Exchange and collective behavior of magnetic impurities in a disordered helical metal

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We study the exchange interaction and the subsequent collective behavior of magnetic impurities embedded in a disordered two-dimensional (2D) helical metal. The exchange coupling follows a statistical distribution whose moments are calculated to the lowest order in \((p_F \ell)^{-1}\), where \(p_F\) is the Fermi momentum of itinerant electrons and \(\ell\) is the mean free path. We find that i) the first moment of the distribution decays exponentially, and ii) the variance of the interaction is long-range, however, it becomes independent of the orientation of the localized magnetic moments due to the locking between spin and momentum of the electrons that mediate the interaction. As consequence, long-range magnetic order tends to be suppressed, and a spin glass phase is stabilized at finite temperatures. The formalism is applied to the surface states of a three-dimensional (3D) topological insulator. The lack of a net magnetic moment in the glassy phase and the full randomization of spin polarization at distances larger than \(\ell\) excludes a spectral gap for surface states. Hence, non-magnetic disorder may explain the dispersion in results for photoemission experiments in magnetically-doped topological insulators.

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A topological insulator is a system that supports metallic edge/surface states within the bulk gap, whose existence and integrity are protected by time-reversal symmetry [1]. In the case of a 3D topological insulator, the surface states disperse as Dirac quasi-particles; the minimal Hamiltonian consists in a Bychkov-Rashba spin-orbit coupling of the form \((\hbar = 1)\)

\[
\mathcal{H}_{BR} = v_F (\sigma \times p)_z,
\]

where \(v_F\) is the Fermi velocity of surface electrons and \(\sigma = (\sigma_x, \sigma_y)\) is a vector of Pauli matrices acting on electron spin. The inclusion of a mass term in the Hamiltonian, \(M\sigma_z\), reflects the breakdown of time-reversal symmetry. This may arise as a Zeeman term due to a weak magnetic field [2] or the proximity exchange coupling to a magnetic thin film [3]. In this scenario, the system becomes a 2D quantum Hall liquid, a perfect platform for several magneto-electric effects [7].

An interesting possibility is the deposition of magnetic adatoms, in such a way that, for high enough concentrations, \(n_m\), and at low enough temperatures, \(T\), a spontaneous ordering opens a gap in the spectrum, being the mass \(M\) proportional to the net magnetic moment in the out-of-plane direction \(\hat{z}\) [4–6]. Experimentally, this possibility does not seem to be completely clear. Angle-resolved photoemission (ARPES) and scanning tunnel spectroscopy (STS), together with X-ray magnetic circular dichroism (XMCD) and magneto-transport experiments generate results that are in agreement with a gap opening in some cases [8] and gapless spectrum in others [9] for similar experimental conditions. In the particular case of Fe adatoms deposited directly on the surface of Bi\(_2\)Se\(_3\), for example, a gap of the order of 100 meV at \(T \sim 15\) K was reported in an ARPES experiment [10], whereas later on, another ARPES experiment in similar conditions of Fe concentration and even lower temperatures reported no gap opening [11], further confirmed by STS and XMCD experiments [12]. This dispersion in experimental results motivates the present study about the impact of disorder in the collective behavior of magnetic adatoms placed on the surface of a 3D topological insulator.

We consider first the problem of the exchange interaction between localized spins associated to magnetic impurities embedded in a weakly disordered helical metal. The Hamiltonian reads in general

\[
\mathcal{H} = \mathcal{H}_{it} + \mathcal{H}_{loc},
\]

where the first terms refers only to the itinerant electrons and the second term describes the coupling with localized
spins,
\[ \mathcal{H}_{\text{loc}} = J_z \sum_{i \in m} s_z (\mathbf{R}_i) \cdot S_i^z + J_\parallel \sum_{i \in m} \mathbf{s} (\mathbf{R}_i) \cdot \mathbf{S}_i, \]
where the sum runs over the magnetic impurities. Here \( S_i^z = \langle S_i^z, S_i^z \rangle \) are the spin operators of the impurity and \( s_z (\mathbf{R}_i) = (s_x (\mathbf{R}_i), s_y (\mathbf{R}_i)) \) are the spin density operators of the metal evaluated at the magnetic impurity sites,
\[ s_\alpha (\mathbf{R}_i) = \sum_{\mathbf{r}} \delta^{(2)} (\mathbf{R}_i - \mathbf{r}) \sigma_\alpha (\mathbf{r}). \]
The exchange is taken to be anisotropic due to the 2D nature and strong spin-orbit coupling of the system. In-plane isotropy is assumed, \( J_x = J_y \equiv J_\parallel \).

The single-particle Hamiltonian for itinerant electrons can be written as \( \mathcal{H}_{\text{it}} = \mathcal{H}_{\text{BR}} + V (\mathbf{r}) \), where the first term corresponds to Eq. (1), and the second describes the effect of disorder, \( V (\mathbf{r}) = \sum_{i \in \text{nn}} \mathcal{V}_{\text{imp}} (\mathbf{r} - \mathbf{R}_i) \). Here \( \mathcal{V}_{\text{imp}} (\mathbf{r}) \) is the potential created by a non-magnetic impurity or any source of disorder that preserves time-reversal symmetry. Since we do not posses a detailed expression for \( \mathcal{V}_{\text{imp}} (\mathbf{r}) \) and the distribution of impurities change from sample to sample, we employ a statistical description in terms of different disorder realizations forming an ensemble of macroscopically identical replicas of the system. Assuming that the typical decay length of the impurity potential is smaller than the mean separation between scattering centers, we consider a Gaussian distribution of disorder configurations, determined by the mean free path \( l \), or equivalently, the scattering time \( \tau = l / v_F \), and characterized by correlators of the form
\[ \langle V (\mathbf{r}_1) V (\mathbf{r}_2) \rangle_{\text{dis}} = \frac{1}{2 \pi \gamma \tau} \delta^{(2)} (\mathbf{r}_1 - \mathbf{r}_2), \]

\[ \langle \mathcal{H}_{\text{RKKY}} \rangle_{\text{dis}} = - \frac{p_F e^{-R / l}}{4 \pi^2 v_F R^2} \sum_{i,j \in m} \left[ J_z^2 \sin (2R \rho_F) S_i^z S_j^z + J_\parallel^2 \sin (2R \rho_F) \left( \mathbf{R} \cdot \mathbf{S}_i \right) \cdot \left( \mathbf{R} \cdot \mathbf{S}_j \right) - J_z J_\parallel \cos (2R \rho_F) (S_i \times S_j)_{\times \mathbf{R}} \right]. \]

\[ J_z \] controls the exchange between the out-of-plane spin components, and \( J_\parallel \) between the projections along the direction linking the two magnetic impurities. The last term is a Dzyaloshinskii-Moriya coupling. In the diffusive regime, the three components decay exponentially as in a conventional 2D electron gas [13]. This must not be interpreted as an exponential suppression of the RKKY interaction, but as the result of the randomization of its characteristic oscillatory tail [15]. Consequently, higher moments of the distribution must be studied.

\[ \langle \chi_{\alpha, \beta} \chi_{\alpha', \beta'} (\mathbf{R} = \mathbf{R}_j - \mathbf{R}_i) \rangle \equiv \langle \chi_{\alpha, \beta} (\mathbf{R}_i, \mathbf{R}_j) \chi_{\alpha', \beta'} (\mathbf{R}_i, \mathbf{R}_j) \rangle_{\text{dis}}, \]
which is analogous to the calculation of the universal conductance fluctuations in a disordered conductor [16]. To the lowest order in \((p_F \ell)^{-1}\), the second moment is given by both diffuson and cooperon ladder contributions, see Fig. 2. In the absence of time-reversal symmetry breaking perturbations
of the localized spins, the interaction becomes independent of the orientation is still a long-range interaction, however, the variance of the Fermi level or the distance between magnetic impu-

rations induced by the presence of localized spins at the helicity of carriers, disorder fully randomizes the po-

trusion to 

triplet modes yields to an exponentially vanishing con-

sequent by the Hamiltonian in Eq. (1). Therefore, the se-
quence of the locking between spin and momentum D

1/2 \sigma_\mu \sigma_\nu | \sigma_\nu \rangle | \sigma_\nu \rangle \right). 

In the diffusive regime, |\omega| \tau \ll 1, with |\omega| \ll 1 and 

sign (|\omega_1|) \neq \text{sign} (|\omega_2|) \text{ (otherwise, the diffusion ladder is zero), the diffuson satisfy the equation}

where D = \frac{1}{2} v_F^2 \tau is the diffusion constant. The triplet modes (\mu, \nu = x, y, z) are coupled by precession terms and only the singlet mode D_{00} remains gapless as consequence of the locking between spin and momentum implied by the Hamiltonian in Eq. [1]. Therefore, the triplet modes yields to an exponentially vanishing contribution to \chi_{\alpha \beta} \chi_{\alpha' \beta'} (R), similarly to what happens in a 2D electron gas with strong spin-orbit scattering [17]. Only the singlet mode contribution survives, leading to

As it is evident from this result, the RKKY coupling is still a long-range interaction, however, the variance of the interaction becomes independent of the orientation of the localized spins, \left( \langle H_{\text{RKKY}} \rangle \right)_{\text{dis}} \sim S_i^x S_j^x. Due to the helicity of carriers, disorder fully randomizes the polarization induced by the presence of localized spins at distances larger than the mean free path. These results have dramatic consequences for the collective behavior of magnetic adatoms on the surface of a 3D topological insulator. For instance, it was suggested in Ref. [5] that different spin models could be implemented by adjusting the Fermi level or the distance between magnetic impurities given that the oscillatory tails of the Ising-like and Dzyaloshinskii-Moriya couplings are out-of-phase [18]. In particular, when only the Dzyaloshinskii-Moriya term is present a non-coplanar spin configuration is expected with a non-zero net ferromagnetic moment. However, when disorder is considered, these phases are expected to be suppressed due to the exponential decay of the first moment of the distribution, since, according to mean field arguments, it is this quantity what determines the critical temperature below which long-range magnetic order is possible. Moreover, what it is expected from Eq. [4] is that localized spins freeze to a non-zero value at low temperatures, but with no spatial correlation between them and zero net magnetization. In such a glassy phase, a global spectral gap is excluded.

We focus now on the collective behavior of magnetic adatoms assuming that their spatial distribution is completely random. The intrinsic case (p_F = 0, \ell \rightarrow \infty) was analyzed in Ref. [2]. Randomness in the position of the magnetic impurities in combination with the form of the interaction in that case makes the in-plane interactions frustrated. Then, different behaviors are expected as a function of \delta \equiv J_\parallel / J_z. For \delta \ll 1, a Ising-like ferromagnetic phase is expected at low temperatures, with magnetization along the out-of-plane axis. Hence, a gap in the spectrum of surface states is expected. In the opposite limit, \delta \gg 1, the frustrated in-plane interactions dominate, giving rise to a spin glass separated from the
Ising ferromagnet by a quantum critical point estimated to be located at $\delta_c \approx 1.3$ [6]. This spin glass phase is expected not to survive at finite temperatures due to the reduced dimensionality of the system. However, this last statement changes if disorder is taken into account. Fluctuations in the exchange interaction due to disorder tend to destroy long-range order, and the system freezes to a glass for sufficiently low but finite temperature.

In order to see this, we analyze in detail the situation when $\delta \ll 1$. The spin Hamiltonian reads

$$H_{\text{RKKY}} = -\sum_{i,j} J_{ij} S_i^z S_j^z,$$

where the couplings $J_{ij}$ follow a certain statistical distribution. The first and second moments can be written in general as

$$\langle J_{ij} \rangle = J_z^2 \sum_{\{R\}} P(\{R\}) \chi_{zz}(R),$$

$$\langle (J_{ij})^2 \rangle = J_z^4 \sum_{\{R\}} P(\{R\}) \chi_{zz}^2(R).$$

Here $P(\{R\})$ describes the statistical distribution of magnetic adatoms over the surface of the topological insulator. We assume for simplicity that they are uniformly distributed, so we can approximate the above equations by [18]

$$\langle J_{ij} \rangle \approx \frac{J_z^2}{A} \int d^2 R \chi_{zz}(R) = \frac{n_m J_z^2 p_F}{4 v_F N} + O\left(\frac{1}{p_F \ell}\right),$$

and similarly for the second moment,

$$\langle (J_{ij})^2 \rangle \approx \frac{J_z^4}{A} \int d^2 R \chi_{zz}^2(R) = \frac{n_m J_z^2 p_F^2}{12 \pi^3 v_F^2 N} \int_{\ell}^{\infty} dR \frac{1}{R^3} = \frac{n_m J_z^2 p_F^2}{24 \pi^3 v_F^2 \ell^2 N}.$$

Here $A$ is the area of the system and $N = n_m A$, the number of localized spins. Note that the integral in Eq. (6) is infrared divergent, but the result of Eq. (4) is strictly valid at $R \gg \ell$; therefore, there is a natural cut-off for this integral determined by the mean free path, which allows to estimate $\langle (J_{ij})^2 \rangle$ to the leading order in $(p_F \ell)^{-1}$.

We assume that higher moments of the distribution do not affect the thermodynamic properties of the system, which can be studied within the Sherrington-Kirkpatrick (SK) model [19]. Following SK, the saddle-point equations reads

$$m = \int_{-\infty}^{\infty} d\alpha e^{-\frac{1}{2} \alpha^2} \tanh \left(\frac{T_{\text{FM}}}{T} m + \frac{T_{\text{SG}}}{T} q^{1/2} \alpha\right),$$

$$q = \int_{-\infty}^{\infty} d\alpha e^{-\frac{1}{2} \alpha^2} \tanh^2 \left(\frac{T_{\text{FM}}}{T} m + \frac{T_{\text{SG}}}{T} q^{1/2} \alpha\right),$$

where $m = \frac{1}{N} \langle S_i^z \rangle_T$ is the magnetization per spin and $q = \frac{1}{N} \langle S_i^z S_j^z \rangle_T$ is the Edwards-Anderson order parameter [20], and we have introduced

$$T_{\text{FM}} = \frac{n_m J_z^2 p_F}{4 v_F k_B},$$

$$T_{\text{SG}} = \frac{J_z^2 p_F}{2 \pi v_F k_B} \sqrt{\frac{n_m n_{\text{dis}}}{6 \pi}}.$$

Here we have taken $\ell \sim n_{\text{dis}}^{-1/2}$, where $n_{\text{dis}}$ represents the concentration of non-magnetic strong scatterers.

For low concentrations of scatterers the model predicts a second order phase transition to a ferromagnetic state at $T = T_{\text{FM}} \propto n_m$. In the opposite limit, fluctuations in the couplings destroy long-range order, and a spin glass phase characterized by $q \neq 0$ but $m = 0$ is stabilized below $T_{\text{SG}} \propto \sqrt{n_m n_{\text{dis}}}$. The phase diagram is shown in Fig. 3. Numerical solution of Eqs. (7)-(8) predicts a region of the parameter space where the system passes from a paramagnetic to a ferromagnetic to a reentrant spin glass phase (dashed line in Fig. 3) as the temperature is reduced. This is strange since at intermediate temperatures, where entropy plays a role, the system is ordered, but as $T \to 0$ one finds that a disordered phase is preferred. This is associated to the instability of the saddle-point solution of the SK model and the replica symmetry breaking. From Parisi’s solution [21] we predict a transition from the spin glass to a modified ferromagnetic phase (sometimes called mixed phase, where the replica symmetry is broken) at $n_{\text{dis}} = \frac{3}{T} n_m$. The boundary between this modified ferromagnetic phase and a conventional Ising ferromagnet can be estimated from the Almeida-Thouless line [22]

$$\frac{T^2}{T_{\text{SG}}} = 1 - 2 q + \int_{\ell}^{\infty} d\alpha e^{-\frac{1}{2} \alpha^2} \tanh^4 \left(\frac{T_{\text{FM}}}{T} m + \frac{T_{\text{SG}}}{T} q^{1/2} \alpha\right).$$

Note that the differences between these two ferromagnetic phases are subtle since the Almeida-Thouless transition is third order in the Ehrenfest sense.
In summary, we have computed the first moments of the distribution for the RKKY interaction in a weakly disorder helical metal and applied the results to infer some trends in the collective behavior of magnetic adatoms on a 3D topological insulator. For $J_z \gg J_H$, our analysis reveals that the ensemble of localized spins freezes to a spin glass phase above the disorder threshold $n_{dis} = 3\pi^2 n_m$. In-plane exchange interactions may help to stabilize this phase for weaker disorder due to frustration. In the spin glass phase, a global spectral gap in the surface states is precluded due to the full randomization of the spin polarization and the absence of a net magnetic moment. This means that an ARPES experiment would report no gap opening in this scenario. Therefore, the presence of disorder may explain the dispersion in results for photoemission experiments in magnetically-doped topological insulators. Nevertheless, a local probe could test the consequences of the breakdown of time-reversal symmetry due to the freezing of the moments of the magnetic adatoms.

Finally, it is worth to mention that the effect of a time-reversal symmetry breaking in the electron gas that mediates the interaction is not taken into account in a self-consistent way. In a recent calculation, the authors of Ref. [23] showed that a gap opening does not change the general structure of the RKKY interaction. Similarly, effects of dissipation can be neglected at first glance based on the argument that the RKKY interaction depends on electronic states deep inside the Fermi sea, not only at the Fermi surface, whereas dissipative phenomena as the Kondo effect is purely a Fermi-surface effect [24]. Therefore, a rearrangement of the low energy part of the spectrum does not dramatically affect the RKKY interaction. Nevertheless, interesting novel physics beyond the scope of this work may appear at low temperatures, particularly in the disordered phase.

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