Digital magnetic recording is based on the storage of a bit of information in the orientation of a magnetic system with two stable ground states. Here we address two fundamental problems that arise when this is done on a quantized spin: quantum spin tunneling and back-action of the readout process. We show that fundamental differences exist between integer and semi-integer spins when it comes to both, read and record classical information in a quantized spin. Our findings imply fundamental limits to the miniaturization of magnetic bits and are relevant to recent experiments where spin polarized scanning tunneling microscope reads and records a classical bit in the spin orientation of a single magnetic atom.

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The effect of $\mathcal{V}$ on the energy levels is considered weak in the sense that it can be described within lowest order Fermi golden rule. We assume that the correlation time of the reservoirs formed by the electron gases at the tip and surface is short enough so that non-markovian effects are negligible\(^{13}\). The dissipative dynamics of the atomic spin described by $\mathcal{H}_{\text{Spin}}$, under the influence of the dissipative coupling to the tip and substrate, is described in terms of a Bloch-Redfield (BR) master equation in which the coupling to the reservoirs is included up to second order in the coupling $V$: $\partial_t \hat{\rho} = -i [\mathcal{H}_{\text{Spin}}, \hat{\rho}] + \mathcal{L} \hat{\rho}$, with $\mathcal{L}$ the Liouvillian that accounts for the Kondo coupling $V$\(^{13}\). This equation describes the evolution of the diagonal terms in the density matrix, the occupations $\rho_{M,M'} = \rho_{M,M'}$, as well as the off-diagonal terms or coherences, and only the diagonal terms $P_M$ survive\(^{13}\).

The relevant scattering rates can written in terms of

$$\gamma_{\eta \eta'}^{aa'}(\epsilon) = T_a T_{a'} \rho_\eta \rho_{\eta'} v^2 \epsilon^2 \pi \epsilon / 2 \hbar,$$  

where $\epsilon$ is some energy scale relevant for the process in question, $a$ can be 0 or $J$, and $\rho_\eta$ is the density of states at the Fermi energy in electrode $\eta$. The elastic conductance has a contribution coming from the spinless tunneling, $g_0 = 2 e^2 T_0 / \epsilon$, which plays no role in the remainder of the manuscript ($\epsilon$ is the (negative) electron charge). From the experimental linear conductance we get that $\gamma_{TS}^{00}(1\text{meV}) = 1 / \epsilon \approx 0.1 - 5\text{GHz}\(^2\).

The spin-readout is based on a second contribution to the elastic conductance coming from elastic exchange between transport electrons and the spin of the magnetic atom that gives rise to spin-valve term in the total conductance\(^3\):

$$G_{el}(V) \approx g_0 \left[ 1 + 2 T_J \langle \hat{S}_z \rangle \right],$$  

where $\langle \hat{S}_z \rangle$ is the expectation value of the electronic spin:

$$\langle \hat{S}_z \rangle = \sum_M P_M(V) \langle M | \hat{S}_z | M \rangle.$$

Thus, for finite tip polarization, the conductance is sensitive to the expected value of the atomic magnetic moment along $z$. Thus, if the quantum spin can be in two different spin states at zero applied field, ideally with $\langle M | \hat{S}_z | M \rangle \parallel$ parallel and antiparallel to the tip moment, then a magnetoresistive readout of a classical bit of information on a quantum spin is possible.

We now discuss the necessary conditions for the existence two ground states. First, $D$ should be negative. To see this, we consider first the idealized case of a quantum spin with $E = 0$. The energy levels are $E_M = D S_z^2$, with $S_z = \pm S, \pm (S - 1) ...$ If $D$ is positive, the ground states doublet would have $S_z = \pm 1/2$ for semi-integer spin, which can give rise to Kondo effect\(^3\), or $S_z = 0$ for integer spin. In both cases the magnetic moment is zero. Second, the spin should be integer. Kramer’s theorem\(^{14}\) states that, at zero field and with $E \neq 0$, integer spin systems have non-degenerate spectrum, but semi-integer spins have, at least, a twofold degeneracy. These zero-field splittings can be interpreted in terms of quantum spin tunneling, which is suppressed for semi-integer $S$\(^2\).

Thus, the $E$ term splits all the doublets of the $E = 0$ spectrum only for integer $S$. Zero field splitting for integer spins has a very important consequence, which derives from the following general result. For zero applied magnetic field, the matrix elements $\langle M | \hat{S} | M' \rangle$ are zero for every non-degenerate eigenstate of $\mathcal{H}_{\text{Spin}}$\(^{13}\). Thus, from Eq. (5) we get that, at zero applied field, it is impossible to have a net magnetic moment for integer spins. In contrast, for semi-integer $S$, an arbitrary small magnetic field along an arbitrary direction $\Omega$ will choose between the two ground states $g_+$ and $g_-$, resulting in $\langle g_{\pm} | \hat{S}_z | \Omega | g_{\pm} \rangle 
eq 0$. These two states provide the physical realization of the two logical states of the classical bit.

IETS confirms this scenario for Fe ($S = 2$) and Mn ($S = 5/2$) on Cu$_2$N\(^2\). In both cases, $D$ is negative ($D_{Fe} = -1.55 \text{meV}$, $D_{Mn} = -39 \mu\text{eV}$). However, in the case of Fe, there is a single ground state due to quantum spin tunneling induced by $E$, with a null average magnetization. In contrast, for Mn, with $S = 5/2$ the in-plane anisotropy does not lift the degeneracy of the ground state doublet for the Mn, see Fig. (4a),b)

The storage of information in $D < 0$ semi-integer spins is limited by spin relaxation, originated both by elastic and inelastic processes. The later, addressed below, are exponentially suppressed when both bias and temperature are smaller than the excitation energy. In contrast, the rate of elastic scattering between the two ground states $g_{\pm}$ due to coupling to the substrate, for semi-integer $S$ is given by

$$\Gamma_{el} = \gamma_{S,S}^{J_J}(k_B T) \sum_{a=x,y,z} |\langle g_- | S_a | g_+ \rangle|^2,$$

with $k_B$ the Boltzmann constant. The wave functions $|g_{\pm}\rangle$ satisfy

$$|g_{\pm}\rangle \propto \left( |\pm S\rangle + \sum_n c_n \left( E / D \right)^{2n} |\pm S \mp 2n\rangle \right),$$

where $n = 1, 2, ..., S - 1/2$, and $c_n$ are dimensionless numbers of order 1. We see that in-plane anisotropy enables the exchange assisted elastic spin flip\(^{16}\) $|\langle g_- | S_a | g_+ \rangle|^2 \propto (E / D)^{2S-1}$. Thus, elastic population scattering is suppressed as either $S$ or $D / E$ increase. Numerical calculation (see Fig. 2b) yield lifetimes in the range of microseconds for $S = 5/2$ and $|D| = 5E$ at 0.4K.

Importantly, the coupling of the atomic spin to the conduction electrons kills the coherence between the two
ground states $g_{\pm}$, which satisfies the equation $\partial_t \rho_{g_{+}, g_{-}} = -\Gamma_{g_{+}, g_{-}}$. The decoherence rate $\Gamma_{g_{+}, g_{-}}$ contains contributions from both the non adiabatic terms, that imply population scattering, like in Eq. (6), and adiabatic terms, which do not involve exchange energy with the reservoir \cite{13}. The substrate mediated rate for the adiabatic term reads as:

$$\Gamma_{g_{+}, g_{-}}^{ad} = \frac{\gamma_{SS} J S_z (k_B T)}{4} \left| \langle g_{+} | S_z | g_{+} \rangle - \langle g_{-} | S_z | g_{-} \rangle \right|^2.$$  

Thus, coupling to the electronic environment kills quantum coherence more efficiently in states with opposite magnetic moments, acting as a path detector \cite{17} and favoring the magneto resistive readout. To leading order in $E/|D|$, we have $\Gamma_{g_{+}, g_{-}}^{ad} \approx \gamma_{SS} J S_z (k_B T) S^2$. In contrast with population scattering, decoherence rate increases for larger $S$ and is independent of $E/|D|$. Thus, larger $S$ favors spin memory but kills quantum effects. The ratio of the adiabatic decoherence rate, Eq. (3), and the elastic population scattering, Eq. (4), reads $S^2 (|D|/E)^{2S-1}$, which is above $10^3$ for Mn in Cu$_2$N.

We now turn our attention to the effect of parity on the process of magnetic recording, based on atomic scale spin transfer torque, which has only been studied for semi-integer spins so far \cite{2, 3}. Current flowing through the spin-polarized tip, transfers angular momentum to the atomic spin. When the transfer rate exceeds the spin relaxation rate, the spin is driven out of equilibrium. In the case of semi-integer $S$ at zero applied magnetic field, this can result in the occupation of one of the two decohered ground states, $g_{\pm}$, and the depletion of the other, giving rise to a net magnetic moment $\langle S_z \rangle$, according to Eq. (5). The population transfer takes place mainly through inelastic excitation of the spin from the ground state doublet $S_z = \pm S$ to the first excited doublet, via spin-flip exchange. The transition rate where an $\uparrow$ (majority) electron from the tip spin-flips and goes to the surface reads (positive applied voltage in our sign convention) \cite{18}:

$$\Gamma_{incl} \approx \gamma_{JS} J S_z (\Delta + eV) |\langle g_{+} | S^+ | x_{+} \rangle|^2,$$

where we have assumed that $|eV| \gg \Delta$, $k_B T$ while $|x_{+}|$ refers to the excited state connected to $g_{+}$. In fact, the efficiency of the process is greatly enhanced when either bias or temperature are lower than the inelastic excitation energy, $\Delta \simeq (2S - 1)|D|$ for half-integer spin $S$.

In the case of integer spins, inelastic excitations also transfer population between the two tunnel-split ground states but, as the expectation value of the magnetic moment in Eq. (5) at zero applied field in both states is null, $\langle S_z \rangle = 0$, no matter which distribution is achieved. In Fig. (1.b,d) we plot $\langle S_z \rangle$, defined in Eq. (5), as a function of a magnetic field for 3 situations: zero bias, +10meV and -10meV, for both Fe and Mn on Cu$_2$N with finite tip polarization. At zero bias, we obtain the equilibrium Brillouin curve \cite{19}. At finite bias spin transfer favors spin alignment parallel ($V < 0$) or antiparallel ($V > 0$) to the magnetic moment of the tip. The striking difference between integer and semi-integer spin is apparent in the figure. For integer spin, the magnetic moment is always null at zero field and the effect of bias is to heat the atomic spin decreasing the absolute value of $\langle S_z \rangle$ with respect to the zero bias case. For semi-integer spin, the atomic spin takes a bias dependent value at zero field. Thus, we find that current driven control of the magnetic moment of a single spin is only possible for semi-integer $S$.

We now address the problem of back-action and the conditions under which a SP-STM can perform the quantized spin readout without perturbing the atomic spin state, avoiding the loss of the classical information. In other words, we look for a quantum non-demolition measurement \cite{9} of the atomic spin using SP-STM, with the caveat that the atomic spin is decohered. The magneto resistive read-out [Eq. (4)] is made possible by the tunneling exchange coupling between the quantum spin and the transport electrons. Specifically, it is based on the non-spin flip or Ising coupling, $S_z \sigma_z$, which does not flip the atomic spin. However, if tunneling exchange is spin-rotational invariant, Eq. (2), the Ising term goes together with the flip-flop terms, $S^+ \sigma^- + S^- \sigma^+$, which induces atomic spin scattering with the selection rule $\Delta S_z = \pm 1$ and are responsible of the recording (spin-transfer torque). Thus, as in many other instances, the reading mechanism entails some degree of back-action on the probed system. The back-action occurs via inelastic spin-flip events, whose rate $\Gamma_{incl}$ takes off when either
bias or temperature provides the excitation energy, and the elastic spin-tunneling assisted spin-flip, whose rate $\Gamma_{el}$ depends only on $k_BT$, see Fig. 2.

The condition for non-demolition readout is that the measuring time $\tau$ is significantly shorter than the spin-lifetime, $\tau^{-1} \gg (\Gamma_{inel} + \Gamma_{el})$. Regardless of the instrumentation, the measuring time has a fundamental limit given by the condition that shot noise $\delta I$ should be smaller than the current contrast, $\Delta I = \Delta GV$. For Poissonian noise, we have $\delta I = \sqrt{\langle \bar{I} \rangle T}$, where $\bar{I}$ is the average current measured during $\tau$. If we define the average time for a single electron passage, $\tau_e = e/\bar{I}$, then, the limit imposed by shot noise is $\tau \gg \tau_e$. In other words, many tunneling events are necessary to perform the magneto-resistant single spin readout.

Current experiments are done with $\bar{I}$ in the range of nA, which yields $\tau_e \sim 0.2$ ns, so that the measuring time is bound by the shot noise, by Ins. State of the art instrumentation requires much larger measuring times. For instance, the use of lock-in introduces a more stringent bound to $\tau$, in the range of 1$\mu$s-1ms [22, 23]. In Fig. 2a) we plot the substrate mediated elastic spin relaxation rate $\Gamma_{ela}^{SS}$ for and ideal spin system with $D = -5|E| = -1$meV and $T = 0.4K$. This relaxation time grows exponentially with the spin $S$. For an experimentally sensible zero bias conductance $G(0) \approx 0.01 G_0$ [22] ($\rho ST_J = 0.1$), relaxation time above 1$\mu$s can be found in system with $S \geq 5/2$ with excitation energies $\Delta \geq 4$ meV at $T = 0.4K$. The bias dependence of the total relaxation rate is shown in Fig. 2b) [13]. In order to realize a non-demolition measurement, the bias should be kept $|eV| \ll \Delta$. As shown in Fig. 2b), relaxation rate in this regime is dominated by the substrate mediated processes, Eqs. [3].

In summary, we have studied the limitations imposed by quantum mechanics to the use of quantum spins to store classical bits of information. We have found that classical information can be stored in quantum semi-integer spins, for which quantum spin tunneling is suppressed and quantum spin torque is possible, with uniaxial anisotropy $D < 0$. The storage time is limited, when un-observed, by the elastic spin-flip rate (Eq. [4]). Magneto-resistive readout induces additional spin scattering given by the rate [9]. Shot noise imposes a lower limit to the measuring time. Increasing $S$, using for instance few atom ferromagnetically coupled spin clusters, rises dramatically both the elastic and back-action lifetimes, as well as the decoherence rate facilitating the magnetic recording on a quantum spin.

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