Theory of single spin detection with STM

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We propose a mechanism for detection of a single spin center on a non-magnetic substrate. In the detection scheme, the STM tunnel current is correlated with the spin orientation. In the presence of magnetic field, the spin precesses and the tunnel current is modulated at the Larmor frequency. The mechanism relies on the effective spin-orbit interaction between the injected unpolarized STM current and the local spin center, which leads to the nodal structure of the spatial signal profile. Based on the proposed mechanism, the strongest spin-related signal can be expected for the systems with large spin-orbit coupling and low carrier concentration.

There is no fundamental principle that precludes the single spin measurement. Possibility of a single spin observation is therefore a question of spatial and temporal resolution. The standard electron spin measurement technique – electron spin resonance – is limited to a macroscopic number of electron spins – $10^{10}$ or more [1]: the state-of-the-art magnetic resonance force microscopy has recently achieved the resolution of about 100 fully polarized electron spins [2]. It has already been shown that optically induced ESR of a single spin is possible [3]. There are proposals for the spin detection using single electron transistor and spin-polarized current [4,5]. In this letter we propose the theoretical basis for the new spin-detection technique – electron spin precession scanning tunneling microscopy (ESP-STM) – capable of single spin detection.

The applications of single spin detection and manipulation range from the study of strongly correlated systems, to nanotechnology, to quantum information processing. In the strongly correlated systems, the ability to detect single spin will allow one to investigate the magnetism on the nano scale by detecting the changes in the spin behavior while entering magnetically ordered states [6]. One could also explore temperature evolution of the magnetic properties of a single paramagnetic atom in the Kondo regime [7]. Magnetic properties of spin centers in superconductors is another area where single spin plays an important role by generating intragap impurity states [8,9]. In nanotechnology, spins can be used as elementary information storage units. In the realm of quantum computing, several architecture proposals rely on the ability to manipulate and detect single spins [10,11]. Here we argue that STM offers a powerful technique to detect a single spin.

The electron spin can be detected through its coupling to the magnetic field, which can either freeze the spin along the field, or make it precess around the field direction. To detect the single-spin signal, the super spatial and temporal resolutions are required. The necessary level of sensitivity can be achieved in the Scanning Tunneling Microscope (STM). It is well known that spatial resolution of the STM is in the sub-Angström range. Therefore, the technique naturally lends itself for the detection of the single spin. In the STM setup, the spin precession in an external magnetic field can be detected through an ac modulation of the tunnel current. In this purely dc configuration, the ac tunnel current at the precession, or Larmor, frequency can be generated due to the effective coupling of the precessing localized spin to the tunneling electrons. Hence we argue that STM can operate as Larmor frequency generator. The experimental setup we consider is given in Fig. 1.

Our work is motivated by experiments of Manassen et al. [12]. In these experiments, STM was used to measure the tunneling current while scanning the surface of Si in the vicinity of a local spin impurity (Fe cluster) or imperfection (oxygen vacancy in Si-O). A small signal in the current power spectrum at the Larmor frequency was detected. The ac signal was spatially localized at the distances on the order of 5 – 10 Å from the spin site. The extreme localization of the signal and the linear scaling of its frequency with the magnetic field prompted Manassen et al. to attribute the detected ac signal to the Larmor precession of a single spin site.

Rather than going into further details of these experiments, we pose here a general question: Under what conditions it is possible to detect the single precessing spin with STM? Surprisingly, we find that a number of mechanisms can generate a detectable ac current. The interaction that provides the coupling of the precessing spin to the tunneling current is the spin-orbit (SO) interaction. Based on our analysis, we argue that single spin detection is within the reach of current STM technology and can be realized in a variety of non-magnetic materials with isolated paramagnetic centers.

Consider a localized magnetic site with spin $S$, $S_z = 1/2$. In the presence of magnetic field, $B$, the spin-up ($E_+^z$) and spin-down ($E_-^z$) energy levels are Zeeman split. At zero temperature, only the lowest energy state (parallel to magnetic field) is occupied; at a finite temperature, or due to an external excitation, the spin may be driven into the mixed state characterized by the wavefunction, $|\psi\rangle = \alpha |\uparrow\rangle + \beta |\downarrow\rangle$, with the time evolution $\alpha = |\alpha| \exp(-iE_+t)\phi(t)$ and $\beta = |\beta| \exp(-iE_-t + i\phi(t))$. The drifting phase $\phi(t)$ determines the spin coherence time $\tau_\phi$ and is related to the ESR spin relaxation time $T_2$. In such a state, the spin expectation value, $\langle S_z |\psi(t)\rangle / \langle \psi(t)|\psi(t)\rangle$, will precesses around the di-
rection of the magnetic field at the Larmor frequency,
\[ h\omega_L = E_\uparrow - E_\downarrow = g\mu_BB, \]
where \( g \) is the gyromagnetic ratio and \( \mu_B \) is the Bohr magneton. In magnetic field of 100 Gauss, this frequency for a free electrons is 280 MHz. We assume hereafter that in the course of the ESP-STM experiment the spin is not in the groundstate either due to a thermal fluctuation or an external excitation by the tunneling current. We also assume that \( \tau_\varphi \) is sufficiently long for the spin precession to be well defined, \( \omega_L\tau_\varphi \gg 1 \). The validity of these assumptions is verified at the end of the paper.

![Experimental setup for the electron spin precession STM.](image)

FIG. 1. Experimental setup for the electron spin precession STM. In the applied magnetic field \( B \), spin of the magnetic atom (e.g. Gd, shown in gold) is precessing around the field direction. When precisely positioned next to the spin site (within a few Å), the STM tip can pick up an ac modulation of the tunnel current.

We present now a mechanism for the ac STM current generation with no ac input based on the time-dependent modification of the tunneling density of states induced near the precessing spin in the presence of applied current. On the time scale of all conduction-electron processes, the spin precession is very slow. This follows from the comparison of the energy associated with the rf spin precession, \( h\omega_L \sim 10^{-6} \) eV, and the typical electronic energy scale in metals or semiconductors, which is on the order of 1 eV. Hence, for every “instantaneous” spin orientation the electronic problem can be solved as if the local moment is static. This is similar to the Born-Oppenheimer approximation for atoms where atomic nuclear dynamics is much slower than the electronic motion. We describe the system of electrons interacting with the local impurity spin by the Hamiltonian
\[ H = H_0 + JS \cdot \sigma(0), \]
where \( J \) is the strength of the exchange interaction between the local spin and conduction electron spin density, \( \sigma(0) = \sigma_{\alpha\beta}c_\alpha^\dagger(0)c_\beta(0) \), on the impurity site. To provide the coupling of the spin orientation to the orbital degrees of freedom of electrons, the non-interacting part of the Hamiltonian, \( H_0 \), should include spin-orbit interaction.

We will focus here on the two-dimensional band of states on the material surface. From the symmetry considerations, the energy of the surface states contains spin-orbit part that is linear both in the electron spin, \( \sigma \), and momentum, \( \mathbf{k} \),
\[ \epsilon_{\alpha\beta}(\mathbf{k}) = \frac{k^2}{2m^*} + g_{SO}|\mathbf{k} \times \sigma_{\alpha\beta}|\hat{n}. \]
Here, \( m^* \) is the band mass of the surface states and \( g_{SO} \) is the SO coupling strength. The cross-product of the spin and momentum is projected onto the normal to the surface, \( \hat{n} \).

The problem specified by equations (2) and (3) can be solved for each instantaneous value of the precessing spin \( S(t) \). Specifically, we are looking for the correction to the conduction electron density of states due to the local spin. The Green function matrix for the conduction electrons is
\[ \tilde{G}_0(\mathbf{k},\omega) = \left[\omega - \epsilon(\mathbf{k})\right]^{-1}. \]

In the presence of the current flow \( \mathbf{j} \) in the system, the equilibrium momentum distribution is shifted by an amount proportional to the current, \( \mathbf{k}_0 = \mathbf{j} m^*/ne \), where \( n \) is the carrier density and \( e \) is electron charge. This shift can be approximately introduced by modifying the electronic Green functions [13],
\[ \tilde{G}(\mathbf{k},\omega) = \left[\omega - \left(\mathbf{k} - \mathbf{k}_0\right)^2/2m^* - g_{SO}|\mathbf{k} \times \tilde{\sigma}|\right]^{-1}. \]

Here, only the kinetic energy momentum is shifted since SO term is explicitly non-invariant under shift \( (\mathbf{k} \rightarrow \mathbf{k} - \mathbf{k}_0) \) as it measures the SO effects due to lattice. The correction to the density of states near the Fermi surface caused by the impurity can be then calculated straightforwardly in the first order in the scattering potential,
\[ \delta N(r) = \frac{1}{\pi} \text{Im} \sum_{\alpha\beta\gamma} G_{\alpha\beta}(r) \mathbf{J}S \cdot \sigma_{\beta\gamma} G_{\gamma\alpha}(-r). \]

Expanding in the strength of the SO interaction relative to the Fermi energy, the \( \mathbf{S} \)-dependent contribution to the density of the surface states obtains in the first order in the exchange coupling and the strength of SO interaction,
\[ \frac{\delta N}{N} = g_{SO} \int \frac{dE}{dE} J_\phi^0(\mathbf{k}_F r)|\mathbf{k}_0 \times \mathbf{S}|\hat{n}. \]

This correction depends on the distance from the spin center, \( r \), through the Bessel function of the first kind, \( J_0(x) \). This result also follows from considering Friedel spin oscillation of electrons coupled to the tunneling DOS in the presence of the SO term [12]. It is time-dependent in the presence of magnetic field since the projection of \( \mathbf{S} \) oscillates at the Larmor frequency [17]. This expression is consistent with general properties of DOS. Since \( N(r,t) \) is a scalar, it should be invariant under time reversal, whereas \( \mathbf{S} \) is odd under time reversal. Hence \( \delta N(r,t) \) can depend only on product of the spin vector with some other vector that is odd under time reversal or on time derivative of spin vector. Possible combinations include \( \delta N \sim (\mathbf{1} \times \mathbf{S})\hat{n} \) (Eq. [5]) and \( \delta N \sim \partial_t S(t) \times r \hat{n} \). Indeed, we
have found mechanism for the latter contribution as well; however, for the experimental conditions of Refs. \[2\] the time-dependent DOS mechanism described here dominates.

The current in the STM tunnel junction is proportional to the single electron density of states in the substrate. Therefore, the ac current contribution relative to the base dc STM current, \(I\), can be estimated as

\[
\frac{\delta I(t)}{I} \sim \frac{\delta N(t)}{N}. \tag{8}
\]

Notice, that the magnitude of the effect is proportional to the current in the system via \(k_0\). Experimentally, the necessary bias current can either be provided externally (by extra leads) or can be injected by STM itself. In the latter case, the direction of the current will vary in space and, in the absence of anisotropy, will flow directly away from the tip-sample junction. From the symmetry considerations, in Eq. (7), \(k_0\) should be oriented from the tip towards the spin center. The magnitude of the equilibrium shift is the 2D case considered here is \(k_0 \sim m^* I/nr\), where \(n\) is the 2D density of the surface carriers. Since \(k_0\) itself is proportional to injected current, the \(S\)-dependent correction to the current is proportional to \(I^2\). For direct tunneling into the spin center, \(k_0\) is parallel to the tunneling axis and hence there is no \(S\)-dependent contribution to the current. This nodal structure is a consequence of the problem symmetry. The magnitude of the time-dependent density of states is independent of the magnetic field orientation. The intuitive understanding of this result can be obtained using an analogue of the spin-dependent Büttiker-Landauer formalism \[18,19\]. The electrical circuit that corresponds to the STM experiment is shown in Fig. 2. When current is injected through the tunnel junction with resistance \(R_T\), it can either flow to the leads directly (left branch with conductance \(\Gamma_3\)) or through the channel affected by the spin center (right branch, \(\Gamma_1\) and \(\Gamma_2\)). We assume here that there are no spin flips. From geometric considerations, the closer tip is laterally to the spin center, the larger is the ratio, \(\Gamma_1/\Gamma_3\). From the form of SO interaction, Eq. (8), the relevant spin \(\sigma\) quantization axis for electrons flowing from the tunneling point to the impurity is perpendicular both to the surface normal, \(\hat{n}\), and the vector connecting these two points, \(k_0\). The essence of the right branch of the circuit is that in the presence of SO interaction the current becomes spin polarized, \(\Gamma_1^S \neq \Gamma_1^\perp\), and the transmission of the local magnetic center is spin sensitive, \(\Gamma_2^S \neq \Gamma_2^\perp\).

Approximately,

\[
\Gamma_1^S = \Gamma (1 + \frac{g_{SO} k_0 \sigma}{E_F}), \tag{9}
\]

\[
\Gamma_2^S = \Gamma (1 + \frac{J \sigma}{E_F}), \tag{10}
\]

where \(E_F\) is the Fermi energy of the substrate, and \(S\) is the projection of spin \(S\) onto the \(\sigma\) quantization axis (above). Similar to the Green function treatment presented above, the expression for \(\Gamma_2^S\) goes beyond linear response since it depends on the current through \(k_0\). The correction to the current due to the spin-selectivity of the circuit is

\[
\frac{\delta I}{I} = \frac{R_M^2 \Gamma}{R_M + R_T} \frac{g_{SO} k_0 J}{E_F} S, \tag{11}
\]

where \(R_M = 1/(\Gamma + \Gamma_3)\). This expression is the same as the previously obtained result, Eqs. (8) and (10).

![FIG. 2. Electrical circuit representing ESP-STM experiment.](image)

From Eq. (11) it is clear that the largest ESP-STM effect can be obtained in the systems with large spin orbit interaction and strong particle-hole asymmetry in the density of states. Small density of surface carriers, \(n\), helps to drive the carrier system out of equilibrium, which is crucial for the ESP-STM effect. All these parameters can be tuned by the choice of the substrate material, as well as the type of the spin center, to achieve large response. For model parameters, \(g_{SO} k_F \sim 0.01\) eV, \(J \sim 1\) eV, \(dN/dE \sim 1\) eV\(^{-2}\), and Fermi surface displacement \(k_0/k_F \sim 0.1\), the effect in the density of states is \(\delta N/N \sim 10^{-3}\), which should be experimentally detectable.

In the above discussion we assumed that the spin dephasing time, \(\tau_\phi\), is long compared to the Larmor period, which implies weak coupling to environment. However, even under ideal circumstances, the measurement will induce spin dephasing. This backaction is an unavoidable side effect of measurement. We will now estimate the backaction of the tunneling electrons on the precessing spin. By applying a canonical transformation, the original Hamiltonian, Eqs. (8) and (9), can be mapped to a model of two reservoirs (\(L\) and \(R\)) of spinless electrons.
coupled through a spin-dependent tunneling matrix element [21].

$$H_{\text{eff}} = H_L + H_R + h_z S_z + \sum_{l \in L, r \in R} c_l^\dagger [T_0 + T_1 S_x] c_r.$$  

Here $T_0$ is the tunneling amplitude that doesn’t involve local spin and $T_1 \simeq g_{so}k_0 J(dN/dE) T_0$ is the spin-dependent part of the tunneling. The $x$ axis is chosen along $\hat{n} \times \mathbf{k}_0$ direction. For spin-1/2 quantum systems, this model has been extensively studied [21][22]. Two important parameters in the model are the electron tunneling rate, $1/\tau_e = \pi T_0^2 N_L N_R V$, and the measurement-induced spin-dephasing rate, $1/\tau_s = \pi T_1^2 N_L N_R V$. It has been demonstrated [22] that under the weak measurement conditions, $h/\tau_e < h_z$, the stationary power spectrum of the tunnel current has a peak at the Lambor frequency, 

$$\langle I_x^2 \rangle / 2eI_0 \simeq \frac{1}{(\omega - \omega_L)^2 + 1/\tau_s^2}$$

For weak measurement, increasing applied voltage does not change the peak value relative to the shot noise level; however it increases the weight under the resonance peak. Under the stationary conditions, the coupling to the current fluctuations also provides the excitation for the spin. For the same parameters as above we find spin dephasing time $1/\tau_s \sim 10^{7} \ldots 10^8 \text{ Hz} \ll \omega_L$. Experimentally, the observed linewidth is narrow, $\sim 500 \text{ KHz} \ll \omega_L$ [12], which is consistent with our effective model. Any extrinsic (unrelated to measurement) dephasing mechanisms will decrease the signal to shot noise ratio.

The applicability of the presented ESP-STM mechanism is not limited to semiconductors. Other systems with stronger spin-orbit interaction, such as Au or Cu surfaces [20] with local magnetic defects may be expected to show a similar effect. Yet another route is to use heavy magnetic atoms, such as Eu or Gd as the local spin impurities with large intrinsic SO interactions. In this case, a result similar to Eq. [10] obtains, with the strength of the effect proportional to the spin-orbit coupling on the impurity site. In general, any form of SO interaction is capable of coupling the precessing local spin to the itinerant electron density of states, which can be measured with STM.

The spin-current coupling mechanism that we described applies to the static (dc) case as well. The dc case can be realized by applying a strong polarizing magnetic field and detecting the variation of the tunneling current as a function of the magnetic field orientation. This, however, is only possible at low temperatures, due to the limited strength of available magnetic fields. The dc measurement is further complicated by the low-frequency (1/f) noise.

In conclusion, we have proposed a mechanism for the novel technique for the single spin detection with STM. It is based on the ac modulation of the tunneling current caused by the effective spin-orbit coupling to the precessing local spin. From the analysis of the coupling mechanism we identified classes of materials and experimental conditions for which significant single spin signal can be expected.

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