Quantum Nonlinear Hall Effect Induced by Berry Curvature Dipole in Time-Reversal Invariant Materials

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Quantum nonlinear Hall effect induced by Berry curvature dipole in time-reversal invariant materials

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It is well-known that a non-vanishing Hall conductivity requires broken time-reversal symmetry. However, in this work, we demonstrate that Hall-like currents can occur in second-order response to external electric fields in a wide class of time-reversal invariant and inversion breaking materials, at both zero and twice the driving frequency. This nonlinear Hall effect has a quantum origin arising from the dipole moment of the Berry curvature in momentum space, which generates a net anomalous velocity when the system is in a current-carrying state. The nonlinear Hall coefficient is a rank-two pseudo-tensor, whose form is determined by point group symmetry. We discus optimal conditions to observe this effect and propose candidate two- and three-dimensional materials, including topological crystalline insulators, transition metal dichalcogenides and Weyl semimetals.

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Introduction—The Hall conductivity of an electron system whose Hamiltonian is invariant under time reversal symmetry is forced to vanish. Crystals with sufficiently low symmetry can have resistivity tensors which are anisotropic, but, Onsager's reciprocity relations [1] force the conductivity to be a symmetric tensor in the presence of time reversal symmetry. Hence, when the electric field is along its principal axes the current and the electric field are collinear, at least to the first order in electric fields. However, this constraint is only about the linear response and does not necessarily enforce the full current to flow collinearly with the local electric field.

In this paper we study a special type of such non-linear Hall-like currents. We will demonstrate that metals without inversion symmetry can have a non-linear Hall-like current arising from the Berry curvature in momentum space. The conventional Hall conductivity can be viewed as the zero order moment of the Berry curvature over occupied states, namely, as an integral of the Berry curvature within the metal's Fermi surface. The effect we discuss here is determined by a pseudo-tensorial quantity that measures a first order moment of the Berry curvature over the occupied states, and hence we call it the Berry curvature dipole. This nonlinear Hall effect has a *quantum* origin arising from the anomalous velocity of Bloch electrons generated by the Berry curvature [2], but it is not expected to be quantized.

In a time reversal invariant system, the Berry curvature is odd in momentum space, $\Omega_a(k) = -\Omega_a(-k)$, and hence its integral weighed by the equilibrium Fermi distribution is forced to vanish, because Kramers pair states at k and $-k$ are equally occupied. However, the second order response is determined by the integral of the Berry curvature evaluated in the *non-equilibrium* distribution of electrons computed to first order in the electric field. Since the non-equilibrium current-carrying distribution is not symmetric under $k \to -k$, the integral of the Berry curvature weighed by it can be finite, leading to a net anomalous velocity and hence a transverse current.

Our study builds upon a seminal work by Moore and Orenstein [3], which predicted a DC photocurrent in quantum wells without inversion symmetry due to the anomalous velocity associated with the Berry phase. The quantum nonlinear Hall effect presented here can be regarded as a generalization of this effect. We predict that an oscillating electric field can generate a transverse current at *both zero and twice the frequency* in two- and three-dimensional materials with a large class of crystal point group symmetries. In particular, the second harmonic generation is a distinctive signature that may facilitate the experimental detection of the quantum nonlinear Hall effect. Additionally, the effect does remain finite in the dc limit of the applied electric field.

General theory—The electric current density is given by the integral of the physical velocity of the electrons, v_a, weighed by their occupation function, $f(k)$:

$$j_a = -e \int_k f(k)\ v_a. \tag{1}$$

For simplicity we imagine a single band system but allow it to be two- or three-dimensional: $\int_k \equiv \int d^dk/(2\pi)^d$. The velocity contains two contributions, namely, the group velocity of the electron wave and the anomalous velocity arising from the Berry curvature [2] ($\hbar = 1$):

$$v_a = \partial_a \epsilon(k) + \varepsilon_{abc}\Omega_b \dot{k}_c, \tag{2}$$

where $\partial_a = \partial/\partial_{k_a}$, ϵ and Ω_b are the energy dispersion and the Berry curvature of the electrons in question:

$$\Omega_a \equiv \varepsilon_{abc}\partial_b A_c,\ \ A_c \equiv -i\langle u_k|\partial_c|u_k\rangle. \tag{3}$$

Within the Boltzmann picture of transport, the canonical momentum of electrons changes in time in response to the external electromagnetic fields. In the absence of external magnetic fields the change of momentum is:

$$\dot{k}_c = -eE_c(t). \tag{4}$$

where $E_c(t) = \Re\{\mathcal{E}_c e^{i\omega t}\}$, with $\mathcal{E}_c \in \mathbb{C}$ the driving electric field which oscillates harmonically in time but is uniform in space. In the relaxation time approximation the Boltzmann equation for the distribution of electrons is [4]:

$$-e\tau E_a \partial_a f + \tau \partial_t f = f_0 - f, \tag{5}$$

where f_0 is the equilibrium distribution in the absence of external fields. We are interested in computing the response to second order in the electric field, hence we expand the distribution up to second order: $f = \Re\{f_0 + f_1 + f_2\}$, where the term f_n is understood to vanish as $\mathcal{E}^n$. One finds a recursive structure:

$$\begin{aligned} f_1 &= f_1^\omega e^{i\omega t},\ f_1^\omega = \frac{e\tau \mathcal{E}_a \partial_a f_0}{1+i\omega\tau}, \\ f_2 &= f_2^0 + f_2^{2\omega} e^{2i\omega t},\ f_2^0 = \frac{(e\tau)^2 \mathcal{E}_a^* \mathcal{E}_b \partial_{ab} f_0}{2(1+i\omega\tau)}, \\ f_2^{2\omega} &= \frac{(e\tau)^2 \mathcal{E}_a \mathcal{E}_b \partial_{ab} f_0}{2(1+i\omega\tau)(1+2i\omega\tau)}. \end{aligned} \tag{6}$$

Writing the current as $j_a = \Re\{j_a^0 + j_a^{2\omega} e^{2i\omega t}\}$, one obtains:

$$\begin{aligned} j_a^0 &= \frac{e^2}{2}\int_k \varepsilon_{abc}\Omega_b \mathcal{E}_c^* f_1^\omega - e\int_k f_2^0 \partial_a \epsilon(k), \\ j_a^{2\omega} &= \frac{e^2}{2}\int_k \varepsilon_{abc}\Omega_b \mathcal{E}_c f_1^\omega - e\int_k f_2^{2\omega} \partial_a \epsilon(k). \end{aligned} \tag{7}$$

The term j_a^0 describes a rectified current while the term $j_a^{2\omega}$ describes the second harmonic. The second terms that appear in Eq. (7) are completely semiclassical and do not require the presence of Berry curvature. However, within the approximation of a constant τ, one finds that these non-linear terms are proportional to the integral of a three-index tensor, $\partial_a \epsilon(k) \partial_{bc} f_0(k)$, which is odd under time reversal and hence they are forced to vanish. Therefore the only surviving terms are those associated with the Berry curvature. By writing $j_a^0 = \chi_{abc} \mathcal{E}_b \mathcal{E}_c^*$, $j_a^{2\omega} = \chi_{abc} \mathcal{E}_b \mathcal{E}_c$, one has:

$$\chi_{abc} = \varepsilon_{adc} \frac{e^3\tau}{2(1+i\omega\tau)} \int_k (\partial_b f_0)\Omega_d. \tag{8}$$

An expression essentially equivalent to that above was obtained in Ref. [7] by Deyo, Golub, Ivchenko and Spivak. The presence of the factor $\partial_b f_0$ will gurantee that only states close to the Fermi surface will contribute to the integral in the low temperature limit, so that this response is a Fermi liquid property [8]. Equation (8) can be rewritten as follows:

$$\chi_{abc} = -\varepsilon_{adc} \frac{e^3\tau}{2(1+i\omega\tau)} \int_k f_0\ (\partial_b \Omega_d). \tag{9}$$

This expression (9) for the nonlinear conductivity tensor, χ_{abc}, is the first main result of this work. It shows that χ_{abc} is proportional to the *dipole moment* of the Berry curvature over the occupied states, defined as:

$$D_{ab} = \int_k f_0\ (\partial_a \Omega_b). \tag{10}$$

It is interesting to note that this tensor is dimensionless in three dimensions. At frequencies above the width of the Drude peak $\omega\tau \gg 1$ and below the interband transition threshold, the prefactor in χ_{abc} becomes independent of the scattering time, so that χ_{abc} directly measures the quantum geometry of the Bloch states. In the dc limit or for linearly polarized electric fields, the Berry curvature dipole term always produces a current that is orthogonal to the electric field $j_a E_a = 0$ [25].

To close this section, we wish to remark that there exist additional second order corrections to the current arising from modifications to Eq. (2) that are intrinsic to the band structure, containing no powers of the scattering time τ [5], however, these contributions vanish for time reversal invariant systems. Other type of rectifications might arise in systems with an inversion asymmetric scattering rate, namely when the scattering from k to k' has a different rate than that from $-k$ to $-k'$, which produces a kind of ratchet effect [6]. These semiclassical Berry-phase independent contributions are distinguished from the quantum nonlinear Hall effect discussed in this work because they are expected to scale as τ^2.

Berry curvature dipole in three dimensions—Let us explore the constraints imposed by crystal point symmetries on the Berry curvature dipole tensor D_{ab}. A point symmetry is described by an orthogonal matrix S. Because the Berry curvature is a pseudovector, the Berry curvature dipole transforms as a pseudotensor. Hence, crystal symmetries impose constraints of the form:

$$D = \det(S) S D S^T. \tag{11}$$

To determine which components of this tensor are non-zero it is convenient to decompose it into symmetric and antisymmetric parts: $D^\pm = (D \pm D^T)/2$, which transform independently under symmetry operations. The antisymmetric part of a pseudotensor transforms as a *vector*, as can be verfied from Eq. (11). The components of this vector can be taken to be $d_a \equiv \epsilon_{abc} D^-_{bc}/2$. Therefore for it to be non-zero the crystal must have a polar axis. From the 32 crystallographic point groups, 10 allow for a polar axis, namely $\{C_n, C_{nv}\}$ with $n = 1, 2, 3, 4, 6$. The vector d_a will be oriented along such axis. The contribution to the current from this antisymmetric part can be written in vector notation as:

$$\begin{aligned} \vec{j}^0 &= \frac{e^3\tau}{2(1+i\omega\tau)} \vec{\mathcal{E}}^* \times (\vec{d} \times \vec{\mathcal{E}}), \\ \vec{j}^{2\omega} &= \frac{e^3\tau}{2(1+i\omega\tau)} \vec{\mathcal{E}} \times (\vec{d} \times \vec{\mathcal{E}}). \end{aligned} \tag{12}$$

Let us now determine which crystals allow for a non-zero symmetric part D^+. We require the crystal to be

inversion asymmetric for otherwise the Berry curvature would be identically zero due to time reversal symmetry. Any real symmetric matrix can be diagonalized and has a real spectrum. Let us denote its eigenvalues and eigenvectors by δ_i, $\mathbf{e}_i$ respectively: $D^+ = \sum_{i=1}^{3} \delta_i \mathbf{e}_i \mathbf{e}_i^T$. All inversion asymmetric crystals without left-handed symmetries allow for D^+ to be non-zero, but might impose constraints on its eigenvectors to lie along the principal symmetry axis and some of its eigenvalues to be degenerate, much in the same way they constrain an ordinary tensor. Such non-centrosymmetric crystal point groups without left-handed symmetries are $\{O, T, C_1, C_n, D_n\}$ with $n = 2, 3, 4, 6$.

However under left-handed symmetries ($\det S = -1$) the transformations of D^+ differ from those of an ordinary tensor. Equation (11) implies that under a left-handed symmetry operation the spectrum goes to minus itself: $\{\delta_1, \delta_2, \delta_3\} \to \{-\delta_1, -\delta_2, -\delta_3\}$. Therefore, for it to remain invariant as a set, it must have the form: $\{\delta_1, \delta_2, \delta_3\} = \{\delta, 0, -\delta\}$. In such case the eigenvectors would transform as $S\mathbf{e}_1 = \pm\mathbf{e}_3$, $S\mathbf{e}_3 = \pm\mathbf{e}_1$, $S\mathbf{e}_2 = \pm\mathbf{e}_2$. Therefore, any crystal with a left-handed symmetry and an n-fold rotation axis with $n \geq 3$ will force the tensor D^+ to identically vanish, since such n-fold rotation would additionally force the eigenvectors contained within the invariant plane to be degenerate. For a mirror symmetry, the null eigenvector has to be parallel to the mirror plane, and the eigenvectors with opposite eigenvalues must be at $\pi/4$ angles from such plane, so that they are swapped under the mirror operation. Therefore, the only non-centrosymmetric crystals with mirror symmetries that allow for a non-zero D^+ are C_{1v} and C_{2v} [26]. For C_{1v} symmetry D^+ has two independent parameters which can be taken to be the positive eigenvalue and the orientation of the null eigen-vector within the mirror plane. For C_{2v} there is only one independent parameter, which can be taken to be the positive eigenvalue, since the null eigenvector is forced to lie along the rotation axis. Finally, the point group S_4, which contains a single left-handed four-fold roto-reflection symmetry, allows for a non-zero D^+, whose null eigenvector is forced to lie along the rotoreflection axis. D^+ has two independet parameters for S_4, which can be taken to be the positive eigenvalue and the orientation of the corresponding eigenvector within the roto-reflection plane.

Berry curvature dipole in two dimensions—In two-dimensional crystals the Berry curvature behaves as a pseudoscalar (only the out-of-plane component is non-zero), hence the Berry curvature dipole behaves as a *pseudo-vector* contained in the two-dimensional plane:

$$D_a = \int_k f_0 \, (\partial_a \Omega_z). \tag{13}$$

This vector has units of length. Therefore symmetry constraints are more severe in two-dimensions. In fact, the largest symmetry of a 2D crystal that allows for a non-vanishing Berry curvature dipole is a single mirror line (i.e. a mirror plane that is orthogonal to the 2D crystal), which would force D_a to be orthogonal to it. In vector notation the current can be written as:

$$\begin{aligned} \vec{j}^0 &= \frac{e^3\tau}{2(1+i\omega\tau)} \hat{z} \times \vec{\mathcal{E}}^* (\vec{D} \cdot \vec{\mathcal{E}}), \\ \vec{j}^{2\omega} &= \frac{e^3\tau}{2(1+i\omega\tau)} \hat{z} \times \vec{\mathcal{E}} (\vec{D} \cdot \vec{\mathcal{E}}). \end{aligned} \tag{14}$$

The presence of a single mirror symmetry would force the linear conductivity tensor to have its principal axes aligned with the mirror line. Consequently, according to Eq.(14), when the driving electric field is aligned with the direction of the Berry curvature dipole vector, $\vec{D}$, all the current that flows orthogonal to it would arise solely from the Berry curvature dipole term.

Candidate materials—Berry curvature often concentrates in small regions in momentum space where two or more bands cross or nearly cross. Therefore, Dirac and Weyl materials are excellent candidates to observe the quantum nonlinear Hall effect predicted in this work. Moreover, since this effect requires a Berry curvature dipole, it is advantageous to choose low-symmetry crystals with *tilted* Dirac or Weyl point (see below). We propose three classes of candidate materials: topological crystalline insulators, two-dimensional transition metal dichalcogenides, and three-dimensional Weyl semimetals.

The surface of topological crystalline insulators (TCIs) hosts massless Dirac fermions protected by mirror symmetries [9, 10]. In particular, the [001] surface of TCIs SnTe, $Pb_{1-x}Sn_xTe$ and $Pb_{1-x}Sn_xSe$, hosts four massless Dirac fermions [11] protected by *two* mirror symmetries. Pairs of Dirac cones with spin-momentum locking are located near the $\bar{X}$ points of the surface Brillouin zone, forming a Kramers pair. At low temperatures the surface undergoes a structural transition into a ferroelectric state and one of the mirror symmetries is spontaneously broken [12, 13], while the other remains intact. As a result two of the surface Dirac cones become massive, while the other two remain massless [14] (see inset in Fig. 1). Since the remaining massless Dirac points have vanishing Berry curvature, it is sufficient to consider the contribution to the Hall current from the two Dirac points that become massive in the distorted crystal structure. They acquire Berry curvatures of opposite signs, because they are mapped into one another by time reversal symmetry. The low energy Hamiltonian for the massive Dirac point located at momenta $\pm\Lambda$ away from $\bar{X}_1$ is given by:

$$H_{s\Lambda} = v_x k_x \sigma_y - s v_y k_y \sigma_x + s\alpha k_y + \beta \sigma_z. \tag{15}$$

where $s = \pm 1$. β is the size of the gap opened by the ferroelectric distortion. This low energy theory coincides with that previously considered in the literature [11, 12, 14], except for the term proportional to α which produces a tilt in the Dirac cones. This tilt is allowed by symmetry

and has been observed in ARPES studies [15], and is required for a nonzero Berry curvature dipole (see below). The dispersion relation for the titled Dirac cone is:

$$\varepsilon_s(k) = s\alpha k_y + \mathrm{sign}(\mu)(\beta^2 + v_x^2 k_x^2 + v_y^2 k_y^2)^{1/2}, \quad (16)$$

where $\mu > 0$ ($\mu < 0$) for conduction (valence) band. The Berry curvature can be found, from Eq. (3), to be:

$$\Omega_s = \frac{\mathrm{sign}(\mu)}{2} \frac{s v_x v_y \beta}{(\beta^2 + v_x^2 k_x^2 + v_y^2 k_y^2)^{3/2}}. \quad (17)$$

At zero temperature the Berry curvature dipole, computed from Eq. (13), reduces to an integral over the region $\varepsilon_s(k) < \mu$. This integral can be computed by performing an area preserving transformation: $k'_y = \sqrt{v_y/v_x}k_y$, $k'_x = \sqrt{v_x/v_y}k_x$, and by noting that the Fermi surface is an ellipse in the primed coordinates: $k_x'^2/\gamma_x^2 + (k'_y + sk_0)^2/\gamma_y^2 = 1$. Where $\gamma_x = \gamma/v$, $\gamma_y = \gamma/\sqrt{v^2 - \alpha'^2}$, $k_0 = \mu\alpha'/(v^2 - \alpha'^2)$, $v = \sqrt{v_x v_y}$, $\gamma = \sqrt{\mu^2 + (v^2 - \alpha'^2)k_0^2 - \beta^2}$, $\alpha' = \alpha\sqrt{v_x/v_y}$. The condition $v^2 > \alpha'^2$ is equivalent to $v_y^2 > \alpha^2$ and is needed for the stability of the tilted Dirac cones. The condition $\mu^2 + (v^2 - \alpha'^2)k_0^2 > \beta^2$ states that the chemical potential is outside the gap, so that there is a finite density of massive Dirac fermions.

The surviving mirror symmetry, that takes $k_y \to -k_y$, dictates that only the y-component of the Berry curvature dipole is non-zero, and is found to be:

$$D_y = \frac{3v^2 n\beta\alpha|\mu|(1+u^2)}{\left[\mu^2(1+u^2)(1+2u^2) - u^2\beta^2\right]^{5/2}}, \quad (18)$$

where $u = \alpha'/\sqrt{v^2 - \alpha'^2}$ and $n = \int_{|\varepsilon_s(k)|<|\mu|} d^2k/(2\pi)^2 = \gamma_x\gamma_y/4\pi$ is the absolute value of the carrier density in each of the massive Dirac cones [27]. Each massive Dirac cone produces an identical contribution to D_y, giving rise to a factor of 2 already included in Eq. (18). This dipole is orthogonal to the ferroelectric displacement direction in our convention. The Berry curvature dipole has the same sign for electrons and holes in this system and vanishes when the chemical potential is in the gap of the massive Dirac fermions. The typical scale of D_y for SnTe TCI is $\hbar\alpha/\beta \sim 3$nm, where we used a Fermi velocity of $v_x \approx v_y \approx 4 \times 10^5$m/s [24], $\beta \approx 10$meV and $\alpha = 0.1v_x$ [28]. The behavior of D_y is depicted in Fig. 1.

Another candidate 2D materials to observe the quantum non-linear Hall effect are monolayer transition-metal dichalcogenides (TMDC). Their large spin-orbit-coupling and lack of an inversion center produces substantial local Berry curvatures [16, 17]. The C_{3v} symmetry of these crystals would force the Berry curvature dipole to vanish. However, uniaxial strain can reduce this symmetry so that a single mirror operation survives, in which case the effect is allowed. In fact, two copies of the model of Eq. (15), each with a different gap, can describe the states

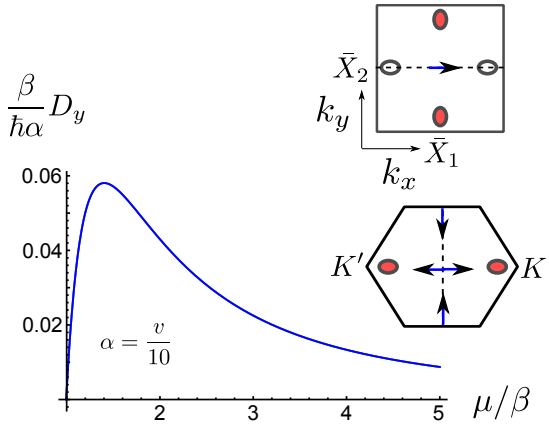

FIG. 1: (color online) Berry curvature dipole dependence on chemical potential μ. Upper inset: surface Brillouin zone of TCI SnTe or (Pb,Sn)Se. The blue arrow indicates the direction of the ferro-electric distortion. Lower inset: Brillouin zone of monolayer TMDC. Dirac points are shifted away from K and K' by shear strain along the directions indicated by the blue arrows. Red circles and dashed lines indicate massive Dirac points and the surviving mirror symmetry respectively.

near charge neutrality within a $k\cdot p$ model [16], and when the shear strain is applied along high-symmetry lines (see inset of Fig. 1). $s = \pm$ would label valleys K and K' in this case. The anisotropic velocity term parametrized by α would be proportional to the strain, much in the same way as in strained graphene [18]. For TMDCs one obtains a scale $\hbar\alpha/\beta \sim 0.2$Å, using a Fermi velocity of $v \approx 4.5 \times 10^5$m/s, a gap $\beta \approx 1.5$eV and $\alpha = 0.1v$.

Last but not least, the Berry curvature dipole induced non-linear Hall effect should be present in a large class of three-dimensional non-centrosymetric crystals. Interesting candidates are the recently discovered Weyl semimetals in the TaAs material class [19–22]. These materials are non-centrosymmtric and have a polar axis, which allows the quantum nonlinear Hall effect described by Eq. (12). When tilted, a Weyl point generates a singular configuration of Berry curvature, with a finite dipole moment whose magnitude can be easily estimated from band structure calculations. In addition, other polar materials such as BiTeI with a strong Rashba-type spin-orbit coupling [23] may also have large Berry curvature dipole moments. These three-dimensional Weyl and Rashba materials provide promising platforms for the observation of the quantum nonlinear Hall effect.

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[25] Note that in the pure dc case, writing $E_c(t) = \mathcal{E}_c$, with $\mathcal{E}_c \in \mathbb{R}$, the current is $j_a = 2\lim_{\omega\to 0} j_a^0$.

[26] Although C_{2h} has only two-fold rotations, the fact that the rotation axis is perpendicular to the mirror plane forces all the elements to vanish.

[27] n is not the full carrier density since there will also be a contribution from the massless Dirac cones.

[28] For an area of $1mm^2$ and radiation of 1 Watt one gets typical currents of 20 nA for parameters of SnTe.