Ordering of Magnetic Impurities and Tunable Electronic Properties of Topological Insulators

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We study collective behavior of magnetic adatoms randomly distributed on the surface of a topological insulator. Interactions of an ensemble of adatoms are frustrated, as the RKKY-type interactions of two adatom spins depend on the directions of spins relative to the vector connecting them. We show that at low temperatures the frustrated RKKY interactions give rise to two phases: an ordered ferromagnetic phase with spins pointing perpendicular to the surface, and a disordered spin-glass-like phase. The two phases are separated by a quantum phase transition driven by the magnetic exchange anisotropy. The ordered phase breaks time-reversal symmetry spontaneously, driving the surface states into a gapped state, which exhibits an anomalous quantum Hall effect and provides a realization of the parity anomaly. We find that the magnetic ordering is suppressed by potential scattering.

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Topological insulators in three dimensions are a class of time-reversal-invariant materials characterized by gapless surface states with Dirac-like dispersion (for a review, see Refs. [1,2] and references therein). These topologically protected states originate from the bulk band inversion induced by strong spin-orbit interactions. The effective low-energy Hamiltonian has the form of the Rashba spin-orbit coupling

$$H_0 = v\vec{n} \cdot \vec{p} \times \vec{\sigma}, \tag{1}$$

where $\vec{\sigma}$ is the electron spin, v is the Fermi velocity, and $\vec{n}$ is the normal vector to the surface, chosen to be along the z direction. The surface states lead to interesting phenomena, including magnetoelectric effect [1–4], large Kerr, and universal Faraday effects [4,5]. In addition, the locking of spin and momentum on the surface [6], evident from Eq. (1), gives rise to electric charging of magnetic textures [7], and opens up new opportunities for spintronics applications [8–10].

The aforementioned physical effects and device applications rely on the ability to induce perturbations that break time-reversal symmetry and open up a gap in the surface states spectrum. A relevant perturbation has the form of a mass term for Dirac electrons,

$$H_1 = m\sigma_z. \tag{2}$$

In principle, such a perturbation can be induced by depositing magnetic films. However, such a method has a significant disadvantage of being irreversible, and likely inducing too strong magnetic fields that can completely destroy surface states. Thus, alternative methods are needed which would allow for a controllable and reversible manipulation of the topological surfaces states.

Here we explore the possibility of gap opening by controlled adsorption of magnetic adatoms. We study the collective behavior of adatom spins, determined by the RKKY-type interactions mediated by the surface states. We argue that depending on the exchange anisotropy of a single impurity spin, the ground state of many spins is either a spin glass (SG), or a ferromagnetically ordered state with spins pointing perpendicular to the surface. The two phases are separated by a quantum phase transition. The phase diagram is summarized in Fig. 1.

In the ferromagnetic phase the average exchange field of impurities induces a mass of Dirac electrons, Eq. (2),

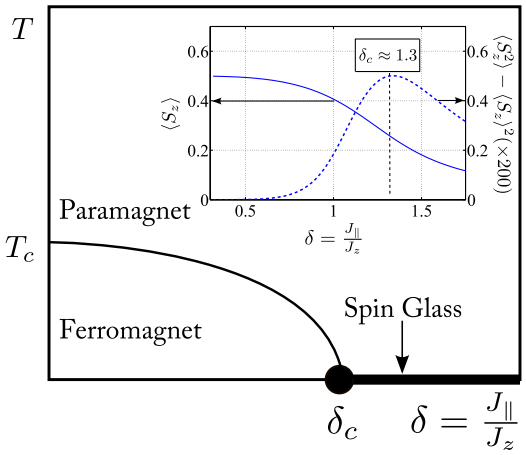

FIG. 1 (color online). Phase diagram of magnetic adatoms. Inset: Magnetization of spins on the topological surface, which are interacting via RKKY interactions, as a function of the exchange anisotropy $\delta = J_\parallel/J_z$. We find the position of the quantum critical point, $\delta_c \approx 1.3$, from the condition that the magnetization is decreased by 50%. This is supported by the fluctuations of the magnetization, which exhibit a maximum at the conjectured transition point, $\delta_c \approx 1.3$. Cluster of 9 spins was considered, and averaging was performed over 150 disorder realizations.

opening up a band gap in the spectrum of the surface states. The mass depends on the concentration of adatoms and their type, and therefore is tunable. We find that this state is favored in a large region of the phase diagram (see below), and persists up to several tens of Kelvin. In contrast, in the SG phase the average value of spin is zero, and on average the time-reversal symmetry is not broken; thus the gap is absent. There is, however, an insignificant disorder broadening which induces finite density of states at the Dirac point [11]. Our work shows that deposition of magnetic impurities provides a route to controllably change the spectral and transport properties of surface electrons.

We start with the analysis of the RKKY interactions, employing the T-matrix description. The interaction energy of two impurity spins located at $\vec{R}_{1,2}$, respectively, is given by [12]

$$\Omega_{12} = -T\sum_{\varepsilon} \mathrm{Tr}\ln[1 - \hat{t}_2(\varepsilon)G_0(\varepsilon,\vec{R})\hat{t}_1(\varepsilon)G_0(\varepsilon,-\vec{R})]. \quad (3)$$

In this expression $\vec{R} = \vec{R}_2 - \vec{R}_1$, ε are fermionic Matsubara frequencies, $\hat{t}_{1,2}(\varepsilon)$ are the low-energy T matrices of the impurities, and $G_0(\varepsilon, 0)$ is the unperturbed Matsubara Green's function of the surface electrons. The trace here is taken over the spin space. Note that there are also single-spin terms in the thermodynamic potential, which correspond to an easy or hard axis anisotropy for spins larger than $1/2$. However, such anisotropy can be shown to be determined by high energies of the order of bandwidth; thus, it cannot be reliably calculated in the present approach, designed to capture low-energy physics.

We consider a model in which the spin of an individual impurity situated at $\vec{r} = \vec{r}_0$ interacts with the surface electrons via anisotropic exchange Hamiltonian [13],

$$H_{\mathrm{ex}} = J_z S_z \sigma_z \delta(\vec{r} - \vec{r}_0) + J_\parallel (S_x\sigma_x + S_y\sigma_y)\delta(\vec{r} - \vec{r}_0), \quad (4)$$

where z is the direction perpendicular to the surface and S_i is the impurity spin operator. The exchange anisotropy, $J_z \neq J_\parallel$, stems from the combination of spin-orbit interactions and breaking of rotational symmetry on the surface. Electrons are described by the Hamiltonian (1), with bandwidth W (for Bi_2Se_3 $W \approx 0.3$ eV [1]), and a short range cutoff $a = v/W$.

The T matrix $\hat{t}(\varepsilon)$ is found using the Lippmann-Schwinger equation:

$$\hat{t}(\varepsilon) = \vec{V}_i\vec{\sigma} + \vec{V}_i\vec{\sigma}G_0(\varepsilon, 0)\hat{t}(\varepsilon), \quad (5)$$

$$\vec{V}_i = (J_\parallel S_i^x, J_\parallel S_i^y, J_z S_i^z). \quad (6)$$

The expression for the unperturbed Matsubara Green's function of the surface electrons, $G_0(\varepsilon, \vec{r})$, for the Hamiltonian (1) reads

$$G_0(\varepsilon,\vec{r}) = -\frac{i\varepsilon}{2\pi v^2}K_0\left(\frac{|\varepsilon|r}{v}\right) - \frac{i|\varepsilon|}{2\pi v^2}K_1\left(\frac{|\varepsilon|r}{v}\right)(\hat{r}\times\vec{\sigma})_z,$$

where $K_{0,1}(x)$ are modified Bessel functions, and $\hat{r}$ is the unit vector in the direction of $\vec{r}$. For $\vec{r} \to 0$ the above equation takes the following form:

$$G_0(\varepsilon, \vec{r} \to 0) \equiv g(\varepsilon)\sigma^0, \qquad g(\varepsilon) = -\frac{i\varepsilon}{2\pi v^2}\ln\frac{W}{|\varepsilon|}. \quad (7)$$

Using the above relations, we get the form of the T matrix,

$$\hat{t} = t^0\sigma^0 + \vec{t}\,\vec{\sigma}, \qquad (t^0, \vec{t}) = \frac{1}{1 - g^2\vec{V}^2}(g\vec{V}^2, \vec{V}). \quad (8)$$

The above form of the T matrix exhibits poles at energies found from the equation $1 - g^2\vec{V}^2 = 0$. In the limit of a very large bare potential, $|\vec{V}| \gg Wa^2$, the resonances are positioned at low energies, $|\varepsilon| \approx 2\pi v^2/|\vec{V}|\ln\frac{W|\vec{V}|}{2\pi v^2}$ [14], similar to the case of graphene (see, e.g., Ref. [12]). It is easy to see that the impurity spin dependent part of the T matrix vanishes in both limits of $|\vec{V}| \to 0$ and $|\vec{V}| \to \infty$. Thus the RKKY interaction reaches a maximum at $r \sim |\vec{V}|/Wa \gg a$ for strong exchange, $\max(J_\parallel, J_z) \gg 2\pi v^2/W$. This effect is missing in the perturbative treatment of RKKY.

Lowest order perturbation theory [15] can be used for moderate exchange values, $\max(J_\parallel, J_z) \lesssim 2\pi v^2/W$. For simplicity, we focus on this case below; RKKY interactions for the case of strong exchange will be discussed elsewhere [11]. At moderate exchange, the RKKY interactions at not too small adatom separation $r \gg a$ can be obtained from the general expression (3) by expanding the logarithm to the lowest order. This gives, up to a small corrections of the order a/r,

$$U_{12}(\vec{r}) = -J_z^2\frac{C}{r^3}S_1^zS_2^z - J_\parallel^2\frac{C}{r^3}(\vec{S}_1\cdot\hat{r})(\vec{S}_2\cdot\hat{r}) + J_\parallel^2\frac{D}{r^3}S_1^\perp S_2^\perp, \quad (9)$$

where $S_{1(2)}^\perp = \vec{S}_{1(2)}\cdot(\hat{r}\times\vec{n})$, $C = \frac{1}{16\pi^3 v}\int d\xi\xi^2(K_0^2(\xi) + K_1^2(\xi)) = \frac{1}{64\pi v}$, $D = \frac{1}{16\pi^3 v}\int d\xi\xi^2(K_1^2(\xi) - K_0^2(\xi)) = \frac{1}{128\pi v} = C/2$. Therefore, the interactions between two impurities have a strongly anisotropic form which stems from the spin-momentum entanglement on the surface. Similar anisotropy occurs in other materials with strong spin-orbit interactions [16].

We analyze the collective behavior of adatoms under the realistic assumption that their spatial distribution is completely random, and the exchange coupling to the surface electrons is not too strong. The positional randomness combined with the form (9) of the RKKY interactions makes the in-plane interactions frustrated: the exchange is antiferromagnetic between components of spins perpendicular to $\vec{r}$, and ferromagnetic for the components of the spins parallel to $\vec{r}$. Instead, the ferromagnetic interactions between z components of spins can be optimized simultaneously. We conclude that for $\delta \equiv J_\parallel/J_z \leq 1$ the ground state of any system of adatoms is a ferromagnet with magnetization along the z axis. In the opposite limit,

$\delta \gg 1$, the frustrated xy interactions dominate, giving rise to the ground state in which spins are frozen in the xy plane, with zero average polarization in the z direction. We expect this phase to be a SG (see below), which is separated from an Ising-type ferromagnet by a quantum critical point. We also expect the ferromagnetic ground state to survive some degree of exchange anisotropy, such that the critical point corresponds to $\delta_c > 1$. As the ferromagnetic ordering breaks discrete Z_2 symmetry, it occurs via a finite-temperature second order phase transition of the Ising type. The SG phase should not exist at finite temperature due to the reduced dimensionality.

The value of δ_c depends on the magnitude of the impurity spin. We choose the most unfavorable for ferromagnetic ordering case of $S = 1/2$, having the largest quantum fluctuations that disordered phases. To estimate the value of δ_c, we have performed numerical simulations on small spin systems. We exactly diagonalized Hamiltonians of randomly distributed spin-1/2 clusters with pairwise interactions given by Eq. (9). The resulting magnetization, averaged over 150 disorder realizations, is illustrated in the inset in Fig. 1. At $\delta \approx 1.3$ magnetization decreases by 50%; we take this point to be the finite-size approximation to the point of quantum phase transition [17]. For $S > 1/2$, we expect $\delta_c > 1.3$ [11].

What is the nature of the disordered phase realized at zero T and $\delta \gtrsim \delta_c$? We expect that at $\delta \to \infty$ (the only interactions are in plane) the frustrated random interaction should lead to a SG phase [18], similarly to the case of 2D bimodal XY model [17], and 3D dipolar dilute magnets and magnetic semiconductors [16,19]. We conjecture that weak ferromagnetic interaction of z spin components does not destabilize the SG phase, and it extends all the way to $\delta = \delta_c$. This hypothesis is supported by the qualitative similarity of our system to the Sherrington-Kirkpatrick model [20], which describes the competition between the nonfrustrated ferromagnetic exchange and the frustrated sign-changing interactions. In the model [20] the ferromagnetic phase is destroyed once the frustrated part of the interactions becomes strong enough. Our model almost certainly exhibits a similar behavior. Experimentally, the zero-temperature SG phase manifests itself in the divergence of the nonlinear magnetic susceptibility as $T \to 0$ [18].

Now we estimate the ferromagnetic ordering temperature. At $\delta \ll 1$, the ordering transition is that of Ising spins randomly distributed in the plane and interacting via $1/r^3$ interactions. The ordering temperature can be estimated as the typical exchange interaction between two neighboring spins. The result for the transition temperature can be read off Eq. (9) by setting $S^{x,y}_{1,2} = 0$ and $r = \sqrt{n_m}$, where n_m is the concentration of impurities. This gives an estimate

$$T_c(n_m) = \alpha \frac{J_z^2}{v} n_m^{3/2}, \tag{10}$$

where α is a numerical coefficient. A Monte Carlo simulation of this model yields $\alpha \approx 0.016$ [21]. We expect this estimate to hold for $\delta \lesssim 1$. For $n_m a^2 \sim 1$, $J_z \sim W a^2$, and $W = 0.3$ eV (band gap of Bi_2Se_3), we obtain $T_c \sim 30$ K, which is in the experimentally observable range.

We now discuss the spectral and transport properties of the Dirac fermions in the presence of adatoms. The ferromagnetic phase is characterized by the spontaneous breaking of the time-reversal symmetry, leading to a band gap. The most dramatic signature of the ferromagnetic ordering, detectable in transport, is that the behavior near the neutrality point changes from metallic to strongly insulating. In contrast, both paramagnetic and SG phases correspond to a gapless state of the Dirac fermions; the randomness of the potential created by adatoms leads to the smearing of the average DOS near the Dirac point [11], similar to the case of graphene [22]. The transport in this case remains metallic.

At the mean-field level, the mass induced by ordering of adatoms is given by

$$m = n_m J_z S. \tag{11}$$

We have calculated density of states (DOS) in the self-consistent T-matrix approximation [23], finding that for the physically relevant case of not too strong exchange, $J_z \lesssim 2\pi v^2/W$, the mean-field result is accurate. The DOS obtained using SCTMA is displayed in Fig. 2. Taking $J_z/a^2 \approx 300$ meV, $na^2 \approx 0.1$, and $S = 1/2$, we obtain the estimate of mass $m \approx 15$ meV, which puts it in the experimentally observable range.

An important consequence of a mass in the spectrum Dirac fermions is the quantized half-integer Hall conductivity [24], even in the absence of magnetic field. Such an anomalous quantum Hall effect (QHE) is a direct consequence of the parity anomaly [25], which so far evaded experimental observation. The magnetic ordering on the surface provides a way to experimentally observe anomalous QHE. This can be done, e.g., in a thin slab geometry, where spins polarize in the same direction on the opposite sides of the slab. Then the half-integer Hall conductivities

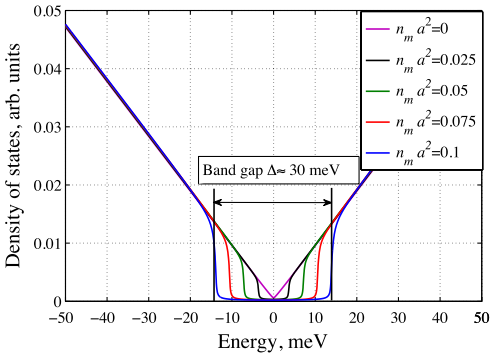

FIG. 2 (color online). Density of states of the topological surface states with a band gap induced by ferromagnetic ordering of adatoms. Density of states for different concentrations of magnetic adatoms. Complete polarization was assumed, and $J_z S = 0.5 W a^2$, $W = 0$, 3 eV.

of the two surface add, giving a quantized Hall conductivity e^2/h. We note that realization of the anomalous QHE state requires a small symmetry-breaking magnetic field, otherwise the system will break into domains of opposite polarization, giving rise to current-carrying domain-wall excitations.

Now we analyze the effect of potential impurities on the interactions and ordering of magnetic impurities. Experimentally, potential disorder is inevitably introduced by chemical doping. The disorder causes an exponential decay of the impurity-averaged RKKY interactions at large distances [11], similar to the case of disordered metals [26]. The relevant length scale $\ell_{\rm dis}$ is given by (twice) the Green's function decay length at $\varepsilon \to 0$. For strong nonmagnetic impurities, it reads [12,22]

$$\ell_{\rm dis} \sim \frac{1}{\sqrt{n_p}} \log \frac{W^2}{v^2 n_p}, \tag{12}$$

where n_p is the density of nonmagnetic impurities, and we neglected factors of order of unity. The sample-specific RKKY interactions are random, have the Dzyaloshinskii-Moriya form, and decay as a power law [26]. Therefore, at large densities of nonmagnetic impurities, $n_p \gg n_m$, the RKKY interaction is frustrated and magnetic ordering is suppressed, while the SG phase is favored at $T = 0$ at any $J_\parallel/J_z$. Thus, magnetic ordering can only be observed in clean samples, $n_p \ll n_m$.

Finally, we discuss the effect of warping and finite doping on the magnetic ordering of adatoms. The warping of the Fermi surface will not change the qualitative behavior; its main effect will be to break the symmetry of the spin interactions down to a group of discrete rotations and to slightly shift the phase boundaries. Similarly, in the doped system, the phase diagram remains unchanged for large enough adatom concentration, such that adatom separation is smaller than the Fermi wave length, $n_m^{-1/2} \ll \lambda_F$. At small adatom concentration $n_m^{-1/2} \gtrsim \lambda_F$, the Fermi surface effects make RKKY coupling sign changing. In this case the RKKY interaction acquires a Dzyaloshinskii-Moriya component [15], which frustrates interactions even more. Thus a finite doping stabilizes the SG phase.

Recently, we have become aware of an experimental work [27], where deposition of magnetic impurities was used as a tool to modify properties of Bi_2Se_3. Above certain density, ferromagnetic ordering, leading to a gap opening, was observed, in agreement with our analysis above. The suppression of ordering at low densities n_m observed experimentally is likely due to potential disorder, which stabilizes SG phase, as discussed above.

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