Spin Current and Magnetoelectric Effect in Noncollinear Magnets

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A new mechanism of the magnetoelectric effect based on the spin supercurrent is theoretically presented in terms of a microscopic electronic model for noncollinear magnets. The electric polarization $\vec{P}_{ij}$ produced between the two magnetic moments $\vec{S}_i$ and $\vec{S}_j$ is given by $\vec{P} \propto \vec{e}_{ij} \times (\vec{S}_i \times \vec{S}_j)$ with $\vec{e}_{ij}$ being the unit vector connecting the sites i and j. Applications to the spiral spin structure and the gauge theoretical interpretation are discussed.

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The interplay between the magnetism and ferroelectricity is an issue as old as the first prediction of the magnetoelectric (ME) effect by Curie [1]. The ME effect is a phenomenon in which the magnetization is induced by the electric field or the electric polarization is induced by the magnetic field. The phenomenological theory of the ME effect was developed by Landau [2] and Dzyaloshinskii [3]. There the symmetry consideration is essential to classify all the possible ME tensors via the magnetic point group. Especially the time-reversal (T) and spatial inversion (I) are the key symmetries to the ME effect. For example, the linear ME effect corresponding to the term $\alpha_{ij}M_iP_j$ ($\vec{P}$: polarization, $\vec{M}$: magnetization) is allowed only when both T and I symmetries are broken. However, on the other hand, the microscopic quantum theory of ME effect has not yet been fully developed, although several scenarios for particular materials have been proposed based on the effective spin Hamiltonian. For Cr_2O_3, the change of the anisotropy energy, exchange, and g value due to the electric field have been proposed for the origin of the parallel ME effect [4,5]. For a spiral spin magnet $ZnCr_2Se_4$, the electric field induced Dzyaloshinskii-Moriya (DM) interaction [6,7] has been proposed as a mechanism for the transverse ME effect [8]. Another well-studied ME material is $(Ga, Fe)O_3$, where the ME coefficient is an order of magnitude larger than in Cr_2O_3 [9]. However, a fully microscopic theory based on the electronic states is still missing. In this Letter, we develop a theory of ME effect starting from the electronic Hamiltonian taking into account the spin-orbit interaction, and show that ME effect and spin current are directly related in noncollinear spin structures such as the spiral state [10,11]. The spin current $\vec{j}_s$ has attracted revived interest recently in the context of spintronics in semiconductors. In contrast to the charge current, it is T even since the spin polarization is also reversed together with the direction. Therefore, from the symmetry point of view, $\vec{j}_s$ belongs to the same class as the electric polarization $\vec{P}$, and it is natural to expect the coupling between these two. In fact, the electric field induced dissipationless spin current driven by the Berry phase curvature has been proposed for the semiconductors [12]. In magnets, the spin current is analogous to the superfluid current, i.e., spin supercurrent, associated with the spin rigidity [13]. The idea is that the spin supercurrent is induced between the two spins with generic nonparallel configurations, which induces the electric polarization. We will describe its gauge theoretical interpretation from the viewpoint of the dual DM [6,7] and/or Aharonov-Casher (AC) effect [14]. Applications to the spiral magnetic structure are also discussed.

We start with the following electronic model with the electron energy levels in the ligand field of $3d$-transition metal. In the octahedral ligand field, the d orbitals are split into e_g and t_{2g} orbitals. The t_{2g} orbitals, i.e., d_{xy}, d_{yz}, and d_{zx}, have energy lower than e_g orbitals. If we take account of the spin degree of freedom, there is sixfold degeneracy in t_{2g} energy level. Because of the on-site spin-orbit interaction, however, this degeneracy is lifted and we have two groups of spin-orbit coupled states, labeled Γ_7 and Γ_8. The twofold degenerate states, i.e., Γ_7, are given by

$$|a\rangle = \frac{1}{\sqrt{3}}(|d_{xy,\uparrow}\rangle + |d_{yz,\downarrow}\rangle + i|d_{zx,\downarrow}\rangle),$$
$$|b\rangle = \frac{1}{\sqrt{3}}(|d_{xy,\downarrow}\rangle - |d_{yz,\uparrow}\rangle + i|d_{zx,\uparrow}\rangle), \quad (1)$$

respectively, where the quantization axis of spin is taken to be the z axis. For the sake of simplicity, we consider only the above two states. However, our method is valid for more general cases and one can easily generalize it to any other spin-orbit coupled situation.

We consider the case where the inversion symmetry exists at the middle point of the two magnetic ions, i.e., there is no DM interaction, and the generic noncollinear magnetic ordering is assumed to be realized by the competing exchange interactions J's. We focus on the ordered ground state properties, and hence the mean field treatment gives a fairly good description of the system. We consider the Hamiltonian: $H_U = -U\Sigma_j \vec{e}_j \cdot \vec{S}_j$, where U is energy

of Coulomb repulsion (we take the unit where $\hbar = 1$ hereafter). Here the magnetic moment at jth site is described by the unit vector $\vec{e}_j = (\cos\phi_j \sin\theta_j, \sin\phi_j \sin\theta_j, \cos\theta_j)$. S_j is the electronic spin operator at site j. For each site j, we restrict the Hilbert space to the two-dimensional one spanned by the above two states, and the effective Hamiltonian is reduced to the 2×2 matrix

$$-\frac{U}{3}\begin{bmatrix} -\cos\theta & \sin\theta e^{-i\phi} \\ \sin\theta e^{i\phi} & \cos\theta \end{bmatrix}. \tag{2}$$

We diagonalize this Hamiltonian matrix to obtain eigenstates $|P\rangle$, $|AP\rangle$ as

$$\begin{aligned} |P\rangle &= \sin\frac{\theta}{2}|a\rangle + e^{i\phi}\cos\frac{\theta}{2}|b\rangle, \\ |AP\rangle &= \cos\frac{\theta}{2}|a\rangle - e^{i\phi}\sin\frac{\theta}{2}|b\rangle. \end{aligned} \tag{3}$$

Here $|P\rangle$ and $|AP\rangle$ mean the spin state parallel and antiparallel to the unit vector $\vec{e}$, and the corresponding eigenvalues are $-\frac{U}{3}$ and $+\frac{U}{3}$, respectively. For convenience, we define the coefficients $A^{i\sigma}$ and $B^{i\sigma}$ and abbreviate the above two states as $|P\rangle = \Sigma_{i\sigma}A^{i\sigma}|d_{i\sigma}\rangle$, and $|AP\rangle = \Sigma_{i\sigma}B^{i\sigma}|d_{i\sigma}\rangle$, where $i = xy, yz, zx$, $\sigma = \uparrow, \downarrow$.

For the moment, we focus on the three atom model as shown in Fig. 1, which represents the bond between the two transition metal ions M1, M2 through the oxygen atom O. We take the hole picture below, where the oxygen orbitals are empty. We assume the generic case of $\vec{e}_1$ and $\vec{e}_2$ including the noncollinear configuration. Each site has two states, i.e., $|P\rangle$ and $|AP\rangle$, mentioned above. So we define $|P\rangle_j$ and $|AP\rangle_j$ $(j = 1, 2)$ corresponding to the magnetic order on each site. Because of the existence of the oxygen atom, there are hopping processes between the M site and the O site. The Hamiltonian for the electron transfer is given by [15] $H_t = H_t^{1-m} + H_t^{2-m} + \text{H.c.}$, with $H_t^{1-m} = +V\Sigma_\sigma(p_{y,\sigma}^\dagger d_{xy,\sigma}^{(1)} + p_{z,\sigma}^\dagger d_{zx,\sigma}^{(1)})$, $H_t^{2-m} = -V\Sigma_\sigma(p_{y,\sigma}^\dagger d_{xy,\sigma}^{(2)} + p_{z,\sigma}^\dagger d_{zx,\sigma}^{(2)})$, where $V(>0)$ is the transfer integral and the superscript j denotes the corresponding site number. Our total Hamiltonian is $H_U + H_t$ and we treat H_t perturbatively. The eight bases needed are $|P\rangle_j$, $|AP\rangle_j$, $(j = 1, 2)$, and $p_{i,\sigma}$, $(i = y, z, \sigma = \uparrow, \downarrow)$. Using the second-order perturbation theory, the four lowest lying states and corresponding perturbed energies are obtained as follows:

$$\begin{aligned} |1\rangle = &\frac{\alpha e^{-i\Delta\phi/2}}{\sqrt{2}|\alpha|}\left(|P\rangle_1 + \frac{V}{\Delta}\sum_\sigma(A_{(1)}^{xy,\sigma}|p_{y,\sigma}\rangle + A_{(1)}^{zx,\sigma}|p_{z,\sigma}\rangle)\right) \\ &+ \frac{1}{\sqrt{2}}\left(|P\rangle_2 - \frac{V}{\Delta}\sum_\sigma(A_{(2)}^{xy,\sigma}|p_{y,\sigma}\rangle + A_{(2)}^{zx,\sigma}|p_{z,\sigma}\rangle)\right), \end{aligned}$$

with $E_1 = -\frac{4}{3}\frac{V^2}{\Delta}(1 + |\alpha|)$,

$$\begin{aligned} |2\rangle = &-\frac{\alpha e^{-i\Delta\phi/2}}{\sqrt{2}|\alpha|}\left(|P\rangle_1 + \frac{V}{\Delta}\sum_\sigma(A_{(1)}^{xy,\sigma}|p_{y,\sigma}\rangle + A_{(1)}^{zx,\sigma}|p_{z,\sigma}\rangle)\right) \\ &+ \frac{1}{\sqrt{2}}\left(|P\rangle_2 - \frac{V}{\Delta}\sum_\sigma(A_{(2)}^{xy,\sigma}|p_{y,\sigma}\rangle + A_{(2)}^{zx,\sigma}|p_{z,\sigma}\rangle)\right), \end{aligned}$$

with $E_2 = -\frac{4}{3}\frac{V^2}{\Delta}(1 - |\alpha|)$, and two other higher energy states. Here $\Delta(>0)$ is the energy difference between the p orbitals and $|P\rangle_j$, $\Delta\phi = \phi_1 - \phi_2$, and we have introduced the complex number $\alpha = \cos\frac{\theta_1}{2}\cos\frac{\theta_2}{2}e^{-i\Delta\phi/2} + \sin\frac{\theta_1}{2} \times \sin\frac{\theta_2}{2}e^{+i\Delta\phi/2}$. Before calculating the expected value of the polarization, it is useful to note that only the following matrix elements are nonzero from the shapes of d and p orbitals;

$$I = \int d^3\vec{r}\, d_{yz}^{(j)}(\vec{r}) y p_z(\vec{r}), \qquad (j = 1, 2),$$

and its cyclic permutations. The integral I is approximately estimated as $I \cong \frac{16}{27}{Z_O}^{5/2}{Z_M}^{7/2}(\frac{Z_O}{2} + \frac{Z_M}{3})^{-6}a_0$, where a_0 is Bohr radius and Z_O/Z_M is the atomic number of O/M. We can easily check the above results by expanding wavefunctions in terms of lattice constant a. So let us now calculate the expected value of polarization in the following two cases.

Double-exchange interaction [16,17].—First, we consider the situation where only one hole is present. In this case, this hole is put into the ground state, determined by the above second-order perturbation theory, and the expected value of polarization, $\langle 1|er|1\rangle/\langle 1|1\rangle$, is given by

$$\vec{P} \cong -\frac{eV}{3\Delta}I\frac{\vec{e}_{12} \times (\vec{e}_1 \times \vec{e}_2)}{|\cos\frac{\theta_{12}}{2}|} \tag{4}$$

where $\vec{e}_{12}$ is the unit vector parallel to the direction of the bond from site M1 to site M2, and θ_{12} is the angle between the two vectors $\vec{e}_1$ and $\vec{e}_2$, i.e., $\vec{e}_1 \cdot \vec{e}_2 = \cos\theta_{12}$. In this case, the spin current is derived from the equation of motion of the electronic spin operator $\vec{S}_i$ [18];

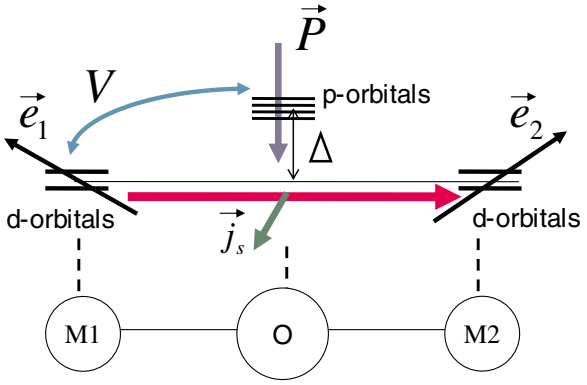

FIG. 1 (color online). The cluster model with two transition metal ions M1, M2 with the oxygen atom O between them. With the noncollinear spin directions $\vec{e}_1$ and $\vec{e}_2$, there arises the spin current $\vec{j}_s \propto \vec{e}_1 \times \vec{e}_2$ between M1 and M2. Here the direction of the vector $\vec{j}_s$ (denoted by the short arrow near the middle of the diagram) is that of the spin polarization carried by the spin current. The direction of the electric polarization $\vec{P}$ is given by $\vec{P} \propto \vec{e}_{12} \times \vec{j}_s$ where $\vec{e}_{12}$ is the unit vector connecting M1 and M2.

$$\vec{j}_s^{\,i} = \frac{i}{\hbar}\sum_{j(>i)}\sum_{\alpha\beta}(t_{ji}d_{j\alpha}^{\dagger}\vec{\sigma}_{\alpha\beta}d_{i\beta} - t_{ij}d_{i\alpha}^{\dagger}\vec{\sigma}_{\alpha\beta}d_{j\beta}) \quad (5)$$

where $\alpha, \beta = a, b$ and $t_{ij} = V^2/\Delta$ is the effective transfer integral between the d orbitals after integrating over the p orbital. Following the above definition, spin current $\vec{j}_s$ is approximately given by $\vec{j}_s \sim (V^2/\Delta)a_0(\vec{e}_1 \times \vec{e}_2)/\cos(\theta_{12}/2)$, and Eq. (4) can be rewritten as $\vec{P} \sim (e/V)\vec{e}_{12} \times \vec{j}_s$. Therefore the spin current is essential to the electric polarization.

Superexchange interaction [19].—Next we consider the case of two holes. From a viewpoint of Hartree-Fock approximation, two holes are put into the ground state $|1\rangle$ and the second low-lying state $|2\rangle$, and the expected value of the polarization is given by

$$\vec{P} \cong -\frac{4e}{9}\left(\frac{V}{\Delta}\right)^3 I\vec{e}_{12} \times (\vec{e}_1 \times \vec{e}_2). \quad (6)$$

In this case, the dominant term comes from the difference of the normalization factor between the two perturbed states, i.e., $\langle 1|1\rangle^{-1} \cong 1 - \frac{2}{3}(\frac{V}{\Delta})^2(1 - |\alpha|)$ and $\langle 2|2\rangle^{-1} \cong 1 - \frac{2}{3}(\frac{V}{\Delta})^2(1 + |\alpha|)$. By using the superexchange interaction J, i.e., $J \cong V^4/\Delta^3$, (6) can be rewritten as $\vec{P} \sim -e(J/V)I\vec{e}_{12} \times (\vec{e}_1 \times \vec{e}_2)$. Again this equation can be interpreted in terms of the spin current $\vec{P} \sim (e/V)\vec{e}_{12} \times \vec{j}_s$ since the spin current $\vec{j}_s \sim J\vec{e}_1 \times \vec{e}_2$ [see the discussion below Eq. (7)]. Let us give here a rough estimate on the order of magnitude for $\vec{P}$. With the lattice constant $a =$ 5 Å, Eq. (6) gives $\vec{P} \sim 10^4 \times (V/\Delta)^3\ \mu\mathrm{C/m}^2$, which is comparable to that obtained in (Ga, Fe)O_3 at $H = 10$ T [9].

For more general models, the magnitude of the electric polarization induced by the spin current depends on the details of the electronic level structure, and most likely is smaller than in the present case, especially for the e_g systems. Also the band structure should be considered for the crystal extending the cluster calculation. However the geometrical relation between the spin current and electric polarization is allowed by symmetry and remains unchanged, and the cluster results are semiquantitatively in agreement with those of the band picture, as will be discussed below.

Applications to the spiral magnets.—Now we turn to the discussion on realistic noncollinear magnets. One of the typical examples is the spiral structure where the direction of the spin rotates along the wavevector $\vec{q}$. Figure 2(a) shows the most generic spiral spin configuration where the spiral axis is along the x axis, the cone axis direction have the angle α measured from the z axis, and the cone angle is β. The angle γ_j for the jth spin direction is measured from the zx plane, and the undistorted spiral means $\gamma_j = qj + \gamma_0$. The spin at site j can be written as $\vec{S}_j = S(\cos\beta\sin\alpha + \sin\beta\cos\alpha\cos\gamma_j, \sin\beta\sin\gamma_j, \cos\beta\cos\alpha - \sin\beta\sin\alpha\cos\gamma_j)$. With this configuration one can easily calculate the spin current $\vec{j}_{sj+1/2}$ and resultant electric polarization $\vec{P}_{j+1/2}$ at each

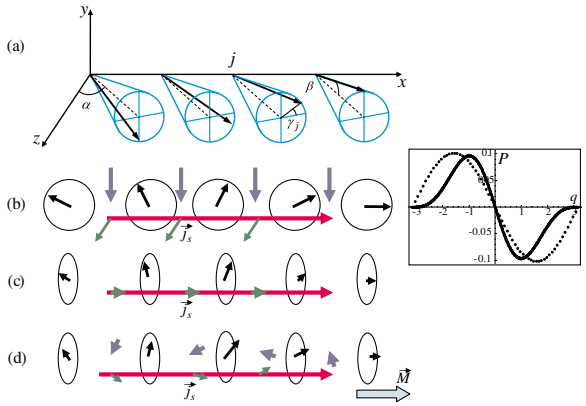

FIG. 2 (color online). (a) Generic spiral spin configuration. (b–d) Some of the specific configurations where the geometrical relation among spins (black arrows), spin current (gray arrows on or below the long right horizontal arrow), and electric polarization [gray arrows above the long right horizontal arrow in (b) and (d)] are shown. The inset shows the comparison of the polarization between the cluster (dotted line) and the periodic array (solid line) at half filling, i.e., superexchange interaction case with the spin configuration in (b).

link connecting j and $j+1$ as $(P_{j+1/2})_x = 0$, $(P_{j+1/2})_y \sim -\cos\beta\sin\beta\sin\alpha[\sin\gamma_{j+1} - \sin\gamma_j] - \sin^2\beta\cos\alpha \times \sin(\gamma_{j+1} - \gamma_j)$, $(P_{j+1/2})_z \sim \cos\beta\sin\beta[\cos\gamma_{j+1} - \cos\gamma_j]$. Therefore only the y component of the uniform electric polarization $\vec{P} = \Sigma_j\vec{P}_{j+1/2}$ is nonzero and is given by $P_y \propto \Sigma_j\sin^2\beta\cos\alpha\sin(\gamma_{j+1} - \gamma_j)$.

Figures 2(b)–2(d) show some typical cases and their spin current and electric polarization. When the spins are within the xy plane as in Fig. 2(b), i.e., $\alpha = 0$, $\beta = \pi/2$, the spin current $\vec{j}_s$ is along the z direction, and the electric polarization $\vec{P}$ is along the y direction for each site. Therefore the total uniform polarization is finite along the y direction. Note that even when the spiral wave number $\vec{q}$ is incommensurate with the lattice periodicity, the uniform polarization, i.e., the ferroelectricity, is realized. This is in sharp contrast to the usual improper ferroelectrics where the polarization is induced by the transition to the commensurate phase. In the inset, we show the results of the cluster and band model. In the latter case, we put the one-dimensional array of the d and p orbitals with the trivial extension of the transfer Hamiltonian H_t. The calculated polarizations are semiquantitatively in agreement with each other. When the spins are in the yz plane [Fig. 2(c)], i.e., $\alpha = \pi/2$, $\beta = \pi/2$, both $\vec{e}_{jj+1}$ and $\vec{j}_s$ are in the x direction and their vector product is zero. Therefore we do not expect any electric polarization induced in this case. Figure 2(d) shows the "conical" spin structure, where the finite x component of the spin is induced starting from the structure in Fig. 2(c), i.e., $\alpha = \pi/2$, $0 < \beta < \pi/2$. In this case, the finite S_x component at each site produces the rotating polarization with the same wave vector q as the spins, but these cancel out to zero with no uniform electric polarization formed.

Now consider the effect of the external electric field E_y along the y direction on the generic spiral configuration. This field induces uniform magnetization (per site), estimated as $\vec{m}_x \propto E_y \sin^2\beta\cos\beta\sin\alpha\cos\alpha\sin q$, and $\vec{m}_z \propto E_y \sin^2\beta\cos\beta\sin q(a + b\cos^2\alpha)$ (a, b: constants). This dependence is consistent with the group theoretical consideration and the experiments on $ZnCr_2Se_4$ (Figures 6 and 7 of Ref. [8]).

Gauge interpretation.—We consider the following Heisenberg spin Hamiltonian for the superexchange case:

$$H_0 = -\sum_{\langle ij\rangle}\left[\frac{J_{ij}}{2}(S_i^+ S_j^- + S_i^- S_j^+) + J_{ij} S_i^z S_j^z\right]. \quad (7)$$

The spin current j_{ij}^s for the z component is defined so as to satisfy $\partial S_i^z/\partial t = (1/i\hbar)[S_i^z, H] = -\Sigma_j j_{ji}^s$ and is given by $j_{ij}^s = iJ_{\perp ij}(S_i^+ S_j^- - S_i^- S_j^+) = J_{\perp ij} S^2 \sin(\theta_i - \theta_j)$ where the spin $(S_j^x, S_j^y) = S(\cos\theta_j, \sin\theta_j)$ is assumed to lie within the XY plane. Here JS^2 corresponds to the spin stiffness, i.e., rigidity. The above equation is analogous to the Josephson equation for superconductors. To go further with this analogy, the next question is "what is the vector potential $\vec{A}_s$ coupled to the spin supercurrent?." The answer to this question can be found in the AC effect [14] and DM interaction [6,7]. The conventional DM interaction [7] is given by $H_{DM} = \Sigma_{\langle ij\rangle}\vec{D}_{ij}\cdot(\vec{S}_i \times \vec{S}_j)$. When the DM vector $\vec{D}_{ij} = D_{ij}\hat{e}_z$, the total Hamiltonian $H_{\text{total}} = H_0 + H_{DM}$ with H_{XY} is written as $H_{\text{total}} = -\Sigma_{\langle ij\rangle}[(\tilde{J}_{ij}/2)\times(e^{-iA_{ij}} S_i^+ S_j^- + e^{iA_{ij}} S_i^+ S_j^-) + J_{ij} S_i^z S_j^z]$ where $\tilde{J}_{ij} e^{iA_{ij}} = J_{ij} + iD_{ij}$. Correspondingly the spin supercurrent is written as $j_{ij}^s = \tilde{J}_{ij} S^2 \sin(\theta_i - \theta_j - A_{ij})$. Therefore the DM vector $\vec{D}$ acts as the vector potential or gauge field to the spin current. In contrast to our model with the frustrated J's, the spiral structure is realized by the DM interaction even without the frustration. In this case, the spin direction is determined so that the total energy is minimized, i.e., $\theta_i - \theta_j - A_s = 0$, and the spin supercurrent $j_{ij}^s \propto \sin(\theta_i - \theta_j - A_s)$ is zero. Therefore our mechanism does not apply in this case. It is well known that the DM interaction exists only when the inversion symmetry is broken at the middle point between the two spins. Therefore when the crystal structure has the inversion symmetry, the external electric field $\vec{E}$ induces the DM interaction. Namely $\vec{D}_{ij} \propto \vec{E} \times \vec{e}_{ij}$, with $\vec{e}_{ij}$ being the unit vector connecting the two sites i and j, which was suggested from symmetry consideration in Ref. [8]. This leads to the coupling between the spin current and the electric field as a "gauge field." Through the relation $\vec{P} = \partial H/\partial\vec{E}$, our calculation above corresponds to a microscopic derivation of this coupling without the symmetry breaking atomic displacement. This form is identical to the AC effect, where the Lorentz transformation of the electric field induces the magnetic field in the moving frame which interacts with the spin moment [20]. However the magnitude of the coupling constant for the AC effect is extremely small in vacuum since it contains the rest mass of the electron $mc^2 \cong 5 \times 10^5$ eV in the denominator. In solids, the electrons are trapped in the strong potential of the atoms with large momentum distribution leading to the enhanced spin-orbit interaction, and the gauge potential A_{ij} could be (a fraction) of the order of unity, e.g., $A_{ij} \sim 2\pi$ as seen above. It is noted here that the gauge field A_{ij} has no gauge degrees of freedom since $\vec{E}$ is the physical field.

In conclusion, we argue, based on the microscopic electronic Hamiltonian, that spin current $\vec{j}_s$ in noncollinear magnets leads to the electric polarization $\vec{P} \propto \vec{e}_{ij} \times \vec{j}_s$ with $\vec{j}_s \propto \vec{S}_i \times \vec{S}_j$, (Eqs. (4) and (6)). By this mechanism we predict a novel ME effect based on spin current induction of electric polarization.

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