# Detecting Larmor Precession of a Single Spin with a Spin-Polarized Tunneling Current *

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*Detection and manipulation of a single spin is essential for spin-based quantum information and computation. We propose an experimental method to detect the Larmor precession of a single spin with a spin-polarized tunneling current. The calculation demonstrates that the modulation of the tunneling current is significantly increased as compared with the non-spin-polarized current. The signal-to-noise ratio in the current power spectrum will be dramatically improved by this method. The longitudinal relaxation time can be measured by temporal control of spin orientation with pulses of magnetic field or radio frequency electromagnetic wave, while the transverse relaxation time can be derived by analyzing the resonant peak profile with a spectrum analyzer. Such ability in characterizing spin relaxation is of particular interest in providing insights into the coherence/decoherence of a single spin.*

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As a potential building block for quantum information and computation,[1,2] single spin has attracted significant interest in the past decade. Various experimental techniques have been developed for single spin detection and control, such as nano-devices based on a single electron quantum dot,[3] magnetic resonance force microscopy,[4] spin-flip spectroscopy,[5] spin polarized scanning tunneling microscope (SP-STM),[6] and electron spin resonance scanning tunneling microscopy (ESR-STM).[7,8] Among these techniques, ESR-STM is unique for its potential ability in accessing spin coherence with the highest spatial resolution. In an ESR-STM, a paramagnetic spin undergoes Larmor precession in a magnetic field and induces a modulation in the tunneling current at a radio frequency, which can be detected with a preamplifier and a spectrum analyzer. Such a modulated signal has been observed in a few systems including defects on the semiconductor surface,[9,10] radicals of an adsorbed organic molecule,[11] and transition metal atoms.[12] However, the reported signal-to-noise (S/N) ratio up to date is mostly poor, and the underlying mechanism is still controversial.[13,14] Further investigations, especially the design of a novel experimental method for achieving a higher S/N ratio, is deeply deserved for the advancement of ESR-STM and the exploration of single spin dynamics.

In this Letter, we propose for the first time an experimental method to detect the Larmor precession of a single spin with a spin-polarized tunneling current. The modulation of tunneling current due to the spin precession is calculated by employing the Tersoff–Hamann model. Experimental methods to derive the longitudinal relaxation time $T_1$ and transverse relaxation time $T_2$ of a single spin are further illustrated. The influence of various experimental parameters, such as temperature and tunneling current magnitude, on the spin precession signal are also discussed. By using a spin-polarized tunneling current, the S/N ratio is significantly increased and the theoretical derivation is also simplified.

Figure 1 shows our proposed schematic diagram of the experimental setup. Compared to the non-spin-polarized STM tip used in previously reported ESR-STM experiments, a spin-polarized STM tip with polarization rate $P$ is implemented here. The spin polarization direction of the tip is parallel to the sample surface, which can be achieved by depositing a magnetic film with specific thickness on a tungsten tip.[15] A relatively small magnetic field (several hundreds of Gauss) perpendicular to the sample surface is generated by an electromagnet. A passive filter separates the direct-current part and the oscillating rf part of the tunneling current. The dc part is fed into normal STM electronics for feedback control and topographic data acquisition. An rf preamplifier and a spectrum analyzer are employed to detect and analyze the precession signal.

The Hamilton of a single spin in magnetic field $\boldsymbol{B}$ can be written as

$$\hat{H} = -\boldsymbol{\mu}\cdot\boldsymbol{B} = \frac{geB}{2m_e}\hat{S}_z, \tag{1}$$

where $\boldsymbol{\mu}$ is the magnetic momentum of an electron, $g$ is the $g$-factor, $e$ is the electron charge, $m_e$ is the electron mass, and $\hat{S}_z$ is the spin operator. The direction of the magnetic field is defined along the $Z$ axis. The time-dependent wave function of the spin can be described with the Bloch sphere model[16] as

$$|\alpha(t)\rangle = \cos\frac{\theta}{2}|0\rangle + \sin\frac{\theta}{2}\exp\Big[i\Big(\varphi + \frac{geB}{2m_e}t\Big)\Big]|1\rangle, \tag{2}$$

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where $|0\rangle$ and $|1\rangle$ are eigenstates for spin-up and spin-down states, respectively; $\theta$ and $\phi$ are polar and azimuth angles in the Bloch sphere, as shown in Fig. 2(a). The spin state undergoes a precession around the $Z$ axis at the Larmor frequency $\omega_0 = geB/(2m_e)$. As mentioned above, the magnetization of the tip is oriented parallel to the sample surface. If we set the $X$ axis along this direction and the tip has a polarization rate $P$, then the mixed tip state is a mixture of $|x^+\rangle$ and $|x^-\rangle$ with probability $\frac{1+P}{2}$ and $\frac{1-P}{2}$, respectively, where $|x^\pm\rangle$ is the spin state along the $\pm X$ direction. By its definition, $P$ is a real number that varies from 0 to 1. In the case of a small magnetic field (several hundreds of Gauss), the tip spin is fixed and does not undergo Larmor procession, and the tip spin density matrix

$$\rho_{\rm tip} = \frac{1}{2}\begin{pmatrix} 1 & P \\ P & 1 \end{pmatrix}$$

can be treated as a constant.

The total tunneling current is calculated by employing the Tersoff–Hamann model.[17]

$$I(V) = \frac{2\pi e}{\hbar}\sum_{\mu,\nu}|M_{\mu\nu}|^2\delta(E_\mu^t - E_\nu^s - eV) \cdot [f(E_\mu^t) - f(E_\nu^s)], \tag{3}$$

where $E_\mu^t(E_\nu^s)$ is the energy level of the tip (sample) state, and $f(E)$ is the Fermi distribution function. The tunneling matrix is given by

$$M_{\mu\nu} = \langle\psi_\mu^t|U_T|\psi_\nu^s\rangle, \tag{4}$$

where $\psi_\mu^t(\psi_\nu^s)$ is the electronic wave function of the tip (sample), and $U_T$ is the potential of the tip. The electronic wave functions can be written as a direct product of the spatial part and spin part,

$$\psi_\nu^s = \begin{pmatrix} \cos\frac{\theta}{2} \\ \sin\frac{\theta}{2}e^{i(\varphi+\omega_0 t)} \end{pmatrix} \otimes \xi(\boldsymbol{r}),$$

$$\psi_\mu^t = \begin{cases} |x^+\rangle \otimes \chi(\boldsymbol{r}), & \text{with probability } \frac{1+P}{2}, \\ |x^-\rangle \otimes \chi(\boldsymbol{r}), & \text{with probability } \frac{1-P}{2}, \end{cases} \tag{5}$$

where $\chi(\boldsymbol{r})$ and $\xi(\boldsymbol{r})$ are the spatial wave functions of the tip and the sample. For simplicity, the spin-flip process during tunneling is ignored in the present consideration, which results in diagonalization of the tip potential $U_T$ in spin space. The tunneling matrix can thus be simplified to

$$|M|^2 = \langle\alpha(t)|\rho_{\rm tip}|\alpha(t)\rangle * |\langle\chi|U_T|\xi\rangle|^2,$$

where the spin-related part $\langle\alpha(t)|\rho_{\rm tip}|\alpha(t)\rangle$ has a form of

$$\frac{1}{2} + \frac{P\sin\theta}{2}\cos(\omega_0 t).$$

The total tunneling current can be written as

$$I = I_0[1 + P\sin\theta\cos(\omega_0 t)], \tag{6}$$

where $I_0$ is defined as the spin-averaged part of the tunneling current. From Eq. (6), it is clear that tunneling current has an oscillating component at the Larmor frequency $\omega_0$, as shown in Fig. 2(b). Under a typical magnetic field of 100 Gauss and assuming a $g$-factor of 2, the Larmor frequency is around 280 MHz. Such a signal can be detected with an rf preamplifier and further analyzed with an rf spectrum analyzer, where the peak frequency and the resonance profile can be derived to reveal the value of $g$-factor and the spin lifetime, respectively. Equation (6) also shows that the amplitude of the oscillating current is proportional to the polarization rate $P$. In the conventional ESR-STM experiment, it is assumed that the current polarization is caused by current fluctuation, which is estimated to be on the order of 0.01.[14] In contrast, a spin polarized tip typically has a polarization rate $P$ of about 0.2–0.3,[6] so our method can dramatically enhance the S/N ratio.

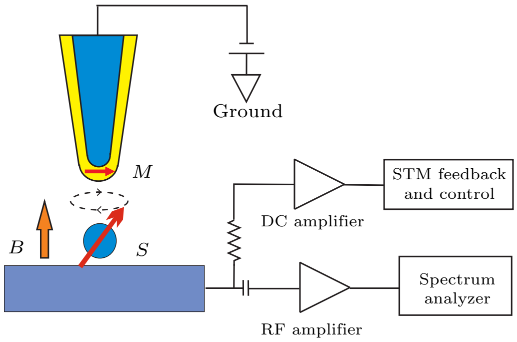


**Fig. 1.** Schematic diagram of the proposed experimental setup. The tip magnetization direction is parallel to the sample surface, while the applied magnetic field is perpendicular to the sample surface. A passive filter separates the dc part and the rf part of the tunneling current. The rf signal is detected and analyzed by an rf preamplifier and a spectrum analyzer.

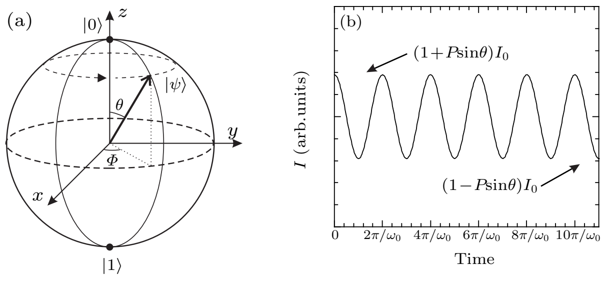


**Fig. 2.** (a) Single spin precession under the Bloch sphere model. (b) Total tunneling current as a function of time. $I_0$ is the dc part of the tunneling current. The oscillating part is at the Larmor frequency $\omega_0$, and its amplitude depends on the tip polarization rate $P$ and the polar angle $\theta$ in the Bloch sphere.

When a single spin is perturbed by its surroundings, its effective $g$-factor will deviate from that of a free electron spin. Since the Larmor frequency changes linearly with the magnetic field, the effective $g$-factor of a single spin can be detected by analyzing the resonance frequency while modulating the magnetic field

$\boldsymbol{B}$ (to eliminate the influence from a stray field),

$$g = \frac{2m_e}{e}\frac{\Delta\omega_0}{\Delta B}. \tag{7}$$

As a result of spin-environment interaction, such as electron-orbital coupling, hyperfine coupling with surrounding nuclear spins, or electron-phonon coupling, the Larmor precession of a single spin has a finite lifetime. In general, there exist two spin relaxation paths, named as longitudinal spin relaxation and transverse spin relaxation. The longitudinal spin relaxation is the process that the spin state evolves from its initial state to a thermal equilibrium state with a time constant $T_1$. The transverse spin relaxation, also known as the spin-spin relaxation, usually results in the exponential decay of transverse magnetization with a time constant $T_2$. For a single spin, the transverse spin relaxation is characterized by a random phase jump of its spin state. By detecting relaxation time $T_1$ and $T_2$, we can obtain the information about spin-substrate interaction, spin-spin interaction, spin-current interaction, and spin coherence/decoherence.

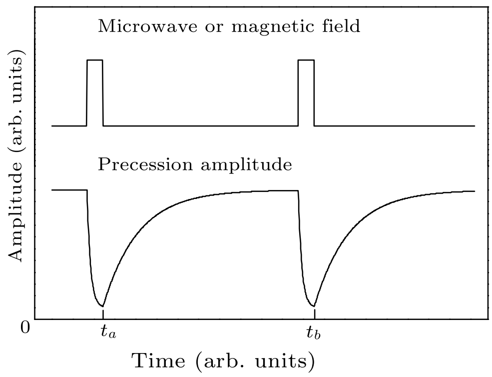


**Fig. 3.** Method to measure the longitudinal relaxation time $T_1$ by monitoring the spin precession signal. A microwave pulse or magnetic field pulse is applied to pump spin into excited state in which the precession signal is suppressed. After the pulse is switched off, the spin relaxes to the thermal equilibrium by longitudinal relaxation and the precession signal recovers to its normal value. The characteristic time of this recovery process is the longitudinal spin relaxation time $T_1$.

Typically, the longitudinal spin relaxation time $T_1$ is much longer than the transverse spin relaxation time $T_2$. It is hard to derive $T_1$ from resonance peak broadening as it is dominated by $T_2$. In STM configuration, $T_1$ can be measured by applying a microwave pulse or a magnetic field pulse, as shown in Fig. 3. The microwave pulse can be generated by an electromagnetic coil or a thin metal wire (about 10 µm in diameter) near the tunneling junction. The pulse width is set to $\pi/2$, which drives a spin state from the $xy$-plane to the $z$-axis[18] as presented in the Bloch sphere. After the pulse is switched off, the spin state and thus the precession amplitude evolve to the equilibrium with time constant $T_1$, as shown in Fig. 3

In the Bloch sphere, the spin state evolution can be characterized with the polar angle $\theta$ that can be derived from the amplitude of the Larmor precession. At the thermally equilibrium state, the polar angle $\theta$ in the Bloch sphere is determined by the Boltzmann distribution

$$\tan^2\frac{\theta}{2} = e^{-\Delta E/2kT}, \tag{8}$$

where $\Delta E = geB\hbar/(2m_e)$ is the Zeeman energy, $k$ is the Boltzmann constant, and $T$ is sample temperature. The temperature dependence of $\sin\theta$, which is linear to the amplitude of the precession signal, is shown in Fig. 4 with a different magnetic field $B$. In an STM operated at a temperature of about 50 mK and under a magnetic field of 200 Gauss, the polar angle $\theta$ is about 89.2°. By turning on a strong magnetic field pulse, say 5 Tesla, the spin can be polarized with $\theta$ decreasing to 8°. After the pulse is switched off, the spin decays with a time constant $T_1$ to the equilibrium state where the angle $\theta$ returns to 89.2°.

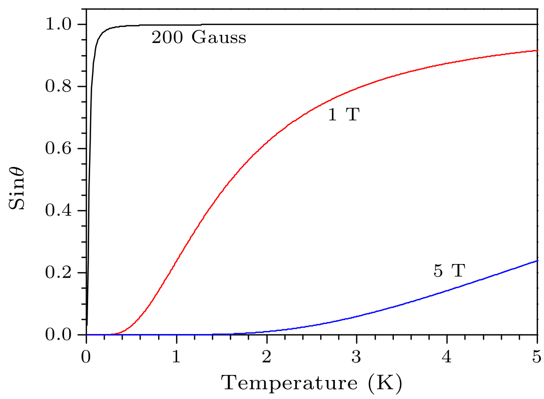


**Fig. 4.** Temperature dependence of the Larmor precession signal under different magnetic fields of 200 Gauss, 1 T, and 5 T.

In contrast to the longitudinal relaxation, the transverse spin relaxation is represented by the random phase jump of the spin precession. Such random phase jump will result in a perturbation in the oscillating tunneling current as follows:

$$I = I_0[1 + P\sin\theta\cos(\omega_0 t + \varphi(t))], \tag{9}$$

where $\varphi(t)$ undergoes random jumps. Figure 5(a) shows the tunneling current as a function of time under transverse spin relaxation. For weak interactions, we can assume that these phase jumps are independent to each other, as in a Poisson process. Therefore, the possibility that two adjacent phase jumps occurring with a time interval $\tau$ is

$$\sigma(\tau) = e^{-\tau/T_2}/T_2, \tag{10}$$

where $T_2$ is the average phase jump interval. The current autocorrelation function can be calculated by

$$\begin{aligned} G(\tau) &= \lim_{T\to\infty}\frac{1}{T}\int_0^T I^*(t)I(t+\tau)dt \\ &= \frac{I_0^2P^2\sin^2\theta}{2}e^{-\frac{|\tau|}{T_2}}\cos(\omega_0\tau). \end{aligned} \tag{11}$$

According to the Wiener–Khinchin theorem, the Fourier transformation of the correlation function gives the power spectrum of the tunneling current at the proximity of $\omega_0$,

$$S(\omega) = \frac{\lambda}{\lambda^2 + (\omega - \omega_0)^2} I_0^2 P^2 \sin^2\theta, \tag{12}$$

with $\lambda = 1/T_2$. Such a power spectrum can be directly measured with the rf spectrum analyzer. The resonance peak position in Eq. (12) is the Larmor frequency, hence this can be used to calculate the effective $g$-factor as mentioned above. The resonance peak has a Lorentz shape broadening with a full width at half maximum (FWHM) of $2\lambda$, as shown in Fig. 5(b). The intensity of the resonance depends on the magnitude of the current, the spin polarization of the tip, and the polar angle of the single spin as determined by the magnetic field and the temperature (Eq. (8)).

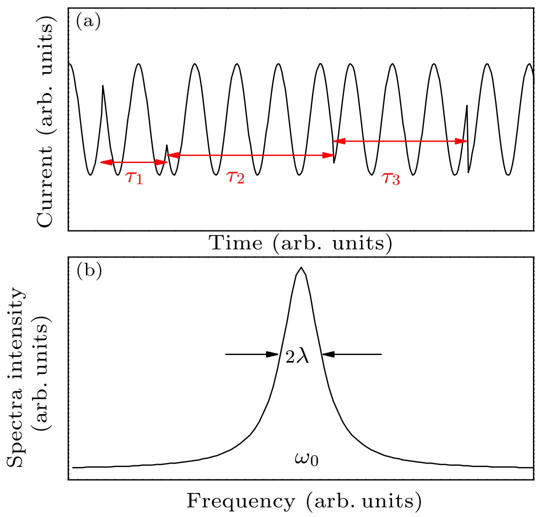


**Fig. 5.** (a) Tunneling current as a function of time under random phase jumps. (b) Power spectrum resulted from transverse spin relaxation. The peak central is at the Larmor frequency, the resonance has a Lorentz line profile with FWHM of $2\lambda$.

Normally, the longitudinal spin relaxation will also contribute to the peak broadening as measured with a spectrum analyzer. The longitudinal relaxation time $T_1$, however, is much longer than $T_2$ in most practical systems. Thus, the spectra broadening and the measured FWHM are mainly determined by the transverse spin relaxation. The influence of longitudinal spin relaxation on the measured spectra profile can be treated as a perturbation and can be subtracted.

One of the sources of transverse spin relaxation in our measurement setup is the spin-polarized tunneling current. The interaction between the spin-polarized tunneling electron and the spin will cause extra phase jumps in spin precession. The mean time interval between two phase jumps is inversely proportional to the tunneling current magnitude $T^* \propto e/I$. The induced peak broadening then is proportional to the current, $\lambda = \alpha_s I/e$, where $\alpha_s$ is the coupling coefficient that characterizes the strength of the spin-current interaction. To understand the spin-current interaction and to elucidate its contribution to the total spectra broadening, it is necessary to obtain an accurate value of $\alpha_s$. This can be achieved by measuring the differential change in spectra broadening while modulating the current magnitude. We have

$$\alpha_s = e\frac{d\lambda}{dI}. \tag{13}$$

For a typical experimental setup, $I{\sim}1$ nA, FWHM$\sim$300 kHz, we estimate that $\alpha_s$ is in the order of $10^{-5}$.

Besides electron spins, the dynamic behavior of a nuclear spin can also be investigated in this proposed setup. Because of its relatively long spin lifetime,[19] nuclear spin has long been proposed as a quantum bit. The hyperfine nuclear-spin interaction makes it possible to detect the behavior of a nuclear spin through an electron. It has been demonstrated that in some materials, such as tellurium donors in silicon,[20] the hyperfine interaction is strong enough to generate an observable change in electron spin resonance. We expect that such modulation in electron precession frequency due to the nuclear spin state can also be detected with our proposed experimental method.

In summary, an experimental method has been proposed to detect the Larmor precession of a single spin with a spin-polarized tunneling current. The signal of precession can be significantly increased by employing a highly spin-polarized current. The spin coherence/decoherence of a single spin center, due to both longitudinal and transverse relaxations, can be revealed with temporal pulse control and spectra analysis. Such an experimental method may in the near future play a significant role in exploring quantum information and computation based on individual electron spin or nuclear spin.

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