Spin Response of Electrons on the Surface of a Topological Insulator

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The surface of a topological insulator hosts a very special form of a quasi-two dimensional metallic system when it is embedded in a topologically trivial medium like the vacuum. The electronic properties of this unusual 2D metal are distinct in many aspects from both the conventional two-dimensional electron gas systems in quantum well heterostructures as well as those of a single layer graphene. In this paper, we study one of these distinct features i.e., the response of the electronic spins to an applied magnetic field perpendicular to the surface. We find an unusual behaviour of the spin magnetization and susceptibility as a function of both the magnetic field and the chemical potential for a generic topological surface. We propose that this behavior could be studied by the recently developed experimental technique called β-NMR which is highly sensitive to the surface electron spins. We explain how this technique could be used to probe for spontaneous magnetic ordering caused by magnetic dopants or interactions discussed in the recent literature.

I. INTRODUCTION

Topologically protected electronic states residing on the surface of topological insulators (TI) form a unique 2D metal distinct from those so far realized in solid state systems. Similar to the low energy electronic states in a single layer graphene, the robust 2D metal on an TI surface has conic branches touching (in the absence of the intrinsic gap) at high-symmetry points in the first Brillouin zone. This resemblance in the energy dispersion is responsible for some common properties between these metallic systems, such as the square-root dependence of Landau Levels (LLs) to the applied magnetic field. However, there are important differences between these systems as a result of the topological nature of the surface states in TIs.

The low energy Dirac-like surface bands of a topological insulator arise due to the mismatch of the bulk topological invariants on the two sides of the surface regardless of how constituent atoms have been arranged on the surface as long as the bulk remains in the topological phase. In the case of a ‘strong’ topological insulator (STI) there is an odd number of Dirac points at any surface and, importantly, they are not spin degenerate. Furthermore, they exhibit a unique spin-momentum locking as a result of the strong spin-orbit interaction in the underlying STI which is essential for the formation of the topologically non-trivial insulator phase with time-reversal symmetry. These properties make electronic surface states in a STI very distinct from those of graphene with a pair of spin degenerate Dirac-like bands near two special points in the Brillouin zone which exist as a result of the unique arrangement of carbon atoms in the honeycomb lattice.

One of the interesting aspects in which these 2D metallic systems behave uniquely is their magnetic response. Here, motivated by the recent progress in the experimental methods and the importance of the spin susceptibility for our understanding of electronic systems, we study the spin response of electrons on the surface of a STI and show that it exhibits interesting features even in a simple non-interacting limit. We find that the characteristic spin-momentum locking of these electrons leads to a unique spin response that is distinct from spin-degenerate systems like graphene and 2DEG. Instead of the oscillatory behaviour of the susceptibility as a function of the chemical potential found in spin degenerate systems, in STIs our work predicts a plateau-type behaviour which arises from the strong correlation between spin and orbital degrees of freedom. In addition we find that the cancellation of the 2D metallic systems in a large perpendicular magnetic field which interferes with the superconducting part of the SQUID and causes noise. A variant of this method using a superconducting pickup coil has been developed in order to study deHaas-van Alphen oscillations of the 2D metallic systems in a large perpendicular magnetic field. High-sensitivity micro-mechanical cantilever magnetometry is another way of measuring electronic magnetization, however, this method measures the magnetization of the whole sample and it is difficult to isolate the contribution from the electrons on a single surface.

The nuclear magnetic resonance (NMR) is another powerful experimental technique which can be used to study electronic spin magnetic response in a bulk
metal. Through the so-called Knight shift in the nuclear resonance peak, it is possible to probe the spin part of the magnetic susceptibility of the electrons as they interact with the resonating nuclei in their proximity. Unfortunately, this method, in its conventional form, fails to be useful in very thin films and 2D metallic systems due to the limitation in the number of available nuclei and the resulting weakness of the signal. Thanks to the progresses made by experimentalists in controlling and implementing high energy beams of unstable ions, an experimental technique, the so-called $\beta$-NMR, has been recently developed to overcome the above limitations. Briefly, in this exotic variety of NMR, unstable radioactive ions such as $^6$Li and $^{11}$Be are implanted in the sample surface. The nuclear spin precession signal is then detected through the products of the beta decay of the radioactive nuclei. Since the ion implantation depth can be controlled by tuning the beam energy it is possible to acquire information about the behaviour of the electronic spins in very thin metallic films. Experiments are currently under way to study the surface magnetic response of STI crystals Bi$_2$Se$_3$ and Bi$_2$Te$_3$ using $\beta$-NMR.

The paper is organized as follows. First we introduce a simple model known to describe the electrons on the surface of a STI in the presence of a perpendicular magnetic field. We review the exact solutions of its eigenvalue problem which has been studied previously in various contexts. In section III, we calculate the spin magnetization and susceptibility assuming that the chemical potential lies inside the gap between positive and negative energy eigenstates. We discuss the magnetic response of the surface as one tunes the magnetic field and the chemical potential for various values of the intrinsic gap. In closing, we explain how a $\beta$-NMR experiment might be able to detect these effects through Knight shift measurements.

II. THE MODEL

Many of the interesting features of the surface states in a topological insulator can be captured, at least qualitatively, using a simple non-interacting Dirac Hamiltonian

$$H = \sum_k \Psi_k^\dag [\hbar v_F \sigma \cdot (\hat{z} \times \mathbf{k}) + \Delta_0 \sigma_z] \Psi_k,$$

where $\Psi_k^\dag = (c_{\uparrow \mathbf{k}}^\dag, c_{\downarrow \mathbf{k}}^\dag)$ and $c_{\sigma \mathbf{k}}^\dag$ is the fermionic creation operator of the spin up(down) states with wave vector $\mathbf{k}$. $\Delta_0$ is the intrinsic gap in the surface spectrum which might be nonzero when the time-reversal symmetry is spontaneously broken due to magnetic ordering. The latter can arise due to the presence of magnetic dopants with spin $S$ in the proximity of the surface exchange-coupled to the electronic spins or as a result of electron-electron interaction.

Turning on a perpendicular magnetic field adds two terms to the above Hamiltonian. One is the minimal coupling of the magnetic vector potential, $\hbar \mathbf{k} \rightarrow \hbar \mathbf{k} + e \mathbf{A}$ (electron charge $-e$). The other is the coupling of the spins to the magnetic field, the Zeeman effect, expressed as $\delta H_z = -g_s \mu_B \hbar \mathbf{B} \cdot \mathbf{s}$, where $g_s$ is the effective electron gyromagnetic constant. In the bulk Bi$_2$Se$_3$ crystal $g_s \approx 30$ (Ref. 33) although much smaller values have been reported for electrons near the surface. In our calculations below we use two representative values, $g_s = 8$ and $g_s = 30$, which yield qualitatively similar results with some interesting differences.

In the continuum limit, the leading order Hamiltonian describing these surface states in the presence of the applied magnetic field, $\mathbf{B} = B_0 \hat{z}$, in the Landau gauge $\mathbf{A} = -(B_0 y, 0)$, can be written in the following form

$$H = \sum_{k_x} \int dy \, \Psi^\dag_{k_x}(y) \mathcal{H}(k_x,y) \Psi_{k_x}(y),$$

where $\Psi^\dag_{k_x}(y)$ is the creation operator for the spinor mode extended along the $x$ direction and localized at $y$. $\mathcal{H}(k_x,y)$ for $B_0 > 0$ is defined as

$$\mathcal{H}(k_x,y) = \begin{pmatrix} \Delta & i e_c a^\dag_{k_x} \\ -i e_c a_{k_x} & -\Delta \end{pmatrix},$$

where $e_c = v_F \sqrt{2 \epsilon_0 B_0}$ and

$$\Delta = \Delta_0 - \frac{g_s \mu_B B_0}{2}.$$

The term in $\Delta$ proportional to the magnetic field is the Zeeman contribution. $a_{k_x}$ is the one-dimensional harmonic oscillator bosonic operator defined as

$$a_{k_x} = \frac{1}{\sqrt{2}} \left( \frac{y}{l_B} + l_B (\partial_y - k_x) \right), \quad a^\dag_{k_x} = \frac{\hbar}{e|B_0|}.$$

Note that varying $k_x$ shifts the position of the localized state produced by the application of $a^\dag_{k_x}$ on the vacuum

![FIG. 1: (Color online) The surface spectrum of a strong topological insulator in the absence of magnetic dopants ($\Delta_0 = 0$), (a) in the absence of the external magnetic field, (b) in the presence of an applied perpendicular magnetic field.](image-url)
state along the $y$ direction. In the $B_0 < 0$ case, $\mathcal{H}(k_x, y)$ can be obtained from the one given for $B_0 > 0$ in Eq. (3) by exchanging the off-diagonal elements and replacing $k_x$ by its time reversed counterpart $-k_x$.

The eigenstates for $B_0 > 0$ and $n > 0$ are given by

$$\phi_{k_x, n}^+(y) = \begin{pmatrix} \cos (\delta_n/2) \varphi_{n-1}(y - k_x l_B^2) \\ -i \sin (\delta_n/2) \varphi_n(y - k_x l_B^2) \end{pmatrix}, \quad (6)$$

$$\phi_{k_x, n}^-(y) = \begin{pmatrix} \sin (\delta_n/2) \varphi_{n-1}(y - k_x l_B^2) \\ i \cos (\delta_n/2) \varphi_n(y - k_x l_B^2) \end{pmatrix}, \quad (7)$$

while for $n = 0$ we have

$$\phi_{k_x, 0}(y) = \begin{pmatrix} 0 \\ \varphi_0(y - k_x l_B^2) \end{pmatrix}. \quad (8)$$

In the above $\cos \delta_n = \Delta/\varepsilon_n^+$ and $\varphi_n$ are the one-dimensional harmonic oscillator eigenstates, i.e., $a^\dagger a \varphi_n = n \varphi_n$. We remark that the $n = 0$ eigenstate in Eq. (8) is very special since it is fully spin polarized. This will have important consequences for the magnetic response discussed below.

The eigenvalues associated with $\phi_{k_x, n}^\pm(y)$ eigenstates are given by

$$\varepsilon_n^\pm = \pm \sqrt{n \varepsilon_c^2 + \Delta^2}, \quad n > 0 \quad (9)$$

and for the fully spin-polarized $n = 0$ eigenstates

$$\varepsilon_0 = -\text{sgn} (B_0) \Delta. \quad (10)$$

Note that the form of eigenstates for $B_0 < 0$ is different from that given in Eq. (6) since the Hamiltonian is different in that case.

### III. SPIN SUSCEPTIBILITY AND MAGNETIZATION

The electronic magnetic moment due to spin is proportional to the spin operator

$$\mu_s = -\gamma_s \mathbf{s} , \quad (\gamma_s = -g_s \mu_B / \hbar) \quad (11)$$

Therefore, to calculate the spin part of the magnetic moment for the eigenstates given in the previous section we only need to find the expectation value of the spin operator. A straightforward evaluation using Eqs. (6) shows that all the electronic states within the same LL contribute equally to the magnetization. For each of them we have

$$\mathcal{M}_{x,n}^\alpha = \mathcal{M}_{y,n}^\alpha = 0 \quad (12)$$

![FIG. 2: (Color online) Spin magnetization $\delta M_s = M_s(T, \mu) - M_s(0,0)$ in units of ($\chi_0$, Tesla) as a function of the chemical potential $\mu$ for a nonmagnetic surface ($\Delta_0 = 0$) at $B_0 = 3.0$ Tesla, $g_s = 8$ (top panel) and 30 (bottom panel). $k_B T = 0.1, 1.0, 5.0$ meV (red, green, blue).](image-url)

The total magnetization for each Landau level can be obtained by multiplying the above quantities by the Landau level degeneracy $L^2/(2\pi l_B^2)$, representing the total number of states with characteristic length $l_B$ that the surface area $L^2$ can accommodate. Since the magnetization contribution computed above for each eigenstate is an explicit function of its energy, we can perform the following integral to find the total magnetization density due to the electronic spins

$$M_s = \int d\varepsilon \, D(\varepsilon) \mathcal{M}_s(\varepsilon) n_F(\varepsilon), \quad (15)$$

where $\mathcal{M}_s(\varepsilon) = g_s \mu_B \Delta/(2\varepsilon)$ is the magnetization of the eigenstate with energy $\varepsilon$ and $n_F(\varepsilon) = 1/[e^{(\varepsilon-\mu)/k_B T} + 1]$ is the Fermi-Dirac distribution function. The electronic density of states associated with the surface states is
where the function to perform integration in Eq. (15) yields we assume $\Lambda = 300$ meV. Using this density of states band becomes degenerate with the bulk bands and here the cutoff can be chosen to be the energy where the surface beyond which the energy exceeds the cutoff energy $\Lambda$. The negative behaviour as a function of chemical potential associated with the high-energy cutoff we choose to display it takes the following form

$$\Delta_{0} = -2, -1, 0, 1, 2 \text{ meV and } k_{B}T = 0.1 \text{meV. } g_s = 8 \text{ (top panel) and } 30 \text{ (bottom panel).}$$

$D(\varepsilon)$. For the Hamiltonian we used in the previous section it takes the following form

$$D(\varepsilon) = \frac{1}{2\pi l_{B}^{2}} \left[ \delta(\varepsilon - \varepsilon_{0}) + \sum_{n>0, \alpha=\pm} \delta(\varepsilon - \varepsilon_{n}^\alpha) \right], \quad (16)$$

where $n_{c} \equiv (\Lambda^{2} - \Delta^{2})/\varepsilon_{0}^{2}$ is the Landau level index beyond which the energy exceeds the cutoff energy $\Lambda$. The cutoff can be chosen to be the energy where the surface band becomes degenerate with the bulk bands and here we assume $\Lambda = 300$ meV. Using this density of states function to perform integration in Eq. (15) yields

$$\frac{M_s}{B_0} = \chi_{0} \left[ -n_F(\varepsilon_0) + \text{sgn}(B_0) \sum_{n=1}^{n_{c}} \frac{n F(\varepsilon_{n}^{+}) - n F(\varepsilon_{n}^{-})}{\varepsilon_{n}/\Delta} \right], \quad (17)$$

where $\chi_{0} \equiv (g_s \mu_B)/2\hbar$.

For a constant magnetic field $B_0$ the magnetization of a single TI surface given by Eq. (17) shows an interesting behaviour as a function of chemical potential $\mu$ illustrated in Fig. 2. In order to avoid ambiguity associated with the high-energy cutoff we choose to display $\delta M_s = M_s(T, \mu) - M_s(0, 0)$, i.e. spin magnetization relative to the neutrality point at $T = 0$. For the negative values of $\mu$ magnetization initially decreases reflecting the fact that the negative-energy surface states exhibit negative spin polarization, as can be seen from Eq. (13). The large jump in $\delta M_s$ near $\mu = 0$ results from electrons filling the fully spin-polarized $n = 0$ Landau level. Further increase in $\mu$ results in increase of $\delta M_s$ now reflecting the fact that the positive-energy surface states exhibit positive spin polarization.

The above behavior is unique to topological insulators for it results from the Landau level structure in a single Dirac point (or more generally an odd number thereof). In a TI the ‘other’ Dirac point is located on the opposite surface where the magnetic field points in the direction opposite relative to the surface normal, see Fig. 1. The contribution of this surface to $\delta M_s$ would be the same.

We emphasize that for a relatively thick TI slab $\beta$-NMR will be sensitive to a single surface facing the beam and the behaviour predicted here is in principle observable, except that continuous tuning of the chemical potential in a crystal might be difficult to achieve.

A much more feasible experiment involves varying magnetic field $B_0$ while keeping $\mu$ constant. We now show that a unique signature of the fully polarized $n = 0$ Landau level still exists in samples with intrinsic magnetic ordering, when the chemical potential resides in the gap (i.e. both the bulk and the surface are insulating in the absence of the field). In his situation we can set $\mu = 0$ in our model. Now consider the effect of the applied magnetic field. Since the gap between the adjacent energy levels in which the Fermi energy is located is fairly large ($\sim 100$ K for a 1T field), at sufficiently low temperatures we can replace the Fermi function in Eq. (17) by the step function

$\chi(\varepsilon - \mu) = 1$ for $\varepsilon > \mu$ and $\chi(\varepsilon - \mu) = 0$ for $\varepsilon < \mu$. The step function $\chi(\varepsilon - \mu)$ is the same as the Heaviside step function $H(\varepsilon - \mu)$.

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Another question that arises has to do with the origin of the electrons that fill the $n = 0$ Landau level upon changing the field through $B_c$. One may wonder where the extra electrons come from in a fully gapped isolated system. The answer lies in the side surfaces which under generic conditions remain gapless and act as a reservoir of electrons. The model we consider here does not capture these states but they reflect themselves in solutions of the Hamiltonian which are not normalizable in an infinite system. These are fully spin polarized with opposite energy and spin direction. In fact they are the particle-hole conjugates of the fully spin polarized LL given in Eq. (8). Taking into account these electronic states and the fact that it is more favourable for electrons with higher energy to be transferred to the negative energy states the counting problem can be resolved.

**IV. THE KNIGHT SHIFT**

We now outline how a $\beta$-NMR experiment can in principle be used to probe some of the physics discussed in the previous section. The valence and conduction electrons in a metal possess magnetic moments arising from both their orbital motion and their spin degrees of freedom. Nuclear magnetic resonance technique can be used as a probe of the spin part of the total magnetization in the presence of the magnetic field by measuring the relative shift in the nuclear resonance peak with respect to the same resonance peak in a reference insulating system. This effect, which is due to the interaction between electronic spins and those of the nuclei, is known in the literature as the Knight shift and has been extensively studied in both theory and experiment. The mobile electrons in a metal interact with the nuclei in their proximity and the Knight shift in the resonance peak of these nuclei can be described by a local Fermi contact interaction term given by $H_{\text{int}} = \frac{-2\pi}{\hbar} \mu_e \cdot \sum_j \mu_i \delta(r - R_i)$ where $R_i$ is the position of the $i$th nucleus and $\mu_i = \gamma_N I_i$ is its magnetic moment. The magnitude of $\gamma_N$, the gyromagnetic ratio, depends on the nucleus quantum state. The nucleus total spin, $I_i$, couples to the applied magnetic field and therefore the position of the peak depends on the magnitude of the total magnetic field experienced by the nucleus which has a contribution due to the interaction with electrons. It turns out that this shift is proportional to the spin susceptibility. The constant of proportionality, known as the hyperfine coupling, can be computed using first principle calculations for the implanted nuclei. On the other hand, if we assume that the presence of the nuclei does not significantly alter the electronic states, then it is possible to approximate the shift for them by taking the expectation value of the aforementioned interaction term using the unperturbed electronic states. This is the lowest order approximation in the perturbative treatment of the interaction term. For metallic systems with spin degenerate bands this shift is proportional to the spin

![Diagram](image)
susceptibility as it can be seen from a simple calculation considering the fact that the spatial and spin degrees of freedom are uncorrelated.

The spin-momentum locking on the surface of a TI along with the energy dependence of the penetration depth can in principle change the above simple physics. Since it is not possible anymore to separate spin and orbital degrees of freedom, one might question the validity of the linear relation between the Knight shift and the spin susceptibility. We devote the rest of this section to addressing this issue by considering a very simple model. We assume that the nuclei do not alter the electronic states around them significantly and computing the Knight shift would then require a first principles calculations.

The field experienced by the ith nucleus due to the interaction with the proximate electrons is given by

$$\delta B_i \equiv - \frac{8\pi}{3} \gamma_e \langle \sigma \delta(r - R_i) \rangle_T,$$  \hspace{1cm} (20)

where $\langle \ldots \rangle_T$ is the expectation value of over electronic states at temperature $T$. Therefore, the effective Hamiltonian for the ensemble of nuclei takes the form

$$H_N^{\text{eff}} = -h\gamma N \sum_i I_i \cdot (B_0 + \delta B_i).$$ \hspace{1cm} (21)

This way, ith nucleus would have a resonance peak $\omega_i = \gamma N (B_0 + \delta B_i)$. The Knight shift is then defined by comparing the resonance frequency with the frequency in a similar material without these electronic states

$$K_i = \frac{\omega_i - \omega_0}{\omega_0} = \frac{B_0 + \delta B_i - B_0}{B_0} = \frac{\delta B_i}{B_0}. \hspace{1cm} (22)$$

The shift in the resonance peak of the nuclei ensemble is the average of the Knight shift from each individual nucleus and is given by

$$K = \frac{1}{N} \sum_i \delta B_i,$$ \hspace{1cm} (23)

where $N$ is the number of the implanted nuclei. Using the electronic eigenstates given in Eq. (8) we get the following expression for $K$

$$-\frac{8\pi\gamma_e}{3N B_0} \sum_{i,k,n,\alpha} \langle |\psi_n^\alpha(z_i)|^2 (|\phi_n^\alpha|^2(R_{Li}) - |\phi_{n+1}^\alpha|^2(R_{Li})) \rangle.$$ \hspace{1cm} (24)

Here $|\psi_n^\alpha(z_i)|^2$ appears as a factor in the realistic 3D electronic wave functions of the surface electrons reflecting the fact that the electrons have an energy dependent penetration depth into the bulk. The nuclei implanted in the system have a spatial probability distribution $P_{\text{Nuc}}(z, r_{\perp})$, which depends on the energy and diameter of the beam of the ions used in the $\beta$-NMR experiment. If the distribution function is known, we can replace the above summation with a 3D integral over the crystal volume

$$\frac{1}{N} \sum_i \int \! dz \, d^2 r_{\perp} P_{\text{Nuc}}(z, r_{\perp}).$$ \hspace{1cm} (25)

Assuming that the distribution is uniform in the plane of the surface, i.e., $P_{\text{Nuc}}(z, r_{\perp}) = P(z)$, we get

$$K = \frac{8\pi}{3\pi f_0 \sum_{n,\alpha} f_n^\alpha \cdot M_{z,n}^\alpha}.$$ \hspace{1cm} (26)

where we have performed the integration over the in-plane degrees of freedom and replaced the summation over $k_x$ with the LL degeneracy. We have also defined the nth LL weight, $f_n^\alpha$, as

$$f_n^\alpha = \int \! dz |\psi_n^\alpha(z_i)|^2 P(z).$$ \hspace{1cm} (27)

Now if we assume that different LL have the same penetration depth we have $f_n^\alpha = f_0$ for all $n < n_c$ and we obtain

$$K = \frac{8\pi f_0}{3B_0} \sum_{n,\alpha} M_{z,n}^\alpha = \frac{8\pi f_0}{3} \lambda_n^e.$$ \hspace{1cm} (28)

We thus recover the linear proportionality of the Knight shift to the surface electronic spin susceptibility under reasonable assumptions. Note that if we relax the assumption that different LL can now have different penetration depths, then the Knight shift would no longer be linearly proportional to the total spin susceptibility. Instead, it would be a weighted superposition of contributions from each individual LL to the spin susceptibility. Nevertheless, the Knight shift will still display the interesting behavior discussed in this study as long as $f_n^\alpha$ is a reasonably slowly varying function of $n$. We should emphasize once again that while above considerations elaborate on the differences caused by the spin-momentum locking and the energy dependent penetration depth, they do not take into account the fact that the electronic wave-functions could be altered by the presence of the implanted nuclei and the hybridization with the adjacent nuclei.

V. CONCLUSIONS

The magnetic response of spins of the Dirac-like electrons on the surface of a topological insulator shows interesting features both in the absence and the presence of an intrinsic gap $\Delta_0$. When the surface states are gapped owing to the time-reversal breaking perturbation (i.e. due to magnetic doping) the $n = 0$ Landau level, which is fully spin polarized, can have positive or negative energy depending on the sign of the intrinsic gap $\Delta_0$ relative to
that of the applied magnetic field. It will therefore be completely filled or empty when the chemical potential is tuned to zero energy. Our study shows that this structure results in an observable jump in the spin susceptibility, measurable e.g. through the $\beta$-NMR Knight shift, as one tunes the applied magnetic field through the critical value $B_c$ given in Eq. (15). The effect may be used as a means to measure the magnitude of the intrinsic gap on the surface of a magnetically doped topological insulator if $\beta$-NMR or another surface-sensitive technique could capture the magnetic response of the electronic spins on the surface. This behaviour is a unique feature of the topological insulator exotic surface states closely related to the special form of the spin-momentum entanglement.

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