Hall vs Ohmic as the only proper generic partition of the nonlinear current

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November 10, 2021

Abstract

The symmetric and antisymmetric parts of the linear conductivity describe the dissipative (Ohmic) and nondissipative (Hall) parts of the current. The Hall current is always transverse to the applied electric field regardless of its orientation; the Ohmic current is purely longitudinal in cubic crystals, but in lower-symmetry crystals it has a transverse component whenever the field is not aligned with a principal axis. In this work, we extend that analysis beyond the linear regime. We consider all possible ways of partitioning the current at any order in the electric field without taking symmetry into account, and find that the Hall vs Ohmic decomposition is the only one that satisfies certain basic requirements. A simple prescription is given for achieving that decomposition.

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1 Introduction

A static electric field applied to a conducting crystal generates a current density that may be written to linear order as

$$j^{(1)}_\alpha = \sigma_{\alpha\beta} E_\beta,$$

where a summation over Cartesian index $\beta$ is implied, and $\sigma_{\alpha\beta}$ is understood to be a function of the magnetic field $B$ if one is present. In general $j^{(1)}$ is not parallel to $E$, but under certain conditions it may contain a part that is always perpendicular to $E$, irrespective of how the field is oriented relative to the crystal axes. This Hall current is described by the antisymmetric part of the conductivity tensor,

$$j^{(1)}_{H,\alpha} = \sigma^H_{\alpha\beta} E_\beta, \quad \sigma^H_{\alpha\beta} = \frac{1}{2} (\sigma_{\alpha\beta} - \sigma_{\beta\alpha}),$$

and the remainder $j^{(1)}_O = j^{(1)} - j^{(1)}_{H}$, given by the symmetric part of the conductivity, is the Ohmic current that gives rise to energy dissipation via Joule heating,

$$j^{(1)}_O,\alpha = \sigma^O_{\alpha\beta} E_\beta, \quad \sigma^O_{\alpha\beta} = \frac{1}{2} (\sigma_{\alpha\beta} + \sigma_{\beta\alpha}).$$

Building on ground-breaking optical studies [1–3], there is at present renewed interest in nonlinear effects in solids arising from broken symmetries [4]. The transport effects that are being actively investigated include unidirectional magnetoresistance (both induced by a magnetic field [5–7] and spontaneous [8]), and various nonlinear Hall effects [9–22].

Despite the surge of interest in nonlinear currents in solids, the basic question of how to extend the Hall vs Ohmic decomposition of Eqs. (2) and (3) to the nonlinear regime has received little attention. With the present work we aim to fill in this gap by providing a sharp answer to that question, and by placing it in the broader context of how to partition the nonlinear current into well-defined parts. Although we will base the discussion on the conductivity tensor, our phenomenological analysis applies equally well to the resistivity tensor.

To motivate the problem, consider the second-order response

$$j^{(2)}_\alpha = \sigma_{\alpha\beta\gamma} E_\beta E_\gamma.$$  

Contrary to the linear conductivity $\sigma_{\alpha\beta}$, the quadratic conductivity $\sigma_{\alpha\beta\gamma}$ is not uniquely defined; adding to it a correction of the form

$$\Delta \sigma_{\alpha\beta\gamma} = -\Delta \sigma_{\alpha\gamma\beta}$$

does not change the current. We will refer to this type of freedom in defining nonlinear conductivities as a “gauge freedom,” and to the unique choice that satisfies $\sigma_{\alpha\beta\gamma} = \sigma_{\alpha\gamma\beta}$ as the “symmetric gauge.” (In the language of nonlinear optics, the symmetric gauge is the one where the conductivity tensor has intrinsic permutation symmetry [23].)

By analogy with Eq. (2), one might attempt to define $\sigma^H_{\alpha\beta\gamma}$ as the part of $\sigma_{\alpha\beta\gamma}$ that is antisymmetric in either the first and second indices,

$$\sigma^{1,2}_{\alpha\beta\gamma} = \frac{1}{2} (\sigma_{\alpha\beta\gamma} - \sigma_{\beta\alpha\gamma}),$$
or in the first and third,
\[ \sigma^{1,3}_{\alpha\beta\gamma} = \frac{1}{2} (\sigma_{\alpha\beta\gamma} - \sigma_{\gamma\alpha\beta}). \] (7)

(Note that we need to choose between these two options, since imposing both conditions would render \( \sigma^{H}_{\alpha\beta\gamma} \) totally antisymmetric, resulting in zero current.) Both choices yield Hall-like transverse currents. However, we will see shortly that not only do they give different currents, but those currents depend on the initial gauge choice for \( \sigma_{\alpha\beta\gamma} \). This problem can be fixed by switching to the symmetric gauge, \( \sigma_{\alpha\beta\gamma} = \frac{1}{2} (\sigma_{\alpha\beta\gamma} + \sigma_{\gamma\alpha\beta}) \), before applying the antisymmetrization (6) or (7). Since the resulting Hall-like conductivities
\[ \sigma^{1,2}_{\alpha\beta\gamma} = \frac{1}{4} (\sigma_{\alpha\beta\gamma} + \sigma_{\alpha\gamma\beta} - \sigma_{\beta\alpha\gamma} - \sigma_{\gamma\beta\alpha}) \] (8)
and
\[ \sigma^{1,3}_{\alpha\beta\gamma} = \frac{1}{4} (\sigma_{\alpha\beta\gamma} + \sigma_{\alpha\gamma\beta} - \sigma_{\beta\alpha\gamma} - \sigma_{\gamma\beta\alpha}) \] (9)
only differ by a gauge transformation, they yield the same current. This modified prescription \([15,20]\) is nevertheless still not quite correct, as we will now show.

As a concrete example, consider the quadratic conductivity obtained by solving the Boltzmann equation at \( B = 0 \) in the constant relaxation-time approximation \([9,12,19]\),
\[ \sigma_{\alpha\beta\gamma} = \frac{e^3}{h^2} \int \frac{d^3k}{(2\pi)^3} \sum_n f_0(\epsilon_{kn}) \left( \partial_\gamma \Omega_{\alpha\beta}^{kn} - (\tau/h) \partial^3_{\alpha\beta\gamma} \epsilon_{kn} \right). \] (10)
Here \( \tau \) is the relaxation time, \( \epsilon_{kn} \) is the band energy, \( f_0(\epsilon_{kn}) \) is the Fermi-Dirac distribution function, \( \Omega_{\alpha\beta}^{kn} = -\Omega_{\beta\alpha}^{kn} \) is the Berry curvature of a Bloch state, and \( \partial_\gamma \) stands for \( \partial/\partial k_\gamma \). The (anomalous) Hall part of the response is given by the first term, and it is conventionally expressed as
\[ j^{(2)}_{H,\alpha} = \frac{e^3}{h^2} D_{\alpha\beta\gamma} E_\beta E_\gamma, \] (11)
where
\[ D_{\alpha\beta\gamma} = \int \frac{d^3k}{(2\pi)^3} \sum_n f_0(\epsilon_{kn}) \partial_\gamma \Omega_{\alpha\beta}^{kn}. \] (12)
Applying to Eq. (10) each of the proposed prescriptions for extracting the Hall part of \( \sigma_{\alpha\beta\gamma} \), we obtain
\[ \left( j^{1,2}, j^{1,3}, j^{1,2} = j^{2,1} \right) = (1,1/2,3/4) j^{(2)}_{H}. \] (13)
for the associated Hall currents. The first prescription gives the full Hall current, but that is accidental: if we make the gauge transformation \( \sigma_{\alpha\beta\gamma} \rightarrow \sigma_{\alpha\gamma\beta} \) in Eq. (10), \( j^{1,2} \) and \( j^{1,3} \) get swapped: \( j^{1,2} \rightarrow j^{1,3} \rightarrow (1/2,1) j^{(2)}_{H} \). Multiplying the right-hand sides of Eqs. (8) and (9) by factors of 4/3 does lead to generally valid expressions for the quadratic Hall conductivity, as will be shown in Sec. 3.

The above strategies constitute attempts to generalize to third-rank tensors the definition in Eq. (2) of an antisymmetric tensor that fail to yield a proper current decomposition. On the other hand, higher-order generalizations of the symmetrization procedure in Eq. (3) are

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1Equation (10) is missing a quadratic Hall-like term coming from the field-induced correction to the Berry curvature \([11]\). We will comment further on that term in Sec. 5.
straightforward, since one can symmetrize over all indices. In the case of the quadratic conductivity, one finds

$$\sigma^{O}_{\alpha\beta\gamma} = \frac{1}{6} \left( \sigma_{\alpha\beta\gamma} + \sigma_{\alpha\gamma\beta} + \sigma_{\beta\alpha\gamma} + \sigma_{\beta\gamma\alpha} + \sigma_{\gamma\alpha\beta} + \sigma_{\gamma\beta\alpha} \right).$$

(14)

It can be readily checked that the power dissipation is fully accounted for by $\sigma^{O}_{\alpha\beta\gamma}$,

$$j^{(Q)} \cdot E = \sigma_{\alpha\beta\gamma} E_\alpha E_\beta E_\gamma = \sigma^{O}_{\alpha\beta\gamma} E_\alpha E_\beta E_\gamma,$$

(15)

which justifies calling it the Ohmic quadratic conductivity. Accordingly,

$$\sigma^{H}_{\alpha\beta\gamma} = \sigma_{\alpha\beta\gamma} - \sigma^{O}_{\alpha\beta\gamma}$$

(16)

describes the dissipationless (Hall) quadratic response. Surprisingly, we could not find any explicit mention of this simple prescription in the growing literature on nonlinear currents in solids.

Let us apply the prescription above to Eq. (10). Since the first term therein is antisymmetric in two indices, it drops out from Eq. (14); and since the second is already totally symmetric, it becomes the full $\sigma^{O}_{\alpha\beta\gamma}$. We immediately conclude that the latter term is Ohmic, and the former is Hall-like.

It is important to note, however, that we not yet proven that Eqs. (14) and (16) give the only valid decomposition of the quadratic current into Ohmic and Hall parts. For example, one could define another partition

$$\tilde{\sigma}^{H}_{\alpha\beta\gamma} = (1 - x)\sigma^{H}_{\alpha\beta\gamma}, \quad \tilde{\sigma}^{O}_{\alpha\beta\gamma} = \sigma^{O}_{\alpha\beta\gamma} + x\sigma^{H}_{\alpha\beta\gamma} \quad (x \in \mathbb{R})$$

(17)

that is not related to that of Eqs. (14) and (16) by any gauge transformation, and again $\tilde{\sigma}^{H}_{\alpha\beta\gamma}$ would describe a dissipationless current, with all the Joule heating coming from $\tilde{\sigma}^{O}_{\alpha\beta\gamma}$.

In the present article, we prove that the partition of the current into Hall and Ohmic parts is in fact unique. To that end, we address the following question:

How many generic ways are there of breaking down the nonlinear current into well-defined separate parts?

(By “generic” we mean a prescription that does not take into account the symmetries of the system.) To address this question, we start by formulating in Sec. 2 the necessary criteria for a well-defined generic partition of the current at arbitrary order in $E$. In Sec. 3 we find the unique decomposition at second order that fulfils those criteria, and confirm that it indeed corresponds to the Hall vs Ohmic decomposition of Eqs. (14) and (16). That decomposition is generalized to arbitrary order in Sec. 4. In Sec. 5 we draw conclusions, and in Appendix A we prove that the Hall vs Ohmic partition of the current is the only generic partition possible at every order in $E$.

2 Criteria for a proper generic partition of the current

Our strategy for partitioning the nonlinear current will be as follows. We start from a conductivity tensor $\sigma_{\alpha_0 \alpha_1 \ldots \alpha_n}$ describing the full $n$-th order response,

$$j^{(n)}_{\alpha_0} = \sigma_{\alpha_0 \alpha_1 \ldots \alpha_n} E_{\alpha_1} \ldots E_{\alpha_n},$$

(18)
and search for an operator $\hat{P}$ that selects part of this current. We want this operator to act order by order in the electric field; this means that its action on the full conductivity tensor should result in a linear combination of versions of that same tensor with different sets of indices,
\[
(\hat{P}\sigma)_{\alpha_0\alpha_1\ldots\alpha_n} = \sum_p a_p \sigma_{p(0)\alpha_p(1)\ldots\alpha_p(n)}.
\] (19)

Here the summation is over all possible mappings
\[
\{0,1,\ldots,n\} \xrightarrow{p} \{p(0),p(1),\ldots,p(n)\}
\] (20)
where $p(n) \in \{0,1,\ldots,n\}$, and $a_p$ are coefficients to be determined. The part of the current selected by $\hat{P}$ can be written symbolically as
\[
(\hat{P}j^{(n)})_{\alpha_0} = (\hat{P}\sigma)_{\alpha_0\alpha_1\ldots\alpha_n} E_{\alpha_1} \ldots E_{\alpha_n}.
\] (21)

We shall require three properties of $\hat{P}$. The first is that it acts on the current as a projector, so that
\[
\hat{P}j^{(n)} = \hat{P}^2j^{(n)};
\] (22)
the second is that the projected current is invariant under gauge transformations of the full $n$-th order conductivity tensor, that is,
\[
\Delta(\hat{P}j^{(n)}) = 0
\] (23)
whenever $\Delta j^{(n)} = 0$, which in turn holds if and only if $\Delta\sigma_{\alpha_0\alpha_1\ldots\alpha_n}$ vanishes under symmetrization over the last $n$ indices.

Finally, we require that the projected current transforms as a vector under rotations of the coordinate system, so that $\hat{P}j^{(n)} \cdot \mathbf{E}$ remains invariant under such transformations. This is justified by the intention to arrive at a generic prescription that is not bound to any particular crystal symmetry, and not even to a specific number of spatial dimensions. This third constraint will be satisfied if the summation in Eq. (19) is restricted to permutation mappings $p$, for which $p(i) \neq p(j)$ whenever $i \neq j$. Conversely, if mappings with $p(i) = p(j)$ for some $i \neq j$ are included, scalar products will not be conserved under rotations.\(^2\) Thus, from here on we shall restrict our attention to permutation mappings, and investigate which operators $\hat{P}$ can satisfy the two conditions expressed by Eqs. (22) and (23).

Before proceeding, we note that if we find some operator $\hat{P}$ that satisfies the conditions above, those conditions will also be satisfied by $\hat{P}' = \hat{1} - \hat{P}$. Thus, any nontrivial operator $\hat{P}$ defines a decomposition of current into two parts (by “nontrivial” we mean an operator such that $\hat{P}j \neq 0$ and $\hat{P}j \neq j$). We will start by applying the above criteria to the second-order response, and then we will generalize to higher orders.

\(^2\)Take for example $\hat{P}_{H\sigma} = (\sigma_{\alpha\alpha} + \sigma_{\beta\beta})/2$. For an electric field lying on the $xy$ plane this gives $\hat{P}_{H\sigma}j^{(1)} \cdot \mathbf{E} = E_x^2\sigma_{xx} + E_y^2\sigma_{yy} + E_xE_y(\sigma_{xx} + \sigma_{yy})$, and the result should be the same in a different coordinate system. However, in a coordinate system that differs by a two-fold rotation about the $y$ axis we obtain $\hat{P}_{H\sigma}j^{(1)} \cdot \mathbf{E} = E_x^2\sigma_{xx} + E_y^2\sigma_{yy} - E_xE_y(\sigma_{xx} + \sigma_{yy})$, which is a different result.
3 Second-order response

Consider an operator \( \hat{P} \) acting on the quadratic conductivity according to Eq. (19),
\[
\hat{P}\sigma_{\alpha\beta\gamma} = a_0^+\sigma_{\alpha\beta\gamma} + a_0^-\sigma_{\alpha\gamma\beta} + a_1^+\sigma_{\gamma\alpha\beta} + a_1^-\sigma_{\beta\alpha\gamma} + a_2^+\sigma_{\beta\gamma\alpha} + a_2^-\sigma_{\gamma\beta\alpha},
\]
and on the quadratic current according to Eq. (21),
\[
\hat{P}j^{(2)}_\alpha = (\hat{P}\sigma_{\alpha\beta\gamma}) E_\beta E_\gamma = (A_0\sigma_{\alpha\beta\gamma} + A_1\sigma_{\beta\alpha\gamma} + A_2\sigma_{\beta\gamma\alpha}) E_\beta E_\gamma.
\]
Here \( A_i = a_i^+ + a_i^- \), and the notation for the coefficients \( a_i^\pm \) in Eq. (24) is as follows: the subscript denotes the position of \( \alpha \) in the permutation of the indices \( \alpha\beta\gamma \), and the superscript gives the parity of the permutation.

Our claim is that \( \hat{P} \) yields a proper generic partition of the current only if it satisfies Eqs. (22) and (23). Let us start with the gauge-invariance condition (23). The projected current (25) remains unchanged under the gauge transformation (5) if and only if
\[
(A_1 - A_2) E_\beta E_\gamma \Delta \sigma_{\beta\alpha\gamma} = 0;
\]
and since this condition must be satisfied for arbitrary \( E \), and since we did not set any rules for permutations involving the first index of \( \Delta \sigma \), it follows that \( A_1 = A_2 \). To impose the idempotency condition (22), we first apply Eq. (24) recursively to find
\[
\hat{P}^2\sigma_{\alpha\beta\gamma} = \tilde{a}_0^+\sigma_{\alpha\beta\gamma} + \tilde{a}_0^-\sigma_{\alpha\gamma\beta} + \tilde{a}_1^+\sigma_{\gamma\alpha\beta} + \tilde{a}_1^-\sigma_{\beta\alpha\gamma} + \tilde{a}_2^+\sigma_{\beta\gamma\alpha} + \tilde{a}_2^-\sigma_{\gamma\beta\alpha},
\]
where
\[
\begin{align*}
\tilde{a}_0^+ &= a_0^+ a_0^- + a_0^- a_0^+ + a_1^- a_1^+ + 2a_2^+ a_1^- + a_2^- a_2^+, \\
\tilde{a}_0^- &= 2a_0^+ a_0^- + a_1^+ a_1^- + a_1^- a_2^+ + a_1^+ a_2^- + a_2^+ a_2^-, \\
\tilde{a}_1^+ &= 2a_0^+ a_1^- + a_0^- a_1^+ + a_0^- a_2^- + a_1^+ a_2^+ + a_2^- a_2^+, \\
\tilde{a}_1^- &= 2a_0^+ a_1^- + a_0^- a_1^+ + a_0^- a_2^- + a_1^+ a_2^- + a_2^- a_2^+, \\
\tilde{a}_2^+ &= 2a_0^+ a_2^- + a_0^- a_1^+ + a_0^- a_2^- + a_1^+ a_1^- + a_1^+ a_2^+, \\
\tilde{a}_2^- &= 2a_0^+ a_2^- + a_0^- a_1^+ + a_0^- a_2^- + a_1^+ a_1^- + a_1^+ a_2^+.
\end{align*}
\]

By analogy with Eq. (25) we have
\[
\hat{P}^2 j^{(2)}_\alpha = (\tilde{A}_0\sigma_{\alpha\beta\gamma} + \tilde{A}_1\sigma_{\beta\alpha\gamma} + \tilde{A}_2\sigma_{\beta\gamma\alpha}) E_\beta E_\gamma
\]
for the twice-projected current, where the coefficients \( \tilde{A}_i = \tilde{a}_i^+ + \tilde{a}_i^- \) are given by
\[
\begin{align*}
\tilde{A}_0 &= A_0^2 + (a_1^- + a_2^+)A_1 + (a_1^+ + a_2^-)A_2, \\
\tilde{A}_1 &= A_0 A_1 + (a_0^+ + a_2^-)A_1 + (a_0^- + a_2^+)A_2, \\
\tilde{A}_2 &= A_0 A_2 + (a_0^+ + a_1^-)A_1 + (a_0^- + a_1^+)A_2.
\end{align*}
\]
Equating (25) and (29), the idempotency condition becomes \( A_i = \tilde{A}_i \) for \( i = 0, 1, 2 \). Substituting Eq. (30) for \( \tilde{A}_i \) and then invoking the gauge invariance condition \( A_1 = A_2 \), we are left with two conditions only,
\[
A_0 = A_0^2 + 2A_1^2, \quad A_1 = (2A_0 + A_1)A_1.
\]
These equations have four solutions. There are two solutions with \( A_1 = 0 \),

\[
\begin{align*}
\hat{P}_0 : (A_0, A_1 = A_2) &= (0, 0) \\
\hat{P}_1 : (A_0, A_1 = A_2) &= (1, 0)
\end{align*}
\]

\( \Rightarrow j^{(2)} = 0 + j^{(2)} \), \hspace{1cm} (32)

which as indicated give the trivial “all or nothing” partition of the current. Then there are two solutions with \( A_1 \neq 0 \),

\[
\begin{align*}
\hat{P}_H : (A_0, A_1 = A_2) &= (\frac{2}{3}, -\frac{1}{3}) \\
\hat{P}_O : (A_0, A_1 = A_2) &= (\frac{1}{3}, \frac{1}{3})
\end{align*}
\]

\( \Rightarrow j^{(2)} = j^{(2)}_H + j^{(2)}_O \), \hspace{1cm} (33)

which give the desired Hall vs Ohmic partition. To show that this is the case, we turn to the condition that defines a Hall-like projected current,

\[
\hat{P}j^{(2)} \cdot E = 0, \hspace{0.5cm} \forall E.
\] \hspace{1cm} (34)

Using Eq. (25) that condition becomes \( A_0 + A_1 + A_2 = 0 \), which is satisfied by \( \hat{P}_H \) but not by \( \hat{P}_O \). This conclude the proof that Eqs. (22) and (23) lead to a partition of the quadratic current into Hall and Ohmic parts. Remarkably, we found that this is in fact the only gauge-invariant and idempotent generic partition possible, apart from the trivial one in Eq. (32).

Since we are still free to adjust the six coefficients \( a_i^\pm \) in Eq. (24) as long as \( A_i = a_i^+ + a_i^- \) maintain the values given in Eq. (33), the Hall and Ohmic quadratic conductivities are highly non-unique. This corresponds precisely to the gauge freedom (5) in defining \( \sigma^H_{\alpha\beta\gamma} \) and \( \sigma^O_{\alpha\beta\gamma} \). One way to fulfill the “Ohmic” conditions in Eq. (33) is by setting all six coefficients in Eq. (24) to \( 1/6 \), which leads to the fully symmetric form in Eq. (14) for the quadratic Ohmic conductivity.

Let us now revisit the prescriptions in Eqs. (8) and (9) for defining the quadratic Hall conductivity, which consist in first symmetrizing the full quadratic conductivity in the last two indices, and then antisymmetrizing the first index with either the second or the third \([15,20]\). When applied to a concrete example in Sec. 2, those prescriptions only recovered three quarters of the full Hall current [see Eq. (13)]. This suggests it may be possible to fix them by multiplying each of Eqs. (8) and (9) by a factor of \( 4/3 \),

\[
\begin{align*}
\sigma^H_{\alpha\beta\gamma}^{(1,2)} &= \frac{4}{3} \sigma^{1,2}_{\alpha\beta\gamma} = \frac{1}{3} \left( \sigma_{\alpha\beta\gamma} + \sigma_{\alpha\gamma\beta} - \sigma_{\beta\alpha\gamma} - \sigma_{\beta\gamma\alpha} \right), \\
\sigma^H_{\alpha\beta\gamma}^{(1,3)} &= \frac{4}{3} \sigma^{1,3}_{\alpha\beta\gamma} = \frac{1}{3} \left( \sigma_{\alpha\beta\gamma} + \sigma_{\alpha\gamma\beta} - \sigma_{\gamma\beta\alpha} - \sigma_{\gamma\alpha\beta} \right).
\end{align*}
\]

\( \sigma^H_{\alpha\beta\gamma}^{(1,2)} \) and \( \sigma^H_{\alpha\beta\gamma}^{(1,3)} \) in the case of Eq. (35), and

\[
\begin{align*}
\sigma^H_{\alpha\beta\gamma}^{(1,2)} &= \frac{4}{3} \sigma^{1,2}_{\alpha\beta\gamma} = \frac{1}{3} \left( \sigma_{\alpha\beta\gamma} + \sigma_{\alpha\gamma\beta} - \sigma_{\beta\alpha\gamma} - \sigma_{\beta\gamma\alpha} \right), \\
\sigma^H_{\alpha\beta\gamma}^{(1,3)} &= \frac{4}{3} \sigma^{1,3}_{\alpha\beta\gamma} = \frac{1}{3} \left( \sigma_{\alpha\beta\gamma} + \sigma_{\alpha\gamma\beta} - \sigma_{\gamma\beta\alpha} - \sigma_{\gamma\alpha\beta} \right).
\end{align*}
\]

Comparing with Eq. (24) we find

\[
a_0^+ = a_0^- = a_1^- = -a_2^+ = \frac{1}{3}, \hspace{1cm} a_1^+ = a_2^- = 0
\]

\( \) in the case of Eq. (35), and

\[
a_0^+ = a_0^- = a_1^- = -a_2^+ = \frac{1}{3}, \hspace{1cm} a_1^+ = a_2^- = 0
\]

\( \) in the case of Eq. (36). Both sets of coefficients satisfy the Hall-like conditions in Eq. (33), and hence Eqs. (35) and (36) are indeed valid expressions for the quadratic Hall conductivity.
To conclude, let us count the number of independent coefficients needed to describe the quadratic Hall conductivity. The full quadratic conductivity can be chosen to be symmetric in the last two indices and hence it has 6 (18) independent components in 2D (3D), while the Ohmic part can be chosen to be fully symmetric and hence it has 4 (10) independent components in 2D (3D). Thus, the Hall part has \( 6 - 4 = 2 \) (18 - 10 = 8) independent components in 2D (3D). Those components can be repackaged as a pseudovector in 2D, and as a traceless rank-2 pseudotensor in 3D [15]. The latter is given by

\[
\chi^H_{\gamma\eta} = \frac{1}{2} \epsilon_{\alpha\beta\gamma} \sigma_{\alpha\beta\eta} \quad \Leftrightarrow \quad \chi^H_{\alpha\beta\gamma} = \frac{4}{3} \epsilon_{\alpha\beta\eta} \chi^H_{\eta\gamma},
\]

where \( \sigma_{\alpha\beta\eta} = \tau_{\alpha\eta\beta} \) is the quadratic conductivity in the symmetric gauge.

### 4 Higher-order responses

At \( n \)-th order in the electric field, the Ohmic conductivity can be chosen as the fully symmetrized conductivity tensor obtained by setting in Eq. (19) \( a_p = 1/(n + 1)! \) for all \( p \),

\[
\sigma^O_{\alpha_0\alpha_1...\alpha_n} = \hat{P}_O \sigma_{\alpha_0\alpha_1...\alpha_n} = \frac{1}{(n + 1)!} \sum_p \sigma_{\alpha(p)\alpha_{p(1)}...\alpha_{p(n)}}.
\]

This generalizes to arbitrary \( n \) the symmetrization procedure of Eqs. (3) and (14) for \( n = 1 \) and \( n = 2 \), respectively.

Let us now show that with the above choice of Ohmic projector, the Hall projector \( \hat{P}_H = 1 - \hat{P}_O \) satisfies Eqs. (22) and (23). We start again with the gauge invariance condition. Since the full \( n \)-th order current is by definition invariant under a gauge transformation \( \Delta \sigma_{\alpha_0\alpha_1...\alpha_n} \), the Hall part is invariant if and only if the Ohmic part is invariant. It is therefore sufficient to show that

\[
\Delta \left( \hat{P}_O j^{(n)}_{\alpha_0} \right) = \left( \hat{P}_O \Delta \sigma_{\alpha_0\alpha_1...\alpha_n} \right) E_{\alpha_1} ... E_{\alpha_n}
\]

vanishes for arbitrary \( E \). But since \( \Delta \sigma_{\alpha_0\alpha_1...\alpha_n} \) must vanish under symmetrization over the last \( n \) indices to ensure that \( \Delta j^{(n)}_{\alpha_0} = 0 \) (see Sec. 2), it also vanishes under full symmetrization by \( \hat{P}_O \). Next, it is clear that \( \hat{P}_O^2 \sigma_{\alpha_0\alpha_1...\alpha_n} = \hat{P}_O \sigma_{\alpha_0\alpha_1...\alpha_n} \) because symmetrization of tensor that is already fully symmetric does not change it further. Therefore,

\[
\hat{P}_O^2 j^{(n)} = \left( 1 - 2 \hat{P}_O + \hat{P}_O^2 \right) j^{(n)} = \left( 1 - \hat{P}_O \right) j^{(n)} = \hat{P}_H j^{(n)}.
\]

Finally, from the \( n \)-th order generalization of Eq. (15) it follows that \( j^{(n)} = j^{(n)} - j^{(n)}_{\alpha_0} \) is dissipationless. Thus we have obtained a solution that satisfies Eqs. (22) and (23) at any order in \( E \), and found that it corresponds to the Hall vs Ohmic partition of the current.

To recapitulate, one can always define the Ohmic part of the \( n \)-th order conductivity as the totally symmetric part, and the Hall part as the remainder. For \( n = 1 \), this procedure reduces to the standard partition of the linear conductivity according to Eqs. (2) and (3). We demonstrated in Sec. 3 that for \( n=2 \) the same procedure leads to the only well-defined (idempotent and gauge-invariant) generic partition of the quadratic current, and in Appendix A we generalize that proof to arbitrary \( n \).

\[\text{In Ref. [15] it is stated that the 3D quadratic Hall conductivity has nine independent components, not eight, and the tracelessness of the associated rank-2 pseudotensor is not stated (even if in the concrete example given in Eq. (8) therein that tensor is indeed traceless). We also note that the second term in that equation is not needed, since the “Berry curvature dipole” tensor is always traceless [13, 14].}\]
5 Conclusions

In this work we have shown how, given a conductivity tensor of arbitrary order \( n \) in the electric field, the current may be uniquely separated into Hall and Ohmic parts

\[
\mathbf{j}^{(n)} = \mathbf{j}_{\text{H}}^{(n)} + \mathbf{j}_{\text{O}}^{(n)}
\]

by taking linear combinations of that conductivity tensor with permuted indices. This separation is insensitive to the particular gauge choice for the conductivity tensor, and applying it multiple times gives the same result as applying it only once. No other generic order-by-order partition of the induced current fulfills these two requirements. Thus, once we have separated the Hall and Ohmic parts we cannot make any further subdivisions of the current into physically meaningful parts without invoking the symmetries of the system.

At linear order in \( \mathbf{E} \), the Hall vs Ohmic decomposition is intimately related to time-reversal symmetry \( T \) by virtue of the Onsager reciprocity relation

\[
\sigma_{\alpha\beta}(\mathbf{B}, \mathbf{M}) = \sigma_{\beta\alpha}(-\mathbf{B}, -\mathbf{M}).
\]

It follows from this relation that the Ohmic part of the linear response is even under time-reversal symmetry \( T \), while the Hall part is odd \([24, 25]\). In the nonlinear regime, this clear separation between Hall and Ohmic responses on the basis of \( T \) symmetry does not always hold. Take for example the quadratic response: the first term in Eq. (10) is Hall-like and \( T \)-even, and the second is Ohmic and \( T \)-odd; however, there is an additional contribution coming from the field-induced change in Berry curvature that is also \( T \)-odd but Hall-like \([11]\). Thus, while in nonmagnetic acentric crystals the quadratic current is purely Hall-like, in magnetically-ordered acentric crystals it generally contains both Hall and Ohmic components that compete with one another \([22]\). We believe that the procedure described in this work could be of use in isolating the Hall and Ohmic components of the nonlinear current measured in such systems.

Funding information Work by S.S.T. was supported by the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation program (ERC-StG-Neupert757867-PARATOP), and by Grant No. PP00P2-176877 from the Swiss National Science Foundation. Work by I.S. was supported by Grant No. FIS2016-77188-P from the Spanish Ministerio de Economía y Competitividad.

A Uniqueness of the partition at arbitrary order

In this Appendix we prove that the Hall vs Ohmic partition of the current described in Sec. 4 is the only valid generic partition at arbitrary order \( n \) in the electric field. We start with the general expression in Eq. (19) for the action of the operator \( \hat{P} \) on the conductivity,

\[
\hat{P}\sigma_{\alpha_0\alpha_1...\alpha_n} = \sum_p a_p\sigma_{\alpha_p(0)\alpha_p(1)...\alpha_p(n)},
\]

(45)
where the sum is over all permutations \( \{p(1), \ldots, p(n)\} \) of \( \{0, 1, \ldots, n\} \). The generalization of Eq. (25) for the action of \( \hat{P} \) on the current reads

\[
\hat{P} J_{\alpha_0}^{(n)} = (A_0 \sigma_{\alpha_0 \alpha_1 \ldots \alpha_n} + A_1 \sigma_{\alpha_1 \alpha_0 \ldots \alpha_n} + \ldots + A_n \sigma_{\alpha_1 \ldots \alpha_n \alpha_0}) E_{\alpha_1} \ldots E_{\alpha_n},
\]

where

\[
A_i = \sum_{p} a_p.
\]

Since they fully determine the projected current, the \( A_i \) are the only physically meaningful parameters, and changes in the parameters \( a_p \) that leave every \( A_i \) invariant amount to gauge transformations.

Recall from Sec. 2 that a general gauge transformation \( \Delta \sigma_{\alpha_0 \ldots \alpha_n} \) of the conductivity tensor must satisfy the condition

\[
\sum_q \Delta \sigma_{\alpha_0 \alpha_q(1) \ldots \alpha_q(n)} = 0,
\]

where the summation is over all permutations \( \{q(1), \ldots, q(n)\} \) of \( \{1, \ldots, n\} \). As stated in Eq. (23), we want the projected current to be invariant under all possible gauge transformations. To make progress, it is sufficient to require at this point invariance under the subset of gauge transformations \( \Delta \sigma_{\alpha_0 \alpha_1 \ldots \alpha_n}^i \) that are antisymmetric under permutation of the indices at positions \( i \) and \( i + 1 \),

\[
\Delta \sigma_{\alpha_0 \ldots \alpha_{i-1} \alpha_{i+1} \alpha_i+2 \ldots \alpha_n}^i = -\Delta \sigma_{\alpha_0 \ldots \alpha_{i-1} \alpha_i+1 \alpha_{i+2} \ldots \alpha_n}^i,
\]

where \( 0 < i < n \). For such transformations, the gauge invariance condition on the projected current (46) takes the form

\[
(A_i - A_{i+1}) \Delta \sigma_{\alpha_{i+1} \ldots \alpha_0 \alpha_{i+1} \alpha_i+2 \ldots \alpha_n}^i E_{\alpha_1} \ldots E_{\alpha_n} = 0.
\]

This condition can hold in general if and only if \( A_i = A_{i+1} \), and by letting the index \( i \) run from 1 to \( n - 1 \) we get

\[
A_1 = A_2 = \ldots = A_n.
\]

Therefore, the two parameters \( A_0 \) and \( A_1 \) fully determine the projected current.

Let us turn now to the idempotency condition (22). Acting with \( \hat{P} \) on both sides of Eq. (45) we obtain the following generalization of Eq. (27),

\[
\hat{P}^2 \sigma_{\alpha_0 \alpha_1 \ldots \alpha_n} = \sum_{p} \tilde{a}_p \sigma_{\alpha_{p(0)} \alpha_{p(1)} \ldots \alpha_{p(n)}} = \sum_{p_1, p_2} a_{p_1} a_{p_2} \sigma_{\alpha_{p(0)} \alpha_{p(1)} \ldots \alpha_{p(n)}},
\]

and hence the idempotency condition becomes \( A_i = \tilde{A}_i \) for \( i = 0, \ldots, n \) where, by analogy with Eq. (47),

\[
\tilde{A}_i \equiv \sum_{p} p(i) = 0 \tilde{a}_i = \sum_{p} \sum_{p_2, p_1} a_{p_1} a_{p_2}.
\]

Solving this equation for arbitrary \( n \) is not as easy as solving it for \( n = 2 \) [Eq. (30)]. But having settled the gauge invariance conditions in Eq. (51), we can now pick a convenient
gauge for the coefficients $a_p$. (This entails no loss of generality, because we study the action of $\bar{P}$ on the physical current, not on a particular form of the conductivity tensor.) We choose the most symmetric gauge compatible with Eq. (51), namely, the gauge where all terms in the summand of Eq. (47) are identical,

$$a_p = \begin{cases} 
A_0/n!, & \text{if } p(0) = 0 \\
A_1/n!, & \text{if } p(0) \neq 0 
\end{cases} \tag{54}$$

Substituting in Eq. (53), the idempotency condition $A_i = \tilde{A}_i$ becomes

$$A_i = \frac{1}{n!^2} \sum_{p} \left( \sum_{p_1,p_2} A_0^2 + \sum_{p_1,p_2} A_0A_1 + \sum_{p_1,p_2} A_1A_0 + \sum_{p_1,p_2} A_1A_1 \right), \tag{55}$$

with $i$ running from 0 to $n$. Due to Eq. (51), the $n$ equations with $1 \leq i \leq n$ are identical, leaving two equations only. These can be written as

$$A_0 = \frac{1}{n!^2} (aA_0^2 + bA_0A_1 + cA_1^2), \quad A_1 = \frac{1}{n!^2} (dA_0^2 + eA_0A_1 + fA_1^2), \tag{56}$$

where the coefficients $a$ to $f$ are the numbers of pairs of permutations $p_1,p_2$ of the set \{0,1,...,n\} that satisfy the conditions

$$a, d : \quad p_1(0) = p_2(0) = 0, \tag{57a}$$
$$b, e : \quad (p_1(0) = 0 \land p_2(0) \neq 0) \lor (p_1(0) \neq 0 \land p_2(0) = 0), \tag{57b}$$
$$c, f : \quad p_1(0) \neq 0 \land p_2(0) \neq 0, \tag{57c}$$

together with

$$a, b, c : \quad p_2(p_1(0)) = 0, \tag{57d}$$
$$d, e, f : \quad p_2(p_1(1)) = 0. \tag{57e}$$

It now becomes a straightforward combinatorial exercise to obtain

$$a = n!^2, \quad d = 0$$
$$b = 0, \quad e = 2 \cdot n!^2$$
$$c = n \cdot n!^2, \quad f = (n-1) \cdot n!^2, \tag{58}$$

which leads to the following generalization of Eq. (31),

$$A_0 = A_0^2 + nA_1^2, \quad A_1 = 2A_0A_1 + (n - 1)A_1^2. \tag{59}$$

Apart from the trivial solutions $\bar{P}_0$ and $\bar{P}_1$ of the same type as in Eq. (32), these equations have the solutions

$$\begin{cases} 
\bar{P}_H : (A_0, A_1 = \ldots = A_n) = \left( \frac{n}{n+1}, -\frac{1}{n+1} \right) \\
\bar{P}_O : (A_0, A_1 = \ldots = A_n) = \left( \frac{1}{n+1}, \frac{1}{n+1} \right) \tag{60}
\end{cases}$$

which generalize Eq. (33). It can be readily verified that the solution for $\bar{P}_O$ is satisfied by Eq. (40). And since $\bar{P}_H$ fulfills the Hall condition (34) but $\bar{P}_O$ does not, we have obtained a unique partition of the $n$-th order current into Hall and Ohmic components.
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