Non-Linear Spin Susceptibility in Topological Insulators

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We theoretically study the effect of impurity resonances on the indirect exchange interaction between magnetic impurities in the surface states of a three dimensional topological insulator. The interaction is composed of an isotropic Heisenberg, and anisotropic Ising and Dzyaloshinskii-Moriya contributions. 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 contribution. We show that the spin-independent component of the impurity scattering can generate large values of the DM term in comparison with the Heisenberg and Ising terms, 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 —The recent decade, there has been a rapidly growing interest in the Dzyalosinski-Moriya (DM) interaction in the field of spintronics [1, 2]. This interaction was initially understood in magnetic insulators [3, 4] but subsequently extended to generally non-centrosymmetric magnetic metals [5]. Moreover, it has been widely studied in hybrid structures such as ferromagnetic (FM)/anti-ferromagnetic (AFM) [6] and FM-bilayer materials [2]. While the isotropic Heisenberg interaction together with the anisotropic Ising contribution in magnetic materials favor collinear alignments of the spins, the DM interaction is associated with the Hamiltonian contribution $\mathbf{D}\cdot(\mathbf{S}_i\times\mathbf{S}_j)$, where the vector $\mathbf{D}$ defines the form of relative rotation of the spins, which 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 [1, 2], which have attracted attention both from fundamental perspectives as well as technological promises for spintronics and quantum computing [1].

Although most of magnetic materials, proposed to be used in the field of spintronics, are based on magnetic metals and insulators, there exists a hope that semiconductors dilutely doped by magnetic impurities can be used in this field as well. The benefit of using semiconductors is that they are more bio-compatible and their properties can be tuned electrically and mechanically more easily [7, 8]. In these materials, the magnetic impurities mostly interact indirectly via the itinerant electrons of the host system, the so-called Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction [9–11]. 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 [12]. As a general role, the RKKY interaction, which is proportional to the spin susceptibility of the host material, scales with the distance R between spins as $R^{-d}\sin(2k_FR)$, where d is the spatial dimension and k_F is the Fermi wavevector. The mentioned long range interaction can lead to FM or AFM ordering of the impurities. In materials with spin-orbit coupling (SOC) [8, 13–16], an effective DM interaction appears between the impurities and new phases such as chiral magnetism and spin glass [14] are possible. Among SOC materials, the surface states of three-dimensional topological insulators (3D TI) defines its own class of materials which can be modelled using a pure Rashba spin-orbit Dirac Hamiltonian which makes it a gapless semimetal. It should be mentioned that, ferromagnetically aligned impurities may provide a gap in the surface states of TI and moreover, the quantum anomalous Hall effect (QAHE) has been observed in a thin version of these materials [17] which makes the magnetically doped TI more fascinating subject to be explored. The RKKY interaction has been studied widely in TI [8, 14, 18] and both collinear and non-collinear terms were reported. Since these terms can be tuned by changing the electronic doping and the distance between impurities, it was proposed to put the magnetic impurities on the TI in any preferred lattice structure and study the preferable spin model [8]. However, the fact that the DM term takes a vanishing magnitude at the Dirac point and consequently small values at a low Fermi energy, makes realizations of exotic phases such as skyrmions challenging [19].

In Dirac materials, such as 3D TI [20], it has been shown that magnetic and non-magnetic impurities produce local resonances near the Dirac point. The existence of these resonances becomes more prominent when they emerge at forbidden energies near the band gap [21] or at low density of electron states (DOS) near the Dirac point [20]. 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 [22]. Besides, notwithstanding the peak according to the potential scattering is a universal feature of Dirac materials [23, 24], 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 2D materials has been investigated recently [25–27], however, restricted to spin-degenerate materials.

In this letter, 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. [26] 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 demonstrate that the spatial decay becomes quadratic in contrast to the cubic decay in the unperturbed scenario. Moreover, the DM interaction becomes finite and non-negligible while the both Heisenberg and Ising components 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 [28–31] $H_0 = \hbar v_F[\mathbf{k}\times\hat{\mathbf{z}}]\cdot\boldsymbol{\sigma}$, where $\boldsymbol{\sigma}$ denotes the vector of Pauli matrices, $\mathbf{k}$ is the momentum, and v_F the Fermi velocity. We, furthermore, model the impurity at $\mathbf{r}_0$ by $H_{\rm imp} = U\delta(\mathbf{r}-\mathbf{r}_0)$, where $U = u\sigma_0 + \mathbf{m}\cdot\boldsymbol{\sigma}$ contains both the non-magnetic (u) and magnetic ($\mathbf{m}$) scattering potentials.

We consider the influence of the local impurity on the surrounding electronic structure in terms of the T-matrix approach [20, 22] for which the full 2×2 matrix Green's function (GF) $\mathbf{G}(\omega,\mathbf{r},\mathbf{r}')$ can be written as

$$\mathbf{G}(\omega,\mathbf{r},\mathbf{r}') = \mathbf{G}_0(\omega,\mathbf{r},\mathbf{r}') + \mathbf{G}_0(\omega,\mathbf{r},\mathbf{r_0})\Big(U^{-1}-\mathbf{G}_0(\omega)\Big)^{-1}\mathbf{G}_0(\omega,\mathbf{r_0},\mathbf{r}'). \quad (1)$$

The local DOS and local spin DOS are related to the GF through $\rho(\mathbf{r},\omega) = -{\rm ImTr}\mathbf{G}(\mathrm{r},\mathrm{r};\omega)/\pi$ and $\mathbf{M}(\mathbf{r},\omega) = -{\rm ImTr}\boldsymbol{\sigma}\mathbf{G}(\mathrm{r},\mathrm{r};\omega)/2\pi$, respectively.

The 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 resonance strongly depends on potential strength. For instance, it appears below (above) the Dirac point for repulsive, $u>0$, (attractive, $u<0$), scattering potential and provides a mechanism for the broken electron-hole symmetry. The magnetic scattering potential can be regarded as to comprise both repulsive and attractive scattering potential, one for each spin channel [20]. Therefore, a pure magnetic scattering potential does not generate a broken electron-hole symmetry, which has a significant influence on the non-linear RKKY interaction, as we shall see below. The emergence of impurity resonances is not restricted to the three-dimensional TIs but is a universal property for all Dirac materials [20, 23, 24, 32–34].

Next, we identify $\mathbf{M}(\mathbf{r}) = \chi(\mathbf{r},\mathbf{r}',m)\cdot\mathbf{m}(\mathbf{r}')$ where χ is the susceptibility tensor, whereas $\mathbf{m}$ is the magnetic scattering potential given in $H_{\rm imp}$. Using this relation together with the definition of magnetization based on the GF in Eq. (1), we obtain the spin susceptibility

$$\chi(\mathbf{r},\mathbf{r}',m) = -\,{\rm Im\,Tr}\int_{-\infty}^{\varepsilon_F}\frac{\boldsymbol{\sigma}\mathbf{G}_0(\mathbf{r},\mathbf{r}';\omega)\boldsymbol{\sigma}\mathbf{G}_0(\mathbf{r}',\mathbf{r};\omega)}{1-2gu+g^2u^2-g^2m^2}\frac{d\omega}{2\pi}, \quad (2)$$

where $g = {\rm Tr}\,\mathbf{G}_0(\mathbf{r},\mathbf{r})$ is the on-site GF.

As mentioned in Ref. [18], the RKKY interaction in 3D TI is strongly direction dependent. By redefining the spin variable according to $\tilde{\mathbf{S}}_m = (S_{mx}\sin(\phi_R), S_{my}\cos(\phi_R), S_{mz})$, the effective Hamiltonian for the RKKY assumes the form

$$H_{\rm RKKY} = J_H S_1\cdot S_2 + \mathbf{J}_{\rm DM}\cdot(\tilde{\mathbf{S}}_1\times\tilde{\mathbf{S}}_2) + J_I(\tilde{\mathbf{S}}_1\cdot\tilde{\mathbf{S}}_2 + \tilde{S}_{1x}\tilde{S}_{2y} + \tilde{S}_{1y}\tilde{S}_{2x}). \quad (3)$$

for which three kinds of pairings between impurities appears with coefficients: Heisenberg, J_H, Dzyaloshinskii-Moriya, $\mathbf{J}_{\rm DM} = J_{\rm DM}(1,-1,0)$ and Ising, J_I. Details of calculating the non-linear RKKY and explicit forms of these different couplings can be found in the supplementary materials.

Results —Within 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 interaction is proportional to the SOC 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 coupling is an odd function of the Fermi energy and, hence, vanishes at the Dirac point [19]. In the following, we focus on the corrections to the RKKY interaction induced by the scattering off the magnetic impurity and the implications thereof.

The spatial dependence of J_i, $i = H, I, DM$ is presented in Fig. 1 for short (a) – (f) and long distances (g) – (h), where we plot the interaction for different values of u ($\mathbf{m}=0$) and $\mathbf{m}$ ($u=0$). The linear response results ($u=0$, $\mathbf{m}=0$) are included for reference and display a strictly cubic spatial decay as well as vanishing DM contribution. Inclusion of impurity scattering, substantially modifies the simply cubic decay of the RKKY interaction. 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 scattering impurity. Second, there is a finite range ($2<R\lesssim8$) of nearly quadratic decay for all interactions. Third, by increasing potential scattering u, the Heisenberg and Ising contributions change sign near the scattering impurity. This behavior is equivalent to a transition between FM and AFM phases. Fourth, although collinear contributions decrease in amplitude as u is increased, the DM 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)).

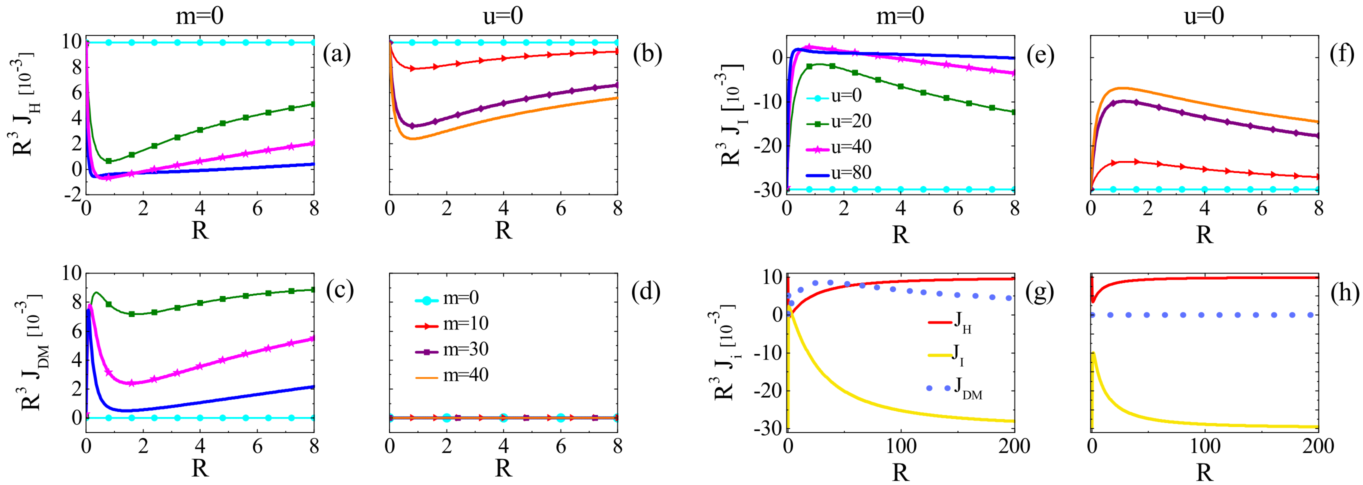

Figure 1: (Color online) Different terms of the RKKY interction multiplied by R^3 as a function of the distance between two impurities at zero doping, $\varepsilon_F = 0$. In panels (a,c,e) $m = 0$ and panels (b,d,f), $u = 0$ while in panel (g), $m = 0$ and $u = 40$ and panel (h), $m = 30$ and $u = 0$. Here, R is scaled by $\hbar v_F$/eV= 2.975Å.

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 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 captured 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 $|\mathbf{m}|$.

While the linear response theory yields a vanishing DM contribution, Fig. 1 (e) shows that it is non-negligible whenever the non-magnetic scattering potential is finite. Note that a mere magnetic scattering potential ($u = 0$, $\mathbf{m} \neq 0$) is not sufficient to provide a finite DM interaction (Fig. 1 (f)). We attribute this property to that a purely magnetic scattering potential preserves the electron-hole symmetry present in Dirac materials. Inclusion of the non-magnetic scattering potential breaks this symmetry which leads to a finite $J_{\rm DM}$. 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 ε_F for varying strengths of the scattering potentials, where the linear response ($\sin 2k_F R$) result is included for reference (dark yellow). 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, inclusion of the scattering potential changes the oscillations and the sign of all terms in a wide range of energies, suggesting that nonlinearity terms cannot be neglected without loosing accuracy in the theoretical description.

It should be noticed that both u and $\mathbf{m}$ tends to reduce the magnitude of the RKKY interactions. However, for a wide range $|\varepsilon_F| < 100$ meV, that non-magnetic impurity scattering enhances the DM contribution while J_H and J_I are suppressed, consistent with the effect of the impurity scattering of the spatial decays.

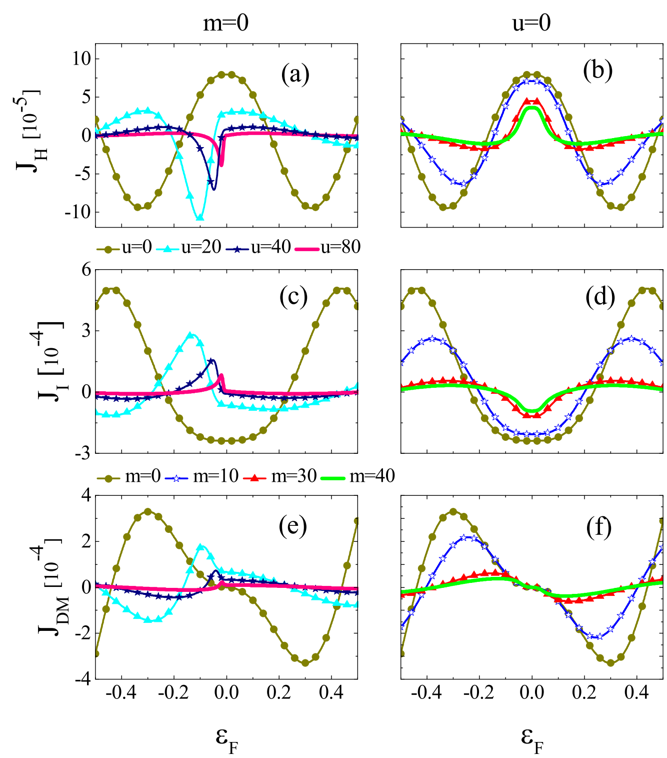

Figure 2: (Color online) Different terms of the RKKY interction as a function of the Fermi energy for distance $R = 2$ between impurities and different values of impurity potential u, m.

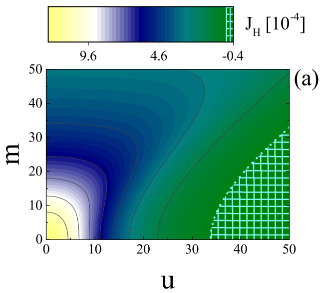

Figure 3: (Color online) The contour plot of the RKKY couplings (a) J_H and (b) J_{DM} in the plane of impurity potential terms u,m at zero doping, $\varepsilon_F = 0$ and $R = 2$.

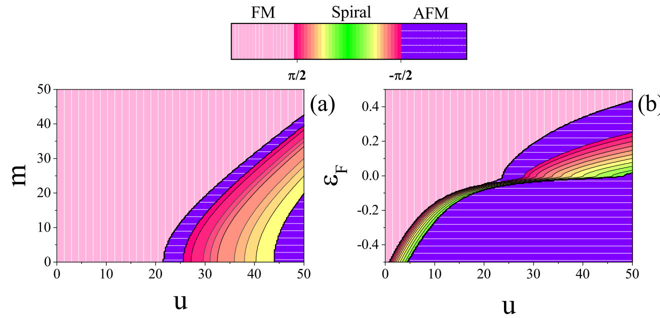

Figure 4: (Color online) The phase diagram of the situation of two magnetic impurities with respect to each other for (a) $\varepsilon_F = 0$ with respect to u,m and (b) for $\mathbf{m} = 0$ in the plane of u,ε_F. In both cases we assumed the relative position of the impurities to be $\mathbf{R} = (2,0)$.

The combined effect of u and m terms on the isotropic and anti-symmetric anisotropic components of the RKKY interaction is plotted in Fig. 3, which shows the parameters (a) J_H and (b) J_{DM} at $R = 2$, and zero doping ($\varepsilon_F = 0$). The rastered region in panel (a), indicates a sign change of the Heisenberg spin-spin interaction such that anti-parallel alignment of the spins is favored over a parallel one, suggesting the possibility of a magnetic phase transition. This region is shifted to higher values of u with increasing distance R, reflecting the fact that the impurity states decay with the distance such that their coupling approaches the linear behaviour. The DM contribution vanishes at $u = 0$ line for all values of $|\mathbf{m}|$, Fig. 3 (b). Moreover, the parameter $J_{\rm DM}$ is a non-monotonic function of u, with a wide peak around a finite value of u. At zero Fermi energy, this parameter is generated by the electron–hole symmetry breaking introduced by a finite u. However, with increasing u, the impurity resonance approaches the Dirac point while its life-time increases which leads to a reduced electron–hole asymmetry. The competition between different couplings opens possibilities to optimize the properties of the magnetic interactions, the resulting effective magnetic field, and the influence of the magnetic impurities on the electronic structure of surface state of the host TI. It has previously been shown that the ratio between the magnetic and non-magnetic scattering potentials strongly influences the possibilities for a gap opening near the Dirac point [22]. The results obtained here, moreover, suggest that a small ratio $|\mathbf{m}|/u$ would favor non-collinear configurations of the magnetic impurities as the interaction between these are dominated by $J_{\rm DM}$. It is then expected that the z-component of the total magnetic field generated by the magnetic impurities is strongly reduced, such that the size of the density gap around the Dirac point is significantly diminished.

In order to better understand the impact of the non-linear interactions and the competition between the different couplings, we investigate the ordering of two impurities in Fig. 4. Following references [16, 35], we find that in presence of the DM term, the magnetic moments become non-collinear, hence, defining an angle $\phi = \arctan(J_{DM}/J_H)$ (here referred to as spiral order) between each other in the plane perpendicular to $J_{\rm DM}$. However, this phase does not necessarily correspond to the ground state of the system and the spins can also be FM or AFM aligned. Figure 4 (a), shows the phases of two impurities located at $\mathbf{r}_1 = (0,0)$ and $\mathbf{r}_2 = (2,0)$, at $\varepsilon_F = 0$ (at which value the linear response theory predicts an FM ground state). The figure shows that inclusion of impurity scattering opens a wide range of the $(u,|\mathbf{m}|)$-plane in which the impurities are either in AFM or in non-collinear configuration, where in the latter case $-\pi/2 < \phi < \pi/2$. Figure 4 (b) illustrates the ordering of the two impurities as a function of u and ε_F, for $m = 0$. The asymmetric behavior about $\varepsilon_F = 0$ is expected due to broken electron–hole symmetry caused by the potential scattering. The details of derivation of the phases, can be found in the supplementary material.

Conclusion —In conclusion, we have developed the theory for indirect spin-spin interactions in Dirac materials by including the influences of the impurity scattering on the local electronic structure. In particular, we have studied the effect of impurity states on the RKKY interaction mediated by the surface states of 3D TIs. We found that for reasonable range of distance between impurities and for experimental accessible values of impurity potential, the impurity states substantially affect the RKKY interactions and completely modifies the picture obtained by means of linear response theory. In particular, the emergence of impurity resonances from both magnetic and non-magnetic scattering potentials tend to reduce the Heisenberg and Ising contributions and may even leads to sign changes of the interactions. In contrast, the DM interaction at zero doping, predicted to vanish in linear response theory, it is not only finite but becomes the dominating interaction for large ratios between the non-magnetic and magnetic scattering potentials. Our results are shown to be stable under finite doping. In terms of our results we predict that the deepened insight to the magnetic interactions may revise the picture concerning the possibilities to create density gaps around the Dirac point using magnetic impurities.

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Suplementary material

In this supplementary material, we first present our formalism based on the T-matrix approach to derive the RKKY interaction beyond the linear response theory that captures the effect of impurity's states. Then we present different terms of magnetic coupling between two impurities on the surface of topological insulators (TI), namely Heisenberg, Ising and Dzyalosinski-Morya (DM). Finally we show how one can use the RKKY coupling to determine the ordering between the moments of two impurities.

RKKY interaction in topological insulator

The RKKY interaction, an indirect exchange interaction between magnetic impurities via the itinerant electrons is mostly studied in the literature within the linear response theory. In this limit, the interaction would be directly proportional to the spin susceptibility of the material as

$$J_{RKKY} \sim m^2 \chi^{\alpha\beta}(r_i, r_j) = \Im \int_{-\infty}^{\varepsilon_F} \frac{d\varepsilon}{\pi} Tr[\sigma^\alpha G_0(r_i, r_j, \varepsilon)\sigma^\beta G_0(r_j, r_i, \varepsilon)], \tag{4}$$

where m shows the strength of the interaction between magnetic impurity and host's electrons, σ is the Pauli matrix for spins, χ^0 is the spin susceptibility of the material, G_0 represents the retarded Green's function and ε_F is the Fermi energy. As one can see in the above formula, the only connection between the RKKY interaction and impurities effect is the m which presents as a prefactor and the coupling between impurities is determined by the pristine sample's susceptibility $\chi^0(r_i, r_j)$ and so the effect of impurity-induced bound states is neglected. Here, we extend the approach introduced in reference [26] for nonlinear charge susceptibility and obtain the spin susceptibility of TI, where the effect of impurity's states is included.

Derivation of non-linear terms

The magnetization $\mathbf{M}$ at point $\mathbf{r}_i$ induced by a magnetic perturbation $\mathbf{m}$ applied at $\mathbf{r}_j$ would relate to the spin susceptibility of the material via

$$\mathbf{M}(\mathbf{r}_i) = \bar{\chi}(\mathbf{r}_i, \mathbf{r}_j, \mathbf{m}) \cdot \mathbf{m}(\mathbf{r}_j), \tag{5}$$

where $\bar{\chi}(\mathbf{r}_i, \mathbf{r}_j, \mathbf{m})$ is the spin susceptibility tensor. Assuming the $\mathbf{m}$ relates to a magnetic impurity placed in the system , it changes the spin-density of itinerant electrons next to itself and the spin local density of states (sLDOS) can be derived from $\mathbf{M}(\mathbf{r}_i, \varepsilon) = -\frac{1}{\pi}\Im Tr\{\boldsymbol{\sigma}\, G(\mathbf{r}_i, \mathbf{r}_i)\}$, which is related to the spin imbalance produced by the magnetic impurity at position $\mathbf{r}_j$ where $G(\mathbf{r}, \mathbf{r}) = G_0(\mathbf{r}, \mathbf{r}, \varepsilon) + \delta G(\mathbf{r}, \mathbf{r}, \varepsilon)$. The total induced magnetization at point $\mathbf{r}_i$ and spin-direction α can be read as

$$\begin{aligned} M^\alpha(\mathbf{r}_i) &= \frac{-1}{2\pi}\Im \int_{-\infty}^{\varepsilon_F} d\varepsilon\, Tr[G(\mathbf{r}_i, \mathbf{r}_i, \varepsilon)\, \sigma^\alpha] = \frac{-1}{2\pi}\Im \int_{-\infty}^{\varepsilon_F} d\varepsilon\, Tr[(\, G_0(\mathbf{r}_i, \mathbf{r}_i, \varepsilon) + \delta G(\mathbf{r}_i, \mathbf{r}_i, \varepsilon)\,)\, \sigma^\alpha] \\ &= \frac{-1}{2\pi}\Im \int_{-\infty}^{\varepsilon_F} d\varepsilon \left\{Tr[G_0(\mathbf{r}_i, \mathbf{r}_i, \varepsilon)\, \sigma^\alpha]\right\} + \frac{-1}{2\pi}\Im \int_{-\infty}^{\varepsilon_F} d\varepsilon \left\{Tr[\delta G(\mathbf{r}_i, \mathbf{r}_i, \varepsilon)\, \sigma^\alpha]\right\}. \end{aligned} \tag{6}$$

In the case of our study, the first term of the latest right hand side of Eq.(6) vanishes for any α-direction so it has no contribution in the magnetization of the system and so the induced magnetization reduces to

$$M^\alpha(\mathbf{r}_i) = \frac{-1}{2\pi}\Im \int_{-\infty}^{\varepsilon_F} d\varepsilon\, Tr[\boldsymbol{\sigma}^\alpha \delta G(\mathbf{r}_i, \mathbf{r}_i, \varepsilon)]. \tag{7}$$

In order to calculate $\delta G(\mathbf{r}_i, \mathbf{r}_i, \varepsilon)$ we used the T-matrix approach for which $\delta G(\mathbf{r}_i, \mathbf{r}_i, \varepsilon) = G^0(\mathbf{r}_i, \mathbf{r}_j, \varepsilon) T(\mathbf{r}_j, \mathbf{r}_j, \varepsilon) G^0(\mathbf{r}_j, \mathbf{r}_i, \varepsilon)$ and T-matrix is defined by $T(r_i, r_i, \varepsilon) = \left[\sigma_0 - U\, G_0(r_i, r_i, \varepsilon)\right]^{-1} U$. Here, U represents the scattering from magnetic impurity including both magnetic and scattering potential and we model it by

$$U = \mathbf{m} \cdot \boldsymbol{\sigma} + u\sigma_0 = \begin{pmatrix} m_z + u & m_x - i\, m_y \\ m_x + i\, m_y & -m_z + u \end{pmatrix} \tag{8}$$

using this form of U, the T-matrix would reduces to

$$T = \begin{pmatrix} \frac{u-g\,u^2+m_z+gm^2}{1-2g\,u+g^2\,u^2-g^2\,m^2} & \frac{m_x-i\,m_y}{1-2g\,u+g^2\,u^2-g^2\,m^2} \\ \frac{m_x+im_y}{1-2g\,u+g^2\,u^2-g^2\,m^2} & \frac{u-g\,u^2-m_z+g\,m^2}{1-2g\,u+g^2\,u^2-g^2\,m^2} \end{pmatrix}, \tag{9}$$

in which, $g = \sum_{\mathbf{k}} G_0(\mathbf{k},\varepsilon)$ and for simplicity, we set $m^2 = m_x^2 + m_y^2 + m_z^2$. By defining the dimensionless quantity $\Upsilon = 1 - 2g\,u + g^2\,u^2 - g^2\,m^2$, we have

$$T = \frac{1}{\Upsilon}\{(u - g\,u^2)\sigma_0 + (g\,\mathbf{m}\,\sigma_0 + \boldsymbol{\sigma})\cdot\mathbf{m}\,\}. \tag{10}$$

So $\delta G_0(\mathbf{r}_i,\mathbf{r}_i,\varepsilon)$ is achieved as

$$\begin{aligned} \delta G(\mathbf{r}_i,\mathbf{r}_i,\varepsilon) &= G_0(\mathbf{r}_i,\mathbf{r}_j,\varepsilon)\,\frac{(u-g\,u^2)\sigma_0 + (g\,\mathbf{m}\,\sigma_0+\boldsymbol{\sigma})\cdot\mathbf{m}}{\Upsilon}\,G_0(\mathbf{r}_j,\mathbf{r}_i,\varepsilon) \\ &= G_0(\mathbf{r}_i,\mathbf{r}_j,\varepsilon)\,\frac{(u-g\,u^2+g\,m^2)\,\sigma_0}{\Upsilon}\,G_0(\mathbf{r}_j,\mathbf{r}_i,\varepsilon) + G_0(\mathbf{r}_i,\mathbf{r}_j,\varepsilon)\,\frac{\boldsymbol{\sigma}\cdot\mathbf{m}}{\Upsilon}\,G_0(\mathbf{r}_j,\mathbf{r}_i,\varepsilon) \end{aligned}. \tag{11}$$

By inserting Eq.(11) into Eq.(7), the magnetization including impurity scattering would be

$$M^\alpha(\mathbf{r}_i) = \frac{-1}{2\pi}\Im\int_{-\infty}^{\varepsilon_F} d\varepsilon\,Tr[\mathcal{G}_0(\mathbf{r}_i,\mathbf{r}_j;\varepsilon)\frac{(u-g\,u^2)\sigma_0 + (g\,\mathbf{m}\,\sigma_0+\boldsymbol{\sigma})\cdot\mathbf{m}}{\Upsilon}\,\mathcal{G}_0(\mathbf{r}_j,\mathbf{r}_i;\varepsilon)\boldsymbol{\sigma}^\alpha]. \tag{12}$$

It can be shown that the terms including σ_0 have no contribution in the trace. Compare Eq.(12) with Eq.(5), the spin susceptibility, $\chi(\mathbf{r}_i,\mathbf{r}_j)$, in the non-linear regime is read as

$$\chi^{\alpha\beta}(\mathbf{r}_i,\mathbf{r}_j) = \frac{-1}{2\pi}\,\Im\int_{-\infty}^{\varepsilon_F} d\varepsilon\,Tr[\sigma^\alpha G_0(\mathbf{r}_i,\mathbf{r}_j,\varepsilon)\,\frac{\sigma^\beta}{\Upsilon}\,G_0(\mathbf{r}_j,\mathbf{r}_i,\varepsilon)], \tag{13}$$

where, β comes from the direction of induced-magnetic field $\mathbf{m}$. Since this integration, based on retarded Green's function, diverges on its lowest limit, using some cut-off is inevitable. As Ref.[36] proposes to avoid divergency, we can rewrite the equation based on Matsubara Green's function. We change the representation of the last equation in terms of Matsubara Green's function in which $\mathcal{G}(\mathbf{r}_i,\mathbf{r}_i;\omega)$ refers to Matsubara Green's function in energy-space and rewrite the tensor of spin susceptibility beyond the linear regime

$$\chi^{\alpha\beta}(\mathbf{r}_i,\mathbf{r}_j) = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega\frac{\sigma^\alpha\mathcal{G}_0(\mathbf{r}_i,\mathbf{r}_j;\omega)\sigma^\beta\mathcal{G}_0(\mathbf{r}_j,\mathbf{r}_i;\omega)}{\Upsilon} \tag{14}$$

Note that while Υ is an even function of m, it is not symmetric for $u > 0$ and $u < 0$ which makes the susceptibility asymmetric.

Explicit form of the Matsubara Green's function in 3D TI

The Matsubara Green's function used in Eq. (14) can be derived by taking the Fourier transformation of the Matsubara k-space Green's function, $\mathcal{G}_0(\mathbf{k};\omega) = [i\omega + \varepsilon_F - H_0(\mathbf{k})]^{-1}$

$$\mathcal{G}_0(\mathbf{r},\mathbf{r}';\omega) = \frac{1}{\Omega_{BZ}}\int d^2\mathbf{k}\,e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\,\mathcal{G}_0(\mathbf{k};\omega) \tag{15}$$

here, the first Brillouin zone is shown by Ω_{BZ}. The unperturbed Matsubara Green's function gets the form of

$$\mathcal{G}_0(\pm R;\omega) = \frac{-2i\pi\omega}{\hbar^2 v_F^2\Omega_{BZ}}\begin{pmatrix} K_0(\frac{(\omega-i\varepsilon_F)\,sgn(\omega)\,R}{\hbar v_F}) & \pm\,e^{-i\phi_R}\;sgn(\omega)K_1(\frac{(\omega-i\varepsilon_F)\,sgn(\omega)\,R}{\hbar v_F}) \\ \pm\,e^{i\phi_R}\;sgn(\omega)K_1(\frac{(\omega-i\varepsilon_F)\,sgn(\omega)\,R}{\hbar v_F}) & K_0(\frac{(\omega-i\varepsilon_F)\,sgn(\omega)\,R}{\hbar v_F}) \end{pmatrix}, \tag{16}$$

where $\pm R = |\mathbf{r}-\mathbf{r}'|, |\mathbf{r}'-\mathbf{r}|$ respectively and $K_{0/1}$ refers to the modified Bessel function. Briefly, we can write Green's function as below

$$\mathcal{G}_0(\pm R;\omega) = \frac{-2i\pi\omega}{\hbar^2 v_F^2\Omega_{BZ}}[K_0(\frac{(\omega-i\varepsilon_F)\;sgn(\omega)\,R}{\hbar v_F})\,\sigma_0 \pm\;K_1(\frac{(\omega-i\varepsilon_F)\;sgn(\omega)\,R}{\hbar v_F})\,(\cos\phi_R\,\sigma_x + \sin\phi_R\,\sigma_y)]. \tag{17}$$

Besides, on-site Green's function is obtained by

$$\mathcal{G}_0^{on-site}(0;\omega) = <\mathbf{0}|\mathcal{G}_0^{on-site}(\mathbf{r},\mathbf{r}';\omega)|\mathbf{0}> = \frac{1}{\Omega_{BZ}} \int_0^{k_c} \int_0^{2\pi} k\, dk\, d\varphi\, \mathcal{G}_0(k;\omega) = g\, \sigma_0, \tag{18}$$

in which $g = \frac{-\pi\,(i\omega+\varepsilon_F)}{\hbar^2 v_F^2 \Omega_{BZ}} \log|1 + \frac{\hbar^2 v_F^2 k_c^2}{(\omega - i\varepsilon_F)^2}|$ and k_c is a momentum cutoff that we suppose it such that $\hbar v_F k_c = 1$ eV, where v_F is the Fermi velocity of electrons on the surface of the topological insulator.

Different components of the spin susceptibility's tensor in 3D TI

In the following, we use analytic form of $\mathcal{G}_0(\pm R;\omega)$, Eq. (17) to obtain all components of spin susceptibility's tensor, Eq. (14) for 3D TI. Different terms of the spin susceptibility tensor can be written as

$$\chi_{zz}(R) = \frac{-4\pi}{\hbar^4 v_F^4 \Omega_{BZ}^2} \int_{-\infty}^{\infty} d\omega\, [(\omega - i\varepsilon_F)^2\, \frac{(K_1^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F}) + K_0^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F}))}{\Upsilon}], \tag{19a}$$

$$\chi_{zy}(R) = \frac{-8\pi}{\hbar^4 v_F^4 \Omega_{BZ}^2} \cos(\varphi_R) \int_{-\infty}^{\infty} d\omega\, i[(\omega - i\varepsilon_F)^2\, sgn(\omega) \frac{K_0(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})\, K_1(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})}{\Upsilon}], \tag{19b}$$

$$\chi_{zx}(R) = \frac{8\pi}{\hbar^4 v_F^4 \Omega_{BZ}^2} \sin(\varphi_R) \int_{-\infty}^{\infty} d\omega\, i[(\omega - i\varepsilon_F)^2\, sgn(\omega) \frac{K_0(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})\, K_1(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})}{\Upsilon}]. \tag{19c}$$

$$\chi_{yz}(R) = -\chi_{zy}(R), \tag{20a}$$

$$\chi_{yy}(R) = \frac{-4\pi}{\hbar^4 v_F^4 \Omega_{BZ}^2} \int_{-\infty}^{\infty} d\omega\, [(\omega - i\varepsilon_F)^2\, \frac{(K_0^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F}) + K_1^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})\cos(2\varphi_R))}{\Upsilon}], \tag{20b}$$

$$\chi_{yx}(R) = \frac{8\pi}{\hbar^4 v_F^4 \Omega_{BZ}^2} \cos(\varphi_R)\sin(\varphi_R) \int_{-\infty}^{\infty} d\omega\, [(\omega - i\varepsilon_F)^2\, \frac{K_1^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})}{\Upsilon}]. \tag{20c}$$

$$\chi_{xz}(R) = -\chi_{zx}(R), \tag{21a}$$

$$\chi_{xy}(R) = \chi_{yx}(R), \tag{21b}$$

$$\chi_{xx}(R) = \frac{-4\pi}{\hbar^4 v_F^4 \Omega_{BZ}^2} \int_{-\infty}^{\infty} d\omega\, [(\omega - i\varepsilon_F)^2\, \frac{(K_0^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F}) - K_1^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})\cos(2\varphi_R))}{\Upsilon}]. \tag{21c}$$

For simplicity, direction dependency of couplings can be extracted by defining the new Spinors as $\tilde{\mathbf{S}}_m = (S_{mx}\sin(\phi_R), S_{my}\cos(\phi_R), S_{mz})$ [18], then the RKKY Hamiltonian including three different coupling terms, Heisenber, Ising and Dzyalosinski-Morya, is achieved by

$$H_{\text{RKKY}} = J_H S_1 \cdot S_2 + \mathbf{J}_{\text{DM}} \cdot (\tilde{\mathbf{S}}_1 \times \tilde{\mathbf{S}}_2) + J_I(\tilde{\mathbf{S}}_1 \cdot \tilde{\mathbf{S}}_2 + \tilde{S}_{1x}\tilde{S}_{2y} + \tilde{S}_{1y}\tilde{S}_{2x}), \tag{22}$$

in which $\mathbf{J}_{DM} = J_{DM}(1,-1,0)$ and

$$\begin{aligned} J_H &= \frac{-4\pi}{\hbar^4\, v_F^4 \Omega_{BZ}^2} \int_{-\infty}^{\infty} d\omega\, (\omega - i\varepsilon_F)^2\, \frac{K_0^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F}) - K_1^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})}{\Upsilon}, \\ J_I &= \frac{-8\pi}{\hbar^4\, v_F^4 \Omega_{BZ}^2} \int_{-\infty}^{\infty} d\omega\, (\omega - i\varepsilon_F)^2\, \frac{K_1^2(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})}{\Upsilon}, \qquad . \\ J_{DM} &= \frac{8\pi}{\hbar^4\, v_F^4 \Omega_{BZ}^2} \int_{-\infty}^{\infty} d\omega\, (\omega - i\varepsilon_F)^2\, \frac{K_0(\frac{(\omega-i\varepsilon_F)\, sgn(\omega)\, R}{\hbar v_F})\, K_1(\frac{\omega\, sgn(\omega)\, R}{\hbar v_F})}{\Upsilon}. \end{aligned} \tag{23}$$

Rotation of spinors

By using the different RKKY interaction terms, one can find the final ordering between two impurities. Let's suppose the impurities to be in x-direction with respect to each other, $R = (R_x, 0)$, then the RKKY Hamiltonian (Eq. (22)) reduces to

$$H = (J_H + J_I)\mathbf{S}_1 \cdot \mathbf{S}_2 - J_I S_{1x} S_{2x} + J_{DM}(S_{1y} S_{2z} - S_{1z} S_{2y}). \tag{24}$$

By assuming S_1 and S_2 as classical vectors, we can write them in the form of $S_{i=1,2} = |S_i|\,(\sin\theta_i \cos\varphi_i, \sin\theta_i \cos\varphi_i, \cos\theta_i)$, so the Hamiltonian can be re-written as

$$H_{RKKY} = |S_1||S_2|\,[(J_H + J_I)(\cos\theta_1\,\cos\theta_2 + \sin\theta_1\,\sin\theta_2 \cos\Delta\varphi) - \sin\theta_1\,\sin\theta_2(J_I \cos\varphi_1\,\cos\varphi_2 - J_{DM}\sin\Delta\varphi)], \tag{25}$$

where $\Delta\varphi = \varphi_2 - \varphi_1$.
To find the minimum energy and configuration of the spinors, we should set $\frac{\partial H}{\partial \theta_i} = 0$ and $\frac{\partial H}{\partial \varphi_i} = 0$, where the first relation binds (θ_1, θ_2) to be one of sets of $(0,0)$, $(0,\pi)$, $(\pi,0)$, (π,π) and $(\pi/2, \pi/2)$. The only possibility of the rotated spinors is $\theta_{1,2} = \pi/2$, in which the values of $\varphi_{1,2}$ are meaningful. By setting $\frac{\partial H}{\partial \varphi_i} = 0$ we obtain

$$\tan\Delta\varphi = \frac{J_{DM}}{J_H - J_I}. \tag{26}$$

By defining the Hessian matrix, we can find which of the above extremum is the true minimum of the system.

$$\begin{aligned} D_1 &= -sgn(J_H - J_I)\sqrt{(J_H - J_I)^2 + J_{DM}^2} \\ D_2 &= J_{DM}^2 + J_I(2J_H - J_I) \end{aligned} \tag{27}$$

Depends on the sign of D_1 and D_2 as bellow, we can obtain the ordering of magnetic impurities.

$$\begin{aligned} &D_1 > 0, D > 0 \rightarrow \textbf{Spiral} \\ &\text{otherwise} \\ &J_H + J_I > 0 \rightarrow \textbf{AFM} \\ &J_H + J_I < 0 \rightarrow \textbf{FM} \end{aligned} \tag{28}$$

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