Electrically detected single-spin resonance with quantum spin Hall edge states

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Detection is most often the main impediment to reduce the number of spins in paramagnetic resonance experiments. Here, we propose another route to carry out electrically detected spin resonance of an individual spin, placed at the edge of a quantum spin Hall insulator (QSHI). The edges of a QSHI host a one-dimensional electron gas with perfect spin-momentum locking. Therefore, the spin relaxation induced by emission of an electron-hole pair at the edge state of the QSHI can generate current. Here, we demonstrate that driving the system with an ac signal, a nonequilibrium occupation can be induced in the absence of applied bias voltage, resulting in a dc measurable current. We compute the dc current as a function of the Rabi frequency Ω, the spin relaxation, and decoherence times T_1, and we discuss the feasibility of this experiment with state-of-the-art instrumentation.

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I. INTRODUCTION

The sensitivity limit of commonly available electron paramagnetic resonance (EPR) spectrometers is in the range of 10^{13} spins [1]. This number can be dramatically reduced in tailored setups [2]. In some special systems, such as nitrogen-vacancy (NV) centers, one is permitted to carry out single-spin resonance using optical readout, made possible both by the fact that NV centers are very good single-photon emitters and their photon yield is spin dependent [3]. Using spin-to-charge conversion, electrically detected single-spin resonance has been demonstrated for defects in field-effect transistors [4], quantum dots [5,6], and single dopants in silicon [7]. Electrically detected single-spin resonance with subatomic spatial resolution has been also demonstrated [8] using electron spin resonance scanning tunneling microscopy (ESR-STM).

Here, we explore the spin-locked edge states of a two-dimensional quantum spin Hall insulator [9,10] (QSHI) to accomplish the electrical readout of the spin resonance of an individual spin sitting on the edge. The edge states of QSHI are predicted to have a one-to-one relation between the propagation direction and the spin orientation along a system-dependent spin quantization axis (see Fig. 1). As a result, pumping spin along this axis entails electrical current flow. As we discuss below, if an externally pumped localized spin is exchange coupled to the spin-locked edges, it will generate a dc current.

Experimental evidence of the spin-locked edge states in QSHI is indirect. In the absence of magnetic impurities, edge states should have no backscattering and therefore a quantized conductance is expected [11–13]. Values of conductance close to $2e^2/h$ were reported in HgTe/CdTe quantum wells [14] and $1T'$ WTe_2 [15,16]. In addition, coherent propagation along the edge with scattering properties consistent with the strong suppression of backscattering have been observed in bismuth bilayers [17,18], and in bismuth nanocontacts [19]. Very relevant for the ensuing discussion, experiments where magnetic atoms have interacted with the edge states in the bismuth bilayer and produced backscattering have been demonstrated [20].

The interplay between local spins and the spin-locked edge states of a QSHI has been widely studied theoretically [21–37]. Several physical realizations of the local spin have been considered, including a confined electron in a quantum dot [29,35], nanomagnets [28], magnetic atoms [26,27], spin chains [36], nuclear spins [25,33,37], and magnetic molecules [32]. Early works focused on the Kondo effect [21,24], and the influence of magnetic impurities on conductance [22]. More recent works have addressed the spin pumping of local moments at the edges by the helical-electron spin current [26–29,31,32,34]. The reverse problem, pumping dc current by an external ac excitation of nuclear spins, has been addressed recently [37]. On a similar standpoint, here we assess whether the paramagnetic spin resonance of an individual spin, in the form of an individual magnetic atom, spin chain, or magnetic molecule, could be carried out.

The rest of this paper is organized as follows. In Sec. II we introduce the basic principles of the electrically detected single-spin resonance in a QSHI. An estimation of the maximum dc current is provided in Sec. III, while the main limiting factors are discussed in Sec. IV. Finally, a brief summary and conclusions are given in Sec. V.

II. ELECTRICALLY DETECTED SINGLE-SPIN RESONANCE IN QSHI EDGE STATES

This work builds on the following idea: At the edge states of QSHI, an electron with spin $-\sigma$ and momentum $-k_F$ can

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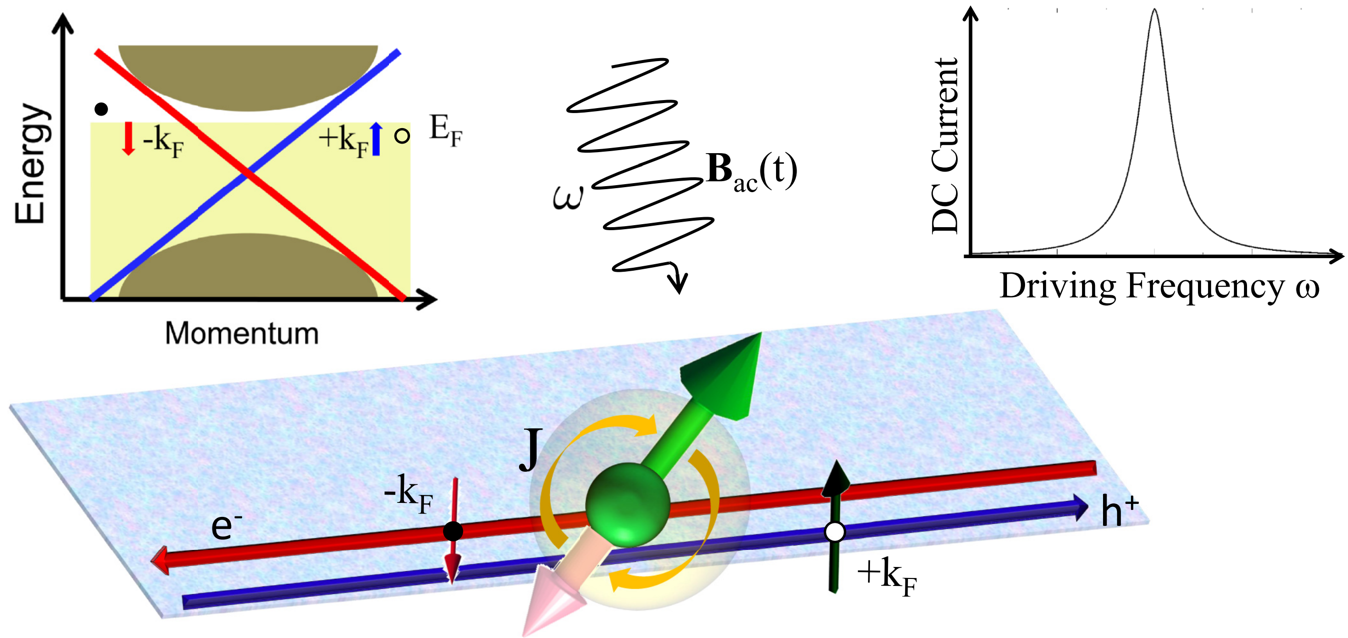

FIG. 1. Scheme of the device proposed for the electrically detected single-spin detection. A local spin S is exchange coupled to one edge state of a QSHI, where momentum direction and spin orientation are locked (see the left inset). When the system is under the action of an external ac magnetic field with frequency ω and intensity determined by the Rabi rate Ω, a frequency-dependent nonequilibrium steady state occupation is established (see the right inset), where one of the spin transitions ΔS_z is favored. This in turns leads to a net dc electrical current along the QSHI edge.

be scattered to a state with momentum $+k_F$ and spin σ by an exchange interaction with a local spin. Here, σ is defined along a material-dependent axis that, without loss of generality, we label as z. Since the total spin has to be conserved in the process, the spin change of the electron has to be compensated by the spin change of the local magnetic moment. Unless otherwise stated, we only consider a local spin whose spin quantization axis is aligned along z, the same quantization axis of the quasiparticle states. Therefore, for a given local-spin transition with a change of spin $\Delta S_z = \pm 1$, the quasiparticles undergo a $\pm 2k_F$ backward-scattering process with $\Delta\sigma = \mp 1$, on account of their helical nature.

Crucially, the electron-hole pairs carry a net current whose sign depends on the sign of $\Delta\sigma$. The extra electron in one branch and the missing electron in the opposite branch contribute to current flow with the *same* polarity. As the edge states are expected to have no backscattering, the electron and the hole will reach the electrodes and contribute to the current. Hence, if we create a stationary nonequilibrium imbalance in the ΔS_z transitions by driving the spin transitions with an external ac driving field, a net dc current will be generated.

We now substantiate the argument mathematically. Let us take for simplicity a local $S = 1/2$ spin moment under the influence of a static magnetic field, $B_{\rm eff} = \hbar\omega_0/(g\mu_B)$, where $B_{\rm eff} = B_z + B_z^{\rm other}$ is the sum of the external field B_z and other contributions that could arise from the interaction of the local spin and its environment. Thus, the stationary spin Hamiltonian can be written as $\mathcal{H}_0 = \frac{\hbar\omega_0}{2}\hat{\tau}_z$, with $\hat{\tau}_z$ the z-Pauli matrix, and $\hbar\omega_0 = \epsilon_1 - \epsilon_0 \geqslant 0$. We label P_0 and P_1 as the probabilities of occupying the ground (0) and excited (1) states, respectively. The relevant spin-exchange process that gives place to spin flips is governed by the Hamiltonian [36]

$$H_{\rm sf}^{\rm QSHI} = \sum_{k,k'} \frac{J}{2L}(\hat{S}^+ L_k^\dagger R_{k'} + \text{H.c.}), \tag{1}$$

where J is the exchange coupling constant, L is the length of the edge, and $L_k^\dagger \equiv c^\dagger_{-(k_F+k),\downarrow}$ and $R_k^\dagger \equiv c^\dagger_{+(k_F+k),\uparrow}$ are the left (right) moving fermion operators, with $c^\dagger_{k\sigma}$ the creation operator of a fermion in the edge channel with spin σ and momentum k. Here, $\hat{S}^\pm = 1/2(\hat{S}^x \pm i\hat{S}^y)$ are the spin-ladder operators.

If we define the rate $\Gamma_{1\to 0}$ and $\Gamma_{0\to 1}$ of the $\Delta S_z = \mp 1$ process, respectively, we can write the electric current flowing to the right as

$$I = 2e(P_1\Gamma_{1\to 0} - P_0\Gamma_{0\to 1}), \tag{2}$$

where e is the elementary charge and P_i are the nonequilibrium occupations of the $i \equiv 0, 1$ states. In equilibrium, the current (2) vanishes because the scattering rates satisfy the detailed balance principle

$$\frac{\Gamma_{1\to 0}}{\Gamma_{0\to 1}} = \frac{P_0^{\rm eq}}{P_1^{\rm eq}} = e^{\beta\hbar\omega_0}, \tag{3}$$

where $1/\beta = k_B T$ and $P_i^{\rm eq}$ are the equilibrium occupations. We shall now demonstrate that if the local spin is driven away from equilibrium by some external force that does not significantly modify the rates, then a net current can occur. If we write $P_i = P_i^{\rm eq} \pm \delta P/2$, where the $+$ $(-)$ sign corresponds to $i = 1$ $(i = 0)$, and taking into account that in thermal equilibrium (without any applied bias voltage) the net current is null, then we have

$$I_{\mathrm{dc}} = e\delta P\Gamma_{1\to 0}(1 + e^{-\beta\hbar\omega_0}). \tag{4}$$

This equation is the starting point of our analysis. It relates the out-of-equilibrium occupations of the two-level system and a net current flow. The direction of the current is established by the chirality of the spin edge and by the sign of the magnetic field. For a fixed edge, the reversal of the magnetic field would lead to current flow in the opposite direction.

Let us consider now the case where the local spin is also under the action of an ac transverse magnetic field $B_x(t) \equiv 2\hbar\Omega/(g\mu_B)\cos(\omega t)$, where Ω is known as the *Rabi frequency* or *flop rate*. When the local spin is driven by $B_x(t)$ with the frequency ω close enough to the natural frequency ω_0, the nonequilibrium occupations P_i can deviate significantly from their equilibrium counterpart P_i^{eq}. In particular, for a two-level system the occupation imbalance $\Delta P = P_0 - P_1$ is given by the steady state solution of the Bloch equations [38,39]. Thus, using the definition of δP, we can write

$$\delta P = \Delta P^{\mathrm{eq}} \frac{\Omega^2 T_1 T_2}{1 + \delta^2 T_2^2 + \Omega^2 T_1 T_2}, \tag{5}$$

where $\Delta P^{\mathrm{eq}} \equiv \tanh(\beta\hbar\omega_0/2)$ is the equilibrium population imbalance and $\delta = \omega - \omega_0$ is the frequency detuning. In addition to the equilibrium imbalance, the nonequilibrium occupation difference, and therefore the induced electrical current, depends on the Rabi flop rate and the two characteristic timescales, the longitudinal relaxation time $T_1 = 1/(\Gamma_{0\to 1} + \Gamma_{1\to 0})$ and the decoherence time T_2, also known as the transversal relaxation time in the language of macroscopic Bloch equations [38]. If we make the substitution of Eq. (5) into the current expression (4), we get

$$I = I_0 \Delta P^{\mathrm{eq}} \frac{\Omega^2 T_1 T_2}{1 + \delta^2 T_2^2 + \Omega^2 T_1 T_2}, \tag{6}$$

where

$$I_0 = \frac{e}{2T_1}. \tag{7}$$

Equation (6) is the main result of this paper. It predicts a dc current flowing at the edge of a quantum spin Hall when a single localized spin is driven with an ac field.

III. ESTIMATE OF MAXIMAL dc CURRENT

The maximal induced dc current is obtained at resonance ($\delta = 0$), when the driving frequency matches the Zeeman frequency, and it is given by

$$I_{\max} = I_0 \Delta P^{\mathrm{eq}}, \tag{8}$$

obtained when $\Omega^2 T_1 T_2 \gg 1$ and assuming T_1 is entirely due to the Kondo exchange mechanism envisioned in Fig. 1. The maximal equilibrium spin polarization $\Delta P^{\mathrm{eq}} = 1$ is achieved only when the low-energy spin state is fully occupied, i.e., $\beta\hbar\omega_0 \gg 1$, where $I_{\max} = I_0$. In other words, the magnitude of the maximal current is determined by T_1 provided $\Omega^2 T_1 T_2 \gg 1$. In this limit, the spin relaxation rate due to the Kondo exchange for a single $S = 1/2$ spin interacting with the spin-locked edge of a QSHI is given by [36]

$$\frac{1}{T_1} \approx \frac{(\rho J)^2 \pi}{16}\omega_0, \tag{9}$$

where ρ is the density of states at the Fermi energy of the edge electrons. Equation (9) is derived taking ρJ as a small parameter. Therefore, an upper bound for the dc current is given by

$$I_{\max}^{\mathrm{theo}} < \frac{e\pi}{32}\omega_0. \tag{10}$$

For a dc field of 1 T ($\omega_0 \approx 1.8 \times 10^{11}$ s^{-1}), the standard for ESR experiments, $I_{\max}$ is in the nA regime for $T \ll 1.3$ K, well within the instrumental state of the art. We note that nuclear Zeeman splitting is three orders of magnitude smaller than its electronic counterpart, and the hyperfine interaction is at least three orders of magnitude smaller than the Kondo exchange. Therefore, nuclear spin relaxation rates, that scale with the square of the hyperfine interaction, will be many orders of magnitude smaller than their electronic counterparts.

Although Eq. (7) naively implies that a T_1 as short as possible is desired, the inequality $\Omega^2 T_1 T_2 \gg 1$ must also hold. Given that $T_2 < 2T_1$, a short T_1 requires a large Rabi coupling Ω. Thus, T_1 must remain above $1/\Omega$ so the maximal current criteria is satisfied. In practice, this leads to the stricter condition

$$I_{\max} < e\Omega. \tag{11}$$

In conventional ESR experiments, the spin is driven by the ac magnetic field of a microwave. Typically, cavities are used to increase the magnitude of the ac field. State-of-the-art values for the ac magnetic field in ESR experiments can be larger than 250 mG [1]. For a spin $S = 1/2$ with $g = 2$ this gives $\Omega \simeq 0.7$ MHz and, from Eq. (11), $I < 120$ fA, well above state-of-the-art current detectors than can detect changes as small as 10 fA [8,40].

Larger values of Ω have been achieved using ESR-STM, where several different driving mechanisms other than the Zeeman interaction with the ac field have been proposed [41–44]. For Ti-H on MgO, and an $S = 1/2$ spin system, ac magnetic fields up to 1 mT have been reported [40], with an induced Rabi frequency $\Omega/2\pi \sim 10$ MHz in continuous mode, while Rabi frequencies up to 30 MHz have been demonstrated in pulsed ESR-STM [45] or using double resonance under large ac voltages [46]. Moreover, these conditions can be achieved while keeping the $\Omega^2 T_1 T_2$ factor larger than one [46–48]. These rates translate into maximal currents up to $\sim$3 pA,

IV. LIMITING FACTORS

Condition (11) is an upper bound for the pumped current generated by the single-spin resonance. In addition to the conditions leading to this maximum current ($\beta\hbar\omega_0 \gg 1$ and $\Omega^2 T_1 T_2 \gg 1$), there are a few factors that could reduce the efficiency of this resonant pumping. For instance, any mechanism that leads to the local-spin relaxation without creation of a $2k_F$ electron-hole pair will decrease the dc current, for a fixed value of the Rabi coupling. There are several mechanisms that can relax the spin. First, suppose the material-dependent spin-momentum locking axis z is not perfectly aligned with the local-spin quantization axis z'. In that case, exchange interactions will relax the local spin in the forward-scattering channel that entails no current. For instance, let us consider a local spin governed by the Hamiltonian $H = DS_z^2 + E(S_x^2 - S_y^2)$, integer spin, and $D < 0$. It can be seen [49] that transitions between the ground state doublet are generated by the S_z operator. Therefore, the Kondo exchange with the QSHI edge states is via the $S_z\sigma_z(0)$ operator, which can only produce forward-scattering spin-conserving transitions. In general, the quantization direction of the edge state will depend on momentum and it can point in directions different than the normal [15,50].

Second, spin-phonon coupling can represent an important source of spin relaxation in paramagnetic crystals [51], including both one-phonon direct relaxation processes, with a typical relaxation rate proportional to T when $\hbar\omega_0 \gg k_BT$, and two-phonon Raman and Orbach processes [38]. Third, the spin relaxation of the current-carrying electron-hole pair, induced by nuclear spins [25], by other magnetic impurities, and with other thermally excited electron-hole pairs in bulk states would reduce the resulting current. Whereas hyperfine interactions are typically weak, the case where more than one magnetic center is present at the edge deserves future attention. One the one hand, having N resonating spins enhances the spin pumping. On the other, electron-hole pairs generated by a given spin can be reabsorbed by the others.

Another limiting factor would be the formation of a Kondo singlet, that would quench the magnetic moment of the local spin, reducing its effective coupling to the external driving force.

V. DISCUSSION AND CONCLUSIONS

Here, we have proposed a mechanism that permits one to envision an electrically detected single-spin resonance of a magnetic impurity coupled to the edge state of a QSHI. We have demonstrated that the spin-momentum locking at the edge states leads to a spontaneous net current when an electron-hole pair is created by the isotropic exchange coupling with a local magnetic moment. If an external ac driving is capable of inducing a departure of the stationary occupations from their equilibrium counterpart, this in turns generates a measurable dc current. We have shown that, with state-of-the-art instrumentation, the upper limit for the generated dc current is given by the Rabi coupling Ω of the local spin to the ac driving fields and presented a thorough discussion of the limiting factors that could reduce this maximum induced current. We estimate that state-of-the-art ESR instrumentation can provide values of Ω that will induce currents within the current sensitivity, with dc currents well above a few tens of fA. Finally, we have proposed several physical realizations, such as magnetic adatoms or molecules attached at the border of a QSHI and probed by a ESR-STM.

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