

Intrinsic Nonlinear Electric Spin Generation in Centrosymmetric Magnets

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We propose an intrinsic nonlinear electric spin generation effect, which can dominate in centrosymmetric magnets. We reveal the band geometric origin of this effect and clarify its symmetry characters. As an intrinsic effect, it is determined solely by the material's band structure and represents a material characteristic. Combining our theory with first-principle calculations, we predict sizable nonlinear spin generation in single-layer MnBi_2Te_4 , which can be detected in experiment. Our theory opens a new route for all-electric controlled spintronics in centrosymmetric magnets which reside outside of the current paradigm based on linear spin response.

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Electric control of spin degree of freedom is a central topic of spintronics. Much theoretical and experimental effort [1–7] has been devoted to generating spin polarization δs by an applied electric field, characterized by a linear spin response tensor α_{ij} with $\delta s_i = \alpha_{ij}E_j$, where i, j are Cartesian indices and the Einstein summation convention is adopted. In a magnet, the generated spin polarization may further induce torques on the magnetization and even cause magnetic reversal [7]. Importantly, since spin is even under space inversion but electric field is odd, the linear effect is constrained to systems with broken inversion symmetry [1–15]. For the class of centrosymmetric magnets, the linear spin response is strictly forbidden in the bulk and may only occur at interfaces when forming heterojunctions with other materials [16–18].

In this Letter, we unveil that sizable nonlinear spin generation can exist in centrosymmetric magnets, thus substantially extending the playing field of spin-charge conversion. Since the linear response is forbidden, the leading contribution is of the second order:

$$\delta s_i = \alpha_{ij\ell} E_j E_\ell, \quad (1)$$

characterized by a nonlinear response tensor $\alpha_{ij\ell}$. We show that $\alpha_{ij\ell}$ contains an *intrinsic* part determined solely by the band structure. For insulators, $\alpha_{ij\ell}$ is closely connected to the electric polarization of Bloch electrons and can be expressed by an important band geometric quantity, the momentum space Berry connection polarizability (BCP). For metals, there is an extra Fermi surface contribution, which involves the BCP in an extended parameter space. We clarify the symmetry properties of the effect.

The intrinsic response has the advantage of allowing a quantitative evaluation. By combining our theory with first-principle calculations, we study the effect in single-layer MnBi_2Te_4 and find a sizable result that can be detected in experiment. Our Letter develops the theory for nonlinear electric spin generation, uncovers the important roles of BCPs in spintronic effects, and opens the door to new nonlinear spintronic device concepts. The approach here also offers a general recipe for investigating other intrinsic nonlinear response properties of Bloch electrons.

Thermodynamic argument for insulators.—We first present a thermodynamic argument, which applies to insulating cases and captures both linear and nonlinear electric spin generation. It also helps to expose the role of electric polarization in the effect.

To evaluate the spin response, a conventional way is to introduce a fictitious (homogeneous) Zeeman-like field \mathbf{h} that couples to spin in the form of $-\hat{s} \cdot \mathbf{h}$, with \hat{s} the spin operator. This auxiliary field is to be distinguished from the genuine magnetization of the system and is set to zero at the end of the calculation [19]. Under the \mathbf{h} field and the electric field, the electronic enthalpy of the magnetic insulator follows the relation $d\mathcal{H} = -s \cdot d\mathbf{h} - \mathbf{P} \cdot d\mathbf{E}$, where s and \mathbf{P} denote the spin magnetization and the electric polarization of electrons, respectively. According to the Maxwell relation, we have

$$\frac{\partial s_i}{\partial E_j} = \frac{\partial P_j}{\partial h_i}. \quad (2)$$

Therefore, the electrically induced spin generation can be extracted from studying the electric polarization.

To include the second-order spin generation, we need the polarization expanded to the first order in the electric field: $\mathbf{P} = \mathbf{P}^{(0)} + \mathbf{P}^{(1)}$. The zero-field part $\mathbf{P}^{(0)} = -\int [d\mathbf{k}] \mathcal{A}(\mathbf{k})$ is well known [20–23] (we set $e = \hbar = 1$), $\mathcal{A}(\mathbf{k}) = \langle u_n(\mathbf{k}) | i\partial_{\mathbf{k}} | u_n(\mathbf{k}) \rangle$ is the intraband Berry connection for a band eigenstate $|u_n(\mathbf{k})\rangle$, $[d\mathbf{k}]$ is a short-hand notation for $\sum_n d\mathbf{k}/(2\pi)^d$ with d the dimension of the system, and the summation is over all occupied bands. Here and hereafter, for simple notations, we drop the band index n whenever appropriate. $\mathbf{P}^{(0)}$ gives the linear spin response with $\delta\mathbf{s} = \partial_{\mathbf{h}} P_j^{(0)} E_j$, reproducing the result in previous works [11,23–25].

On the other hand, the second-order spin response is contained in $\mathbf{P}^{(1)}$, and the polarization is linear in the \mathbf{E} field [26–28]. It can be expressed as $\mathbf{P}^{(1)} = -\int [d\mathbf{k}] \mathbf{a}(\mathbf{k})$ [29] in terms of the field-induced Berry connection $a_i = G_{ij} E_j$, where

$$G_{ij} = 2\text{Re} \sum_{n' \neq n} \frac{(v_i)_{nn'} (v_j)_{n'n}}{(\epsilon_n - \epsilon_{n'})^3} \quad (3)$$

is known as the BCP for the state $|u_n(\mathbf{k})\rangle$ [29], $(v_i)_{nn'}$ is the interband velocity matrix element, and ϵ_n is the unperturbed energy for $|u_n(\mathbf{k})\rangle$. Combining this result with Eq. (2), we immediately find the second-order spin polarization

$$\delta\mathbf{s} = \partial_{\mathbf{h}} \left[\frac{1}{2} \mathbf{E} \cdot \mathbf{P}^{(1)} \right] \quad (4)$$

with the nonlinear response tensor

$$\alpha_{ij\ell} = -\frac{1}{2} \partial_{h_i} \int [d\mathbf{k}] G_{j\ell}(\mathbf{k}) \quad (5)$$

expressed nicely in terms of the momentum space BCP of occupied states. Recent studies have highlighted the role of this BCP in nonlinear charge transport phenomena [29–36], whereas our result here unveils its significance in the nonlinear spin generation effect.

The thermodynamic argument reveals the important role of electric polarization in spin response. Particularly, the term in the bracket of Eq. (4) is just the material-dependent part of the electric energy density in a dielectric that is of E^2 order. For centrosymmetric systems, the zero-field polarization $\mathbf{P}^{(0)}$ vanishes, so as expected, the linear spin response must also vanish. Meanwhile, $\mathbf{P}^{(1)}$ can be nonzero regardless of the inversion symmetry, so that the nonlinear spin response would become dominant in centrosymmetric systems.

It is also important to note that the argument above applies only to insulators. For metals, the electric polarization ceases to be well defined; hence we need a more general approach to the problem. As we shall see, in a

magnetic metal, there will be additional nonlinear contributions from the Fermi surface.

Intrinsic nonlinear spin generation.—To establish a general result which is also applicable to metallic cases, we develop a semiclassical theory for Bloch electrons in the nonlinear response regime. In Refs. [29,37], Gao *et al.* extended the semiclassical theory to second-order accuracy. Nonetheless, the formulation there is focusing on the charge degree of freedom, but does not explicitly handle spin. Here, we add this missing piece. As is detailed in the Supplemental Material [38], within the extended semiclassical framework, we derive the following spin expectation value corrected to second order of the E field for an electron wave packet centered at $|u_n(\mathbf{k})\rangle$:

$$s_n(\mathbf{k}) = -\partial_{\mathbf{h}} \tilde{\epsilon}_n + \mathbf{\Omega}_{\mathbf{h}\mathbf{k}} \cdot \mathbf{E}. \quad (6)$$

Here, $\tilde{\epsilon}_n = \epsilon_n - (1/2)G_{ij}E_iE_j$ is the field-corrected band energy, and $(\mathbf{\Omega}_{\mathbf{h}\mathbf{k}})_{ij} = \partial_{h_i}(\mathcal{A}_j + a_j) - \partial_{k_j}(\mathfrak{A}_i + \mathbf{a}_i)$ is the field-corrected Berry curvature in the hybrid $k-h$ space. The definitions of the Berry connections \mathfrak{A} and \mathbf{a} are analogous to their counterparts \mathcal{A} and \mathbf{a} in k space. Specifically, $\mathfrak{A}_i = \langle u_n(\mathbf{k}) | i\partial_{h_i} | u_n(\mathbf{k}) \rangle$, and $\mathbf{a}_i = \mathfrak{G}_{ij} E_j$ can be expressed using an h -space BCP

$$\mathfrak{G}_{ij} = -2\text{Re} \sum_{n' \neq n} \frac{(s_i)_{nn'} (v_j)_{n'n}}{(\epsilon_n - \epsilon_{n'})^3}, \quad (7)$$

where the numerator involves the interband matrix elements of spin and velocity operators. Like G_{ij} , \mathfrak{G}_{ij} is gauge invariant, so it is also an intrinsic band geometric property.

With the spin polarization for each state, the total spin polarization in the system can be obtained as

$$\mathbf{s} = \int [d\mathbf{k}] s_n(\mathbf{k}) f_n(\mathbf{k}), \quad (8)$$

where f is the electron distribution function. The second-order spin response is obtained by inserting the expression in Eq. (6) and retaining terms that are of E^2 order. Here, we are particularly interested in the intrinsic contribution that involves only the Fermi distribution function $f_0(\epsilon_n)$ of the unperturbed band structure. The intrinsic nonlinear response tensor is obtained as

$$\alpha_{ij\ell}^{\text{int}} = -\frac{1}{2} \partial_{h_i} \int [d\mathbf{k}] G_{j\ell} f_0 - \int [d\mathbf{k}] (s_i G_{j\ell} + v_j \mathfrak{G}_{i\ell}) f'_0, \quad (9)$$

where s_i (v_j) are the intraband spin (velocity) matrix elements for $|u_n(\mathbf{k})\rangle$.

Equation (9) is the key result of this Letter. First, it applies to both insulators and metals. Compared with Eq. (5), Eq. (9) contains an additional Fermi surface term

TABLE I. Constraints on the intrinsic nonlinear spin response tensor elements from magnetic point group symmetries. “✓” (“×”) means that the element is symmetry allowed (forbidden). Here, we choose to symmetrize the second and the third tensor indices, by defining $\alpha_{i(xy)} \equiv \frac{1}{2}(\alpha_{ixy} + \alpha_{iyx})$, and we omit the superscript “int” in the table. Symmetry operations \mathcal{T} , \mathcal{PT} , $C_3\mathcal{T}$, and $S_6\mathcal{T}$ forbid all the elements here; they are not listed.

	\mathcal{P}	C_2^z	C_3^z, S_6^z	$C_{4,6}^z, S_4^z$	$C_{2,4,6}^x, S_4^x$	C_3^x, S_6^x	σ_z	σ_x	$C_2^z\mathcal{T}$	$C_4^z\mathcal{T}, S_4^z\mathcal{T}$	$C_6^z\mathcal{T}$	$C_2^x\mathcal{T}$	$C_4^x\mathcal{T}, S_4^x\mathcal{T}$	$C_6^x\mathcal{T}$	$\sigma_z\mathcal{T}$	$\sigma_x\mathcal{T}$
α_{xxx}	✓	×	$-\alpha_{xyy}$	×	✓	×	×	✓	×	$-\alpha_{xyy}$	×	×	×	×	✓	×
$\alpha_{x(xy)}$	✓	×	α_{yxx}	×	×	×	×	✓	×	α_{yxx}	✓	×	×	×	✓	✓
α_{xyy}	✓	×	✓	×	✓	×	×	✓	×	✓	×	×	✓	×	✓	×
α_{yxx}	✓	×	✓	×	×	×	×	✓	×	✓	✓	×	×	×	✓	✓
$\alpha_{y(xy)}$	✓	×	α_{xyy}	×	✓	×	×	✓	×	α_{xyy}	×	×	✓	×	✓	×
α_{yyy}	✓	×	$-\alpha_{yxx}$	×	×	×	×	✓	×	$-\alpha_{yxx}$	✓	×	×	✓	✓	✓
α_{zxx}	✓	✓	✓	✓	×	×	✓	×	×	×	×	✓	×	×	×	✓
$\alpha_{z(xy)}$	✓	✓	×	×	✓	✓	✓	×	×	×	×	×	✓	×	×	×
α_{zyy}	✓	✓	α_{zxx}	α_{zxx}	×	✓	✓	×	×	$-\alpha_{zxx}$	×	✓	×	✓	×	✓

(the second term). For the case of an insulator, the Fermi surface term vanishes, and the result recovers [Eq. (5)], confirming the consistency between the two approaches. Second, $\alpha_{ij\ell}^{\text{int}}$ is suppressed by time reversal symmetry, and is nonzero only for magnetic systems. In Eq. (9), the effect of magnetic ordering on $\alpha_{ij\ell}^{\text{int}}$ is through the eigenstates $|u_n(\mathbf{k})\rangle$, which are for the magnetic band structure fully considering the ordering. Third, as an intrinsic contribution, Eq. (9) is a genuine material property, determined solely by the material’s band structure. We mention that the h derivative in the first term of Eq. (9) can be done straightforwardly to obtain an expression involving only the spin and velocity matrix elements [see Eq. (S6) in the Supplemental Material [38]] of the band structure. Hence, the response can be readily evaluated in first-principles calculations.

Symmetry property.—The intrinsic nonlinear response $\alpha_{ij\ell}^{\text{int}}$ is allowed by the inversion symmetry. Meanwhile, other magnetic crystalline symmetries also put constraints on the form of $\alpha_{ij\ell}^{\text{int}}$, which we analyze here.

Since spin is a time reversal (\mathcal{T}) odd pseudovector and the electric field is a \mathcal{T} even vector, $\alpha_{ij\ell}^{\text{int}}$ transforms as a third-rank \mathcal{T} odd pseudotensor, which respects

$$\alpha_{i'j'\ell'}^{\text{int}} = \eta_T \det(O) O_{i'i} O_{j'j} O_{\ell'\ell} \alpha_{ij\ell}^{\text{int}}. \quad (10)$$

Here O is a point group operation, and the factor $\eta_T = \pm 1$ is connected with the character of $\alpha_{ij\ell}^{\text{int}}$ being \mathcal{T} odd: $\eta_T = -1$ for primed operations, i.e., the magnetic symmetry operations of the form $R\mathcal{T}$ with R a spatial operation, and $\eta_T = +1$ for nonprimed operations.

Assuming the applied electric field is in the $x - y$ plane, the constraints from different magnetic point group symmetries are listed in Table I. This offers useful guidance for analyzing the nonlinear spin response for a particular material. For example, consider a ferromagnet which preserves the inversion and a horizontal mirror σ_z , with

its magnetization along the z direction. Then, Table I tells us that for an applied in-plane electric field, the generated nonlinear spin polarization must be out of plane, along the magnetization direction. For the purpose of electric control of the magnetization direction, one may want the induced spin polarization to have a component normal to the magnetization, such that it can generate a torque, which means that the desired material should not have a horizontal mirror plane.

A material example.—We demonstrate the implementation of our theory in first-principle calculations to study a concrete material. Guided by the symmetry constraints in Table I, we consider the effect in single-layer MnBi_2Te_4 . MnBi_2Te_4 in its bulk and two-dimensional few-layer forms has attracted considerable research interest recently, because it provides a platform for realizing various types of topological states [52–62]. Our focus here is on its single layer, which has been successfully fabricated in experiment, either by exfoliation from the bulk or by molecular beam epitaxy growth [52,54,62]. Its crystal structure is shown in Figs. 1(a) and 1(b), characterized by a hexagonal lattice with the space group $P\bar{3}m1$ (No. 164) and the point group D_{3d} . It consists of seven atomic layers, stacked in the sequence of Te-Bi-Te-Mn-Te-Bi-Te. Previous works have established that the ground state of single-layer MnBi_2Te_4 is a topologically trivial ferromagnetic semiconductor with out-of-plane magnetization [53], and the Curie temperature is about 15.2 K [63]. The ground-state magnetic configuration possesses a magnetic point group of $\bar{3}m'$. Importantly, the inversion symmetry is preserved, which forbids the linear spin generation. According to Table I, the symmetries C_3^z , $C_2^x\mathcal{T}$, and $\sigma_x\mathcal{T}$ further enforce the following relations among elements of the nonlinear response tensor: $\alpha_{yxx}^{\text{int}} = \alpha_{xxy}^{\text{int}} = \alpha_{xyx}^{\text{int}} = -\alpha_{yyy}^{\text{int}}$ and $\alpha_{zxx}^{\text{int}} = \alpha_{zyy}^{\text{int}}$.

It follows that the nonlinear spin response of single-layer MnBi_2Te_4 is specified by two independent elements $\alpha_{yxx}^{\text{int}}$ and $\alpha_{zxx}^{\text{int}}$. To see this more clearly, we assume the electric field is along an in-plane direction that makes an angle θ

from the x axis, i.e., $\mathbf{E} = E(\cos \theta, \sin \theta, 0)$. The induced out-of-plane nonlinear spin polarization takes the form of $\delta s_z = \alpha_{zx}^{\text{int}} E^2$, which is independent of the field direction. Meanwhile, the induced in-plane spin polarization can be expressed as [38]

$$(\delta s_{\parallel}, \delta s_{\perp}) = \alpha_{yxx}^{\text{int}} (\sin 3\theta, \cos 3\theta) E^2, \quad (11)$$

where δs_{\parallel} and δs_{\perp} denote the components parallel and perpendicular to the electric field, respectively. It is interesting to note that the in-plane nonlinear response is characterized by a single element $\alpha_{yxx}^{\text{int}}$, and exhibits an angular dependence with $2\pi/3$ periodicity. The induced in-plane spin polarization is perpendicular to the equilibrium magnetization; hence it can be more readily detected in experiment and can exert spin torques on the magnetization [7]. We shall focus on $\alpha_{yxx}^{\text{int}}$ in the following discussion.

Next, we evaluate the intrinsic nonlinear spin response tensor by combining our theory with first-principles calculations (calculation details are presented in the Supplemental Material [38]). Figure 1(d) shows the calculated band structure of single-layer MnBi_2Te_4 . The system

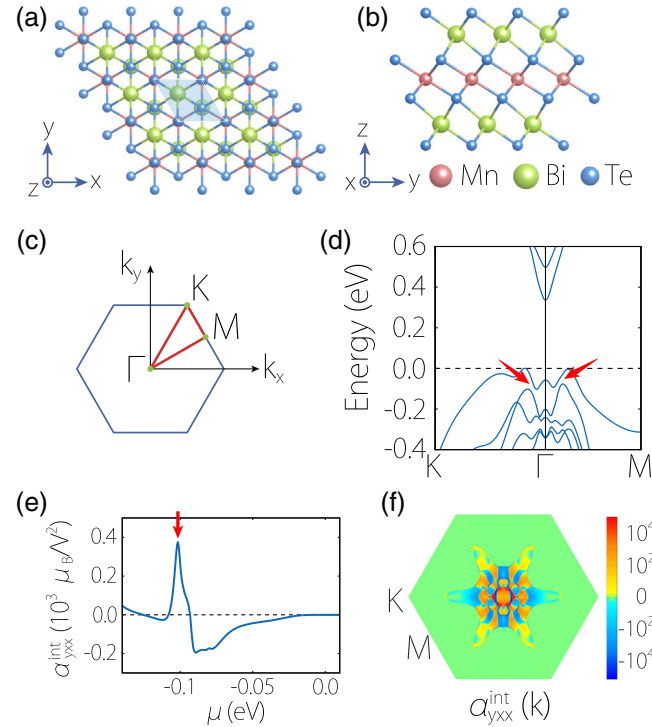


FIG. 1. (a) Top and (b) side views of the lattice structure of single-layer MnBi_2Te_4 . (c) shows the Brillouin zone. (d) Calculated band structure (spin-orbit coupling is included). (e) Calculated nonlinear response tensor element $\alpha_{yxx}^{\text{int}}$ versus the chemical potential μ . (f) Distribution of $\alpha_{yxx}^{\text{int}}(\mathbf{k})$, i.e., the integrand of Eq. (9), in the momentum space for chemical potential at -0.1 eV [marked by the red arrow in (e)]. The unit is μ_B/V^2 per unit cell. In the calculation, the temperature is set to 8 K.

is a ferromagnetic semiconductor with an indirect gap of 337 meV, which agrees with previous result [53]. We have computed all the relevant tensor elements for $\alpha_{ij\ell}^{\text{int}}$ according to Eq. (9). The results comply with the symmetry constraints discussed above. As mentioned, we focus on the in-plane spin generation. The obtained $\alpha_{yxx}^{\text{int}}$ as a function of the chemical potential is plotted in Fig. 1(e). Within the large band gap, the value of $\alpha_{yxx}^{\text{int}}$ is small but nonzero. It is $\sim -0.036 \mu_B/\text{V}^2$, with μ_B as the Bohr magneton. The response is greatly enhanced by hole doping, especially when the chemical potential is shifted to band near degeneracy regions in the valence bands. Because Eqs. (3) and (7) show that the BCPs are generally large around band near degeneracies, it follows that $\alpha_{ij\ell}^{\text{int}}$, involving integrals of BCPs, must also be peaked when the chemical potential is aligned in such regions. Particularly, in Fig. 1(e), a peak of $375 \mu_B/\text{V}^2$ is observed around -0.1 eV, which can be attributed to the small gap regions marked by red arrows in Fig. 1(d). At the peak, we plot the k -resolved contribution to $\alpha_{yxx}^{\text{int}}$, i.e., the integrand in Eq. (9), in Fig. 1(f). The distribution shows an even function with respect to both the x and y axis, and is peaked around the small-gap regions.

Consider the hole doped case with the chemical potential ~ -0.1 eV and a moderate applied electric field of 1 kV/cm which is readily achievable in experiment [5]. The induced in-plane spin magnetization in single-layer MnBi_2Te_4 is $\sim 0.4 \times 10^{-5} \mu_B/\text{nm}^3$. Note that this value is comparable to the linear spin generation (less than $10^{-6} \mu_B/\text{nm}^3$) already measured in noncentrosymmetric ferromagnetic systems [5,6,11]. Hence, it should be detectable in experiment, e.g., by magneto-optical Kerr spectroscopy or anisotropic magnetoresistance. Moreover, since it was shown that induced transverse spin polarization of this magnitude is able to drive magnetization dynamics [5,6,11], we expect our proposed effect can produce sizable spin torques [7] and useful for spintronics applications. Finally, we note that the response can be further enhanced by more than 1 order of magnitude ($\sim 10^{-4} \mu_B/\text{nm}^3$) at higher doping levels ~ -0.16 eV [38]. In practice, the doping can be readily controlled for 2D materials by gating [64,65].

Discussion.—We have proposed the nonlinear electric spin generation effect, which is the leading response in centrosymmetric magnets. The focus here is on the intrinsic contribution, which can be quantitatively evaluated for each material. For insulators, it captures the total response, whereas for metals, there are additional extrinsic contributions from scattering processes at the Fermi surface. The extrinsic contributions are in principle also contained in Eq. (8) and can be extracted by solving the distribution function, e.g., from the Boltzmann equation. As resulting from carrier scattering, they usually involve the relaxation time parameter. A systematic study of the extrinsic effect is

an interesting topic to explore in future works. In practice, the intrinsic and extrinsic parts can be separated by their different scaling with the relaxation time and distinct symmetry constraints, analogous to cases in nonlinear charge transport [33–35,66–68].

We have demonstrated the implementation of our theory with first-principles calculations. This will guide the experimental study and facilitate the search for nonlinear spintronic material platforms. The effect should exist in conventional ferromagnets like fcc Ni and Co, which preserve the inversion symmetry. We also expect the recently fabricated 2D centrosymmetric magnets, such as 1T-MnSe₂ [69], CrI₃ [70,71], and 1T-VSe₂ [72], would be good candidates, due to their great tunability. Moreover, the effect also exists in centrosymmetric antiferromagnets. For fully compensated antiferromagnets, such as MnF₂ and RuO₂ [38,73,74], the spin polarization will be induced on a zero magnetization background, which could be readily detected in experiment.

Finally, we note that intrinsic second-order responses of other observables that correspond to local operators, such as charge current [29], spin current, and pseudospin, admit a formulation similar to the theory developed here (see the Supplemental Material [38]). Therefore, our finding not only serves as a building block for the emerging field of nonlinear spintronics, but also forms the basis for exploring rich intrinsic nonlinear response properties of Bloch electrons.

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