A Non-Demolition Single Spin Meter

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We present the theory of a single spin meter consisting of a quantum dot in a magnetic field under microwave irradiation combined with a charge counter. We show that when a current is passed through the dot, a change in the average occupation number occurs if the microwaves are resonant with the on-dot Zeeman splitting. The width of the resonant change is given by the microwave induced Rabi frequency, making the quantum dot a sensitive probe of the local magnetic field and enabling the detection of the state of a nearby spin. If the dot-spin and the nearby spin have different g-factors a non-demolition readout of the spin state can be achieved. The conditions for a reliable spin readout are found.

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Single-shot readout of individual spins lies at the heart of many future technologies, from spintronics and quantum computation to single molecule spin resonance. There have been several recent proposals and demonstrations of single spin readout, all with restricted applicability, relying on the details of the physical system in which the spin is housed [1] or requiring certain special features, such as optical activity [2], nuclear spins [3] or a large detector-system interaction [4]; others are too slow to achieve a measurement in a single attempt [5]. Here we introduce a versatile spin meter that can achieve reliable determination of the spin state. Our electrical method is non-invasive, in contrast to standard techniques that destroy the spin in the course of the measurement [6]. It consists of a quantum dot under microwave illumination and can in principle be used to detect arbitrarily weakly coupled spins in one shot.

Our scheme could be used to read out an arbitrary spin, and we will focus our discussion on a system that cannot be measured by any means proposed so far: a molecular spin that is not optically active. Single molecule magnets exhibit a range of quantum phenomena [7] and represent a possible implementation of spin qubits [8]. One of the leading candidates is a fullerene with an endohedral nitrogen (N@C\textsubscript{60}), whose electron spin has a remarkably long coherence time [9]. However, the very property that makes the spin coherence time so long, i.e. weak interaction of the spin with the environment, also makes the readout difficult. One path to single spin readout is to place the molecule in a tiny gap between metal contacts; recent electrical current measurements through an endohedral fullerene show spin dependent features in the current voltage characteristic of the device [10]. The problem with this approach is the unpredictable tunnel coupling of the molecule to the metal leads. In this Letter, we shall show that this difficulty can be overcome by spatially separating the electrical readout from the molecular spin that is measured.

The set-up for our proposed spin meter is shown schematically in Fig. 1. A general obstacle in the sort of device we are proposing is the wide range of relevant energy scales, which at first seem to preclude sensitive spin measurement. An estimate of the dipole-dipole interaction energy between two electron spins is \( J \approx 100 r/r_0 \text{MHz} \), where \( r_0 = 1 \text{ nm} \). For a successful measurement, the current flow through the dot has to be sensitive to a corresponding energy change. The energy resolution of the dot is usually governed by the width of a typical electronic energy level, which depends on the tunnelling rate and, more importantly, by the temperature dependent width of the Fermi function for the electrons in the leads. With attainable temperatures in standard dilution refrigerators (\( \sim 10 \text{ mK} \)) the energy resolution of the dot becomes \( \sim 1 \text{ GHz} \), clearly not enough to resolve the dipole-dipole interaction.

This obstacle can be overcome by considering spin resonance effects that are introduced by applying microwave radiation. Recently there has been an increasing interest in

Figure 1: An implementation of the spin meter concept. The central part of the device is a quantum dot that is defined by electrical gates which pinch off a one-dimensional conductor, such as a nanowire or a carbon nanotube. The electrical transport through the quantum dot can be controlled with high precision by varying the source and drain potentials and the potentials of the extra finger gates placed at the junctions. The charging state of the dot can be sensed through an electrometer charge counter which is coupled to the quantum dot by a conducting bridge, and a back gate can control the number of electrons on the quantum dot. A single spin, e.g. in a fullerene, is placed close to the quantum dot. If there is no electron hopping between the dot and the fullerene, then the main coupling mechanism between the electron spin on the dot and the fullerene spin is the dipole-dipole interaction; there may also be an exchange interaction that could be tuned by inserting a conducting molecule between the fullerene and the nanotube. In such a setup no current will flow directly through the fullerene and thus the spin will not be disturbed.
methods for electrical detection of spin resonance [11, 12]. Mozyrsky and Martin showed that the current in a quantum channel next to an impurity spin depends on frequency of the incident microwave frequency [13]. We showed that a similar resonant change in the current takes place in transport through a quantum dot [14]. The line-width of the resonance introduces a new energy scale into the energy dependence of the dot occupations that can be much smaller than the temperature, subsequently enabling the detection of much smaller shifts in energy. Typical microwave intensities correspond to frequencies \( \sim 1 \text{ MHz} \) but can be made smaller, provided that the lifetime of the electron on the dot is sufficient to allow for equalisation of the spin populations on resonance, i.e. the tunnelling rate should be smaller than the microwave induced Rabi frequency. Given a controllable tunnelling rate the sensitivity of the dot detector can be increased by lowering the microwave intensity and also the tunnelling rate, with the lower limit set by the coherence time of the spin on the dot.

The simplest model capturing all the relevant physics consists of two spin 1/2 particles, one situated on the quantum dot (the flying spin), the other nearby (the stationary spin). In a magnetic field the energy levels for spin up and spin down for each of the spins will be split by the Zeeman energy, but in general the splitting will be different for the flying and the stationary spin whose g-factors are typically unequal. Thus any incident narrow-band microwave radiation can only be resonant with one of the spins. We choose it to be resonant or nearly resonant with the flying spin. The dipole-dipole and exchange coupling between the two spins give rise to an effective Ising interaction, since flip-flop processes are suppressed by the Zeeman terms. The resulting Hamiltonian in the rotating frame after the rotating wave approximation can be written as

\[
H = \frac{\Delta_{0S}}{2} \sigma_z(S) + \frac{\Delta_{0F}}{2} \sigma_z(F) + \frac{\Omega}{2} \sigma_x(F) + \frac{J}{2} \sigma_z(S) \sigma_z(F),
\]

where \( S \) labels the stationary spin and \( F \) the flying spin, \( \Delta_{0S/F} = g_{S/F} \mu_B B_z - \hbar \omega_0 \) with \( B_z \) the magnetic field in the \( z \)-direction, \( \mu_B \) the Bohr magneton, \( g_{S/F} \) the g-factor for the stationary/flying spin respectively and \( \omega_0 \) the microwave frequency. The Rabi frequency due to an oscillating magnetic field in the \( x \)-direction for the flying spin is \( \Omega \); we can neglect the equivalent term for the off-resonant static spin.

The resulting energy level structure is shown in Fig. 2. Decreasing the magnetic field will shift the upper pair of levels towards resonance, with the Rabi frequency dominating their splitting. Meanwhile, the splitting of the lower pair increases. Close to resonance the eigenstates become products of the static spin state with linear superpositions of flying spin up and flying spin down:

\[
|a\rangle_F = a_s \uparrow + a_d \downarrow, \quad |b\rangle_F = b_s \uparrow + b_d \downarrow, \quad s = \uparrow, \downarrow
\]

Figure 2: The level structure of the two spin system in the rotating frame for \( \Delta_{0F} = 0 \). After decreasing the magnetic field the upper two levels (stationary spin down subspace) move into resonance. The difference in the Zeeman splittings is given by \( \delta \Delta_z = \Delta_{0S} - \Delta_{0F} \).

We can derive the rate equations for the stationary spin up subspace, as

\[
\dot{p}_{\uparrow a} = \frac{1}{2} \left( |a_1|^2 \gamma_1^\uparrow + |a_1|^2 \gamma_1^\downarrow \right) p_{\uparrow 0} - \frac{1}{2} \left( |a_1|^2 \gamma_1^\downarrow + |a_1|^2 \gamma_1^\uparrow \right) p_{\uparrow a},
\]

where \( p_{\uparrow a} \) is the probability of finding the stationary spin in state \( \uparrow \) and the flying spin in state \( |a\rangle_F \) and \( p_{\uparrow 0} \) is the probability of finding the stationary spin in state \( \uparrow \) and no electron on the dot, \( \gamma_{\uparrow1}^\downarrow \) are the tunnelling rates onto the dot from either spin up or spin down leads and \( \gamma_{\downarrow1}^\uparrow \) are the tunnelling rates off the dot for different spin populations. They are given by

\[
\gamma_{\uparrow1}^\downarrow = \gamma_{\downarrow1}^\uparrow, \quad \gamma_{\downarrow1}^\uparrow = \gamma_{\uparrow1}^\downarrow + \gamma_{\uparrow1}^\uparrow
\]

where the tunnelling rates from the left and right leads are given by

\[
\gamma_{L\downarrow} = \gamma_0 f(\mu_L - \hbar \omega_0), \quad \gamma_{L\uparrow} = \gamma_0 (1 - f(\mu_L - \hbar \omega_0))
\]
\[ \gamma_{L} = \gamma_{0} f(\mu_{L} + h\omega_{0}), \quad \gamma_{L} = \gamma_{0} [1 - f(\mu_{L} + h\omega_{0})], \]

with similar equations for the right lead, \( f(e) = 1/(1 + \exp(\beta e)) \) being the Fermi function and the chemical potentials in the left and right lead are given by \( \mu_{L/R} = \mu \pm V_{g} \pm V_{sd}/2 \) with \( \mu \) being the chemical potential, \( V_{g} \) the gate voltage and \( V_{sd} \) the bias voltage. A similar rate equation holds for the population of other energy eigenstate

\[ \dot{p}_{1b} = \left( |b|^{2} \gamma_{1}^{<} + |b|^{2} \gamma_{1}^{>} \right) p_{10} - \left( |b|^{2} \gamma_{1}^{<} + |b|^{2} \gamma_{1}^{>} \right) p_{1b}. \]  

(2)

Additionally the individual probabilities have to add up to the probability to find the stationary spin in a spin up state \( p_{1} = p_{1a} + p_{1b} + p_{10} \). The equivalent rate equations can be derived for the stationary spin down subspace and \( p_{1} + p_{1} = 1 \).

The stationary state can be obtained for each stationary spin subspace from \( \dot{p}_{1a/b} = 0 \) and \( \dot{p}_{1a/b} = 0 \) and we obtain the average population on the quantum dot as a function of the applied magnetic field,

\[ \langle n \rangle = 1 - p_{1} L(\Delta_{01}) - p_{1} L(\Delta_{01}) \]  

(3)

with

\[ L(\Delta) = \frac{\Delta^{2}r_{\infty} + \Omega^{2}r_{0}}{\Delta^{2} + \alpha^{2}\Omega^{2}}, \]  

(4)

The coefficients in the Lorentzian Eq. (4) are given by

\[ r_{\infty} = \frac{(1 - f_{+}^{2})^{2} - f_{0}^{2}}{1 + f_{+}^{2} - f_{0}}, \quad r_{0} = \frac{(1 - f_{+})^{2}}{1 + f_{+}^{2} - f_{0}}, \]

and

\[ \alpha^{2} = \frac{1 - f_{0}^{2}}{1 + f_{+}^{2} - f_{0}^{2}} \]

with \( f_{+} = \frac{1}{2} [f_{1L} + f_{1R} + f_{1L} + f_{1R}], \quad f_{-} = \frac{1}{2} [f_{1L} + f_{1R} - f_{1L} - f_{1R}] \) and \( f_{1L} = f(\mu_{L}+h\omega_{0}), \quad x = L, R \).

The behaviour of the average dot population, Eq. (3), as a function of the detuning is shown in Fig. 4. If we want to distinguish spin up from spin down we can monitor the population at resonance, say \( \Delta_{0F} = J \) and consider the change in population from a pure spin up state \( (p_{1} = 1) \) to a pure spin down state \( (p_{1} = 1) \)

\[ \Delta n = L(0) - L(2J). \]

The dependence of \( \Delta n \) on the spin-spin interaction strength \( J \) and the temperature is shown in Fig. 5. The change in occupation number decreases for increasing temperature and decreasing spin-spin interaction strength. In order to detect a small change in the dot occupation number a certain number of electron tunnelling events have to take place. To detect a change of \( \Delta n \) more than \( 1/\Delta n \) electron have to tunnel, therefore the electron tunnelling rate needs to be much larger than the stationary spin relaxation time. In order to see

a resonance of the spin on the quantum dot two things have to be fulfilled: The Rabi frequency has to be stronger than any spin relaxation, intrinsic or tunnelling induced, and the thermal spin polarisation has to be significantly different from zero. For a non-demolition measurement the difference in the spin Zeeman splittings between stationary and flying spin has to be much larger than the Rabi frequency and the spin-spin interaction.

How does this translate to experimental parameters? With a spin coherence time of \( 1 \mu s \) on the carbon nanotube quantum dot we need a Rabi frequency of at least \( \Omega = 10 \text{ MHz} \) to drive the spin into saturation, and thus effect a change of the on dot spin occupation. Given the standard ESR microwave frequency of \( \omega_{0} = 10 \text{ GHz} \) and the corresponding magnetic field of \( 350 \text{ mT} \), at dilution refrigerator temperature \( (T = 50 \text{ mK}) \) we can read out a spin with a relaxation time of \( 0.1 \text{ ms} \) and a
coupling of $J = 3$ MHz. To achieve such a coupling strength purely by dipole-dipole interaction the spin must not be further than 1 nm from the nanotube and the length of the dot must be $\approx 100$ nm. Such a setup would for example allow the readout of a molecular magnet spin [8]. Using a conductive molecule to attach the spin to the nanotube could increase the coupling strength or allow longer distances between nanotube and spin.

Quantum dots in nanotubes have been fabricated (see e.g. [15, 16]), also with integrated charge counters [17, 18], and on-chip microwave resonators reaching Rabi frequencies of $\sim 10$ MHz have been demonstrated (e.g. [19, 20]). We therefore conclude that our scheme is realisable with current technology.

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