Storage of Classical Information in Quantum Spins

F. Delgado[1,2] and J. Fernández-Rossier[1,2]

[1]*International Iberian Nanotechnology Laboratory (INL), Avenida Mestre José Veiga, 4715-330 Braga, Portugal*
[2]*Departamento de Física Aplicada, Universidad de Alicante, 03690 San Vicente del Raspeig, Spain*
(Received 6 December 2011; published 8 May 2012)

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 backaction of the readout process. We show that fundamental differences exist between integer and semi-integer spins when it comes to both reading and recording classical information in a quantized spin. Our findings imply fundamental limits to the miniaturization of magnetic bits and are relevant to recent experiments where a spin-polarized scanning tunneling microscope reads and records a classical bit in the spin orientation of a single magnetic atom.

Recent experimental breakthroughs have laid the foundations for atomic-scale data storage, showing the capability to read and manipulate the spin of a single magnetic atom with a spin-polarized scanning tunneling microscope (SP-STM) [1–3]. Readout is based on tunneling magnetoresistance at the atomic scale [1]: for a fixed spin polarization in the tip, the resistance is higher when the magnetic adatom spin is antiparallel to it. Spin manipulation is based on spin-transfer torque at the atomic scale [2,4]: angular momentum is transferred from the spin-polarized tunneling electrons to the magnetic atom. When the magnetic atom is weakly coupled to the conducting substrate, which can be done thanks to a monoatomic layer of insulating material [5], the spin of the magnetic atom is quantized, and it can be described by a single spin Hamiltonian, identical to that of single molecule magnets [6], as revealed by inelastic electron tunneling spectroscopy (IETS) [2,5,7–9].

In this Letter, we address two fundamental questions that arise when considering magnetic recording in the quantum limit, i.e., the storage and readout of a classical bit of information on the magnetization of a quantized spin. First, what is the role played by spin parity in the readout and control operations of a quantized spin? Here we show that the two physically different stable states with opposite magnetization required in digital magnetic recording appear only in the case of semi-integer spins [10], for which quantum spin tunneling [6,11,12] is forbidden. We also demonstrate that zero-field current-induced single atom spin switching is only possible for semi-integer spins. The second question is how can the magnetoresistive single spin readout be performed without disturbing the spin state? Here we study the problem of the backaction, akin to the quantum nondemolition [13] problem on a decohered qubit.

The physical system of interest consists of a magnetic atom with a quantized spin S [2,5,7–9]. The magnetic atom is probed and controlled by a SP-STM. The quantized spin

of an atomic-scale nanomagnet on a surface can be described with a single ion Hamiltonian [2,5,7–9]:

$$\mathcal{H}_{\text{Spin}} = D\hat{S}_z^2 + E(\hat{S}_x^2 - \hat{S}_y^2) + g\mu_B \hat{\vec{S}} \cdot \vec{B}, \quad (1)$$

where D and E define the uniaxial and in-plane magnetic anisotropy. Eigenvalues and eigenfunctions of $\mathcal{H}_{\text{Spin}}$ will be denoted by E_M and $|M\rangle$, respectively. The above Hamiltonian accounts for the measured IETS in $S = 1$ Fe phthalocyanine [14] and Mn-12-acetate ($S = 10$) [15], and transition metal adatoms: Co ($S = 3/2$) [7], Fe ($S = 2$), and Mn ($S = 5/2$) [5].

The Hamiltonian of the total system features a single ion Hamiltonian exchange coupled to the transport electrons [4,16,17], $\mathcal{H} = \mathcal{H}_T + \mathcal{H}_S + \mathcal{H}_{\text{Spin}} + \mathcal{V}$, where $\mathcal{H}_T + \mathcal{H}_S = \sum_{\lambda,\sigma} \epsilon_\sigma(\lambda) c_{\lambda,\sigma}^\dagger c_{\lambda,\sigma}$ describes the tip and surface electrodes, with quantum numbers $\lambda \equiv (\vec{k}, \eta)$ and σ, the momentum $\vec{k}$, electrode ($\eta = T, S$), and spin projection σ along the tip polarization axis. We assume a spin-polarized tip with polarization $\vec{\mathcal{P}}_T$ and a spin-unpolarized substrate S. The $\mathcal{V}$ term describes interactions between tip, surface, and the magnetic atom:

$$\mathcal{V} = \sum_{\lambda,\lambda',\sigma,\sigma'} \left(\mathcal{T}_{\lambda,\lambda'}^{(0)} \frac{\delta_{\sigma\sigma'}}{2} + \mathcal{T}_{\lambda,\lambda'} \vec{S} \cdot \frac{\vec{\sigma}_{\sigma\sigma'}}{2} \right) c_{\lambda,\sigma}^\dagger c_{\lambda'\sigma'}, \quad (2)$$

with $\vec{\sigma}$ the Pauli matrices vector and $\vec{S}$ the magnetic atom spin. Equation (2) describes both spin-independent tunneling, described by the $\mathcal{T}_{\lambda,\lambda'}^{(0)}$ term, and spin-dependent processes, described by the $\mathcal{T}_{\lambda,\lambda'}$ term, where carriers can either remain in the same electrode, providing the most efficient atomic-spin relaxation channel, or switch sides, which gives rise to spin-dependent tunneling current. Neglecting their momentum dependence, the exchange integrals can be written as $\mathcal{T}_{\lambda\lambda'}^{(0)} = v_\eta v_{\eta'} T_0$ and $\mathcal{T}_{\lambda,\lambda'} = T_J v_\eta v_{\eta'}$, with the spinless T_0 and spinfull T_J tunneling

matrix elements, while ν_T and ν_S are two dimensionless parameters that describe the strength of the tip-atom and surface-atom single-particle hoppings.

In the weak coupling regime, the effect of $\mathcal{V}$ can be accounted for within the 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 [18]. The dissipative dynamics of the atomic 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 $\mathcal{V}$: $\partial_t \hat{\rho} = -\frac{i}{\hbar}[\mathcal{H}_{\text{Spin}}, \hat{\rho}] + \mathcal{L}\hat{\rho}$, with $\mathcal{L}$ the Liouvillian that accounts for the Kondo coupling $\mathcal{V}$ [18]. This equation describes the evolution of the diagonal terms in the density matrix, the occupations $P_M \equiv \rho_{M,M}$, as well as the off-diagonal terms or coherences, $\rho_{M,M'}$. In steady state, the density matrix described by the BR master equation only contains diagonal terms [18].

It is convenient to define the following scattering rate

$$\gamma_{\eta,\eta'}^{aa'}(\epsilon) = T_a T_{a'} \rho_\eta \rho_{\eta'} \nu_\eta^2 \nu_{\eta'}^2 \frac{\pi\epsilon}{2\hbar}, \tag{3}$$

where ϵ is some energy scale relevant for the process in question, $a \equiv 0, J$, and ρ_η is the density of states at the Fermi energy in electrode η.

The elastic conductance has a contribution coming from the spinless tunneling, $g_0 \equiv 2e^2 \partial_\epsilon \gamma_{TS}^{00}(\epsilon)$, which plays no role in the remainder of the manuscript (e is the elementary charge). From the experimental linear conductance we get that $\gamma_{TS}^{00}(1 \text{ meV}) = I/e \sim 0.1\text{–}5$ GHz [2]. The spin read-out is based on a second contribution to the elastic conductance coming from elastic exchange between transport electrons and the spin S, which gives rise to a spin-valve term in the total conductance [4]:

$$G_{\text{el}}(V) \approx g_0 \left[1 + 2\frac{T_J}{T_0}\langle \vec{S}\rangle \cdot \vec{\mathcal{P}}_T \right], \tag{4}$$

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

$$\langle \vec{S}\rangle = \sum_M P_M(V)\langle M|\vec{S}|M\rangle. \tag{5}$$

Thus, for finite tip polarization, the conductance is sensitive to the expected value $\langle \vec{S}\rangle$. Thus, if the quantum spin at zero-applied field can be in two different spin states with different $\langle M|\vec{S}|M\rangle$, ideally 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 of two degenerate ground states. First, the spin should be semi-integer. Kramer's theorem [10] states that, at zero field and with $E \neq 0$, integer spin systems have a nondegenerate 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 [11]. 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 $\langle M|\vec{S}|M\rangle$ are zero for every nondegenerate eigenstate of $\mathcal{H}_{\text{Spin}}$ [19]. This has far reaching consequences: at zero field, quantized integer spins do not have net magnetic moment, making them invisible to both magnetoresistive detection and magnetic imaging.

Second, for semi-integer spin D should be negative, so that the zero-field ground state doublet is not the $S_z = \pm 1/2$, which might result in a Kondo effect [7], but the doublet with maximal $\pm S_z$ for which the Kondo temperature is exponentially reduced [20]. For semi-integer S and $D < 0$, an arbitrary small magnetic field along an arbitrary direction $\hat{\Omega}$ will choose between the two ground states "+" and "−," resulting in $\langle \pm|\vec{S}\cdot\hat{\Omega}|\pm\rangle \neq 0$. These two states provide the physical realization of the two logical states of the classical bit.

In the following, we illustrate our discussion with the case of Fe and Mn deposited on a decoupling monolayer of Cu_2N on Cu. IETS on this system [5] portraits Fe as $S = 2$ spin with $D_{\text{Fe}} = -1.55$ meV and $E_{\text{Fe}} = 0.31$ meV and single ground states, $|+_{\text{Fe}}\rangle \simeq |+2\rangle + |-2\rangle$, and Mn as a $S = 5/2$ spin with $D_{\text{Mn}} = -39$ μeV and $E_{\text{Mn}} = 7$ μeV and two degenerate ground states at zero field. A scheme of their energy levels is shown in Figs. 1(a) and 1(b).

Even if the two ground states of a semi-integer spin are protected from quantum spin tunneling, we have to consider how their exchange coupling to the electrons in the substrate affects their capability to store a classical bit. Whereas inelastic scattering is exponentially suppressed when both bias and temperature are smaller than the excitation energy, elastic scattering is unavoidable and has a profound influence, giving rise both to decoherence and to population scattering.

For the two ground states $\pm$ of the quantized spin, to behave as a classical bit, quantum coherence between them needs to be suppressed [21]. Within the BR approach, the coherence between the relevant off-diagonal element of the density matrix element satisfies the equation $\partial_t \rho_{+,-} = -\gamma_{+,-}\rho_{+,-}$ [18]. The decoherence rate $\gamma_{+,-}$ contains contributions from both the inelastic terms and the elastic or pure dephasing terms, which do not involve population transfer [18]. The substrate-mediated pure dephasing rate, $\gamma_{+,-}^{el} = 1/T_2^{el}$, is given by:

$$\gamma_{+,-}^{\text{el}} = \frac{\gamma_{SS}^{JJ}(k_B T)}{4}|\langle +|S_z|+\rangle - \langle -|S_z|-\rangle|^2, \tag{6}$$

with k_B the Boltzmann constant. This formula has an appealing physical interpretation: the electrons in the reservoir act as a spin *which-path detector* [22] that, due to
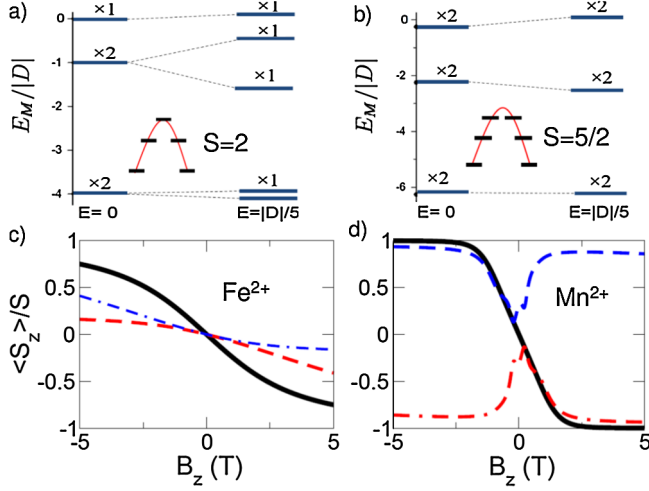
FIG. 1 (color online).   Schematic evolution of the energy spectrum and degeneracies versus E for: (a) integer spin $S = 2$ and (b) half-integer spin $S = 5/2$. Magnetization curves of the Fe (c) and Mn (d) adatoms probed with a spin-polarized tip with $\mathcal{P}_T = 0.74$ for three applied biases: $V = 0$ (thick solid line), $V = -10$ meV (dashed line), and $V = +10$ meV (dotted-dashed line). Here $T = 0.5$ K, $T_J/T_0 = 0.5$, $v_\eta = 1$, and $\rho_T = \rho_S$.

FIG. 2 (color online).   (a) Substrate-mediated elastic spin relaxation rate Γ^{el} in units of $\gamma_0 \equiv \gamma_{S,S}^{JJ}(1 \text{ meV})$ for an ideal half-integer spin system with $D = -5|E| = -1$ meV and $T = 0.4$ K. (b) Total relaxation rate ($\Gamma^{inel} + \Gamma^{el}$) for $S = 5/2$ at three different temperatures. Here $\mathcal{P}_T = -1$, $v_S = 1$, $v_T = 0.7$, $T_J/T_0 = 0.5$, and $\rho_S^2 T_J^2 = 0.01$.

spin-conserving exchange, is sensitive to the different spin orientation of the ground states of the magnetic atom and destroys the coherence between them [23]. The ground states are given by $|g_\pm\rangle \propto [|\pm S\rangle + \sum_n c_n(\frac{E}{D})^{2n}|\pm S \mp 2n\rangle]$, where $n = 1, 2, \ldots, S - \frac{1}{2}$, and c_n are dimensionless numbers of order 1, so that we have $\gamma_{+,-}^{el} \simeq \gamma_{SS}^{JJ}(k_B T)S^2$, plus corrections of order $(E/|D|)^2$.

In addition, elastic exchange to the substrate electrons causes population transfer between the two ground states, and thereby memory loss. The corresponding rate $\Gamma^{el} = 1/T_1^{el}$ is given by

$$\Gamma^{el} = \gamma_{S,S}^{JJ}(k_B T) \sum_{a=x,y,z} |\langle -|S_a|+\rangle|^2. \qquad (7)$$

The elastic scattering matrix element can be approximated by $|\langle -|S_a|+\rangle|^2 \propto (E/D)^{2S-1}$, so that population scattering is only possible due to the combined action of exchange and in-plane anisotropy [20]. The ratio between decoherence and population scattering rate yields $T_1^{el}/T_2^{el} \approx S^2(|D|/E)^{2S-1}$, 4 orders of magnitude larger than the minimal value $T_1/T_2 = 1/2$, in a system without pure dephasing. This ratio is approximately 6×10^3 for Mn on Cu$_2$N. Numerical calculations, shown in Fig. 2(a), yield population lifetimes in the range of microseconds for $S = 5/2$ and $|D| = 5E = 1$ meV at 0.4 K.

We now turn our attention to the effect of parity on the process of magnetic recording, based on atomic-scale spin-transfer torque. The 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
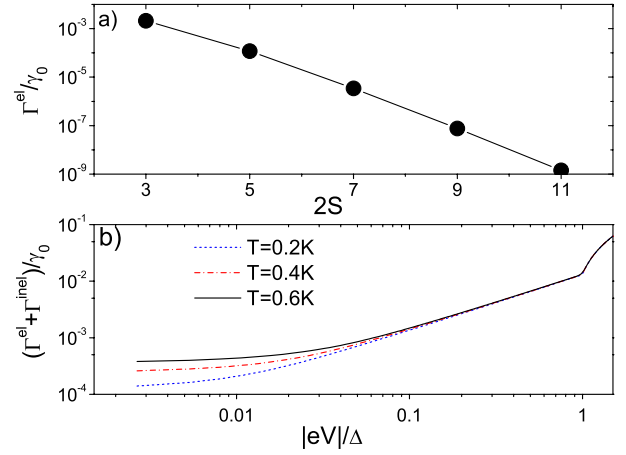
semi-integer S at zero-applied magnetic field, this can result in the preferential occupation of one of the two ground states and the depletion of the other producing a net magnetic moment $\langle S_z \rangle$, see Eq. (5). The population transfer between the two ground states takes place through inelastic excitation of the excited state doublets by exchanging the spin with the transport electrons [2,4]. The transition rate where a majority electron from the tip spin flips and goes to the surface reads (positive applied voltage in our sign convention) [24]:

$$\Gamma^{inel} \approx \gamma_{TS}^{JJ}(|\Delta + eV|)|\langle +|S^+|x_+\rangle|^2, \qquad (8)$$

where we have assumed that $|eV| \gg \Delta$, $k_B T$ while $|x_+\rangle$ refers to the excited state connected to $|+\rangle$. In fact, the efficiency of the process is greatly enhanced when either bias or temperature are higher 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 field is null in both states, then $\langle S_z \rangle = 0$, no matter which nonequilibrium distribution is achieved.

In Figs. 1(c) and 1(d) we plot $\langle S_z \rangle$, defined in Eq. (5), as a function of a magnetic field for 3 situations: zero bias, $+10$ meV, and -10 meV, for both Fe and Mn on Cu$_2$N with finite tip polarization. At zero bias, we obtain a paramagnetic equilibrium magnetization curve, with significantly larger slope for the semi-integer case. 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. Hence, we find that zero-field current driven control of the magnetic moment of a single spin is only possible for semi-integer spin.

We now address the problem of backaction 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 nondemolition measurement [13] of the atomic spin using SP-STM, with the caveat that the atomic spin is decohered. The magnetoresistive readout, 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, due to the spin-rotational invariance of the tunneling exchange, 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 permits the recording through the spin-transfer torque. Thus, as in many other instances, the reading mechanism entails some degree of backaction on the probed system. The backaction occurs via inelastic spin-flip events, Γ^{inel}, and elastic spin tunneling assisted by spin flip, Γ^{el}. The rate Γ^{inel} takes off when either bias or temperature provides the excitation energy, while Γ^{el} depends only on $k_B T$, see Eq. (7).

A nondemolition readout of the spin requires a measuring time τ significantly shorter than the spin lifetime, $\tau \ll T_1 \equiv (\Gamma^{\text{inel}} + \Gamma^{\text{el}})^{-1}$. Regardless of the instrumentation, the measuring time has a fundamental limit given by the condition that shot noise δI should be smaller than the current, $\delta I / \bar{I} \ll 1$, where $\bar{I}$ is the average current measured during τ. For Poissonian noise, we have $\delta I = (\frac{e}{\tau} \bar{I})^{1/2}$, which defining the average time for a single electron passage, $\tau_e = e / \bar{I}$, translates into the condition $\tau \gg \tau_e$. In other words, many tunneling events are necessary to perform the magnetoresistive 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 below by 1 ns. However, state of the art instrumentation requires much larger measuring times, in the range of 1 μs–1 ms [25,26]. Our numerical simulations, summarized in Fig. 2(a), show that at $T = 0.4$ K, in order to have a zero-current spin lifetime longer than the state of the art measuring time, we would require systems with $S \geq 5/2$ and excitation energies $\Delta \gtrsim 4$ meV, assuming an experimentally attainable [2] zero bias conductance $G(0) \approx 0.01 G_0$. In order to satisfy the $T_1 \gg \tau \sim 1$ μs criteria, for Mn on Cu$_2$N, temperature should be reduced below 10 mK. We note that Co on Cu$_2$N has $S = 3/2$ and $\Delta \simeq 5$ meV, but positive D [7]. Figure 2(b) also shows that, during the magnetoresistive readout, the inelastic scattering can be kept 1 order of magnitude below the elastic one

provided that $|eV|$ is kept below 0.1Δ. All things considered, the nondemolition readout is almost within reach of state of the art techniques.

In summary, we have studied the limitations imposed by quantum mechanics to store a classical bit of information in the magnetization direction of a quantized spin. We have considered two different types of quantum effects that limit classical magnetic recording: spin parity and backaction during a magnetoresistive readout. We have found that only semi-integer spins with an easy axis can have two ground states with opposite magnetization which can be read and written using a SP-STM. The storage time is limited, when unobserved, by the elastic spin-flip rate. Magnetoresistive readout induces additional inelastic spin scattering. In addition, shot noise imposes a lower limit to the measuring time when doing a nondemolition measurement of the quantum spin.

[1] R. Wiesendanger, Rev. Mod. Phys. **81**, 1495 (2009).

[2] S. Loth, K. von Bergmann, M. Ternes, A. F. Otte, C. P. Lutz, and A. J. Heinrich, Nature Phys. **6**, 340 (2010).

[3] S. Loth, S. Baumann, C. P. Lutz, D. M. Eigler, and A. J. Heinrich, Science **335**, 196 (2012).

[4] F. Delgado, J. J. Palacios, and J. Fernández-Rossier, Phys. Rev. Lett. **104**, 026601 (2010).

[5] C. Hirjibehedin, C.-Y. Lin, A. F. Otte, M. Ternes, C. P. Lutz, B. A. Jones and A. J. Heinrich, Science **317**, 1199 (2007).

[6] D. Gatteschi, R. Sessoli, and J. Villain, *Molecular Nanomagnets* (Oxford University Press, New York, 2006).

[7] A. F. Otte, M. Ternes, K. von Bergmann, S. Loth, H. Brune, C. P. Lutz, C. F. Hirjibehedin and A. J. Heinrich1, Nature Phys. **4**, 847 (2008).

[8] S. Loth, C. P. Lutz and A. J Heinrich, New J. Phys. **12**, 125021 (2010).

[9] A. A. Khajetoorians, B. Chilian, J. Wiebe, S. Schuwalow, F. Lechermann, and R. Wiesendanger, Nature (London) **467**, 1084 (2010).

[10] H. A. Kramers, Proc. Amsterdam Acad. **33**, 959 (1930).

[11] D. Loss, D. P. DiVincenzo, and G. Grinstein, Phys. Rev. Lett. **69**, 3232 (1992).

[12] W. Wernsdorfer and R. Sessoli, Science **284**, 133 (1999).

[13] V. B. Braginsky and F. Y. Khalili, Rev. Mod. Phys. **68**, 1 (1996).

[14] N. Tsukahara *et al.*, Phys. Rev. Lett. **102**, 167203 (2009).

[15] S. Kahle *et al.*, Nano Lett. **12**, 518 (2012).

[16] F. D. Novaes, N. Lorente, and J.-P. Gauyacq, Phys. Rev. B **82**, 155401 (2010).

[17] J. Fransson, Nano Lett. **9**, 2414 (2009).

[18] C. Cohen-Tannoudji, G. Grynberg, and J. Dupont-Roc, *Atom-Photon Interactions* (Wiley and Sons, Inc., New York, 1998).

[19] M. J. Klein, Am. J. Phys. **20**, 65 (1952).

[20] C. Romeike, M. R. Wegewijs, W. Hofstetter, and H. Schoeller, Phys. Rev. Lett. **96**, 196601 (2006).

[21] W. H. Zurek, Phys. Today **44**, No. 10, 36 (1991).

[22] E. Buks, R. Schuster, M. Heiblum, D. Mahalu and V. Umansky, Nature (London) **391**, 871 (1998).

[23] A. Stern, Y. Aharonov, and Y. Imry, Phys. Rev. A **41**, 3436 (1990).

[24] F. Delgado and J. Fernández-Rossier, Phys. Rev. B **82**, 134414 (2010).

[25] P. A. Sloan, J. Phys. Condens. Matter **22**, 264001 (2010).

[26] S. Guo, J. Hihath, and N. Tao, Nano Lett. **11**, 927 (2011).