Disorder-induced nonlinear Hall effect with time-reversal symmetry

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The nonlinear Hall effect has opened the door towards deeper understanding of topological states of matter. Disorder plays indispensable roles in various linear Hall effects, such as the localization in the quantized Hall effects and the extrinsic mechanisms of the anomalous, spin, and valley Hall effects. Unlike in the linear Hall effects, disorder enters the nonlinear Hall effect even in the leading order. Here, we derive the formulas of the nonlinear Hall conductivity in the presence of disorder scattering. We apply the formulas to calculate the nonlinear Hall response of the tilted 2D Dirac model, which is the symmetry-allowed minimal model for the nonlinear Hall effect and can serve as a building block in realistic band structures. More importantly, we construct the general scaling law of the nonlinear Hall effect, which may help in experiments to distinguish disorder-induced contributions to the nonlinear Hall effect in the future.
The Hall effects refer to a transverse voltage in response to a current applied in a sample of metal or semiconductor. The family of the classical and quantized Hall effects is one of the mainstreams of modern condensed matter physics, leading to the full spectrum of the search on the topological states of matter and many practical applications. All previous Hall effects are in the linear-response regime, that is, the transverse voltage is linearly proportional to the driving current, and a measurable Hall voltage requires that time-reversal symmetry is broken by magnetic fields or magnetism. The recently discovered nonlinear Hall effect does not need time-reversal symmetry breaking but inversion symmetry breaking, significantly different from the known linear Hall effects.

The nonlinear Hall effect depends on the higher-order properties of the Berry curvature, thus not only can bring our knowledge to the next level but also may help device applications. More importantly, by adjusting the measurements to the nonlinear regime, unconventional transport phenomena can be explored in a great number of emergent materials in which discrete and crystal symmetries are broken. The disorder effects have been a large part of the research on the linear Hall effects, such as the localization in the quantized Hall effects, the extrinsic mechanisms of the anomalous, planar, and nonlinear Hall effects.

In this work, we use the Boltzmann formalism to derive the formulas of the nonlinear Hall conductivity, which help to identify the scaling laws of the nonlinear Hall effect, and comparably important. The disorder-induced contribution is comparably important. In the linear Hall effects, the extrinsic mechanisms of the anomalous, planar, and nonlinear Hall effects.

is the electron charge, \( l = (\eta, k) \) labels a state in band \( \eta \) with wave vector \( k \) and \( f_i \) is the corresponding distribution function. The current up to the second-order of the \( ac \) electric field can be found as \( J_a = Re\{j_a^{(0)} + j_a^{(1)} e^{i\omega t} + j_a^{(2)} e^{i2\omega t}\} \), with

\[
\begin{align*}
J_a^{(0)} &= \xi_{abc} e^b E^c, \\
J_a^{(1)} &= \sigma_{ab} e^b, \\
J_a^{(2)} &= \chi_{abc} e^b E^c,
\end{align*}
\]

respectively, where \( \{a, b, c\} \in \{x, y, z\} \). Table 1 summarizes our main results for the anomalous Hall response tensor \( \sigma_{ab} \) and the double-frequency nonlinear Hall response tensor \( \chi_{abc} \) (see Methods). We have assumed that \( \omega \tau \ll 1 \), because \( \omega \) is about tens of Hertz and the relaxation time \( \tau \) is about picoseconds in experiments. This low-frequency limit is one of the differences from the nonlinear optics. The disorder-induced zero-frequency response \( \sigma_{ab} \) is identical with the double-frequency response \( \chi_{abc} \) in the \( \omega \tau \ll 1 \) limit. Away from the \( \omega \tau \ll 1 \) limit, the double- and zero-frequency nonlinear Hall conductivities have different frequency dependence, thus are different in general. For a complete description, we list the \( \omega \)-dependent full expressions with and without time-reversal symmetry (Supplementary Note 3), which would be helpful for understanding the recently proposed high-frequency rectification and gyorotropic Hall effects.

According to how disorder works, the formulas are classified in terms of intrinsic (\( in \)), side-jump (\( sj \)), skew-scattering (\( sk \)) contributions. The formulas in Table 1 can be applied to different models to calculate the nonlinear Hall responses.
Tilted 2D massive Dirac model. Now we apply Table 1 to calculate the nonlinear Hall conductivity in the presence of disorder scattering, for the tilted 2D massive Dirac model (see Methods). The model gives the symmetry-allowed minimal description of the non-Gaussian scattering. The side-jump and skew-scattering contributions are due to the coordinates shift and antisymmetric scattering, respectively. Here \( \varepsilon_{\text{sk}} \) is the anti-symmetric tensor, we define \( \delta_{j} = \delta_{i,j}, \delta_{j} = \delta_{i,j} \), and \( g_{ij} \) is the general relaxation time and \( \varepsilon_{ij} \) is the anti-symmetric tensor, we define \( \delta_{j} = \delta_{i,j}, \delta_{j} = \delta_{i,j} \). The side-jump and skew-scattering becomes the strongest contribution, which is similar to the behaviors in the anomalous Hall effect. All contributions vanish as \( \varepsilon_{\text{sk}} \to \infty \). These behaviors can be seen in Fig. 2e.

The zero-frequency nonlinear Hall response was not addressed experimentally. In the \( \omega \tau \ll 1 \) limit, \( \xi_{\text{xy}} = \xi_{xy} \). According to symmetry, \( \xi_{\text{yy}} = 0 \). In the \( \tau \ll \omega \) limit, the electric field becomes time independent \( E_{\text{n}}(t) = E_{\text{n}} \), and the nonlinear Hall response becomes a \( \omega \tau \ll 1 \) dc current \( I_{\text{dc}} = \xi_{\text{dc}} \chi_{\text{dc}} \chi_{\text{xy}} E_{\text{dc}} E_{\text{x}} \), which means that for the Dirac model tilted along the x direction [Eq. (2)], an \( x \)-direction electric field can generate a \( dc \) nonlinear Hall current along the \( x \) direction. As a result, the measured Hall conductivity will be proportional to the electric field

\[
\sigma_{\text{xy}}^{\text{N}} = 2\chi_{\text{xy}}^{\text{E}} E_{\text{x}}.
\]

In contrast, if the electric field is along the \( y \) direction, there is no such a Hall signal because \( \chi_{\text{yy}} = 0 \), as required by the \( y \)-direction mirror reflection symmetry. This indicates that the \( dc \) nonlinear Hall signal \( \sigma_{\text{xy}}^{\text{N}} \) has one-fold angular dependence. This \( dc \) Hall signal can exist in the presence of time-reversal symmetry, which has been observed in the nonmagnetic Weyl–Kondo semimetal Ce\(_3\)Bi\(_4\)Pd\(_3\).25

### Scaling law of the nonlinear Hall effect

It is of fundamental importance to distinguish the different contributions to the nonlinear Hall signal in experiments. For the anomalous Hall effect, distinguishing different contributions is based on the scaling law of the transverse Hall signal to the longitudinal signal.20,21,26. For the nonlinear Hall effect, a scaling law can be constructed as well. We adopt the quantity \( V_{f}^{2} / (V_{x}^{2})^{2} = \xi_{\text{xy}}^{2} \rho_{\text{xx}}^{2} \) or \( \chi_{\text{xy}}^{\text{E}}^{2} \rho_{\text{xx}}^{2} \) as the experimental scaling variable,21,26, where \( V_{f}^{2} \) and \( V_{x}^{2} \) refer to the nonlinear Hall (zero- or double-frequency) and linear longitudinal voltage, respectively. To measure the nonzero \( \chi_{\text{xy}}^{\text{E}}^{2} \rho_{\text{xx}}^{2} \) the driving electric current is applied along the \( x \) direction and the nonlinear Hall voltage is measured along the \( y \) direction. An advantage of this variable is that the intrinsic and side-jump parts become disorder independent.

To account for multiple sources of scattering,21,26, we consider the scaling law of nonlinear Hall effect in a general manner. For simplicity, we assume no correlation between different scattering sources, thus each source contributes to the total resistivity independently, as dictated by Matthiessen’s rule \( \rho_{\text{xx}} = \sum \rho_{i}^{2} \), where \( \rho_{i} \) is the contribution of the \( i \)th type of disorder scattering to the longitudinal resistivity. According to Table 1, the general scaling law of the nonlinear Hall effect can be obtained as
contributions to nonlinear Hall conductivity. As an example, we consider two major scattering sources, one static (e.g., by using multi-step samples), and one dynamic (e.g., phonons). Furthermore, at finite temperatures, it is more convenient to rewrite the scaling law as

\[ \frac{V_N}{V_x^2} \approx (C_1 \rho_{xx0} + C_2 \rho_{xx0}^2 + C_3 \rho_{xx0} \rho_{xxT} + C_4 \rho_{xxT}^2), \] (8)

\[ \frac{V_N}{V_x^2} - C_1 \sigma_{xx0}^{-1} \sigma_{xx}^2 \approx (C_2 + C_4 - C_3) \sigma_{xx0}^{-1} \sigma_{xx}^2 + (C_3 - 2C_4) \sigma_{xx0}^{-1} \sigma_{xx}^{-1} \] (10)

This enables all four scaling parameters to be experimentally extracted from the total nonlinear Hall conductivity (e.g., by using multi-step samples). Furthermore, at finite temperatures, it is more convenient to rewrite the scaling law as

\[ \frac{V_N}{V_x^2} = C_1 \rho_{xx0} + C_2 \rho_{xx0}^2 + C_3 \rho_{xx0} \rho_{xxT} + C_4 \rho_{xxT}^2, \] (8)

so that the residual resistivity due to static impurities at zero temperature

\[ \rho_{xx0} = \frac{1}{e^2 \hbar^2 \pi^2} \int \frac{d^2 \mathbf{k}}{\Omega_2} \frac{1}{\epsilon_k - \epsilon_0}, \] (6)

\[ C_1 = C_{sk} + C_0^0 + C_{sk1}, \]
\[ C_2 = C_{sl} + C_{sl1} + C_{sl2} + C_{sl3}, \]
\[ C_3 = C_{s} + C_{s1} + C_{s2} + C_{s3} \]
\[ C_4 = C_{s} + C_{s1} + C_{s2} + C_{s3} \]

with four scaling parameters:

\[ C_1 = C_{sk}^1, C_2 = C_{sl} + C_{sl1} + C_{sl2} + C_{sl3}, \]
\[ C_3 = 2C_{sl} + C_{sl1} + C_{sl2} + C_{sl3} \]
\[ C_4 = C_{s} + C_{s1} + C_{s2} + C_{s3} \]

\[ C_{1,2,3,4} \text{can be extracted from experiments.} \]

\[ \rho_{xx0} = \frac{1}{e^2 \hbar^2 \pi^2} \int \frac{d^2 \mathbf{k}}{\Omega_2} \frac{1}{\epsilon_k - \epsilon_0}, \] (6)

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experimental data with the parabolic function, one can in principle extract the information of the rest scaling parameters, as shown in Fig. 3. Equation (10) can be reorganized as \( V_{xx}^{\prime \prime} / V_{xx}^{\prime} - c_{\text{ax}}^{-2} \sigma_{xx}^{}\sigma_{xx}^{\prime} \simeq (C_{2} - C_{3} + C_{4}) \sigma_{xx}^{2} + (C_{2} - 2C_{4}) c_{\text{ax}}^{-2} \sigma_{xx}^{}\sigma_{xx}^{\prime} + C_{4} \). In the anomalous Hall effect, the second term on the right has been argued to be negligible in both the high-temperature limit (\( c_{\text{ax}} \gg \sigma_{xx} \)) and the low-temperature limit (\( c_{\text{ax}} \approx \sigma_{xx} \)). This linear scaling behavior has been observed in thin films of WTe\(_{2}\). Nevertheless, Eq. (10) shows that the linear scaling behavior with \( \sigma_{xx}^{2} \) may become invalid in the high-conductivity regime\(^{21,26}\). In the nonmagnetic Weyl–Kondo semimetal Ce\(_{2}\)Bi\(_{4}\)Pd\(_{3}\), a linear scaling behavior of \( \sigma_{xx}^{2} \) is observed as a function of \( T \), representing the T-matrix\(^{27}\). 

\[
\sigma_{xx}^{N} \simeq C_{1} \sigma_{xx}^{2} + (C_{2} - C_{3} + C_{4}) \sigma_{xx}^{2} + (C_{2} - 2C_{4}) \sigma_{xx}^{2} + C_{4} \sigma_{xx},
\]

(11)

for a fixed electric field. According to the conductivity scaling law, the observed linear behavior\(^{25}\) indicates the dominance of the scaling parameter \( C_{4} \). According to Eq. (9), \( C_{4} \) is contributed mainly by the intrinsic mechanism and the dynamical scattering processes (e.g., Fig. 2b, c).

**Methods**

**Boltzmann formalism in the nonlinear regime.** In the Boltzmann formalism, the distribution function \( f \) can be found from the standard Boltzmann equation\(^{27}\), which reads

\[
\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla f = \mathcal{L}_{e}(f)
\]

in the spatially uniform case. Here \( \mathcal{L}_{e}(f) \) represents the elastic disorder scattering by static defects or impurities. The elastic disorder scattering can be decomposed as the intrinsic, side-jump, and skew-scattering parts (Supplementary Note 1)

\[
\mathcal{L}_{e}(f) = \mathcal{L}_{s}^{\text{inel}}(f) + \mathcal{L}_{s}^{\text{el}}(f) + \mathcal{L}_{s}^{\text{skew}}(f).
\]

(12)

The intrinsic part is contributed by symmetric scatterings, in which incoming and outgoing states are reversible in a scattering event. The side-jump part is resulting from the coordinates shift during scattering processes. The skew-scattering part is contributed by anti-symmetric scatterings, in which exchanging incoming and outgoing states yields a minus sign. Specifically, \( \mathcal{L}_{s}^{\text{inel}}(f) = -\sum_{\mathbf{k} \neq \mathbf{r}} \mathcal{O}_{s}^{}(\mathbf{f} - \mathbf{f}_{\mathbf{r}}) \), \( \mathcal{L}_{s}^{\text{el}}(f) = -\sum_{\mathbf{k} \neq \mathbf{r}} \mathcal{O}_{s}^{}(\mathbf{f} + \mathbf{f}_{\mathbf{r}}) \), where \( \mathcal{O}_{s} \) and \( \mathcal{O}_{l} \) represent the symmetric and antisymmetric parts of the scattering rate \( \mathcal{O}_{s} = (2n/\hbar)T_{s}^{\text{el}}(\partial \varepsilon_{\mathbf{f}} / \partial \varepsilon_{\mathbf{f}_{\mathbf{r}}}) \) with \( T_{s} \) representing the T-matrix\(^{28}\), \( \mathcal{O}_{l} = (2n/\hbar)T_{l}^{\text{el}}(\partial \varepsilon_{\mathbf{f}} / \partial \varepsilon_{\mathbf{f}_{\mathbf{r}}}) \), where the coordinates shift \( \varepsilon_{\mathbf{f}} \) is defined in Table I. The expression of \( \mathbf{r} \) and \( \mathbf{k} \) can be found from the semiclassical equations of motion\(^{28,29}\)

\[
\mathbf{i} = \mathbf{v} - \mathbf{k} \times \mathbf{v} + \mathbf{v}^{\prime}, \quad \mathbf{k} = \frac{\mathbf{c}}{\hbar} \mathbf{E},
\]

(14)

where \( \mathbf{v} = \partial_{\mathbf{x}} \psi / \hbar \mathbf{k} \) is the group velocity, \( \mathbf{H} \) is the Berry curvature\(^{15,28}\), and \( \mathbf{v}^{\prime} \) is the side-jump velocity\(^{28}\) (see Table 1). To solve the Boltzmann equations up to the second order of \( \mathbf{E} \), we adopt the relaxation time approximation\(^{27}\) for the intrinsic scattering parts \( \mathcal{L}_{s}^{\text{el}}(f) = (f_{\mathbf{f}}^{\prime} - f_{\mathbf{f}})T_{s}^{\text{el}}(\partial \varepsilon_{\mathbf{f}} / \partial \varepsilon_{\mathbf{f}}) \), where \( f_{\mathbf{f}}^{\prime} \) is the Fermi distribution function and \( T_{s}^{\text{el}} \) represents the relaxation time. Usually, in good metal regime, \( T_{s} \) is treated as a constant that can be determined by experiments. For systems with large anisotropy, \( T_{s} \) can have a significant angular dependence\(^{28,29}\). With the above equations, the current up to the second-order responses to the applied electric field can be obtained.

**Tilted 2D massive Dirac model with disorder.** We use the tilted 2D massive Dirac model in Eq. (2) to calculate the nonlinear Hall conductivity in Fig. 2. The model describes two energy bands (denoted as \( \pm \)) with the band dispersions \( \varepsilon_{\pm} = \pm m^{*}\mathbf{k}^{2}/2m^{*} \), where \( m^{*} \equiv k_{x}^{2} + k_{y}^{2} \). In the \( x-y \) plane, the Berry curvature behaves like a pseudoscalar, with only the \( z \) component

\[
\Omega_{z}^{\pm} = \frac{\pi m^{*}^{2}}{2m^{*}k^{2} + m^{*}k^{4}}/2.
\]

To consider the disorder effect, we expanded the scattering rate up to the fourth order in the disorder strength as \( \mathcal{S}_{\pm} = \mathcal{S}_{\pm}^{(2)} + \mathcal{S}_{\pm}^{(3)} + \mathcal{S}_{\pm}^{(4)} \). Here \( \mathcal{S}_{\pm}^{(n)} \) is pure symmetric and of order \( n \mathbf{V}_{\mathbf{f}}^{\pm} \) with \( n \mathbf{V}_{\mathbf{f}}^{\pm} \) refers to the concentration of disorder. Figure 2a corresponds to the contribution to \( \mathcal{S}_{\pm}^{(2)} \), which is non-Gaussian and of order \( n \mathbf{V}_{\mathbf{f}}^{\pm} \). Figure 2b corresponds to \( \mathcal{S}_{\pm}^{(3)} \) within non-crossing approximation, which is Gaussian and of order \( n^{2} \mathbf{V}_{\mathbf{f}}^{\pm} \). Thus, \( \mathcal{S}_{\pm}^{(2)} \) is the leading symmetric contribution, \( \mathcal{S}_{\pm}^{(3)} \) and \( \mathcal{S}_{\pm}^{(4)} \) contain the leading non-Gaussian and Gaussian antisymmetric contribution to the scattering rate. Considering all the leading contributions, we identify that \( \mathcal{S}_{\pm}^{(2)} = \mathcal{S}_{\pm}^{(2)} \) and \( \mathcal{S}_{\pm}^{(3)} = \mathcal{S}_{\pm}^{(3)} + \mathcal{S}_{\pm}^{(4)} \), where \( \mathcal{S}_{\pm}^{(3)} \) and \( \mathcal{S}_{\pm}^{(4)} \) represent the antisymmetric parts of the third and fourth order scattering rate, respectively (Supplementary Note 4).
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Author contributions
Z.Z.D. did the calculations with assistance from C.M.W., S.L., and H.-Z.L. Z.Z.D. and H.-Z.L. wrote the manuscript with assistance from C.M.W., S.L., and X.C.X. H.-Z.L. and X.C.X. supervised the project.

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