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Carrier mediated ferromagnetism on the surface of a topological insulator

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We study the effect of magnetic doping at the surface of a three dimensional topological insulator (TI) on emergence of ferromagnetic ordering at the TI-surface assuming the exchange coupling between the Dirac fermions and the dilute magnetic ions. We show that this coupling results in an uniaxial magnetic anisotropy with out-of-plane magnetization direction. It is found that the system under consideration is unstable with respect to a spontaneous uniform magnetization along the easy axis, which is accompanied by opening a gap in a spectrum of the Dirac surface states. In the framework of a mean-field approach, we study the possibility of ferromagnetic order on the magnetically doped surface of TI at different temperatures and positions of the chemical potential.

Recently, topological insulators (TIs) have attracted particular interest [1, 2] due to their important potential applications in quantum computing and spintronics [3, 4]. In these materials strong spin-orbital coupling causes the inversion of the conduction and valence bands that results in emergence of gapless surface or edge states of helical Dirac fermions without spin degeneracy. In the 1980s related seminal ideas were applied to a surface of crystal with strong spin-orbit interaction [5] and an interface between narrow gap semiconductors where the Dirac mass changes sign [6]. More recently the two-dimensional TI, known as a quantum spin Hall insulator, was observed in HgTe/CdTe quantum well structures [7]. Current researches are mostly focused on binary and ternary three-dimensional TIs such as Bi_2Te_3, Bi_2Se_3, and Sb_2Te_3 and thallium based compounds [8–14]. The Dirac-like surface states that occur in these materials were experimentally confirmed and thoroughly studied using scanning tunneling microscopy and angle-resolved photoemission spectroscopy [1, 15, 16].

The unique feature of the surface of TI is an exceptional sensitivity to perturbations breaking the time reversal invariance [1, 17]. Upon applying a perpendicular magnetic field, the notable properties of the surface of TI can dramatically change because an energy gap is induced in the helical state spectrum. One of the mechanisms to gap the topological surface states is the exchange coupling of these states with spin-polarized states of a ferromagnetic layer adjacent to the surface [17–20]. In particular, if the Fermi level is in the energy gap, a half integer quantized Hall effect can be induced on the surface of three-dimensoinal TI with the proximity to a magnetic insulator film [1, 21]. Another way is magnetic doping of the surface. However, the effect of interaction between magnetic impurities and the surface states of a TI on a collective behavior of localized impurity moments is a topic that is poorly explored. To the best of our knowledge, recent investigations are essentially restricted to a theoretical consideration of peculiarity of the indirect coupling between two localized magnetic moments on the surface of TI mediated by the helical metal, with an emphasis on Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction mechanism [20, 22–27].

At low temperature, the bulk TI doped with magnetic impurities can have a long-range ferromagnetic (FM) order [28–32]. At the surface of TI, dilute magnetic impurities can form a collective ordered state at temperature above the bulk Curie temperature provided that an exchange coupling between them mediated by the surface states is stronger than an exchange coupling in the bulk. Furthermore, moments of magnetic ions adsorbed on the surface of TI can also form a collective ordered state due to an exchange interaction via the helical states when the Fermi energy is close to the Dirac point. In any case, with or without a bulk FM order, the breaking of the time reversal symmetry at the surface of TI results in a gap opening at the Dirac point.

Recently Chen et al. [30] introduced magnetic and nonmagnetic dopants into a three-dimensional TI Bi_2Se_3 and performed angle-resolved photoemission spectroscopy measurements of electronic structure of the samples. They found a broken dispersion of the

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surface states at the Dirac point, so that the gap value appears to be from ~ 44 meV to ~ 50 meV increasing with the increase of the magnetic dopant concentration from $x = 0.12$ to $x = 0.16$ for iron doped samples $(Bi_{1-x}Fe_x)_2Se_{3.7}$ and ~ 7 meV for a manganese doped sample $(Bi_{0.99}Mn_{0.01})_2Se_3$. It was noticed that the Fermi level resided just inside both the surface and bulk gaps. In Ref. [33], by using Fourier transform scanning tunneling spectroscopy, the emergence of new scattering channels due to broken time reversal symmetry has been clearly observed in the Fe-doped Bi_2Te_3 single crystals with a nominal doping of 0.25%. The origin of these processes is attributed to local regions of FM correlated domains or enhanced surface ferromagnetism mediated by the surface states. In [34], strong modification of the surface states on the (111) surface of Bi_2Se_3 with chemisorbed Fe atoms has been studied. It was shown that an energy gap $\Delta \approx 100$ meV appears in the spectrum of helical electrons with increasing Fe coverage above $\gtrsim 20$ percent. The authors of [34] explained this result in terms of the FM ordering of the local magnetic moments of iron ions in the direction perpendicular to the surface. In contrast, angle-resolved photoemission spectroscopy studies of electronic structure and the scattering rates upon adsorption of impurities on the surface of Bi_2Se_3 have shown a remarkable insensitivity of the topological surface to both non-magnetic (Cs, Rb) and magnetic (Gd) ions in the low concentration regime; no evidence for an opening a gap at the Dirac point has been also found [35]. The recent work [36] also revealed that the topological surface states of Bi_2Se_3 and Bi_2Te_3 were stable against the deposition of Fe without opening a band gap.

The situation with properties of the magnetically doped surface of TI remains unclear and controversial – both theoretically and experimentally. The aim of the present paper is to elucidate possibility of the FM ordering in this system. For this purpose we study a collective behavior of localized impurity moments and spin polarization of the helical electrons within the framework of the mean-field scenario analogous to that for the carrier mediated FM ordering in dilute magnetic semiconductors [37]. It is accepted [20, 22–27] that two-dimensional electrons on the surface of TI mediate an exchange coupling of RKKY-type between the local moments i and j, $I(\boldsymbol{\rho}_{ij}) \propto \cos(2k_{\mathrm{F}}\rho_{ij})\rho_{ij}^{-2}$, which oscillates with the double Fermi wave vector k_{F} and decays with distance between impurities ρ_{ij}. Besides the coupling $I(\boldsymbol{\rho}_{ij})$ depends on the directions of the moments relative to the vector $\boldsymbol{\rho}_{ij}$. Below we show that the physics of magnetic ordering associated with chirality of the single-particle states on the surface of TI can be caught even without special analysis of mechanism and details of the pair exchange interaction $I(\boldsymbol{\rho}_{ij})$. On the one hand, we suppose that the helical electrons mediate an exchange interaction among magnetic impurities, which can leads to a FM state on the surface of TI. On the other hand, the helical states undergo a great modification due to an appearance of the long-range FM order. In this context we self-consistently calculate the energy gap in density of the surface states as well as the average magnetization of magnetic impurities.

To describe the helical metallic states on the surface of three-dimensional TI, we consider the Hamiltonian of noninteracting electrons in the following form:

$$H_0 = \int d\mathbf{r} \sum_{\alpha,\beta} \psi_\alpha^\dagger(\mathbf{r}) iv \left(\sigma^x_{\alpha\beta} \frac{\partial}{\partial y} - \sigma^y_{\alpha\beta} \frac{\partial}{\partial x} \right) \psi_\beta(\mathbf{r}), \quad (1)$$

where ψ_α is the electron field, v is the Fermi velocity, $\mathbf{r} = (x, y)$ is the two-dimensional vector, the z direction is perpendicular to the surface, the Pauli matrices $\sigma^l_{\alpha\beta}$ $(l = x, y, z)$ are introduced; we use $\hbar = 1$. The unique feature of (1) is that the electron spin and momentum are intimately locked, which originates from strong spin-orbit coupling. The surface states are implied to reside inside the bulk band gap.

We assume that the coupling between magnetic impurities carrying localized moment $\mathbf{S}_i$ and the helical electron spin takes the form

$$H_{ex} = a^2 \int d\mathbf{r} \sum_{\alpha,\beta} \psi_\alpha^\dagger(\mathbf{r}) \sum_i J(\boldsymbol{\sigma} \cdot \mathbf{S}_i)_{\alpha\beta} \delta(\mathbf{r} - \mathbf{R}_i) \psi_\beta(\mathbf{r}), \quad (2)$$

where a is the lattice constant of the surface. The impurities are randomly located on the surface at different positions $\mathbf{R}_i$. For simplicity, we treat the magnetic ions as classical local moments, which are merely dissimilar in orientations $\mathbf{S}_i$, i.e., $|\mathbf{S}_i| = S$. The matrix element of an exchange interaction J depends on the overlap between wave functions of the surface and impurity states. Prefactors such as the Bohr magneton or the Landé factor are implied to be included into J.

At finite density of magnetic ions on the surface, we expect them to order ferromagnetically below a certain critical temperature T_c that will be estimated below. We consider the system in the virtual crystal approximation and treat the interaction (2) within the mean-field approximation, which leads to the replacement $(\mathbf{S} \cdot \mathbf{s}) \to (\langle \mathbf{S} \rangle \cdot \mathbf{s}) + (\mathbf{S} \cdot \langle \mathbf{s} \rangle) - (\langle \mathbf{S} \rangle \cdot \langle \mathbf{s} \rangle)$, where $\mathbf{s} = \frac{1}{2} \psi_\alpha^\dagger \boldsymbol{\sigma}_{\alpha\beta} \psi_\beta$ is spin density of the topological surface state carriers. Then an effective mean-field Hamiltonian describing electrons can be obtained by replacing the

localized moments which are randomly distributed and oriented with a classical continuous magnetization $\langle \mathbf{S} \rangle$:

$$H_0^{\text{eff}} = H_0 + n_m a^2 \int d\mathbf{r} \sum_{\alpha,\beta} \psi_\alpha^\dagger(\mathbf{r}) \sum_i J(\boldsymbol{\sigma} \cdot \langle \mathbf{S} \rangle)_{\alpha\beta} \psi_\beta(\mathbf{r}), \tag{3}$$

where n_m is the averaged density of the moments on the surface. In the present study we consider the mean-field expectation value $\langle \mathbf{S} \rangle$ as spatially invariable. Then the Hamiltonian (3) is translational invariant and, in momentum space, its Green function reads

$$G_{\alpha\beta}(\mathbf{k}, \omega) = \frac{(\omega + \mu)\sigma^0_{\alpha\beta} + v(\sigma^y_{\alpha\beta} K_x - \sigma^x_{\alpha\beta} K_y) + \sigma^z_{\alpha\beta} \Delta}{(\omega + \mu)^2 - \varepsilon(\mathbf{K})^2}, \tag{4}$$

where σ^0 is the unity matrix, μ is the chemical potential. The in-plane components of magnetization $\langle \mathbf{S}_\parallel \rangle = \mathbf{e}_x \langle S_x \rangle + \mathbf{e}_y \langle S_y \rangle$ shifts the Dirac cone from a center of the Brillouin zone of the surface states: $k_x \to K_x = k_x + \Lambda_x / v$, $k_y \to K_y = k_y - \Lambda_y / v$, where $\mathbf{k} = (k_x, k_y)$ is a plane moment, $\Lambda_{x,y} = n_m a^2 |J| \langle S_{x,y} \rangle$. In contrast, out-of-plane uniform exchange field $\mathbf{e}_z \langle S_z \rangle$ opens in the spectrum the gap

$$\Delta = n_m a^2 |J| \langle S_z \rangle, \tag{5}$$

and splits the Dirac band into subbands above and below the gap with dispersions $\varepsilon(\mathbf{K}) = \pm\sqrt{\Delta^2 + v^2 K^2}$, $\mathbf{K} = (K_x, K_y)$, and the labels $\pm$ distinguish bands with positive and negative energies. The magnetic order associated with the nontrivial field $\mathbf{e}_z \langle S_z \rangle$ breaks the time reversal symmetry $\mathbf{k} \to -\mathbf{k}$ and $\boldsymbol{\sigma} \to -\boldsymbol{\sigma}$ of the Hamiltonian (1) and (2) and polarizes the helical electrons along the $\mathbf{z}$-axis. In turn this order of the magnetic dopants on the surface suggests FM interaction among the localized moments that is mediated by the helical surface state.

Having known the Green function (4), the grand canonical potential for the ground state of the helical electrons in the field $\langle \mathbf{S} \rangle$ can be written as

$$\Omega(\langle \mathbf{S} \rangle) = \int \frac{a^2 d\mathbf{k}}{(2\pi)^2} \varepsilon(\mathbf{K}) - \mu N, \tag{6}$$

where the integral is running over filled states $\varepsilon(\mathbf{K}) < \mu$, $N = \int a^2 d\mathbf{k} / (2\pi)^2$ is the number of particles. Under the condition $\Delta^2 + \Lambda^2 > \mu^2$, after an angle integration the potential $\Omega(\langle \mathbf{S} \rangle)$ is given by

$$\Omega(\langle \mathbf{S} \rangle) = \frac{-2}{\pi W^2} \int_0^W d\xi \xi \sqrt{\Delta^2 + (|\Lambda| + \xi)^2} \times \\ \times \mathbb{E}\left[\sqrt{\frac{4|\Lambda|\xi}{\Delta^2 + (|\Lambda| + \xi)^2}} \right] - \frac{\mu}{2}, \tag{7}$$

where $\mathbb{E}(\kappa)$ is the complete elliptic integral of the second kind with modulus κ, $W = \sqrt{2\pi} v / a$ is the band cut-off energy being symmetric on the particle and hole sides.

The expression (7) is obtained at a given orientation of the magnetization $\mathbf{S}$. Let us suppose that its magnitude is taken fixed, $|\mathbf{S}| = S$, and consider a small deviations, $\langle S_{x,y} \rangle^2 \ll S^2$, around a uniformly magnetized along the $\mathbf{z}$-axis state, $\mathbf{e}_z \langle S_z \rangle$, such that $\langle \mathbf{S} \rangle = \mathbf{e}_z \langle S_z \rangle + \langle \mathbf{S}_\parallel \rangle$. To leading order in the small parameter Δ / W one obtains from (7) the electron energy of the system with respect to that in the absence of the average magnetization:

$$\Omega(\langle \mathbf{S} \rangle) - \Omega_0 = -\frac{\Delta_0^2}{2W} \left(1 - \frac{\langle \mathbf{S}_\parallel \rangle^2}{2S^2} \right), \tag{8}$$

where $\Omega_0 = \Omega(\langle \mathbf{S} \rangle = 0)$, $\Delta_0 = n_m a^2 |J| S$ is the gap in the saturation state at $T = 0$. In Fig. 1 we represent the behavior of $\Omega(\langle \mathbf{S} \rangle) - \Omega_0$ for arbitrary angle θ between the vector $\langle \mathbf{S} \rangle$ and the $\mathbf{e}_z$-axis ($\Delta = \Delta_0 \sin\theta$, $\Lambda = \Delta_0 \cos\theta$) obtained numerically from Eq. (7).

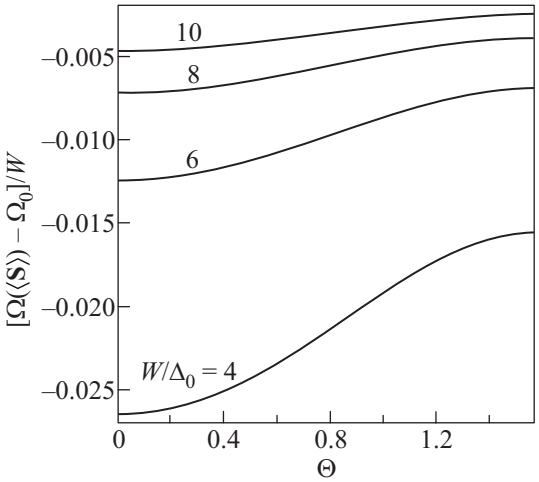

Fig. 1. The grand canonical potential for the ground state of the helical electrons versus the angle θ between the magnetization vector $\langle \mathbf{S} \rangle$ and the $\mathbf{e}_z$-axis for four values of Δ_0, $W/4$, $W/6$, $W/8$, and $W/10$

As is seen from Eq. (8), under the exchange field applied along the $\mathbf{e}_z$-direction, the helical electrons lower their energy by the value $\Delta^2/2W$ due to the opening of the gap. Figure 1 shows that the helical electrons are easily polarizable along axis perpendicular to the surface of TI, while a deviation of the collective magnetization from the easy axis $\mathbf{e}_z$ is energetically unfavourable. Such the situation is enhanced with the increasing magnetization. We thus conclude that random magnetic impurities will tend to align parallel to the normal to the TI surface. We expect that the FM ordering occurs via a

finite-temperature second order phase transition of the Ising type. Below we investigate this phase transition in more detail.

The interaction between carrier spins and local moments can be described as a self-consistent process in which carrier spins see an effective magnetic field produced by the local moments. Correspondingly, the exchange splitting introduced by the magnetic dopants can be estimated following (5). In turn the impurity magnetization $\langle \mathbf{S} \rangle = \mathbf{e}_z \langle S \rangle$ is influenced by the spin polarization of the helical electrons $\langle \mathbf{s} \rangle = \mathbf{e}_z \langle s \rangle$. The response of the classical localized moment to an effective field produced by the carrier spins $B_{\text{eff}} \sim J\langle s \rangle$ follows Boltzmann statistics:

$$\langle S \rangle = S\mathcal{L}\left(\frac{JS\langle s \rangle}{T}\right), \tag{9}$$

where $\mathcal{L}(w)$ is the Langevin function, T is temperature, we use $k_{\text{B}} = 1$.

The electron spin polarization is defined as

$$\langle s \rangle = T \sum_{n=-\infty}^{\infty} Tr \int \frac{a^2 d\mathbf{k}}{(2\pi)^2} \sigma^z_{\alpha\beta} G_{\beta\alpha}(\mathbf{k}, \omega_n), \tag{10}$$

where the trace is taken over spin indices. We introduce the Matsubara frequency $\omega_n = \pi T(2n+1)$ into the Green function (4) in common manner $\omega \to i\omega_n$. It is not difficult to show that the polarization is equal to

$$\langle s \rangle = \frac{\Delta T}{W^2} \ln\left[\frac{\cosh(\frac{\sqrt{W^2+\Delta^2}+\mu}{2T}) \cosh(\frac{\sqrt{W^2+\Delta^2}-\mu}{2T})}{\cosh(\frac{\Delta+\mu}{2T}) \cosh(\frac{\Delta-\mu}{2T})}\right]. \tag{11}$$

At zeroth temperature one has

$$\langle s \rangle = \frac{\Delta(\sqrt{W^2+\Delta^2} - \max\{|\mu|, \Delta\})}{W^2}. \tag{12}$$

The position of the chemical potential with respect to the Dirac point depends on the detailed electrostatics of the surface and, strictly speaking, will change with the gapping, i.e. $\mu = \mu(\Delta)$. In what follows the chemical potential is assumed to be unchanged, which could be achieved either when the Dirac point is close to edge of a bulk band with high density of states or via control of the surface charge with a gate. The equations (5), (9), and (11) constitute the self-consistent mean-field approximation for the description of the magnetic phase transition to a state with nontrivial order parameter, being the gap Δ. Indeed, inserting (11) into (9) and taking into account the relation (5), one obtains the equation for the gap Δ as a function of temperature T and the chemical potential μ. Implying that the FM-transition is second-order phase transition, in the vicinity of the critical temperature $T \lesssim T_c$, we are able to obtain an analytical expression for the order parameter as a function of temperature and chemical potential. We restrict ourselves to the weak interaction approximation that is natural for the given statement of the problem, $n_m a^2 (JS/W)^2 \ll 1$. Under the stipulation that $\Delta \ll T$ and simultaneously $T \ll W$, the self-consistent equation reduces to

$$\left[\frac{\Delta}{2T\cosh(\mu/2T)}\right]^2 = \ln\left[\frac{\cosh(W/T)+\cosh(\mu/T)}{1+\cosh(\mu/T)}\right] - \frac{W}{T_c^0}. \tag{13}$$

Above the curve $T_c(\mu)$, in the paramagnetic phase, $\Delta = 0$. The magnetic phase diagram $T_c(\mu)$ is determined by setting the right side of equation (13) to be equal to zero. The maximal temperature of the onset of magnetization $T_c = T_c^0$ is reached at $\mu = 0$. We can estimate this temperature as

$$T_c^0 = T_c(\mu = 0) = \frac{n_m a^2}{3} \frac{(JS)^2}{W}. \tag{14}$$

It is worthy to note that the ratio $T_c^0/\Delta_0 \simeq |J|S/W$ is rather a small value. Under the condition $\mu \ll T_c^0$, one obtains from (13) simple estimations for the Curie temperature, $T_c(\mu)$, and the gap, $\Delta(T, \mu)$, respectively:

$$T_c(\mu) = T_c^0 - \frac{\mu^2}{2W}, \tag{15}$$

$$\Delta^2 = 4W[T_c(\mu) - T]. \tag{16}$$

When the chemical potential is tuned relatively far from the Dirac point, $\mu \gg T_c^0$, the order parameter increases sharply (nevertheless, continuously) with temperature decreasing

$$\Delta^2 = W \exp\left[\frac{\mu}{T_c(\mu)}\right] \frac{T_c(\mu)}{T_c^0} [T_c(\mu) - T], \tag{17}$$

and the dependence $T_c(\mu)$ acquires the linear shape

$$T_c(\mu) = T_c^0\left(1 - \frac{\mu}{W}\right). \tag{18}$$

Figures 2 and 3 illustrate the typical behavior $\Delta(T, \mu)$ and $\langle s \rangle(T, \mu)$, respectively, produced with a numerical solution of the above equations (5), (9), and (11). From these figures it follows that there is a certain boundary $T_c(\mu, n_m)$ separating the gapped FM-state from gapless paramagnetic state. From this point of view one can roughly interpret the presence or absence of the gap in the spectrum of the surface states in the spectroscopy data [34–36] as a consequence of the conditions under which the measurements are carried out.

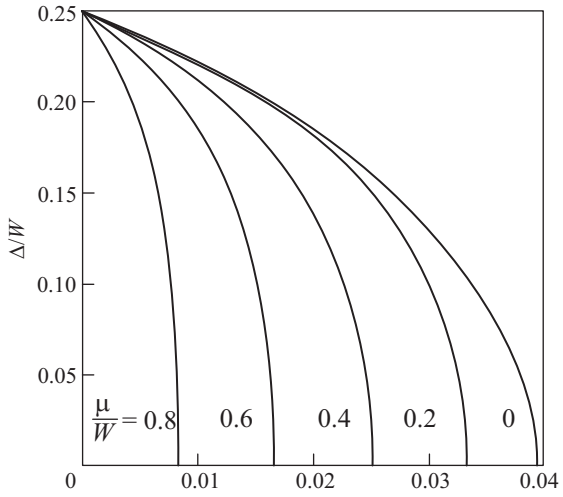

Fig. 2. The behavior of the FM order parameter Δ as a function of temperature T at different levels of the chemical potential μ ($\mu/W = 0$, 0.2, 0.4, 0.6, and 0.8) for given values of the magnetic ion concentration $a^2 n_m = 0.5$ and the exchange coupling $|J|S/W = 0.5$

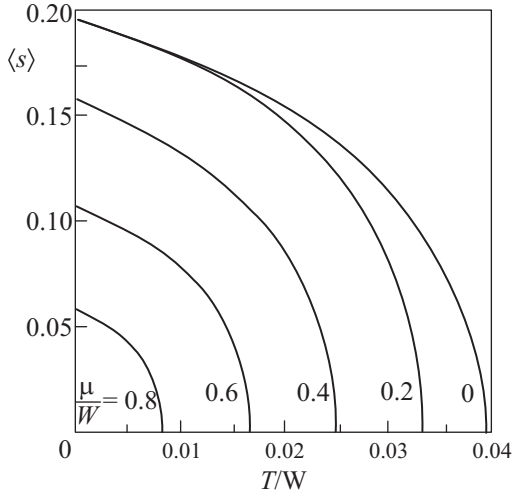

Fig. 3. The behavior of the helical electron spin polarization $\langle s \rangle$ as a function of temperature T at different levels of the chemical potential μ ($\mu/W = 0$, 0.2, 0.4, 0.6, and 0.8) for given values of the magnetic ion concentration $a^2 n_m = 0.5$ and the exchange coupling $|J|S/W = 0.5$

To summarize, in the present work we have studied the problem of FM ordering on the magnetically doped surface of TI at the mean-field level. A more comprehensive analysis of the modification of the local spin texture caused by a single local impurity on the surface of a three-dimensional TI and the pair exchange interaction mediated by the surface states is beyond our description. Generally speaking, it is necessary to carry out self-consistent calculation of the relevant exchange integral $I(\boldsymbol{\rho}_{ij})$ taking into account a gapping of the surface states in the FM-phase. Note, when the Fermi level is in the band gap, $|\mu| < \Delta$, solely the superexchange mechanism (due to electron excitations through the gap) gives rise to the indirect exchange coupling between impurity moments, which could lead to appearance of a magnetically ordered insulator on the surface of TI. In the case of $|\mu| > \Delta$, the scattering of carriers on impurity disorder on the surface causes a depression of both magnitude and length of the RKKY interaction via the helical states. As a result, the Curie temperature and average magnetization acquire negative augments in the parameter $(\Delta\tau)^{-1}$, where the inverse time of momentum relaxation can be estimated as $\tau^{-1} \simeq a^2 n_m |\mu| U^2/W^2$, U is an effective potential of electron-impurity interaction. If the scattering is such strong that $\Delta\tau \simeq 1$ (at the same time $|\mu|\tau > 1$), it may be expected that a gapless FM-state appears on the surface. It is possible a similar state was observed in works [35, 36].

Another important direction for future study of properties of the surface of three-dimensional TI is the effect of warping [15] on magnetic ordering upon variable filling of the surface states. An instability of the helical electrons caused by the formation of a spin-density wave state due to nesting of the Fermi surface for a certain position of chemical potential was predicted in [38]. In this case the competition between different magnetic configurations can remarkably change the phase diagram. It seems also desirable to compliment and extend the above analysis by presenting a contribution of magnetic thermal fluctuations to thermodynamics of the system.

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