Theory of nonlinear Hall effects: Modified semiclassics from quantum kinetics

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(Received 29 May 2019; published 28 October 2019)

We propose a modified Boltzmann nonlinear electric-transport framework which differs from the nonlinear generalization of the linear Boltzmann formalism by a contribution that has no counterpart in linear response. This contribution follows from the interband-coherence effect of dc electric fields during scattering and is related to the interband Berry connection. As an application, we demonstrate it in the second-order nonlinear Hall effect of the tilted massive Dirac model. The intuitive Boltzmann constructions are confirmed by a quantum kinetic theory, which shows that the arbitrary nth-order nonlinear dc response up to the first three leading contributions in the weak disorder potential is handled by the same few gauge-invariant semiclassical ingredients.

DOI: 10.1103/PhysRevB.100.165422

I. INTRODUCTION

The nonlinear response to an applied electric field in crystalline solids has attracted revived interest, owing to the essential role played by the quantum geometry of the Bloch wave function [1–4]. In the optical high-frequency regime of the electric field, the shift current photogalvanic effect and second harmonic generation have been shown to be related to the Berry connection of each involved Bloch band [5,6]. In the low-frequency regime, higher-order moments of the Berry curvature in momentum space emerge in the nonlinear anomalous Hall responses in the absence of magnetic field, such as the Berry curvature dipole [3,7–15] and quadrupole [16] in second- and third-order Hall responses, respectively. In particular, the second-order nonlinear response dominates the anomalous Hall effect in time-reversal-invariant crystals that break inversion symmetry, and has been observed in few-layer WTe_2 [17,18].

The quantum geometry of the Bloch electron also influences its scattering with disorder. A prominent case is the linear anomalous Hall effect [19], where nonzero Berry connection and curvature imply the presence of two asymmetric scattering effects termed as skew scattering and side jump [20]. A Boltzmann transport formalism for the linear anomalous Hall effect has been established [21–25]. It has been generalized phenomenologically in the recent efforts to understand the second-order nonlinear response in the low-frequency limit [26–29]. A basic question then arises: Is this framework valid in nonlinear responses? The existing quantum transport theories [30,31] have not settled this issue. Moreover, there exist two different proposals [26,28] to generalize phenomenologically the side-jump contribution to the second-order nonlinear response. More importantly, one should worry whether there is another contribution that is missed in the direct generalization of the aforementioned semiclassical formalism. If there is, can the nonlinear response still be grasped by the few gauge-invariant semiclassical ingredients as in the linear response?

In this paper we address all the above concerns in the dc limit by developing a recursive quantum kinetic theory for an arbitrary nth-order (finite n) electric current response. We focus on the first three leading-order contributions in the weak disorder potential $\hat{V}$ (namely, V^{-2n}, V^{-2n+1}, V^{-2n+2} in the nth-order electric transport), which are usually sufficient to account for both the longitudinal and transverse transport in the regime $\hbar/\tau < \Delta$ (τ is the scattering time, Δ is the band splitting around the Fermi level). Remarkably, we find that an arbitrary order nonlinear response retains the same structure as the linear response, except for a contribution resulting from the electric-field-induced interband virtual transition during the scattering. This contribution only contributes to a nonlinear response and is related to the interband Berry connection. A modified Boltzmann nonlinear-response framework thus emerges, establishing a consistency between the Boltzmann and quantum kinetics in nonlinear (Hall) electric transport. As an application, we show the aforementioned contribution in the second-order nonlinear Hall effect of the two-dimensional (2D) tilted massive Dirac model.

Our paper is organized as follows. In Sec. II we set forth the modified Boltzmann theory for nonlinear electric transport, which is applied to the model calculation of the second-order nonlinear Hall effect in Sec. III. The quantum kinetic theory that underlies the Boltzmann formulation is outlined in Sec. IV, with the main ideas and results elaborated. Finally, we compare our theory to other existing theories in Sec. V and conclude this paper in Sec. VI. The detailed derivation of our quantum kinetic theory is presented in the Supplemental Material [32] (see also Ref. [33] therein) for the convenience of interested readers.

II. MODIFIED SEMICLASSICS

The outcomes of the quantum kinetic approach are found to correspond to a semiclassical Boltzmann way of understanding the nonlinear transport. In this section we first describe the latter framework, considering its great physical transparency and simplicity.

In the Boltzmann description of electronic transport in crystalline solids, the charge current density is given by

$$\boldsymbol{j} = e\sum_l F_l \boldsymbol{v}_l, \tag{1}$$

where the occupation function F_l of the Bloch state $|l\rangle = |\eta \boldsymbol{k}\rangle$, with η the band index and $\boldsymbol{k}$ the crystal momentum, and the velocity $\boldsymbol{v}_l$ are two central quantities. In the perturbative treatment for the weak electric field, F_l can be expanded in terms of ascending powers (denoted by n) of the electric field $\boldsymbol{E}$, namely,

$$F_l = \sum_{n\geqslant 0} F_{n,l}, \tag{2}$$

where $F_{n,l} \propto E^n$ is the occupation function responsible for the nth-order electric transport.

In the conventional Boltzmann recipe the driving term by the applied electric field and the collision term by scattering are clearly separated in the steady-state Boltzmann equation [34]

$$-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}} F_l = \sum_{l'}\left(\omega^{(2)}_{l'l}F_l - \omega^{(2)}_{ll'}F_{l'}\right). \tag{3}$$

The semiclassical scattering rate, regarded to be independent of the electric field, is given by the golden rule

$$\omega^{(2)}_{l'l} = \omega^{(2)}_{ll'} = \frac{2\pi}{\hbar}W_{l'l}\delta(\epsilon_l - \epsilon_{l'}), \quad W_{l'l} = \langle |V_{ll'}|^2\rangle_c. \tag{4}$$

Here, $W_{l'l}$ is the scattering matrix element, and $\langle\cdots\rangle_c$ stands for the disorder average. In the constant relaxation time approximation the collision term on the right-hand side of Eq. (3) reduces to F_l/τ, where $1/\tau \sim V^2$, and the recursive solution of this equation yields the scaling $F_{n,l} \sim E^n\tau^n \sim E^nV^{-2n}$.

A. Modification to scattering by electric field

In the conventional Boltzmann equation the scattering process is independent of the electric field. But this is not true in general. A prominent example is the linear anomalous Hall current originating from the work done by the electric field during scattering. The key ingredient here is the coordinate shift of a semiclassical electron during any scattering process [20],

$$\delta\boldsymbol{r}_{l'l} = \mathcal{A}_{l'} - \mathcal{A}_l - (\partial_{\boldsymbol{k}} + \partial_{\boldsymbol{k}'})\arg V_{l'l}, \tag{5}$$

where $\mathcal{A}_l = \langle u_{\eta\boldsymbol{k}}|i\partial_{\boldsymbol{k}}|u_{\eta\boldsymbol{k}}\rangle$ is the intraband Berry connection, with $|u_{\eta\boldsymbol{k}}\rangle$ the periodic part of the Bloch state. This picture implies that the energy conservation condition in the golden rule [Eq. (4)] is modified to be

$$\delta(\epsilon_l - \epsilon_{l'} + e\boldsymbol{E}\cdot\delta\boldsymbol{r}_{l'l}) \simeq \delta(\epsilon_l - \epsilon_{l'}) + \frac{\partial\delta(\epsilon_l - \epsilon_{l'})}{\partial\epsilon_l}e\boldsymbol{E}\cdot\delta\boldsymbol{r}_{l'l}. \tag{6}$$

The direct generalization of this semiclassical construction into nonlinear responses leads to an occupation function which scales as $F_{n,l} \sim E^n\tau^{n-1} \sim E^nV^{-2n+2}$. This $F_{n,l}$ yields an important contribution to the second-order nonlinear Hall effect [26,27,29]. Note that the first-order expansion in the above equation is already sufficient to obtain the $F_{n,l}$ of order of V^{-2n+2}.

We reveal in the following that there is another electric-field-induced effect during scattering, which only contributes to nonlinear responses. The intuitive motivation is that not only the energy conservation delta function but also the scattering matrix element $W_{l'l}$ of the semiclassical scattering rate should be corrected by the $\boldsymbol{E}$ field. This term is an interband-coherence (interband virtual transition) effect of the $\boldsymbol{E}$ field during scattering. More precisely, the Bloch states involved in the scattering have to be dressed by the electric field, thus $V_{ll'} \to \langle\tilde{l}|\hat{V}|\tilde{l}'\rangle$, where $|\tilde{l}\rangle = |l\rangle + |\delta^{\boldsymbol{E}}l\rangle$ is the $\boldsymbol{E}$-field-dressed Bloch state, and

$$|\delta^{\boldsymbol{E}}l\rangle = -e{\sum_{l''}}'|l''\rangle\frac{\boldsymbol{E}\cdot\mathcal{A}_{l''l}}{\epsilon_l - \epsilon_{l''}} \tag{7}$$

arises from the electric-field-induced interband virtual transition [35]. $\mathcal{A}_{ll'} = \langle u_{\eta\boldsymbol{k}}|i\partial_{\boldsymbol{k}}|u_{\eta'\boldsymbol{k}}\rangle$ is the interband Berry connection. Hereafter the notation $\sum'$ means that all the index equalities should be avoided in the summation. In order to obtain the $F_{n,l}$ of order of V^{-2n+2}, it is sufficient to retain

$$W_{l'l} \to W_{l'l} + \delta^{\boldsymbol{E}}W_{l'l}, \tag{8}$$

where the $\boldsymbol{E}$-field-corrected scattering matrix element is linear in $\boldsymbol{E}$ and reads

$$\begin{aligned}\delta^{\boldsymbol{E}}W_{l'l} &= 2\,\mathrm{Re}\langle V_{ll'}(\langle l'|\hat{V}|\delta^{\boldsymbol{E}}l\rangle + \langle\delta^{\boldsymbol{E}}l'|\hat{V}|l\rangle)\rangle_c \\ &= -e\boldsymbol{E}\cdot{\sum_{l''}}'2\,\mathrm{Re}\left\langle\frac{V_{ll''}V_{l''l'}\mathcal{A}_{l''l}}{\epsilon_l - \epsilon_{l''}} + \frac{V_{ll'}\mathcal{A}_{l'l''}V_{l''l}}{\epsilon_{l'} - \epsilon_{l''}}\right\rangle_c.\end{aligned} \tag{9}$$

In Fig. 1 we show schematically the physical processes described by $\delta^{\boldsymbol{E}}W_{l'l}$ in a two-band system. The Fermi level is assumed to locate at the conduction band. Because of the presence of the vertical interband virtual transition induced by the electric field, these scattering processes involve an off-shell Bloch state away from the Fermi surface.

Collecting Eqs. (4), (6), (8), and (9), the $\boldsymbol{E}$-field-corrected scattering rate takes the following form,

$$\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l} = \delta^{\boldsymbol{E}}_1\omega^{(2)}_{l'l} + \delta^{\boldsymbol{E}}_2\omega^{(2)}_{l'l}, \tag{10}$$

where

$$\delta^{\boldsymbol{E}}_1\omega^{(2)}_{l'l} = \frac{2\pi}{\hbar}W_{l'l}\frac{\partial\delta(\epsilon_l - \epsilon_{l'})}{\partial\epsilon_l}e\boldsymbol{E}\cdot\delta\boldsymbol{r}_{l'l}, \tag{11}$$

$$\delta^{\boldsymbol{E}}_2\omega^{(2)}_{l'l} = \frac{2\pi}{\hbar}\delta^{\boldsymbol{E}}W_{l'l}\delta(\epsilon_l - \epsilon_{l'}). \tag{12}$$

While $\delta^{\boldsymbol{E}}_1\omega^{(2)}_{l'l}$ has been well known, $\delta^{\boldsymbol{E}}_2\omega^{(2)}_{l'l}$ is proposed in the context of the nonlinear Hall effect. We note here that interband virtual processes are also indispensable in $\delta^{\boldsymbol{E}}_1\omega^{(2)}_{l'l}$ through the coordinate shift $\delta\boldsymbol{r}_{l'l}$ [20].

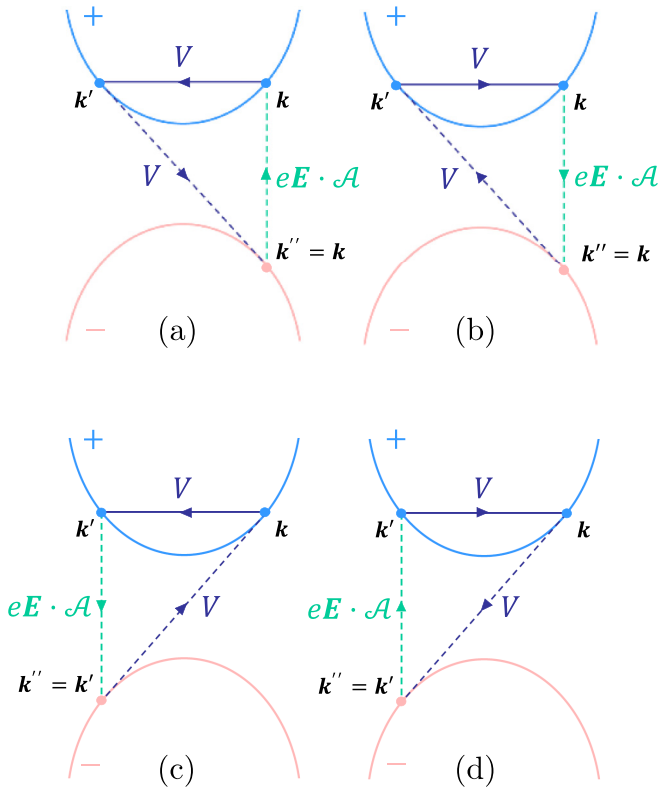

FIG. 1. Schematics of Eq. (9) describing the electric-field-induced interband virtual processes during scattering in two-band systems. The contributions from (a) and (b) to $\delta^{\boldsymbol{E}}W_{l'l}$ are complex conjugated, as are (c) and (d).

B. Boltzmann equation for nonlinear responses

Taking into account the effect of the $\boldsymbol{E}$ field during scattering, the Boltzmann equation (3) is modified to be

$$-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F_l=\sum_{l'}\left(\omega^{(2)}_{l'l}+\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l}\right)(F_l-F_{l'}). \tag{13}$$

In combination with Eq. (2), it can be written in a recursive form,

$$\begin{aligned}&-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F_{n-1,l}-\sum_{l'}\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l}(F_{n-1,l}-F_{n-1,l'})\\&\quad=\sum_{l'}\left(\omega^{(2)}_{l'l}F_{n,l}-\omega^{(2)}_{ll'}F_{n,l'}\right)\quad(n\geqslant 1),\end{aligned} \tag{14}$$

where the effect of the electric field during scattering appears as an effective driving term in the Boltzmann equation for $F_{n,l}$, and $F_{n,l}$ is accurate to the third leading order of the weak disorder potential, namely, the order of V^{-2n+2}.

In a linear response, $n=1$, $F_{0,l}$ is the Fermi distribution and $(F_{0,l}-F_{0,l'})\delta(\epsilon_l-\epsilon_{l'})=0$, thus $\delta^{\boldsymbol{E}}_2\omega^{(2)}_{l'l}$ does not contribute to the Boltzmann equation. This explains why this term is absent in the Boltzmann theory of linear response [22]. By contrast, it constitutes a basic ingredient of the Boltzmann description of nonlinear responses.

Higher-order disorder corrections to $\omega^{(2)}_{l'l}$ on the right-hand side of Eq. (14) are included through replacing $V_{ll'}$ by the T matrix $T_{ll'}$ [22,25]. The golden rule thus yields $\omega_{ll'}=\omega^{(2)}_{ll'}+\omega^{(3)}_{ll'}+\omega^{(4)}_{ll'}$ up to the first three leading orders of the disorder potential. Hereafter the superscript (i) means the order in the disorder potential. It is easy to check that such corrections to $\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l}$ are not needed provided that the electric current is considered up to the three leading orders of the disorder potential.

In the considered case the occupation function is the sum of the leading (L), subleading (SL), and subsubleading (SSL) contributions,

$$F_{n,l}=F^{\rm L}_{n,l}+F^{\rm SL}_{n,l}+F^{\rm SSL}_{n,l}, \tag{15}$$

where $F^{\rm L}_{n,l}$, $F^{\rm SL}_{n,l}$, and $F^{\rm SSL}_{n,l}$ are of order of V^{-2n}, V^{-2n+1}, and V^{-2n+2}, respectively. The semiclassical occupation functions with a positive exponent of V can be neglected in the weak disorder regime, thus in equilibrium $F_{0,l}=F^{\rm L}_{0,l}$ is just the Fermi distribution and $F^{\rm SL}_{0,l}=F^{\rm SSL}_{0,l}=0$. This point is in fact implicit in previous works on the semiclassical Boltzmann theories for the linear and nonlinear anomalous Hall effects [22,26–29].

Therefore, the Boltzmann equation can be cast into the following three equations,

$$-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F^{\rm L}_{n-1,l}=\sum_{l'}\omega^{(2)}_{l'l}\left(F^{\rm L}_{n,l}-F^{\rm L}_{n,l'}\right), \tag{16}$$

$$\begin{aligned}-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F^{\rm SL}_{n-1,l}&=\sum_{l'}\omega^{(2)}_{l'l}\left(F^{\rm SL}_{n,l}-F^{\rm SL}_{n,l'}\right)\\&\quad+\sum_{l'}\left(\omega^{(3)as}_{l'l}F^{\rm L}_{n,l}-\omega^{(3)as}_{ll'}F^{\rm L}_{n,l'}\right),\end{aligned} \tag{17}$$

and

$$\begin{aligned}&-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F^{\rm SSL}_{n-1,l}-\sum_{l'}\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l}\left(F^{\rm L}_{n-1,l}-F^{\rm L}_{n-1,l'}\right)\\&\quad=\sum_{l'}\omega^{(2)}_{l'l}\left(F^{\rm SSL}_{n,l}-F^{\rm SSL}_{n,l'}\right)\\&\qquad+\sum_{l'}\left(\omega^{(4)as}_{l'l}F^{\rm L}_{n,l}-\omega^{(4)as}_{ll'}F^{\rm L}_{n,l'}\right),\end{aligned} \tag{18}$$

which are of order of V^{-2n+2}, V^{-2n+3}, and V^{-2n+4}, respectively. In a linear response, $n=1$, these three equations just reduce to the familiar ones in the study of the linear anomalous Hall effect [22]. In line with the Boltzmann recipe for the linear response [22,35], the antisymmetric part $[\omega^{\rm as}_{l'l}\equiv(\omega_{l'l}-\omega_{ll'})/2]$ of $\omega_{ll'}$, namely, $\omega^{(3)\rm as}_{ll'}$ and $\omega^{(4)\rm as}_{ll'}$, yields the skew scattering contribution to nonequilibrium phenomena, while the inessential symmetric part of $\omega^{(3)}_{ll'}$ and $\omega^{(4)}_{ll'}$ has been suppressed in the above three equations.

$F^{\rm SL}_{n,l}$ arises from the conventional skew scattering induced by non-Gaussian disorder, thus it can also be labeled by $F^{\rm csk}_{n,l}$. $F^{\rm SSL}_{n,l}$ comprises contributions from the skew scattering induced by Gaussian disorder (through $\omega^{(4)\rm as}_{l'l}$) and the electric-field-corrected scattering rate, thus can be decomposed into

$$F^{\rm SSL}_{n,l}=F^{\rm Gsk}_{n,l}+F^{\rm a}_{n,l}, \tag{19}$$

with

$$-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F^{\rm Gsk}_{n-1,l}=\sum_{l'}\omega^{(2)}_{l'l}\big(F^{\rm Gsk}_{n,l}-F^{\rm Gsk}_{n,l'}\big)+\sum_{l'}\big(\omega^{(4)\rm as}_{l'l}F^{\rm L}_{n,l}-\omega^{(4)\rm as}_{ll'}F^{\rm L}_{n,l'}\big) \tag{20}$$

and

$$-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F^{\rm a}_{n-1,l}-\sum_{l'}\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l}\big(F^{\rm L}_{n-1,l}-F^{\rm L}_{n-1,l'}\big)=\sum_{l'}\omega^{(2)}_{l'l}\big(F^{\rm a}_{n,l}-F^{\rm a}_{n,l'}\big). \tag{21}$$

Here, $F^{\rm a}_{n,l}$ can be further decomposed as $F^{\rm a}_{n,l}=F^{\rm a1}_{n,l}+F^{\rm a2}_{n,l}$, in correspondence to Eq. (10). We note again that $F^{\rm a2}_{1,l}=0$ in the linear response.

C. Electric current in the semiclassical framework

It has been well known that $\boldsymbol{v}_l$ is not equal to the usual group velocity $\boldsymbol{v}^0_l$, but contains corrections from interband virtual transitions induced by both the electric field and scattering [21,35]: $\boldsymbol{v}_l=\boldsymbol{v}^0_l+\boldsymbol{v}^{\rm bc}_l+\boldsymbol{v}^{\rm sj}_l$. Here, $\boldsymbol{v}^{\rm bc}_l=\frac{e}{\hbar}(\partial_{\boldsymbol{k}}\times\mathcal{A}_l)\times\boldsymbol{E}$ and $\boldsymbol{v}^{\rm sj}_l=\sum_{l'}\omega^{(2)}_{l'l}\delta\boldsymbol{r}_{l'l}$ are the Berry-curvature anomalous velocity and side-jump velocity, respectively [20].

Therefore, the nth-order electric current is given by

$$\begin{aligned}\boldsymbol{j}_n=&\,e\sum_l F^{\rm L}_{n,l}\boldsymbol{v}^0_l+e\sum_l F^{\rm csk}_{n,l}\boldsymbol{v}^0_l\\&+e\sum_l F^{\rm Gsk}_{n,l}\boldsymbol{v}^0_l+e\sum_l F^{\rm a1}_{n,l}\boldsymbol{v}^0_l+e\sum_l F^{\rm a2}_{n,l}\boldsymbol{v}^0_l\\&+e\sum_l F^{\rm L}_{n,l}\boldsymbol{v}^{\rm sj}_l+e\sum_l F^{\rm L}_{n-1,l}\boldsymbol{v}^{\rm bc}_l\end{aligned} \tag{22}$$

up to the first three leading orders of the weak disorder potential. The third term on the second line is absent in all recent works on the semiclassical Boltzmann theory of the second-order nonlinear anomalous Hall effect [26–29].

Both $F^{\rm a1}_{n,l}$ and $\boldsymbol{v}^{\rm sj}_l$ are related to the coordinate shift, thereby the sum of these two terms is usually referred to as the side-jump contribution [20–22,26–29]. However, $F^{\rm a1}_{n,l}$ has nothing to do with the sideways shift, which is the original meaning of side jump [36]. Accordingly, in the following the terminology "side jump" is only assigned to the $\boldsymbol{v}^{\rm sj}_l$ term.

III. MODEL CALCULATION IN SECOND-ORDER NONLINEAR HALL EFFECT

To be specific, we illustrate the contribution from the $F^{\rm a2}_{n,l}$ term in the second-order nonlinear Hall effect in inversion-breaking nonmagnetic materials [3,17,18]. To obtain an analytic result, we follow the previous publications involving $\delta^{\boldsymbol{E}}_1\omega^{(2)}_{l'l}$ [26,27,29] to take the constant relaxation time so that $\sum_{l'}\omega^{(2)}_{l'l}(F^{\rm a2}_{2,l}-F^{\rm a2}_{2,l'})=F^{\rm a2}_{2,l}/\tau$ and

$$F^{\rm a2}_{2,l}=-\tau\sum_{k'}\frac{2\pi}{\hbar}\delta^{E}W_{l'l}\delta(\epsilon_l-\epsilon_{l'})\big(F^{\rm L}_{1,l}-F^{\rm L}_{1,l'}\big). \tag{23}$$

Here, $F^{\rm L}_{1,l}$ solves the conventional Boltzmann equation (16), reading $F^{\rm L}_{1,l}=-\frac{e}{\hbar}\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}F^{\rm L}_{0,l}\tau$ in the constant relaxation time approximation. When the $\boldsymbol{E}$ field is applied in the x direction, the resultant transverse current is

$$j^a_y=e\sum_l F^{\rm a2}_{2,l}v^0_{l,y}\equiv\Xi^a_{yxx}E_xE_x, \tag{24}$$

where Ξ^a_{yxx} is the corresponding second-order response coefficient. One can show that Ξ^a_{yxx} can be nonzero when the inversion symmetry is broken, even if the time-reversal symmetry remains. This character is the same as the known contributions of order of τ to the second-order nonlinear Hall effect [27–29,31].

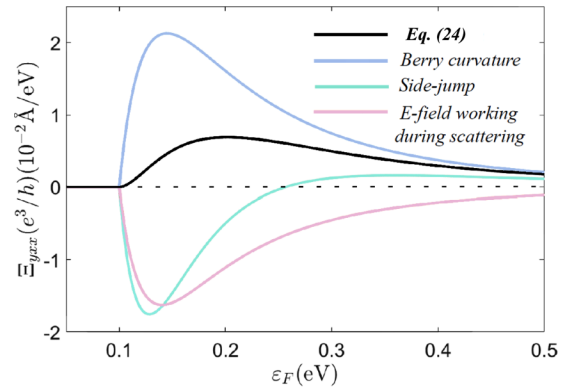

FIG. 2. The second-order nonlinear Hall responses in the 2D tilted massive Dirac model that are beyond the conventional Boltzmann equation, from the Berry curvature and side-jump velocities, the $\boldsymbol{E}$ field working during scattering $e\boldsymbol{E}\cdot\delta\boldsymbol{r}_{l'l}$, and the interband effect of the $\boldsymbol{E}$ field during scattering [Eq. (24)]. Parameters are chosen as $t=0.1$ eV Å, $v=1$ eV Å, $\Delta=0.1$ eV, $n_iV_0^2=10^2$ (eV Å)^2. $h=2\pi\hbar$ is the Planck constant.

Let us take the 2D tilted massive Dirac model [3],

$$\hat{H}_0=tk_x+v(k_x\sigma_x+k_y\sigma_y)+\Delta\sigma_z, \tag{25}$$

with scalar disorder as a concrete example, which is the minimal model of the considered effect [3,17,27,29,31]. $\sigma_{x,y,z}$ are the Pauli matrices, and the gapped Dirac cone is tilted along the x direction. Here, one can consider the contribution from only one Dirac cone because, as addressed in Refs. [3,27], taking into account that from another one of the pair of Dirac cones simply doubles the obtained result.

When the Fermi level only intersects the upper (+) band we have

$$\delta^{\boldsymbol{E}}W_{l'l}=\frac{W_{\boldsymbol{k}'\boldsymbol{k}}}{2}e\boldsymbol{E}\cdot\left(\frac{\Omega_{+\boldsymbol{k}'}}{\Delta_{\boldsymbol{k}}}-\frac{\Omega_{+\boldsymbol{k}}}{\Delta_{\boldsymbol{k}'}}\right)\hat{\boldsymbol{z}}\times(\boldsymbol{k}'-\boldsymbol{k}), \tag{26}$$

where $l=+\boldsymbol{k}$, $l'=+\boldsymbol{k}'$, $W_{\boldsymbol{k}'\boldsymbol{k}}=W_{l'l}/|\langle u_l|u_{l'}\rangle|^2$, $\Omega_{+\boldsymbol{k}}$ is the Berry curvature of the upper band, and $\Delta_{\boldsymbol{k}}=\epsilon^+_{\boldsymbol{k}}-\epsilon^-_{\boldsymbol{k}}$. To compare with other contributions obtained analytically for this model, we also assume weak anisotropy $t\ll v$ and take [29,31] $1/\tau=n_iV_0^2(\epsilon_F^2+3\Delta^2)/(4\hbar v^2\epsilon_F)$ in the presence of pointlike impurities of density n_i, for which $W_{\boldsymbol{k}'\boldsymbol{k}}=n_iV_0^2$. It follows that

$$\Xi^a_{yxx}=-\frac{e^3}{2\pi\hbar}\frac{t\Delta}{n_iV_0^2}\frac{3v^2\big(\epsilon_F^2-\Delta^2\big)^2}{\epsilon_F^3\big(\epsilon_F^2+3\Delta^2\big)^2} \tag{27}$$

up to the first order of t. As shown in Fig. 2, Ξ^a_{yxx} is of the similar magnitude to the previously identified contributions

[3,26,29] that are also beyond the conventional Boltzmann recipe [37].

IV. BOLTZMANN TRANSPORT EMERGING FROM QUANTUM KINETICS

In this section we place the intuitive Boltzmann framework on the foundation of quantum kinetics, by extending the density-matrix equation of motion approach of Kohn and Luttinger [23,38] to nonlinear responses.

A. Basic formulations

In the single-electron Hamiltonian $\hat{H}_T = \hat{H}_0 + \hat{V} + \hat{H}_{\boldsymbol{E}}$, $\hat{H}_0$ is the equilibrium disorder-free one, $\hat{V}$ is the potential produced by randomly distributed impurities, and the $\boldsymbol{E}$-field term $\hat{H}_{\boldsymbol{E}} = -e\boldsymbol{E}\cdot\boldsymbol{r}e^{st}$ is switched on adiabatically from the remote past $t=-\infty$. The physical situation is obtained by taking the limit $s\to 0^+$ [38]. In the case of a weak $\boldsymbol{E}$ field, the single-particle density matrix is decomposed into $\hat{\rho}_T = \sum_{n\geqslant 0}\hat{\rho}_n$, where $\hat{\rho}_0$ is its equilibrium value, and $\hat{\rho}_n \propto E^n$ satisfies $\hat{\rho}_n(t\to-\infty)=0$ for $n\geqslant 1$. Then the quantum Liouville equation reduces to $[\hat{H}_0+\hat{V},\hat{\rho}_0]=0$ and

$$i\hbar\frac{\partial\hat{\rho}_n}{\partial t} = [\hat{H}_{\boldsymbol{E}},\hat{\rho}_{n-1}] + [\hat{H}_0+\hat{V},\hat{\rho}_n] \quad (n\geqslant 1), \tag{28}$$

where $[\hat{H}_{\boldsymbol{E}},\hat{\rho}_{n-1}]$ enables the recursion from $\hat{\rho}_{n-1}$ to $\hat{\rho}_n$. Utilizing the ansatz [38] $\hat{\rho}_n = \hat{f}_n e^{nst}$, where $\hat{f}=\sum_{n\geqslant 0}\hat{f}_n$ is the single-particle density matrix at the time of interest $t=0$, in the Bloch representation of $\hat{H}_0$ we have ($n\geqslant 1$)

$$(\epsilon_l-\epsilon_{l'}-i\hbar ns)f_{n,ll'} = \sum_{l''}(f_{n,ll''}V_{l''l'}-V_{ll''}f_{n,l''l'}) + e\boldsymbol{E}\cdot[\boldsymbol{r},\hat{f}_{n-1}]_{ll'}. \tag{29}$$

When $l=l'$ the equation of motion (29) reduces to (see more details in Refs. [25,38,39])

$$0 = C_{n,l} + \sum_{l'}{}'(f_{n,ll'}V_{l'l}-V_{ll'}f_{n,l'l}), \tag{30}$$

otherwise we have ($f_{n,l}\equiv f_{n,ll}$)

$$f_{n,ll'} = \frac{C_{n,ll'}}{\epsilon_l-\epsilon_{l'}-i\hbar ns} + \frac{f_{n,l}-f_{n,l'}}{\epsilon_l-\epsilon_{l'}-i\hbar ns}V_{ll'} + \sum_{l''}{}'\frac{f_{n,ll''}V_{l''l'}-V_{ll''}f_{n,l''l'}}{\epsilon_l-\epsilon_{l'}-i\hbar ns} \quad (l\neq l'), \tag{31}$$

where V_{ll} is absorbed into H_0 and then $V_{ll}=0$ [38]. Here,

$$C_{n,l} = e\boldsymbol{E}\cdot\{i\partial_{\boldsymbol{k}}f_{n-1,l} + [\mathcal{A},\hat{f}_{n-1}]_{ll}\}, \tag{32}$$

and

$$C_{n,ll'} = e\boldsymbol{E}\cdot\{i(\partial_{\boldsymbol{k}}+\partial_{\boldsymbol{k}'})f_{n-1,ll'} + [\mathcal{A},\hat{f}_{n-1}]_{ll'}\}. \tag{33}$$

According to Eq. (31), in the case of weak disorder potential $f_{n,ll'}$ is generally one order of V higher than $f_{n,l}$. Thereby, Eq. (31) can be solved by an iterative procedure, which yields the expression for $f_{n,ll'}$ in terms of $f_{n,l}$ [23,38]. Substituting this solution into Eq. (30) leads to an equation only concerning the diagonal element $f_{n,l}$ for $n\geqslant 1$. The disorder average of this latter equation yields a Boltzmann-type equation for $\langle f_{n,l}\rangle_c$, provided that one assumes $f_{n,l}$ is self-averaged, i.e., $\langle f_{n,l}VV\rangle_c = \langle f_{n,l}\rangle_c\langle VV\rangle_c$. This assumption plays a similar role to that of the assumption of molecular chaos in deriving the classical Boltzmann equation from the classical Liouville equation [40]. One can then identify $\langle f_{n,l}\rangle_c$ with the occupation function used in the Boltzmann framework,

$$F_{n,l}\equiv\langle f_{n,l}\rangle_c, \quad F_l\equiv\langle f_l\rangle_c = \sum_{n\geqslant 0}\langle f_{n,l}\rangle_c. \tag{34}$$

On the other hand, the nth-order charge current in the density-matrix formulation is given by

$$\boldsymbol{j}_n = \boldsymbol{j}_n^{\rm d} + \boldsymbol{j}_n^{\rm od}, \tag{35}$$

where $\boldsymbol{j}_n^{\rm d} = e\sum_l\langle f_{n,l}\rangle_c\boldsymbol{v}_l^0$ and $\boldsymbol{j}_n^{\rm od} = e\sum_{ll'}{}'\langle f_{n,ll'}\rangle_c\boldsymbol{v}_{l'l}^0$ are the band-diagonal and band-off-diagonal (the matrix element of velocity operator $\boldsymbol{v}_{l'l}^0$ is diagonal in $\boldsymbol{k}$) responses, respectively.

B. Band-diagonal response

We first illustrate the aforementioned iterative procedure in its lowest order, where $C_{n,l} = ie\boldsymbol{E}\cdot\partial_{\boldsymbol{k}}f_{n-1,l}$, and $f_{n,ll'}$ is given by the second term on the right-hand side of Eq. (31). Plugging them into Eq. (30) leads to, after disorder average, the most conventional Boltzmann equation (16). Then, up to the third-order iteration the Boltzmann equations (17) and (18) also emerge after the disorder average, thus

$$\boldsymbol{j}_n^{\rm d} = e\sum_l F_{n,l}^{\rm L}\boldsymbol{v}_l^0 + e\sum_l F_{n,l}^{\rm csk}\boldsymbol{v}_l^0 + e\sum_l F_{n,l}^{\rm Gsk}\boldsymbol{v}_l^0 + e\sum_l F_{n,l}^{\rm a1}\boldsymbol{v}_l^0 + e\sum_l F_{n,l}^{\rm a2}\boldsymbol{v}_l^0. \tag{36}$$

Most details of the iteration procedure have in fact already been presented in previous papers [23,25,38,39], and are also provided in the Supplemental Material [32] for the convenience of the interested readers.

It is apparent that in the higher orders of the iteration $C_{n,l}$ contains the combination effect of the electric field and disorder, which leads finally to the additional driving term related to $\delta^{\boldsymbol{E}}\omega_{l'l}^{(2)}$. In the linear anomalous Hall effect, only the coordinate-shift-related component, namely, $\delta_1^{\boldsymbol{E}}\omega_{l'l}^{(2)}$, survives in the resulting Boltzmann equation, as has been elaborated in Refs. [23,25,39]. On the other hand, in nonlinear responses the additional driving term related to $\delta_2^{\boldsymbol{E}}\omega_{l'l}^{(2)}$ is derived from the quantum kinetics, as is detailed in the Supplemental Material [32].

Lastly, we emphasize that the equilibrium density matrix deserves separate discussions. It should be obtained from the definition of the single-particle density matrix [32], and the leading value is $f_{0,ll'}=\delta_{ll'}f_{0,l}$, with $f_{0,l}$ the Fermi distribution. Note that the equilibrium density matrix is also altered by disorder and thus does not coincide with $f_{0,l}$. Through the $C_{1,ll'}$ term, the disorder-induced corrections to $f_{0,ll'}$ incorporate the effect of the $\boldsymbol{E}$ field during scattering into linear response. The neglect of this fact would lead to the absence of the $e\boldsymbol{E}\cdot\delta\boldsymbol{r}_{l'l}$ contribution to $f_{1,l}$. In fact, this is one of the main differences between the Kohn-Luttinger approach and another quantum kinetic approach employed recently to study the linear and nonlinear anomalous Hall effects [31,41]. More detailed discussions on this issue are presented later.

TABLE I. Correspondence of Boltzmann transport to the band-off-diagonal (od) and band-diagonal (d) responses of density matrix. $\boldsymbol{v}_l^{\rm bc}$ and $\boldsymbol{v}_l^{\rm sj}$ are the Berry-curvature and side-jump velocities, respectively. $\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l}$ is the $\boldsymbol{E}$-field-corrected scattering rate. $\omega^{(3)\rm as}_{ll'}$ and $\omega^{(4)\rm as}_{ll'}$ yield the skew scattering.

Semiclassical ingredients	Density-matrix response
$\boldsymbol{v}_l^{\rm bc}$	$\boldsymbol{j}_n^{\rm od}$
$\boldsymbol{v}_l^{\rm sj}$	$\boldsymbol{j}_n^{\rm od}$
$\delta^{\boldsymbol{E}}\omega^{(2)}_{l'l}=\delta_1^{\boldsymbol{E}}\omega^{(2)}_{l'l}+\delta_2^{\boldsymbol{E}}\omega^{(2)}_{l'l}$	$\boldsymbol{j}_n^{\rm d}$
$\omega^{(3)\rm as}_{ll'}$, $\omega^{(4)\rm as}_{ll'}$	$\boldsymbol{j}_n^{\rm d}$

C. Band-off-diagonal response

The leading nonzero contribution to the off-diagonal response $\boldsymbol{j}_n^{\rm od}$ is of $O(V^{-2n+2})$, given by the second-order iteration of Eq. (31),

$$\langle f_{n,ll'}\rangle_c=\frac{e\boldsymbol{E}\cdot\mathcal{A}_{ll'}\left(F^{\rm L}_{n-1,l'}-F^{\rm L}_{n-1,l}\right)}{\epsilon_l-\epsilon_{l'}-i\hbar ns}+\sum_{l''}{}'\frac{\langle V_{ll''}V_{l''l'}\rangle_c}{\epsilon_l-\epsilon_{l'}-i\hbar ns}\times\left[\frac{F^{\rm L}_{n,l}-F^{\rm L}_{n,l''}}{\epsilon_l-\epsilon_{l''}-i\hbar ns}-\frac{F^{\rm L}_{n,l''}-F^{\rm L}_{n,l'}}{\epsilon_{l''}-\epsilon_{l'}-i\hbar ns}\right],\tag{37}$$

and can be readily cast into [32]

$$\boldsymbol{j}_n^{\rm od}=e\sum_l F^{\rm L}_{n-1,l}\boldsymbol{v}_l^{\rm bc}+e\sum_l F^{\rm L}_{n,l}\boldsymbol{v}_l^{\rm sj}.\tag{38}$$

Here, $\boldsymbol{v}_l^{\rm bc}$ and $\boldsymbol{v}_l^{\rm sj}$ coincide respectively with the Berry-curvature anomalous velocity and side-jump velocity [35].

Summing up, our quantum kinetic theory shows that arbitrary nth-order response retains the same form of Eqs. (35), (36), and (38), namely, the semiclassical Boltzmann result Eq. (22), up to the first three leading-order contributions in the weak disorder potential. The correspondence of the basic ingredients in the Boltzmann theory to the density-matrix response is summarized in Table I. It is worthwhile to remind here that interband virtual processes play the essential role also in the band-diagonal response of the density matrix.

V. COMPARISON TO OTHER THEORIES

We start by noting that a similar idea to the intuitive consideration leading to the $\boldsymbol{E}$-field-influenced scattering matrix element Eq. (9) also appeared in two publications by Tarasenko [42,43]. In these two papers a nonlinear current arises due to the $\boldsymbol{E}$-field-induced admixture of excited conduction- and valence-band states to the ground-subband wave function in quantum wells. In fact, one can find that our Eqs. (7)–(9) are quite similar to Eqs. (13)–(15) in Ref. [42]. The difference is that, in Refs. [42,43] the electric-field component E_z (in the z direction of the quantum well) mixes the quantum-confined states, whereas in the present work the electric field E_x mixes the Bloch states of electrons.

Next, we compare our theory to the previous works on the Boltzmann formulation of the nonlinear Hall effect [26–29]. All these works just generalized the Boltzmann theory for the linear anomalous Hall effect [22] directly and phenomenologically into the nonlinear response. First, the $\delta_2^{\boldsymbol{E}}\omega^{(2)}_{l'l}$ term proposed in the present study has no counterpart in the linear response and thus is beyond such a direct generalization of the linear theory. Second, our quantum theory supports the form of the coordinate-shift-related $\delta_1^{\boldsymbol{E}}\omega^{(2)}_{l'l}$ speculated intuitively in Refs. [26,27,29], which differs from the one proposed in Ref. [28].

Our theory is also different from the other quantum kinetic one of the nonlinear Hall effect posted recently [31]. This latter theory is based on the nonlinear generalization of a linear-response density-matrix theory [41]. Thus in the following we discuss first the difference between this linear-response theory and ours, and then that between the theory of Ref. [31] and ours.

The whole second line of Eq. (36), which arises from the band-diagonal response of the density matrix, is missed in the linear-response theory of Ref. [41]. First, in this theory the equilibrium density matrix is identified to be just the Fermi distribution. However, as we have stressed in the last paragraph of Sec. IV B, the equilibrium density matrix is not equal to the Fermi distribution and has the disorder-induced correction. It is this correction that leads finally to the coordinate-shift-related $\delta_1^{\boldsymbol{E}}\omega^{(2)}_{l'l}$ and thus to $F^{\rm al}_{1,l}$ [23,25,39]. Second, the theory of Ref. [41] only considers the lowest-order Born approximation in calculating the scattering rate, thus $F^{\rm Gsk}_{1,l}$ is missed. The theory of Ref. [41] was shown to work well for the linear anomalous Hall effect in the spin-polarized Rashba model and for the spin Hall effect in the Rashba model. However, the peculiarity of the Rashba models in fact plays a basic role in this success: In the spin-polarized Rashba model $F^{\rm Gsk}_{n,l}+F^{\rm al}_{n,l}=0$ in the case of scalar pointlike impurities within the noncrossing approximation for $F^{\rm Gsk}_{n,l}$ [44], whereas in the Rashba model the diagonal element of the spin-current operator $\boldsymbol{j}^s$ in the Bloch representation is zero $[(\boldsymbol{j}^s)^0_l=0]$ [45]. Therefore, when applied to another model, such as the two-dimensional gapped Dirac model, one can check that the theory of Ref. [41] cannot reproduce the same anomalous Hall conductivity as the previous theories [21].

Now we turn to the theory of Ref. [31]. In the case of linear response, this theory still misses the disorder-induced correction to the equilibrium density matrix, thus missing the contribution from $F^{\rm al}_{1,l}$. To be more specific, one can check that the side-jump conductivity in the first equation of Eq. (26) of Ref. [31] is in fact only one half of the side-jump conductivity defined in Ref. [21]. Another half, namely, the contribution from $F^{\rm al}_{1,l}$, disappears: It is contained in neither the first nor the second equation of Eq. (26) of Ref. [31].

Because the linear-response density matrix is vital in producing the nonlinear-response one, the aforementioned difference makes the nonlinear theory of Ref. [31] also different from ours. While the band-off-diagonal response Eq. (38) is produced in Ref. [31], the second line of the band-diagonal response Eq. (36) is not. This means that only the Berry-curvature dipole and side-jump velocity contributions to the second-order nonlinear Hall effect proposed in the previous semiclassical theory [26–29] have been identified in the quantum kinetic theory of Ref. [31]. At the present stage only our theory establishes the consistency between the Boltzmann and quantum kinetics in nonlinear responses.

VI. CONCLUSION

In conclusion, we have proposed a modified Boltzmann framework for nonlinear electric transport, and identified an interband-coherence effect induced by dc electric fields during scattering. This effect has no counterpart in linear response, and thus is missed in the previous nonlinear Boltzmann formalism for the nonlinear Hall effect which is just the direct generalization of the linear Boltzmann theory. The proposed Boltzmann formulation has been confirmed by a quantum kinetic theory. This theory also shows that an arbitrary nth-order nonlinear response to a dc electric field, up to the first three leading contributions in the weak disorder potential, is handled by the same few gauge-invariant semiclassical ingredients.

ACKNOWLEDGMENTS

We thank T. Chai for helpful discussions. Q.N. is supported by DOE (DE-FG03-02ER45958, Division of Materials Science and Engineering) on the transport formulation in this work. C.X. is supported by NSF (EFMA-1641101) and Welch Foundation (F-1255).

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