

Nonlinear anomalous Edelstein response at altermagnetic interfaces

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In altermagnets, time-reversal symmetry breaking spin polarizes electronic states, while total magnetization remains zero. In addition, at altermagnetic surfaces Rashba–spin orbit coupling is activated due to broken inversion symmetry, introducing a competing spin-momentum locking interaction. Here we show that their interplay leads to the formation of complex, chiral spin textures that offer nonlinear spin-to-charge conversion properties. Whereas altermagnetic order suppresses the canonical linear in-plane Rashba-Edelstein response, we establish the presence of an *anomalous* transversal Edelstein effect for planar applied electric and magnetic field or, alternatively, an in-plane magnetization. Additionally, we predict a purely electric-field-driven nonlinear out-of-plane magnetization. We compute the anomalous response within a general altermagnet *d*-wave model, with parameters extracted from the *ab initio* electronic structure of an altermagnetic bilayer. Our results suggest altermagnetic surfaces as a promising platform for unconventional spintronic functionalities.

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I. INTRODUCTION

Altermagnets (AMs) are a class of magnetic materials with a total magnetization that vanishes by symmetry while at the same time Kramers’ degeneracy is lifted [1–11]. Unlike antiferromagnets, for which the vanishing of the magnetization is enforced by the combined action of time-reversal and translation/inversion symmetry, in AMs this involves an additional rotation or mirror operation. This symmetry operation causes the band structure to be spin polarized, also in the absence of spin-orbit coupling (SOC) [1]. The presence of spin-polarized bands and Fermi surfaces makes AMs potentially appealing as spin-polarized current sources for antiferromagnetic spintronics [5,12–17]. On the basis of magnetic symmetries [1], an extensive set of candidate materials exhibiting collinear altermagnetism have been identified from electronic structure calculations, both three dimensional (3D) [8] and 2D [18]. While experimentally the spin splitting of electronic bands has been observed by photoemission spectroscopy [19–23], exploration of AM (magneto-) transport

properties for antiferromagnetic spintronic purposes, which capitalizes on their strong exchange interactions [24], has only just begun and has so far focused on harnessing their anomalous and spin Hall effects [9,13,15,20,25–28]. For instance it has recently been shown that altermagnetic spin splitting is at the origin of a spin Hall effect in RuO₂ [12]. In this context, we identify natural possibilities for spin-to-charge conversion processes at AM interfaces that fundamentally go beyond the canonical linear Rashba-Edelstein effect [29–31], namely the generation of an in-plane magnetization in response to a planar electric field caused by antisymmetric spin texture at the Fermi surface. Indeed, we show that this response is actually suppressed by AM. Instead, the competition between Rashba SOC (RSOC) and AM triggers *anomalous* nonlinear Edelstein effects (EEs): we will show that a magnetization transverse to the interface is induced by an in-plane applied electric and magnetic field (or an in-plane magnetization)—or alternatively as nonlinear magnetic response to purely planar electric fields. This spin-to-charge conversion in electric and magnetic field can be depicted by an in-plane current forcing the magnetic moments out of the plane, as schematically indicated in Fig. 1. Also the purely electric nonlinear case will be understood in such basic terms.

It should be noted that, while AM may be strictly defined in the limit of vanishing SOC, it still defines an essential energy scale, determining for instance the altermagnetic anomalous Hall response [9] and electronic properties related to the topology of altermagnetic electronic structures [25]. At interfaces and in 2D, the RSOC is induced by inversion-symmetry

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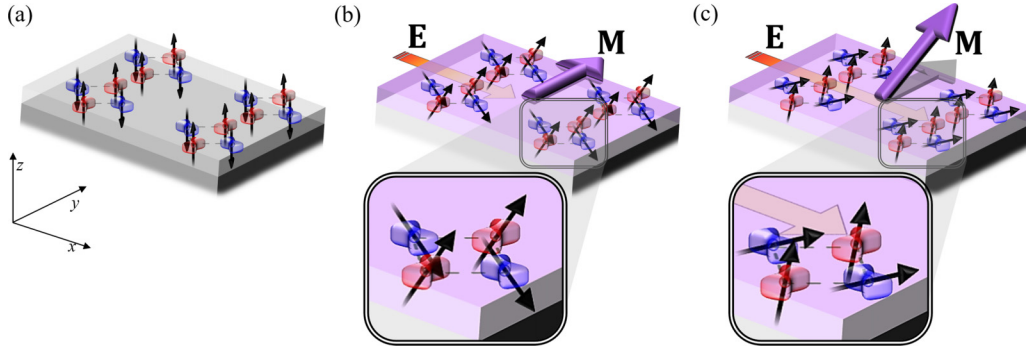


FIG. 1. Schematic representation of the transversal, nonlinear altermagnetic EE. (a) Out-of-plane spin arrangement in the absence of driving current with zero net magnetization. (b) An in-plane driving current tilts the out-of-plane spins resulting in an in-plane transverse magnetic field. (c) A stronger current is able to further tilt the spins resulting in a nonlinear magnetization, which is also out of plane.

breaking and naturally produces a spin-momentum locking without net magnetization. Indeed, the coexistence of AM spin splitting and the RSOC has been pointed out [32–34], but so far not investigated as a mechanism to achieve an anomalous Rashba-Edelstein response and nonlinear spin-to-charge conversion.

The standard linear EE originating from RSOC locks the spin of electrons in plane and orthogonal to their momentum, respecting spinful time-reversal symmetry. Then, an applied in-plane electric field results in an imbalance in the spin density at the Fermi surface and a net in-plane magnetization orthogonal to the induced current. Interestingly, in the presence of only RSOC, the second-order nonlinear EE [35–37] vanishes by symmetry. However, it may be triggered by, for instance, introducing orbital degrees of freedom [35] or a time-dependent setting [36]. In contrast to RSOC, AM breaks time-reversal symmetry, so that in collinear AMs opposite momenta carry the same spin orientation. This renders the momentum-space magnetic textures generated by RSOC and AM fundamentally different: the two interactions compete.

II. ALTERMAGNETIC RASHBA MODEL

To explore the physical consequences of this competition, we start by considering the 2D effective continuum Hamiltonian

$$\hat{H} = \frac{k^2}{2m} + \gamma(k_x^2 - k_y^2)\hat{\sigma}_z + \alpha\hat{z} \cdot (\vec{\sigma} \times \mathbf{k}) - \mu, \quad (1)$$

where $\hat{\sigma}_i$ is the Pauli matrix in the direction $\hat{i}$, α is the RSOC interaction, γ is the altermagnetic d -wave coupling with symmetry $[C_2][C_{4z}]$ [1], μ is the chemical potential, and $\hbar$ is set to unity. The resulting bands and their spin texture for given chemical potential are illustrated in Figs. 2(a) and 2(b). Note that the chirality of the resulting noncoplanar spin texture is determined by $\text{sgn}(\gamma\alpha)$. To maintain contact with real materials, we choose the numerical values of the altermagnetic coupling γ and the effective mass m by explicitly comparing our $\mathbf{k} \cdot \mathbf{p}$ model with the local band structure close to the Γ point of a RuO₂ bilayer. Details of *ab initio* calculation and band-structure fitting are given in Appendix B. We use $m = 0.152 \text{ eV}^{-1} \text{ \AA}^{-2}$ and $\gamma = 1.881 \text{ eV \AA}^2$. While the magnetic nature of RuO₂ is a debated question [38–42], thin films of RuO₂ [43–47] have been characterized as d -wave

altermagnets. Here, we consider this system just as a representative d -wave altermagnetic model, in which both the band filling and the effective RSOC can, in principle, be tuned via gating.

A. Linear Edelstein responses

The linear EE is defined through $M_j = \chi_{ij}E_i$, where E_i is the electric field along the direction $\hat{i}$ and χ_{ij} is the Edelstein susceptibility. The spin accumulation M_j can be obtained in the Boltzmann framework as

$$M_j(\mu) = -\mu_b \sum_{\mathbf{k}, v} |e| (\vec{\Lambda}_{\mathbf{k}}^v \cdot \mathbf{E}) \delta[\varepsilon_{\mathbf{k}}^v - \mu] [v s_j(\mathbf{k})], \quad (2)$$

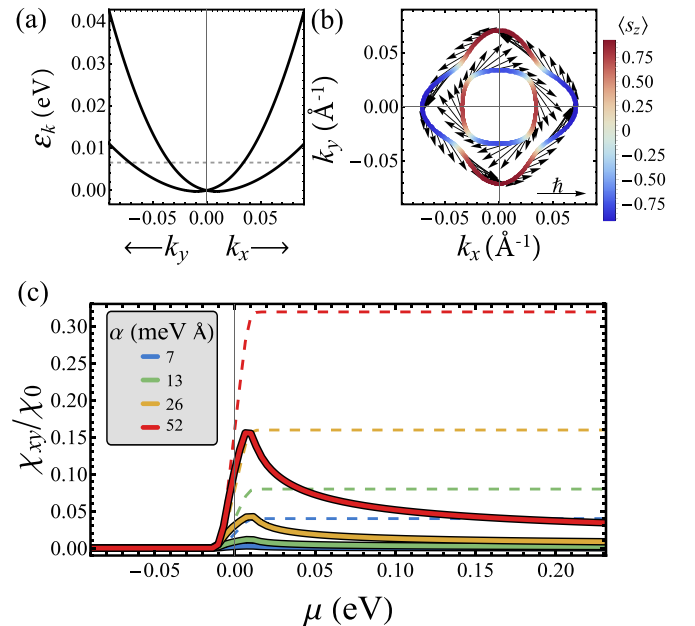


FIG. 2. Competing altermagnetic and Rashba spin-orbit interactions. (a) Electronic dispersion resulting from the continuum Hamiltonian (1) $\alpha = 52 \text{ meV \AA}$. (b) Fermi surface and spin structure for fixed chemical potential [dashed line in panel (a)]. (c) Linear Edelstein susceptibility χ_{xy}/χ_0 ($\chi_0 = 2.44 \times 10^2 \mu_B \text{ \AA V}^{-1}$) for different α . The dashed lines correspond to the pure RSOC case ($\gamma = 0$).

where we made μ explicit in the Dirac delta since we treat it as an external parameter, μ_b is the Bohr magneton, $\varepsilon_{\mathbf{k}}$ are the eigenvalues with band index $v = \pm$ that, for the Hamiltonian in Eq. (1), are given by

$$\varepsilon_{\mathbf{k}}^{\pm} = \frac{k^2}{2m} \pm \sqrt{\alpha^2(k_y^2 + k_x^2) + \gamma^2(k_x^2 - k_y^2)^2} = \frac{k^2}{2m} \pm \delta\varepsilon, \quad (3)$$

and s_j is the j component of the spin vector $\mathbf{s} = (\alpha k_y, -\alpha k_x, \gamma(k_x^2 - k_y^2))/\delta\varepsilon$. The mean free path is computed within the relaxation-time approximation, $\bar{\Lambda}_{\mathbf{k}}^v = \tau_{\mathbf{k}}^v \mathbf{v}_{\mathbf{k}}^v$, with $\tau_{\mathbf{k}}^v$ and $\mathbf{v}_{\mathbf{k}}^v = \nabla_{\mathbf{k}} \varepsilon_{\mathbf{k}}^v$ being the transport lifetime and the group velocity, respectively.

Supposing, for simplicity, that $\tau_{\mathbf{k}}^v = \tau$, and taking $\mathbf{E} = E_0 \hat{i}$, Eq. (2) can be written in terms of the susceptibility per unit cell, χ_{ij} , as

$$\chi_{ij} = -a\chi_0 \sum_{v=\pm} \int d^2\mathbf{k} v s_j(\mathbf{k}) \delta(\varepsilon_{\mathbf{k}}^v - \mu) v_i^v(\mathbf{k}), \quad (4)$$

where $\chi_0 = \frac{\tau|e|\mu_b S_{\text{cell}}}{4\pi^2 a}$, with a being the lattice parameter, S_{cell} the area of the unit cell, and $\tau = 10^{-12}$ s the typical order of magnitude of oxides [30].

The conventional EE is governed by χ_{xy} and RSOC alone is sufficient to induce it, as shown for our continuum Hamiltonian in Fig. 2(c). The symmetry group of the Hamiltonian $[C_2||C_{4z}]$ with broken inversion symmetry enforces $\chi_{xx} = \chi_{yy} = 0$, $\chi_{xy} = -\chi_{yx}$, and $\chi_{iz} = 0$ for $i = x, y$. Without AM and in-plane magnetic field ($\gamma = 0$), the linear Edelstein susceptibility χ_{xy} scales with the RSOC α and quickly saturates for large chemical potential μ ; see dashed lines in Fig. 2(c). Here, we vary α in a range around 0.05 eV Å, which is a realistic range of values for oxides [48–51], and may be experimentally achieved by, e.g., a tunable top gate or interfacial electric field [52]. Clearly, a finite altermagnetic coupling, γ , significantly suppresses the EE, particularly at higher μ , and this for any value of γ leads to a pronounced peak in χ_{xy} in the vicinity of the band bottom. Thus, as we expected on the basis of symmetry considerations, AM and the conventional EE strongly compete.

B. Definition of nonlinear Edelstein responses

Clearly, the RSOC, with its in-plane spin-momentum locking, cannot alone produce an out-of-plane, transverse Edelstein response, i.e., χ_{xz} vanishes identically. Even if the γ term tends to align spins out of plane, RSOC in the presence of altermagnetism is not sufficient to induce a transversal response. Such an anomalous transversal response actually requires altermagnetism and higher orders in the fields, as we will detail in the following. One may distinguish two types of nonlinear (second order) responses, one involving an in-plane electric and magnetic field and one involving only in-plane electric fields to second order. In the following, we focus solely on the extrinsic contribution arising from impurity scattering. For such a model, the intrinsic interband Edelstein contribution, originating from the off-diagonal terms [53], vanishes by symmetry (see details in Appendix F). Other

intrinsic effects, such as those related to quantum geometry [54] or deviations from the rigid-band approximation [55,56], could in principle be considered, but they are expected to be more relevant for complex multiband structures. Here, we highlight the standard contribution, which should also dominate for thin films on a substrate, where the constant-scattering-time approximation is valid [57].

The case of combined electric and magnetic fields is most easily considered by introducing a Zeeman term in Eq. (1), orthogonal to the applied electric field E_x , of the form $H_h = h_y \sigma_y$ and then using Eq. (4). The H_h term corresponds to an effective homogeneous spin splitting, which may arise from a Zeeman field or from weak ferromagnetism induced by Dzyaloshinskii-Moriya (DM) interactions, generally allowed by AM symmetries [18,58–60]. The presence of the *in-plane* magnetic field spontaneously breaks the $\mathcal{TC}_{4z}$ symmetry, allowing out-of-plane Edelstein response. This response is nonlinear as it depends both linearly on electric field and magnetic coupling and vanishes when either one is zero (see Appendix A). Incorporating $\mathbf{h}$ directly into the Hamiltonian offers the advantage that it is easy to go beyond its linear response.

To assess the nonlinear EE involving only electric fields, it is more convenient to introduce the second order Edelstein susceptibility of the form $M_j = \tilde{\chi}_{ij} E_i^2$, with

$$\tilde{\chi}_{ij} = -\tilde{\chi}_0 \sum_{v=\pm} \int d^2\mathbf{k} v \frac{\partial s_j(\mathbf{k})}{\partial k_i} \delta(\varepsilon_{\mathbf{k}}^v - \mu) v_i^v(\mathbf{k}), \quad (5)$$

where $\tilde{\chi}_0 = \frac{\mu_b e^2 \tau^2}{4\pi^2} S_{\text{cell}}$ (see Appendix D for the full derivation). The $\mathcal{TC}_{4z}$ symmetry does not rule out an out-of-plane response and enforces $\tilde{\chi}_{xxz} = -\tilde{\chi}_{yyz}$ (while it easily excludes an in-plane spin response $\tilde{\chi}_{iij} = 0$ for $i, j = x, y$).

III. CALCULATION OF NONLINEAR EDELSTEIN RESPONSES

Having established that, due to the interplay of RSOC with AM symmetry allowing anomalous nonlinear EEs, we now determine them in an experimentally relevant parameter range. For the effective in-plane field h_y resulting from weak ferromagnetism, we choose $h_y = 40$ meV as a benchmark value [18]. The first effect of the in-plane Zeeman coupling is to lift the degeneracy at Γ , as shown in Fig. 3(a), in contrast with the simple Rashba system, for which the in-plane magnetic field can only move the degeneracy point. Moreover, as mentioned above, it breaks the symmetry of the system both in the shape of the Fermi surface and in the chiral structure of the spin, as one can see in Fig. 3(b). The resulting Edelstein susceptibility tensor χ is shown in Fig. 3(c). While χ_{xx} vanishes, χ_{xy} depends nonmonotonically on chemical potential and reaches its maximum after occupying the first band. Compared to the case $h_y = 0$, χ_{xy} grows slower to its maximum, but reaches a value similar to the situation with pure RSOC. Moreover, the in-plane magnetic field is able to induce a substantial out-of-plane anomalous transversal response χ_{xz} due to the large altermagnetic coupling, with the transversal component reaching larger values than the in-plane response, even larger than the canonical linear RSOC situation.

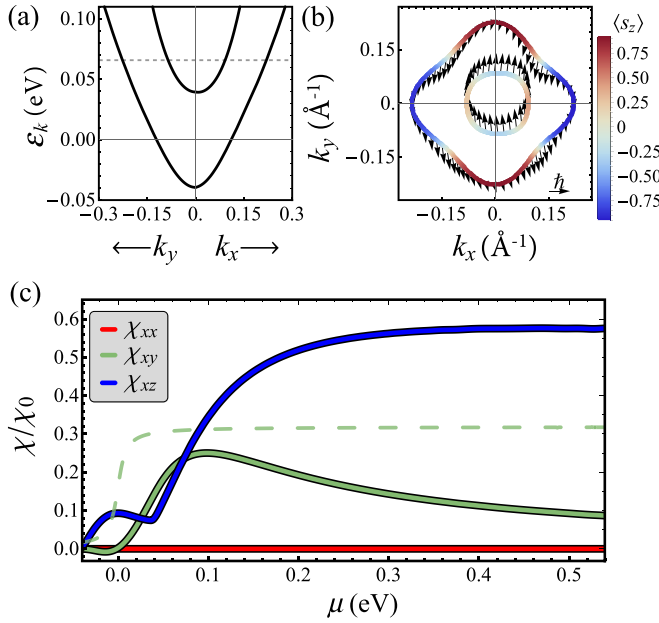


FIG. 3. Anomalous nonlinear EE due to a combined planar magnetic and electric field. (a) Electronic dispersion from Hamiltonian (1) with $\alpha = 52$ meV Å and $h_y = 40$ meV. (b) Fermi surface and spin structure for fixed chemical potential; see the dashed line in (a). (c) Edelstein response χ_{xi}/χ_0 ($\chi_0 = 2.44 \times 10^2 \mu_B \text{ Å V}^{-1}$) for the directions $i = x, y, z$. The green dashed line corresponds to the pure RSOC case ($\gamma = h_y = 0$).

The nonmonotonic behavior of χ_{xz} as a function of μ derives from the competition and complex interplay of the different interactions. At low filling, even an infinitesimal h_y breaks the TC_{4z} symmetry, leading to a characteristic threshold behavior in the response. Increasing μ , the out-of-plane response is first slightly suppressed and later saturates to a constant value in the altermagnetic-dominated regime. Crucially, all three ingredients are required to exhibit the out-of-plane response: the altermagnetic interaction enforces an out-of-plane magnetization, the RSOC induces the spin-momentum locking, and the weak ferromagnetism breaks its otherwise symmetric pattern required for this nonlinear Edelstein response.

On the other hand, without inducing any modification to the energy spectrum, i.e., the electronic bands and the spin structure correspond to the one shown in Fig. 2, an out-of-plane, second-order EE is obtained. The computed Edelstein susceptibilities from Eq. (5), for different values of the RSOC, are shown in Fig. 4. One observes a signature peak right above the band crossing at $\epsilon = 0$, which decreases with the increasing of α , and then a steady decrease of $\tilde{\chi}_{xxz}$ for larger μ . This anomalous response is characteristic of altermagnetic systems with RSOC and vanishes for either $\alpha = 0$ or $\gamma = 0$ (see Appendix D for details). Therefore, this nonmonotonic behavior allows one to individuate the location of band crossing due to RSOC and the strength of the coupling in altermagnetic surfaces. A detailed analysis is presented in Appendix C. When generalized to finite frequencies, the nonlinear susceptibility $\tilde{\chi}_{xxz}$ induces second-harmonic generation. This, in principle, offers an experimentally attractive route to detect

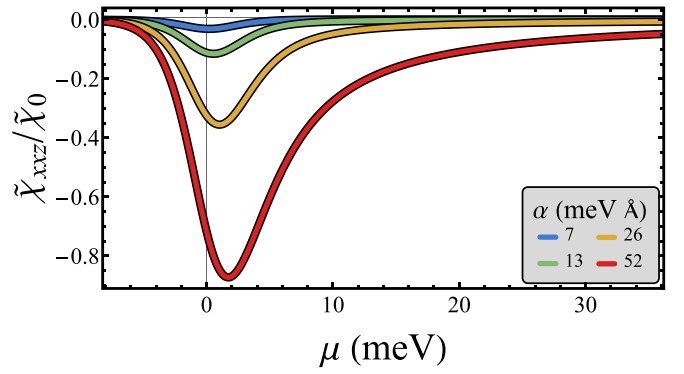


FIG. 4. Second-order electric Edelstein response. $\tilde{\chi}_{xxz}/\tilde{\chi}_0$ (with $\tilde{\chi}_0 = 2.36 \times 10^6 \text{ Å}^2 \text{ V}^{-2}$) versus μ for different values of α .

such a nonlinear response. In our case, the rectifying and doubled frequency terms $\langle \tilde{\chi}_{xxz} \rangle = \text{Re}[\langle \tilde{\chi}_{xxz}^0 \rangle + \langle \tilde{\chi}_{xxz}^{2\omega} \rangle e^{2i\omega t}]$ appear, which are related to Eq. (5) as $\langle \tilde{\chi}_{xxz}^0 \rangle = \tilde{\chi}_{xxz}/2(1 + i\omega\tau)$ and $\langle \tilde{\chi}_{xxz}^{2\omega} \rangle = \tilde{\chi}_{xxz}/2(1 + i\omega\tau)(1 + 2i\omega\tau)$.

IV. SUMMARY AND OUTLOOK

Using a continuum description for AMs based on *ab initio* electronic band structure calculations, we have shown that the interplay of altermagnetic coupling with an interfacial RSOC gives rise to the appearance of out-of-plane spin polarizations throughout the Brillouin zone via nonlinear EEs. While the in-plane linear response can be strongly suppressed with the band filling, the nonlinear Edelstein susceptibilities exhibit strong nonmonotonic behavior as a function of the chemical potential. The nonlinear responses we determined involve a term that depends on the in-plane electric and the magnetic fields and a second-order term in the electric field. For the former term, an anomalous out-of-plane magnetization is generated, exceeding the in-plane EE. For the latter term, the out-of-plane magnetization is the only response allowed and it is enhanced when the chemical potential is close to the crossing of the electronic bands due to RSOC. In both cases, it is remarkable that out-of-plane magnetization is obtained in response only to in-plane fields. All these features can be exploited to fully characterize the altermagnetic nature of the electron gas at the interface. Indeed, the behavior of the magnetization with the filling can be directly related to the presence of the spin-orbit, DM interaction, and band crossings. To isolate the essential physics, we focused on a minimal model capturing the local interplay between altermagnetic exchange and Rashba spin-orbit coupling near the Γ point. This approach reveals the core mechanism underlying the anomalous Edelstein response in altermagnets. Other band crossings at high-symmetry points can be described within the same framework (see Appendix E) and while a full multiband analysis may introduce quantitative differences, it does not alter the fundamental physical picture. The uncovered *anomalous* nonlinear altermagnetic Edelstein responses may be used experimentally to induce efficient spin-to-charge conversion at altermagnetic interfaces leading to electronic and spintronic applications.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [61].

APPENDIX A: SPIN-TEXTURE ANALYSIS

In this Appendix we analyze in detail the spin texture of the Hamiltonian including also the Zeeman coupling

$$\hat{H} = \frac{k^2}{2m} + \gamma(k_x^2 - k_y^2)\hat{\sigma}_z + \alpha\hat{z} \cdot (\vec{\sigma} \times \mathbf{k}) + h_y\hat{\sigma}_y - \mu. \quad (\text{A1})$$

The eigenvalues of this 2×2 matrix are easily obtained, as well as the direction of the spin quantization axis

$$\varepsilon^\pm = \frac{k^2}{2m} \pm \sqrt{(h_y - \alpha k_x)^2 + \alpha^2 k_y^2 + \gamma^2(k_x^2 - k_y^2)^2}, \quad (\text{A2})$$

$$\mathbf{s} = \frac{(\alpha k_y, h_y - \alpha k_x, \gamma(k_x^2 - k_y^2))}{\sqrt{(h_y - \alpha k_x)^2 + \alpha^2 k_y^2 + \gamma^2(k_x^2 - k_y^2)^2}}. \quad (\text{A3})$$

For $\gamma = 0$, the band structure exhibits a crossing at $\mathbf{k} = (h_y/\alpha, 0)$. However, the spin texture is tilted along the y direction, distorting the in-plane chirality. When $\gamma \neq 0$, the s_z component of the spin becomes an odd function of $\mathbf{k}$, as the parity symmetry is broken by the denominator of $\mathbf{s}$.

Notably, for $\alpha = 0$, the presence of h_y alone does not break the even symmetry of s_z [i.e., $s_z(\mathbf{k}) = s_z(-\mathbf{k})$]. This highlights the need for all three ingredients—Rashba spin-orbit coupling (α), Zeeman field (h_y), and anisotropic coupling (γ)—to generate an out-of-plane Edelstein effect. In Fig. 5 we show the values of s_y and s_z as a function of k_x for $k_y = 0$ for $\alpha \neq 0$ and by varying γ and h_y . In case of a pure Rashba coupling only s_y is present and is constant with a change of sign at $k_x = 0$, while when a Zeeman field is present along the line $k_y = 0$ the

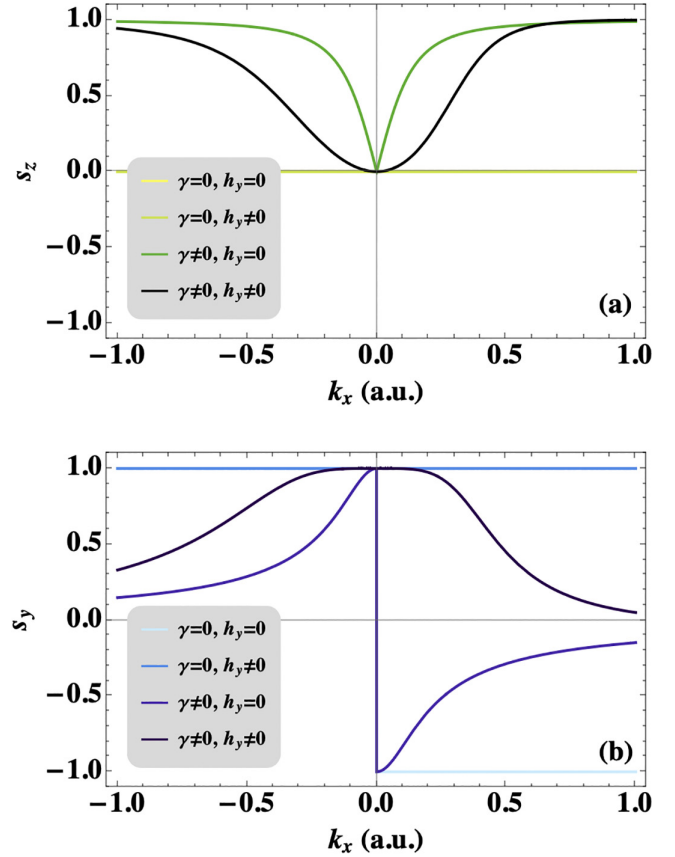


FIG. 5. s_z (a) and s_y (b) as a function of k_x and $k_y = 0$ with benchmark values of $\alpha \neq 0$ and by varying γ and h_y . The values of α , γ , and h_y have been chosen to make the plot clearer.

spin is constant. When the altermagnetic coupling is turned on, the even (odd) symmetry in k_x is present for s_z (s_y), which is broken while $h_y \neq 0$.

APPENDIX B: *AB INITIO* RuO₂ BILAYER BAND STRUCTURE AND PARAMETER FIT

Here we estimate the strength of the altermagnetic coupling γ by explicit comparison with the local band structure of a RuO₂ bilayer. Based on the bulk crystal structure, we construct a bilayer oriented along the c axis of RuO₂ bulk, containing two Ru ions related by fourfold rotoinversion symmetry. The altermagnetic bilayer can be represented with the space group $Cmm2$ (no. 35), with a unit cell having $a = 6.35$ Å. For this structure, we perform density-functional calculations in the generalized gradient approximation based on the FPLO code [62]. We use a k -points mesh having $16 \times 16 \times 1$ subdivisions along with a tetrahedron method for Brillouin zone integrations. Figure 6 shows the band structure in the absence of RSOC. We apply our effective two-band model to describe the two bands closest to the Fermi energy at Γ . The parameter γ and m of Hamiltonian (A1) (for $\alpha = 0$ and $h_y = 0$) can be extracted from DFT data of the RuO₂ bilayer. We note here that our model has a spin-ordering rotation of 45° with respect to the RuO₂, which simplifies the analysis of the Edelstein susceptibility tensor. In Fig. 6 we show the DFT data of RuO₂ and the fit of the band structure for the

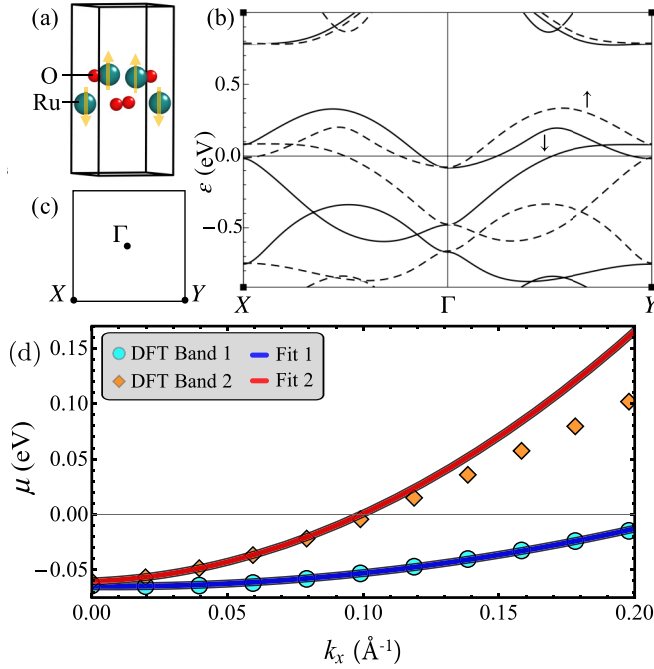


FIG. 6. (a)–(c) Bilayer of RuO₂ and its electronic structure density-functional calculations. (a) Crystal structure. (b) Band structure along high symmetry points indicated in (c). (d) Fit of the bands crossing the Fermi level nearby Γ along the direction ΓY .

two bands crossed by the Fermi level. The two bands are well fit by

$$\begin{aligned} E_- &= 1.412 \text{ eV } \text{\AA}^2, & k_x^2 &= 0.023 \text{ eV } \text{\AA}, & k_x &= 0.065 \text{ eV}, \\ E_+ &= 5.175 \text{ eV } \text{\AA}^2, & k_x^2 &= 0.094 \text{ eV } \text{\AA}, & k_x &= 0.060 \text{ eV}. \end{aligned} \quad (\text{B1})$$

By comparison with the analytical expression of the eigenvalues of Hamiltonian (A1) we obtain $m = 0.152 \text{ eV}^{-1} \text{\AA}^{-2}$ and $\gamma = 1.881 \text{ eV } \text{\AA}^2$.

APPENDIX C: PARAMETRIC DEPENDENCE OF THE EDELSTEIN RESPONSE

This Appendix details the dependence of the Edelstein responses on the RSOC parameter (α) and the Zeeman coupling (h_y). In Fig. 7, we show the behavior of the EE for different h_y . The in-plane response is even in h_y , while the out-of-plane response is odd in h_y . The inversion of the magnetic field $h_y \rightarrow -h_y$ can be canceled by a corresponding change in sign $k_x \rightarrow -k_x$, which leaves the eigenvalues of the Hamiltonian unchanged. With such a transformation, the group velocity $v_x \rightarrow -v_x$, and the same goes for the in-plane spin $s_y \rightarrow -s_y$, while the out-of-plane spin, determined by the altermagnetic term, which is even in k_x , remains unchanged $s_z \rightarrow s_z$. Therefore, from the definition of the EE susceptibility, we see that the in-plane response is even, while the out-of-plane response is odd in h_y . Correspondingly, the out-of-plane response, which vanishes for $h_y \rightarrow 0$, must have a linear behavior $\chi_{xz} \propto h_y$ for very small fields. In Fig. 7 we show χ as a function of the Zeeman coupling for a fixed

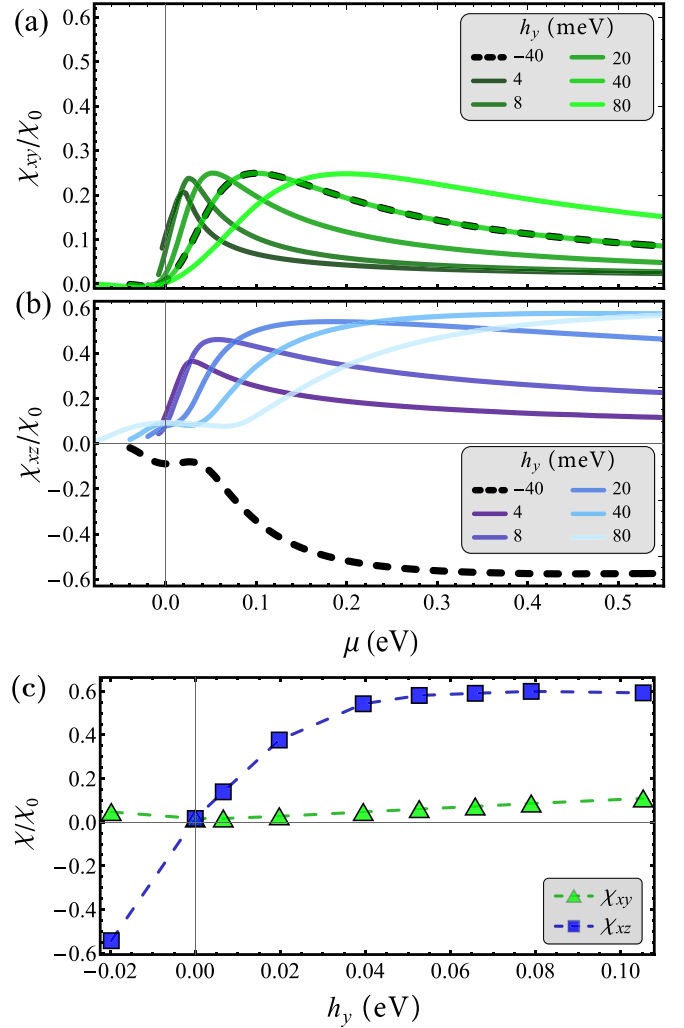


FIG. 7. (a) χ_{xy} and (b) χ_{xz} as a function of the chemical potential for $\alpha = 52 \text{ meV } \text{\AA}$ and different values of the in-plane Zeeman field h_y . (c) Edelstein susceptibility behavior with Zeeman coupling for $\alpha = 52 \text{ meV } \text{\AA}$ and for fixed chemical potential $\mu = 1.05 \text{ eV}$ and $\gamma = 1.881 \text{ eV } \text{\AA}^2$. Lines are guides to the eye.

value of the chemical potential $\mu/h_y \gg 1$, in order to show the behavior of the χ in the saturation region.

Second-order Edelstein susceptibility. The ratio between the RSOC and the altermagnetic energy scales is determined by $|\alpha|/|k_F \gamma|$, with k_F to be the Fermi momentum. It is a measure for their competition, which is strongest when $|\alpha|/|k_F \gamma| \simeq 1$. For the value $|\alpha|/|k_F \gamma| \simeq 1$ we observe that the second-order anomalous susceptibility has a peak. In Fig. 8 we show the second-order anomalous Edelstein susceptibility for different values of γ . It tends to peak at a small value of the chemical potential close to the band crossing at Γ (see Fig. 2 of the main text). The peak is substantially broadened when $|\gamma k_F| > |\alpha|$. Indeed the peak shape of the second-order response is lost for small values of γ ($\gamma = 0.2 \text{ eV } \text{\AA}^2$). Within the considered range of μ values, the maximum explored Fermi momentum corresponds to $k_F = 0.22 \text{ \AA}^{-1}$, so that $|\alpha| > |\gamma k_F|$. However, since the nonrelativistic spin splitting is expected to be larger (as seen from DFT

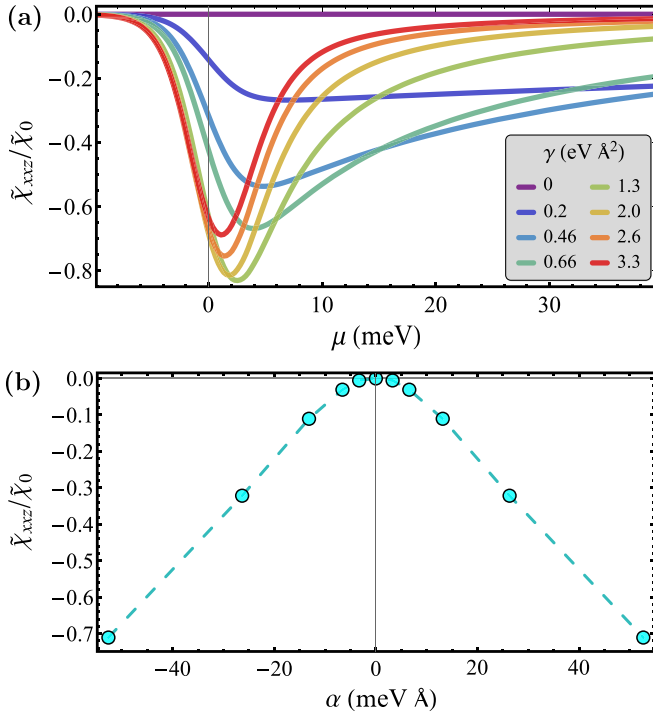


FIG. 8. (a) Second-order Edelstein susceptibility as a function of the chemical potential for $\alpha = 52$ meV Å and different values of γ . (b) Second-order Edelstein susceptibility $\tilde{\chi}_{xxz}$ as a function of α for $\mu = 0$ and $\gamma = 1.881$ eV Å². The line is a guide to the eye.

calculation), the peak-shape feature might be used as a probe of the band crossing due to the Rashba coupling.

Finally, to quantify the effect of the RSOC, we show in Fig. 8 the dependence on α of the second-order Edelstein susceptibility. For a fixed chemical potential the susceptibility is an even function of α , showing also a sign change for α close to zero.

APPENDIX D: BOLTZMANN APPROACH TO NONLINEAR EE

The first- and second-order Edelstein reads

$$M_k = \chi_{ik} E_i + \tilde{\chi}_{ijk} E_i E_j, \quad (D1)$$

where χ_{ik} and $\tilde{\chi}_{ijk}$ are the linear and second-order Edelstein susceptibilities, respectively. In the Boltzmann approach, the magnetization is obtained as

$$M_k = \mu_B \sum_{n \text{ occ}} \int \frac{d^2 \mathbf{k}}{(2\pi)^2} s_k^n(\mathbf{k}) f(\mathbf{k}), \quad (D2)$$

with s_k^n the spin component of band n and $f(\mathbf{k})$ the distribution function.

In the Boltzmann framework, we expand $f = f_{\text{th}} + \varphi^{(1)} + \varphi^{(2)} + \dots$ in powers of $\mathbf{E}$, with the lowest-order f_{th} being the Fermi distribution function, and for homogeneous, stationary transport with constant relaxation time τ , the first- and second-order corrections read

$$\varphi^{(1)} = |e| \tau \mathbf{E} \cdot \nabla_{\mathbf{k}} f_{\text{th}}, \quad \varphi^{(2)} = e^2 \tau^2 \frac{\partial}{\partial k_j} \left(\frac{\partial f_{\text{th}}}{\partial k_i} \right) E_i E_j. \quad (D3)$$

The second-order susceptibility is then given by

$$\tilde{\chi}_{xxz} = -\mu_B e^2 \tau^2 S_{\text{cell}} \sum_{\nu} \int \frac{d^2 \mathbf{k}}{4\pi^2} \nu \frac{\partial s_z(\mathbf{k})}{\partial k_x} \delta(\epsilon_{\mathbf{k}}^{\nu} - \mu) v_x^{\nu}(\mathbf{k}). \quad (D4)$$

An estimate of the prefactor yields

$$\tilde{\chi}_0 = \frac{\mu_B e^2 \tau^2}{4\pi^2} S_{\text{cell}} \simeq 2.36 \times 10^{-14} \frac{\text{m}^2}{\text{V}^2}. \quad (D5)$$

We adopt the rigid band approximation, which is adequate for the purpose of our model. In principle, for a more complex multiband structure, corrections to the eigenstates due to a strong (or oscillating) electric field could be considered [55,56], but these effects are not critical in our approach. In the absence of Rashba coupling it is straightforward to see that $\frac{\partial s_z}{\partial k_x} = 0$ and therefore $\chi_{xxz} = 0$. We use these expressions to determine the nonlinear Edelstein response shown in the main text.

APPENDIX E: TIGHT-BINDING MODEL

In this Appendix we highlight the role of the crossing at high-symmetry points and the multiband crossings on the linear Edelstein response. We construct a minimal tight-binding model from the irreducible representations of the alternating magnetic symmetry group adopted in the main text, including the inversion-breaking Rashba term

$$\begin{aligned} H(\mathbf{k}) = & [-2t(\cos k_x + \cos k_y) - \mu] \sigma_0 \tau_0 \\ & - 2\gamma(\cos k_x - \cos k_y) \sigma_z \tau_x - 2t_1(\cos k_x + \cos k_y) \sigma_0 \tau_x \\ & + \alpha[\sin k_x \sigma_y - \sin k_y \sigma_x] \tau_x. \end{aligned} \quad (E1)$$

Here t_1 has been included to split the fourfold degeneracy. For $t_1 = 0$ and $t = 1/2m$ the expansion at $\mathbf{k} \approx 0$ is equivalent to Hamiltonian (A1). The band structure is shown in Fig. 9, together with the Edelstein response function for varying filling of the bands. The dominant contribution to the Edelstein response comes from the band crossings, as one could expect. In particular, the band crossings at the Γ points, which give the largest contributions, are well described by our $\mathbf{k} \cdot \mathbf{p}$ local models, providing a concrete example of the local nature of the Edelstein response. At large fillings, a specular contribution comes from the band crossings at X and Y, due to a change of chirality of the spin texture close to the corner of the Brillouin zone. Band crossings at M and M' do not produce an Edelstein response, since the Rashba operator has vanishing matrix elements between the doubly degenerate eigenstates. This is immediately visible by expanding the Hamiltonian (E1) $\mathbf{q}_{\Delta \mathbf{k}} = \mathbf{k} - \Delta \mathbf{k}$ with $\Delta \mathbf{k} = (\pm\pi, 0)$

$$\begin{aligned} H(\mathbf{q}_{(\pi,0)}) \approx & t(-q_x^2 + q_y^2) \sigma_0 \tau_0 + \gamma(4 - q_x^2 - q_y^2) \sigma_z \tau_x \\ & + t_1(-q_x^2 + q_y^2) \sigma_0 \tau_x + \alpha(-q_x \sigma_y - q_y \sigma_x) \tau_x, \end{aligned} \quad (E2)$$

so for $\mathbf{q} \rightarrow 0$ the doubly degenerate eigenstates with $\sigma_z \tau_x = +1$ (or $\sigma_z \tau_x = -1$) lead to a zero matrix element for the RSOC.

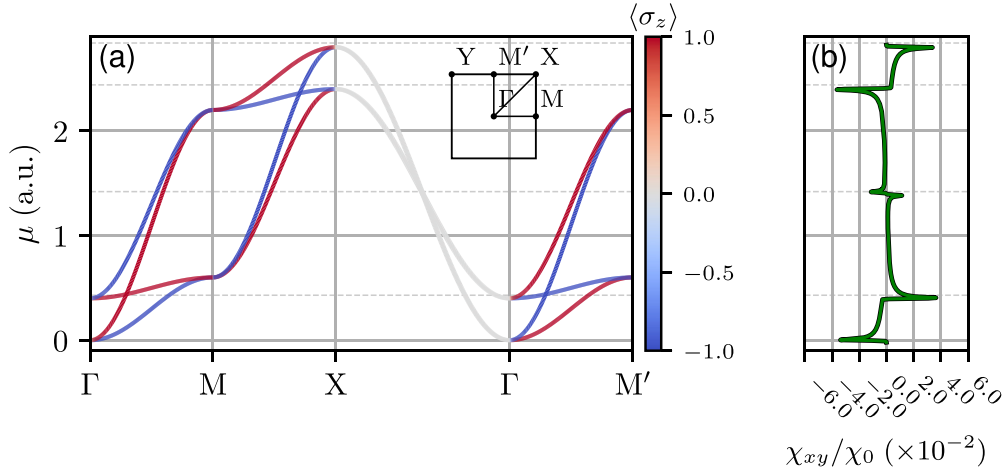


FIG. 9. (a) Band structure of the tight-binding model of Hamiltonian (E1) using $t = 0.6$, $\gamma = 0.2$, $t_1 = 0.05$, and $\alpha = 0.01$. The energy has been shifted in order to set the minimum of the bands to $\mu = 0$. (b) Edelstein susceptibility χ_{xy}/χ_0 as a function of the chemical potential.

Another band crossing occurs at $k_x = k_y = \pi/2$; a local expansion gives

$$\begin{aligned} H(\mathbf{q}_{(\pi/2, \pi/2)}) &\approx 2t(q_x + q_y)\sigma_0\tau_0 + 2\gamma(q_x - q_y)\sigma_z\tau_x \\ &+ 2t_1(q_x + q_y)\sigma_0\tau_x + \alpha((1 - q_x^2)\sigma_y \\ &+ (-1 + q_y^2)\sigma_x)\tau_x. \end{aligned} \quad (\text{E3})$$

For $q \rightarrow 0$, the Rashba term reduces to $\alpha(\sigma_y - \sigma_x)\tau_x$, so the crossing is gapped by the RSOC which enforces an in-plane spin. At finite $\mathbf{q}$, the spin tilts out of plane due to the alternating terms, enforcing a spin-momentum locking and inducing a symmetry-allowed Edelstein response, which is clearly visible at half filling, albeit of lower intensity than the response coming from the Γ and X points.

APPENDIX F: INTRINSIC INTERBAND CONTRIBUTION

In this section, we discuss the role of the interband intrinsic contribution to the Edelstein effect in our model. Its general definition [53] is

$$\chi_{ij}^{\text{int}} = e\hbar \int \frac{d^2\mathbf{k}}{(2\pi)^2} \sum_{m \neq n} \text{Im} \left[\frac{s_j^{nm}(\mathbf{k}) v_i^{mn}(\mathbf{k})}{(\varepsilon_{\mathbf{k}}^n - \varepsilon_{\mathbf{k}}^m)^2} \right] [f(\varepsilon_{\mathbf{k}}^n) - f(\varepsilon_{\mathbf{k}}^m)], \quad (\text{F1})$$

where $s_j^{nm}(\mathbf{k}) = \langle u_n(\mathbf{k}) | \sigma_j | u_m(\mathbf{k}) \rangle$ and $v_i^{mn}(\mathbf{k}) = \langle u_m(\mathbf{k}) | \partial_{k_i} H(\mathbf{k}) | u_n(\mathbf{k}) \rangle$ are the off-diagonal matrix elements of the Pauli matrix and group velocity operators over the eigenstates of the Hamiltonian.

This contribution has to be compared with the extrinsic Edelstein susceptibility

$$\chi_{ij}^{\text{ext}} = e\tau \sum_n \int_{\text{BZ}} \frac{d^2\mathbf{k}}{(2\pi)^2} \left(-\frac{\partial f_0}{\partial \varepsilon_{\mathbf{k}}^n} \right) v_i^n(\mathbf{k}) s_j^n(\mathbf{k}), \quad (\text{F2})$$

where all quantities are expressed in standard units. From these definitions, it becomes immediately clear that the intrinsic contribution can dominate over the extrinsic one only over those regions of the Brillouin zone where $\varepsilon_m - \varepsilon_n \lesssim \hbar/\tau$ (excepting cases where the extrinsic contribution vanishes due to some symmetry, which we have established not to be the case for the components of the Edelstein susceptibility we determine). Since $\hbar/\tau \sim 0.7$ meV, whereas all the energy scales within our model Hamiltonian are significantly larger, clearly the interband intrinsic contribution can become competitive only close to near-degeneracy points. Let us therefore consider such contribution. If Zeeman terms are included, the spectrum is fully gapped, with typical gaps larger than ~ 50 meV. In this regime, the intrinsic susceptibility is always negligible compared to the extrinsic one. The only case requiring more careful analysis is the gapless limit at $\mathbf{k} = 0$. By considering Hamiltonian (1), and using Eq. (F1), one obtains for the integrand of $\chi_{ix} = e\hbar \int I(s_i) d^2\mathbf{k}$

$$I(s_z) = -\frac{1}{4\pi^2} \frac{k_y \alpha^2}{4[(k_x^2 + k_y^2)\alpha^2 + (k_x^2 - k_y^2)^2 \gamma^2]^{3/2}} \quad (\text{F3})$$

and

$$I(s_y) = k_x k_y \mathcal{F}(k_x^2, k_y^2), \quad (\text{F4})$$

where $\mathcal{F}(k_x^2, k_y^2)$ is an even function of $\mathbf{k}$. Both integrands vanish upon Brillouin-zone integration by symmetry, so the intrinsic contribution is absent in this two-band model.

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