Non-Linear Spin Susceptibility in Topological Insulators

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We revise the theory of the indirect exchange interaction between magnetic impurities beyond the linear response theory to establish the effect of impurity resonances in the surface states of a three dimensional topological insulator. The interaction is composed of an isotropic Heisenberg, anisotropic Ising and Dzyaloshinskii-Moriya-type of couplings. We find that all three contributions are finite at the Dirac point, which is in stark contrast to the linear response theory which predicts a vanishing Dzyaloshinskii-Moriya-type contribution. We show that the spin-independent component of the impurity scattering can generate large values of the Dzyaloshinskii-Moriya-type coupling in comparison with the Heisenberg and Ising types of coupling, while these latter contributions drastically reduce in magnitude and undergo sign changes. As a result, both collinear and non-collinear configurations are allowed magnetic configurations of the impurities.

**Introduction** — Three-dimensional topological insulators (3D TI), materials with insulating bulk states and two-dimensional gapless surface states have attracted a huge attention during the last decade, both for their fundamentally interesting properties as well as potential applications. Fascinating novel effects such as quantum anomalous Hall effect (QAHE) and topological superconductivity have been observed in these materials and other unprecedented effects have been proposed in the fields of electronics and spintronics. More specifically, since the surface states of these materials follow a pure Rashba-type Hamiltonian, modifications of their band dispersion can be invoked by proximity of a ferromagnet material or magnetic impurities. In the latter case the QAHE has been experimentally observed which makes the field of dilute magnetic TIs an important area of research. It worth to mention that, although this experiment has been observed in magnetic TIs by several groups, the nature of the coupling between the impurities and their alignment is still under vigorous debate.

In dilute magnetic semiconductors, the magnetic impurities mostly interact indirectly via the itinerant electrons of the host system, the so-called Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. This interaction allows control of the magnetic properties by tuning the electronic properties of the system, which is most desirable in the field of spintronics. As a general rule, the RKKY interaction, which is proportional to the spin susceptibility of the host material, scales with the distance $R$ between spins as $R^{-d} \sin(2k_F R)$, where $d$ is the spatial dimension and $k_F$ is the Fermi wave-vector. This long range interaction can lead to ferromagnetic (FM) or antiferromagnetic (AFM) ordering of the impurities. In materials with spin-orbit coupling, an effective Dzyaloshinskii-Moriya (DM) type of interaction appears between the impurities. While the isotropic Heisenberg (H)-type interaction together with the anisotropic Ising (I)-type contribution in magnetic materials favour collinear alignment of the spins, the DM-type interaction is associated with the Hamiltonian $\mathbf{D} \cdot (\mathbf{S} \times \mathbf{S})$, where the vector $\mathbf{D}$ defines the form of relative rotation of the spins, favors a perpendicular orientation of the spins with respect to each other. The competition between these collinear and non-collinear interactions may result in exotic phases such as skyrmions, helices, and chiral domain walls. The RKKY interaction in TI has been studied widely and both collinear and non-collinear terms were reported. The importance of this interaction in TI is that the magnetic moments can couple to each other up to many nanometers in contrast to many Angstroms in magnetic semiconductors. Since these terms can be tuned by changing the electronic doping and the distance between the impurities, it was proposed to deposit magnetic impurities on TIs in any desirable lattice structure or random distribution and study the resulting spin model. However, the fact that the DM-type interaction vanishes at the Dirac point makes realization of exotic phases such as skyrmions challenging.

In Dirac materials, such as 3D TI, it has been shown that magnetic and non-magnetic impurities generate local resonances near the Dirac point. The existence of these resonances becomes more prominent when they emerge at forbidden energies near the band gap or at low density of electron states (DOS) near the Dirac point. Note that a magnetic impurity comprises both a magnetic and a non-magnetic scattering potential. While the former potential generates both electron and hole resonance peaks located symmetrically around the Dirac point, the latter breaks the electron-hole symmetry and creates only an electron or hole resonance, depending on whether it is attractive or repulsive. Recent studies suggest that the gap induced by magnetic impurities may be destroyed by the accompanied non-magnetic scattering. Besides, notwithstanding the peak according to the potential scattering is a universal feature of Dirac materials, the effect of magnetic term would differ in different materials with respect to their spin properties. The effect of impurity resonances on indirect spin-spin coupling in two-dimensional (2D) materials has been investigated recently, however, restricted to spin-degenerate materials. Although the distances between magnetic impurities in dilute magnetic TIs are sufficiently large to suppress direct interaction between them, it is not larger than that their induced impurity states have an influence on their indirect interaction. This observation and the importance of the impurity states near the vanishing DOS...
at the Dirac point motivate the present Rapid Communication.

Here, we investigate the effect of the impurity resonances in the TI surface states on the RKKY interaction. First, we extend the formalism introduced in Ref. 35 and calculate the spin susceptibility beyond the linear response theory and subsequently investigate the effects of both non-magnetic and magnetic scattering potentials on the RKKY interaction. We show that the non-magnetic scattering potential enhances the electron density near the Dirac point, which significantly modifies the properties of the interaction. In particular, we find a quadratic spatial decay in contrast to the cubic obtained in linear response. Moreover, the DM-type interaction becomes finite and non-negligible while the both H and I-types of coupling are reduced and even their sign change in some range of parameters. We show, furthermore, that our findings are not restricted to the Dirac point but is important at finite doping. Finally, we present the application of our results to the final phase of two impurities on the surface of TI.

Theoretical modeling —The surface states of the 3D TI around the Γ point can be described by the effective Hamiltonian 37–40 $H_0 = \hbar v_F (\mathbf{k} \times \hat{\mathbf{z}}) \cdot \sigma$, where $\sigma$ denotes the vector of the Pauli matrices corresponding to the real spin, $\mathbf{k}$ is the momentum, and $v_F$ presents the Fermi velocity. We, furthermore, model the impurity at $\mathbf{r}_0$ by $H_{\text{imp}} = U \delta (\mathbf{r} - \mathbf{r}_0)$, where $U = u r_0 + m \cdot \sigma$ contains both the non-magnetic ($u$) and magnetic ($m$) scattering potentials. The latter, relates to the spin of the impurity, $S$, via $m = \hbar J S / 2$, where $J_s$ is the coupling constant between impurity and itinerant electron spins.

We approach the RKKY interaction beyond linear response theory, by identifying the local magnetization $M(\mathbf{r}) = \chi(\mathbf{r}, \mathbf{r}') \cdot \mathbf{m}(\mathbf{r}')$ where $\chi$ is the susceptibility tensor and $\mathbf{m}$ indicates the magnetic scattering potential given in $H_{\text{imp}}$. By using this relation together with the definition of magnetization based on the spin local density of states (LDOS), $M(\mathbf{r}, \mathbf{r}; \varepsilon) = -3 \Tr [\sigma \mathbf{G}(\mathbf{r}, \mathbf{r}; \varepsilon)] / 2 \pi$, we capture the effect of impurity states in the spin susceptibility tensor. Here, $\mathbf{G}(\mathbf{r}, \mathbf{r}; \varepsilon)$ is the on-site perturbed Green’s function (GF) which can be obtained by using the $T$-matrix approach 40 as below

$$G(\mathbf{r}, \mathbf{r}'; \varepsilon) = G_0(\mathbf{r}, \mathbf{r}'; \varepsilon) + G_0(\mathbf{r}, \mathbf{r}_0; \varepsilon) (U^{-1} - G_0(\varepsilon))^{-1} G_0(\mathbf{r}_0, \mathbf{r}'; \varepsilon). \tag{1}$$

It should be highlighted that scattering off the impurity potential $u$ leads to the emergence of a resonance near the Dirac point, where the position and width of the impurity resonances strongly depend on potential strength, which provides a mechanism for breaking of the electron-hole symmetry. The magnetic scattering potential can be regarded as comprising both repulsive and attractive scattering potentials, one for each spin channel 40. Therefore, a pure magnetic scattering potential preserves electron-hole symmetry, which has a significant influence on the non-linear RKKY interaction, as we shall see below. After some algebra (see Supplemental material 41), the non-linear spin susceptibility tensor can be written as

$$\chi(\mathbf{r}, \mathbf{r}') = -\text{Im} \int_{-\infty}^{\infty} \frac{d\varepsilon}{2\pi} \Tr \left[ \frac{\sigma \mathbf{G}_0(\mathbf{r}, \mathbf{r}'; \varepsilon) \sigma \mathbf{G}_0(\mathbf{r}, \mathbf{r}; \varepsilon)}{1 - 2gu + g^2u^2 - g^2m^2} \right]. \tag{2}$$

where $g(\varepsilon) = \text{Tr} \left[ \int d\mathbf{k} \mathbf{G}_0(\varepsilon, \mathbf{k}) \right] / 2$. As mentioned in Ref. 27, the RKKY interaction in 3D TI is strongly direction dependent. By redefining the spin variable according to $\mathbf{S}_m = (S_m \sin \varphi_R, S_m \cos \varphi_R, S_m)$ where $\varphi_R$ is the angle of the relative distance between impurities, the effective RKKY Hamiltonian assumes the form

$$H_{\text{RKKY}} = J_H S_1 \cdot S_2 + J_{\text{DM}} (S_1 \times S_2) + J_I (S_1 \cdot S_2 + \frac{m_1 S_2 + S_1 S_2}{2}). \tag{3}$$

for which three kinds of pairing between impurities appear with coefficients: H-type, $J_H$, DM-type, $J_{\text{DM}} = J_{\text{DM}}(1, -1, 0)$ and I-type, $J_I$. See Supplemental Material 41 for details.

Results —Within the linear response theory, at zero Fermi energy the RKKY interaction for 2D Dirac materials decays as $R^{-3}$ with unchanged sign, in contrast to other 2D material for which it decays as $R^{-2}$. Moreover, the DM-type interaction is proportional to the spin-orbit coupling and its sign depends on the helicity. Hence, due to the electron-hole symmetry in the TI and opposite helicity in conduction and valance bands, the DM-type coupling is an odd function of the Fermi energy and, hence, vanishes at the Dirac point 29. In the following, we present the corrections to the RKKY interaction induced by the scattering off the magnetic impurity and the implications thereof. In this paper, the energies are scaled by the band cut-off, $\Lambda = 1\ eV$, and $m_u$ by $\Lambda \tilde{t}^2$. Here, $\tilde{t}$ is the short range cut-off introduced by $\lambda \equiv \hbar v_F / \Lambda$ which scales the distance ($R$) between impurities. In all figures, three couplings, $J_s$, are presented in units of $(4\pi J_s \Lambda^{-1})^2$.

The spatial dependence of $J_s$, $i = H, I, DM$ is presented in Fig. 1 for short ($a$) – ($f$) and long distances ($g$) – ($h$), where we plot the interaction for different values of $u$ ($m_0 = 0$) and $m_u$ ($u = 0$). The linear response results ($u = 0$, $m_0 = 0$) are included for reference and display a strictly cubic spatial decay as well as vanishing DM contribution. The impurity scattering, substantially modifies the simply cubic decay of the RKKY interaction as it locally changes the doping of the system. First, we notice that a finite $u$, Fig. 1 ($a$), ($c$), ($e$), ($g$), leads to that all contributions acquire a non-monotonic spatial dependence with strong variation near the impurity. Second, there is a finite range ($2 < R < 8$) of nearly quadratic decay for all interactions. Third, by increasing scattering potential $u$, the H and I contributions change sign near the impurity. This behaviour is equivalent to a transition between FM and AFM phases. Fourth, although collinear contributions decrease in amplitude as $u$ is increased, the DM-type interaction becomes the dominating contribution for large $u$, which is expected to have severe implications on the effective magnetic field exerted by the magnetic impurities on the TI surface states. Fifth, the spatial decay of the impurity resonances leads to that the non-linearity vanishes for large distances, such that the interaction approaches the linear response result (see Fig. 1 ($g$), ($h$)). This is consistent with previous ab-initio results 42, where impurities hosting d-electrons were found to reduce the effective exchange interaction.

It should be noticed, while the combination of a finite magnetic and vanishing non-magnetic scattering potential, see Figs. 1 ($b$), ($d$), ($f$), ($h$), yields a vanishing DM-type contribution, the non-monotonic spatial dependence of $J_H$ and $J_I$
remain as before. In this limit, one can expect an FM formation of the magnetic impurities. Although some of these behaviours are established also for \( m = 0 \), the effect of the magnetic potential is smaller than the \( u \) term. In particular, the sign of the interaction remains intact with growing \( |m| \).

While the linear response theory yields a vanishing DM-type contribution, Fig. 1 (c) shows that it is non-negligible whenever the non-magnetic scattering potential is finite. Note that a mere magnetic scattering potential \( (u = 0, m \neq 0) \) is not sufficient to provide a finite DM-type interaction (Fig. 1 (d)). We attribute this property to that a purely magnetic scattering potential preserves the electron-hole symmetry present in Dirac materials. The non-magnetic scattering potential breaks this symmetry by introducing local doping which leads to a finite \( J_{\text{DM}} \). We expect that this property can be used in spintronics devices with electrical tunability. The plots in Fig. 1 suggest that the scattering potential \( u \) can make this contribution dominating over \( J_H \) and \( J_I \), something which may have an impact on the functionality.

At finite doping, \( \varepsilon_F \neq 0 \), the interaction parameters acquire an oscillating dependence on the Fermi wave vector and distance \( R \) between the spin moments. The plots in Fig. 2 show the dependencies of \( \varepsilon_F \) for varying strengths of the scattering potentials, where the linear response (\( \sin 2k_F R \)) result is included for reference (dark yellow curve). The plots in the left panels clearly show the electron-hole symmetry breaking caused by a finite \( u \) while the right panels show that it is preserved under purely magnetic scattering potentials. Importantly, the scattering potential changes the oscillations and the sign of all terms in a wide range of energies, suggesting that non-linearity terms cannot be neglected without losing accuracy in the theoretical description. It should be noticed that both \( u \) and \( m \) tend to reduce the magnitude of the RKKY interactions. However, for a wide range \( |\varepsilon_F| < 100 \text{ meV} \), that non-magnetic impurity scattering enhances the DM-type contribution while \( J_H \) and \( J_I \) are suppressed, consistent with the effect of the impurity scattering of the spatial decays. The simultaneous effect of \( u \) and \( m \) terms on the isotropic and antisymmetric anisotropic components of the RKKY interaction is plotted in Fig. 3, which shows the parameters (a) \( J_H \) and (b) \( J_{\text{DM}} \) at \( R = 2 \), and at zero doping \( (\varepsilon_F = 0) \). The rastered region in panel (a), indicates a sign change of the H type spin-spin interaction such that anti-parallel alignment of the spins.
configuration, where in the latter case predicts an FM ground state). The figure shows that inclusion necessarily correspond to the ground state of the system and plane perpendicular to \( \phi \) between each other in the ferromagnetic impurities is strongly reduced, such that the interaction between these are dominated by \( J \). The RKKY interaction mediated by the surface states of 3D TIs and found that the impurity states substantially affect the RKKY interaction and intensively modify the picture obtained from linear response theory. In particular, the emergence of impurity resonances from both magnetic and non-magnetic scattering potentials tend to reduce the effective magnetic field, reflecting the fact that magnetic and non-magnetic scattering potentials tend to reduce the electronic structure. In particular, we have studied the electronic structure. In particular, we have studied the electronic structure. In particular, we have studied the electronic structure. In particular, we have studied the electronic structure. In particular, we have studied the electronic structure.

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Figure 3: (Color online) The contour plot of the RKKY couplings (a) \( J_H \) and (b) \( J_{DM} \) in the plane of impurity potential terms \( u,m \) at zero doping, \( \varepsilon_F = 0 \) and \( R = 2 \).

Figure 4: (Color online) The phase diagram of the situation of two magnetic impurities with respect to each other for (a) \( \varepsilon_F = 0 \) with respect to \( u,m \) and (b) for \( m = 0 \) in the plane of \( u,\varepsilon_F \). In both cases, we assumed the relative position of the impurities to be \( R = (2,0) \).
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41 See Supplemental Material at [URL will be inserted by publisher] for details of calculation and explicit forms of the non-linear RKKY couplings.

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