Boltzmann transport from density matrix theory: interband and intraband coherences

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To account for the anomalous/spin Hall conductivities and spin-orbit torque in the zeroth order of electron scattering time in strongly spin-orbit coupled systems, the Boltzmann transport theory in the case of weak disorder-potentials has been augmented by adding some interband coherence effects by hand. In this work these interband coherence terms are derived systematically from analyzing the equation of motion of the single-particle density matrix in the Bloch representation. Interband elements of the out-of-equilibrium density matrix are related to only one part of interband-coherence responses. Disorder-induced off-diagonal elements of the equilibrium density matrix are shown to be vital in producing the coordinate-shift anomalous driving term in the modified Boltzmann equation. Moreover, intraband coherence is inherent in the Boltzmann equation, whose contribution to anomalous/spin Hall conductivities is parametrically the same as the interband coherence.

I. INTRODUCTION

The Boltzmann transport theory has been generally accepted as a qualitatively good and intuitive starting point in discussing nonequilibrium phenomena in weakly disordered crystals [1], including recent focuses on the anomalous Hall effect [2, 3], spin Hall effect [4, 7], valley Hall effect [8] and spin-orbit torque [9, 10]. In applications to these phenomena, the coherence between Bloch states in different bands, i.e., interband coherence, caused by both the electric field and disorder has been incorporated via semiclassical constructions (Berry-curvature anomalous velocity [12] and scattering-induced coordinate-shift [13, 14]) or semi-phenomenological arguments (scattering-induced interband-coherence dressing of carrier states [6, 10]). The resultant contribution to the linear-response coefficients in aforementioned nonequilibrium phenomena is of the zeroth order of the electron scattering time, and experiments usually suggest the dominance of this sort of contribution in moderately dirty transition metal samples [15, 16].

Despite the practical success, microscopic understanding of this augmented Boltzmann formalism has not been complete. Although Kohn and Luttinger have laid the foundation for the Boltzmann formalism of the anomalous Hall effect on a density matrix perturbation theory in the case of weak impurity potentials sixty years ago [19, 20], their classical paper is too complicated to be absorbed by most researchers in the modern community of spin and valley Hall effects and spin-orbit torque. Kohn and Luttinger, in the very early years of modern transport theories, aimed to lay a foundation not only for the Boltzmann theory of electrical conductivity but also for the whole metallic conduction theory. From the modern point of view, this aim is beyond the scope of the Kohn-Luttinger density matrix approach, which only works in limited parameter regime and suffers from the lack of a systematic renormalization scheme [21, 22]. Therefore, this approach is much less employed than other quantum transport approaches based on Green’s functions (e.g., Kubo-Streda and Keldysh [2]), into which systematic renormalization procedures can be incorporated.

On the other hand, if one only aims to derive, in the case of weak disorder-potentials, the Boltzmann formalism which was born in the Bloch representation of the disorder-free equilibrium single-particle Hamiltonian, then the density matrix approach is still the most intuitive starting point. In fact, when constructing the modified Boltzmann theory of anomalous Hall effect, Sinitsyn et al. [13] noticed the correspondence between the semiclassical coordinate-shift effects and the sum of some gauge-dependent equations of Luttinger [20]. However, the Boltzmann theory for the anomalous Hall effect cannot be directly used to account for spin Hall effect (when the spin is not conserved due to strong band-structure spin-orbit coupling) and spin-orbit torque. Recently Xiao et al. have argued that the scattering-induced interband-coherence dressing of carrier states contributes to the spin Hall conductivity and spin-orbit torque, playing the role of the side-jump velocity in the anomalous Hall effect. But this semi-phenomenological construction has not been confirmed by the density matrix theory.

In this work we show that, all the aforementioned interband-coherence terms added-by-hand in the Boltzmann formalism can be derived systematically from a density matrix perturbation analysis with respect to the weak disorder-potential [19, 20], in the case of Bloch electrons in non-degenerate multiple-bands scattered by weak Gaussian static disorder.

In particular, the disorder-dependent part of the interband elements of the out-of-equilibrium density-matrix leads to the disorder-induced interband-coherence dressing of Bloch states in the Boltzmann theory [6, 10]. The latter is the only part of interband coherence responses that arises from interband elements of the out-of-equilibrium density matrix. We emphasize this because it is sometimes misunderstood that the conventional Boltzmann formalism [1] only misses the interband elements of the out-of-equilibrium density matrix. In fact, the combination of the disorder-induced off-diagonal (in the Bloch representation \( |l\rangle = |\eta k\rangle \) with \( \eta \) the band index and \( k \) the momentum) elements of the \( \text{equilibrium} \) density matrix and the diagonal perturbations by the electric field leads to the coordinate-shift induced anomalous driving term...
in the modified Boltzmann equation \[23\].

The interband coherence process also occurs in the conventional Boltzmann equation as long as the scattering amplitude is calculated up to the second Born order \[8, 14\]. This so-called intrinsic-skew-scattering contribution \[14\] arises from asymmetric differential scattering cross-section on rare impurity pairs separated by distances of the order of the Fermi wavelength \[24–27\]. Therefore, contributions to the differential cross-section from both crossed and noncrossed impurity-lines of this two-impurity complexes are parametrically the same \[23, 20\]. Previous Boltzmann theories \[3, 14, 23\] only addressed the noncrossed contribution which involves an interband virtual scattering process. In this work we show explicitly that the crossed contribution involves not only interband but also intraband virtual scattering processes. The latter means coherence between Bloch states in the same band but with different energies, and an intermediate state of the virtual process lies away from the Fermi surface (off-shell).

The above main ideas are analyzed in Sec. II, with the main results given in Eqs. \((9) – (12)\), Eq. \((19)\) and Eqs. \((20) – (27)\). The paper is concluded by some discussions in Sec. III. Some calculation details are presented in the Appendix.

II. DERIVATION AND ANALYSIS

A. Preliminaries: Density matrix approach

In this subsection we just outline the basic framework of the density matrix equation-of-motion approach proposed by Kohn and Luttinger \[14, 20\] in the case of weak-potential static disorder.

We introduce the notation \(\hat{A}\) to stand for the representation of operator \(\hat{A}\) in the second-quantized formalism. For a single-carrier operator, i.e., \(\hat{A} = \sum_{\mathbf{i}} \hat{A}_{\mathbf{i}}\) where \(\hat{A}_{\mathbf{i}}\) depends only on the dynamical variables of the \(i\)-th carrier, one has \(\hat{A} = \sum_{n\nu} A_{n\nu} a_{\nu}^{\dagger} a_{\nu}^{\prime}\) where \(A_{n\nu}\) are the matrix elements in the \(n\) representation of single-carrier space, \(a_{\nu}^{\dagger}\) (or \(a_{\nu}\)) is the creation (annihilation) operator on the single-carrier eigenstate \(|\nu\rangle\). The expectation value of \(\hat{A}\) is given by \(\langle \hat{A} \rangle = Tr\left(\hat{\rho}_{T} \hat{A}\right)\), where \(Tr\) denotes the trace operation in the occupation-number space, and the many-particle density matrix \(\hat{\rho}_{T}\) in the occupation-number representation is governed by the quantum Liouville equation \(i\hbar \frac{\partial}{\partial t} \hat{\rho}_{T} = \left[\hat{H}_{T}, \hat{\rho}_{T}\right]\). The expectation value of a single-carrier operator \(\hat{A}\) can then be expressed in terms of \(\hat{A}\) and a single-carrier operator \(\hat{\rho}_{T}\):

\[
\langle A \rangle = \sum_{nn'} A_{nn'} \langle \hat{\rho}_{T} \rangle_{nn'} = tr\left(\hat{A} \hat{\rho}_{T}\right),
\]

\[
\langle \hat{\rho}_{T} \rangle_{nn'} = tr\left(\hat{\rho}_{T} a_{n}^{\dagger} a_{n'}\right). \tag{1}
\]

Here \(tr\) denotes the trace in single-carrier Hilbert space. As Kohn and Luttinger have noticed \[19\], when the total Hamiltonian is a single-carrier operator \(\hat{H}_{T} = \sum_{nn'} \left(\hat{H}_{T}\right)_{nn'} a_{n}^{\dagger} a_{n'}\), the equation of motion for \(\langle \hat{\rho}_{T} \rangle_{nn'}\) reads \(i\hbar \frac{\partial}{\partial t} \langle \hat{\rho}_{T} \rangle_{nn'} = \left[\hat{H}_{T}, \hat{\rho}_{T}\right]_{nn'}\). The \(n\) representation in the single-carrier Hilbert space is arbitrary thus

\[
i\hbar \frac{\partial}{\partial t} \hat{\rho}_{T} = \left[\hat{H}_{T}, \hat{\rho}_{T}\right] \tag{2}
\]

with the operators acting on the single-carrier space. \(\hat{\rho}_{T}\) satisfies \(\langle \hat{\rho}_{T} \rangle_{nn} = \langle N_{n} \rangle \geq 0\) and \(tr \hat{\rho}_{T} = N\), where \(N_{n} = a_{n}^{\dagger} a_{n}\) and \(N = \sum_{n} N_{n}\). Although normalized to the carrier number \(N_{c}\) instead of 1, \(\hat{\rho}_{T}\) is often referred to as the single-particle density matrix, the diagonal elements of which represent the average occupation numbers of single-particle eigenstates rather than occupation probability. This character implies that \(\hat{\rho}_{T}\) is a quantum-statistics generalization of the single-particle density function described by the classical Boltzmann equation, and the diagonal elements of \(\hat{\rho}_{T}\) may compose with a Boltzmann-type transport equation. This observation motivates one to split the quantum Liouville equation in the Bloch representation into diagonal and off-diagonal parts in the following.

The single-carrier Hamiltonian reads \(\hat{H}_{T} = \hat{H}_{0} + \hat{H}' + \hat{H}_{F}\), where \(\hat{H}_{0}\) is the single-particle free Hamiltonian, \(\hat{H}' = \lambda \hat{V}\) with \(\lambda\) a dimensionless parameter and \(\hat{V}\) the disorder potential, and the field term \(\hat{H}_{F} = \hat{H}_{1} e^{i t}\) with \(\hat{H}_{1} = -e \hat{E} \cdot \hat{r}\) arises from the electric field adiabatically switched-on from the remote past \(t = \infty\). The infinitesimal positive \(s\) in \(\hat{H}_{F}\) can be taken to be the same as the \(s\) which appears as a regularization factor in the \(T\)-matrix theory of the Boltzmann formalism \[14, 23\]. This is because the physical situation is obtained by taking the limit \(s \to 0^{+}\). We remind that a similar note on the infinitesimal positive \(s\) has appeared in the derivation of Kubo-Streda linear response formula with respect to the uniform static electric field \[28\].

In the linear response regime one can thus decompose \(\hat{\rho}_{T}\) into \[19\]

\[
\hat{\rho}_{T} = \hat{\rho} + \hat{f} e^{i t}, \tag{3}
\]

where \(\hat{\rho}\) is the equilibrium density matrix, \(\hat{f}\) is the out-of-equilibrium density matrix linear in the electric field at the time of interest \((t = 0)\). The linear response of a single-particle observable \(A\) thus reads \[29\]

\[
\delta A = tr\left(\hat{f} \hat{A}\right) = \sum_{l} \langle f_{l} \rangle A_{ll} + \sum_{l'w} \langle f_{l'w} \rangle A_{lw} \tag{4}
\]

in the eigenbasis of \(\hat{H}_{0}\), where the index \(l\) denotes the Bloch state. Hereafter \(\langle . \rangle\) stands for disorder average, and the notation \(\sum'\) means that all the index equalities should be avoided in the summation. For anomalous and spin Hall conductivities in the presence of weak Gaussian disorder, the leading contribution of \(\delta A\) is of \(O(\lambda^{0})\).
It is noticed that, $\rho_{ll'} \neq \rho_{ll'}^{(0)} \delta_{ll'}$ with $\rho_{ll'}^{(0)}$ the Fermi distribution function, because $\rho_{ll'}$ is altered by disorder and thus even possesses off-diagonal elements. The neglect of this fact would lead to the absence of an important interband-coherence process (Sec. II. C). In fact, $\hat{\rho}$ can be expanded in the Bloch representation as:

$$\rho_{ll'} = \rho_{ll'}^{(0)} + \rho_{ll'}^{(1)} + \rho_{ll'}^{(2)} + ..., \quad (5)$$

where the superscript means the order of $\lambda$. Here $\rho_{ll'}^{(0)} = \rho_{ll'}^{(0)}(\delta_{ll'}$ is known from the definition $\rho_{ll'}^{(0)} = Tr \left( \alpha_{l}^{\dagger} \alpha_{l} \rho^{(0)} \right)$. Disorder-induced corrections $\rho_{ll'}^{(1),(2)}$ can be obtained from an iterative solution to the quantum Liouville equation $\left[ \hat{H}_{0} + \hat{H}', \hat{\rho} \right] = 0$ obeyed by the equilibrium single-particle density matrix. The iteration gives $\left[ \hat{H}_{0}, \hat{\rho}^{(1)} \right] = \left[ \hat{\rho}^{(0)}, \hat{H}' \right]$ and $\left[ \hat{H}_{0}, \hat{\rho}^{(2)} \right] = \left[ \hat{\rho}^{(1)}, \hat{H}' \right]$, thus

$$\rho_{ll'}^{(1)} = \rho_{ll'}^{(0)} - \frac{\rho_{ll'}^{0}}{d_{ll'}} \hat{H}_{ll'}, \quad (6)$$

and $\rho_{ll'}^{(2)} = \sum_{l''} H_{ll'}^{l''} \frac{H_{l''}^{l''}}{d_{ll''}} \left( \delta_{l''l''} - \rho_{l''l''}^{0} - \rho_{l''l''}^{0} \right)$. Here $\rho_{ll}^{(1)} = H_{ll}^{l} \partial_{l''} \rho_{ll}^{(0)} = 0 \quad (H_{ll}^{l} = 0, \text{ see below})$ has been used, and $\rho_{ll'}^{(2)}$ can be obtained from $\lim_{\lambda \rightarrow 0} \rho_{ll'}^{(2)}$. Hereafter $d_{ll'} = \epsilon_{l} - \epsilon_{l'}$, $d_{ll'}^{\pm} = d_{ll'} \pm i \hbar \epsilon_{l}$ with $\epsilon_{l}$ the energy.

Now we turn to the out-of-equilibrium density matrix, which satisfies the quantum Liouville equation

$$d_{ll'} f_{ll'} = \sum_{l''} (f_{l''l'} H_{l''l'} - H_{l'l''} f_{l''l'}) + C_{ll'}, \quad (7)$$

in the Bloch representation. $C_{ll'} = \left[ \hat{\rho}, \hat{H}_{ll'} \right]$, combines the electric field and equilibrium density matrix, reading

$$C_{ll'} = i e E \cdot \left[ \partial_{l} + \partial_{l'} \right] \rho_{ll'} + [\mathbf{J}, \hat{\rho}_{ll'}] \quad (8)$$

for $l \neq l'$ and $C_{ll} = i e E \cdot \left[ \partial_{l} \rho_{ll'} + [\mathbf{J}, \hat{\rho}_{ll'}] \right]$, where $[\mathbf{J}, \hat{\rho}_{ll'}] = \sum_{l''} J_{ll''} \rho_{ll''} - \rho_{ll''} J_{ll''}$. Here $d_{ll'} = i \frac{\partial}{\partial \lambda} \hat{H}_{ll'} + i J_{ll'}$ and $J_{ll'} = \delta_{kk'} \langle u_{l} | \partial_{l} | u_{l'} \rangle$ are used. $|l\rangle$ is the Bloch state, $l = (\eta, k)$. Equation (7) can be split into

$$d_{ll'} f_{ll'} = \sum_{l''} (f_{l''l'} H_{l''l'} - H_{l'l''} f_{l''l'}) + (f_{l} - f_{l'}) H_{ll'} + C_{ll'}, \quad (9)$$

for $l \neq l'$, and

$$- i \hbar s f_{l} = \sum_{l''} (f_{l''l'} H_{l''l'} - H_{l'l''} f_{l''l'}) + C_{l}. \quad (10)$$

Here $H_{ll'}$, which is the first-order energy correction in the bare quantum mechanical perturbation theory, has been absorbed into $H_{kk'}$, thus $H_{kk'} = 0$ hereafter.

In the case of weak disorder-potential, an iterative analysis of Eqs. (9) and (10) in terms of the parameter $\lambda$ is possible. First a starting point for this iteration is needed. To do this one has to assume that, the most conventional Boltzmann equation (where the scattering amplitude is obtained under the lowest-order Born approximation) gives the leading order contribution to $f_{l}$, i.e., $f_{l}$ starts from the order of $\lambda^{-2}$. In other words, we demand the most conventional Boltzmann equation is at least qualitatively correct as a leading approximation of longitudinal electronic transport. This is always true for the electrical conductivity in the metallic regime where the Fermi energy is much larger than the disorder-induced band broadening. However, this assumption may break down for spin-orbit torques when there are multiple spin-orbit-split bands on the Fermi surface if the minimal interband splitting is smaller than the disorder-induced band broadening (weak spin-orbit-coupling regime), even in the metallic regime \[30\]. In that case the field-like torque may not be captured by the conventional Boltzmann equation (detailed discussions in Ref. \[31\]). Therefore, a necessary condition for the validity of the Boltzmann formalism is that the minimal interband splitting around the Fermi level is smaller than the disorder-induced band broadening.

Then $s_{f} \rightarrow 0$ when $s \rightarrow +^{0}$, if the electric field is turned on much more slowly than the scattering time \[19\] \[22\]. And $f_{ll'}$ starts from the order of $\lambda^{-1}$. Thus an order-by-order analysis with respect to the weak disorder potential follows:

$$f_{l} = f_{l}^{(-2)} + f_{l}^{(-1)} + f_{l}^{(0)} + ..., \quad (11)$$

$$f_{ll'} = f_{ll'}^{(-1)} + f_{ll'}^{(0)} + f_{ll'}^{(1)} \ldots \quad (l \neq l')$$

$$C_{ll'} = C_{ll'}^{(0)} + C_{ll'}^{(1)} + C_{ll'}^{(2)} + ... \; .$$

The iteration yields an equation only concerning the diagonal element $f_{l}$ and a series of equations expressing $f_{ll'}$ in terms of $f_{l}$ \[3] \[20\], e.g., $f_{ll'}^{(1)} = \frac{f_{ll'}^{(0)} - f_{ll'}^{(-2)}}{d_{ll'}} H_{ll'}$, and

$$f_{ll'}^{(2)} = \sum_{l''} H_{ll''} \frac{H_{l''l''}}{d_{ll''}} \left[ \frac{f_{l''l''}^{(-2)}}{d_{ll''}} - \frac{f_{l''l''}^{(-2)} - f_{l''l''}^{(2)}}{d_{ll''}} \right]$$

$$+ \frac{f_{l'}^{(-1)} - f_{l'}^{(1)}}{d_{ll'}} H_{ll'} + C_{ll'}^{(0)} \; . \quad (12)$$

The required transport equation for $\langle f_{l} \rangle$ is obtained by disorder averaging. In so doing, one assumes that $f_{l}$ does not contain any physically important, rapidly varying exponential factors, thus in the thermodynamic limit $\langle f_{l}^{(-2)} H_{ll'} \rangle = \langle f_{l}^{(-2)} \rangle \langle H_{ll'} \rangle$, $\langle H_{ll'} H_{l''l''} f_{l''l''}^{(-2)} \rangle = \langle H_{ll'} H_{l''l''} \rangle \langle f_{l''l''}^{(-2)} \rangle$. Only when this assumption is true, the semiclassical distribution function and thus the Boltzmann formalism can be defined. The validity of this assumption has been confirmed \[22\], but beyond the
we throw away all the terms which still exist in the case where \( \hat{D} \) the scattering \([13, 31, 32]\). Thus one can recognize that the quantity contains combination effects of the electric field and disorder. These three equations just constitute the modified T-matrix in the single-particle scattering theory. The symmetric parts \( (\omega^2_{l''} - \omega^2_{l'}) \) of \( \omega^2_{l''} \) and \( \omega^2_{l'} \) only contribute to the spin-orbit induced Hall transport in \( O(\lambda^2) \), and thus can be neglected in the case of weak disorder potential. In contrast, the anti-symmetric parts \( (\omega^2_{l''} - \omega^2_{l'}) \) of \( \omega^2_{l''} \) and \( \omega^2_{l'} \) contribute to the spin-orbit induced Hall transport in \( O(\lambda^2) \), the leading order in the case of Gaussian disorder. Assuming isotropic systems \( \sum \rho_{l''} \), one has

\[
\begin{align*}
\omega^2_{l''} = 2\frac{\pi}{\hbar} \left| (T_{l''})^2 \right| \delta (d_{l''}) = \omega^2_{l''} + \omega^2_{l'} + ..., \quad (17)
\end{align*}
\]

where \( T_{l''} \) is the T-matrix in the single-particle scattering theory. The symmetric parts \( (\omega^2_{l''} = \frac{1}{2} (\omega^2_{l'} + \omega^2_{l''}) \) and \( \omega^2_{l''} \) of the higher-order scattering rates \( \omega^2_{l''} \), \( \omega^2_{l'} \), and \( \omega^2_{l''} \) only contribute to the spin-orbit induced Hall transport in \( O(\lambda^2) \), and thus can be neglected in the case of weak disorder potential. In contrast, the anti-symmetric parts \( (\omega^2_{l''} = \frac{1}{2} (\omega^2_{l'} - \omega^2_{l''}) \) of \( \omega^2_{l''} \) and \( \omega^2_{l'} \) contribute to the spin-orbit induced Hall transport in \( O(\lambda^2) \), the leading order in the case of Gaussian disorder. Assuming isotropic systems \( \sum \rho_{l''} \), one has

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\end{align*}
\]
respectively. Due to $v_{ll'} \delta_{kk'} = -\frac{1}{\hbar} d_{ll'} \mathbf{J}_{ll'}$ for $l \neq l'$, we have
\[ \sum_{ll'} C_{ll'}^{(0)} A_{ll'} / d_{ll'} = \sum_{ll} \rho_{ll}^{(0)} \delta_{ll} A_{ll}, \]
where
\[ \delta_{ll} A_{ll} = -\hbar E \sum_{ll'} 2 \text{Im} \langle u_{ll} | \hat{\psi} | u_{ll'} \rangle \delta_{kk'} A_{ll'} / d_{ll'} \] (20)
coincides with the correction to $A_{ll}$ due to electric-field-induced interband mixing of Bloch states introduced in the Boltzmann theory [6, 9, 10]. Besides, by interchanging the indices $l$ and $l'$ here and there and some simple algebra, we find
\[ \sum_{ll'} \langle H_{ll'}^{1} H_{ll'}^{1} \rangle \left( \frac{f^{(-2)}_{l} - f^{(-2)}_{l'}}{d_{ll'}^{2}} - \frac{f^{(-2)}_{l'} - f^{(-2)}_{l}}{d_{ll'}} \right) A_{ll} \]
\[ = \sum_{ll'} \langle H_{ll'}^{1} H_{ll'}^{1} \rangle A_{ll} / d_{ll'} + c.c. \]
\[ + \sum_{ll'} \langle f^{(-2)}_{l} \rangle \langle H_{ll'}^{1} H_{ll'}^{1} \rangle A_{ll'} \left( \frac{1}{d_{ll'}} - \frac{1}{d_{ll}} \right) \frac{1}{d_{ll'}} \]
\[ = \sum_{l} \langle f^{(-2)}_{l} \rangle \delta^{xx} A_{ll}, \]
where
\[ \delta^{xx} A_{ll} = \sum_{ll'} \left[ 2 \text{Re} \frac{\langle H_{ll'}^{1} H_{ll'}^{1} \rangle A_{ll}}{d_{ll'}^{2} d_{ll'}^{2}} + \frac{\langle H_{ll'}^{1} H_{ll'}^{1} \rangle A_{ll'}}{d_{ll'}^{2} d_{ll'}} \right] \] (22)
coincides with the correction to $A_{ll}$ due to disorder-induced interband mixing of Bloch states introduced semi-phenomenologically in the Boltzmann theory [6, 10]. Therefore, in the weak disorder-potential regime
\[ \sum_{ll'} \langle f^{(-2)}_{ll'} \rangle A_{ll} = \sum_{l} \rho_{ll}^{(0)} \delta_{ll} A_{ll} + \sum_{l} \langle f^{(-2)}_{l} \rangle \delta_{ll} A_{ll}. \] (23)
In the anomalous Hall effect ($\hat{A} = \hat{\psi}$), $\delta_{ll} v_{ll}$ and $\delta^{xx} v_{ll}$ are just the Berry-curvature anomalous velocity and side-jump velocity [11, 10, 13], respectively. Thus the anomalous driving term [16] is just $e \mathbf{E} \cdot \delta^{xx} \mathbf{v}_{ll} \delta_{ll} \rho_{ll}^{(0)}$, of which the interband-coherence nature is apparent.

We also note that in a recent alternative density matrix treatment to interband-coherence responses [33], where only interband elements of the out-of-equilibrium density-matrix are considered, their main results (Eqs. (45), (47) and (48) in Ref. [33]) just correspond to our Eq. (23).

C. Anomalous driving term and off-diagonal elements of the equilibrium density matrix

Now we look into the anomalous driving term $C^{(0)}_{ll'}$. $C^{(0)}_{ll'}$, $C^{(1)}_{ll'}$, and $C^{(2)}_{ll'}$ contain both the diagonal ($\hat{D}$, $\hat{\psi}$, and $\mathbf{J}_{ll}$) and off-diagonal ($\mathbf{J}_{ll'}$) components of the electric-field perturbation $\hat{H}_{ll}$ in the Bloch representation, as well as diagonal and off-diagonal components of the equilibrium density matrix. We notice that, if we demand that only the diagonal (also band-diagonal) components of the electric-field perturbation contribute to the final form of $C^{(0)}_{ll'}$, i.e., we only preserve $C^{(0)}_{ll'} \sim 0, C^{(2)}_{ll'} \sim ie \mathbf{E} \cdot \partial_{k} \rho^{(2)}_{ll}$ and $C^{(1)}_{ll'} \sim ie \mathbf{E} \cdot (\hat{D} \rho^{(1)}_{ll} + (\mathbf{J}_{ll} - J_{ll'}) \rho^{(1)}_{ll'})$ (here $l \neq l'$), then we directly arrive at Luttinger’s expression for $C^{(0)}_{ll'}$. If we further demand that only the off-diagonal elements of the equilibrium density matrix survive in the final form of $C^{(0)}_{ll'}$, thus $C^{(2)}_{ll'} \sim 0$ and
\[ C^{(0)}_{ll'} = \sum_{ll'} \left[ \langle C^{(1)}_{ll'} H_{ll'}^{1} \rangle / d_{ll'} - c.c. \right] \] (24)
with
\[ C^{(1)}_{ll'} = ie \mathbf{E} \cdot (\hat{D} \rho^{(1)}_{ll} + (\mathbf{J}_{ll} - J_{ll'}) \rho^{(1)}_{ll'}), \] (25)
and throw away the trivial renormalization terms, we again obtain Eq. (16). Therefore, the anomalous driving term (16) adopted in the modified Boltzmann formalism results in fact from the combination of off-diagonal elements of the equilibrium density matrix and the diagonal electric-field perturbations.

D. Interband and intraband virtual scattering contributions to intrinsic-skew-scattering

In previous Boltzmann theories [3, 10], the anti-symmetric part $\omega^{(4)}_{ll'}$ of $\omega^{(4)}_{ll'}$ was calculated within the noncrossing approximation [14, 23]. Here we show that the crossed part of $\omega^{(4)}_{ll'}$ corresponds to the recently identified Hall contribution of X and $\Psi$ diagrams in the Kubo diagrammatic approach [24, 25].

Starting from Eq. (A11) we have (set $\lambda = 1$ hereafter)
\[ \omega^{(4)}_{ll'} = -\frac{2\pi}{\hbar} \delta (d_{ll'}) \sum_{ll'} \left[ \text{Im} \langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle \text{Im} \frac{1}{d_{ll'}, d_{ll'}} \right. \]
\[ + \text{Im} \langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle \text{Im} \frac{1}{d_{ll'}, d_{ll'}} \]
\[ \left. + \text{Im} \langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle \text{Im} \frac{1}{d_{ll'}, d_{ll'}} \right]. \]

When taking the disorder average in the case of Gaussian disorder, there exist both the non-crossed (nc) and crossed (c) contributions [14, 25], thus $\omega^{(4)}_{ll'} = \omega^{(4)}_{ll'}^{nc} + \omega^{(4)}_{ll'}^{c}$. For example, $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ contains noncrossed contribution [14, 25] $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ and crossed contribution $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ [24, 25] corresponding to the so-called X diagram [24], while $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ implies $l = l'$ and thus does not contribute to $\omega^{(4)}_{ll'}$. $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ contains noncrossed contributions [14, 25] $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ and $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ as well as crossed contribution $\langle V_{ll'} V_{ll'} V_{ll'} V_{ll'} \rangle$ [24, 25].
Alternatively, we have
\[ \omega_{4l'}^{\alpha} = -\frac{(2\pi)^2}{\hbar} \delta(d_{l'}) \sum_{l'',l'''} \delta(d_{l''}) \left[ \Im \langle V_{l'l''} V_{l''l'''} V_{l'''}l'l' \rangle \right. \]
\[ + \Im \langle V_{l'l''} V_{l''l'''} V_{l'''}l'l' \rangle \] 
from which we see that the \( l, l' \) and \( l'' \) states lie on the mass shell, while the \( l''' \) state does not \[27\]. This means that virtual off-shell scattering are indispensable for intrinsic-skew-scattering in the presence of Gaussian disorder. This observation was first made in analyzing the Feynman diagrams \[25-27\].

To be definite, we consider slowly varying scalar impurity potentials \[26\], then
\[ V_{ll'} = V_{kk'} \left[ \delta_{ll'} + \delta_{ll'} J_{l''l'(k)} + \frac{1}{2} \delta_{ll'} \delta_{ll'} J_{l''l''l'''l'''}(k) + \ldots \right], \]
where \( J_{l''l'(k)} = k_{l''} - k_{l'} \), \( J_{l''l''l'''l'''}(k) = \langle u_{ll'} \delta_{ll'} | u_{l''l'''} \rangle \) and \( J_{l''l''l'''l'''}(k) = \langle u_{ll'} \delta_{ll'} \delta_{ll'} \delta_{ll'} | u_{l''l'''} \rangle \). The Einstein summation convention is used hereafter for the indices \( \mu, \nu \).

The noncrossed part contributes
\[ \omega_{4l'}^{\alpha-nc} = \frac{(2\pi n_{im} V_0^2)}{2\hbar} \sum_{l',l''} \delta_{ll'} \delta_{ll'} \delta(d_{l'}) \]
\[ \times \sum_{l''} \delta_{ll'} \delta_{ll'} \delta_{ll'} (\delta_{ll'} + \delta_{ll'} + \delta_{ll'}) \]
\[ \times \Im \left[ J_{ll'}^{\eta\eta'}(k) J_{\eta\eta'}^{\eta\eta}(k) \right], \tag{26} \]
where \( V_0 \) is the averaged scattering strength, \( n_{im} \) is the impurity density, \( \langle k \times k \rangle_{\mu\nu} \equiv k_{l''} k_{l'} - k_{l'} k_{l''} \). Thus an interband off-shell scattering \( \eta'' \neq \eta \) is unavoidable in each term of the noncrossing intrinsic-skew-scattering \[11, 14\]. More specifically, in Eq. \[(26)\] for \( \omega_{4l'}^{\alpha-nc} \), the contribution related to \( \delta_{ll'} \delta_{ll'} + \delta_{ll'} \) corresponds to the sum of the middle parts of the \( l' \) Feynman diagrams in the last row (first and second rows) of Fig. 11 in Ref. \[14\].

For the crossed contribution, we get
\[ \omega_{4l'}^{\alpha-c} = -\frac{(2\pi n_{im} V_0^2)}{2\hbar} \delta_{ll'} \delta_{ll'} \delta(d_{l'}) \]
\[ \times (k \times k')_{\mu\nu} \delta(d_{l'}) \sum_{l''} \delta_{ll'} \delta_{ll'} (\delta_{ll'} + \delta_{ll'} + \delta_{ll'}) \]
\[ \times \left\{ \Im \left[ J_{ll'}^{\eta\eta'}(k) J_{\eta\eta'}^{\eta\eta}(k) - \delta_{\eta\eta'} \Omega_{\mu\nu}(k) \right] \right\}, \tag{27} \]
which contains both intraband \((\eta'' = \eta)\) and interband \((\eta'' \neq \eta)\) terms. \( \Omega_{\mu\nu}(k) \) is the momentum-space Berry curvature. We note that Eq. \[(27)\] was already obtained by Luttinger sixty years ago \[24\], but has been unnoticed \[8, 13\] in the recent researches on the crossed contribution to anomalous and spin Hall effects. But Luttinger missed the noncrossing term \( \omega_{4l'}^{\alpha-nc} \). In Eq. \[(27)\], the contributions related to \( \delta_{ll'} \delta_{ll'} \) and \( \delta_{ll'} \delta_{ll'} + \delta_{ll'} \) correspond to the middle parts of the X diagram and two \( \Psi \) diagrams \[21, 22, 27\], respectively. The presence of crossed intrinsic-skew-scattering reveals the fact that, not only interband off-shell processes but also intraband off-shell processes are important for anomalous and spin Hall effects as well as related phenomena.

### III. DISCUSSION

We conclude by discussing certain limitations of the present consideration.

In the case of weak disorder-potential, the off-diagonal response only concerns the lowest nonzero order of \( \langle f_{l'l'} \rangle \), while the analysis of \( \langle f_{l'l'} \rangle \) has to go to higher orders in the perturbation expansion in terms of the disorder potential. Because this expansion is basically a bare perturbation theory, some trivial renormalization terms are unavoidable in high orders of this expansion. These terms should be eliminated systematically by a renormalization procedure, if one aims at placing the density matrix equation-of-motion theory as a nonperturbative description in the entire metallic region. Such a renormalization treatment has been shown for non-relativistic free electrons \[21, 22\], but has never been done for Bloch electrons according to our literature knowledge. On the other hand, we only regard the density matrix approach as a foundation of the Boltzmann theory in the case of a very weak disorder potential. In this case the aforementioned trivial renormalization effects are much smaller high-order corrections (at least \( O(\lambda^3) \) for spin-orbit induced transport coefficients), and can thus be neglected.

The present consideration shows that the modified Boltzmann formalism \[3, 10, 13, 14\] only works in the presence of weak disorder-potentials. This is much more limited than the usually accepted regime of (qualitative) validity of a Boltzmann theory \( \hbar/\tau < \Delta \) (this is a necessary condition for the validity of the Kohn-Luttinger expansion), where \( \tau \) is the electron lifetime and \( \Delta \) is the minimal interband-splitting around the Fermi level \[30, 34\]. This implies that there exists another sort of modified Boltzmann formalism in the presence of very dilute impurities, because large \( \tau \) can be obtained by not only weak disorder potential but also small impurity density. In fact this kind of theory also started from Luttinger and Kohn \[35\] who tried to formulate the transport equation according to the order of impurity density. But this Boltzmann theory is complicated enough because it is nonperturbative with respect to the disorder potential, especially when concerning the distribution function in the zeroth order of impurity density \[35\]. In particular, this nonperturbative nature indicates that the Gaussian disorder approximation cannot well describe dilute impurities with strong scattering potentials. New characteristics of transport, differing from those discussed above in moderately dirty samples with weak disorder-potentials,
are naturally expected in the regime of dilute impurity and strong disorder-potential (as revealed in recent Kubo diagrammatic calculations). 

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Appendix A: Calculation details

The expressions for $\omega_{ll'}^{(4)}$ and $\omega_{ll'}^{(4)}$ are

$$\omega_{ll'}^{(4)} = \frac{2\pi}{\hbar} \sum_{ll'} \delta (d_{ll'}) \left[ \frac{\langle H_{ll'}^{(4)} \rangle}{d_{ll'}} \delta (d_{ll'}) + \frac{\langle H_{ll'}^{(4)} \rangle}{d_{ll'}} \delta (d_{ll'}) + \frac{\langle H_{ll'}^{(4)} \rangle}{d_{ll'}} \delta (d_{ll'}) \right],$$

$$\omega_{ll'}^{(4)} = \frac{2\pi}{\hbar} \sum_{ll'} \delta (d_{ll'}) \left[ \frac{\langle H_{ll'}^{(4)} \rangle}{d_{ll'}} \delta (d_{ll'}) + \frac{\langle H_{ll'}^{(4)} \rangle}{d_{ll'}} \delta (d_{ll'}) + \frac{\langle H_{ll'}^{(4)} \rangle}{d_{ll'}} \delta (d_{ll'}) \right].$$

(A1)

In fact, obtaining this equation is not easy, if one does not expect Eq. [17] in advance. Starting from the expressions for $C_{ll'}^{(0)}$, $C_{ll'}^{(1)}$, and $C_{ll'}^{(2)}$ presented in the main text, after some delicate steps we recover Luttinger’s expression [20] for $C_{ll'}^{(2)}$:

$$C_{ll'}^{(2)} = i e E \left[ \frac{\partial \rho_{ll}}{\partial \epsilon_l} \delta (d_{ll'}) + \sum_{l'} \left[ \langle H_{ll'}^{(2)} \rangle \delta (d_{ll'}) \frac{\partial \rho_{ll}}{\partial \epsilon_l} + \frac{\langle H_{ll'}^{(2)} \rangle}{\delta (d_{ll'})} \frac{\partial \rho_{ll}}{\partial \epsilon_l} + \frac{\langle H_{ll'}^{(2)} \rangle}{\delta (d_{ll'})} \frac{\partial \rho_{ll}}{\partial \epsilon_l} \right] \right],$$

which can also be expressed as

$$C_{ll'}^{(2)} = i e E \sum_{ll'} \omega_{ll'}^{(2)} \delta (d_{ll'}) \left( - \frac{\partial \rho_{ll}}{\partial \epsilon_l} \delta (d_{ll'}) + i e E \left[ \frac{\partial \rho_{ll}}{\partial \epsilon_l} \delta (d_{ll'}) \frac{\partial \rho_{ll}}{\partial \epsilon_l} + \frac{\langle H_{ll'}^{(2)} \rangle}{\delta (d_{ll'})} \frac{\partial \rho_{ll}}{\partial \epsilon_l} + \frac{\langle H_{ll'}^{(2)} \rangle}{\delta (d_{ll'})} \frac{\partial \rho_{ll}}{\partial \epsilon_l} \right] \right).$$

The second term on the right hand side still exists in the case of non-relativistic free electrons without spin-orbit coupling. It contains some trivial renormalization effects (e.g., $\epsilon_{l}^{(2)}$ is the second-order energy correction in the bare quantum mechanical perturbation theory) and is not important to spin-orbit induced transport. Thus we only preserve $C_{ll'}^{(2)} = i e E \sum_{ll'} \omega_{ll'}^{(2)} \delta (d_{ll'}) \left( - \frac{\partial \rho_{ll}}{\partial \epsilon_l} \right)$ which is of qualitative importance for spin-orbit induced transport [2, 10, 14].

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