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Surface magnetic ordering in topological insulators with bulk magnetic dopants

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We show that a three-dimensional topological insulator doped with magnetic impurities in the bulk can have a regime where the surface is magnetically ordered but the bulk is not. This is in contrast to conventional materials where bulk ordered phases are typically more robust than surface ordered phases. The difference originates from the topologically protected gapless surface states characteristic of topological insulators. We study the problem using a mean-field approach in two concrete models that give the same qualitative result, with some interesting differences. Our findings could help explain recent experimental results showing the emergence of a spectral gap in the surface state of Bi_2Se_3 doped with Mn or Fe atoms, but with no measurable bulk magnetism.

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I. INTRODUCTION

In recent years, the field of topological insulators (TIs) has attracted much attention and research in the condensed-matter community.[1–3] The advance has been rapid, on both the theoretical and experimental fronts; however, many challenges still remain. Among these perhaps the most important is gaining experimental control over the bulk and surface conduction in three-dimensional TIs. In this paper we address one aspect of this challenge that is associated with a magnetically induced excitation gap in the topologically protected surface states.

Topological insulators are bulk insulators in two or three dimensions with strong spin-orbit coupling (SOC) and protected gapless surface states.[4–9] The topological protection of the surface states arises due to time-reversal invariance (TRI). The surface states are conducting, have a characteristic linear (Dirac) dispersion, and exhibit spin-momentum locking. The most studied and most promising three-dimensional (3D) TI is the semiconducting thermoelectric Bi_2Se_3, with a relatively large band gap of $\sim$0.3 eV and a simple surface state consisting of a single Dirac cone.[10,11] The spectrum of Bi_2Se_3 and other TIs has been studied using angle-resolved photoelectron spectroscopy[12–14] (ARPES) and scanning tunneling microscopy,[15–17] showing that the surface states form an almost ideal Dirac cone, illustrated in Fig. 1(a), familiar from studies of graphene.[18]

Breaking TRI, for example by adding magnetic dopants (as we discuss), is expected to open a gap in the spectrum of the surface states. The resulting spectrum then resembles that of a "massive" Dirac fermion [Fig. 1(b)]. There is considerable interest in having a system with an odd number of massive Dirac fermions, since it is predicted to exhibit many interesting topological phenomena, including the half quantum Hall effect on the surface ($e^2/2h$ Hall conductance),[19] the image magnetic monopole (an electric charge adjacent to a TI results in the field of a magnetic monopole embedded in the TI),[20,21] and a Kerr-Faraday angle quantization in units of the fine structure constant.[22,23] A tunable gap would also allow the control of the surface transport and could, in addition, lead to unique practical applications associated with purely electric control of the surface magnetization.[24,25]

A signature of the massive Dirac fermion has been observed recently using ARPES in magnetically doped Bi_2Se_3,[26,27] although the interesting effects associated with it have yet to be seen in a laboratory. A surprising feature of these experiments is that the gap in the surface spectrum appears *without* bulk magnetic ordering, even though the dopants are uniformly distributed everywhere in the 3D sample. These findings raise several important questions concerning the precise conditions under which TRI-breaking perturbations open up a gap. Can a gap open in the surface state of a TI in a TRI-broken phase which, however, lacks global magnetic ordering? Although we know of no systematic study of this problem, simple arguments suggest that unordered magnetic moments do not open a gap. Consider creating such a disordered state from a uniform two-dimensional (2D) ferromagnet (FM) in the surface of a TI by introducing domains with opposite magnetization (taken to point in the direction perpendicular to the surface). It is well known that the resulting domain walls carry topologically protected gapless fermionic modes.[28] As the number of the domains grows so does the density of the low-energy fermion modes, ultimately presumably recovering the 2D gapless state characteristic of the system with unbroken TRI. The above argument thus suggests that uniform magnetic ordering over large domains is necessary to gap out the surface modes in a TI.

In this paper, we lay out the hypothesis that a temperature window exists in which the surface of a magnetically doped TI is magnetically ordered but the bulk is not. We present a simple and intuitive argument why this is so, and we back it up via a mean-field calculation for two simple tight-binding TI models: a cubic-lattice regularized Bi_2Se_3 and a model on the perovskite lattice. Our results show that indeed a sizable regime such as that described above could exist in real TIs, and this indicates a possible physical explanation for the results seen in experiments.[26,27]

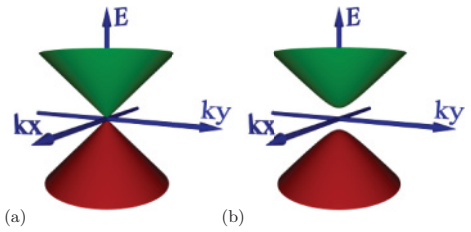

FIG. 1. (Color online) (a) Energy spectrum of a massless Dirac fermion. The bottom (red) cone is the valence band, fully occupied at half filling. The top (green) cone is the conduction band, which is assumed to be vacant. (b) Massive Dirac fermion. This figure shows how the opening of a gap tends to lower the free energy.

II. SURFACE MAGNETIC ORDERING IN TOPOLOGICAL INSULATORS

A. Surface doping

The most natural way to attempt to open up a gap in the surface state of a TI is to coat the surface with a ferromagnetic material, with magnetization perpendicular to the surface. Theoretically, this causes a gap to open up, proportional to the magnetization of the FM coating.[28,29] To illustrate this point, consider the effective low-energy Hamiltonian for electrons on the surface of a 3D TI that lies parallel to the x-y plane,

$$\mathcal{H}_0 = v(k_x\sigma_y - k_y\sigma_x), \tag{1}$$

where v is the Fermi velocity, and σ_i are Pauli matrices in the spin subspace. If we coat this surface with a ferromagnetic coating with magnetization $\boldsymbol{M} = M\hat{z}$ then we get an additional term in the Hamiltonian,

$$\mathcal{H} = \mathcal{H}_0 + JM\frac{\sigma_z}{2}, \tag{2}$$

where J is the exchange-coupling strength. Since $\mathcal{H}$ is a sum of anticommuting matrices we can write the spectrum immediately:

$$E_k = \pm\sqrt{v^2k^2 + (JM/2)^2}, \tag{3}$$

where $k^2 = k_x^2 + k_y^2$.

We see that a gap of size JM has opened up. However, even though from a theoretical standpoint this proposition seems promising, experimentally it has proven very difficult to fabricate a sample with the requisite properties. Two key challenges need to be overcome: First, to observe most of the interesting surface phenomena, one requires the surface to remain insulating; however, most ferromagnets in nature are metallic. Second, for a ferromagnet in a thin-film geometry, the magnetization vector usually lies in the plane, whereas a perpendicular magnetization is required to open up a gap in the TI surface state. To the best of our knowledge, this has yet to be achieved in an experiment, although some theoretical work has been done on this topic.[30]

B. Bulk doping

If surface doping with magnetic impurities fails, it is natural to try bulk doping. In ARPES experiments[26,27] it was found that doping the bulk with nonmagnetic impurities (such as Ca, Sn, and Tl) did not result in a gap in the Dirac cone, as expected since they do not break TRI. Conversely, doping with magnetic impurities, for example $Bi_{2-x}Fe_xSe_3$, resulted in a spectral gap that increased with the concentration of magnetic dopants x, with a gap of 60 meV for $x = 0.25$ (the bulk gap for Bi_2Se_3 is $\sim$0.3 eV). For the magnetic dopants Fe and Mn it was found that, at least for small x, the bulk was paramagnetic, whereas for the undoped samples the bulk was found to be diamagnetic. The magnetization measurements were not sensitive to the surface.

This raises the question of magnetic ordering in the bulk versus the surface. In general, ordered phenomena in lower dimensions are more fragile ($T_c^{3D} > T_c^{2D}$), for example, the XY model and Heisenberg models: in one dimension they do not order at any temperature, in two dimensions they order only for $T = 0$, and in three dimensions they order for $T < T_c$. This is also the case for the superconducting order and general stability of lattices. However, in the case of a TI, we argue that it is possible that $T_c^{\text{bulk}} < T_c^{\text{surf}}$. Therefore, there is a regime $T_c^{\text{bulk}} < T < T_c^{\text{surf}}$ in which the bulk is unordered (paramagnetic) and the surface is ordered (for example, ferromagnetic).

To illustrate why this could be the case, first recall that magnetic ordering with the magnetization perpendicular to the TI surface implies opening of a gap in the spectrum of the surface states. This can be seen directly from Eqs. (2) and (3). Now consider the ungapped surface spectrum, assuming half filling, so that the surface valence band is fully occupied and the conduction band is empty [Fig. 1(a)]. Gapping the surface states causes the occupied states to move down in energy [Fig. 1(b)], so the total kinetic energy decreases. Therefore, the formation of a surface gap is favorable. If the chemical potential is shifted either up or down then the net gain in kinetic energy is diminished and we expect T_c^{surf} to decrease.

Contrast this with the situation in the insulating bulk, which is gapped to begin with. In an ordinary insulator with negligible spin-orbit coupling it is not possible to generate magnetization in the initially spin-degenerate bands without first closing the gap. Equivalently, one may recall that the spin susceptibility of an ordinary insulator with a negligible spin-orbit coupling vanishes. In the bulk of a topological insulator the situation is more complicated as a result of the strong spin-orbit coupling that is necessary for the occurrence of the topological phase. In this case, magnetic susceptibility can be significant[31] and can lead to bulk magnetic states at nonzero temperatures. Nevertheless, in this study we find that generically the surface critical temperature for the formation of magnetic order exceeds the bulk critical temperature. We remark that a similar situation has been found in a theoretical study of the transition-metal oxide Na_2IrO_3, which was predicted to be a layered quantum spin Hall system.[32]

Equivalently, we can imagine integrating out the electrons, and we consider that the range of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the dopants is inversely proportional to the gap. Since the gap is small for the surface and large for the bulk, the range of the interaction is much longer for the surface, and hence long-range order is expected to be stronger on the surface.[33]

III. FORMALISM

A. Mean-field theory for the bulk system

We consider first just the 3D bulk of the material and study the coupling of electrons to magnetic dopants within a mean-field approximation. We assume that the density of impurities is low enough that we can neglect impurity-impurity interactions and that there is no clustering. This is supported by experiments[34] on doped Bi_2Te_3. Therefore, we add an onsite electron-impurity exchange interaction term[31]

$$H = H_e + J\sum_I \boldsymbol{S}_I \cdot \boldsymbol{s}_I, \tag{4}$$

where H_e is the Hamiltonian for the electrons in a TI to be discussed in Sec. IV, J is an exchange-coupling constant, the sum extends over all impurity sites I, and $\boldsymbol{S}_I$ and $\boldsymbol{s}_I$ are the spin operators of the impurity and electron on site I, respectively.

We define the average magnetization of the impurities and electrons as $\boldsymbol{M}$ and $\boldsymbol{m}$. The impurity magnetization is averaged over all impurities and over all sites. This virtual crystal approximation can be pictured as "spreading out" the localized magnetic moment of the impurities so that there is a moment on all sites. We assume that the fluctuations around the mean are small and can be neglected, so that the interaction term is decoupled:

$$J\sum_I \boldsymbol{S}_I \cdot \boldsymbol{s}_I \simeq J\boldsymbol{M}\cdot\sum_i \boldsymbol{s}_i + J\boldsymbol{m}\cdot\sum_I \boldsymbol{S}_I - NJ\boldsymbol{M}\cdot\boldsymbol{m}, \tag{5}$$

where N is the number of sites. We assume that the magnetization is in the z direction, so $\boldsymbol{M} = M\hat{z}$ and $\boldsymbol{m} = m\hat{z}$. Then the mean-field Hamiltonian becomes $H^{\mathrm{MF}} = H_e^{\mathrm{MF}} + H_{\mathrm{imp}}^{\mathrm{MF}} - NJMm$, where

$$\begin{aligned} H_e^{\mathrm{MF}} &= \sum_{\boldsymbol{k}} \Psi_k^\dagger \left[\mathcal{H}_e(\boldsymbol{k}) + JM\frac{\sigma_z}{2} - \mu\right]\Psi_k, \\ H_{\mathrm{imp}}^{\mathrm{MF}} &= Jm\sum_I \boldsymbol{S}_I^z, \end{aligned} \tag{6}$$

and $\mathcal{H}_e(\boldsymbol{k})$ is a matrix in spin and orbital spaces. Since the mean-field Hamiltonian is decoupled, we can write the energies as a sum

$$E(\boldsymbol{k},\lambda_I) = E_e(\boldsymbol{k}) + E_{\mathrm{imp}} - NJMm. \tag{7}$$

The first term reflects the energies of the mean-field electron Hamiltonian H_e^{MF}. The second term is a sum over the expectation values of the spin operator in the z direction at each impurity site $E_{\mathrm{imp}} = Jm\sum_I \lambda_I$, where λ_I is the component of the spin in the z direction on impurity I. The last term gives the overall shift in energy following from the mean-field decoupling Eq. (5).

To find the magnetizations, we calculate the expectation values of the electron and impurity spins in the ensemble defined by H^{MF} and find the equations

$$\begin{aligned} M &= -2SxB_S(\beta JmS), \\ m &= \frac{1}{N}\sum_{\boldsymbol{k},i}\left(U^\dagger\frac{\sigma_z}{2}U\right)_{ii} f(E_i), \end{aligned} \tag{8}$$

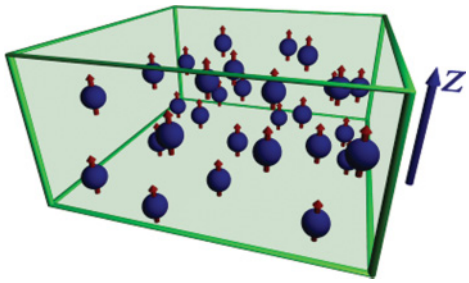

FIG. 2. (Color online) A 3D TI in a slab geometry with a low density of randomly positioned magnetic impurities.

where $B_S(y)$ is the Brillouin function[35]

$$B_S(y) = \frac{2S+1}{2S}\coth\frac{2S+1}{2S}y - \frac{1}{2S}\coth\frac{1}{2S}y, \tag{9}$$

and where U is the matrix that diagonalizes H_e^{MF} (it is a function of J, M, and $\boldsymbol{k}$) and $f(E)$ is the Fermi function. The chemical potential μ can be determined by summing over the average occupation for each energy (given by the Fermi-Dirac distribution) and equating to the total number of states. The number of states per site is $n = \sum_k f(E_e)/N$. For example, for half filling ($\mu = 0$) at $T = 0$ for a model with four states per site, we have $n = 2$ since only half of those are occupied. The equations for m, M, and μ are coupled nonlinear equations which can be solved self-consistently by an iterative procedure.

B. Adding the surfaces

Consider now a system with open boundary conditions in the z direction and periodic boundary conditions in the x and y directions (see Fig. 2). The advantage of this setup is that, in addition to the bulk, we can also investigate the magnetization near the two surfaces of the sample. We take the real space Hamiltonian, and Fourier transform it only in x and y, keeping the dependence on z in real space. The problem is now a one-dimensional (1D) problem in z, as opposed to the zero-dimensional (0D) problem before.

Once again, we rewrite the interaction term and then define the magnetizations as z-dependent fields. The mean-field Hamiltonian is now $H^{\mathrm{MF}} = H_e^{\mathrm{MF}} + H_{\mathrm{imp}}^{\mathrm{MF}} - N_\perp J\sum_j M_j \cdot m_j$, where j labels the layers and

$$\begin{aligned} H_e^{\mathrm{MF}} &= \sum_{\boldsymbol{k}_\perp} \Psi_{k_\perp}^\dagger \left[\mathcal{H}_e(\boldsymbol{k}_\perp) + J\boldsymbol{M}\otimes\frac{\sigma_z}{2} - \mu\right]\Psi_{k_\perp}, \\ H_{\mathrm{imp}}^{\mathrm{MF}} &= J\sum_{j,I\in j} m_j \cdot \boldsymbol{S}_I. \end{aligned} \tag{10}$$

Here $\mathcal{H}_e(\boldsymbol{k}_\perp)$ is a matrix in indices labeling the z coordinate in addition to spin and orbital spaces; $\boldsymbol{M}$ is a vector of length L_z (the number of sites in the z direction), $N_\perp = N/L_z$ is the number of sites in the x-y plane, and by $\boldsymbol{M}\otimes\sigma_z$ we mean that $\boldsymbol{M}$ is expanded to a diagonal matrix of dimension L_z and then we perform an external product with σ_z. In $H_{\mathrm{imp}}^{\mathrm{MF}}$ the summation is over layer index j and dopant sites I in layer j.

We now diagonalize the full electron Hamiltonian $\mathcal{H}_e(\boldsymbol{k}_\perp) + J\boldsymbol{M}\otimes\sigma_z/2 - \mu$ and find a matrix U which diagonalizes it. We calculate the electron and impurity magnetizations of each layer, which are given by the sum over the expectation values of the spin operators

$$\begin{aligned} M_i &= -2Sx B_S(\beta J m_i S), \\ m_i &= \frac{1}{N_\perp}\sum_{l,j}\left(U_{il}^\dagger \frac{\sigma_z}{2} U_{il}\right)_{jj} f(E_j), \end{aligned} \tag{11}$$

where U_{il} is a $d\times d$ block of U, and d is the dimension of the Hamiltonian in the bulk ($d=4$ for Bi_2Se_3 and $d=8$ for perovskite; the models are defined below). The chemical potential can be determined as before by solving $n = \sum_{\boldsymbol{k}_\perp,\alpha} f(E_\alpha(\boldsymbol{k}_\perp))/N$, where α runs from 1 to $L_z d$. In the limit of a macroscopic crystal that is homogeneously doped, we expect to find that the doping for each layer is equal, and hence x does not vary in our sample. It would be straightforward to consider a spatially varying dopant density x but we do not pursue this here.

C. Estimate of the surface critical temperature

Consider a ferromagnetically ordered 2D surface of a TI. We can make a rough estimate of the critical temperature. The effective Hamiltonian is given by Eqs. (1) and (2), and the energies are given by Eq. (3). The impurity and electron magnetizations are described by the coupled equations

$$\begin{aligned} M &= -2Sx B_S(\beta J m S), \\ m &= \frac{1}{NJ}\sum_k \frac{\partial E_k}{\partial M} f(E_k-\mu). \end{aligned} \tag{12}$$

For $T\to T_c^{\rm surf}$ we have $m, M\to 0$, so we expand the above equations to first order in M and m. We make use of $\partial E/\partial M = J^2 M/4E$ and the asymptotic form of the Brillouin function $B_S(y)\simeq y(S+1)/3S$ for $y\ll 1$, and we find

$$\begin{aligned} M &= -\frac{2}{3}S(S+1)\beta J x m, \\ m &= -\frac{JM}{4N}\sum_k \frac{1}{E_k}[1-f(E_k+\mu)-f(E_k-\mu)]. \end{aligned} \tag{13}$$

We can evaluate the sum by converting it into an integral and imposing a momentum cutoff Λ. Assuming $E_k\simeq \pm v|k|$ (since $M\to 0$ at the critical point) we find a simple result[30,36] for exact half filling (i.e., $\mu=0$),

$$T_c^{\rm surf} \simeq \pi \frac{S(S+1)}{3k_B}\left(\frac{\Lambda a}{2\pi}\right)^2 \frac{J^2 x}{(\Lambda v)}. \tag{14}$$

For $S=5/2$, $J=0.5$ eV, $x=0.05$, $v=2\lambda_\perp a$, and a cutoff $\Lambda a/(2\pi)\simeq 1/5$ we find $T_c^{\rm surf}\simeq 100$ K. Away from half filling, when $\beta\mu\ll 1$, there is a small correction of $\sim -\mu^2/(4k_B v\Lambda)$ to the result in Eq. (14).

A simple result can also be found for the case $\beta\mu\gg 1$, which is relevant for most values of μ inside the bulk gap,

$$T_c^{\rm surf} \simeq \pi \frac{S(S+1)}{3k_B}\left(\frac{\Lambda a}{2\pi}\right)^2 \frac{J^2 x}{(\Lambda v)^2}(\Lambda v - |\mu|). \tag{15}$$

As expected, $T_c^{\rm surf}$ is seen to decrease away from half filling.

To complete the argument we would now like to give a similar simple estimate for $T_c^{\rm bulk}$. Unfortunately, the bulk critical temperature is not easy to estimate, since unlike the topologically protected surface state, whose physics is simple and universal, the bulk of a TI can be complicated and $T_c^{\rm bulk}$ will generally depend on the details of the band structure and other factors. For Bi_2Se_3 with 5% concentration of Cr dopants, Ref. 31 estimates $T_c^{\rm bulk}\simeq 70$ K using first-principles numerical calculations. Comparing with our rough estimate for $T_c^{\rm surf}$ given above we see a clear indication that a $T_c^{\rm bulk} < T_c^{\rm surf}$ regime can be easily obtained.

IV. MODELS AND RESULTS

Below we present our results for the two tight-binding models we considered in this study: a model for Bi_2Se_3 regularized on a simple cubic lattice[37] and a model on the perovskite lattice.[38] These simple tight-binding models with spin-orbit coupling exhibit nontrivial topological invariants for a broad range of model parameters and have been used widely in the literature to study the physical properties of topological insulators.

A. Effective model for Bi_2Se_3 regularized on the cubic lattice

We consider electrons hopping on a simple cubic lattice with two orbitals per site [Fig. 3(a)]. The form of the Hamiltonian and the parameters (see Fig. 4) were chosen to fit the dispersion near the center of the Brillouin zone obtained by the first-principles calculation[10,37,39] for Bi_2Se_3. The electron Hamiltonian is given by

$$\begin{aligned} \mathcal{H}_e(\boldsymbol{k}) &= d_4 + \sum_\mu d_\mu \Gamma_\mu, \\ d_0 &\equiv m_k = \epsilon - 2\sum_i t_i\cos(k_i a_i), \\ d_i &\equiv -2\lambda_i \sin(k_i a_i), \\ d_4 &\equiv \gamma_0 - 2\sum_i \gamma_i \cos(k_i a_i), \end{aligned} \tag{16}$$

where we have chosen the gamma matrices so that $\Gamma_0 = \tau_1\otimes\sigma_0$, $\Gamma_1 = -\tau_3\otimes\sigma_2$, $\Gamma_2 = \tau_3\otimes\sigma_1$, $\Gamma_3 = \tau_2\otimes\sigma_0$, and τ_i are Pauli matrices in the orbital subspace and σ_i are Pauli matrices in the spin subspace. The energies are

$$E_e = d_4 + \gamma\sqrt{d_1^2+d_2^2+\left[\sqrt{d_0^2+d_3^2}+\delta\tilde{M}\right]^2}, \tag{17}$$

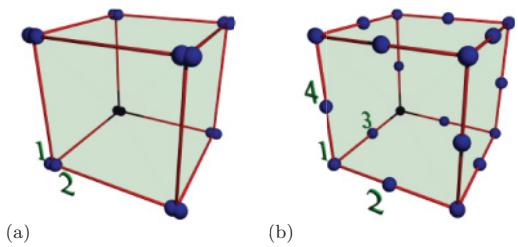

FIG. 3. (Color online) (a) A unit cell of the discretized Bi_2Se_3 model, a simple cubic lattice with two orbitals per site. (b) A unit cell of the perovskite lattice, which can be described as edge-centered cubic. The four basis sites are labeled.

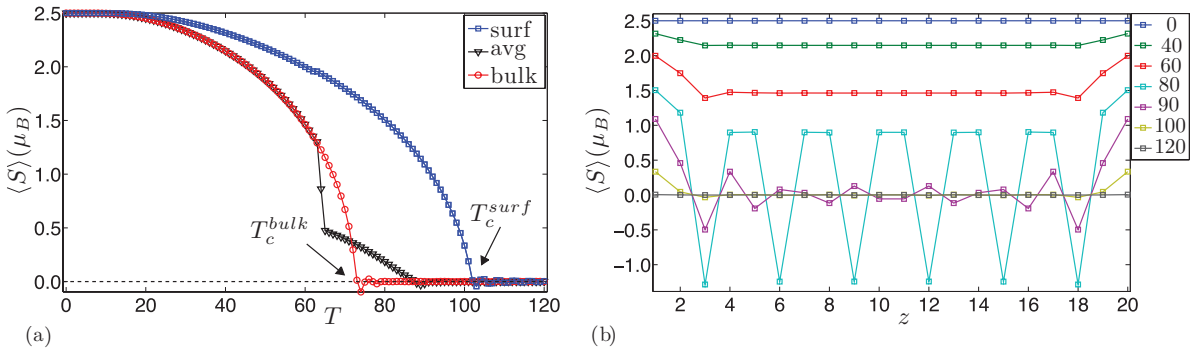

FIG. 4. (Color online) (a) Impurity spin expectation value as a function of temperature for the discretized Bi_2Se_3 lattice. We plot the order parameter on the surfaces ("surf"), the averaged bulk result for the 14 middle sites ("avg"), and the results of the separate bulk calculation ("bulk"). (b) Impurity spin expectation value as a function of the z coordinate for different temperatures. For both plots the parameters are $L_x = L_y = 40$, $L_z = 20$, $S = 5/2$, $J = 0.5$ eV, $x = 0.05$, and the rest of the parameters are chosen to fit the Bi_2Se_3 dispersion close to the Γ point based on first-principles calculations:[10,37,39] $\gamma_0 = 0.3391$ eV, $\gamma_\perp = 0.0506$ eV, $\gamma_z = 0.0717$ eV, $\epsilon = 1.6912$ eV, $t_\perp = 0.3892$ eV, $t_z = 0.2072$ eV, $\lambda_\perp = 0.2170$ eV, and $\lambda_z = 0.1240$ eV.

where $\tilde{M} \equiv JM/2$, and $\gamma,\delta = \pm 1$. Note that if there are no impurities ($J = 0$) then we get the "clean" doubly degenerate electron spectrum $E_0 = d_4 \pm \sqrt{\sum_\mu d_\mu^2}$.

The results for the magnetization in this model are presented in Fig. 4. The bulk is ferromagnetically ordered up to $T_c^{\rm bulk} \simeq 73$ K, and the surface remains FM ordered up to $T_c^{\rm surf} \simeq 102$ K for the two surfaces [see Fig. 4(a)]. Therefore, the window in which the surface is ordered and the bulk is unordered (paramagnetic) is $\simeq$29 K. We plot the magnetization in the z direction in Fig. 4(b). Here the effect can be seen clearly. If we ramp up the temperature from $T = 0$ at first, all spins, regardless of being in the bulk or surface, are fully polarized. As we increase the temperature, the magnetization of the bulk drops faster than the magnetization of the surface. Eventually we cross $T_c^{\rm bulk}$, at which stage the magnetization of the bulk is zero, but the magnetization of the surface is finite. If we increase the temperature further, the magnetization of the surface drops, but the magnetization of the bulk remains at zero. Once we cross $T_c^{\rm surf}$, the magnetization of both the bulk and the surface vanish—thermal fluctuations have broken the ordered phases.

In samples with open surfaces we observed spatial fluctuations in the bulk magnetization as the temperature approached $T_c^{\rm bulk}$ from above, e.g., the $T = 80$ K curve in Fig. 4(b). We attribute these to the large magnetic susceptibility of the bulk, which diverges at $T \to T_c^{\rm bulk}$, and the proximity of the ordered surfaces. The correct $T_c^{\rm bulk}$ can be obtained from the bulk calculation with periodic boundary conditions, the results of which are plotted in Fig. 4(a).

We plot the critical temperature for the surface and the bulk as a function of the chemical potential in Fig. 5. The surface critical temperature is maximal when the chemical potential intersects the surface Dirac point, which happens for $\mu \simeq 0.09$ eV, and falls on both sides, as expected. The result agrees well with the linear dependence of the surface critical temperature on μ predicted in Eq. (15). The bulk critical temperature is approximately constant within the bulk gap, as expected; since there are no bulk states within the gap, changing the chemical potential should not change the critical temperature.

B. Perovskite lattice model

To ascertain whether the results obtained for the Bi_2Se_3 model are in fact generic, we investigate another simple lattice model known to give a robust TI. The perovskite lattice, or edge-centered cubic lattice, consists of atoms on a simple cubic lattice with a four-point basis, corresponding to atoms at the center of each edge [Fig. 3(b)]. Tight-binding electrons on this lattice (with spin-orbit coupling) were recently shown to form a TI.[38] The momentum-space electron Hamiltonian, written in the basis of four sublattice sites indicated in Fig. 3(b), is $\mathcal{H}_e = \mathcal{H}_0 + \mathcal{H}_{\rm SO}$. Here $\mathcal{H}_0$ is the hopping part,

$$\mathcal{H}_0 = -2t \begin{pmatrix} 0 & \cos k_x & \cos k_y & \cos k_z \\ \cos k_x & 0 & 0 & 0 \\ \cos k_y & 0 & 0 & 0 \\ \cos k_z & 0 & 0 & 0 \end{pmatrix}, \quad (18)$$

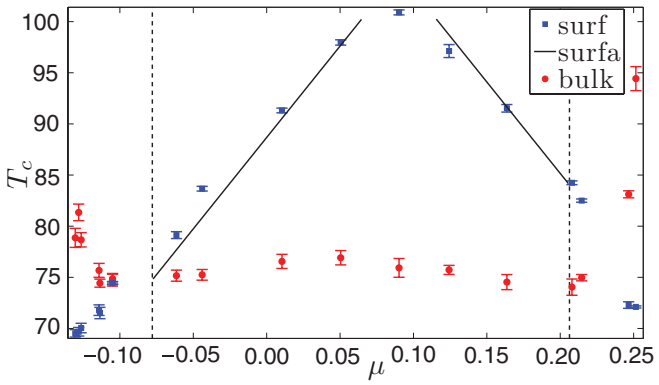

FIG. 5. (Color online) Critical temperature as a function of the chemical potential μ for the discretized Bi_2Se_3 lattice. We plot the surface critical temperature (blue squares) and the approximate surface temperature from Eq. (15) (solid black line). In addition, we plot the bulk critical temperature as obtained from an average over the bulk sites (red circles). The error bars were obtained by fitting the magnetization below the critical temperature to the known behavior for a second-order phase transition, $M \propto (T_c - T)^{1/2}$. The parameters are as in Fig. 4, except for $L_z = 10$. The cutoff was visually chosen for best fit: $\Lambda a/2\pi = 0.215$. The vertical dashed lines mark the bulk gap.

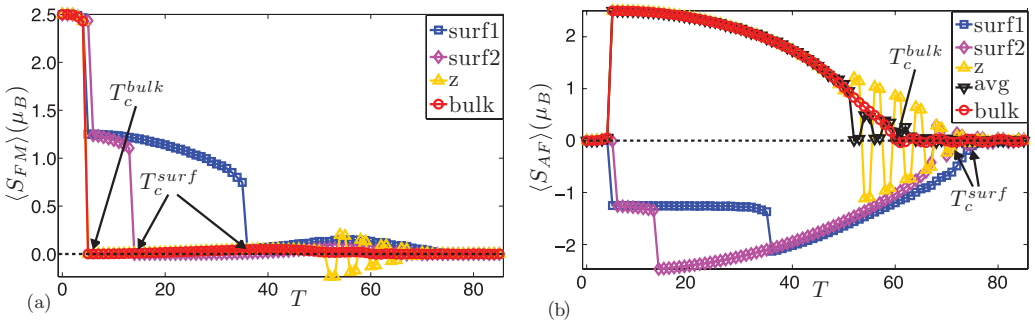

FIG. 6. (Color online) (a) Ferromagnetic order parameter as a function of temperature for the perovskite lattice, with parameters $L_x = L_y = 20$, $L_z = 10$, $S = 5/2$, $J = 0.25$ eV, $t = 1$ eV, $\lambda = 0.2$ eV, and $x = 0.05$. We plot the order parameter on each of the surfaces ("surf1" and "surf2"), the result for the site at $z = L_z/2$ ("z"), and the results of the separate bulk calculation ("bulk"). (b) Antiferromagnetic order parameter as a function of temperature for the perovskite lattice, with parameters as in (a). Here we also plot the average over the four central sites ("avg").

and $\mathcal{H}_{\rm SO}$ is the spin-orbit part, $\mathcal{H}_{\rm SO} = \sum_\mu \sigma_\mu \otimes \mathcal{H}_{\rm SO}^\mu$, and

$$\mathcal{H}_{\rm SO}^x = 4\lambda i \sin k_y \sin k_z \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & -1 & 0 \end{pmatrix}, \qquad (19)$$

and similarly for y and z.

The results for magnetization in this model are presented in Fig. 6. In this case we observe that for a large range of temperatures the magnetizations on basis sites 1 and 4 are similar and opposite in sign from the magnetizations on basis sites 2 and 3 (which are equivalent due to a rotational symmetry around the z axis). This motivates the definition of a FM order parameter $S_i^{\rm FM} = (S_{i,1} + S_{i,2} + S_{i,3} + S_{i,4})/4$ and an antiferromagnetic (AF) order parameter $S_i^{\rm AF} = (S_{i,1} - S_{i,2} - S_{i,3} + S_{i,4})/4$ (where $S_{i,\ell}$ is the expectation value of the impurity spin on basis site ℓ of lattice site i). We see that the system is ferromagnetically ordered up to $T \simeq 4.5$ K, where it becomes AF ordered. The bulk remains AF ordered until $T_c^{\rm bulk} \simeq 60.5$ K, and the surface remains AF ordered until $T_c^{\rm surf} \simeq 72$ and 78 K for the two surfaces. Therefore, the maximum window in which the surface is ordered and the bulk is unordered (paramagnetic) is $\simeq$17.5 K. Note that the difference in surfaces results from the fact that the unit cell is not symmetric under reflection along z: the "top" of the system ends with basis site 4 and the bottom ends with basis site 1 [see Fig. 3(a)]. In addition, we see that the surfaces undergo an additional partial phase change signified by discrete jumps of the order parameter seen at $T \simeq 4.5$, 5.5, 13.5, and 35.5 K, which we claim as additional evidence that the bulk and surface differ.

V. CONCLUSIONS

We have demonstrated that magnetically doped topological insulators can have a sizable window where the bulk is paramagnetic and the surface is magnetically ordered. Our conclusions are based on general arguments that involve only universal properties of the topologically protected surface states and on numerical calculations performed on simple lattice models of 3D topological insulators. Physically, these results are in accord with the intuition that the metallic state on the surface should be more susceptible to magnetic ordering than the insulating bulk, although this expectation is only partially borne out in systems with strong spin-orbit coupling.

The results reported in this study rely on two key approximations: (i) the mean-field decoupling of the exchange interaction between electrons and magnetic dopants indicated in Eq. (5), and (ii) the "virtual crystal" approximation which replaces the localized magnetic moments of dopant atoms by their average over all lattice sites. We have attempted to bypass the latter approximation by solving the problem in real space without averaging over sites. This procedure gives reasonable results for $T_c^{\rm bulk}$ compared to those obtained in the virtual crystal but is numerically more costly. The system sizes that we could simulate did not allow us to unambiguously determine the surface critical temperature. Nevertheless, based on these results we feel that, in a large crystal, approximation (ii) will not lead to a significant error in the determination of the critical temperatures.

Since fluctuations around the mean-field result are typically stronger in two than in three dimensions, they will likely reduce the size of the temperature window between $T_c^{\rm bulk}$ and $T_c^{\rm surf}$. On the other hand, our mean-field calculation did not include electron-electron interactions, which would tend to stabilize the long-range magnetic order and thereby strengthen the ordered phases. It was shown recently that electron-electron interactions can lead to spontaneous breaking of TRI on the surface of a TI,[40] even in the absence of magnetic dopants. Thus the interactions might in fact strengthen the effect found in our study, although a more detailed investigation would be needed in order to obtain a quantitative result. Overall, in our opinion it is very likely that the combined effect of the magnetic dopants and the electron-electron interactions can account for the experimentally observed surface excitation gap without bulk magnetic order in Mn- and Fe-doped Bi_2Se_3.

We note that the critical temperature for the bulk magnetic ordering of the magnetically doped topological insulator $Bi_{2-x}Mn_xTe_3$ was recently measured[34] to be 9–12 K for $x = 0.04$–0.09. This critical temperature is smaller than our results and those of Ref. 31, which estimate $T_c^{\rm bulk} \simeq 70$ K for Cr-doped Bi_2Se_3 using first-principles numerical calculations. We chose our coupling constant J based on the first-principles results, so we expect that for a reduced coupling constant the temperature window would shift to lower temperatures.

As noted above, our arguments are universal; hence the exact details of the material and coupling might alter the result quantitatively but should not change it qualitatively. Taking the above finding for $Bi_{2-x}Mn_xTe_3$ as a guideline, one may surmise that T_c^{bulk} for Fe- and Mn-doped Bi_2Se_3 lies in a similar range of temperatures. It is then entirely possible that the ARPES experiments,[26,27] performed at $\sim$20 K, detected surface magnetic ordering without bulk magnetism as advocated in this paper. Careful surface-sensitive magnetic measurements, as proposed recently,[41] might be able to probe this intriguing phenomenon directly.

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[1] M. Z. Hasan and C. L. Kane, Rev. Mod. Phys. **82**, 3045 (2010).
[2] J. E. Moore, Nature (London) **464**, 194 (2010).
[3] X.-L. Qi and S.-C. Zhang, Rev. Mod. Phys. **83**, 1057 (2011).
[4] C. L. Kane and E. J. Mele, Phys. Rev. Lett. **95**, 226801 (2005).
[5] C. L. Kane and E. J. Mele, Phys. Rev. Lett. **95**, 146802 (2005).
[6] B. A. Bernevig and S.-C. Zhang, Phys. Rev. Lett. **96**, 106802 (2006).
[7] M. König, S. Wiedmann, C. Brüne, A. Roth, H. Buhmann, L. W. Molenkamp, X.-L. Qi, and S.-C. Zhang, Science **318**, 766 (2007).
[8] B. A. Bernevig, T. L. Hughes, and S.-C. Zhang, Science **314**, 1757 (2006).
[9] L. Fu and C. L. Kane, Phys. Rev. B **76**, 045302 (2007).
[10] H. Zhang, C.-X. Liu, X.-L. Qi, X. Dai, Z. Fang, and S.-C. Zhang, Nat. Phys. **5**, 438 (2009).
[11] Y. Xia *et al.*, Nat. Phys. **5**, 398 (2009).
[12] D. Hsieh *et al.*, Nature (London) **460**, 1101 (2009).
[13] Y.-L. Chen *et al.*, Science **325**, 178 (2009).
[14] Y. Xia *et al.*, Nat. Phys. **5**, 398 (2009).
[15] T. Hanaguri, K. Igarashi, M. Kawamura, H. Takagi, and T. Sasagawa, Phys. Rev. B **82**, 081305(R) (2010).
[16] P. Roushan *et al.*, Nature (London) **460**, 1106 (2009).
[17] T. Zhang *et al.*, Phys. Rev. Lett. **103**, 266803 (2009).
[18] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, Rev. Mod. Phys. **81**, 109 (2009).
[19] A. M. Essin, J. E. Moore, and D. Vanderbilt, Phys. Rev. Lett. **102**, 146805 (2009).
[20] X.-L. Qi, R. Li, J. Zang, and S.-C. Zhang, Science **323**, 1184 (2009).
[21] J. Zang and N. Nagaosa, Phys. Rev. B **81**, 245125 (2010).
[22] W.-K. Tse and A. H. MacDonald, Phys. Rev. Lett. **105**, 057401 (2010).
[23] J. Maciejko, X.-L. Qi, H. D. Drew, and S.-C. Zhang, Phys. Rev. Lett. **105**, 166803 (2010).
[24] T. Yokoyama, Y. Tanaka, and N. Nagaosa, Phys. Rev. B **81**, 121401(R) (2010).
[25] I. Garate and M. Franz, Phys. Rev. Lett. **104**, 146802 (2010).
[26] L. A. Wray, S.-Y. Xu, Y. Xia, D. Hsieh, A. V. Fedorov, Y. S. Hor, R. J. Cava, A. Bansil, H. Lin, and M. Z. Hasan, Nat. Phys. **7**, 32 (2011).
[27] Y. L. Chen *et al.*, Science **329**, 659 (2010).
[28] L. Fu and C. L. Kane, Phys. Rev. Lett. **100**, 096407 (2008).
[29] X.-L. Qi, T. L. Hughes, and S.-C. Zhang, Phys. Rev. B **78**, 195424 (2008).
[30] Q. Liu, C-X. Liu, C. Xu, X-L. Qi and S.-C. Zhang, Phys. Rev. Lett. **102**, 156603 (2009).
[31] R. Yu, W. Zhang, H.-J. Zhang, S.-C. Zhang, X. Dai, and Z. Fang, Science **329**, 61 (2010).
[32] A. Shitade, H. Katsura, J. Kuneš, X.-L. Qi, S.-C. Zhang, and N. Nagaosa, Phys. Rev. Lett. **102**, 256403 (2009).
[33] R. R. Biswas and A. V. Balatsky, Phys. Rev. B **81**, 233405 (2010).
[34] Y. S. Hor *et al.*, Phys. Rev. B **81**, 195203 (2010).
[35] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Saunders, Philadelphia, 1976).
[36] T. Jungwirth, J. Sinova, J. Mašek, J. Kučera, and A. H. MacDonald, Rev. Mod. Phys. **78**, 809 (2006).
[37] L. Fu and E. Berg, Phys. Rev. Lett. **105**, 097001 (2010).
[38] C. Weeks and M. Franz, Phys. Rev. B **82**, 085310 (2010).
[39] R. Li, J. Wang, X.-L. Qi and S.-C. Zhang, Nat. Phys. **6**, 284 (2010).
[40] Y. Baum and A. Stern, Phys. Rev. B **85**, 121105(R) (2012).
[41] M. M. Vazifeh and M. Franz, e-print arXiv:1201.2424.