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# Electric field control of spin dynamics in a magnetically active tunnel junction

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## Abstract

The dynamics of a single spin embedded in a tunnelling junction is studied. Within a nonequilibrium Keldysh Green's function technique, we derive a quantum Langevin equation describing the spin dynamics. In the high temperature limit, it reduces to a Bloch equation, for which the spin relaxation rate, as determined by the temporal fluctuation, is linearly proportional to the temperature. In the opposite limit, the relaxation rate depends on the applied voltage, in contrast to the case of a spin in an equilibrium environment. We also show that spin-flip transition processes during electron tunnelling convert the applied electric field (i.e. voltage bias) into an effective magnetic field. Consequently, the dynamics of the spin, otherwise precessing along the static magnetic field, will have either a frequency shift proportional to the dc bias or a magnetic resonance driven indirectly by an ac electric field at the Larmor frequency  $\omega_L$ . An experiment to measure this effect is also proposed.

There is considerable experimental [1–3] and theoretical [4–10] interest in the coupling of a single spin to transport electrons. These studies are helpful in understanding the mechanism for the detection and manipulation of a single spin [11], a crucial element in spintronics and spin-based quantum information processing. Experimentally, a modulation in the tunnelling current has been observed by scanning tunnelling microscopy (STM) using a spin-unpolarized electron beam [1–3]. This observation opens up the possibility of an alternative single-spin detection technique. Theoretically, several explanations for current modulation have been proposed, including spin-orbit coupling [5], the role of the current itself [7], and magnetic scattering [8]. So far, an understanding of the mechanism for this phenomenon is still an open issue [9, 10]. The measurement of a single spin by electron spin resonance was proposed in [4]. We note that the coupling between a single spin and a supercurrent in Josephson junctions has also been studied recently [12, 13].

In earlier works involving normal conducting leads, the spin dynamics itself was treated as an isolated system, and the back-action of the transport current on the spin dynamics has not been considered. It is well known that the back action from conduction electrons is detrimental to spin coherence. However, this is the only way to manipulate the spin *electrically*. In this

paper, we concentrate on the dynamics of a single spin embedded in a normal tunnel junction. A quantum Langevin equation is derived for the single-spin dynamics. In the high-temperature limit, it reduces to a Bloch equation, for which the spin relaxation rate, as determined by the temporal fluctuation, is linearly proportional to the temperature. In the opposite limit, the relaxation rate depends on the applied voltage, in contrast to the case of a spin in an equilibrium environment. More interestingly, we show that spin-flip transition processes during electron tunnelling convert the applied electric field (i.e. bias voltage) into an effective magnetic field. Consequently, the dynamics of the spin, otherwise precessing around the static magnetic field, is tuned indirectly by an ac electric field at frequency  $\omega_0$ . This also has an implication that magnetic resonance will occur at  $\omega_0 = \omega_L$ , where  $\omega_L$  is the Larmor frequency due to the applied magnetic field. The signal at resonance can be picked up by magnetic resonance force microscopy (MRFM).

The model system under consideration consists of two normal metallic leads coupled to each other by a single spin  $\mathbf{S}$ . In the presence of a magnetic field  $\mathbf{B}$ , the spin precesses around the field direction. The system Hamiltonian can be written as:

$$H = H_L + H_R + H_S + H_T. \quad (1)$$

The first two terms are the Hamiltonians for electrons in the left and right leads,

$$H_{L(R)} = \sum_{k(p);\sigma} E_{k(p)} c_{k(p),\sigma}^\dagger c_{k(p),\sigma} \quad (2)$$

where we have denoted the electron creation (annihilation) operators in the left (L) lead by  $c_{k\sigma}^\dagger$  ( $c_{k\sigma}$ ) and those in the right (R) lead by  $c_{p\sigma}^\dagger$  ( $c_{p\sigma}$ ). The quantities  $k$  ( $p$ ),  $\sigma$  are momentum and spin indices, and  $E_{k(p),\sigma}$  are the single particle energies of the conduction electrons. The Hamiltonian of a free spin in the presence of the magnetic field is given by:

$$H_S = -g\mu_B \mathbf{B} \cdot \mathbf{S}, \quad (3)$$

where  $g$  and  $\mu_B$  are the gyromagnetic ratio and Bohr magneton of the conduction electron. The two leads are weakly coupled via the tunnelling Hamiltonian:

$$H_T = \sum_{k,p;\sigma,\sigma'} [T_{\sigma\sigma'}(k,p) c_{k\sigma}^\dagger c_{p\sigma'} + \text{H.c.}], \quad (4)$$

where the matrix elements  $T_{\sigma\sigma'}(k,p)$  transfer electrons through a magnetically active tunnelling barrier. When a spin is embedded in the tunnelling barrier, the tunnelling matrix becomes a spin operator [6, 8]:

$$\hat{T}_{\sigma\sigma'} = T_0 \delta_{\sigma,\sigma'} + T_1 \mathbf{S} \cdot \boldsymbol{\sigma}_{\sigma\sigma'} + T_2 \sigma_{\sigma\sigma'}^x. \quad (5)$$

Here the first term describes spin-independent tunnelling. The second term describes the process originating from the direct exchange coupling  $J$  of the conduction electron to the localized spin  $\mathbf{S}$ . Typically, the ratio between  $T_1$  and  $T_0$ , i.e.  $T_1/T_0$ , scales as  $J/\Phi$ , where  $\Phi$  is a spin-independent tunnelling barrier; see [14] for more details. For the spin of localized electron, it is implied that the localized level is far below the Fermi surface of the conduction electrons in both electrodes, that is,  $\epsilon_d \ll E_F$ , while the Coulomb repulsion  $U$  is so large that  $\epsilon_d + U \gg E_F$ . Therefore, the voltage bias should not exceed this energy level difference. We also take, for convenience, the respective amplitudes to be momentum independent (although it is not required). We further allow a weak external magnetic field  $B \sim 10^2\text{--}10^4$  G, which is applied along the  $z$ -direction in the  $z$ - $x$  plane perpendicular to the electron tunnelling direction ( $y$ -axis). The last term describes the spin-flip transition in the magnetically active tunnelling barrier between the leads.

When a time-dependent voltage bias is applied across the tunnelling barrier, such that  $V(t) = V_{\text{dc}} + V_{\text{ac}} \cos(\omega_0 t)$ , where  $V_{\text{dc}}$  and  $V_{\text{ac}}$  are the dc and ac components, and  $\omega_0$  is

the frequency of the ac field, a dipole will be formed around the barrier region through the accumulation or depletion of electron charge. This process results in the time dependence of single-particle energies,  $E_k = \epsilon_k + W_L(t)$  and  $E_p = \epsilon_p + W_R(t)$ , with the constraint  $W_L(t) - W_R(t) = eV_{ac} \cos(\omega_0 t)$ . However, the occupation of each state in the respective contact remains unchanged and is determined by the distribution established before the time dependence is turned on. Therefore, the chemical potentials on the left  $\mu_L$  lead and on the right lead  $\mu_R$  differ by the dc component of the applied voltage bias,  $\mu_L - \mu_R = eV_{dc}$ . The tunnelling junction with the spin then has two time scales: the Larmor precession frequency of the spin  $\omega_L = g\mu_B B$  and the characteristic frequency  $\omega_0$  of the ac field.

By performing a gauge transformation,

$$\hat{U} = e^{-i \int_0^t (\mu_L + W_L(t')) \hat{N}_L dt'} e^{-i \int_0^t (\mu_R + W_R(t')) \hat{N}_R dt'}, \quad (6)$$

with  $\hat{N}_{L(R)} = \sum_{k(p), \sigma} c_{k(p)\sigma}^\dagger c_{k(p)\sigma}$ , we obtain a new Hamiltonian:  $K = K_L + K_R + K_S + K_T$ , where  $K_{L(R)} = \sum_{k(p), \sigma} \xi_{k(p)\sigma} c_{k(p)\sigma}^\dagger c_{k(p)\sigma}$ ,  $K_S = H_S$  and

$$K_T = \sum_{kp, \sigma\sigma'} (\hat{T}_{\sigma\sigma'} e^{i\phi(t)} c_{k\sigma}^\dagger c_{p\sigma'} + \text{H.c.}),$$

with  $\phi(t) = \int_0^t [eV_{dc} + eV_{ac} \cos(\omega_0 t')] dt'$ . Note that  $\xi_{k(p)} = \epsilon_{k(p)} - \mu_{L(R)}$ , the energy being measured with respect to the different chemical potential on each side of the tunnel junction.

We now derive the effective action via the Keldysh technique. If all external fields are the same on both forward and backward branches of the Keldysh contour ( $C$ ) then  $\mathcal{Z} = \text{Tr} T_C \exp[-i \oint_C dt K_T(t)] = 1$ , where the trace is over both the fermionic bath and the local spin degrees of freedom. We first take a partial trace in  $\mathcal{Z}$  over the lead fermions (the bath) to obtain an effective spin action. In the present situation, this action represents the interaction of the magnetic spin with a nonequilibrium environment. The tunnelling contribution to the resulting spin action reads  $i\delta S = -\frac{1}{2} \oint_C dt \oint_C dt' \langle T_C K_T(\mathbf{S}(t), t) K_T(\mathbf{S}(t'), t') \rangle$ , much in the spirit of [15].

For brevity we introduce  $A_{\sigma\sigma'} \equiv \sum_{k,p} c_{k\sigma}^\dagger c_{p\sigma'}$ . The tunnelling Hamiltonian of a voltage biased junction reads

$$K_T(\mathbf{S}(t)) = \hat{T}_{\sigma\sigma'}(t) A_{\sigma\sigma'} \exp(i\phi) + \text{H.c.} \quad (7)$$

For nonmagnetic normal metallic contacts such as those we are considering here, the correction to the effective action for the spin dynamics is given by:

$$i\delta S = -i \oint_C dt \oint_C dt' \hat{T}_{\sigma\sigma'}(t) \hat{T}_{\sigma\sigma'}(t') \mathcal{D}_c(t, t') e^{i(\phi(t) - \phi(t'))}, \quad (8)$$

where  $\mathcal{D}_c(t, t') \equiv -i \langle T_C A_{\sigma\sigma'}(t) A_{\sigma\sigma'}^\dagger(t') \rangle$ , which is spin independent.

We perform the standard Keldysh manipulations, defining upper and lower spin fields  $\mathbf{S}^{u,l}$  residing on the forward/backward contours and reducing the time ordered integral over the Keldysh contour to the integral over forward running time at the cost of making the Green's function  $G$  a  $2 \times 2$  matrix. We then perform a rotation to the classical and quantum components

$$\mathbf{S}^{cl} \equiv (\mathbf{S}^u + \mathbf{S}^l)/2, \quad \mathbf{S}^q \equiv \mathbf{S}^u - \mathbf{S}^l, \quad \mathbf{S}^{cl} \cdot \mathbf{S}^q = 0, \quad (9)$$

which makes the matrix Green's functions uniquely determined by the retarded (R), advanced (A), and Keldysh (K) components  $D^{R(A)}(t, t') = \mp i\theta(\pm t \mp t') \langle [A_{\sigma\sigma'}(t), A_{\sigma\sigma'}^\dagger(t')]_{\mp} \rangle$  and  $D^K(t, t') = -i \langle [A_{\sigma\sigma'}(t), A_{\sigma\sigma'}^\dagger(t')]_{+} \rangle$ . The procedure leads to  $\delta S = \delta S_{cl} + \delta S_q$ , where

$$\delta S_{cl} = \int \int dt dt' [K_{12}(t, t') \mathbf{S}^q(t) \cdot \mathbf{S}^{cl}(t') + K_{sf}(t, t') S_x^q(t)], \quad (10)$$

and

$$\mathcal{S}_q = \int \int dt dt' K_{22}(t, t') \mathbf{S}^q(t) \cdot \mathbf{S}^q(t'). \quad (11)$$

By noting that the drift velocity of the electrons is determined by the voltage bias, the kernels in equations (10) and (11) are given by, respectively,

$$\begin{aligned} K_{12}(t, t') &= -2T_1^2 [D^R(t, t')e^{i(\phi(t)-\phi(t'))} + D^A(t', t)e^{-i(\phi(t)-\phi(t'))}] \\ &= -4T_1^2 \theta(t-t') \sum_{k,p} [f(\xi_k) - f(\xi_p)] \\ &\quad \times \sin[(\xi_k - \xi_p)(t-t') + (\phi(t) - \phi(t'))], \end{aligned} \quad (12)$$

$$\begin{aligned} K_{sf}(t, t') &= -2T_1 T_2 [D^R(t, t')e^{i(\phi(t)-\phi(t'))} + D^A(t', t)e^{-i(\phi(t)-\phi(t'))}] \\ &= -4T_1 T_2 \theta(t-t') \sum_{k,p} [f(\xi_k) - f(\xi_p)] \\ &\quad \times \sin[(\xi_k - \xi_p)(t-t') + (\phi(t) - \phi(t'))], \end{aligned} \quad (13)$$

and

$$\begin{aligned} K_{22}(t, t') &= -T_1^2 D^K(t, t')e^{i(\phi(t)-\phi(t'))} = iT_1^2 \sum_{k,p} [f(\xi_k) + f(\xi_p) - 2f(\xi_k)f(\xi_p)] \\ &\quad \times e^{i[(\xi_k - \xi_p)(t-t') + (\phi(t) - \phi(t'))]}. \end{aligned} \quad (14)$$

To describe the dynamics of the spin properly, we employ the path integral representation for the spin fields. In addition to the term  $-\oint H_S(t) dt$ , the action for a free spin also contains a Wess–Zumino–Witten–Novikov (WZWN) term [16], i.e.  $\mathcal{S}_{\text{WZWN}}$ , which describes the Berry phase accumulated by the spin as a result of motion of the spin on the sphere. We generalize this action for nonequilibrium dynamics within the Keldysh contour formalism, which can be expressed as [12]

$$\mathcal{S}_{\text{WZWN}} = \frac{1}{S} \int dt \mathbf{S}^q \cdot (\mathbf{S}^{cl} \times \partial_t \mathbf{S}^{cl}). \quad (15)$$

The total effective spin action is given by:

$$\mathcal{S}_{\text{eff}} = \mathcal{S}_{\text{WZWN}} + g\mu_B \int dt \mathbf{B} \cdot \mathbf{S}^q(t) + \delta\mathcal{S}_{cl} + \mathcal{S}_q. \quad (16)$$

As seen from equations (13) and (14), the first three terms on the right-hand side of equation (16) are real, which determine the quasi-classical equation of motion, while  $\mathcal{S}_q$  is imaginary, which stands for the fluctuations of the spin field  $\mathbf{S}^q$ . This means that the quantum effects have indeed been included even in the semi-classical approximation. We perform the Hubbard–Stratonovich transformation with an auxiliary stochastic field  $\boldsymbol{\xi}(t)$  to decouple the quadratic term in  $\mathcal{S}_q$ . The total effective action is rewritten as:

$$\begin{aligned} \mathcal{S}_{cl} &= \mathcal{S}_{\text{WZWN}} + g\mu_B \int dt [\mathbf{B} + \boldsymbol{\xi}(t)] \cdot \mathbf{S}^q(t) + \int \int dt dt' [K_{12}(t, t') \mathbf{S}^q(t) \cdot \mathbf{S}^{cl}(t') \\ &\quad + K_{sf}(t, t') S_x^q(t)], \end{aligned} \quad (17)$$

where the fluctuating random magnetic field satisfies the correlation function

$$(g\mu_B)^2 \langle \xi_i(t) \xi_j(t') \rangle = -2i K_{22}(t, t'). \quad (18)$$

Here we have assumed that the fluctuations of the three component of the field are independent and  $\langle \xi_i(t) \xi_j(t') \rangle = \langle \xi(t) \xi(t') \rangle$  with  $i = x, y, z$ .

As the spin dynamics is much slower compared to electronic processes, we set  $\mathbf{S}^{cl}(t') \simeq \mathbf{S}^{cl}(t) + (t' - t)d\mathbf{S}^{cl}/dt$ . The variational equations  $\delta S_{cl}/\delta \mathbf{S}^g(t) = 0$  imply that

$$\frac{d\mathbf{n}}{dt} = \alpha(t)\mathbf{n} \times \frac{d\mathbf{n}}{dt} + g\mu_B\mathbf{n} \times [\mathbf{B}_{\text{eff}} + \boldsymbol{\xi}(t)], \quad (19)$$

where henceforth we denote  $\mathbf{S}^{cl}$  by  $\mathbf{S} = S\mathbf{n}$ . Here we find that the coefficient  $\alpha(t)$  is given by

$$\alpha(t) = 4T_1^2 N_0^2 \sum_{nm} \int_{-D}^D \int_{-D}^D dE dE' [f(E) - f(E')] \times \frac{J_n(eV_{\text{ac}}/\omega_0) J_m(eV_{\text{ac}}/\omega_0) \sin(m-n)\omega_0 t}{(E - E' + eV_{\text{dc}} + m\omega_0)^2}, \quad (20)$$

where  $N_0$  is the density of states at the Fermi energy of the conducting leads (we assume that both leads are identical) and  $D$  is half of the bandwidth, and  $J_n$  is the  $n$ th Bessel function. At zero temperature and small bias, equation (20) reduces to

$$\alpha(t) = -8ST_1^2 N_0^2 (\omega_0/D) \sin(\omega_0 t). \quad (21)$$

From equation (20), it is evident that  $\alpha$  approaches zero with increasing temperature. Therefore, for a large band-width, the first term on the right-hand side of equation (19) drops out. This result is different from the case of a dc-biased superconducting tunnel junction, where  $\alpha(t)$  is finite, leading to spin nutation [12]. The effective magnetic field is given by  $\mathbf{B}_{\text{eff}} = \mathbf{B} + b_{\text{sf}}\hat{\mathbf{x}}$  with

$$g\mu_B b_{\text{sf}} = -4\pi T_1 T_2 N_0^2 \sum_{nm} J_n(eV_{\text{ac}}/\omega_0) J_m(eV_{\text{ac}}/\omega_0) [eV_{\text{dc}} + n\omega_0] \sin \omega_0 (n-m)t, \quad (22)$$

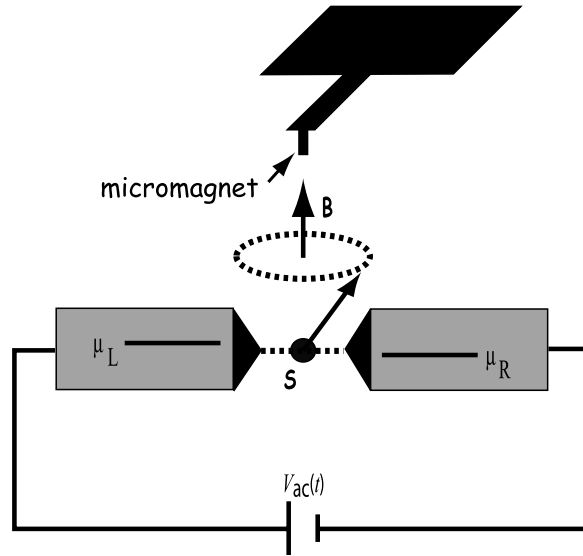
where  $\hat{\mathbf{x}}$  is the unit vector along the  $x$ -direction. Equation (19) is general in describing single-spin dynamics. We now look into the damping effect due to the tunnelling electrons. From equation (18), the power spectrum of the fluctuating magnetic field is given by  $\tilde{\chi}(\omega) = 2\pi T_1^2 N_0^2 \sum_n J_n^2(eV_{\text{ac}}/\omega_0) (\omega + n\omega_0 + eV_{\text{dc}}) \coth[(\omega + n\omega_0 + eV_{\text{dc}})/2T]$ . For the ac case, the fast oscillating part in time is irrelevant to the damping effect and has been dropped out. At high temperatures,  $\tilde{\chi}(\omega) = 4\pi T T_1^2 N_0^2$ , which is frequency independent, i.e. the time correlation is extremely short ranged. Under this circumstance, we can safely apply the Redfield approach [17], and map the above Langevin equation to the Bloch equation:

$$\frac{d\mathbf{S}}{dt} = g\mu_B [\mathbf{S} \times \mathbf{B}_{\text{eff}}] + \frac{\mathbf{S}_0 - \mathbf{S}}{T_1}, \quad (23)$$

where  $\mathbf{S}_0 = \chi_0 \mathbf{B}_{\text{eff}}$ , with  $\chi_0$  being the static magnetic susceptibility. Here we have also used the fact that the longitudinal and transverse spin relaxation time [17]:

$$\frac{1}{T_1} = \frac{1}{T_2} = 8\pi T T_1^2 N_0^2. \quad (24)$$

In a weak coupling measurement, a reasonable value of  $T_1 N_0$  is about  $10^{-3}$ . For a temperature  $T \sim 100$  K,  $1/T_{1(2)} \sim 10$  MHz. At zero temperature, the quantum fluctuation dominates in the dissipation. Therefore, equation (19) can be regarded as a quantum version of the Langevin equation. In this limit,  $\tilde{\chi}(\omega) = 2\pi T_1^2 N_0^2 \sum_n J_n^2(eV_{\text{ac}}/\omega_0) |\omega + n\omega_0 + eV_{\text{dc}}|$  is frequency dependent, the Redfield approach is inapplicable and we are unable to arrive at a Bloch equation. However, we can still estimate the spin relaxation rate to be  $2\pi T_1^2 N_0^2 (\omega_L + eV_{\text{dc}})$  for the dc case, and  $2\pi T_1^2 N_0^2 (\omega_L + eV_{\text{ac}})$  for the ac case. The crossover temperature to the quantum regime is given by  $T_c \approx (\omega_L, eV_{\text{ac}}, eV_{\text{dc}})$ , below which frequency dependence of the relaxation rate is expected. For a voltage bias of about 1 meV, the spin relaxation rate is about  $10^4$  Hz. Therefore, in both limits, the spin relaxation rate is much smaller than the Larmor frequency  $\omega_L \sim 500$  MHz for a magnetic field of 180 G.



**Figure 1.** Schematic illustration of a tunnel junction in combination with MRFM: a magnetic spin coupled to two conducting leads. The static magnetic field  $\mathbf{B}$  polarizes the spin, while an ac electric field generates an effective alternating magnetic field through the spin–flip transition during the tunnelling process of conduction electrons. A micromagnet mounted on a nanomechanical cantilever serves to couple the resonator to the single spin in the tunnelling junction.

Equations (19)–(24) constitute the central results of the paper. Several significant consequences can be concluded: the spin–flip transition process in tunnelling converts the electric field (i.e. voltage bias) into a magnetic field, which plays the role of an additional torque on the spin. When a dc voltage bias ( $V_{ac} = 0$ ) is applied, the Larmor frequency of the precessing spin will shift from  $\omega_L = g\mu_B B$  to  $\tilde{\omega}_L = g\mu_B \sqrt{B^2 + b_{sf}^2}$ . Interestingly, the frequency shift away from the expected value was indeed observed by STM experiments [1]. Whether this is the effect of the spin–flip transitions remains to be seen. By taking  $T_2 N_0 \sim 10^{-2}$  and  $eV_{dc} = 1$  meV, the effective magnetic field induced by the spin–flip transition will be of the order of 10 G, which gives a frequency shift close to the experimental value. Our result may provide a natural explanation for the observation. We also point out that the frequency shift should still be observable for a low concentration of diluted spins because it is identical for each individual spin. When an ac voltage bias ( $V_{dc} = 0$ ) is applied, the system is analogous to a spin in a static magnetic field plus an alternating magnetic field—a standard nuclear magnetic resonance (NMR) or electron spin resonance (ESR) setup [17]. In conventional ESR experiments, a number of  $10^5$  electrons is needed to generate a measurable electromotive force. The measurement of a single-spin dynamics goes beyond the conventional ESR technique. Here we propose a new experimental technique to monitor the single-spin dynamics. It has two key elements, as shown in figure 1: a tunnel junction with a single electron spin embedded in the tunnelling barrier. The static magnetic field polarizes the single spin, while the applied ac electric field across the tunnel junction serves to tune the spin dynamics. Above the tunnel junction is positioned an MRFM, which consists of a magnetic particle mounted on a nanomechanical cantilever. The magnetic particle is coupled to the single spin through a magnetic force [18]:

$$\mathbf{F}(\mathbf{r}, t) = -[\mathbf{m}_e(t) \cdot \nabla]\mathbf{B}(\mathbf{r}), \quad (25)$$

where  $\mathbf{r}$  is the distance between the micromagnet and the single spin,  $\mathbf{m}_e(t) = g\mu_B\mathbf{S}(t)$  is the magnetic moment of the single spin, and the total magnetic field  $\mathbf{B}$  consists of the external magnetic field  $\mathbf{H}$  and the magnetic field  $\mathbf{H}_t(\mathbf{r})$  generated by the micromagnet mounted on the mechanical cantilever. By assuming that  $|\partial H_t^z/\partial z| = 10^4 \text{ G } \mu\text{m}^{-1}$  and that the single spin is a localized electron, we estimate that the force signal will be 10 aN. For a mechanical cantilever with a force sensitivity  $F_n \sim 100 \text{ zN}$ , a bandwidth of the order of  $10^4 \text{ Hz}$ , which might be determined by the intrinsic frequency of the cantilever, is important for single-electron spin detection by MRFM. In view of the progressing improvement in the MRFM technique, a test for our results is within experimental reach. The direct observation of the magnetic resonance signal from a single spin, *in the absence of an externally applied alternating magnetic field*, will provide strong evidence of spin-flip transitions across the tunnelling junction, which will be very helpful in understanding the mechanism for the tunnelling current modulation observed in STM [1–3].

We have studied the back-action effect of tunnelling current on the single-spin dynamics. The spin-flip transition in the tunnelling process of conduction electrons generates an effective magnetic field, which manipulates the motion of the single spin. In addition, we have considered the dissipation effect of the tunnelling electrons, which naturally leads to a Bloch equation at high temperatures, while in the opposite limit the relaxation rate depends on the combination of the Larmor frequency and the applied voltage bias. Our conclusion can also be applied to a magnetic cluster for which a single spin operator can still be defined if the constituent spins have strong magnetic correlation.

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