Noise Spectroscopy of a Single Spin with Spin Polarized STM

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ABSTRACT

We propose a novel way for detecting the dynamics of a single spin center on a non-magnetic substrate. In the detection scheme, the STM tunnel current is correlated with the spin orientation. As a consequence, the *noise* in a spin polarized STM tunneling current gives valuable spectroscopic information on the spin dynamics of a single magnetic atom. This is an example of "noise spectroscopy" in which by probing the noise of a small quantum system we may directly monitor its dynamics.

Keywords: noise spectroscopy, single spin, spin polarized STM

1 INTRODUCTION

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 ($\geq 10^{10}$) of electron spins [1]. Recent experiments on a spin polarized STM [2] opened new possibilities of investigation of magnetic systems at spatial resolutions of the Angstrom scale. The bulk of the experiments performed to date were on magnetically ordered states, such as antiferromagnets.

Here we propose to use a spin polarized STM tunneling current to gain spectroscopic information on a single magnetic atom on a non-magnetic surface. The set up is similar to the one used in a recent ESR-STM experiments. A scheme is depicted in Fig.(1). We will consider a magnetic atom of spin $\mathbf{S}$ placed on an otherwise non-magnetic substrate. As we will explain in the text, the noise in the current flowing from the STM tip will allow us, in certain instances, to immediately measure the single spin time dependent susceptibility. This is yet another case where the spectroscopy of the noise in the current easily allows us (where other methods often fail) to directly probe the highly disordered quantum states of a microscopic system (in this case a single spin).

The outline of this article is as follows: In section(2), we derive the general relation between the noise $\langle|\delta I(\omega)|^2\rangle$

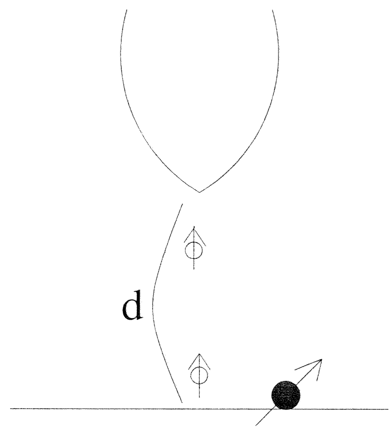

Figure 1: The experimental setup. The surface tip separation distance is d. The single magnetic atom on the surface is marked by a solid circle. The spin polarized current emanating from the STM tip is schematically shown by the open spin up polarized ovals.

in the current and the single spin susceptibility $\chi(\omega)$. Under quite general circumstances, the noise in the current is directly proportional to the single spin susceptibility, so that measuring the noise in the current immediately gives us valuable information about the single spin susceptibility. In Section(3), we will elaborate on the origin of the spin dependent tunneling matrix elements that form much of the backbone of our proposal. In Section(4), we will discuss the decoherence resulting from backaction effects. Here, we will also relate the spin scattering relaxation rate and DC charge transport and estimate how large the various quantities might be in realistic setups. We will conclude, in Section(5), by evaluating the typical signal to noise ratios for our experimental proposal.

2 HOW TO PROBE THE SPIN SUSCEPTIBILITY VIA A MEASUREMENT OF THE NOISE

There is a major difference between probing a single spin (as discussed here) and probing a macroscopic magnetic system. In the case of a single atomic spin $\mathbf{S}(t)$, we perform a measurement on a microscopic state of the that is quantum and highly fluctuating due to interactions with its environment. The experiments that we envision are done at high enough temperatures and sufficiently low magnetic fields. In such instances, the spin state is disordered. The tunneling experiment discussed here is a specific example of *noise spectroscopy*, wherein we extract spectroscopic information, such as the single spin relaxation time T^* from the measurement of the noise in the current. The main idea of noise spectroscopy in STM is that the time dependence of the tunneling current will have a characteristic relaxation time that is directly related to spin relaxation time T^*. The time varying tunneling current $I(t)$ will have a fluctuating component, apart from its average DC value I_0. Current noise is given by the Fourier transform of the time-dependent fluctuations of the electrical current leading to the power spectra $\langle|\hat{I}(\omega)|^2\rangle$ within the frequency domain. Here, we will focus on the power spectrum of the tunneling current and argue that it will be proportional to the local spin susceptibility,

$$\chi(\omega) = \langle \mathbf{S}_\omega \cdot \mathbf{S}_{-\omega} \rangle = \frac{1/T^*}{\omega^2 + 1/T^{*2}}, \tag{1}$$

of the single atom on the substrate. T^* denotes the relaxation time to the polarization axis of the single local spin (for a macroscopic collection of spins, this would become the familiar T^1 of NMR gauging the relaxation time of the average magnetization to the polarization axis). A long time exponential relaxation for $\chi(t) \equiv \langle S(t)S(0)\rangle \sim \exp[-t/T^*]$, has as its Fourier transform the Lorentzian of Eqn.(1).

Examples of noise spectroscopy include the noise NQR measurements [3], noise in Faraday rotation [4] and, recently, noise spectroscopy of a local spin dynamics in STM [5]. The central feature present in all of these examples is that the quantum system is not driven by external fields. Rather, the noise in the signal itself (e.g. thermal, shot, ...) sans any applied polarizing field allows us to extract spectroscopic information! We will consider the excess noise produced by a single spin whose time dependent quantum state we wish to probe and suggest that over a certain parameter range, the excess noise generated by the single spin may overwhelm the noise in the current in its absence.

To make matters concrete, consider the tunneling between two contacts in a presence of a localized spin $\mathbf{S}$. The Hamiltonian of this system assumes the form

$$\begin{aligned} \hat{H} = \{&\sum_{n\sigma} \epsilon_{Ln\sigma} c^\dagger_{Ln\sigma} c_{Ln\sigma} \\ &+ \sum_{nn'\alpha\beta} c^\dagger_{Ln\alpha}[t_0 + t_1 \hat{\mathbf{S}} \cdot \hat{\boldsymbol{\sigma}}_{\alpha\beta}] c_{Rn'\beta} \\ &+ \sum_{nn'\alpha\beta} c^\dagger_{Ln\alpha}[t_L \hat{\mathbf{S}} \cdot \hat{\sigma}_{\alpha\beta}] c_{Ln'\beta}\} + (L \to R). \end{aligned} \tag{2}$$

In the above, $\hat{\boldsymbol{\sigma}}_{\alpha\beta}$ is the Pauli matrix vector with matrix indices α and β, the fermionic $c_{\lambda n\sigma}, c^\dagger_{\lambda n\sigma}$ are the annihilation and creation operators of electrons in the n-th eigenstate of the lead $\lambda = L, R$ with $\sigma = \pm 1$ the (up/down) spin polarization label. The left lead (L) is the STM tip, and the right lead (R) refers to the surface. The wavefunctions of our system are superpositions of the direct product states $|\psi_L\rangle \otimes |\psi_S\rangle \otimes |\psi_R\rangle$ - the direct product of the state of the left contact, the impurity spin, and the right contact. The tunneling matrix $\hat{t}$ present in the second term of Eqn.(2) couples all of these different states. It has two contributions: the term proportional to t_0 describes the spin independent tunneling while the term proportional to t_1 (and to $t_{L,R}$ in the third term) depicts the spin dependent contributions arising from the exchange interaction for electrons tunneling to the magnetic atom. The third term couples the each lead (L or R) to the impurity spin. Only the second term in Eqn.(2) will give rise to net current flow from the left to the right contact. In section(3), we will explain the origin and elaborate on the magnitude of the spin independent and dependent terms.

Henceforth, we will assume that the tunneling electrons are partially spin polarized. There are several situations where such interactions may materialize. One is a ferromagnetically coated tip. The tip then has an excess chemical potential difference $2(\delta\mu_\sigma)$ between the two different spin polarizations: $\epsilon_{Ln\sigma} = \epsilon_{Ln} + \sigma\delta\mu_\sigma$. Yet another possible realization is that the tip is antiferromagnetic. As the tunneling occurs out of the last atom on the tip, even an antiferromagnetically ordered tip lead to spin polarized tunneling. As may be seen by perusing the spin dependent contribution to $\hat{t}$, the tunneling electrons generate a magnetic field and exert torques on the localized spin $\mathbf{S}$ on the surface [7]. In the aftermath this leads to corrections to the spin dynamics. To lowest order in t_1 and $t_{L,R}$, however, such effects are not present.

To make our expressions slightly more concise, we will employ the Heisenberg representation and absorb the time dependence in all operators $\{\hat{O}\}$, i.e. $\hat{O}(t) = \exp[iHt]\hat{O}_S \exp[-iHt]$, with $\hat{O}_S = \hat{O}(t=0)$ the operator in the Schroedinger representation which we now forsake. All expectation values $\langle\hat{O}(t)\rangle$ that appear will represent $\langle\psi(0)|\hat{O}(t)|\psi(0)\rangle$ with $\psi(t=0)$ the zero time wave function of the Schroedinger representation which does not evolve within the Heisenberg formulation.

By directly computing dN_L/dt we find that the electronic current

$$\hat{I} = -ie \sum_{nn'\alpha\beta} c^\dagger_{Ln\alpha}[t_0\delta_{\alpha\beta} + t_1\hat{\mathbf{S}}(t) \cdot \hat{\boldsymbol{\sigma}}_{\alpha\beta}] c_{Rn'\beta} + h.c., \tag{3}$$

with e the electronic charge.

We see that the tunneling current has a part that depends on the localized spin,

$$\delta\hat{I}(t) = et_1\hat{\mathbf{S}}(t) \cdot \hat{\mathbf{I}}_{spin}(t), \tag{4}$$

where

$$\hat{\mathbf{I}}_{spin}(t) = -ic^{\dagger}_{L\alpha}\hat{\boldsymbol{\sigma}}_{\alpha\beta}c_{R\beta} + h.c. \tag{5}$$

is the spin polarization dependent contribution to the electronic current. We assume that there is a non-vanishing *steady* spin polarized current component tunneling from the tip to the surface, aligned along (or defining) the z axis: $\langle \hat{I}^i_{spin}(t)\rangle = \delta_{i,z}A^{\frac{1}{2}}$ + time dependent fluctuations, with a finite $A \neq 0$. To lowest order in t, the electronic current-current correlation function originating from the spin dependent part that we wish to probe,

$$\langle\{\delta\hat{I}(t), \delta\hat{I}(t')\}\rangle = e^2 t_1^2 \langle \hat{S}^i(t)\hat{S}^j(t')\rangle\langle \hat{I}^i_{spin}(t)\hat{I}^j_{spin}(t')\rangle + (t \leftrightarrow t'), \tag{6}$$

where $\{\}$ denotes a symmetrized correlator, $i, j = x, y, z$ denote the spin components. To lowest non-trivial order in t_1, we need to treat the two temporal correlation functions $\chi(t-t') = \langle \hat{S}^z(t)\hat{S}^z(t')\rangle$ and $C(t-t') = \langle \hat{I}^i_{spin}(t)\hat{I}^j_{spin}(t')\rangle \to \langle \hat{I}^i_{spin}(t)\rangle\langle \hat{I}^j_{spin}(t')\rangle = \delta_{i,z}\delta_{j,z}A$ (for $|t-t'| \to \infty$) independently [6]. To make connection with the main proposal in this paper, we note that, when Fourier transformed, the symmetrized correlator $\langle\{\delta\hat{I}(t), \delta\hat{I}(t')\}\rangle$ in Eqn.(6) is none other than the current noise spectrum at various frequencies originating from the local spin. For small $(t_1/t_0) \ll 1$ (the experimental situation), $\mathcal{O}(\langle\{\delta\hat{I}(t), \delta\hat{I}(t')\}\rangle = (t_1/t_0)^2 I_0^2$ (with I_0 the magnitude of the electronic current).

In evaluating the spin current correlator $C(t)$, we ignore the fluctuating contributions present for short times (large frequencies). The spin current correlator $C(t)$ has a finite, asymptotic, long time value, A, that reflects the spin polarized DC current out emanating from the STM tip [6]. In Fourier space, the current power spectrum is given by a convolution of the two power spectra associated with $\mathbf{S}$ (i.e. $\chi(\omega)$) and $\boldsymbol{\sigma}$ (the spin current correlator $C(\omega)$):

$$\langle|\delta\hat{I}(\omega)|^2\rangle = e^2 t_1^2 \int \frac{d\omega_1}{2\pi}\chi(\omega_1)C(\omega-\omega_1) + (\omega \to -\omega). \tag{7}$$

At low frequencies, $C(\omega) \simeq 2\pi A\delta(\omega)$, and, consequently,

$$\langle|\delta\hat{I}(\omega)|^2\rangle = 2Ae^2 t_1^2 \chi(\omega) + ... \tag{8}$$

The ellipsis in Eqn.(8) refer to the contribution to the convolution of Eqn.(7) from the finite frequency (short time) contributions to $C(t)$.

Eqn.(8) is our central result. It vividly illustrates how the spectroscopy of the *noise* in the tunneling current $\langle|\delta I(\omega)|^2\rangle$ allows us to directly probe the spectrum of spin fluctuations encapsulated in $\chi(\omega)$. The spin polarized tunneling current provides a reference frame with respect to which we may measure the fluctuations of the localized spin $\mathbf{S}(t)$.

3 THE ORIGIN OF THE SPIN DEPENDENT TUNNELING

We now elaborate on the origin and magnitude of the spin dependent tunneling matrix elements of Eq.(2). The spin dependence of the tunneling originates from the direct exchange dependence of the tunneling barrier [5]. The overlap of the electronic wave functions of the tip and surface, separated by a distance d is exponentially small and is given by a *spin dependent* tunneling matrix element,

$$\hat{t} = \gamma \exp[-\sqrt{\frac{\Phi - J\mathbf{S}(t)\cdot\hat{\boldsymbol{\sigma}}}{\Phi_0}}], \tag{9}$$

where we explicitly include the direct exchange between tunneling electron spin $\boldsymbol{\sigma}$ and the local spin $\mathbf{S}$. Here, J is the exchange interaction between the tunneling electrons and the local precessing spin $\mathbf{S}$. In the above, $\hat{t}$ is to be understood as a matrix in the internal spin indices, and Φ is the tunneling barrier height. Typically, Φ is a few eV. As a canonical value we may assume $\Phi = 4eV$, $\Phi_0 = \frac{\hbar^2}{8md^2}$ is related to the distance d between the tip and the surface [8]. As the exchange term in the exponent is small compared to the barrier height, we may expand the exponent in JS. Explicitly, $\hat{t}$ may be written as

$$\hat{t} = t_0 + t_1\hat{\boldsymbol{\sigma}}\cdot\mathbf{S}(\mathbf{t}), \tag{10}$$

where,

$$t_0 = \gamma \exp(-(\Phi/\Phi_0)^{1/2})\cosh[\frac{JS}{2\Phi}\sqrt{\frac{\Phi}{\Phi_0}}], \tag{11}$$

describes spin independent tunneling. The spin dependent amplitude,

$$t_1 = \gamma \exp(-(\Phi/\Phi_0)^{1/2})\sinh[\frac{JS}{2\Phi}\sqrt{\frac{\Phi}{\Phi_0}}]. \tag{12}$$

For estimates we may employ the typical rule of thumb $t_1/t_0 \simeq \frac{JS}{2\Phi} \ll 1$.

4 BACKACTION EFFECT OF THE TUNNELING CURRENT ON THE SPIN

We may use the tunneling Hamiltonian of Eq.(2) to estimate the decay rate of the localized spin state resulting from the spin scattering interaction associated with t_1. To second order this calculation is equivalent to a simple application Fermi's golden rule for the up-down spin flip rate leading to $\frac{1}{\tau_s} = \pi t_1^2 N_L N_R eV$, with V is the voltage applied between the left and right electrodes. Similarly, the DC tunneling current I_0 is given by the tunneling rate of conduction electrons $\frac{1}{\tau_e} =$

$\pi t_0^2 N_L N_R eV$, where $N_{L,R}$ denotes the density of states at the Fermi level of the tip and surface respectively [9].

Comparing these simple results, we find

$$\frac{1}{\tau_s} = (\frac{t_1}{t_0})^2 \frac{1}{\tau_e} \simeq (\frac{JS}{2\Phi})^2 \frac{1}{\tau_e}. \quad (13)$$

The important outcome of this analysis is that the current induced broadening predicts a spin relaxation rate $\frac{1}{\tau_s} \propto I_0$ which may be experimentally tested. This result has a very simple interpretation: the impinging foreign electron tunneling rate for a DC current of magnitude $I_0 = 1nA$ is given by $\frac{1}{\tau_e} \sim 10^{10} Hz$. By contrast, the probability to produce a spin flip, sparked by the tunneling electrons, is proportional to t_1^2, which leads to Eq.(13) for the linewidth. The full intrinsic line width is further enhanced by the coupling of the spin to the environment (e.g. the interaction between the spin and the substrate phonons) which may indeed further increase the spin flip rate. The net observed linewidth,

$$(T^*)^{-1} \simeq \tau_s^{-1} + \tau_{env}^{-1}, \quad (14)$$

includes both backaction contributions (τ_s^{-1}) and aforementioned linewidth broadening due to coupling to the environment (τ_{env}^{-1}). The backaction relaxation time scales set a trivial upper bound on the net relaxational linewidth of the single impurity spin.

Given the typical values of the parameters in Eqn.(13), we estimate $\frac{1}{\tau_s} \simeq 4 \times 10^6 Hz$. Future experiments will help to clarify the linewidth dependence on the various parameters.

5 A SIZABLE SIGNAL TO NOISE RATIO

The anticipated signal (the excess noise induced by the impurity spin) to noise (the noise already present in the absence of the impurity spin- the shot noise) ratio can be quite significant.

To obtain estimates of orders of magnitude we may employ Eqs.(2,3, 5,8). In the final analysis, we obtain

$$\frac{\langle |\delta I(\omega \to 0)|^2 \rangle}{\langle |\delta I_{shot}^2(\omega \to 0)| \rangle} \sim (\frac{J}{2\Phi})^2 \frac{T^*}{\tau_e} \sim 1 - 100, \quad (15)$$

where the DC current $I_0 = \frac{e}{\tau_e}$. To quote typical numbers, $J \simeq 0.1 - 1$ eV, and as noted earlier, $\Phi \simeq 4$ eV. Typical relaxation times employed in the above are $T^* \sim 10^{-8} - 10^{-6}$ seconds and $\tau_e \sim 10^{-10}$ seconds (1nA). This sizable ratio offers promise to such a single spin detection experiment and related small system applications.

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REFERENCES

[1] M. Farle, Rep. Prog. Phys. **61**, 755 (1998).

[2] M. Bode, M. Getzlaff and R. Wiesendanger Phys. Rev. Lett. 81, 4256 (1998).

[3] T. Sleator, E. L. Hahn, C. Hilbert and J. Clarke, Phys. Rev. Lett. 55, 1742 (1985); T. Sleator, E. L. Hahn, C. Hilbert and J. Clarke, Phys. Rev. B 36, 1969 (1987).

[4] E. B. Aleksandrov and V. S. Zapasskii, Sov. Phys. JETP 54, 64 (1981). T. Mitsui, Phys. Rev. Lett. 84, 5292 (2000).

[5] ESR-STM as a noise spectroscopy is discussed in A.V. Balatsky, Y. Manassen and R. Salem, Phil. Mag. **B 82**, 1291, (2002); A.V. Balatsky, Y. Manassen and R. Salem, Phys. Rev **B 66**, 195416, (2002)..

[6] The value of A is determined by the microscopic details of the tip. If we assume a ferromagnetic tip then $A \propto 2N_L \delta\mu$ is determined by the chemical potential splitting for different spin components. We will treat A as a phenomenological parameter in our analysis.

[7] For related, spin-transfer torques see e.g. D. Ralph, Science, **291**, 999 (2001)

[8] J. Tersoff and N.D. Lang, "Theory of Scanning Tunneling Microscopy", in Methods of Experimental Physics, v 27, 1, Academic Press, 1993.

[9] A. N. Korotkov, D. V. Averin, and K. K. Likharev, Phys. Rev. **B 49**, 7548 (1994). See also D. Mozyrsky, L. Fedichkin, S. A. Gurvitz, G. P. Berman, cond-mat/0201325.