = 2\) and begin with a polarized state of nuclear spins \(I_{el} = 1\). \(I_{el} = 1\).

An electron with spin \(j\) tunnels into the dot and is spin-tipped by the hyperfine interaction. Then the state of nuclear spins becomes \((j = 1, j = 2)\). \(I_{el} = 1\). \(M = 0\), where \(J = M = 0\), where \(J = 1\) and \(M = 0\), respectively. This
is an entangled state. After the electron tunnels out of the dot, the next electron is injected with $j \neq i$ (or $j^*i$). The spin–ip probability of the second electron is proportional to $\frac{j_i}{j \neq i}$, 1; $i^*$ $j$, $j_0$; $j^*(j^*$ $i)$. This value is twice the probability in the case of non-entangled states $j_1 = 2l_1$ or 1 $= 2l_1$. In general, the capability of state $j ; M$ to ip an electron spin is $\langle J \ M \ J \rangle$. (1) Spin is never s nipped, and ejected out of the dot. After the electron injection, say, $j \neq i$, the time evolution of the dot state by $H_{el} = 2$ $M$ + 1 + $J$, $M$+1; $j \neq i$ is $\langle J \ M \ J \rangle$. The number of such states is given by

$$
K \langle N ; J \rangle = N! \left( \frac{N - 1}{(N - 1)(N - 2)} \right) \left( \frac{N - 2}{(N - 2)(N - 3)} \right) \cdots \left( \frac{1}{1} \right).
$$

A fier the injection of the $n$th electron, say, $j \neq i$, the time evolution of the dot state by $H_{el} = 2$ $M$ + 1 + $J$, $M$+1; $j \neq i$ is

$$
e^{-i \beta \hbar t} \langle 0 | \hat{J} | M \rangle \langle J \ M \ J \rangle = \frac{K \langle N ; J \rangle}{N!} \sum_{j \neq i} X C_{j ; M}^{(0)} (j ; M) (J \ M + 1) e^{i \beta \hbar t}.$$

The probability of finding an electron with spin $i$ at $t = 0$, and $j \neq i$ at $t = \beta \hbar t$, is given by

$$P^{(0)} = \langle 0 | \hat{J} | M \rangle \langle J \ M \ J \rangle.$$

The electron tunnels out the dot immediately after the spin is s nipped. Then the wavefunction shrinks to

$$P^{(1)} = \frac{1}{P^{(0)}} X C_{j ; M}^{(0)} (j ; M) (J \ M + 1) e^{-i \beta \hbar t}.$$

The state of nuclear spins becomes

$$C_{j ; M}^{(n)} = \frac{X C_{j ; M}^{(0)} (j ; M) (J \ M + 1)}{P^{(n)}},$$

$$C_{j ; M}^{(n)} = \frac{X C_{j ; M}^{(0)} (j ; M) (J \ M + 1)}{P^{(n)}},$$

$$f^{(n)} = \frac{1}{P^{(n)}} X C_{j ; M}^{(0)} (j ; M) (J \ M + 1).$$

Figure 1 shows the distribution of the total spin $J$ and ip of $z$ components $M$. The spin–ip processes increase the weight of larger $J$ and smaller $M$. Although $M$ changes less faster than $J$, this means that the total nuclear spins tend to develop in the plane perpendicular to the easy-axis of electron spins.

This entangled state enhances the spin–ip probability. For the $n + 1$th electron, the probability is given by

$$P^{(n)} = \frac{1}{P^{(n)}} X C_{j ; M}^{(n)} (j ; M) (J \ M + 1).$$

The probability increases with $n$ linearly ($P^{(n)} = n P^{(0)}$) and finally saturates ($P^{(n)} = N = 2 P^{(0)}$).
and we consider the case in the presence of magnetic field: of calculating the current in the previous situations, now we consider the case in the presence of magnetic field:

In absence of easy-axis: Until now, we have assumed the presence of easy-axis of electron spins. In the absence of the axis, the spin of the nth electron is oriented in an arbitrary direction, \( \cos \theta_i \mathbf{J} + \sin \theta_i \mathbf{e} \mathbf{J} \). In this case, the previous formulation can be applied after the rotation of \( F_{J+1}^{(n)} \) and \( \mathbf{g} \) in Eq. (8) are replaced by the averaged values of \( F_{J+1}^{(n)} \) and \( \mathbf{g} \). They develop by

\[
F_{J+1}^{(n)} = \frac{1}{2} \left[ \mathbf{J} (J+1) \right]^{1/2}; \quad (12)
\]

where

\[
\mathbf{g}^{(n)} = \frac{1}{2} \left[ \mathbf{J} (J+1) \right]^{1/2}; \quad (13)
\]

As in the previous case, components of larger \( J \) increase with \( n \), which enlarge the spin-\( \mathbf{p} \) probability by the factor of \( 2J(J+1) = 3 \). The probability is given by \( P_{J} = \mathbf{C}_{J+1}^{(n)} \mathbf{J} \), which is shown by broken line in Fig. 2(a). This behaves as \( P_{J} = (2n+3)P_{J}^{(0)} \) for \( n \lt N = 2 \), and \( P_{J} = (N=3)P_{J}^{(0)} \) for \( N = 2 \), with \( n \) with \( J \).

Leakage current: The enhancement of the spin-\( \mathbf{p} \) probability is reached by the leakage current. Instead of calculating the current in the previous situations, now we consider the case in the presence of magnetic field:

Zeeman energy for electrons is much larger than the hyperfine interaction, whereas that for nuclear spins is negligible. Then \( H_{hf} \) can be treated as a perturbation. The magnetic field makes an easy-axis of electron spins. We also take into account the electron-phonon interaction for the energy conservation. The Hamiltionian in the dot reads \( H = H_{el} + H_{ph} \), where \( H_{el} = P_{el} (a_{J} a_{J}^{+}) \), and \( H_{ph} = ~q_{J} q_{J}^{+} - a_{J}^{+} a_{J} \). Thus the current is given by \( I(t) = e \mathbf{J} \), and \( N = 100 \). In the presence of easy-axis, we take the geometry mean between upper and lower signs in Eq. (13).

FIG. 1: The distribution of (a) the total spin \( J \) and (b) its z component, in the state of nuclear spins. An easy-axis of electron spins is assumed. (a) \( P_{J} = \mathbf{C}_{J+1}^{(n)} \mathbf{J} \), and (b) \( P_{J} = \mathbf{C}_{J+1}^{(n)} \mathbf{J} \), where \( n \) is the number of transported electrons accompanied by the spin-\( \mathbf{p} \). The number of nuclear spins is \( N = 100 \). In Eq. (13), we take the geometric mean between upper and lower signs.

FIG. 2: (a) The spin-\( \mathbf{p} \) probability \( P_{J}^{(n)} \) with \( xed t \), as a function of \( n \) (number of transported electrons accompanied by the spin-\( \mathbf{p} \)). (b) The spin-\( \mathbf{p} \) current \( I(t) \) as a function of time \( t \). The cases in the presence and absence of easy-axis of electron spins are indicated by solid and broken lines, respectively. The number of nuclear spins is \( N = 100 \). In the presence of easy-axis, we take the geometric mean between upper and lower signs in Eq. (13).
spin–singlet or–triplet formation with an electron trapped in the other dot, in double dot systems. In this case, the easy-axis of electrons would be absent. (If the crystal electric field is weak enough.) The perturbation with respect to $H_{0} + \hat{H}_{el}$, changes the nuclear spin state following Eq. (1). The spin–triplet rate is $\left\langle n \right| G \left| n \right\rangle$. The leakage current $I(t)$ is represented by broken line in Fig. 2(b). It is written approximately as

$$I(t) = \frac{e}{2} \left\langle 0 \right| e^{2} \left\langle t \right| t_{\text{sat}} \left( t \right)$$

where $t_{\text{sat}} = 3 \ln (N =2) = \left\langle 0 \right|$. The enhancement of the current is less prominent than in the presence of easy-axis.

Discussion: We have proposed the current induced entanglement of nuclear spins in quantum dots. The current accompanying the spin in the dots gradually increases compared with the total spin of nuclei, which significantly enhances the spin relaxation of electrons. As a result, the leakage current in the spin–blockade region grows drastically with time and finally saturates. This mechanism is not relevant in the optical experiments which examine the spin relaxation of single excitations.

The saturation time of the leakage current is of the same order as $1 = \left\langle 0 \right|$, spin relaxation time with non-entangled state of nuclear spins, which is 100 ns or much larger $2.3$. The current is enhanced during the dephasing time $T_{2}$. When the entangled state is broken, the current is suppressed. This possible would result in a current–excitation with time $\left\langle t \right|$ which has been observed recently $3$. The characteristic time of the excitation is $T_{2}$.

A possible origin of the dephasing is the dipole–dipole interaction between nuclear spins. Note that, among $N$ nuclear spins participating in the formation of total spin $\frac{j}{2}$; $M_{i}$, this interaction conserves the total spin, and hence does not destroy the entangled state. The interaction between one of the $N$ spins and a spin surrounding the dot results in the dephasing. Estimating $T_{2}$ in our case and analyzing evolution of nuclear spins after the dephasing are beyond the present paper.

We discuss the validity of our simple models. We have disregarded the spatial variation of the envelope wavefunction of electrons in the quantum dot. Although the entanglement of nuclear spins is generally seen in the presence of the variation (see Eq. (4) in Ref. $[15]$), the enhancement of the spin relaxation is more evident with larger total spin $J$. Hence the capability of spin–triplet electron is determined by the e–ective number $N_{e}$ of nuclear spins which feel the same electron field, ($\left\langle n \right| G \left| n \right\rangle \simeq$ const. since $J_{nax} = N_{e} = 2$). The generalization of nuclear spins is an interesting problem.

If the contribution from higher energy levels of electrons in quantum dots is not negligible, through electron–phonon interaction $2,3$ or spin–orbit (SO) interaction $14$, it should be taken into account carefully. Particularly, the coexistence of hyperfine and SO interactions would complicate the evolution of the nuclear spin state because the terms of $n \rightleftharpoons m$ are mixed on the right side of Eq. (3). We have also disregarded higher-order tunneling processes $3$, which could play a role in the spin relaxation of electrons $4$. However, the processes do not influence the entanglement of nuclear spins discussed here.

Finally, we comment an analogy between our mechanism and the Dicke effect of superradiance $3,19$. The spontaneous emission of photons is enhanced from $N$ atoms with two levels (pseudo-spin $S = 1/2$) if all of them are excited initially. This is due to the formation of pseudo-spin of total spin $J = N/2$. Starting from $j_{i} / J$, the state of $N$ atoms changes like a cascade, $j_{i} / J_{i} j_{1} / J_{1}$, emitting photons. A similar effect has been proposed for the emission of phonons from $N$ equivalent quantum dots $13,20$. The atom $s$ (quantum dots) correspond to the nuclear spins in our model, while the emission of phonons (phonons) to the spin–triplet of electrons. A main difference is the initialization, $N$ excited states have to be prepared by the pumping in the Dicke effect, whereas the initialization is not necessary in our mechanism.

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[13] In double quantum dots, it depends on the spin direction whether the spin–blockade takes place. If the spin of an incident electron in the first dot is parallel to that of the electron trapped in the second dot, the former electron cannot tunnel in the second dot due to the Pauli exclusion principle. If the spins are antiparallel, the spin–blockade does not happen; the electron tunnels in the second dot and finally out to lead the immediate. The total current...
is the sum of the currents with and without spin flip. We consider the former current only.

[14] We assume that k_B T is larger than E_z. Otherwise, the spin flip of electrons only from j' to j'' is available by \( H_{el} + H_{ph} \), which increases the components of larger J and M. The other spin could be caused by e.g., higher-order tunnel processes between the dot and leads [17].

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