

Exchange-based noise spectroscopy of a single precessing spin with scanning tunnelling microscopy

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ABSTRACT

Electron spin resonance–scanning tunnelling microscopy is an emerging technique which is capable of detecting the precession of a single spin. We discuss a mechanism based on direct exchange coupling between the tunnelling electrons and the local precessing spin \mathbf{S} . We claim that, since the number of tunnelling electrons in a single precessing period is small (about 20), one may expect a net temporary polarization within this period which will couple via exchange interaction to the localized spin. This coupling will modulate the tunnelling barrier with the Larmor frequency of the precessing spin ω_L . This modulation, although randomly changing from cycle to cycle, will produce an elevated noise in the current at ω_L . We find that for relevant values of parameters the signal-to-noise ratio in the spectral characteristic is 2–4 and is comparable with the values of the signal-to-noise ratio reported by Manassen and co-workers and by Durkan and Welland. The magnitude of the current fluctuation is a relatively weak increasing function of the dc and the magnetic field. The linewidth produced by the back action effect of tunnelling electrons on the precessing spin is also discussed.

§ 1. INTRODUCTION

There is a growing realization that the technique of electron spin resonance (ESR)–scanning tunnelling microscopy (STM) is capable of detecting the precession of a single surface spin by modulating the tunnelling current at the Larmor frequency. This technique was successful in measuring Larmor frequency modulations in defects in semiconductor surfaces (Manassen *et al.* 1989, 1993, 2000) and in paramagnetic molecules (Durkan and Welland 2002). The increasing interest in this technique is due to the possibility of detecting and manipulating a single spin (Manoharan 2002).

In ESR–STM we have the unique ability to correlate the spectroscopic information with the spatial information, detected on the atomic level, by STM, or to

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manipulate the position of the spin centres on the atomic level (Crommie *et al.* 1993, Yazdani *et al.* 1997, Madhavan *et al.* 1998). Spatial resolution of ESR–STM is what makes this technique interesting and different from the alternative technique of optically detected magnetic resonance spectroscopy in a single molecule (Koehler *et al.* 1993, Wrachtrup *et al.* 1993a, b).

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 local density of states as a result of the precession (Balatsky and Martin 2001). Another is the interference between two resonant tunnelling components through the magnetic-field-split Zeeman levels (Muzyrsky *et al.* 2002). Both of these mechanisms rely on a spin–orbit coupling to couple a local spin \mathbf{S} to the conduction electrons and have assumed *no* spin polarization of tunnelling electrons. Recently, however, Durkan and Welland (2002) observed a strong signal in a system with a substantially smaller spin–orbit coupling than assumed in the calculations (Balatsky and Martin 2001, Muzyrsky *et al.* 2002). Motivated by these experiments we addressed the following question: what is the role of the *direct* exchange interaction between the localized spin and the tunnelling electrons? This interaction has a tremendous influence on the physics of conducting substances when magnetic impurities are present (Li *et al.* 1998, Manoharan *et al.* 2000); so the following is a natural question to ask here: does exchange interaction play a role in ESR–STM also?

We find that direct Heisenberg exchange interaction between the localized spin and the conduction electrons is capable of producing modulation of the tunnelling current. The qualitative difference compared with the previous models is that we consider temporal fluctuations in the spin polarization of the electrons that are tunnelling between the tip and the surface. Spin–orbit interaction is irrelevant for this consideration. At first sight, since the experiments performed so far (Manassen *et al.* 1989, 1993, 2000, Durkan and Welland 2002) were with a non-magnetic tip, an exchange-related mechanism for ESR–STM does not look plausible. The tunnelling current has no spin polarization; thus, how can the exchange interaction between the tunnelling electrons and the local spin centre create a current modulation at ω_L ? We argue in this paper that, although the spin polarization of the tunnelling electrons is zero in the long-time limit, it is not zero on the scale of the period of the precession, typically $1/\omega_L \approx 2$ ns. On this time scale there are very few electrons that pass near the localized spin. There exists a temporary spin polarization of the tunnelling electrons, which may interact, through exchange interaction with the localized spin centre.

It is important to point out that the ESR–STM technique is 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 the detection of magnetic resonance with noise spectroscopy (Alexandrov and Zpasskii 1981, Sleator *et al.* 1985a, b, Mitsui 2000). Natural questions to be answered are as follows. What kind of noise spectroscopy are we performing here and what are its characteristics? Is it possible to obtain a noise-related signal from an exchange interaction between the tunnelling electrons and the localized surface spin centre?

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

$$\hat{\Gamma} = \Gamma_0 \exp \left[- \left(\frac{\Phi - JS(t) \cdot \hat{\sigma}}{\Phi_0} \right)^{1/2} \right], \quad (1)$$

where we consider the spin $\mathbf{S}(t)$ in the magnetic field $\mathbf{B} // \mathbf{z}$, precessing with the Larmor frequency $\omega_L = g\mu_B B$, $\hat{\Gamma}$ is understood as a matrix in spin indexes, Φ is the tunnelling barrier height, typically a few electronvolts (we assume that $\Phi = 4 \text{ eV}$) and $\Phi_0 = \hbar^2/8md^2$ is the energy related to the distance d between tip and surface (Tersoff and Lang 1993). The exchange term in the exponent is small compared with the barrier height and we can expand the exponent in JS . Explicitly $\hat{\Gamma}$ can be written as

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

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

$$\delta I(t) \propto \mathbf{n}(t) \cdot \boldsymbol{\sigma}(t), \quad (3)$$

in a scalar product $\mathbf{n}(t) \cdot \boldsymbol{\sigma}(t) = n^z(t)\sigma^z(t) + n^x(t)\sigma^x(t) + n^y(t)\sigma^y(t)$; only a transverse part, which depends on the x and y components of the localized spin and the spin of the tunnelling electrons, will describe precession in a magnetic field ($\mathbf{B} // \mathbf{z}$ is assumed). We shall focus on the transverse terms below. To make the argument as simple as possible we shall 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 the period $T = 2\pi/\omega_L$. It is convenient to introduce a time average of the current over T , $\Delta I = (1/N) \sum_{i=1}^N \delta I(t_i)$, where the sum from $i=1$ to N is over the number of electrons that will tunnel between the tip and the surface in time T , with an average $\bar{N} = I_0 T$, which is dependent on the dc in the system I_0 :

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

This fluctuation will determine the power spectrum of the tunnelling current at the Larmor frequency. Then the dispersion of the current, which depends on the precessing components, is given by the dispersion of the quantity $\sum_{i,j=1}^N n_x(t_i) n_x(t_j) [\sigma^x(t_i) \sigma^x(t_j)]$. Since the spin wavefunctions of the tunnelling electrons are not correlated between different tunnelling events we find that

$$\left(\sum_{i=1}^N \sigma^x(t_i) n_x(t_i) \right)^2 + (x \rightarrow y) \propto \bar{N}. \quad (5)$$

Therefore the dispersion of the current at the Larmor frequency due to spin fluctuations of the tunnelling electrons is

$$\frac{\langle \Delta I^2 \rangle}{I_0^2} \propto \langle (n^x)^2 \rangle \frac{\bar{N}}{\bar{N}^2} + (x \rightarrow y) \propto \frac{1}{\bar{N}}, \quad (6)$$

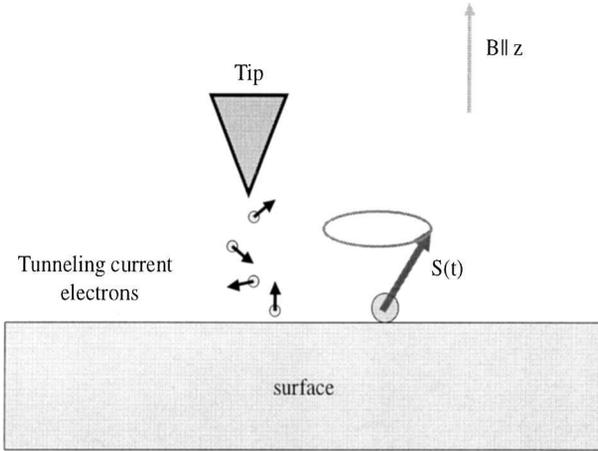


Figure 1. Schematics of the ESR–STM experiment. Tunnelling electrons carry spin that on average is zero. However, the fluctuations in the spin polarization of the tunnelling electrons on the time scale of the precession T will be non-zero and will scale as $1/\bar{N}$. $\bar{N} = 1/eI_0T$ is the average number of electrons tunnelling between the tip and the surface during one precession cycle. Once the tip is positioned close to the localized spin, the exchange interaction between the localized spin and the tunnelling electrons will modulate the tunnelling current.

where the result is normalized to the dc magnitude. We find that the magnitude of the fluctuations $(\langle \Delta I^2 \rangle)^{1/2}$ is on the scale of a few per cent of the dc for experimentally relevant values of parameters (see equation (11)).

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

$$(\langle \Delta I^2 \rangle)^{1/2} \propto (I_0 B)^{1/2}. \quad (7)$$

This is in qualitative agreement with the experimental observations, although more precise characterization is needed.

We shall now give a derivation of the results. Consider the set-up that is used in ESR–STM (figure 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 $\mathbf{S} = n\mathbf{S}$, is typically of the order of a fraction of an electronvolt. Hence the effective barrier, seen by the tunnelling electron, will depend on the spin of the conduction electron.

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

§2. SPIN-DEPENDENT TUNNELLING

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

The Hamiltonian that we consider describes a spin-dependent tunnelling matrix element between the tip (left electrode, indicated by the subscript L) and the surface (right electrode, indicated by the subscript R):

$$H = \sum_{\mathbf{k}, \alpha} \epsilon(\mathbf{k}) c_{L\alpha}^\dagger(\mathbf{k}) c_{L\alpha}(\mathbf{k}) + (\text{L} \rightarrow \text{R}) + \sum_{\mathbf{k}, \mathbf{k}'} c_{L\alpha}^\dagger(\mathbf{k}) \Gamma_{\alpha\beta} c_{R\beta}(\mathbf{k}'). \quad (8)$$

We assume that the magnetic field is along the \mathbf{z} axis: $\mathbf{B} // \mathbf{z}$. The tunnelling current operator will contain the spin-independent part that we omit hereafter and the spin-dependent part:

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

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 at a given bias V : $I_0 = \gamma_0 V$. The current-current correlator, normalized to the dc, is then

$$\frac{\langle \delta \hat{I}(t) \delta \hat{I}(t') \rangle}{I_0^2} = \left\{ \sinh \left[\frac{JS}{2\Phi} \left(\frac{\Phi}{\Phi_0} \right)^{1/2} \right] \right\}^2 \sum_{i,j=x,y,z} \langle n^i(t) n^j(t') \rangle \overline{\sigma^i(t) \sigma^j(t')}. \quad (10)$$

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

To estimate the magnitude of the current fluctuations due to the coupling to the localized spin we shall take $J \approx 0.1$ eV. This is typical for an exchange interaction in semiconductors and metals (for example Bhattacharjee (1992)). The barrier height $\Phi \approx 4$ eV and the spin $S = \frac{1}{2}$. To estimate $\Phi_0 = \hbar^2/8md^2$, we assume a typical tunnelling distance $d = 4 \text{ \AA}$. This yields $\Phi_0 \approx 0.1$ eV. For these parameters we find that

$$\frac{(\langle \Delta I^2 \rangle)^{1/2}}{I_0} \approx \frac{2}{N^{1/2}} \sinh \left[\frac{JS}{2\Phi} \left(\frac{\Phi}{\Phi_0} \right)^{1/2} \right] \approx 0.01, \quad (11)$$

with $\Gamma_1 = 0.02\gamma_0$. The magnitude of the fluctuation is in the 10 pA range for a tunnelling current $I_0 = 1$ nA and is within the observed range (Manassen *et al.* 1989, 1993, 2000, Manassen 1997, Durkan and Welland 2002). This is the magnitude of the fluctuating current in the time domain. Experimental values are observed in the frequency domain. We shall address the spectrum of the current below.

§3. BACK ACTION EFFECT OF THE TUNNELLING CURRENT ON THE SPIN

One can use the tunnelling Hamiltonian equation (8) to estimate the decay rate of the localized spin state due to interaction Γ_1 . To second order this calculation is equivalent to the Fermi golden rule calculation and we have $1/\tau_s = \pi \Gamma_1^2 N_L N_R$ eV. Similarly, the tunnelling dc I_0 is given by the tunnelling rate of conduction electrons: $1/\tau_e = \pi \gamma_0^2 N_L N_R$ eV, where N_L and N_R are the densities of states at the Fermi level

of the tip and surface respectively (Korotkov *et al.* 1994). One finds by combining these two equations that

$$\frac{1}{\tau_s} = \frac{1}{\tau_e} \frac{\Gamma_1^2}{\gamma_0^2} \approx 4 \times 10^{-4} \frac{1}{\tau_e}. \quad (12)$$

This result has a simple interpretation. The electron tunnelling rate $1/\tau_e \approx 10^{10}$ Hz gives the attempt rate for the tunnelling electrons. The probability to flip the localized spins is proportional to Γ_1^2 , which gives equation (12) for the linewidth. We estimate that $1/\tau_s \approx 4 \times 10^6$ Hz. This estimate is within an order of magnitude of the reported linewidth (Manassen *et al.* 1989, 1993, Manassen 1997, Durkan and Welland 2002). Given the uncertainty in the parameters used, we believe this is a reasonable result; for example, if we take $J = 0.05$ eV, we shall find that $(\langle \Delta I^2 \rangle)^{1/2}/I_0 \approx 10^{-2}$ and the linewidth will change by a factor of 4; $1/\tau_s \approx 10^6$ Hz. Obviously, future experiments will help to clarify the linewidth dependence on J , B and other parameters.

§ 4. SPECTRAL DENSITY OF THE CURRENT

The Fourier transform of the current–current correlator will give a power spectrum of the current fluctuation (equation (10)):

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

where $\langle (n^i)_{\omega}^2 \rangle \approx \gamma/[(\omega - \omega_L)^2 + \gamma^2]$ is the power spectrum of $\mathbf{n}(t)$ fluctuations and $\langle \sigma^i \rangle_{\omega}^2$ is the power spectrum of $\sigma^i(t)$. We have assumed that the tunnelling electron spins were uncorrelated between different tunnelling events. We can rewrite the current as a sum over discrete tunnelling events:

$$I(t) = I_0 \sum_{i=1}^N \sinh [(JS/2\Phi)(\Phi/\Phi_0)^{1/2}] n^x(t_i) \sigma^x \delta(t - t_i) + (x \rightarrow y),$$

which has a Fourier transform

$$I_\omega = I_0 \sum_{i=1}^N \sinh [(JS/2\Phi)(\Phi/\Phi_0)^{1/2}] n^x(t_i) \sigma^x \exp(i\omega t_i) + (x \rightarrow y):$$

$$\overline{\langle I_\omega^2 \rangle} \approx I_0^2 \left\{ \sinh \left[\frac{JS}{2\Phi} \left(\frac{\Phi}{\Phi_0} \right)^{1/2} \right] \right\}^2 \sum_{i,j=1}^N \langle n^x(t_i) n^x(t_j) \rangle \exp[i\omega(t_i - t_j)]. \quad (14)$$

Using the fact that N is large and averaging over the distribution of t_i , we obtain for a spectral power density in the interval $\Delta\omega \leq \gamma \ll \omega_L$ †

$$\overline{\langle I_\omega^2 \rangle} \approx I_0^2 \left\{ \sinh \left[\frac{JS}{2\Phi} \left(\frac{\Phi}{\Phi_0} \right)^{1/2} \right] \right\}^2 \frac{\Delta\omega}{\pi} \frac{\gamma}{(\omega - \omega_L)^2 + \gamma^2}. \quad (15)$$

† More precisely, in order to observe a peak at the Larmor frequency it is required that the power spectrum of the tunnelling electron spin is not white noise. Since the localized moment can create a spin polarization in the tunnelling electrons because of the spin dependent tunnelling barrier, it is plausible that the power spectrum is not white noise and has a peak at zero frequency with long enough relaxation time.

The peak of this function at the Larmor frequency is a consequence of the narrow window $\Delta\omega$. In the opposite limit of very wide bandwidth a signal will be unobservable. It is useful to relate this spectral density to the shot noise power spectrum $\langle I_{\text{shot}}^2(\omega) \rangle = 2eI_0 \Delta\omega$. We have

$$\frac{\overline{\langle I_{\omega}^2 \rangle}}{\langle I_{\text{shot}}^2(\omega) \rangle} = \left\{ \sinh \left[\frac{JS}{2\Phi} \left(\frac{\Phi}{\Phi_0} \right)^{1/2} \right] \right\}^2 \frac{1/\tau_e \gamma}{(\omega - \omega_L)^2 + \gamma^2}. \quad (16)$$

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

$$\frac{\overline{\langle I_{\omega}^2 \rangle}}{\langle I_{\text{shot}}^2(\omega) \rangle} = \left\{ \sinh \left[\frac{JS}{2\Phi} \left(\frac{\Phi}{\Phi_0} \right)^{1/2} \right] \right\}^2 \frac{1}{\tau_e \gamma} \approx 2-4. \quad (17)$$

We see that the signal is large and certainly detectable and is close to what has been observed experimentally. We used $\gamma \approx 1/\tau_s = 1$ MHz for the linewidth, $1/\tau_e = 10^{10}$ 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, a current in nanostructures with no STM tunnelling current.

As a direct outcome of this analysis we discuss the possible use of a paramagnetic tip. A tip of this sort either can be created accidentally by absorption of paramagnetic atoms on the tip from the surface or can be prepared by evaporating a thin metallic layer on it (Bode *et al.* 1998). In this case the signal is expected to be stronger. The spin polarization is larger by a factor of $\bar{N}^{1/2}$ if the spin polarization in the tip persists long enough owing to paramagnetism. The intensity of the signal is expected to increase by a similar factor. On the other hand, the linewidth is also expected to increase accordingly. It is obvious nevertheless that, if experiments with a paramagnetic tip give a stronger but broader signal, it will be a strong indication that the scenario described here is correct. There are many possibilities to modify the tip material, from working with an antiferromagnetic tip, to a superconducting (at low temperatures) tip (e.g. a Nb tip) to take advantage of the Meissner effect, and to create a signal with stronger intensities.

§5. CONCLUSION

In this paper we have shown that the temporal spin polarization of the tunnelling 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 with those 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 the ESR–STM signal from well-defined defects or atoms on the surface; observation of spin–spin interactions from neighbouring spins. After all these results have been obtained, it might be possible to prove that the ultimate goal, a single spin, could indeed be 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 and other milestones will result in a very powerful technique with a broad range of applications.

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