Theory of Single-Spin Detection with a Scanning Tunneling Microscope

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No fundamental principle precludes the measurement of a single spin, and therefore the capability to make such a measurement simply depends on our ability to develop a detection method of sufficient spatial and temporal resolution. The standard electron spin detection technique—electron spin resonance—is limited to a macroscopic number of electron spins (10^{10} or more) (Farle 1998). A state-of-the-art magnetic resonance force microscope has recently detected about a hundred fully polarized electron spins (Bruland et al. 1998). We argue that scanning tunneling microscopy offers a powerful technique to detect a single spin and propose the theoretical basis for the new spin-detection technique, which we call spin precession by scanning tunneling microscopy.

The capability to routinely detect and manipulate a single spin would be remarkably useful, with applications ranging from the study of strongly correlated systems to nanotechnology and quantum information processing. For example, we could investigate magnetism on the nanoscale in a strongly correlated system by detecting changes in the spin behavior as the system enters the magnetically ordered state (Heinze et al. 2000). We could also fully explore the magnetic properties of a single paramagnetic atom in the Kondo regime (Manoharan et al. 2000). Magnetic properties of spin centers in superconductors are another area where a single spin plays an important role, since it can generate intragap impurity states (Salkola et al. 1997, Yazdani et al. 1997). With regard to nanotechnology, the ability to manipulate a single spin could open the door to single-spin-based information storage devices, whereas in the realm of quantum computing, it could help bring to fruition several specific computing architectures (Kane 1998, Loss and DiVincenzo 1998).

Our theoretical investigation of spin precession–scanning tunneling microscopy has in part been motivated by the experiments of Yshay Manassen et al. (1989), in which a defect structure (an oxygen vacancy) in oxidized silicon was interrogated with a scanning tunneling microscope (STM). The STM operated in the presence of an external magnetic field, and a small alternating current (ac) signal in the power spectrum of the tunneling current was detected at the spin's precession, or Larmor, frequency. The ac signal was spatially localized at distances of about 5–10 angstroms from the spin site. The extreme localization of the signal and the linear scaling of its frequency with the magnetic field prompted Manassen to attribute the detected ac signal to the Larmor precession of a single-spin site. Whereas that interpretation was somewhat controversial, the later work by Manassen et al. (2000) and more recent work by Colm Durkan and Mark Welland (2002) support the notion that STM can indeed sense a single spin.

From a theoretical perspective, it was not clear how the spin could generate an ac component in the STM's tunneling current. As outlined below, however, the precessing spin causes an ac modulation of the surface density of states near the spin site, provided a dc current flows through the surface. In fact, that current can be the tunneling current that flows between the STM tip and the surface. Thus, the tunneling current, which is proportional to the surface density of states, plays two roles in spin detection by scanning tunneling microscopy: It provides a means to couple the precessing spin to

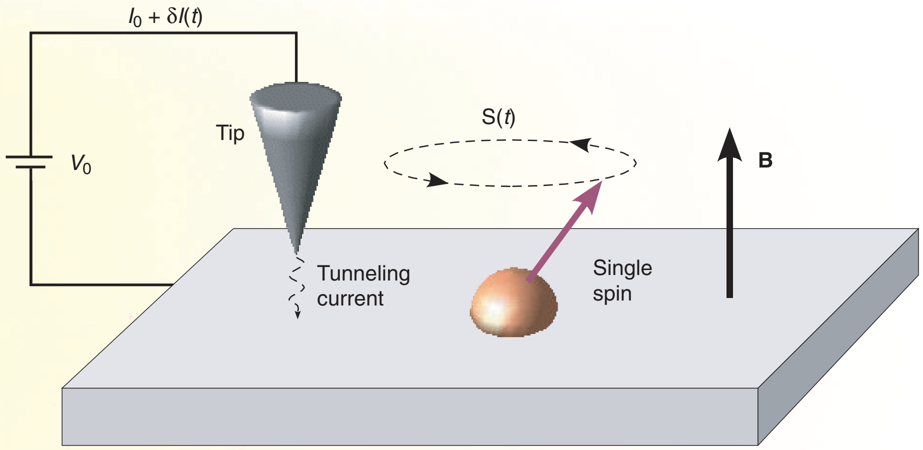

Figure 1. Experimental Setup for Electron Spin Precession by Scanning Tunneling Microscopy
In the applied magnetic field B, the spin of the magnetic atom (for example, gadolinium, shown in gold) is precessing around the field line. The STM tip is precisely positioned within a few angstroms of the spin site. The dc tunneling current I_0, between the STM tip and the sample, can acquire an ac component, $\delta I(t)$, that signals the presence of the precessing spin.

the density of states and a means to detect the ac modulation of those states. The experimental setup that we consider is shown in Figure 1. A general discussion of the principles underlying scanning tunneling microscopy can be found on page 303.

Before analyzing the effect of the STM, consider a localized magnetic site with spin **S** (spin 1/2), on the surface of a substrate. In the presence of a magnetic field, **B**, the energy levels of the spin-up and spin-down states (denoted by $E_\uparrow$ and $E_\downarrow$, respectively) are Zeeman-split. At a finite temperature, or as a result of an external excitation, the spin may be driven into the mixed state characterized by the wave function

$$|\psi(t)\rangle = \alpha(t)\,|\uparrow\rangle + \beta(t)\,|\downarrow\rangle \ , \tag{1}$$

where

$$\alpha(t) = |\alpha| \exp(-iE_\uparrow t) \ , \text{ and}$$
$$\beta(t) = |\beta| \exp(-iE_\downarrow t + i\phi(t)) \ .$$

The phase $\phi(t)$ determines the spin coherence time τ_ϕ and is related to the spin relaxation time T_2 measured by electron spin resonance.

In the state given by Equation (1), the spin, with an expectation value of

$$\frac{\langle \psi(t)|\mathbf{S}|\psi(t)\rangle}{\langle \psi(t)|\psi(t)\rangle} \ , \tag{2}$$

will precess around a magnetic field line at the Larmor frequency ω_L,

$$\hbar\omega_L = E_\uparrow - E_\downarrow = \gamma B \tag{3}$$

where γ is the gyromagnetic ratio. (See the box "Spin Manipulation with Magnetic Resonance" on page 288.) In a magnetic field of 100 gauss, this frequency is 280 megahertz for a free electron.

If we consider what happens on the surface, then the precession of the local moment will be coupled to the orbital motion of electrons via the spin-orbit interaction. The details of the spin-orbit coupling depend on the specific material. In general, however, the interaction of the conduction electrons with the local impurity spin can be described by the Hamiltonian

$$H = H_0 + J\,\mathbf{S}\bullet\sigma(0)\ \ , \tag{4}$$

where J is the strength of the exchange interaction between the local spin **S**, and the spin density of the conduction electrons, $\sigma(0) = \sigma_{\alpha\beta}\, c^{\dagger}_{\alpha}(0)\, c_{\beta}(0)$, on the impurity site. Here, $c^{\dagger}_{\alpha}(0)$, $c_{\beta}(0)$ are the electron creation/destruction operators with spin α and β, respectively, and $\sigma_{\alpha\beta} = (\sigma^{x}_{\alpha\beta}, \sigma^{y}_{\alpha\beta}, \sigma^{z}_{\alpha\beta})$ is a vector of Pauli matrices. The unperturbed Hamiltonian H_0 describes the surface without the spin impurity. Based on symmetry, the energy of the unperturbed surface states contains a spin-orbit part that is linear both in the conduction-electrons' spin, σ, and their momentum, **k** (Bychkov and Rashba 1984).

$$\varepsilon(\mathrm{k}) = \frac{k^2}{2m^*} + \gamma_{\mathrm{SO}}\left[\mathbf{n} \times \mathbf{k}\right] \cdot \hat{\sigma}\ \ , \tag{5}$$

where m^* is the band mass of electrons in the substrate, **n** is a unit vector normal to the surface, and γ_{SO} is a parameter that characterizes the strength of the surface spin-orbit coupling. The problem specified by Equations (4) and (5) can be solved for each instantaneous value of the precessing spin $\mathbf{S}(t)$. The solution, however, does not lead to a time-dependent conduction-electron density of states $N(\mathbf{r}, t)$ because the effects of the precessing spin average to zero. In that case, the tunneling current would remain constant.

To extend the model, we account for the fact that the tunneling current injects electrons into the sample, and those electrons can flow to the spin site. In the presence of a current density $\boldsymbol{j}$ flowing through the surface, the equilibrium momentum distribution **k** is shifted by an amount, $\mathbf{k}_0 = \boldsymbol{j}m^*/ne$, where n is the carrier density and e is the electron charge. This shift can be introduced into a Green's function matrix for the conduction electrons, $\hat{G}_0(\mathbf{k},\omega)$,

$$\hat{G}_0(\mathbf{k},\omega) = \left[\omega - \frac{(\mathbf{k} - \mathbf{k}_0)^2}{2m^*} - \gamma_{\mathrm{SO}}\left[\mathbf{n} \times \mathbf{k}\right] \cdot \hat{\sigma}\right]^{-1} . \tag{6}$$

We expand the matrix in γ_{SO} relative to the Fermi energy. Then, to first order in both the exchange coupling J and γ_{SO}, we obtain an **S**-dependent contribution to the density of the surface states:

$$\frac{\delta N}{N} = \gamma_{\mathrm{SO}}\, J\, \frac{dN}{dE}\, J_0^2(k_F r)\left[\mathbf{k}_0 \times \mathbf{S}\right]_n\ . \tag{7}$$

This correction depends on the distance from the spin center, r, through the Bessel function of the first kind, $J_0(x)$. The correction is time dependent in the presence of a magnetic field because the projection of **S** oscillates at the Larmor frequency. The magnitude of the correction is proportional to the current density in the system (through $\mathbf{k}_0$).

The total (ac plus dc) tunneling current I, between the STM tip and the sample is proportional to the single-electron density of states in the substrate. Therefore, the ac component $\delta I(t)$, normalized to the tunneling current, can be estimated as

$$\frac{\delta I(t)}{I} = \frac{\delta N(t)}{N} \quad . \tag{8}$$

We have focused on the case in which an STM injects current into the system, but in principle, the current can also be provided externally (through extra leads attached to the substrate), and the ac current can be detected with some ultrasensitive current measurement device.

It is also important to note that the electron density of states $N(\mathbf{r}, t)$ is a scalar and should be invariant under time reversal, whereas $\mathbf{S}$ is odd under time reversal. Hence, $\delta N(\mathbf{r}, t)$ can depend only on the product of the spin vector with some other vector that is odd under time reversal. In Equation (7), that vector is the current density, that is, $\delta N \sim [\mathbf{k}_0 \times \mathbf{S}]_n$. Another possibility is that the correction to the density of states depends on the time derivative of the spin vector, that is, $\delta N \sim \partial_t \mathbf{S}(t)$. We have also found a mechanism for this possibility.

Our conjecture of how an STM can detect single spins is based on the ac modulation of the density of surface states that results from a current-induced spin-orbit coupling to the precessing local spin. The changing state density is observed as the ac component to the tunneling current. ■

Alexander Balatsky received his Ph.D. in 1987 from the Landau Institute for Theoretical Physics, where he then worked as a researcher until 1989. From 1989 to 1991, he was at the University of Illinois at Urbana-Champaign, where he became a visiting resident assistant professor in 1990. In 1991, he joined Los Alamos National Laboratory as a J. R. Oppenheimer Fellow and is currently a technical staff member in the Theoretical Division.

Further Reading

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