

**Supplemental Material for
“Gyrotropic Magnetic Effect and the Magnetic Moment on the Fermi Surface”**

I. KUBO FORMULA FOR NATURAL GYROTROPY IN CRYSTALS

The electronic structure of the crystal is treated at the independent-particle level taking into account the spin-orbit interaction. The Pauli Hamiltonian has the form^{S1,S2}

$$\mathcal{H}_0 = \frac{p^2}{2m_e} + V(\mathbf{r}) + \frac{\hbar}{4m_e^2} (\partial_{\mathbf{r}} V \times \mathbf{p}) \cdot \boldsymbol{\sigma}, \quad (\text{S1})$$

where $V(\mathbf{r}) = V(\mathbf{r} + \mathbf{R})$ is the periodic crystalline potential, \mathbf{p} is the canonical momentum, and $\boldsymbol{\sigma}$ is the vector of Pauli matrices. The kinematic momentum associated with \mathcal{H}_0 is

$$\boldsymbol{\pi} = \frac{m_e}{i\hbar} [\mathbf{r}, \mathcal{H}_0] = \mathbf{p} + \frac{\hbar}{4m_e} \boldsymbol{\sigma} \times \partial_{\mathbf{r}} V, \quad (\text{S2})$$

and it satisfies $[r_i, \pi_j] = [r_i, p_j] = i\hbar\delta_{ij}$.

In the presence of an electromagnetic field with vector potential $\mathbf{A}(\mathbf{r}, t)$ the Hamiltonian becomes^{S2}

$$\mathcal{H} = \frac{1}{2m_e} (\mathbf{p} + e\mathbf{A})^2 + V(\mathbf{r}) + \frac{\hbar}{4m_e^2} [\partial_{\mathbf{r}} V \times (\mathbf{p} + e\mathbf{A})] \cdot \boldsymbol{\sigma} + g_s \frac{\mu_B}{2} (\partial_{\mathbf{r}} \times \mathbf{A}) \cdot \boldsymbol{\sigma}, \quad (\text{S3})$$

where m_e and $-e$ are the electron mass and charge, $g_s = 2.0023$ is the spin g factor of the electron, and $\mu_B = e\hbar/2m_e$ is the Bohr magneton. Expanding Eq. (S3) and comparing with Eq. (S2) we find $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I + \mathcal{O}(A^2)$, where

$$\mathcal{H}_I = \frac{e}{2} (\mathbf{v} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{v}) + \frac{g_s e}{2m_e} (\partial_{\mathbf{r}} \times \mathbf{A}) \cdot \mathbf{S}. \quad (\text{S4})$$

Here $\mathbf{v} = \boldsymbol{\pi}/m_e$ is the velocity operator without the field,^{S2} and $\mathbf{S} = (\hbar/2)\boldsymbol{\sigma}$ is the spin operator. The first and second terms describe the orbital and spin (Zeeman) couplings, respectively. The interaction Hamiltonian of Eq. (S4) neglects orbital terms quadratic in \mathbf{A} , which do not contribute to the linear response of interest to us.

Consider an optical field $\mathbf{A}(t, \mathbf{r}) = \mathbf{A}(\omega, \mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r} - i\omega t}$. The current-density operator in the Fourier representation is

$$\mathbf{j} = -\frac{e}{2V} \{ [\mathbf{v} + (e/m_e)\mathbf{A}]e^{-i\mathbf{q}\cdot\mathbf{r}} + e^{-i\mathbf{q}\cdot\mathbf{r}}[\mathbf{v} + (e/m_e)\mathbf{A}] \} - i\frac{g_s e}{2m_e V} \mathbf{q} \times \mathbf{S}e^{-i\mathbf{q}\cdot\mathbf{r}}, \quad (\text{S5})$$

where V is the volume of the crystal. The first term is the orbital current, comprising paramagnetic (\mathbf{v}) and diamagnetic (\mathbf{A}) contributions;^{S3,S4} the diamagnetic term appears because the total orbital current is given by the velocity operator in the presence of the optical field, $\mathbf{v}^{\text{tot}} = (1/i\hbar)[\mathbf{r}, \mathcal{H}] = \mathbf{v} + (e/m_e)\mathbf{A}$. The last term in Eq. (S5) is the current density associated with the induced spin magnetization.

Expressing the total current density induced at linear order by the optical field as

$$j_i(\omega, \mathbf{q}) = \Pi_{ij}(\omega, \mathbf{q})\mathbf{A}(\omega, \mathbf{q}) \quad (\text{S6})$$

we find, following the standard perturbative calculation^{S4,S5} and setting $\hbar = 1$,

$$\Pi_{ij}(\omega, \mathbf{q}) = -\frac{e^2}{m_e} \sum_n \int [d\mathbf{k}] f_{\mathbf{k}n}^0 \delta_{ij} - e^2 \sum_{n,m} \int [d\mathbf{k}] \frac{f_{\mathbf{k}-\mathbf{q}/2,n}^0 - f_{\mathbf{k}+\mathbf{q}/2,m}^0}{\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + \omega + i\eta} M_{\mathbf{k}nm,ij}(\mathbf{q}), \quad (\text{S7})$$

where the first term gives the diamagnetic response. $[d\mathbf{k}] \equiv d^3k/(2\pi)^3$, $f_{\mathbf{k}n}^0 = f(\epsilon_{\mathbf{k}n})$ is the occupation of the Bloch eigenstate $|\psi_{\mathbf{k}n}\rangle = e^{i\mathbf{k}\cdot\mathbf{r}}|u_{\mathbf{k}n}\rangle$ in equilibrium, η is a positive infinitesimal, and the matrix element $M_{\mathbf{k}nm,ij}(\mathbf{q})$ reads

$$M_{\mathbf{k}nm,ij}(\mathbf{q}) = I_{\mathbf{k}mn,i}^*(\mathbf{q})I_{\mathbf{k}mn,j}(\mathbf{q}) \quad (\text{S8a})$$

$$\mathbf{I}_{\mathbf{k}mn}(\mathbf{q}) = \mathbf{I}_{\mathbf{k}mn}^{\text{orb}}(\mathbf{q}) + \mathbf{I}_{\mathbf{k}mn}^{\text{spin}}(\mathbf{q}) \quad (\text{S8b})$$

$$\mathbf{I}_{\mathbf{k}mn}^{\text{orb}}(\mathbf{q}) = \langle \psi_{\mathbf{k}+\mathbf{q}/2,m} | e^{i\mathbf{q}\cdot\mathbf{r}} \mathbf{v} + \mathbf{v} e^{i\mathbf{q}\cdot\mathbf{r}} | \psi_{\mathbf{k}-\mathbf{q}/2,n} \rangle / 2 = \langle u_{\mathbf{k}+\mathbf{q}/2,m} | \partial_{\mathbf{k}} H_{\mathbf{k}} | u_{\mathbf{k}-\mathbf{q}/2,n} \rangle \quad (\text{S8c})$$

$$\mathbf{I}_{\mathbf{k}mn}^{\text{spin}}(\mathbf{q}) = -(ig_s/2m_e) \langle u_{\mathbf{k}+\mathbf{q}/2,m} | \mathbf{q} \times \mathbf{S} | u_{\mathbf{k}-\mathbf{q}/2,n} \rangle. \quad (\text{S8d})$$

In Eq. (S8c) $H_{\mathbf{k}} \equiv e^{-i\mathbf{k}\cdot\mathbf{r}}\mathcal{H}_0e^{i\mathbf{k}\cdot\mathbf{r}}$, and we used $\partial_{\mathbf{k}}H_{\mathbf{k}} = \mathbf{v}_{\mathbf{k}} \equiv e^{-i\mathbf{k}\cdot\mathbf{r}}\mathbf{v}e^{i\mathbf{k}\cdot\mathbf{r}}$ and $\mathbf{v}_{\mathbf{k}\pm\mathbf{q}/2} = \mathbf{v}_{\mathbf{k}} \pm \mathbf{q}/2m_e$.^{S1} At $\mathbf{q} = 0$, Eq. (S7) reduces to the Kubo formula for the optical conductivity $\sigma_{ij}(\omega, 0) = (1/i\omega)\Pi_{ij}(\omega, 0)$ in the electric-dipole approximation.^{S4,S5} The dissipative (anti-Hermitian) part of Eq. (S7) corresponds to Eq. (40) in Ref. S6.

At nonabsorbing frequencies the response is purely reactive (i.e., Π_{ij} is Hermitian^{S7}) and we can set $\eta = 0$ in Eq. (S7). Optical gyrotropy is described by the antisymmetric part $\Pi_{ij}^A = (\Pi_{ij} - \Pi_{ji})/2$ at $\mathcal{O}(q)$.^{S8} We therefore take

$$\Pi_{ij}^A(\omega, \mathbf{q}) = -ie^2 \sum_{n,m} \int [d\mathbf{k}] \frac{f_{\mathbf{k}-\mathbf{q}/2,n}^0 - f_{\mathbf{k}+\mathbf{q}/2,m}^0}{\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + \omega} \text{Im} M_{\mathbf{k}nm,ij}(\mathbf{q}), \quad (\text{S9})$$

and Taylor expand it to first order in \mathbf{q} , in order to capture natural gyrotropy:

$$\Pi_{ij}^A(\omega, \mathbf{q}) = \Pi_{ij}^A(\omega, 0) + \Pi_{ijl}^A(\omega)q_l + \dots \quad (\text{S10})$$

Our goal is to calculate $\Pi_{ijl}^A(\omega \ll \epsilon_{\text{gap}}/\hbar)$, with $\epsilon_{\text{gap}}/\hbar$ the threshold for interband absorption. This low-frequency regime where the semiclassical description holds can be viewed the $\omega \rightarrow 0$ limit of the Kubo formula (Ref. S9, p. 253). In Sec. III we calculate $\Pi_{ijl}^A(\omega \rightarrow 0)$ from Eq. (S9) in the static limit (setting $\omega = 0$ before sending $\mathbf{q} \rightarrow 0$), and in the uniform limit (sending $\mathbf{q} \rightarrow 0$ before $\omega \rightarrow 0$). The uniform-limit results are valid in the frequency range $1/\tau \ll \omega \ll \epsilon_{\text{gap}}/\hbar$ where intraband as well as interband absorption is negligible; the effects of intraband scattering at lower frequencies are included heuristically in Sec. IV.

For future reference, we collect below the expressions for all \mathbf{q} -dependent quantities in Eq. (S9) up to $\mathcal{O}(q)$. In particular, we expand the matrix-element part as

$$\text{Im} M_{\mathbf{k}nm,ij}(\mathbf{q}) \simeq \text{Im} M_{\mathbf{k}nm,ij}(0) + \text{Im}(M_{\mathbf{k}nm,ijl})q_l, \quad (\text{S11})$$

and consider separately the orbital and spin contributions, and the intraband ($m = n$) and interband ($m \neq n$) parts. Using a simplified notation where $|u_{\mathbf{k}n}\rangle \rightarrow |n\rangle$ and $\partial_{k_i} \rightarrow \partial_i$, and defining $v_{ni} = \partial_i \epsilon_n$ and $S_{ni} = \langle n | S_i | n \rangle$, we find

$$f_{\mathbf{k}-\mathbf{q}/2,n}^0 - f_{\mathbf{k}+\mathbf{q}/2,m}^0 \simeq (f_n^0 - f_m^0) - (1/2)(v_{nl}\partial f/\partial\epsilon_n + v_{nm}\partial f/\partial\epsilon_m)q_l \quad (\text{S12})$$

$$\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + \omega \simeq (\epsilon_n - \epsilon_m + \omega) - (1/2)(v_{nl} + v_{ml})q_l \quad (\text{S13})$$

$$\text{Im} M_{\mathbf{k}nn,ij}(0) = \text{Im}(v_{ni}v_{nj}) = 0 \quad (\text{S14})$$

$$\text{Im} M_{\mathbf{k}n\neq m,ij}(0) = -(\epsilon_n - \epsilon_m)^2 \text{Im}(\langle n | \partial_i m \rangle \langle m | \partial_j n \rangle) \quad (\text{S15})$$

$$\text{Im} M_{\mathbf{k}nn,ijl}^{\text{orb}} = v_{nj} \text{Im}(\langle n | \partial_i H | \partial_l n \rangle - (i \leftrightarrow j)) \quad (\text{S16})$$

$$\text{Im} M_{\mathbf{k}n\neq m,ijl}^{\text{orb}} = (1/2)(\epsilon_n - \epsilon_m) \text{Im}[-\langle \partial_l n | \partial_i H | m \rangle \langle m | \partial_j n \rangle + \langle n | \partial_i H | \partial_l m \rangle \langle m | \partial_j n \rangle] - (i \leftrightarrow j) \quad (\text{S17})$$

$$\text{Im} M_{\mathbf{k}nn,ijl}^{\text{spin}} = -(g_s/2m_e)\epsilon_{ipl}S_{np}v_{nj} - (i \leftrightarrow j) \quad (\text{S18})$$

$$\text{Im} M_{\mathbf{k}n\neq m,ijl}^{\text{spin}} = -(g_s/2m_e)(\epsilon_n - \epsilon_m)\epsilon_{ipl} \text{Re}(\langle n | S_p | m \rangle \langle m | \partial_j n \rangle) - (i \leftrightarrow j), \quad (\text{S19})$$

where we have abbreviated $\partial f/\partial\epsilon|_{\epsilon=\epsilon_{\mathbf{k}n}}$ as $\partial f/\partial\epsilon_n$, and $H_{\mathbf{k}}$ as H .

II. SOME USEFUL IDENTITIES

We list here some identities that will be used in subsequent manipulations (Eq. (S24) has already been used in connection with Eqs. (S14)-(S19) above),

$$\langle n | \partial_i m \rangle = -\langle \partial_i n | m \rangle \quad (\text{S20})$$

$$\sum_m |\partial_i m \rangle \langle m| = -\sum_m |m \rangle \langle \partial_i m| \quad (\text{S21})$$

$$(\partial_i H)|m \rangle = (\epsilon_m - H)|\partial_i m \rangle + v_{mi}|m \rangle \quad (\text{S22})$$

$$\sum_m \langle \partial_i n | H | \partial_l m \rangle \langle m | \partial_j n \rangle = -\sum_m \epsilon_m \langle \partial_i n | m \rangle \langle \partial_l m | \partial_j n \rangle \quad (\text{S23})$$

$$\langle n | \partial_i H | m \rangle = -(\epsilon_n - \epsilon_m)\langle n | \partial_i m \rangle + v_{ni}\delta_{nm} \quad (\text{S24})$$

$$\langle n | \partial_i H | \partial_l n \rangle = \sum_m (\epsilon_n - \epsilon_m)\langle \partial_i n | m \rangle \langle m | \partial_l n \rangle + v_{ni}\langle n | \partial_l n \rangle \quad (\text{S25})$$

$$\langle n | \partial_i H | \partial_l m \rangle = \epsilon_n \langle \partial_i n | \partial_l m \rangle - \langle \partial_i n | H | \partial_l m \rangle + v_{ni}\langle n | \partial_l m \rangle. \quad (\text{S26})$$

Equations (S20)-(S22) are obtained by differentiating $\langle n | m \rangle = \delta_{nm}$, the completeness relation $\sum_m |m \rangle \langle m| = \mathbb{1}$, and $H|m \rangle = \epsilon_m|m \rangle$, respectively. Equation (S23) follows from Eq. (S21), and Eqs. (S24)-(S26) follow from Eq. (S22).

III. LOW-FREQUENCY RESPONSE WITHOUT DISSIPATION

A. Orbital part

1. Static limit

Here, Π_{ijl}^A denotes the orbital part of the purely reactive response of Eq. (S9) at $\mathcal{O}(q)$, calculated by setting $\omega = 0$ before expanding in powers of \mathbf{q} . We decompose it into interband and intraband parts,

$$\Pi_{ijl}^A = \Pi_{ijl}^{A,\text{inter}} + \Pi_{ijl}^{A,\text{intra}}. \quad (\text{S27})$$

The interband part is calculated by setting $n \neq m$ in Eq. (S9), and then using Eqs. (S12), (S13), (S15) and (S17),

$$\begin{aligned} \Pi_{ijl}^{A,\text{inter}} = & -\frac{ie^2}{2} \sum_{n,m \neq n} \int [d\mathbf{k}] \text{Im} \left\{ (v_{nl} \partial f / \partial \epsilon_n + v_{nm} \partial f / \partial \epsilon_m) \langle n | \partial_i m \rangle \langle m | \partial_j n \rangle (\epsilon_n - \epsilon_m) \right. \\ & - (f_n^0 - f_m^0) (v_{nl} + v_{ml}) \langle n | \partial_i m \rangle \langle m | \partial_j n \rangle \\ & \left. + (f_n^0 - f_m^0) [-\langle \partial_l n | \partial_i H | m \rangle \langle m | \partial_j n \rangle + \langle n | \partial_i H | \partial_l m \rangle \langle m | \partial_j n \rangle - (i \leftrightarrow j)] \right\}. \quad (\text{S28}) \end{aligned}$$

The first, second, and third lines are obtained by differentiating the occupation factors, energies, and matrix elements in Eq. (S9), respectively, and Eq. (S24) was used to cancel an energy denominator in the last term. The summation over m can be extended to include $m = n$, and exchanging dummy indices $n \leftrightarrow m$ in terms containing f_m renders them equal to the corresponding f_n terms. Using Eq. (S20) in some terms we obtain

$$\begin{aligned} \Pi_{ijl}^{A,\text{inter}} = & -ie^2 \sum_{n,m} \int [d\mathbf{k}] \left\{ -\frac{\partial f}{\partial \epsilon_n} v_{nl} \text{Im}(\langle \partial_i n | m \rangle \langle m | \partial_j n \rangle) (\epsilon_n - \epsilon_m) + f_n^0 (v_{nl} + v_{ml}) \text{Im}(\langle \partial_i n | m \rangle \langle m | \partial_j n \rangle) \right. \\ & \left. + f_n^0 \text{Im}[-\langle \partial_l n | \partial_i H | m \rangle \langle m | \partial_j n \rangle + \langle n | \partial_i H | \partial_l m \rangle \langle m | \partial_j n \rangle - (i \leftrightarrow j)] \right\}. \quad (\text{S29}) \end{aligned}$$

At $\omega = 0$ and for $n = m$, the fraction in Eq. (S9) becomes $\partial f / \partial \epsilon_n$. Using Eq. (S16) we find for the intraband part

$$\Pi_{ijl}^{A,\text{intra}} = -ie^2 \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) v_{nj} \text{Im} \langle n | \partial_i H | \partial_l n \rangle - (i \leftrightarrow j), \quad (\text{S30})$$

which can be rewritten with the help of Eq. (S25) as

$$\Pi_{ijl}^{A,\text{intra}} = -ie^2 \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) \text{Im} [-v_{nj} \langle \partial_l n | m \rangle \langle m | \partial_i n \rangle - (i \leftrightarrow j)] (\epsilon_n - \epsilon_m). \quad (\text{S31})$$

Adding Eqs. (S29) and (S31) gives Π_{ijl}^A as a sum of three types of terms, $\Pi_{ijl}^A = T_1 + T_2 + T_3$. They are

$$T_1 = -ie^2 \sum_{n,m} \int [d\mathbf{k}] \frac{\partial f_n^0}{\partial \epsilon_n} \text{Im} [-v_{nl} \langle \partial_i n | m \rangle \langle m | \partial_j n \rangle - v_{nj} \langle \partial_l n | m \rangle \langle m | \partial_i n \rangle + v_{ni} \langle \partial_l n | m \rangle \langle m | \partial_j n \rangle] (\epsilon_n - \epsilon_m) \quad (\text{S32a})$$

$$T_2 = -ie^2 \sum_{n,m} \int [d\mathbf{k}] f_n^0 \text{Im} [\langle n | \partial_i H | \partial_l m \rangle \langle m | \partial_j n \rangle - \langle \partial_l n | \partial_i H | m \rangle \langle m | \partial_j n \rangle - (i \leftrightarrow j)] \quad (\text{S32b})$$

$$T_3 = -ie^2 \sum_{n,m} \int [d\mathbf{k}] f_n^0 (v_{nl} + v_{ml}) \text{Im}(\langle \partial_i n | m \rangle \langle m | \partial_j n \rangle). \quad (\text{S32c})$$

Writing $v_{nl} \partial f / \partial \epsilon_n = \partial_l f_n^0$ in T_1 and integrating by parts yields

$$\begin{aligned} T_1 = & -ie^2 \sum_{n,m} \int [d\mathbf{k}] f_n^0 \left\{ (\epsilon_n - \epsilon_m) \text{Im} [-\langle \partial_j n | \partial_l m \rangle \langle m | \partial_i n \rangle + \langle \partial_l n | \partial_j m \rangle \langle m | \partial_i n \rangle + \langle \partial_j n | \partial_i m \rangle \langle m | \partial_l n \rangle - (i \leftrightarrow j)] \right. \\ & + (v_{nl} - v_{ml}) \text{Im}(\langle \partial_i n | m \rangle \langle m | \partial_j n \rangle) \\ & - (v_{nj} - v_{mj}) \text{Im}(\langle \partial_i n | m \rangle \langle m | \partial_l n \rangle) \\ & \left. + (v_{ni} - v_{mi}) \text{Im}(\langle \partial_j n | m \rangle \langle m | \partial_l n \rangle) \right\}. \quad (\text{S33}) \end{aligned}$$

In order to facilitate the collection of terms, we have adopted the following conventions: in $\text{Im}(\langle \partial_a n | \partial_b m \rangle \langle m | \partial_c n \rangle)$ we set $c = i, l$ but never $c = j$, and in $\text{Im}(\langle \partial_a n | m \rangle \langle m | \partial_b n \rangle)$ we choose “ $a < b$ ” where “ $i < j < l$.”

Expanding the term T_2 using Eq. (S26) followed by Eq. (S23),

$$T_2 = -ie^2 \sum_{n,m} \int [d\mathbf{k}] f_n^0 [- (v_{nj} + v_{mj}) \text{Im}(\langle \partial_i n | m \rangle \langle m | \partial_l n \rangle) - \epsilon_n \text{Im}(\langle \partial_j n | \partial_l m \rangle \langle m | \partial_i n \rangle) - \epsilon_m \text{Im}(\langle \partial_j n | \partial_l m \rangle \langle m | \partial_i n \rangle) - \epsilon_m \text{Im}(-\langle \partial_l n | \partial_j m \rangle \langle m | \partial_i n \rangle - \langle \partial_j n | \partial_i m \rangle \langle m | \partial_l n \rangle) - (i \leftrightarrow j)]. \quad (\text{S34})$$

Consider the identity obtained by multiplying Eq. (S21) on the left with $\langle \partial_l n |$ and on the right with $| \partial_j n \rangle$. It implies that the second term in Eq. (S34) [combined with its “ $-(i \leftrightarrow j)$ ” partner] vanishes upon summing over m , and so $-\epsilon_n$ therein can be changed into ϵ_n . Likewise, we can substitute $-\epsilon_m$ in the second line with $\epsilon_n - \epsilon_m$, to find

$$T_2 = -ie^2 \sum_{n,m} \int [d\mathbf{k}] f_n^0 [- (v_{nj} + v_{mj}) \text{Im}(\langle \partial_i n | m \rangle \langle m | \partial_l n \rangle) + (\epsilon_n - \epsilon_m) \text{Im}(\langle \partial_j n | \partial_l m \rangle \langle m | \partial_i n \rangle - \langle \partial_l n | \partial_j m \rangle \langle m | \partial_i n \rangle - \langle \partial_j n | \partial_i m \rangle \langle m | \partial_l n \rangle) - (i \leftrightarrow j)]. \quad (\text{S35})$$

Adding Eqs. (S32c), (S33), and (S35) we obtain, upon invoking the completeness relation,

$$\Pi_{ijl}^A = ie^2 \sum_n \int [d\mathbf{k}] f_n^0 [v_{nl} \Omega_{n,ij} - (i \leftrightarrow l) - (j \leftrightarrow l)], \quad (\text{S36})$$

where $\Omega_{n,ij} = -2\text{Im}(\langle \partial_i n | \partial_j n \rangle) = -\Omega_{n,ji}$ is the Berry curvature. Because the quantity [...] in the previous equation is totally antisymmetric, it can be written as $C_n \varepsilon_{ijl}$ with $C_n = (1/6) \varepsilon_{ijl} [\dots] = \mathbf{\Omega}_n \cdot \mathbf{v}_n$. Thus,

$$\boxed{\Pi_{ijl}^A = (ie^2/\hbar) \varepsilon_{ijl} \sum_n \int [d\mathbf{k}] f_n^0 (\mathbf{\Omega}_n \cdot \mathbf{v}_n) = 0} \quad (\text{orbital, static limit}) \quad (\text{S37})$$

where we have restored \hbar . Writing $j_i = \Pi_{ijl}^A A_j q_l$ for the current and $B_i = -i \varepsilon_{ijl} A_j q_l$ for the field, we arrive at Eqs. (1) and (2). The vanishing of Eq. (S37) was demonstrated in Ref. S10, and an alternate proof is given in the main text.

2. Uniform limit

We expand Eq. (S9) in powers of \mathbf{q} keeping ω finite, and send $\omega \rightarrow 0$ at the end. This change in the order of limits compared to the static case does not affect the calculation of the interband term $\Pi_{ijl}^{A,\text{inter}}$, but the intraband term now vanishes. To show this, define $F \equiv f_{\mathbf{k}-\mathbf{q}/2,n}^0 - f_{\mathbf{k}+\mathbf{q}/2,n}^0$ and $G \equiv \epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,n} + \omega$, so that

$$\Pi_{ijl}^{A,\text{intra}} = -ie^2 \lim_{\omega \rightarrow 0} \sum_n \int [d\mathbf{k}] \left[(F/G)|_{\mathbf{q}=0} \text{Im} M_{\mathbf{k}nn,ijl}^{\text{orb}} + \partial_l (F/G)|_{\mathbf{q}=0} \text{Im} M_{\mathbf{k}nn,ij}^{\text{orb}}(0) \right], \quad (\text{S38})$$

where

$$(F/G)|_{\mathbf{q}=0} = (f_n^0 - f_n^0)/(\epsilon_n - \epsilon_n + \omega) = 0, \quad (\text{S39a})$$

$$\partial_l (F/G)|_{\mathbf{q}=0} = -(1/\omega) v_{nl} \partial f / \partial \epsilon_n, \quad (\text{S39b})$$

and $\text{Im} M_{\mathbf{k}nn,ij}^{\text{orb}}(0) = 0$ according to Eq. (S14). Thus $\Pi_{ijl}^{A,\text{intra}} = 0$, and so the response in the uniform limit is purely interband. Since the interband term is independent of the order of limits, and the net response vanishes in the static limit, the uniform-limit response is given by minus the intraband term calculated in the static-limit. Using Eq. (S22) in Eq. (S30) we find, after some cancellations,

$$\Pi_{ijl}^A = (ie^2/\hbar) \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) v_{ni} \text{Im}(\langle \partial_j n | H - \epsilon_n | \partial_l n \rangle) - (i \leftrightarrow j). \quad (\text{S40})$$

B. Spin part

1. Static limit

We again start with the interband part, and collect $\mathcal{O}(q)$ spin contributions to Eq. (S9). Since $\text{Im}M_{\mathbf{k}n \neq m, ij}(0)$ is purely orbital [Eq. (S15)], the only contribution comes from Eq. (S19), yielding

$$\Pi_{ijl}^{A, \text{inter}} = (ig_s e^2 / 2m_e) \sum_{n,m} \int [d\mathbf{k}] (f_n^0 - f_m^0) \varepsilon_{ipl} \text{Re}(\langle n | S_p | m \rangle \langle m | \partial_j n \rangle) - (i \leftrightarrow j). \quad (\text{S41})$$

Exchanging $n \leftrightarrow m$ indices in one term and using the completeness relation gives

$$\begin{aligned} \Pi_{ijl}^{A, \text{inter}} &= (ig_s e^2 / 2m_e) \sum_n \int [d\mathbf{k}] f_n^0 \varepsilon_{ipl} \partial_j S_{np} - (i \leftrightarrow j) \\ &= -(ig_s e^2 / 2m_e) \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) \varepsilon_{ipl} v_{nj} S_{np} - (i \leftrightarrow j). \end{aligned} \quad (\text{S42})$$

For the intraband part we use Eq. (S18), which leads to the expression above, but with the opposite sign. Thus,

$$\Pi_{ijl}^A = \Pi_{ijl}^{A, \text{inter}} + \Pi_{ijl}^{A, \text{intra}} = 0. \quad (\text{S43})$$

2. Uniform limit

The steps in the derivation are similar to those carried out in Sec. III A 2 for the orbital contribution. The intraband part of the spin contribution vanishes, and the interband part is the same as in the static limit [Eq. (S42)], so that

$$\Pi_{ijl}^A = -(ig_s e^2 / 2m_e) \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) \varepsilon_{ipl} v_{nj} S_{np} - (i \leftrightarrow j). \quad (\text{S44})$$

IV. LOW-FREQUENCY RESPONSE WITH DISSIPATION

The low-frequency ($\hbar\omega \ll \epsilon_{\text{gap}}$) natural gyrotropy response was evaluated in Secs. III A 2 and III B 2 in the clean limit $\omega\tau \gg 1$. It is well known that at $\mathbf{q} = 0$ the Drude formula for the *complex* optical conductivity (i.e., including both reactive and dissipative parts) can be recovered heuristically from the intraband part of the Kubo formula (S7) for a pristine metal, by interpreting the positive infinitesimal η therein as a scattering rate $1/\tau$.^{S11} Here we apply the same procedure to extend our calculation of natural gyrotropy in metals to arbitrarily low frequencies compared to $1/\tau$. The first step is to restore η in the denominator of Eq. (S9),

$$\Pi_{ij}^A(\omega, \mathbf{q}) = -ie^2 \sum_{n,m} \int [d\mathbf{k}] \frac{f_{\mathbf{k}-\mathbf{q}/2, n}^0 - f_{\mathbf{k}+\mathbf{q}/2, m}^0}{\epsilon_{\mathbf{k}-\mathbf{q}/2, n} - \epsilon_{\mathbf{k}+\mathbf{q}/2, m} + \hbar\omega + i\hbar\eta} \text{Im} M_{\mathbf{k}nm, ij}(\mathbf{q}), \quad (\text{S45})$$

and we have also brought back \hbar for clarity. Multiplying and dividing by $\hbar\omega$ and then using the identity

$$\frac{1}{\hbar\omega(\hbar\omega + \Delta)} = \frac{1}{\Delta} \left(\frac{1}{\hbar\omega} - \frac{1}{\hbar\omega + \Delta} \right) \quad (\text{S46})$$

with $\Delta = \epsilon_{\mathbf{k}-\mathbf{q}/2, n} - \epsilon_{\mathbf{k}+\mathbf{q}/2, m} + i\hbar\eta$ we find

$$\begin{aligned} \Pi_{ij}^A(\omega, \mathbf{q}) &= -ie^2 \sum_{n,m} \int [d\mathbf{k}] \frac{f_{\mathbf{k}-\mathbf{q}/2, n}^0 - f_{\mathbf{k}+\mathbf{q}/2, m}^0}{\epsilon_{\mathbf{k}-\mathbf{q}/2, n} - \epsilon_{\mathbf{k}+\mathbf{q}/2, m} + i\hbar\eta} \text{Im} M_{\mathbf{k}nm, ij}(\mathbf{q}) \\ &\quad + ie^2 \hbar\omega \sum_{n,m} \int [d\mathbf{k}] \frac{f_{\mathbf{k}-\mathbf{q}/2, n}^0 - f_{\mathbf{k}+\mathbf{q}/2, m}^0}{\epsilon_{\mathbf{k}-\mathbf{q}/2, n} - \epsilon_{\mathbf{k}+\mathbf{q}/2, m} + i\hbar\eta} \frac{\text{Im} M_{\mathbf{k}nm, ij}(\mathbf{q})}{\hbar(\omega + i\eta) + \epsilon_{\mathbf{k}-\mathbf{q}/2, n} - \epsilon_{\mathbf{k}+\mathbf{q}/2, m}}. \end{aligned} \quad (\text{S47})$$

Consider first the dissipative response, given by the real part of the above expression. The semiclassical condition $\hbar\omega \ll \epsilon_{\text{gap}}$ excludes interband absorption, and so we can set $m = n$. The contribution from the first line is

$$-\pi e^2 \sum_n \int [d\mathbf{k}] \left(f_{\mathbf{k}-\mathbf{q}/2,n}^0 - f_{\mathbf{k}+\mathbf{q}/2,n}^0 \right) \delta(\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,n}) \text{Im} M_{\mathbf{k}nm,ij}(\mathbf{q}), \quad (\text{S48})$$

and it vanishes identically because whenever the first factor is nonzero the second factor is zero, and vice-versa. Thus, intraband absorption comes entirely from the second line in Eq. (S47),

$$\text{Re} \Pi_{ij}^A(\omega \ll \epsilon_{\text{gap}}/\hbar, \mathbf{q}) = \text{Re} \left\{ i e^2 \hbar \omega \sum_n \int [d\mathbf{k}] \frac{\partial f}{\partial \epsilon_n} \frac{\text{Im} M_{\mathbf{k}nn,ij}(\mathbf{q})}{\hbar(\omega + i\eta) + \epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,n}} \right\}. \quad (\text{S49})$$

The dissipative part of natural gyrotropy is given by the $\mathcal{O}(q)$ terms in this equation. Since $\text{Im} M_{\mathbf{k}nn,ij}(0) = 0$ according to Eq. (S14), the only contribution comes from Taylor expanding the numerator with $\mathbf{q} = 0$ in the denominator. Identifying η with $1/\tau$ and assuming it is constant across the Fermi surface (FS) we find

$$\text{Re} \Pi_{ijl}^A(\omega \ll \epsilon_{\text{gap}}/\hbar) = e^2 \text{Re} \left(\frac{\omega\tau}{1 - i\omega\tau} \right) G_{ijl}, \quad (\text{S50})$$

where

$$G_{ijl} = \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) \text{Im} M_{\mathbf{k}nn,ijl} = G_{ijl}^{\text{orb}} + G_{ijl}^{\text{spin}} \quad (\text{S51a})$$

$$G_{ijl}^{\text{orb}} = (1/\hbar) \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) v_{ni} \text{Im} \langle \partial_j n | H - \epsilon_n | \partial_l n \rangle - (i \leftrightarrow j) \quad (\text{S51b})$$

$$G_{ijl}^{\text{spin}} = -(g_s/2m_e) \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_n) \varepsilon_{ipl} v_{nj} S_{np} - (i \leftrightarrow j). \quad (\text{S51c})$$

To obtain the orbital term we used Eqs. (S16) and (S22), and for the spin term we used Eq. (S18).

The reactive part can be recovered from the Kramers-Krönig relation^{S12} for $\sigma_{ijl} = (1/i\omega)\Pi_{ijl}$, the $\mathcal{O}(q)$ part of the effective conductivity defined in Eq. (S59) below. The result for the full (complex) response at low frequencies is

$$\boxed{\Pi_{ijl}^A(\omega \ll \epsilon_{\text{gap}}/\hbar) = \frac{e^2 \omega \tau}{1 - i\omega\tau} G_{ijl}} \quad (\text{orbital} + \text{spin, uniform limit}) \quad (\text{S52})$$

which can be viewed as a Drude-like formula for natural gyrotropy in metals. At $\omega\tau \ll 1$ the response becomes purely dissipative and at $\omega\tau \gg 1$ it becomes purely reactive, reducing to the result from the rigorous Kubo-formula calculation in the clean limit, Eqs. (S40) and (S44). Equation (8) is the combination of Eqs. (S51) and (S52).

V. ORBITAL GME IN TWO-BAND MODELS: COMPARISON WITH THE PREVIOUS LITERATURE

In the main text we obtained an expression [Eq. (17)] for the orbital GME current $\mathbf{j}^{\mathbf{B}} = \bar{\alpha}^{\text{GME}} \mathbf{B}$ in a clean Weyl semimetal with two isotropic Weyl points (WPs). Here we consider a generic two-band model, and show how to recover, starting from the FS formula for α^{GME} , the expression given in Refs. S13 and S14 for $\bar{\alpha}^{\text{GME}} \equiv \text{tr}(\alpha^{\text{GME}})/3$ (but keeping in mind that in anisotropic models the traceless part is generally nonzero; specific traceless pieces will be considered in Secs. VIA 3 and VID).

In two-band models the orbital moment is given by Eq. (15), and it is simply related to the Berry curvature according to $\mathbf{m}_{\mathbf{k}t}^{\text{orb}} = (e/\hbar) t d_{\mathbf{k}} \mathbf{\Omega}_{\mathbf{k}t}$.^{S15} Inserting this expression into Eq. (9) and taking the trace gives, at $\omega\tau \gg 1$,

$$\bar{\alpha}^{\text{GME}} = -\frac{e^2}{3\hbar} \sum_{t=\pm} \int [d\mathbf{k}] (\partial f / \partial \epsilon_{\mathbf{k}t}) t d_{\mathbf{k}} \mathbf{v}_{\mathbf{k}t} \cdot \mathbf{\Omega}_{\mathbf{k}t}. \quad (\text{S53})$$

Replacing $\mathbf{v}_{\mathbf{k}t} \partial f / \partial \epsilon_{\mathbf{k}t}$ with $\partial_{\hbar\mathbf{k}} f_{\mathbf{k}t}^0$ and integrating by parts yields two terms; the one containing $\partial_{\mathbf{k}} \cdot \mathbf{\Omega}_{\mathbf{k}t}$, Eq. (S56) below, vanishes identically, leaving

$$\bar{\alpha}^{\text{GME}} = \frac{e^2}{3\hbar} \sum_{t=\pm} \int [d\mathbf{k}] f_{\mathbf{k}t}^0 \mathbf{\Omega}_{\mathbf{k}t} \cdot \partial_{\hbar\mathbf{k}} (t d_{\mathbf{k}}). \quad (\text{S54})$$

Writing $td_{\mathbf{k}}$ as $\epsilon_{\mathbf{k}t} - (\epsilon_{\mathbf{k}+} + \epsilon_{\mathbf{k}-})/2$ and using Eq. (2) to eliminate a term we find

$$\bar{\alpha}^{\text{GME}} = -\frac{e^2}{3\hbar} \sum_{t=\pm} \int [d\mathbf{k}] f_{\mathbf{k}t}^0 \frac{\mathbf{v}_{\mathbf{k}+} + \mathbf{v}_{\mathbf{k}-}}{2} \cdot \boldsymbol{\Omega}_{\mathbf{k}t}. \quad (\text{S55})$$

The expression in Refs. S13 and S14 amounts to 3/2 times Eq. (S55) minus 1/2 times Eq. (S53), divided by two. To understand the division by two, recall from Eq. (6a) that α^{GME} describes the current response to the optical \mathbf{B} field. The optical \mathbf{E} field contributes an equal amount to the component of the natural gyrotropy current along \mathbf{B} (see Sec. VIA below), bringing our result in accord with Refs. S13 and S14. While strictly correct, the above expression for $\bar{\alpha}^{\text{GME}}$ in two-band models is not physically transparent. It fails to recognize the FS nature of the effect, and has led to the erroneous identification of the Berry curvature as the key quantity governing it.

Between Eqs. (S53) and (S54) we dropped a term containing the divergence of the Berry curvature,

$$\frac{e^2}{3\hbar^2} \sum_{t=\pm} \int [d\mathbf{k}] f_{\mathbf{k}t} (\epsilon_{\mathbf{k}t} - \bar{\epsilon}_{\mathbf{k}}) (\boldsymbol{\partial}_{\mathbf{k}} \cdot \boldsymbol{\Omega}_{\mathbf{k}t}). \quad (\text{S56})$$

The Berry curvature $\boldsymbol{\Omega}_{\mathbf{k}t} = \boldsymbol{\partial} \times \mathbf{A}_{\mathbf{k}t}$ is divergence-free except at the WPs $\epsilon_{\mathbf{k}t} = \bar{\epsilon}_{\mathbf{k}}$, which act as monopole sources and sinks. The above expression is proportional to the sum over all WPs, and over the two bands, of $f_{\mathbf{k}t}$ times $\epsilon_{\mathbf{k}t} - \bar{\epsilon}_{\mathbf{k}}$ times δ -function singularities. Since $f_{\mathbf{k}t} = 0, 1$ and $\epsilon_{\mathbf{k}t} - \bar{\epsilon}_{\mathbf{k}} = 0$ are the same for the two bands at a WP, the summation only acts on the last factor. But $\sum_{t=\pm} \boldsymbol{\partial}_{\mathbf{k}} \cdot \boldsymbol{\Omega}_{\mathbf{k}t} = 0$, because each WP contributes twice with opposite signs: once as a source term in one band, and another time as sink in the other band: see, e.g., Eq. (10) in Ref. S16. Thus, each WP gives a vanishing net contribution to the expression in Eq. (S56).

VI. NATURAL OPTICAL ACTIVITY OF METALS IN THE SEMICLASSICAL LIMIT

A. Gyrotropic response in equilibrium

1. Phenomenological relations

The GME in metals is conveniently formulated in terms of the dual tensors $\Pi_{ijl}^A = -\Pi_{jil}^A$ and α_{ij}^{GME} , related to one another by Eq. (5). Instead, the gyrotropic response of a bulk medium is usually discussed in terms of the antisymmetric part of the relative permittivity tensor, $\epsilon_{ij}^A(\omega, \mathbf{q}) = [\epsilon_{ij}(\omega, \mathbf{q}) - \epsilon_{ji}(\omega, \mathbf{q})]/2$, expanded to $\mathcal{O}(q)$:^{S8}

$$\epsilon_{ij}^A(\omega, \mathbf{q}) = \epsilon_{ij}^A(\omega, 0) + i\gamma_{ijl}^A(\omega)q_l + \dots \quad (\text{S57})$$

The T -odd tensor $\epsilon_{ij}^A(\omega, 0)$ describes magneto-optical gyrotropic effects such as Faraday rotation and magnetic circular dichroism, and the T -even tensor $\gamma_{ijl}^A(\omega) = -\gamma_{jil}^A(\omega)$ describes natural gyrotropy effects including natural optical rotation and natural circular dichroism. It is also useful to introduce a dimensionless rank-2 tensor $g_{ij}(\omega)$ dual to $\gamma_{ijl}^A(\omega)$ according to^{S8}

$$\gamma_{ijl}^A = (c/\omega)\epsilon_{ijm}g_{ml} \quad (\text{S58a})$$

$$g_{ij} = (\omega/2c)\epsilon_{ilm}\gamma_{lmj}^A, \quad (\text{S58b})$$

where c is the speed of light. In order to convert between Π_{ijl}^A and α_{ij}^{GME} on one hand and γ_{ijl}^A and g_{ij} on the other, consider the effective conductivity tensor $\sigma_{ij}(\omega, \mathbf{q})$ satisfying^{S6,S7}

$$j_i(\omega, \mathbf{q}) = \sigma_{ij}(\omega, \mathbf{q})E_j(\omega, \mathbf{q}). \quad (\text{S59})$$

Using $\mathbf{E} = i\omega\mathbf{A}$ and comparing with Eq. (S6) gives $\Pi_{ij} = i\omega\sigma_{ij}$. Hence the tensors ϵ_{ij} , σ_{ij} , and Π_{ij} are related by^{S7}

$$\epsilon_{ij}(\omega, \mathbf{q}) = \delta_{ij} + \frac{i}{\omega\epsilon_0}\sigma_{ij}(\omega, \mathbf{q}) = \delta_{ij} + \frac{1}{\omega^2\epsilon_0}\Pi_{ij}(\omega, \mathbf{q}). \quad (\text{S60})$$

Expanding the antisymmetric part to $\mathcal{O}(q)$ and comparing with Eq. (S57) yields the relation

$$\gamma_{ijl}^A = -(i/\omega^2\epsilon_0)\Pi_{ijl}^A \quad (\text{S61})$$

between the two rank-3 natural gyrotropy tensors. The relation between their duals g_{ij} and α_{ij}^{GME} follows from combining Eqs. (S58b), (S61), and (5a). The result is (note the transposed indices)

$$g_{ij} = \frac{1}{\omega c \epsilon_0} (\tilde{\alpha}_{ji}^{\text{GME}} - 2\bar{\alpha}^{\text{GME}} \delta_{ij}), \quad (\text{S62})$$

where $\bar{\alpha}^{\text{GME}} \delta_{ij}$ and $\tilde{\alpha}_{ij}^{\text{GME}}$ are the trace piece and the traceless part of α_{ij}^{GME} , respectively.

2. Optical rotation in high-symmetry metals

Let us specialize to a metal with cubic symmetry or higher, and express the rotatory power ρ in terms of α^{GME} . In a medium with such high symmetry the tensors Π_{ijl}^A and γ_{ijl}^A are totally antisymmetric.^{S8,S17} From Eq. (5) we get $\Pi_{ijl}^A = -2i\bar{\alpha}^{\text{GME}} \epsilon_{ijl}$ and $\alpha_{ij}^{\text{GME}} = \bar{\alpha}^{\text{GME}} \delta_{ij}$, Eq. (6) gives $\mathbf{j}^{\text{B}} = i\mathbf{q} \times \mathbf{M}^{\text{E}} = \bar{\alpha}^{\text{GME}} \mathbf{B}$, and finally $\gamma_{ijl}^A = \gamma \epsilon_{ijl}$ with

$$\gamma = -(2/\epsilon_0 \omega^2) \bar{\alpha}^{\text{GME}} \quad (\text{S63})$$

according to Eq. (S61). The standard formula^{S18,S19}

$$\rho = (\omega^2/2c^2) \text{Re } \gamma \quad (\text{S64})$$

for the rotatory power of a high-symmetry medium becomes

$$\rho = -(1/\epsilon_0 c^2) \text{Re } \bar{\alpha}^{\text{GME}}. \quad (\text{S65})$$

This expression, together with Eq. (11), implies that the rotatory power of a clean metal goes to a constant at low frequencies. Instead, the rotatory power of an insulator decreases as ω^2 , because γ goes to a constant, as follows from Eq. (S61) and the ω^2 scaling at low frequencies of $\text{Im } \Pi_{ijl}^A = \omega \text{Re } \sigma_{ijl}^A$ (see, e.g., Ref. S6).

As an application of Eq. (S65), consider a Weyl semimetal with isotropic Weyl nodes described by the effective 2-band Hamiltonian (see main text)

$$H_{\mathbf{k}\nu} = \epsilon_\nu \mathbb{1} + \chi_\nu \hbar v_F \mathbf{k} \cdot \boldsymbol{\sigma}. \quad (\text{S66})$$

The orbital contribution to $\bar{\alpha}^{\text{GME}}$ from a pair of nodes of opposite chirality is given in the clean limit by the prefactor in Eq. (17), yielding

$$\boxed{\rho = (2\alpha/3hc)(\epsilon_L - \epsilon_R)} \quad (\text{S67})$$

per node pair in the frequency range $1/\tau \ll \omega \ll \epsilon_{\text{gap}}/\hbar$, where $\alpha = e^2/4\pi\epsilon_0\hbar c \approx 1/137$ is the fine-structure constant. This result, given as Eq. (18) in the main text, can be extended to lower frequencies in the constant relaxation-time approximation by inserting a factor of $\text{Re}[i\omega\tau/(i\omega\tau - 1)] = \omega^2\tau^2/(1 + \omega^2\tau^2)$ on the right-hand-side. Therefore $\rho \propto \omega^2$ at the lowest frequencies in dirty metals, the same low-frequency behavior as in molecules^{S20} and insulators.^{S6,S18}

3. Transverse GME in polar metals

In a ‘‘pyroelectric’’ metal with polar axis $\hat{\mathbf{c}}$, the optical activity tensors g_{ij} and α_{ij}^{GME} acquire an antisymmetric part that does not contribute to optical rotation,^{S17} but which gives rise to an inverse GME [Eq. (6b)] of the form^{S21}

$$\mathbf{M}^{\text{E}} = \frac{1}{1 - i\omega\tau} \frac{\tau e}{8\pi^2 \hbar} \mathbf{E} \times \boldsymbol{\delta}, \quad (\text{S68})$$

with $\boldsymbol{\delta} \parallel \hat{\mathbf{c}}$. This characteristic magnetoelectric response of polar metals has been noted.^{S22} It should occur in the Weyl semimetal TaAs, and in the low-temperature phase of the ‘‘ferroelectric’’ metal LiOsO₃. Using Eq. (11) we find

$$\boxed{\boldsymbol{\delta} = \sum_{n,a} \int_{S_{na}} dS (\hat{\mathbf{v}}_F \times \mathbf{m}_{\mathbf{k}n})} \quad (\text{S69})$$

The (polar) vector $\boldsymbol{\delta} = \boldsymbol{\delta}^{\text{orb}} + \boldsymbol{\delta}^{\text{spin}}$ vanishes in metals without a polar axis, and its magnitude in a given polar metal reflects the extent to which the polar character of the structure is transmitted to the conduction electrons.

$\boldsymbol{\delta}^{\text{orb}}$ vanishes for an asymmetrically-confined 2D metal with the polar axis normal to the plane, because in 2D systems $\mathbf{m}_{\mathbf{k}n}^{\text{orb}}$ always points out of plane; the Rashba spin-orbit interaction generates an in-plane tangential spin texture that leads to a $\boldsymbol{\delta}^{\text{spin}} \parallel \hat{\mathbf{c}}$, and an in-plane spin polarization normal to \mathbf{E} appears: this is the inverse spin-galvanic effect in 2D Rashba systems.^{S23} In 3D polar metals (including strained ‘‘piezoelectric’’ metals^{S24}), the dissipative magnetoelectric effect of Eq. (S68) has an orbital-magnetization contribution that does not require spin-orbit coupling.

B. Nonequilibrium optical gyrotropy in Weyl semimetals driven by the chiral anomaly

It was pointed out in Ref. S25 that the chiral-anomaly mechanism leads to circular dichroism for light propagating inside a Weyl semimetal in the presence of external electric and magnetic fields with $\mathbf{E}_{\text{ext}} \cdot \mathbf{B}_{\text{ext}} \neq 0$. In the following we use our microscopic formulation of natural gyrotropy in metals to calculate the induced response at low frequencies.

For Weyl nodes described by Eq. (S66), we obtain from Eqs. (11) and (15) the following expression for the orbital contribution to $\alpha_{ij}^{\text{GME}}(\omega) = \bar{\alpha}^{\text{GME}}(\omega)\delta_{ij}$ from the spherical pocket enclosing the ν -th node, with chemical potential μ_ν ,

$$\bar{\alpha}_\nu^{\text{GME}}(\omega) = \frac{1}{3} \frac{e}{(2\pi)^2 \hbar} \frac{i\omega\tau}{i\omega\tau - 1} (4\pi k_F^2) \left(\mp \chi_\nu \frac{ev_F}{2k_F} \right) = \mp \frac{1}{3} \frac{e^2}{h^2} \frac{i\omega\tau}{i\omega\tau - 1} \chi_\nu \hbar v_F k_F = \frac{1}{3} \frac{e^2}{h^2} \frac{i\omega\tau}{i\omega\tau - 1} \chi_\nu (\epsilon_\nu - \mu_\nu). \quad (\text{S70})$$

The minus (plus) sign in the intermediate expressions corresponds to $\epsilon_\nu < \mu_\nu$ ($\epsilon_\nu > \mu_\nu$).

Consider a minimal model where $\nu = L, R$. In the equilibrium situation ($\mu_L = \mu_R = \epsilon_F$) depicted in Fig. 1(b) in the main text, Eq. (S70) reduces to Eq. (16) for $\omega\tau \gg 1$. Here we are interested in the scenario where the background $\mathbf{E}_{\text{ext}} \cdot \mathbf{B}_{\text{ext}}$ field pumps charge across the nodes, leading to $\mu_L \neq \mu_R$. Following Ref.S25 we assume $\omega\tau \ll 1$, and for simplicity we set $\epsilon_L = \epsilon_R \equiv \epsilon_W$ as in Fig. 1(a), so that the equilibrium gyrotropic response of Eq. (S67) vanishes. Combining Eqs. (S63) and (S70) and taking the imaginary part yields

$$\text{Im } \gamma_\nu(\omega \ll 1/\tau) = -\frac{\chi_\nu(\mu_\nu - \epsilon_W)e^2\tau}{6\pi^2\epsilon_0\hbar^2\omega}. \quad (\text{S71})$$

Summing over WPs and using $\sum_\nu \chi_\nu = 0^{\text{S26}}$ gives for the orbital contribution to the nonequilibrium circular dichroism

$$\text{Im } \gamma(\omega \ll 1/\tau) = \frac{(\mu_L - \mu_R)e^2\tau}{6\pi^2\epsilon_0\hbar^2\omega}. \quad (\text{S72})$$

Equation (S71) agrees with Eq. (11) of Ref. S25, except possibly for the numerical prefactor. Exact agreement was not expected, since the calculation in Ref. S25 was based on an incomplete formulation of natural gyrotropy in metals in terms of the Berry curvature. On the other hand, semiquantitative agreement for two-band models seems plausible, in view of the simple relation between the Berry curvature and the orbital moment in such models.^{S15}

Consider now the optical rotation under nonequilibrium conditions. According to Eq. (S70) $\text{Re } \gamma(\omega \ll 1/\tau) = 0$, suggesting an absence of optical rotation at the lowest frequencies.^{S25} Note, however, that Eq. (S70) was obtained starting from Eq. (S40), which assumes an equilibrium situation where the nondissipative gyrotropic current vanishes in the static limit [Eq. (S37)]. In the presence of a chemical-potential imbalance, a new reactive term appears: the optical \mathbf{B} field induces a dissipationless current at $\mathcal{O}(q)$ via Eq. (3), leading to optical rotation even for $\omega \ll 1/\tau$. Interestingly, this is a genuine Berry-curvature – as opposed to orbital-moment – contribution to the (nonequilibrium) optical gyrotropy in Weyl semimetals.

C. Reciprocity relation for a gyrotropic metal with a smooth interface

There is a general reciprocity principle for electromagnetic fields interacting with time-reversal (T) invariant media in equilibrium that requires the optical rotation to vanish in the reflection geometry.^{S27,S28} Thus, while for a T -breaking material there are in general optical rotation effects both in transmission (Faraday) and in reflection (Kerr), optical rotation in T -invariant materials occurs in transmission only. At first glance this is surprising, because it implies a constraint on the nonlocal response functions imposed by T symmetry when the gyrotropic coefficient varies in space, for example at an interface.^{S29–S31} This has been actively discussed recently in the context of optical rotation measured in reflection on cuprate superconductors.^{S19,S28,S32}

We show in the following how this constraint appears in our treatment of natural optical activity in metals. While the validity of the constraint is not in question, i.e., it had to be satisfied, it seems worthwhile to explain how it arises, given the recent interest in the nonlocal constitutive relation in spatially varying media with natural optical activity.

A T -invariant spatially-dispersive medium with a boundary or an interface is described, taking into consideration only the first derivatives with respect to coordinates, by the following constitutive relation,

$$D_i(\omega, \mathbf{r}) = \epsilon_0 \left[\epsilon_{ij}^S(\omega, \mathbf{r}) E_j(\omega, \mathbf{r}) + \gamma_{ijl}^A(\omega, \mathbf{r}) \partial_{r_l} E_j(\omega, \mathbf{r}) + E_j(\omega, \mathbf{r}) \partial_{r_l} \lambda_{ijl}^A(\omega, \mathbf{r}) \right]. \quad (\text{S73})$$

This is Eq. (8) in Ref. S29, with the T -breaking parameter \mathbf{B}_{ext} set to zero. Under those conditions, the response tensors satisfy $\epsilon_{ij}^S(\omega, \mathbf{r}) = \epsilon_{ji}^S(\omega, \mathbf{r})$, $\gamma_{ijl}^A(\omega, \mathbf{r}) = -\gamma_{jil}^A(\omega, \mathbf{r})$, and $\lambda_{ijl}^A(\omega, \mathbf{r}) = -\lambda_{jil}^A(\omega, \mathbf{r})$. (More generally, in a T -breaking material the constitutive relation can be split into even and odd parts with respect to \mathbf{B}_{ext} , and Eq. (S73)

then corresponds to the T -even part.) For an infinite macroscopically homogeneous medium the response tensors are independent of \mathbf{r} , and only the first two terms survive in Eq. (S73). The last term is an additional contribution coming from the spatial inhomogeneity near the interface. The reciprocity constraint takes the form^{S29}

$$\lambda_{ijl}^A(\omega, \mathbf{r}) = \frac{1}{2}\gamma_{ijl}^A(\omega, \mathbf{r}), \quad (\text{S74})$$

which in turn implies vanishing optical rotation in reflection: see Ref. S28 and references cited therein.

To obtain Eq. (S74) for a metal subject to a low-frequency optical field, recall from the discussion around Eq. (7) that the natural gyrotropy of metals at low frequencies is fully determined by the magnetoelectric response of the medium, i.e., by the dynamic polarization $\mathbf{P}^{\mathbf{B}}$ and magnetization $\mathbf{M}^{\mathbf{E}}$ induced by the optical fields \mathbf{B} and \mathbf{E} respectively. In the main text, the T -even magnetoelectric response of a metal was calculated at $\hbar\omega \ll \epsilon_{\text{gap}}$ using a semiclassical Boltzmann formalism, and the result was of the form

$$P_i^{\mathbf{B}}(\omega, \mathbf{r}) = (i/\omega)\alpha_{ij}^{\text{GME}}(\omega, \mathbf{r})B_j(\omega, \mathbf{r}) \quad (\text{S75a})$$

$$M_i^{\mathbf{E}}(\omega, \mathbf{r}) = -(i/\omega)\alpha_{ji}^{\text{GME}}(\omega, \mathbf{r})E_j(\omega, \mathbf{r}), \quad (\text{S75b})$$

with α^{GME} given in the long-wavelength limit by Eq. (11). (The calculation was done for a bulk metal where α^{GME} is independent of \mathbf{r} , but the semiclassical approach remains valid if we assume a smooth interface with a spatial variation that is slow on the scale of the mean free path.) Writing the constitutive relation in terms of the auxiliary fields,

$$\mathbf{D}(\omega, \mathbf{r}) = \epsilon_0\mathbf{E}(\omega, \mathbf{r}) + \mathbf{P}(\omega, \mathbf{r}) + (i/\omega)\partial_{\mathbf{r}} \times \mathbf{M}(\omega, \mathbf{r}), \quad (\text{S76})$$

and using Eq. (S75) together with $B_j = -(i/\omega)\epsilon_{jlp}\partial_{r_l}E_p$, we find for the spatially-dispersive part of $D_i(\omega, \mathbf{r})$

$$\underbrace{(1/\omega^2) [\epsilon_{ilp}\alpha_{jp}^{\text{GME}}(\omega, \mathbf{r}) - \epsilon_{jlp}\alpha_{ip}^{\text{GME}}(\omega, \mathbf{r})]}_{\epsilon_0\gamma_{ijl}^A(\omega, \mathbf{r})} \partial_{r_l}E_j(\omega, \mathbf{r}) + E_j(\omega, \mathbf{r})\partial_{r_l} \underbrace{[(1/\omega^2)\epsilon_{ilp}\alpha_{jp}^{\text{GME}}(\omega, \mathbf{r})]}_{\epsilon_0\lambda_{ijl}(\omega, \mathbf{r})}. \quad (\text{S77})$$

We have identified the tensors γ_{ijl}^A and $\lambda_{ijl} = \lambda_{ijl}^S + \lambda_{ijl}^A$ by comparing with Eq. (S73), noting that the term containing $\lambda_{ijl}^S = \lambda_{jil}^S$ was included in the first term of Eq. (S73).^{S29} The expression for γ_{ijl}^A is consistent with Eqs. (S61) and (5a) for a bulk medium, and by inspection we obtain Eq. (S74), with $2\lambda_{ijl}^A(\omega, \mathbf{r}) \equiv \lambda_{ijl}(\omega, \mathbf{r}) - \lambda_{jil}(\omega, \mathbf{r})$. In conclusion, our microscopic formulation of natural gyrotropy in metals is consistent with the general reciprocity principle.

D. Berry-curvature contributions

In Ref. S33, the natural optical activity of clean metals in equilibrium was studied using a semiclassical ‘‘Berry-Boltzmann’’ approach, and a combination of the band velocity and the Berry curvature was shown to give a traceless contribution to the tensor g_{ij} defined in Eq. (S58). Here we show how that contribution appears as part of the full microscopic expression in terms of the intrinsic magnetic moment. We start by setting $\omega\tau \gg 1$ in Eq. (9) to get

$$\alpha_{ij}^{\text{GME}} = -e \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_{\mathbf{k}n}) v_{\mathbf{k}n,i} \left(m_{\mathbf{k}n,j}^{\text{spin}} + m_{\mathbf{k}n,j}^{\text{orb}} \right) \equiv \alpha_{ij}^{\text{spin}} + \alpha_{ij}^{\text{orb}}, \quad (\text{S78})$$

and note that Eq. (10) for $\mathbf{m}_{\mathbf{k}n}^{\text{orb}}$ contains two terms, one of which involves the Berry curvature,

$$\mathbf{m}_{\mathbf{k}n}^{\text{orb}} = (e/2\hbar)\text{Im}\langle \partial_{\mathbf{k}} u_{\mathbf{k}n} | \times H_{\mathbf{k}} | \partial_{\mathbf{k}} u_{\mathbf{k}n} \rangle + (e/2\hbar)\epsilon_{\mathbf{k}n}\boldsymbol{\Omega}_{\mathbf{k}n} \equiv \mathbf{m}_{\mathbf{k}n}^{\text{H}} + \mathbf{m}_{\mathbf{k}n}^{\Omega}. \quad (\text{S79})$$

Accordingly we write $\alpha^{\text{orb}} = \alpha^{\text{H}} + \alpha^{\Omega}/2$, with

$$\alpha_{ij}^{\text{H}} = -\frac{e^2}{2\hbar}\epsilon_{jlp} \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_{\mathbf{k}n}) v_{\mathbf{k}n,i} \text{Im}\langle \partial_l u_{\mathbf{k}n} | H_{\mathbf{k}} | \partial_p u_{\mathbf{k}n} \rangle \quad (\text{S80a})$$

$$\alpha_{ij}^{\Omega} = -\frac{e^2}{\hbar} \sum_n \int [d\mathbf{k}] (\partial f / \partial \epsilon_{\mathbf{k}n}) v_{\mathbf{k}n,i} \epsilon_{\mathbf{k}n} \Omega_{\mathbf{k}n,j}. \quad (\text{S80b})$$

The reason for writing $\alpha^{\Omega}/2$ is that α^{H} can be further decomposed as (see derivation at the end)

$$\alpha_{ij}^{\text{H}} = \alpha_{ij}^{\Omega}/2 + \Delta\alpha_{ij}^{\text{orb}} \quad (\text{S81a})$$

$$\Delta\alpha_{ij}^{\text{orb}} = \frac{e^2}{2\hbar^2}\epsilon_{jlp} \sum_{n,m} \int [d\mathbf{k}] f_{\mathbf{k}n}^0 (1 - f_{\mathbf{k}m}^0) \partial_i [(\epsilon_{\mathbf{k}n} + \epsilon_{\mathbf{k}m}) \text{Im}\langle \partial_l u_{\mathbf{k}n} | u_{\mathbf{k}m} \rangle \langle u_{\mathbf{k}m} | \partial_p u_{\mathbf{k}n} \rangle], \quad (\text{S81b})$$

so that finally

$$\boldsymbol{\alpha}^{\text{orb}} = \boldsymbol{\alpha}^{\Omega} + \Delta\boldsymbol{\alpha}^{\text{orb}}. \quad (\text{S82})$$

The decomposition (S82) has two noteworthy features. Firstly, the Berry-curvature contribution is traceless,

$$\text{tr}(\boldsymbol{\alpha}^{\Omega}) = \frac{e^2\epsilon_F}{2\hbar^2} \sum_{n,a} \int_{S_{na}} dS (\hat{\mathbf{v}}_F \cdot \boldsymbol{\Omega}_{\mathbf{k}n}) = \frac{\pi e^2\epsilon_F}{\hbar^2} \sum_{n,a} C_{na} = 0, \quad (\text{S83})$$

for the same reason that the chiral magnetic effect vanishes when all Fermi sheets are in chemical equilibrium (see main text). Secondly, $\boldsymbol{\alpha}^{\Omega}$ and $\Delta\boldsymbol{\alpha}^{\text{orb}}$ are separately gauge-invariant, but they are *not* separately invariant under a shift of the zero of energy. Nevertheless, $\boldsymbol{\alpha}^{\Omega}$ can be further decomposed into two traceless parts, only one of which depends on the zero of energy, by replacing $v_{\mathbf{k}n,i}\partial f/\partial\epsilon_{\mathbf{k}n}$ in Eq. (S80b) with $(1/\hbar)\partial_i f_{\mathbf{k}n}^0$ and integrating by parts,

$$\alpha_{ij}^{\Omega} = \alpha_{ij}^{\Omega,1} + \alpha_{ij}^{\Omega,2} \quad (\text{S84a})$$

$$\alpha_{ij}^{\Omega,1} = \frac{e^2}{\hbar^2} \sum_n \int [d\mathbf{k}] f_{\mathbf{k}n}^0 \epsilon_{\mathbf{k}n} \partial_i \Omega_{\mathbf{k}n,j} \quad (\text{S84b})$$

$$\alpha_{ij}^{\Omega,2} = \frac{e^2}{\hbar} \sum_n \int [d\mathbf{k}] f_{\mathbf{k}n}^0 v_{\mathbf{k}n,i} \Omega_{\mathbf{k}n,j}. \quad (\text{S84c})$$

The trace of $\boldsymbol{\alpha}^{\Omega,1}$ vanishes for the same reason as Eq. (S56), and the trace of $\boldsymbol{\alpha}^{\Omega,2}$ vanishes according to Eq. (2). $\boldsymbol{\alpha}^{\Omega,1}$ clearly depends on the zero of energy, but $\boldsymbol{\alpha}^{\Omega,2}$ does not.

The Berry-curvature contribution identified in Ref. S33 amounts to $\boldsymbol{\alpha}^{\Omega,2}$. To see this, convert Eq. (S84c) into a (traceless) contribution to g_{ij} using Eq. (S62),

$$g_{ij}^{\Omega,2} = \frac{e^2}{\hbar\omega c\epsilon_0} \sum_n \int [d\mathbf{k}] f_{\mathbf{k}n}^0 v_{\mathbf{k}n,j} \Omega_{\mathbf{k}n,i}. \quad (\text{S85})$$

To compare with Ref. S33 the expression above should be multiplied by $ic\epsilon_0$,^{S34} leading to Eq. (10) therein.^{S35}

In closing, we reiterate that even if Berry-curvature contributions can be identified (see also Sec. V), the magnetic moment on the FS provides a more basic and compact description of the low-frequency natural gyrotropy of metals.

1. Derivation of Eq. (S81)

Integrating Eq. (S80a) by parts and inserting a complete set of states gives, in the condensed notation used earlier,

$$\begin{aligned} \alpha_{ij}^{\text{H}} &= \frac{e^2}{2\hbar^2} \epsilon_{jlp} \sum_{n,m} \int [d\mathbf{k}] f_n^0 \partial_i [\epsilon_m \text{Im}(\langle \partial_l n | m \rangle \langle m | \partial_p n \rangle)] \\ &= \frac{e^2}{2\hbar^2} \epsilon_{jlp} \sum_{n,m} \int [d\mathbf{k}] [f_m^0 - f_m^0(1 - f_n^0) + f_n^0(1 - f_m^0)] \partial_i [\epsilon_m \text{Im}(\langle \partial_l n | m \rangle \langle m | \partial_p n \rangle)]. \end{aligned}$$

Exchanging n and m in the first and second terms,

$$\begin{aligned} \alpha_{ij}^{\text{H}} &= \frac{e^2}{2\hbar^2} \epsilon_{jlp} \sum_{n,m} \int [d\mathbf{k}] \left\{ f_n^0 \partial_i [\epsilon_n \text{Im}(\langle \partial_l m | n \rangle \langle n | \partial_p m \rangle)] \right. \\ &\quad \left. - f_n^0(1 - f_m^0) \partial_i [\epsilon_n \text{Im}(\langle \partial_l m | n \rangle \langle n | \partial_p m \rangle)] + f_n^0(1 - f_m^0) \partial_i [\epsilon_m \text{Im}(\langle \partial_l n | m \rangle \langle m | \partial_p n \rangle)] \right\}. \quad (\text{S86}) \end{aligned}$$

Using Eq. (S20) to combine the second and the third terms, and to recast the first in terms of the Berry curvature, we arrive at Eq. (S81) after an integration by parts:

$$\begin{aligned} \alpha_{ij}^{\text{H}} &= -\frac{e^2}{2\hbar} \sum_n \int [d\mathbf{k}] (\partial f/\partial\epsilon_n) v_{n,i} (\epsilon_n \Omega_{n,j}) \\ &\quad + \frac{e^2}{2\hbar^2} \epsilon_{jlp} \sum_{n,m} \int [d\mathbf{k}] f_n^0 (1 - f_m^0) \partial_i [(\epsilon_n + \epsilon_m) \text{Im}(\langle \partial_l n | m \rangle \langle m | \partial_p n \rangle)]. \quad (\text{S87}) \end{aligned}$$

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