Theory of Nonlinear Response for Charge and Spin Currents

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The nonlinear Hall effect, which is the second-order harmonic charge Hall effect from the Berry curvature dipole in momentum space, has received much attention recently. As the responses to higher harmonics of the driving ac electric field are prominent and measurable, we develop a general nonlinear theory by taking the charge and spin currents as well as the longitudinal and transverse effects into account. We introduce the expansion order of the electric field and Berry curvature multipole moment, where the Berry curvature dipole is a particular one manifesting itself at the second harmonic order and the second expansion order of the electric field. There are four cases with conserving or breaking the time-reversal symmetry (TRS) and inversion symmetry (IS). We find a specific “selection rule” that only longitudinal odd harmonic order charge currents exist for conserving both the TRS and IS, and with breaking both symmetries, all harmonic order charge and spin currents are nonzero. With conserving TRS and breaking IS, the charge Hall current exists at even harmonic order, and the longitudinal charge current occurs at odd harmonic order. Only the longitudinal spin current survives at even harmonic order. With breaking TRS and conserving IS, only odd harmonic order charge and spin currents can appear. Moreover, we observe that every harmonic order charge current contains a series of infinite-order expansion of the electric field. We further show that the Berry curvature dipole and quadrupole can be determined by measuring the second and fourth harmonic order currents in experiments. This may open a way to explore the higher responses of an ac driving system.

Introduction.— The Hall effect is closely related to the topological properties of condensed matter [1–3]. It describes that a transverse charge current can be induced by a longitudinal applied electric field \(E\) in a perpendicular external magnetic field \(B\). The induced charge current is a linear response to \(E\) on account of breaking the time-reversal symmetry (TRS). For a TRS system, as the Berry curvature is odd in momentum space \([3]\), \(\Omega(\vec{k}) = -\Omega(-\vec{k})\), its integral weight by the equilibrium Fermi distribution is forced to vanish. Thus, the linear Hall current disappears because of the limitation of the TRS. However, it was proposed that a nonlinear charge Hall effect, i.e. the second-order harmonic charge Hall (2HCH) current from the Berry curvature dipole (BCD) in momentum space can exist in the broken inversion symmetry (IS) \([4–6]\), which has been experimentally confirmed \([7, 8]\). Recently, the 2HCH effect has been studied in many materials, such as Weyl semimetals \([6–10]\), topological insulators \([11, 12]\), transition metal dichalcogenides family \([13, 14]\), strained graphene \([15]\), ferromagnetic materials with \(T_{1/2}\) symmetry \([16]\), ferroelectric metals \([17–21]\) and piezoelectric-like device \([22]\). Similarly, as the linear response of the spin Hall effect is connected with Berry phase \([23, 24]\), the nonlinear spin Hall effect, i.e. the second-order harmonic spin Hall effect (2HSH) is also discussed in the transition metal dichalcogenides \([25]\), topological Dirac semimetals \([26]\), and two-dimensional (2D) Rashba-Dresselhaus system \([27, 28]\), etc.

Within the linear response theory, Onsager \([29]\), Kubo \([30]\), and Luttinger \([31]\) explained how to relate the coefficients of the driving fields to microscopic quantities, but the higher-order terms are less clear. Since the second order response can be measured by observing the response signal with 2\(\omega\) frequency (\(\omega\) is the frequency of the external field) by a lock-in amplifier in a phase sensitive way \([7]\), the other order signals with \(n\omega\) frequency can be measured as well. It is desired to study the higher orders even to the infinite order response for charge and spin transport either in longitudinal or in transverse direction. On one hand, the infinite-order expansion exposes the general hierarchical structure of response theory, especially by symmetry regulation. On the other hand, the higher order response shows the sensitivity of nonlinear transport clearly, which is more helpful for us to understand the distribution of Berry curvature in \(k\) space (Berry curvature dipoles, quadrupoles, and so on). Therefore, we present a general formalism for the nonlinear responses of charge and spin current by introducing harmonic order and the expansion order of the electric field. Conserving or breaking the TRS or IS, we discuss four cases and demonstrate the conditions for existence or disappearance of the even or odd harmonic order charge and spin currents. We find every harmonic order contains an infinite expansion order of the electric field in which the lowest expansion order of the electric field has the same order as that of the harmonic order. To characterize this, we introduce the Berry curvature multipole moment in
If we define a scalar $\Lambda_n$ where is the Levi-Civita symbol, and the second term of (Eq. 5) is $j^{\text{dc}}_a = \sum_{m=0}^{\infty} j_m^{\text{dc}}(E^m) = \sum_{m=0}^{\infty} e \int \mathbf{j}_m^{\text{dc}} \mathbf{F}_m$, which is independent of $E$ and should disappear at equilibrium. However, the other terms contain higher orders of $E$. For example, the second term is proportional to $E^2$, which means a nonlinear dc response (the second expansion order of the electric field) due to the ac driving external electric field and was obtained previously [5]. One may note that even in this case ($m = 0$) there are higher expansion orders of the electric field dc charge current up to infinite. Equation (5) is also valid for $E = 0$, where all components of current are zero. For $m = 1$ (the first harmonic order), the first expansion order of the electric field is the $m$-th harmonic order current in the form of

$$j^{m\omega} = \frac{e^2}{2\hbar} \left[ \hat{z} \times \hat{E}(\Lambda_{m-1}) + \hat{z} \times \hat{E}^* (\Lambda_{m+1}) \right] - e \hat{V}_m. \quad (4)$$

The first term in square bracket is the $m$-th harmonics ac Hall current density (i.e., transverse current), which we call $j^{m\omega}_L$. The second term is the longitudinal current, which we refer to as $j^{m\omega}_N$. Next we introduce the expansion order of the electric field current for the $m$-th harmonic order current in the form of

$$j^{m\omega}_m = j^{m\omega}_a (E^m) + j^{m\omega}_a (E^{m+2}) + j^{m\omega}_a (E^{m+4}) + \cdots. \quad (5)$$

It shows that the $m$-th harmonic order current density contains an infinite order of the electric field (the lowest expansion order of the electric field is the $m$-th) current density. For $m = 0$ (the zeroth harmonic order), the first term in Eq. (5) is $j^{0\omega}_a (E^0) = -\sum_{I} e \int \mathbf{j}_m^{\text{dc}} \mathbf{F}_m$, which is independent of $E$ and should disappear at equilibrium. However, the other terms contain higher orders of $E$. For example, the second term is proportional to $E^2$, which means a nonlinear dc response (the second expansion order of the electric field) due to the ac driving external electric field and was obtained previously [5]. One may note that even in this case ($m = 0$) there are higher expansion orders of the electric field dc charge current up to infinite. Equation (5) is also valid for $E = 0$, where all components of current are zero. For $m = 1$ (the first harmonic order), the first expansion order of the electric field is the $m$-th harmonic order current in the form of

$$j^{m\omega}_m = j^{m\omega}_a (E^m) + j^{m\omega}_a (E^{m+2}) + j^{m\omega}_a (E^{m+4}) + \cdots. \quad (5)$$

This equation describes the connection between the $m$-th harmonic order spin current density and the expansion order of the electric field spin current density.

Symmetry analysis.— let us consider the general form of the charge [Eq. (3)] and spin current [Eq. (6)]. We first consider the Hamiltonian with the TRS, satisfying $H(k, \sigma) = H(-k, -\sigma)$, and every eigenstate has a
TABLE I. A “selection rule” for charge and spin currents in four symmetry cases.

| Case | TRS | IS | Charge current | Spin current |
|------|-----|----|----------------|--------------|
| I    | Yes | No | $j_{a}^{2m\omega} = \mathcal{J}_{L_a}^{2m\omega}$, $j_{a}^{(2m+1)\omega} = \mathcal{J}_{L_a}^{(2m+1)\omega}$ | $\mathcal{J}_{a,s_{a}}^{2m\omega} = \sum \mathcal{J}_{k} \mathcal{J}_{a,s_{a}}^{2m\omega}$, $j_{a}^{(2m+1)\omega} = 0$ |
| II   | No  | Yes| $j_{a}^{2m\omega} = 0$, $j_{a}^{(2m+1)\omega} = \mathcal{J}_{L_a}^{(2m+1)\omega}$ | $\mathcal{J}_{a,s_{a}}^{2m\omega} = 0$, $j_{a}^{(2m+1)\omega} = \sum \mathcal{J}_{k} \mathcal{J}_{a,s_{a}}^{2m\omega}$ |
| III  | Yes | Yes| $j_{a}^{2m\omega} = 0$, $j_{a}^{(2m+1)\omega} = \mathcal{J}_{L_a}^{(2m+1)\omega}$ | $\mathcal{J}_{a,s_{a}}^{2m\omega} = 0$, $j_{a}^{(2m+1)\omega} = 0$ |
| IV   | No  | No | $j_{a}^{m\omega} = \mathcal{J}_{L_a}^{m\omega}$ | $\mathcal{J}_{a,s_{a}}^{m\omega} = \sum \mathcal{J}_{k} \mathcal{J}_{a,s_{a}}^{m\omega}$ |

TRS partner that carries opposite momentum and opposite spin. The Berry curvature $\Omega_{b}$ and the group velocity $v_{k}^{a}$ are thus both odd ($\Omega_{b} \rightarrow -\Omega_{b}$; $v_{k}^{a} \rightarrow -v_{k}^{a}$). The distribution function $f_{b}^{a} \propto (\mathcal{E}_{b}\partial_{\mathcal{E}_{b}})^{n} f_{a}$ and $\mathcal{J}_{a}^{m\omega}$ is even (odd) for even (odd) $n$. The even and odd harmonic order charge current give different results

$$j_{a}^{2m\omega} = \mathcal{J}_{L_a}^{2m\omega}, \quad j_{a}^{(2m+1)\omega} = \mathcal{J}_{L_a}^{(2m+1)\omega}.$$  \hspace{1cm} (8)

A remarkable conclusion can be drawn from Eq. (8) that only even harmonic order Hall currents and odd harmonic order longitudinal currents exist in the presence of TRS. The so-called nonlinear Hall current (i.e., the 2HCH current) due to the contribution of the Berry curvature dipole [5, 6] is just the leading-order non-vanishing term in the total charge Hall current, i.e., the first equation in Eq. (8). Similarly, under the TRS the spin current operator remains intact ($\mathcal{J}_{a,s_{a}} \rightarrow \mathcal{J}_{a,s_{a}}$), and we get

$$j_{a}^{2m\omega} \neq 0, \quad j_{a}^{(2m+1)\omega} = 0.$$  \hspace{1cm} (9)

Comparing Eq. (8) with Eq. (9), one may uncover that the odd-order distribution function, i.e., $\mathcal{J}_{2\ell+1}^{m\omega}$, where $\ell$ is an integer, only contributes to the charge current; whereas the even-order distribution function, i.e., $\mathcal{J}_{2\ell}^{m\omega}$, only contributes to the spin current under the TRS. If we focus on the longitudinal charge and spin current, we find that the disappearance and appearance of the charge and spin current are just opposite, occurring in alternative odd and even harmonic order ways[27].

We discuss the second case in which the Hamiltonian satisfies $H(k, \sigma) = H(-k, \sigma)$ under the IS. The Berry curvature $\Omega_{b}$ is conserved ($\Omega_{b} \rightarrow \Omega_{b}$), and we have

$$j_{a}^{(2m+1)\omega} = j_{L_a}^{(2m+1)\omega} + j_{L_a}^{(2m+1)\omega} \neq 0, j_{a}^{2m\omega} = 0.$$  \hspace{1cm} (10)

The spin current density can be obtained

$$j_{a}^{2m\omega} = 0, \quad j_{a}^{(2m+1)\omega} \neq 0.$$  \hspace{1cm} (11)

If the TRS and IS exist simultaneously, the Berry curvature and the spin current operator must disappear throughout the Brillouin zone. Hence there is no Hall phenomenon and only the longitudinal current is left, yielding

$$j_{a}^{(2m+1)\omega} = j_{L_a}^{(2m+1)\omega} \neq 0.$$  \hspace{1cm} (12)

According to the above analysis, Table I summarizes a “selection rule” for the charge and spin current in four cases with different symmetries. It can be seen that case I is just the case discussed in Refs. [5, 6].

A tilted 2D massive Dirac model.— Now we illustrate this general theory by studying a tilted 2D massive Dirac
where \( E \) is up to the fourth harmonic order \( \{ \text{model} \} \). The Hamiltonian is instructive to study the case in a weak external electric field. For each \( 2m \)-th harmonic order charge current density, it generally contains infinite terms of expansion order of the electric field charge current density whose order is larger than and equal to \( 2m \). However, after neglecting the higher expansion order of the electric field charge current density, we only keep the \( 2m \)-th expansion order of the electric field (the leading order in this case), and have [34]

\[
\hat{\mathcal{J}}_{y}^{2m} \propto \mathcal{E}_{x}^{2m} \sum_{I} \int_{k} \left( \partial^{2m-1} \Omega_{i}^{j} \right) f_{I}^{0}. \tag{15}
\]

It says that any \( 2m \)-th (even) harmonic order current density is mainly contributed from \( 2m \)-th expansion order of the electric field charge current density, which is induced by a \( 2m \)-th multipole moment of Berry curvature (the integration). The higher multipole moment of Berry curvature is introduced in this work and emerges naturally. For a weak driving electric field, this multipole moment of Berry curvature can be explored in principle by measuring the higher harmonic order response charge current in experiments.

When the driving electric field is in the \( x \) direction and only a single mirror symmetry exists, the BCD and BCQ are forced to be orthogonal to the mirror line [5, 6]. Meanwhile, the charge current in Eq. (14) can be expressed in vector notations as

\[
\hat{\mathcal{J}}_{y}^{2m} \propto \mathcal{E}_{x}^{2m} \sum_{I} \int_{k} \left( \partial^{2m-1} \Omega_{i}^{j} \right) f_{I}^{0}. \tag{15}
\]

In addition to the band dispersion, Berry curvature, BCD, BCQ and their density distributions in momentum space are shown in Fig. 1. If there is no titling \( (t = 0) \), the BCD [6] and BCQ are both zero [see Figs. 1(c) and (e)], which is due to the symmetric band [Fig. 1(a)]. For nonzero titling, a negative peak of BCD [Fig. 1(c)] appears near the conduction band edge \( (E_{f} \approx 0.12 \text{ eV}) \), while the BCQ [Fig. 1(e)] displays a positive peak \( (E_{f} \approx 0.10 \text{ eV}) \) followed by a negative one \( (E_{f} \approx 0.21 \text{ eV}) \). The different features are mainly caused by different distributions of the BCD and BCQ densities in \( k \) space [Figs. 1(d) and (f)]. Although both have the symmetry of \( C_{2z} \), the BCD has one node at the center of the Dirac cone \( (k = 0) \) as it contains only the first derivative of Berry curvature to \( k_{z} \). Analogously, the BCQ develops three nodes which are distributed symmetrically around \( k = 0 \) since it contains the third derivative of Berry curvature. We emphasize that the BCD and BCQ characterized by \( \vec{D} \) and \( \vec{Q} \) in a vector form in Eq. (16) can be directly determined by measuring the second and fourth harmonic order current density in experiments.
Figure 2 shows the Fermi energy $E_f$ dependence of the 2HCH current density in Fig. 2(a) and the second harmonic order longitudinal spin current density in Fig. 2(b). For small $\omega \tau$ (slow field, dirty limit), the 2HCH current density [Fig. 2(a)] develops a negative and a positive peak with increasing $E_f$. The negative peak is due to the effect of BCQ around the bottom of the conduction band. With increasing $\omega \tau$ to larger than 1 (fast field, clean limit), the negative peak gradually shrinks and disappears eventually, indicating a weakening effect of BCQ for larger $\omega \tau$. Another feature is that the 2HCH current tends to be independent of $\omega$ for very small $\omega \tau$. For the spin current, the model shows that there is only longitudinal even harmonic order spin current density and the transverse even harmonic order, and longitudinal and transverse odd harmonic order spin current densities are all zero. The reason is that the broken inversion symmetry due to tilting in the $x$ direction only releases the spin degeneracy in this direction, leading to the nonvanishing nonlinear longitudinal spin current (even harmonic order). Figure 2(b) only shows the nonvanishing second harmonic order longitudinal spin current densities varying from a zero value ($E_f$ in the band gap) to a positive peak ($E_f$ in conduction band). With further increasing $E_f$, it turns negative and gradually saturates. The saturation is due to a full effect of spin degeneracy breaking. Increasing $\omega \tau$ leads to an enhancement of this positive to negative transition.

Conclusions and Discussions.—We develop a theory of nonlinear responses driven by an ac electric field for charge and spin transport in the presence of nonvanishing Berry curvature. We introduce the harmonic order and the expansion order of the electric field and find that every harmonic order current is composed of an infinite expansion order of the electric field, and the lowest expansion order of the electric field is equal to the harmonic order. Therefore, the nonlinear charge Hall effect studied previously just results from the second harmonic order and second expansion order of the electric field. To characterize the effects of a higher expansion order of the electric field, we introduce the Berry curvature multipole moment in which the so-called Berry curvature dipole is derived from the second expansion order of the electric field. We identify a “selection rule” for the nonlinear charge and spin currents for four cases with respect to the TRS and IS. Finally, we show the role of the Berry curvature quadrupole moment by studying a specified model. A vector formalism of the charge current is given, indicating the Berry curvature quadrupole may be inferred in experiments via this equation. In real applications, the order of harmonics may not be very large in some cases. For a material with an energy gap and the Fermi level is lying in the middle of the gap, a driving harmonics with high enough order may induce interband transitions, which is beyond the scope of our paper. Another case which deserves to mention is that when the energy bands are higher above the Fermi level, a resonant interband transition could still be happening for a large enough $n$. As our theory may be more important in slow fields and in dirty doping cases, there is plenty of space to test the predicted effect from higher harmonics in realistic applications.

This work is supported in part by the National Key R&D Program of China (Grant No. 2018YFA0305800), and the NSFC (Grants No. 11974348 and No.11834014). It is also supported by the Fundamental Research Funds for the Central Universities, and the Strategic Priority Research Program of CAS (Grants No. XDB28000000, and No. XDB33000000).

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