ESR-STM of a single precessing spin: Detection of exchange-based spin noise

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Electron Spin resonance scanning tunneling microscopy (ESR-STM) is an emerging technique which is capable of detecting the precession of a single spin. We discuss the mechanism of ESR-STM based on a direct exchange coupling between the tunneling electrons and the local precessing spin **S**. We claim that since the number of tunneling electrons in a single precessing period is small (~ 20), one may expect a net temporary polarization within this period that will couple via exchange interaction to the localized spin. This coupling will randomly modulate the tunneling barrier and create a dispersion in the tunneling current which is a product of a Larmor frequency component due to the precession of the single spin and the dispersion of the spin of the tunneling electrons. This noise component is spread over the whole frequency range for random white noise spin polarization of electrons. In the opposite case where the power spectrum of the spins of the tunneling electrons has a peak at zero frequency an elevated noise in the current at ω_L will appear. We discuss the possible source of this spin polarization. We find that for relevant values of parameters the signal-to-noise ratio in the spectral characteristic is 2–4 and is comparable to the reported signal to noise ratio.[1,2] The magnitude of the current fluctuation is a relatively weak increaing function of the dc current and magnetic field. The linewidth produced by the back action effect of tunneling electrons on the precessing spin is also discussed.

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There is a growing realization that the technique of electron spin resonance scanning tunneling microscopy (ESR-STM) is capable of detecting the precession of a single surface spin by modulating the tunneling current at the Larmor frequency. This technique was successful in measuring Larmor frequency modulations in defects in semiconductor surfaces[1] and in paramagnetic molecules.[2] The increasing interest in this technique is due to the possibility to detect and manipulate a single spin.[3]

The alternative technique that allows one to detect single spin is the optically detected magnetic resonance (ODMR) spectroscopy in a single molecule.[5] In comparison, ESR-STM has the unique ability to correlate spectroscopic information with spatial information, detected at the atomic level. It also allows one to manipulate the position of the spin centers at the atomic level.[4]

There have been several proposals for the mechanism of detection. One is a polarization of the mobile carriers through spin-orbit coupling and modulation of the LDOS as a result of the precession.[6] Another one is the interference between two resonant tunneling components through the magnetic-field-split Zeeman levels.[7] Both of these mechanisms rely on a spin-orbit coupling to couple a local spin **S** to the conduction electrons and have assumed *no spin polarization of tunneling electrons*. Recently, however, Durkan and Welland[2] observed a strong signal in a system with a substantially smaller spin-orbit coupling than what was assumed in the calculations.[6,7] Motivated by these experiments we addressed a question: what is the role of the *direct* exchange interaction between the localized spin and the tunneling electrons. The exchange interaction has a tremendous influence on the physics of conducting substances when magnetic impurities are present[8] and it is natural to ask here: Does exchange interaction play a role in ESR-STM also?

We find that a direct Heisenberg exchange interaction between the localized spin and conduction electrons is capable of producing a modulation of the tunneling current. The qualitative difference compared with the previous models is that we consider temporal fluctuations of the spin polarization of the electrons that are tunneling between the tip and surface. The spin-orbit interaction is irrelevant for this consideration. We argue in this paper that although the spin polarization of the tunneling electrons is zero in the long time limit, it is not zero on the scale of the period of the precession, typically $1/\omega_L \sim 2$ ns. On this time scale there are very few electrons that pass nearby the localized spin. There exists a temporary spin polarization of the tunneling electrons which may interact through an exchange interaction with the localized spin center.

It is important to point out that the ESR-STM technique performs a *noise spectroscopy*. We do not drive the single spin with an external coherent rf field, and we are basically detecting an incoherent phenomenon (we avoid here the question of the meaning of this concept on a single-particle level). There have been several demonstrations in the past of detecting magnetic resonance with noise spectroscopy.[9] We argue that it is possible to get a noise-related signal from an exchange interaction between the tunneling electrons and the localized surface spin center.

The overlap of the electron wave function in the tip and surface, separated by a distance d, is exponentially small and is given by a *spin-dependent* tunneling matrix element

$$\hat{\Gamma} = \Gamma_0 \exp\left[- \sqrt{\frac{\Phi - J\mathbf{S}(t)\hat{\boldsymbol{\sigma}}}{\Phi_0}} \right], \tag{1}$$

where we consider the spin $\mathbf{S}(t)$ in the magnetic field $B||z$, precessing with the Larmor frequency $\omega_L = g\mu_B B$, $\hat{\Gamma}$ is understood as a matrix in spin indexes, Φ is the tunneling barrier height, Φ is typically few eV, and we assume $\Phi = 4$ eV, and $\Phi_0 = \hbar^2/8md^2$ is the energy related to the distance between tip and surface d.[11] The exchange term in the exponent is small compared to the barrier height and we can expand the exponent in JS. Explicitly $\hat{\Gamma}$ can be written as

$$\hat{\Gamma} = \Gamma_0 \exp[-(\Phi/\Phi_0)^{1/2}]\left[\cosh\left(\frac{JS}{2\Phi}\sqrt{\frac{\Phi}{\Phi_0}}\right) + \hat{\boldsymbol{\sigma}}\mathbf{n}(\mathbf{t})\sinh\left(\frac{JS}{2\Phi}\sqrt{\frac{\Phi}{\Phi_0}}\right)\right], \tag{2}$$

where Γ_0 describes spin-independent tunneling in the absence of J. Note that the dynamics of the spin is now absorbed in the time dependence of the unit vector $\mathbf{n}(t):\mathbf{S} = \mathbf{n}S$. Let us now give a simple qualitative description of the effect we address here. Leaving aside the constants we see that the tunneling conductance has a part that depends on the localized spin,

$$\delta I(t) \sim \mathbf{n}(t)\boldsymbol{\sigma}(t), \tag{3}$$

and in a scalar product $\mathbf{n}(t)\boldsymbol{\sigma}(t) = n^z(t)\sigma^z(t) + n^x(t)\sigma^x(t) + n^y(t)\sigma^y(t)$ only a transverse part, which depends on the x,y components of the localized spin and the spin of the tunneling electrons, will describe precession in a magnetic field ($B||z$ is assumed). We will focus on the transverse terms below. To make the argument as simple as possible we will assume at the moment that the spin $\mathbf{S}(t)$ is a simple periodic function of time, $n_x(t) = n_\perp \cos(\omega_L t), n_y(t) = n_\perp \sin(\omega_L t)$, with period $T = 2\pi/\omega_L$. It is convenient to introduce a time average of the current over T, $\Delta I = 1/N \Sigma_{i=1}^{N} \delta I(t_i)$, where the sum over $i = 1 \ldots N$ is over the number of electrons that will tunnel between the tip and surface in time T, with an average $\overline{N} = I_0 T$, which is dependent on the dc current in the system I_0:

$$\Delta I = \frac{1}{N}\sum_{i=1}^{N} \sigma^x(t_i) n^x(t_i) + (x \to y). \tag{4}$$

This term represents the fluctuations of tunneling current due to the interaction with the single precessing spin. Then the dispersion of the current, which depends on the precessing components, is given by the dispersion of the quantity $\Sigma_{i,j=1}^{N} n_x(t_i) n_x(t_j)[\sigma^x(t_i)\sigma^x(t_j)]$. If the spin-wave functions of the tunneling electrons are not correlated between different tunneling events, we find

$$\left(\sum_{i=1}^{N} \sigma^x(t_i) n_x(t_i)\right)^2 + (x \to y) \sim \overline{N}. \tag{5}$$

Therefore the dispersion of the current due to the exchange interaction between the localized precessing spin and the spin of the tunneling electrons is

$$\frac{\overline{\langle \Delta I^2 \rangle}}{I_0^2} \sim \langle (n^x)^2 \rangle \frac{\overline{N}}{\overline{N}^2} + (x \to y) \sim \frac{1}{\overline{N}}, \tag{6}$$

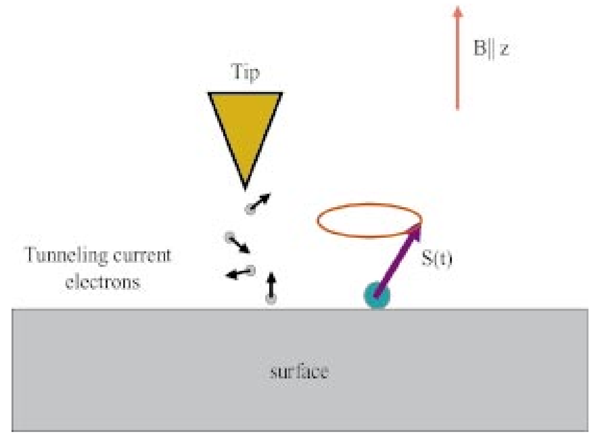

FIG. 1. (Color) Schematics of the ESR-STM experiment. The fluctuations in the spin polarization of the tunneling electrons at the time scale of the precession, T, will be nonzero and will scale as $1/\overline{N}$. $\overline{N} = 1/eI_0T$ is the average number of electrons tunneling between the tip and surface during one precession cycle. Once the tip is positioned close to the localized spin, the exchange interaction between the localized spin and the tunneling electrons will modulate the tunneling current. The conditions in which this random modulation will create a ω_L peak are discussed in the text.

where the result is normalized to the dc current magnitude. We find that the magnitude of the fluctuations $(\overline{\langle \Delta I^2 \rangle})^{1/2}$ is on the scale of a few percent of the dc current for experimentally relevant values of the parameters; see Eq. (11). If the spins of the tunneling electrons are totally uncorrelated, this noise component will be smeared over the whole frequency range. However, as we show below, if some tunneling electron spin polarization exists, a strong noise peak will appear at ω_L.

We argue that this simple mechanism is in agreement with several experimental observations, such as the intensity of the signal and the signal's linewidth. From Eq. (6) we can immediately conclude that the mean square fluctuation of the spin-dependent current is a *weak increasing function of both the magnetic field and dc current* with power $\frac{1}{2}$:

$$(\overline{\langle \Delta I^2 \rangle})^{1/2} \sim (I_0 B)^{1/2}. \tag{7}$$

We will now give a derivation of the results. Consider the setup that is used in ESR-STM, Fig. 1. Since the tip is very close to the magnetic site, we assume that the Heisenberg exchange coupling between conduction electrons that tunnel across the barrier and the localized spin $\mathbf{S} = \mathbf{n}S$ is typically on the order of a fraction of eV. Hence the effective barrier, seen by the tunneling electron, will depend on the spin of the conduction electron.

Let us first discuss the relevant time scales of the problem. For $I_0 = e/\tau_e = 1$ nA current the electron tunneling rate is $1/\tau_e \sim 10^{10}$ Hz. The electron precession frequency at field $B \sim 200$ G is about $\omega_L/2\pi = 500$ MHz, $T = 2 \times 10^{-9}$ sec. Per single precession cycle there are about $\overline{N} = 20$ electrons that tunnel between the tip and the surface. As we indicated above, the fluctuation of the electron spin is appreciable, $\sim (\overline{N})^{1/2} \sim 4$, for such a small number of electrons.

(a) *Spin-dependent tunneling*. We model the effect of the Heisenberg interaction as a spin-dependent tunneling barrier. For practical purposes we can assume that the precessing localized spin $\mathbf{S}(t)$ is slow compared with the typical tunneling time of electrons.

The Hamiltonian we consider describes a spin-dependent tunnneling matrix element between the tip (L electrode) and the surface (R electrode):

$$H=\sum_{\mathbf{k},\alpha}\epsilon(\mathbf{k})c^{\dagger}_{L\alpha}(\mathbf{k})c_{L\alpha}(\mathbf{k})+(L\rightarrow R)$$

$$+\sum_{\mathbf{k},\mathbf{k}'}c^{\dagger}_{L\alpha}(\mathbf{k})\Gamma_{\alpha\beta}c_{R\beta}(\mathbf{k}'). \quad (8)$$

We assume that the magnetic field is along the z axis: $B||z$. The tunneling current operator will contain a spin-independent part that we omit hereafter and a spin-dependent part

$$\delta\hat{I}(t)=\Gamma_1\mathbf{n}(t)\boldsymbol{\sigma}, \quad (9)$$

where $\Gamma_1=\gamma_0\sinh[(JS/2\Phi)(\Phi/\Phi_0)^{1/2}]$. We introduced a renormalized $\gamma_0=\Gamma_0\exp[-(\Phi/\Phi_0])^{1/2}]$ that determines the dc current at a given bias V: $I_0=\gamma_0V$. The current-current correlator, normalized to dc current, is then

$$\frac{\overline{\langle\delta\hat{I}(t)\delta\hat{I}(t')\rangle}}{I_0^2}=\left[\sinh\left(\frac{JS}{2\Phi}(\Phi/\Phi_0)^{1/2}\right)\right]^2$$

$$\times\sum_{i,j=x,y,z}\langle n^i(t)n^j(t')\rangle\overline{\sigma^i(t)\sigma^j(t')}. \quad (10)$$

We explicitly separate the averaging over the dynamics of the localized spin $\langle AB\rangle$ and the averaging over the ensemble of the tunneling electrons $\overline{AB}$. For the spin dynamics we use $\langle n^x(t)n^x(t')\rangle\sim\cos[\omega_L(t-t')]\exp(-\gamma|t-t'|)$ and similarly for y component. For the averaged-over-time T current-current correlator we will have a result similar to Eq. (10) with $\delta I\rightarrow\Delta I$ [see definition in Eq. (4) and above]. This brings an additional factor of $1/\overline{N}$.

To estimate the magnitude of the current fluctuations due to the coupling to the localized spin we will take $J\sim0.1$ eV. This is typical for an exchange interaction in semiconductors and metals.[13] The barrier height $\Phi\simeq4$ eV, spin $S=1/2$. To estimate $\Phi_0=\hbar^2/8md^2$ we assume a typical tunneling distance $d=4$ Å. This yields $\Phi_0\simeq0.1$ eV. For these parameters we find

$$\frac{(\overline{\langle\Delta I^2\rangle})^{1/2}}{I_0}\simeq\frac{2}{\sqrt{\overline{N}}}\sinh\left(\frac{JS}{2\Phi}(\Phi/\Phi_0)^{1/2}\right)\simeq0.01 \quad (11)$$

and $\Gamma_1=0.02\gamma_0$. The magnitude of the fluctuation is in the 10 pA range for a tunneling current of $I_0=1$ nA and is within the observed range.[1,2] This is a magnitude of the fluctuating current in the time domain due to the exchange interaction between the precessing single spin and the tunneling electrons. This current fluctuation will give a peak at the Larmor frequency once there exists some spin polarization in the tunneling current on a time scale of the relaxation time of the single spin. For finite spin polarization, the size of the noise component will be larger also. Thus the value of 10 pA represents a minimal intensity. Actual signals will increase with the degree of spin polarization of the tunneling electrons.

(b) *Back action effect of the tunneling current on the spin*. One can use the tunneling Hamiltonian, Eq. (8), to estimate the decay rate of the localized spin state due to the interaction Γ_1. To second order this calculation is equivalent to the Fermi golden rule calculation and we have $1/\tau_s=\pi\Gamma_1^2N_LN_R$ eV. Similarly, the dc tunneling current I_0 is given by the tunneling rate of conduction electrons, $1/\tau_e=\pi\gamma_0^2N_LN_R$ eV, where $N_{L,R}$ is the density of states at the Fermi level of the tip and surface, respectively.[12] One finds, by combining these two equations,

$$\frac{1}{\tau_s}=\frac{1}{\tau_e}\frac{\Gamma_1^2}{\gamma_0^2}\simeq4\times10^{-4}\frac{1}{\tau_e}. \quad (12)$$

This result has a simple interpretation: The electron tunneling rate $1/\tau_e\sim10^{10}$ Hz gives the attempt rate for the tunneling electrons. The probability to flip the localized spins is proportional to Γ_1^2, which gives Eq. (12) for the linewidth. We estimate $1/\tau_s\simeq4\times10^6$ Hz. This estimate is within an order of magnitude of the reported linewidth.[1,2] Given the uncertainty in the parameters used we believe this is a reasonable result; for example, if we take $J=0.05$ eV, we will find $(\overline{\langle\Delta I^2\rangle})^{1/2}/I_0\sim10^{-2}$ and the linewidth will change by factor of 4, $1/\tau_s\simeq10^6$ Hz. The linewidth will increase with the increased spin polarization of tunneling electrons. Future experiments will help to clarify the linewidth dependence on J,B, and other parameters.

In the above discussion, we have assumed that the dynamics of the local spin is controlled by the magnetic field only and no decoherence mechanism, except back action, is included. In practice there are other sources of dephasing of a precessing spin that will add to the back action effect of tunneling electrons and details will depend on the specific material. In this context we point out that the ESR linewidths are quite narrow for magnetic centers in semiconductors and insulators even at room temperatures, typically few MHz.[14] In the case of a single spin the linewidth will be narrower as the inhomogeneous broadening is not an issue in this case.

For any source of decoherence, be it back action scattering or interaction with the environment, the localized spin will be scattered from the ground state and produce mixed states with nonzero $\langle S_x\rangle,\langle S_y\rangle$, required to have precessing spin. No phase coherence between different precessing spins is required as we are looking at the single site.

(c) *Spectral density of the current*. The Fourier transform of the current-current correlator will give a power spectrum of the current fluctuation, Eq. (10):

$$\frac{\overline{\langle I_\omega^2\rangle}}{I_0^2}=\left[\sinh\left(\frac{JS}{2\Phi}(\Phi/\Phi_0)^{1/2}\right)\right]^2 \times\sum_{i=x,y,z}\int\frac{d\omega_1}{2\pi}\langle(n^i)^2_{\omega-\omega_1}\rangle\overline{(\sigma^i)^2_{\omega_1}}, \quad (13)$$

where $\langle(n^i)^2_\omega\rangle\simeq\gamma/[\omega-\omega_L)^2+\gamma^2]$ is the power spectrum of $\mathbf{n}(t)$ fluctuations and $\overline{(\sigma^i)^2_\omega}\simeq\gamma_\sigma/[(\omega)^2+\gamma_\sigma^2]$ is the power spectrum of $\sigma^i(t)$ which we approximate as a Lorenzian at zero frequency with the width given by the maximum $\gamma_m=\max(\gamma,\gamma_\sigma)$. We get for a spectral power density

$$\overline{\langle I_\omega^2\rangle\simeq I_0^2}\left[\sinh\left(\frac{JS}{2\Phi}(\Phi/\Phi_0)^{1/2}\right)\right]^2\frac{\gamma_m}{(\omega-\omega_L)^2+\gamma_m^2}. \quad (14)$$

Hereafter we omit the subscript m in γ_m for simplicity. We assume that $\gamma_\sigma\leqslant\gamma$ and $\gamma_m\simeq\gamma$. It is useful to relate this spectral density to the shot noise power spectrum $\langle I^2_{shot}(\omega)\rangle=2eI_0\Delta\omega$. We have

$$\frac{\overline{\langle I_\omega^2\rangle}}{\langle I^2_{shot}(\omega)\rangle}=\left[\sinh\left(\frac{JS}{2\Phi}(\Phi/\Phi_0)^{1/2}\right)\right]^2\frac{1/\tau_e\gamma}{(\omega-\omega_L)^2+\gamma^2}. \quad (15)$$

At the Larmor frequency we find that signal-to-noise ratio is

$$\frac{\overline{\langle I_\omega^2\rangle}}{\langle I^2_{shot}(\omega)\rangle}=\left[\sinh\left(\frac{JS}{2\Phi}(\Phi/\Phi_0)^{1/2}\right)\right]^2\frac{1}{\tau_e\gamma}\simeq 2-4, \quad (16)$$

and we see that signal is large and certainly detectable and is close to what has been observed experimentally. We used $\gamma\sim 1/\tau_s=1$ MHz for a linewidth and $1/\tau_e=10^{10}$ Hz and $\sinh[(JS/2\Phi)(\Phi/\Phi_0)^{1/2}]=0.02$ for our values of the parameters. We also point out that the above analysis could be equally applied to other configurations—say, the current in nanostructures with no STM tunneling current.

In order to get a well-defined signal at ω_L a *transverse* spin polarization must exist in the tunneling current (longitudinal components cannot interact with the time-dependent components of the single spin). There are several possibilities in which such a polarization might be created: The first obvious possibility is due to the absorption of a paramagnetic atom or cluster on the tip. The magnetic moment at the edge of the tip will feel a strong magnetic anisotropy which will tend to force it in the direction of the easy axis (not necessarily in the z direction). Such a paramagnetic tip can be a source of spin-polarized tunneling electrons in the transverse direction. One may think of such a polarization even in the absence of a paramagnetic tip. The time-independent component of the single spin (which is in the z direction) may introduce a transverse polarization in points where the spin polarization changes from parallel to antiparallel. Such a phenomenon occurs, for example, with a dominant quadrupolar exchange interactions where J goes through zero as a function of distance.[15] Further work is required to understand the mechanisms of polarizing the tunneling electrons.

As a direct outcome of this analysis we discuss the possible use of a paramagnetic tip. A tip of this sort can be prepared by evaporating a thin magnetic layer on it.[10] Working with such tips may enable a more well-defined experiment where the spin polarization of the tunneling electrons will be dependent on the type of paramagnetic material deposited on the tips.

There are many possibilities to modify the tip material, from working with a antiferromagnetic tip, to a superconducting (at low temperatures) tip (for example made by Nb) to take advantage of the Meissner effect, and to create a signal with stronger intensities.

In this paper we have shown that the temporal spin polarization of tunneling electrons can interact through the Heisenberg exchange interaction with the precessing spin. We have shown that such a mechanism can create an elevated noise level at the Larmor frequency with an intensity and linewidth which are comparable to what is detected experimentally.

The potential scientific merit of this technique is very large. Several milestones have to be achieved on different spin systems to bring this technique to maturity: Detection of the hyperfine couplings, observation of ESR-STM signal from well-defined defects or atoms on the surface, and observation of spin-spin interactions from neighboring spins. After all these results are shown it might be possible to prove that the ultimate goal, a single spin, could be indeed detected. It also would be very interesting to observe the effect of an external excitation field on the signal (excitation and saturation). Successful achievement of these milestones will result in a very powerful technique with a broad range of applications.

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