A thermodynamic measure of the Magneto-electric coupling in the 3D topological insulator

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We show that the magneto-electric coupling in 3D (strong) topological insulators is related to a second derivative of the bulk magnetization. The formula we derive is the non-linear response analog of the Streda formula for Hall conductivity (P. Streda, J. Phys. C: Solid State Physics, 15, 22 (1982)), which relates the Hall conductivity to the derivative of the magnetization with respect to chemical potential. Our finding allows one to extract the magneto-electric coefficient by measuring the magnetization, while varying the chemical potential and one more perturbing field, a unique method never attempted before in the experimental search for the magneto-electric effect. The relation we find also makes transparent the effect of disorder on the magneto-electric response, which occurs only through the density of states, and has no effect when the system is gapped.

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The Quantum Hall effect (QHE) was the first experimental instance where a transport coefficient (the Hall conductivity in $d = 2$) was quantized. Finding an analog system in $d = 3$ had remained an unrealized dream of the condensed matter community, for many years. Recently, however, this has changed. The discovery of the topological insulator (TI) and specifically the 3D strong topological insulator (STI) have finally realized the dream of a $d = 3$ analog of the QHE. In the context of the QHE, Streda et al. proved an insightful relation between the Hall conductivity, and a second derivative of a thermodynamic potential

$$\sigma_{\text{Hall}} = \frac{1}{2} (\sigma_{xy} - \sigma_{yx}) = -\frac{\partial \rho}{\partial B} \bigg|_\mu = -\frac{\partial M}{\partial \mu} \bigg|_B ,$$

where $\rho$ is the charge density, $M$ the orbital magnetization per unit volume (perpendicular to the 2D system), $B$ is the external magnetic field, and $\mu$ the chemical potential. Here and throughout the manuscript we use units where $\hbar = e = \epsilon = 1$.

Motivated by the analogy between the $d = 2$ QHE and $d = 3$ STI, in this paper we show that the magneto-electric effect, most of them relying on the surface states in the STI and on the Witten effect. The magneto-electric effect at the surface appears as a consequence of the material boundary, where $P_3$ has a sharp

$$\chi = \frac{2\pi}{3} \frac{\partial^2 \rho}{\partial B^a \partial (\partial_a \phi)} = -\frac{2\pi}{3} \frac{\partial^2 M_a}{\partial \mu \partial (\partial_a \phi)} .$$

There are a number of merits to this result, similar to those of $\chi$. First, $\chi$ suggests we can measure the Hall conductivity in the QHE, by doing a thermodynamic measurement - vary the chemical potential through a back gate, and measure the magnetization of the sample. Calculating the derivative of the magnetization with respect to the gate voltage should give $\sigma_{\text{Hall}}$. Similarly, with $\chi$ we can measure $\chi$ by simultaneously varying the chemical potential $\mu$ and the gradient $(\partial_a \phi)$, while measuring the magnetization of the sample. Second, the effects of disorder on (1) and (3) are entirely included in the density of states (DOS) $D(\epsilon)$ through the particle density $\rho = \int d\epsilon f(\epsilon) D(\epsilon)$, where $f(\epsilon)$ is the Fermi Dirac distribution. Disorder will reduce the effective gap in the spectrum, compared with the clean limit, but otherwise will not change anything for an insulator, as long as the chemical potential remains in the gap.

The magneto-electric effect is usually formulated as an anomalous term appearing in the action for the electromagnetic fields in an insulator $S_{EB} = \frac{1}{2\pi} \int d\tau d^3 r P_3 E \cdot B$, with $P_3$ the magneto-electric coefficient. Under inversion, $E$ is odd, and under time reversal $B$ is odd, so $P_3$ is odd under both. Ref. showed that $P_3$ takes on values modulo 1, and so in a material with either time reversal or inversion symmetry (or both), it can take on the values $P_3 = 0, \frac{1}{2}$. The value $P_3 = \frac{1}{2}$ then characterizes the STI. This value can in principal be measured as

$$P_3 = \frac{1}{2} = \frac{\partial M_a}{\partial E^b} = \frac{2\pi}{\partial B^a} ,$$

where the Latin letters $a, b = x, y, z$ denote spatial directions. Here and throughout the manuscript we will use the Einstein summation convention.

At this point in time, a number of materials have been identified as topological insulators using spectroscopy to characterize their unique surface states (an odd number of Dirac points). However, thus far measuring the magneto electric coefficient has proved challenging. The materials, by and large, have proven rather poor insulators, with significant carrier concentration, in some cases even a bulk Fermi surface appearing. In a bulk metal DC electric fields are screened, making it impossible to measure directly.

Many other indirect ways have been proposed to detect the magneto-electric effect, most of them relying on the surface states in the STI and on the Witten effect. The magneto-electric effect at the surface appears as a consequence of the material boundary, where $P_3$ has a sharp
that the \( \sigma \) sea) \( \sigma \) surface) \( \sigma \) between contributions from states at the Fermi energy (the Fermi energy is well-defined for both an insulator and a metal because it is defined through a current response to an external field \( \mathbf{J} \). For an insulator, \( \sigma^{I} = 0 \), and the total Hall conductivity is reduced to \( \mathbf{S} \).

The result \( \mathbf{S} \) can be anticipated from the following considerations. In an insulator, with no dissipative currents, the only currents possible are persistent currents related to the orbital magnetization \( \mathbf{M} \). The electric field is found from \( \mathbf{E} = -\nabla \mu(\mathbf{x}) \). Assuming the magnetization is an entirely local function of the intensive thermodynamic quantities \( M_a = M_b(\mathbf{T}, \mathbf{B}, \mu(\mathbf{x})) \), we find

\[
J^a = e^{abc} \partial_b M_c = e^{abc} \frac{\partial M_c}{\partial \mu} \partial \mu = -e^{abc} \frac{\partial M_c}{\partial \mu} E_b ,
\]

resulting in \( \sigma^{xy} = -\frac{\partial M}{\partial \mu} \). The second equality in \( \mathbf{S} \) is due to a Maxwell relation.

As mentioned earlier, \( \mathbf{S} \) indicates the \( \sigma^{II} \) contribution can be found by doing a thermodynamic measurement. However, the measured magnetization will include both the orbital and Zeeman contributions to the magnetization, while \( \mathbf{S} \) involves the orbital magnetization alone. For an insulator one can argue that the magnetization due to Zeeman coupling does not vary with chemical potential, and therefore measuring the derivative of the total magnetization, will give the same result as if we were measuring the orbital magnetization alone. For a metal on the other hand, the Zeeman effect magnetization can depend on the chemical potential, for instance in Pauli paramagnetism. Therefore, measuring the total magnetization will only give a quantitatively accurate measure of \( \sigma^{II} \) in an insulator. Still, it will be useful in finding qualitative differences - it will exhibit quantization when the bulk is gapped. While such a measurement is conceptually straightforward, in practice it is more difficult than measuring Hall effect through electric currents. However, it has been carried out \cite{11,12}.

Next we will present the analog of \( \mathbf{S} \) in the \( d = 3 \) STI.

In order to deal with a possibly gapless spectrum, as well as with disorder, we will have to formulate the magneto-electric coupling in a slightly different way from Refs. \cite{11,12}. The magneto-electric effect relates the magnetization to an applied electric field \( \mathbf{M} = \frac{P_3}{2\pi} \mathbf{E} \) equivalent to \( \mathbf{S} \). The Hall conductivity is well-defined for both an insulator and a metal because it is defined through a current response to an external field \( J_x = \sigma^{xy} E_y \). For an insulator, \( \mathbf{J} = \nabla \times \mathbf{M} \), and using Faraday’s law \( \nabla \times \mathbf{E} = -\partial_t \mathbf{B} \), we get

\[
\mathbf{J} = \frac{1}{2\pi} \nabla P_3 \times \mathbf{E} - \frac{P_3}{2\pi} \partial_t \mathbf{B} .
\]

The only DC (static) response comes from the first term

\[
\mathbf{J} = \frac{1}{2\pi} \nabla P_3 \times \mathbf{E} .
\]
Formulated in this way, we see $P_3$ should be regarded as an external field.

In fact, any inhomogeneous external field $\phi$ with the same symmetry properties of $P_3$, namely odd under inversion and time reversal, should suffice to generate such a current response. We therefore write $J = \frac{\pi}{2}\nabla \phi \times E$, which is just (2). The response coefficient $\chi$ is the generalization of the magneto-electric coupling in the insulator, and a more faithful analog of the Hall conductivity - it can now be defined and calculated for metals as well. For an insulator, $\chi$ will be quantized, yet the precise value of $\chi$ depends on how the field $\phi$ is defined and coupled to the system, and therefore by itself will not attain a universal quantized value. This is the one sacrifice we have to make in the alternate formulation of the magneto electric response. On the other hand, it will prove a more robust quantity to measure, in a system that may be gapless, and most importantly it will exhibit quantization in an insulator - the key qualitative feature we are after.

Next we will derive (3). The derivation in the body of this manuscript is not rigorous, and does not apply to gapless systems. It is presented here for the sake of brevity and clarity. In the supplementary material we will derive the result with some assumptions, while a general rigorous derivation is left for a future publication. Using charge conservation $\partial_t \rho = - \nabla \cdot J$, as well as Faraday’s law $\nabla \times E = - \partial_t B$ we find from (2)

$$\rho = - \frac{\chi}{2\pi} \nabla \phi \cdot B. \quad (9)$$

The second term from (7) has no contribution since $\nabla \cdot B = 0$. Taking $\partial_t \phi = h_c$, the differential form, (9) becomes

$$\chi \delta \phi = - 2\pi \frac{\partial^2 \rho}{\partial B^\mu \partial h_b} = - 2\pi \frac{\partial^2 M_a}{\partial \mu \partial h_b}, \quad (10)$$

where we used the same Maxwell relation as in (3). Note that the magneto-electric response is found from the magnetization parallel to the direction of the auxiliary field gradient. This should not be surprising as the magneto-electric effect should not care about whether the system is isotropic or not. In an isotropic system, the absence of any other directional- ity necessitates this outcome. Contracting the $a, b$ indices in $d = 3$, we arrive at (11).

A rigorous proof of (11), as well as a generalization to gapless systems can be derived with some effort. Following standard response theory technique, we can find the nonlinear response analog of the Bastin Formula

$$\chi^I = \frac{4\pi}{4!} e^{abc} \text{Re} \{ \text{Tr} [\delta(H) (v_b G_R v_\phi - v_\phi G_R v_b) G_R v_c G_R v_a] \}$$

$$\chi^{II} = 2\pi \frac{\epsilon_{\mu \nu \lambda \tau}}{4!} \int_{-\infty}^{+\infty} \frac{d\epsilon}{2\pi i} f(\epsilon) \text{Tr} [v_{\phi} G_{R\mu} G_{R\nu} G_{R\lambda} G_{R\tau} G_{R}] + c.c., \quad (11)$$

where $v_a$ are the velocity operators, $f(\epsilon)$ is the Fermi Dirac distribution, $\delta(x)$ is the Dirac delta function, and $G_R = [\epsilon + i\delta - H]^{-1}$ is the retarded Green’s functions, which can include any random potential. The chemical potential is included in $H = \ldots - \mu$, so that the Fermi energy is at $\epsilon = 0$. The trace is over all degrees of freedom of the system - real (or momentum) space coordinates, and internal degrees of freedom. Also, in the expression for the contribution $\chi^{II}$, all Green’s functions depend on the frequency $\epsilon$, while in the expression for $\chi^I$, all Green’s functions have $\epsilon = 0$. Finally, the velocity $v_{\phi}$ is the conjugate operator to the auxiliary field $H = H_0 + \int \phi(x) v_{\phi}$. The full details of this derivation we leave for a future publication. In the supplementary material we provide a limited derivation, appropriate for an insulator, with the field $\phi = h_a x^a$ coupled to a momentum independent $v_{\phi}$.

Much like in the case of conductivity, the form (11) distinguishes between Fermi surface contributions $\chi^I$, which vanish for an insulator, and the contribution $\chi^{II}$, which turns out to be the second derivative of the orbital magnetization, satisfying (5)

$$\chi^{II} = - \frac{2\pi}{3} \frac{\partial^2 M_a}{\partial \mu \partial h_b} \cdot (12)$$

As we already discussed in the introduction, our result (12), much like the Streda formula (3), indicates that $\chi$ can be obtained by measuring the 2nd derivative of the (orbital) magnetization. As noted earlier, at least in the insulating case, the Zeeman contribution to the magnetization should not vary with the chemical potential, and measuring the full magnetization instead of the orbital magnetization alone, will yield the same result. It is then conceptually straightforward to measure magnetization, and vary the chemical potential. The auxiliary field gradient $h_a$, on the other hand, is at this point an abstract object we defined for our theoretical needs. We turn our attention now to discussing how $\phi$ can be realized. First, given that $\phi$ must be odd under time reversal and inversion, it can appear when anti-ferromagnetic (AFM) order is present in the material. It is not unimaginable that a topological insulator material could be stuffed with magnetic atoms that realize AFM order in the material. Second, we need $\phi$ to vary (slowly) in space, as illustrated in Fig. 1. This can occur naturally in AFM order, as it tends to form magnetic domains. More difficult will be controlling and varying the strength of the AFM field. This can be done by changing the temperature of the system, and for better control of it, to be sufficiently close to the critical temperature of the AFM order. We mention in passing that if the material lacks inversion symmetry, a field breaking time reversal alone should suffice to generate $\phi$. However, a vast majority of the currently known topological insulators are inversion symmetric, and we therefore focus on this case in the current manuscript.

We now turn to a concrete example for realizing $\phi$ as a spatially-varying Zeeman field in Bi$_2$Se$_3$. The effective low energy continuum model derived for Bi$_2$Se$_3$ in Ref. 51 involves electrons in two orbitals, originating in the p-orbitals of different atoms (Bi and Se respectively). As a consequence, the two orbitals in general will have a different gyromagnetic ratio when coupled to a Zeeman field. Indeed, if the magnetic field $b$ is applied in the direction of the trigonal axis of the Bi$_2$Se$_3$ crystal (z-direction in the notation of Ref. 51), one
finds $H_{\text{Zeeman}} = b \sigma_3 (g_0 + g_3 \tau_3)$, where $\sigma_{1,2,3}$ are the Pauli matrices of the electron spin, and $\tau_{1,2,3}$ are the Pauli matrices describing the orbital degree of freedom. If the magnetic field $b$ varies on the length scale of atomic distances, it will effectively break inversion symmetry in the crystal and allow a more general Zeeman coupling to occur

$$H_{\text{Zeeman}} = b_{\text{FM}} \sigma_3 (g_0 + g_3 \tau_3) + b_{\text{AFM}} \sigma_3 (g_1 \tau_1 + g_2 \tau_2). \quad (13)$$

Here $b_{\text{FM}}$ and $b_{\text{AFM}}$ are “ferromagnetic” (FM) and “antiferromagnetic” (AFM) fields, respectively. The field $b_{\text{AFM}}$ is odd under both time-reversal and inversion, and is therefore a suitable realization of $\phi$. Generating it may require antiferromagnetic order, though any magnetic order that varies strongly on microscopic scales (ferrimagnetism, spin spirals etc.) will suffice. We also note in passing that we neglect the orbital coupling of the magnetic field we apply here. With a sufficiently weak Zeeman field the flux through a unit cell of the solid will be small, and we can safely neglect it. Using (11) we calculate $\chi^{II}$ for Bi$_2$Se$_3$, with all the numerical parameters we need (apart from $g_1$) taken from Ref. [51]. Varying the chemical potential, we find the values plotted in Fig. [2].

The most striking feature in plotting $\chi^{II}$ versus chemical potential is the plateau in its value while $\mu$ is in the gap. Once $\mu$ is outside the gap, $\chi^{II}$ is no longer quantized.

In conclusion, we have found that the magneto-electric coupling in topological insulators and their gapless counterparts, can be related to a third derivative of a thermodynamic potential. Most interestingly, this implies that the topological effects could be measured by probing either charge-density or magnetization in equilibrium, and in the bulk, rather than from non-equilibrium transport properties of the surface. Our result (5), suggests a conceptually simple way to measure the magneto-electric response, by measuring magnetization, while varying the chemical potential and the auxiliary field $\phi$, or measuring the charge density while varying the magnetic field and $\phi$. Moreover, our formula holds regardless of whether the system is gapless or gapped, clean or disordered. The measurement we propose, however, is challenging. First and foremost, realizing the auxiliary field is difficult, in the case of Bi$_2$Se$_3$, requiring the introduction of microscopic AFM order to the bulk of the material, and carefully controlling it. Controlling the chemical potential may also be challenging, given that we wish to probe 3D systems. Varying the chemical potential is needed not only to calculate the derivative in (5), but also to detect the most clear cut evidence for a topological state - the plateau in $\chi^{II}$, as illustrated in Fig. [2].

Finally, the magnetization in our formula is the orbital magnetization, ignoring the Zeeman contributions to the magnetization. In a metal, the Zeeman contribution can vary with the chemical potential, but in an insulator, it will not. Therefore measuring the full magnetization, instead of the orbital magnetization, will yield $\chi^{II}$ in the insulating state, but in the metal it will yield $\chi^{II}$ plus some corrections. However, the key qualitative feature is the plateau in $\chi^{II}$ in the insulating state, which will still show up when measuring the total magnetization instead of the orbital magnetization. It is the quantization of a response coefficient that signifies a topological incompressible state. Despite these difficulties, our findings allow a unique conceptual approach to measuring the magneto-electric coupling, and it is our sincere hope this insight will be put to use in the lab.

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\begin{figure}[h!]
\centering
\includegraphics[width=0.4\textwidth]{fig2.png}
\caption{Plot of $\chi^{II}/g_1$ numerically calculated for Bi$_2$Se$_3$. The $\chi^{II}$ value is quantized as long as the chemical potential $\mu$ is in the bulk gap. Once $\mu$ is outside the gap, $\chi^{II}$ is no longer quantized.}
\end{figure}
