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Abstract. We consider the detection of the electron spin of a doped atom, i.e., nitrogen or phosphorus, caged in a fullerene by the currently available technique of the scanning tunnelling microscope (STM), which actually corresponds to the readout of a qubit in fullerene-based quantum computing. Under the conditions of polarized STM current and Coulomb blockade, we investigate the tunnelling matrix elements involving the exchange coupling between the tunnelling polarized electrons and the encapsulated polarized electron, and calculate the variation of the tunnelling current with respect to different orientations of the encapsulated electron spin. The experimental feasibility of our scheme is discussed taking account of some imperfect factors.
1. Introduction

Quantum information processing (QIP) based on solid-state materials has attracted much attention over the past few years due to the potential of scalability. Since the qubits are encoded in individual electron spins in most solid-state QIP schemes, how to efficiently detect a single electron spin has recently become a focus. Although there is no fundamental restriction for the single spin detection, it seems that, to do the qubit readout very well, we expect further development of detection techniques with sufficient spatial and temporal resolution.

Some preliminary experiments have reached the single spin level so far. For example, scanning tunnelling microscope (STM) electron spin resonance (ESR) has demonstrated single molecule ESR spectroscopy of iron impurities in silicon [1], although theoretical work remains to clarify the best description of the effects [2]. A very recent STM experiment under low temperature and with a high magnetic field has shown the spin flip spectra of a single manganese atom [3], and the magnetic resonance force microscope, which utilizes a cantilever driven by spins oscillating in resonance, has successfully detected a single surface electron spin [4]. Moreover, it has been reported that micro-superconducting quantum interference devices are capable of distinguishing large spin difference ($\Delta m_S \sim 30$) [5].

There have been some theoretical proposals for single spin detection in the candidate systems of QIP [6, 7]. The present study will focus on the detection of a single electron spin of an encapsulated atom inside a fullerene (i.e., C$_{60}$), which corresponds to the readout of a qubit in the performance of quantum computing based on the endohedral fullerenes N@C$_{60}$ or P@C$_{60}$ [8, 9]. The electron spin inside the fullerene plays the role of a qubit or an auxiliary qubit. As the electrons of the doped atoms cannot escape the C$_{60}$ cage while preserving their spin states, we must develop special proposals for the spin detection, different from those in [6, 7]. In fact, there have been some schemes in this respect, for example, using a modified single molecule transistor (SMT) [10], a nanomagnet molecule Fe$_8$ [11], and a shuttling device [12]. However, no relevant experiment has yet been achieved.

In the present paper, we investigate the possibility of detecting a single electron spin inside the fullerene by STM. STM is a mature technique available to manipulate single atoms with high precision at low temperature [13]. Although no reliable evidence has been shown to achieve a single spin detection, there have been some experiments [3, 14, 15] and theoretical proposals [16]–[18] regarding single spin problems with the STM. Our scheme is different from those proposals in the following points: firstly, the electron spin to be detected, in our case, is assumed to be well polarized along an always-on magnetic field, while we do not know whether the spin is up- or down-polarized. This corresponds to the final result of real quantum computing,
i.e., the qubits returning to product states from entanglement and superposition after the elaborately designed logic gates have been carried out. So our detection requires to know the orientation of the spin polarization. Secondly, as the tunnelling through the air is different from that through a fullerene, our treatment is to separate the whole tunnelling process into three consecutive steps. Thirdly, due to the C_{60} cage, the local electron spin (i.e., the spin to be detected inside the cage) is well protected. As a result, different from the case in \[16\]–[18] with the electron spin on the surface, the main error in our case is from the vibration of the fullerene, instead of the spin scattering, due to the tunnelling electrons.

The conditions we employ in the treatment below include Coulomb blockade \[19\] and the polarized current in STM \[20\], both of which have been achieved experimentally. The Coulomb blockade restricts the fullerene to be charged by no more than one electron, which means an electron from the STM tip could jump on the C_{60} only after the previous electron sitting on the C_{60} has jumped away. This has been demonstrated in a recent experiment \[19\] for SMT. So we will suppose that this Coulomb blockade works throughout our scheme under a suitable bias voltage of the STM. The polarized current in STM implies that the electrons going out of the STM tip are well polarized, i.e., up- or down-polarized. It is reported in \[20\] that high-quality polarized current is already available in STM. So we suppose below that the tunnelling electrons out of the STM tip are up-polarized. In the presence of a magnetic field, the tunnelling electrons will couple to the caged electron by exchange interaction due to the spin degrees of freedom involved \[21\]. Moreover, the total electron spin of the doped atom N or P are \(S = \frac{3}{2}\) with four Zeeman levels \(|\pm \frac{3}{2}\rangle\) and \(|\pm \frac{1}{2}\rangle\) \[9\]. It has been demonstrated that quantum gating can be performed independently with electron spin states \(|\pm \frac{3}{2}\rangle\) or \(|\pm \frac{1}{2}\rangle\) \[22\]. For simplicity, however, we will focus below on the discussion about the caged electron spins \(|\pm \frac{1}{2}\rangle\). The case regarding \(|\pm \frac{3}{2}\rangle\) could be obtained simply by enlarging the variation of the tunnelling current by three times.

In the next section, we will explore the spin-dependent tunnelling matrix elements. Then a specific calculation of the tunnelling will be made in section 3 which also includes discussions about the experimental imperfection. The last section is for the conclusion.

2. Spin-dependent tunnelling

To clarify our description below, we simplify the system shown in figure 1 to be a doped fullerene sandwiched by two leads L and R, as shown in figure 2. So the Hamiltonian is,

\[ H = \sum \epsilon_{\lambda,k,l} c_{\lambda,k,l}^\dagger c_{\lambda,k,l} + H_c + \sum (t_{\lambda,l} c_{\lambda,k,l}^\dagger a_{n,l} + t_{\lambda,l}^* c_{\lambda,k,l} a_{n,l}^\dagger), \]

where \(c_{\lambda,k,l}^\dagger (c_{\lambda,k,l})\) creates (annihilates) an electron in the lead \(\lambda = L, R\), with \(k\) being the momentum of the electron, \(l = \pm 1\) for (up/down) spin polarization, and \(\epsilon_{\lambda,k}\) are energies regarding the Fermi energy. The operator \(a_{n,l}^\dagger (a_{n,l})\) is related to the electron on the fullerene in the orbital \(n\). \(t_{\lambda,l}\) is the tunnelling coefficient regarding \(\lambda = L\) or \(R\). \(H_c\) accounts for the fullerene including its energy and the Coulomb blockade term \((\sum_k a_k^\dagger a_k)(\sum_k a_k^\dagger a_k - 1)\) as well as the exchange coupling \(-J\hat{S} \cdot \hat{\sigma}\) with \(\hat{\sigma}\) and \(\hat{S}\) the spin degrees of freedom of the electrons outside and inside the fullerene, respectively. Since our interest is in the tunnelling modification due to the electrons’ exchange interaction, instead of the electro-phonon \[23\] or spin-phonon resonances,
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Figure 1. Schematic of the detection of a single caged electron spin by STM, where the black dots mean the electrons, and $\sigma$ and $S$ correspond to the spin degrees of freedom regarding the electrons tunnelling and caged, respectively. The caged electron spin can be $\pm 1/2$ or $\pm 3/2$; which pair of spins employed in quantum computing is known before the detection. So what we want to detect is whether the spin is up (i.e., $1/2$ or $3/2$) or down (i.e., $-1/2$ or $-3/2$). The gap between the fullerene and the substrate is due to van der Waals interaction [19].

Figure 2. (a) Schematic of our treatment to model the spin-dependent tunnelling by three steps, and (b) the work functions in different steps are assumed.

we may follow the idea in [16]–[18] and focus on the spin-dependent tunnelling matrix elements. To this end, we separate the tunnelling into three steps (see figure 2). The first step is from the left lead to the fullerene, the second one is for tunnelling through the fullerene, and the third accounts for the tunnelling from the fullerene to the right lead.
Both the first and the third steps can be modelled as electronic tunnelling through capacitors with $\Phi_i = W_id_i$, where $i = 1, 3$, $\Phi_i$ is the work function, $d_i$ is the tunnelling gap, and $W_i$ represents the constant regarding the free space permittivity, the dielectric constant and the area of the lead. By using WKB approximation, we could reach the spin-dependent tunnelling matrix elements as below\textsuperscript{4},

$$\hat{T}_1 \propto \exp\left\{-\sqrt{\frac{8m}{\hbar^2}} \int_0^{x_1} \sqrt{\Phi_1 - J\hat{S} \cdot \hat{\sigma}} \, dx\right\},$$

(2)

and

$$\hat{T}_3 \propto \exp\left\{-\sqrt{\frac{8m}{\hbar^2}} \int_{x_2}^{x_3} \sqrt{\Phi_3 - J\hat{S} \cdot \hat{\sigma}} \, dx\right\},$$

(3)

where $J$ is the strength for the exchange coupling between the tunnelling electrons and the caged electron due to the spin degrees of freedom of the electrons, which is proportional to the integral below

$$\int\int \Psi_1^*(r_1)\Psi_2^*(r_2)\frac{e^2}{r_{12}}\Psi_1(r_2)\Psi_2(r_1) \, dr_1 \, dr_2,$$

(4)

with $\Psi_1(r_i)$ and $\Psi_2(r_j)$ the wavefunctions of the tunnelling and caged electrons at the position $r_i$ and $r_j$, respectively, and $r_{12} = |r_1 - r_2|$. For simplicity, we have omitted in equations (2) and (3) the unimportant constants regarding the spin-independent tunnelling rate in the absence of $J$. Note that we have also made an assumption here that, due to the Coulomb blockade and the low temperature limits, each electron from the left lead would jump to the ground state of $C_{60}$ and the tunnelling amplitude is mainly dependent on the exchange interaction, instead of the electronic momenta. Since both spins of the tunnelling and caged electrons are well polarized, we may simplify $J\hat{S} \cdot \hat{\sigma}$ to be $J\sigma_z$ with $J$ being 1 meV [21]. As a result, straightforward algebra for equation (2) yields the corresponding tunnelling rates,

$$T_1 \propto \exp\left\{-\frac{2}{3} \sqrt{\frac{8m W_1}{\hbar^2 x_1^3/2}} \left[ x_1 \pm \frac{J}{W_1} \right] \left[ \frac{1}{W_1 x_1} \right]^{3/2}\right\},$$

where the signs ‘±’ correspond to the up/down polarization of the caged electron spin. We have approximately omitted in the above equation the high-order terms regarding $J/W_1$ because they

\textsuperscript{4} It is a common understanding from the standard text book of quantum mechanics that the WKB method yields the tunnelling amplitude to be proportional to $\exp\left(-\sqrt{\frac{8m}{\hbar^2}} \int \sqrt{V - E} \, dx\right)$, where $V$ and $E$ are, respectively, the potential barrier and the energy of the tunnelling particles. Due to spin degrees of freedom involved, the potential is spin dependent in our case. So as in [16, 17], we replace $(V - E)$ by $(\Phi - J\hat{S} \cdot \hat{\sigma})$ with $\Phi$ and $J$ being work function and the strength of the exchange coupling, respectively. The difference in expressions of the tunnelling matrix elements in [16, 17] from our study is due to the constant potential barrier supposed in their treatment.
are comparatively negligible (i.e., \( J/W_1 \sim x_1/1000 \) to the numbers shown later). Similarly, we have

\[
T_3 \propto \exp \left\{ -\frac{2}{3} \sqrt{\frac{8mW_3}{\hbar^2}} \left[ x_3^{3/2} \left( 1 \pm \frac{3J}{2W_3x_3} \right) - x_2^{3/2} \left( 1 \pm \frac{3J}{2W_3x_2} \right) \right] \right\}
\]

\[
\approx \exp \left\{ -\frac{2}{3} \sqrt{\frac{8mW_3}{\hbar^2}} x_3^{3/2} \left[ 1 \pm \frac{3J}{2W_3x_3} \right] \right\} \exp \left\{ \frac{2}{3} \sqrt{\frac{8mW_3}{\hbar^2}} x_2^{3/2} \left[ 1 \pm \frac{3J}{2W_3x_2} \right] \right\}.
\]

The second step can be considered as tunnelling through a constant potential barrier. This is because the fullerene could be modelled as a spherical capacitor. Due to the equipotential surface of the spherical capacitor, the work function \( \Phi_2 \) should be a constant, which yields the tunnelling rate as

\[
T_2 \propto \exp \left\{ -\sqrt{\frac{8md_2^2}{\hbar^2}} (\Phi_2 \pm J) \right\}.
\]

Since \( J \) is smaller than \( \Phi_1 \) by 1000 times, we may write the terms regarding \( J \) as

\[
\exp(\pm D_i) = \cosh(D_i) \pm \sinh(D_i) \approx 1 \pm D_i,
\]

where \( D_1 = J\sqrt{8mx_1/(W_1 \hbar^2)} \) is from \( T_1 \), \( D_2 = J\sqrt{8mx_3/(W_3 \hbar^2)} \) and \( D_3 = J\sqrt{8mx_2/(W_3 \hbar^2)} \) are from \( T_3 \), and \( D_4 = \sqrt{2mJd_2}/\sqrt{\Phi_2 \hbar^2} \) is from \( T_2 \).

Since the tunnelling current is proportional to the multiplication of the tunnelling rates above, we may simply write the tunnelling current as

\[
I = I_0 \prod_{i=1}^{4} (1 \pm D_i),
\]

where

\[
I_0 \propto \exp \left\{ -\sqrt{\frac{32mW_1}{9\hbar^2}} x_1^{3/2} \right\} \exp \left\{ -\sqrt{\frac{32mW_3}{9\hbar^2}} (x_3^{3/2} - x_2^{3/2}) \right\} \exp \left\{ -\sqrt{\frac{8m\Phi_2}{\hbar^2}} d_2 \right\},
\]

is the spin-independent tunnelling current, which could be detected in the absence of the magnetic field. In the following calculation, we assume \( I_0 \) to be 1 nA [19] if \( d_1 = 0.23 \) nm.

3. Discussion

From [19] we know that, \( d_2 = 0.7 \) nm is the size of the fullerene, and \( d_3 = 0.27 \) nm is due to van der Waals interaction between the fullerene and the substrate. We suppose that \( \Phi_1(x) \) is 2 eV between \( x = 0 \) and \( x = x_1 \), \( \Phi_2 = 1 \) eV is constant, and \( \Phi_3(x) = 1 \) eV is from \( x = x_2 \) to \( x_3 \). Then we can obtain by direct calculation that \( \Delta I_{\pm} = I_{\pm} - I_0 = \pm 16 \) pA, where \( I_{\pm} \) corresponds,
respectively, to ± in equation (7). The current of 16 pA is detectable with the present STM technology. Slightly changing \(d_1\), we may estimate \(\Delta I = \Delta I_+ - \Delta I_-\), which changes drastically with respect to \(d_1\), as shown in figure 3. So it is evident that the difference between spin-up and spin-down of the caged electron is distinguishable from the tunnelling current as long as \(d_1\) is around 0.24 nm.

As mentioned above, because the local spin in our case is protected by the fullerene cage, the main source of error is the vibration of the fullerene due to the tunnelling electrons, which yields a shift \(\delta = 3\) pm equivalent to the energy variation 5 meV [19]. Because this vibrational degree of freedom is far detuned from other characteristic frequencies, and also there is no evidence of vibration–spin coupling, we did not consider this vibration in above treatment. The tunnelling current, however, is very sensitive to the distance variation. So we have to strictly assess the influence due to the position shift of the fullerene on the tunnelling current. To this end, we consider the tunnelling gaps regarding the first and the third steps to be changed time-dependently, while the second step remains unchanged. So we have the modification of the tunnelling rates,

\[
T_1 \propto \exp \left\{ -\frac{2}{3} \sqrt{\frac{8mW_1}{\hbar^2}} x_1^{3/2} \left( 1 + \frac{3\Delta}{2x_1} \pm \frac{3J}{2W_1x_1} \right) \right\},
\]

\[
T_3 \propto \exp \left\{ -\frac{2}{3} \sqrt{\frac{8mW_3}{\hbar^2}} x_3^{3/2} \left( 1 \pm \frac{3J}{2W_3x_3} \right) \right\} \exp \left\{ \frac{2}{3} \sqrt{\frac{8mW_3}{\hbar^2}} x_2^{3/2} \left( 1 - \frac{3\Delta}{2x_2} \pm \frac{3J}{2W_3x_2} \right) \right\}.
\]

Suppose \(\Delta = (\delta/2) \cos(\omega t)\) with \(\omega = 1\) THz [19]. As in [16], we introduce the average current over the time \(T\), i.e., \(\langle I \rangle = (1/N) \sum_{i=1}^{N} I(t_i)\), where the sum from \(i = 1\) to \(N\) is over the number of the tunnelling electrons in the time \(T\), and \(I(t_i)\) is proportional to the tunnelling rate for each
electron. So $\langle I \rangle$ can be written as

$$\langle I \rangle = I_0 \prod_{i=2,4} (1 \pm D_i) \left[ 1 \pm D_1 + (\delta/2x_1) \sqrt{8mW_1/\hbar^2} \int_0^T (1/T) \cos(\omega t) dt \right]$$

$$\times \left[ 1 \pm D_3 - (\delta/2x_2) \sqrt{8mW_3/\hbar^2} \int_0^T (1/T) \cos(\omega t) dt \right].$$

(11)

As the terms regarding $\delta/x_{1(2)}$ are comparable to $D_{1(3)}$, we have to eliminate the vibrational influence. To this end, we may detect the current during a time period $T = k\pi/\omega$ with $k = 1, 2, \ldots$, in order to average out the terms regarding $\delta$. This would in principle make the fullerene vibration, due to the tunnelling of the electrons, of negligible effect on the original current in our observation. However, due to the intrinsically stochastic property of the electron tunnelling, the efficiency of the above trick depends on the statistical deviation $\sigma$ of a normal distribution $1/(\sqrt{2\pi}\sigma) \exp[-(t - t_0)^2/(2\sigma)^2]$, where $t_0$ is the mean tunnelling time of each electron we desire [10]. To carry out our scheme optimally, we require $\sigma$ to be as small as possible.

Besides, there are other possible imperfection in implementing our scheme. In real quantum computing, the qubits are not usually well polarized at the end of the computing operations. For example, the C$_{60}$-caged electron-spin would probably be in superposition $F|\uparrow\rangle + G|\downarrow\rangle$ ($F \gg G$ or $F \ll G$) due to some unpredictable error sources in quantum computing. The estimated current dispersion due to this imperfect polarization is [16], e.g., $F \gg G$,

$$\frac{\sqrt{\langle \Delta I^2 \rangle}}{I_0} \approx \frac{2G}{\sqrt{N}} \sum_{i=1}^4 D_i,$$

(12)

in which $G$ is much smaller than one and $N$ is of the order of a hundred in an observation over a period of $T = 100$ ns. So this current dispersion is too small to affect our spin detection. In addition, the feedback effect of the tunnelling electrons on the caged spin should also be addressed. This estimate could be done by the Fermi golden rule from the second-order perturbation [16, 17], which yields the spin decay rate $1/\tau_s = |\sum_{i=1}^4 D_i|^2/\tau_e$ with $1/\tau_e$ the tunnelling rate of the electrons. As we suppose $I_0 = 1$ nA, implying $1/\tau_e = 10^{10}$ Hz, we have $\tau_s \geq 10^{-6}$ s, which is much longer than our detection time. So this spin decay is also negligible.

Normally, the tunnelling current in usual use of STM is of the order of pA to nA, which implies that the tunnelling rate of the mobile electrons is from one electron every 100 ns to one electron every 100 ps. We have noticed that the observation of an individual electron-jump is within the reach of the current technique by the $I$–$V$ characteristic and by d$I$/d$V$ plot [24], and the minimum current the STM is able to distinguish is of the order of 0.1 pA [25].

A recent experiment [26] has shown the availability of a controllable manipulation of a single C$_{60}$ by STM. Moreover, in the absence of tunnelling of the electrons outside the fullerene, the decoherence time of the caged electron-spin is about 1 s at 7°K [9], much longer than our implementation time. So decoherence is neglected in our treatment above, while there are no experimental data for dephasing of the tunnelling electrons. A recent study [27] has shown that the spin dephasing time of the bulk two-dimensional electron gas at low temperature could be 150 ns. In our case, we require the dephasing time of the tunnelling electrons to be longer than 100 ns. Considering the data in [27] and the difference between the bound and unbound electrons, our requirement should be satisfied at very low temperature.
Compared to a previous study [10] using a modified SMT, the present scheme is, to some extent, similar but much simpler. In [10], spin flips of the mobile electrons by microwave pulses are necessary. Since the electron tunnelling is intrinsically stochastic, the decoherence due to imperfect spin flip yields the main infidelity. In contrast, no spin flip is needed in the present scheme. So the dephasing of the mobile electron is not a serious problem in our scheme. Furthermore, a magnetic field gradient $\partial B/\partial z = 4 \times 10^6 \text{T m}^{-1}$ is essential to [10], while such a magnetic field being highly stable in time and very large and homogeneous in spatial gradient is still challenging with the present technique. In contrast, the constant magnetic field required in the present scheme is fully within the reach of the current technique. In short, the present scheme is better than in [10], and the advantages are from a combination of STM and SMT: STM makes sure that the tiny variation of the current can be distinguished, and the Coulomb blockade in the SMT experiment guarantees that the change of the tunnelling current corresponds to the fullerene charged by only a single polarized electron.

4. Conclusion

In summary, we have explored the possibility of detecting the electron spin of the doped atom in a fullerene cage by STM, assisted by the Coulomb blockade and polarized current. Unlike in previous studies, we separated the whole tunnelling process into three parts. After presenting the spin-dependent tunnelling matrix elements, we have shown by specific calculation the feasibility of this detection with current STM technique. The detrimental influences from some imperfect factors have also been considered in our treatment. Since the STM is widely employed in atomic and molecular control in various systems, we argue that the main idea of the present study, i.e., the use of the STM device along with the Coulomb blockade and the polarized current, could be in principle applied to other candidate systems of QIP for detection of a single electron spin.

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