Rectification of Spin Current in Inversion-Asymmetric Magnets with Linearly-Polarized Electromagnetic Waves

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We theoretically propose a method of rectifying spin current with a linearly-polarized electromagnetic wave in inversion-asymmetric magnetic insulators. To demonstrate the proposal, we consider quantum spin chains as a simple example; these models are mapped to fermion (spinon) models via Jordan-Wigner transformation. Using a nonlinear response theory, we find that a dc spin current is generated by the linearly-polarized waves. The spin current shows rich anisotropic behavior depending on the direction of the electromagnetic wave. This is a manifestation of the rich interplay between spins and the waves; inverse Dzyaloshinskii-Moriya, Zeeman, and magnetostriction couplings lead to different behaviors of the spin current. The resultant spin current is insensitive to the relaxation time of spinons, a property of which potentially benefits a long-distance propagation of the spin current. An estimate of the required electromagnetic wave is given.

Introduction

Manipulation of magnetic states and spin current is a key subject in spintronics [1]. In conductive materials, the charge current is often used for such purposes; magnetic domain walls are moved by spin-transfer effect [2], and spin Hall effects are used to generate spin current [3-6]. The concept of spintronics is also applied to magnetic insulators. They have several advantages over the metallic materials: Magnetic excitations typically have longer life time and no ohmic loss. In these magnets, the electromagnetic wave is a “utility tool”. Recent studies demonstrate that magnetic states and excitations can be controlled by electromagnetic waves. For instance, laser control of magnetizations [7-12], magnetic interactions [13], and magnetic textures [14-19], spin-wave propagation by focused light [20, 21], etc. have been extensively studied both experimentally and theoretically. These studies demonstrated that the electromagnetic wave has a high potentiality of controlling the magnetic states and opened a subfield utilizing lights, called opto-spintronics [9-22].

In contrast, the manipulation of the spin current carried by magnetic excitations is limited to ferromagnets; spin pumping with the electromagnetic wave is often used to generate the spin current [23-25]. On the other hand, other magnetic states (antiferromagnetic, spiral, spin liquid states, etc.) potentially have different advantages over ferromagnets. Therefore, a method for the generation and manipulation of spin current in these materials opens up an interesting possibility for spintronics. For this purpose, the usage of electromagnetic waves is desirable because of the highly precise and ultra-fast control.

A main issue, however, lies in moving the magnetic excitations using the electromagnetic field; the magnetic excitations do not accelerate/drift by the electromagnetic field because they are chargeless. This problem is potentially solved by utilizing the nonlinear response of magnetic insulators [Fig. 1(a,b)]. In the nonlinear optics of noncentrosymmetric electron systems [26-28], a non-trivial dynamics of electrons during the transition process induce a “shift” of the particle position [29, 32]. Recent experiments investigating this mechanism find the current propagates faster than the quasi-particle velocity [33, 34]. In addition, it is insensitive to the quasi-particle relaxation time;
this is a beneficial property considering the heating by the electromagnetic waves reduces the relaxation time. A spin current with such interesting properties is potentially possible if the shift mechanism of magnetic excitations is generated by the electromagnetic waves.

To investigate the control of spin current by the nonlinear response, we explore the generation of spin current by the shift mechanism in a quantum spin chain model [Fig. 1(a)]. We show that the spin current is indeed generated by simply applying a linearly-polarized electromagnetic wave if the system possesses one of the three kinds of spin-light couplings: inverse Dzyaloshinskii-Moriya (DM), Zeeman, and magnetostriction couplings. These couplings give rise to rich features in the frequency dependence and anisotropy. Interestingly, the spin current is generated by a different transition process from the electronic photogalvanic effect. The estimate of the magnitude of spin current shows our proposal gives an observable spin current with a reasonable strength of electromagnetic wave.

Noncentrosymmetric spin chains — An \( S = 1/2 \) spin chain with staggered exchange and the magnetic field is used to study the photovoltaic effect of spin current. The Hamiltonian reads

\[
H = \sum_i J(1 \mp (-1)^i \delta)(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) - \sum_i (h + (-1)^i h_s) S_i^z.
\]

(1)

Here, \( S_i^{x,y,z} \) are \( S = 1/2 \) spin operators on site \( i \), \( J \) is the exchange coupling whose energy scale is usually in gigahertz (GHz) or terahertz (THz) regime, \( h \) is the uniform magnetic field along \( z \) axis, and \( h_s \) is the staggered magnetic field. This model has a wide range of applications. An obvious application is to the one-dimensional (1D) dimerized XY spin chains with two alternating ions [Fig. 1(a)]. In this case, the staggered magnetic field \( h_s \) appears as a consequence of different \( g \) factors for the odd- and even-site spins [35–40]. The model can also be viewed as the effective model for a Néel ordered Ising-like spin chain [41–43] at zero temperature \( T = 0 \) under a staggered magnetic field, in which the Ising interaction \( J \sum_{i} S_{i}^{z} S_{i+1}^{z} \) is treated via the mean-field approximation \( \langle S_{i}^{z} \rangle = \langle S_{i}^{+} S_{i}^{-} \rangle = \langle S_{i}^{-} S_{i}^{+} \rangle \). For the Néel ordered state, the field \( (-1)^i h_s \) is the sum of the external staggered field and the mean field \( J \langle S_{i}^{z} \rangle = (-1)^i J M_s \) (\( M_s \) is the staggered magnetization). Furthermore, Eq. (1) can also be applied to three-dimensional antiferromagnets of weakly coupled spin chains under a staggered field [Fig. 1(b)].

Treating the inter-chain coupling by a mean-field theory [44–47] gives an effective one-dimensional (1D) model, Eq. (1). Namely, in this system, the staggered field \( h_s \) is renormalized by the inter-chain Néel order. Note that the dimerization parameter \( \delta \) and the staggered field \( h_s \) break site-center and bond-center inversion symmetries, respectively. Such a noncentrosymmetric nature is necessary for a photogalvanic effect.

The spin model in Eq. (1) is mapped to a fermion model using Jordan-Wigner (JW) transformation [48–50]. By introducing fermion operators \( c_i \equiv e^{-i\pi \sum_{j=1}^{i-1} S_j^y} S_i^- \) and \( \bar{c}_i \equiv S_i^+ e^{i\pi \sum_{j=1}^{i-1} S_j^y} \), Eq. (1) is fermionized as 

\[
H = \sum_i J(1 + (-1)^i \delta) \frac{1}{2} (c_i^{\dagger} c_i + c_i c_i^{\dagger}) + (h + (-1)^i h_s) n_i. \tag{2}
\]

Here, \( S_i^\pm \equiv S_i^x \pm i S_i^y \) are the ladder operators and \( n_i \equiv c_i^{\dagger} c_i \) is the number operator for the fermions at \( i \)th site. Figures 1(c) and 1(d) show the band structure of the JW fermions. The model has a band gap \( \Delta_{\pm} = 2\sqrt{J^2 + h_s^2} \) for \( |\delta| < 1 \) [Fig. 1(d)], while the gap is \( \Delta_0 = 2\sqrt{J^2 + h_s^2} \) if \( |\delta| > 1 \). The model is gapless only if \( h_s = 0 \) [Fig. 1(c)]. Therefore, the ground state is robust against a magnetic field as long as \( h < \Delta/2 \), where \( \Delta = \min(\Delta_0, \Delta_{\pm}) \). We focus on the weak \( \delta \) region of this model in the rest of this work.

The spin current operator for \( S^z \) is defined from the continuity equation. The current density operator reads

\[
J_{sc} \equiv \frac{1}{L} \sum_i J(1 + (-1)^i \delta)(S_{i+1}^x S_i^y - S_i^x S_{i+1}^y). \tag{3}
\]

(3)

where \( L \) is the number of sites; here, we set the Planck constant \( \hbar = 1 \).

Inverse DM coupling — External electromagnetic waves couple to spins in several different forms. First, we consider the coupling of the electric field to the electric dipole induced by the inverse DM mechanism [51–55]:

\[
H_{IDM} = E_y(t) \sum_i (p + (-1)^i p_s) (S_i \times S_{i+1})^z. \tag{4}
\]

(4)

Here, the chain is along the \( x \) axis, \( p \pm p_s \) is the coefficient for the ferroelectric polarization of odd and even bonds, and \( E_y(t) = E_y \cos(\omega t) \) is the oscillating electric field along the \( y \) axis with frequency \( \omega \) (typically, GHz or THz). Note that at a special point \( p_s/p = \delta \), the term \( H_{IDM} \) is analogous to the linear-order coupling of the electrons to the vector potential. We will comment on this case later.

The spin current conductivity is calculated using a quadratic response formula similar to that for photovoltaic effects [56]. The formula reads

\[
\sigma(\omega) = \sum_{\alpha,\beta,\gamma} \int \frac{dk}{2\pi} \frac{1}{\omega - \varepsilon_{\alpha}(k) - \varepsilon_{\beta}(k) + i/(2\tau)} \times \left[ \frac{B_{\alpha\beta}(k)J_{\alpha\beta}(k)}{\varepsilon_{\alpha}(k) - \varepsilon_{\gamma}(k) - i/(2\tau)} - \frac{J_{\beta\gamma}(k)B_{\beta\gamma}(k)}{\varepsilon_{\gamma}(k) - \varepsilon_{\beta}(k) - i/(2\tau)} \right]. \tag{5}
\]

(5)

where \( \varepsilon_{\alpha}(k) \) is the eigenenergy of an \( \alpha \)-band state with momentum \( k \) and \( f_\alpha(k) \equiv (1 + e^{\varepsilon_{\alpha}(k)/k_B T})^{-1} \) is the fermion distribution for \( |\alpha k\rangle \). \( \tau \) is the relaxation time of JW fermions, \( J_{\alpha\beta}(k) \equiv \langle \alpha k| J_{sc} |\beta k\rangle \), and \( B_{\alpha\beta}(k) \equiv \langle \alpha k| H_{IDM} |\beta k\rangle \). Hereafter, we will mainly consider the \( T = 0 \) case of the model in Eq. (2). The conduction and
valence bands [Fig. 1(c) and (d)] respectively correspond to \( \alpha = + \) and \(-\). We focus on the real part of \( \sigma^{(2)}(\omega) \) because only the real part contributes to the spin current. With these simplifications, Eq. (5) becomes

\[
\Re[\sigma^{(2)}(\omega)] = \frac{1}{\pi} \Re \left\{ \sum_k \frac{B_{+-}(k)J_{+-}(k)}{\omega^2 - (\varepsilon_{+,k} - \varepsilon_{-,k} - i/(2\tau))^2} \right\},
\]  

(6)

provided that \( \varepsilon_{\pm,k} \) and \( |B_{+-}(k)|^2 \) are even with respect to \( k \).

Using Eq. (6), the nonlinear conductivity in the \( \tau \to \infty \) limit becomes

\[
\Re[\sigma^{(2)}(\omega)] = \text{sgn}(1 - \delta^2) \frac{h_s(p_s - p\delta)(p - p\delta)}{2\pi\omega^2 J^2(1 - \delta^2)^2} \times \sqrt{(\omega^2 - \Delta_{\alpha}^2)(\Delta_{\alpha}^2 - \omega^2)},
\]  

(7)

when \( \Delta \leq \omega \leq W \equiv \max(\Delta_0, \Delta_\pm) \). On the other hand, no spin current appears for a frequency \( \omega < \Delta \) or \( W < \omega \), which implies that an inter-band optical transition is necessary for the spin current. Figure 2(a) shows the result for \( J = 1, \delta = 1/3 \), and \( h_s = 1/10 \). The conductivity becomes zero when \( h_s = 0 \) or \( p_s = \delta = 0 \) and is proportional to \( h_s(p_s - p\delta) \). These features reflect the symmetry property of the conductance. The model becomes inversion symmetric when \( h_s = 0 \) or \( p_s = \delta = 0 \), and therefore, the conductivity vanishes. For the noncentrosymmetric chain, the inversion operation imposes following relations: \( \sigma^{(2)}(\omega; \delta, h_s, p_s) = -\sigma^{(2)}(\omega; -\delta, h_s, -p_s) \) and \( \sigma^{(2)}(\omega; \delta, h_s, p_s) = -\sigma^{(2)}(\omega; -\delta, -h_s, p_s) \) [57]. Hence, the lowest order terms in the symmetry-breaking parameters are proportional to \( h_s(\delta) \) or \( h_s p_s \). Another important feature is that the spin current vanishes when \( \delta = p_s/p \).

This is a well-known result in the photocurrent; the photocurrent induced by the linear-coupling terms vanishes in two-band models [56]. In contrast, in a finite spin current appears in our case because \( B_{\alpha\beta}(k) \) is generally different from the current operator.

We find that the nonlinear conductance in Eq. (7) shows a characteristic structure when the frequency is close to \( \Delta \), i.e., close to the lowest frequency with non-zero \( \Re[\sigma^{(2)}(\omega)] \). The asymptotic form of \( \Re[\sigma^{(2)}(\omega)] \) reads \( \sigma^{(2)}(\omega) \propto \sqrt{\delta\omega} \), where \( \delta\omega = \omega - \Delta \) [57]. This frequency dependence is related to the momentum dependence of \( g(k) \equiv B_{+-}(k)J_{+-}(k)[B_{+-}(k) - B_{++}(k)] \) at the band edge. The real part of \( g(k) \) is always zero in our model. Therefore, Eq. (6) becomes

\[
\sigma^{(2)}(\omega) = \frac{1}{8} \Re[\rho(\varepsilon_{+}(k_0 + k_\omega))\rho(\varepsilon_{+}(k_0 + k_\omega))],
\]  

(8)

where \( \rho(\varepsilon) \) is the density of states (DOS) and \( k_\omega > 0 \) is a wavenumber such that \( \omega = \varepsilon_{+}(k_0 + k_\omega) - \varepsilon_{-}(k_0 + k_\omega) \). Here, \( k_0 \) is the location of the band bottom; it is \( k_0 = \pi/2 \) \( (k_0 = 0) \) when \( 1 > \delta^2 \) \( (1 < \delta^2) \). By definition, \( \delta\omega = \varepsilon_{+}(k_0 + k_\omega) - \varepsilon_{-}(k_0 + k_\omega) - \Delta \) and \( k_\omega \to 0 \) when \( \delta\omega \to 0 \). The asymptotic form \( g(k_0 + k_\omega) \propto k_0^0 \) makes \( \sigma^{(2)}(\omega) \propto \delta\omega^{-1/2} \) through the relations \( \delta\omega \propto k_\omega^2 \) and \( \rho \propto 1/\sqrt{\delta\omega} \). For the present case, \( g(k_0 + k_\omega) \propto k_\omega^0 \) leads to \( \sigma^{(2)}(\omega) \propto \sqrt{\delta\omega} \). In other words, the asymptotic form of \( \sigma^{(2)}(\omega) \) reflects \( g(k) \), i.e., \( B_{\alpha\beta}(k) \). As shown below, different asymptotic form of \( g(k) \) and \( \sigma^{(2)}(\omega) \) appears for different kinds of spin-light couplings.

Zeeman coupling — The Zeeman coupling also contributes to the spin current. We here consider an oscillating magnetic field \( B(t) = B \cos(\omega t) \) parallel to the magnetic moments. The Hamiltonian reads:

\[
H_Z = -B(t) \sum_i (\eta - (-1)^i \eta_\alpha) S_i^z.
\]  

(9)

This is in contrast to the case of usual spin pumping [23–25], in which an oscillating magnetic field perpendicular to the magnetic moment is considered. The spin current is calculated using Eq. (6) by the replacement \( B_{\alpha\beta}(k) \to \).
\[ \langle \alpha k | H_Z | \beta k \rangle \]. The result reads

\[
\sigma^{(2)}(\omega) = \frac{8 \text{sgn}(1 - \delta^2) \delta J^4 h_s \eta_s^2}{\pi \omega^2 \sqrt{(\omega^2 - \Delta_s^2)(\Delta_s^2 - \omega^2)}}, \tag{10}
\]

at \( T = 0 \) and \( \tau \to \infty \). The photocurrent depends on the staggered magnetic field \( \eta_s \) and not to \( \eta \). This follows from the form of the two-band equation in Eq. (6). Naively, three terms appear for \( H_Z \), which are proportional to \( \eta_s^2 \), \( \eta_s \eta_s \), and \( \eta_s^3 \). However, the \( \eta_s \eta_s \) term has \( B_{\alpha \beta}^{(2)}(k) = \eta_s^2 \alpha \beta \) for one of the two \( B_{\alpha \beta}^{(2)}(k)'s \) in Eq. (6) \([B_{+ -}(k) \text{ or } B_{- +}(k) - B_{+ +}(k)]\). As \( B_{+ -}(k) = B_{- +}(k) - B_{+ +}(k) = 0 \) for \( B_{\alpha \beta}^{(2)}(k) = \eta \hat{S}_s \alpha \beta \), the \( \eta_s \eta_s \) term vanishes. Similarly, the \( \eta_s^3 \) term also vanishes. Hence, only the staggered magnetic field contributes to the spin current.

A notable difference from the inverse DM case appears at the lower edge of the spectrum at \( \omega = \Delta \). The conductivity shows a divergence; the asymptotic form is \( \sigma^{(2)}(\omega) \propto 1/\sqrt{\omega \Delta} \). The divergence is a consequence of the asymptotic form of \( g(k) \), which behaves differently from the asymmetric exchange case; \( g(k) \) for the Zeeman coupling become a constant when \( \omega \ll \Delta \). The substitution of \( g(k) \) into Eq. (5) gives the asymptotic form \( \sigma^{(2)}(\omega) \propto \rho(k_\omega) \propto \omega^2 \Delta^{-1/2} \). Hence, the divergence reflects the structure of the DOS.

Magnetostriction effect — Magnetostriction effect also leads to a coupling between local exchange interaction and an external electromagnetic field \([54, 55, 58, 61]\); the Hamiltonian reads

\[
H_{ms} = E_x(t) \sum_i \{ A + (-1)^i A_x \} (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y). \tag{11}
\]

Here, \( A \) and \( A_x \) are the uniform and staggered magnetostriction terms, respectively, and \( E_x(t) = E_x \cos(\omega t) \) is the oscillating electric field along the \( x \) axis. \( A (A_x) \) is the magnetostriction effect for \( J (J^\delta) \).

The solution for \( H_{ms} \) at \( T = 0 \) and \( \tau \to \infty \) reads

\[
\sigma^{(2)}(\omega) = -\frac{\text{sgn}(1 - \delta^2) h_s}{4 \pi \omega^2 J^2 (1 - \delta^2)^2 \sqrt{(\omega^2 - \Delta_s^2)(\Delta_s^2 - \omega^2)}} \times \left\{ A(\Delta_s^2 - \omega^2) + A_x \delta (\omega^2 - \Delta_s^2) \right\} \times \left\{ A_s(\Delta_s^2 - \omega^2) + A_{\delta} \delta (\omega^2 - \Delta_s^2) \right\}. \tag{12}
\]

Figure 2(c) shows the \( \omega \) dependence of \( \sigma^{(2)}(\omega) \) for \( J = 1, \delta = 1/3, \) and \( h_s = 1/10 \). Unlike the other two cases, the asymptotic structure at \( \omega \sim \Delta \) changes depending on \( A \) and \( A_x \). When \( \delta^2 < 1 \), a divergent structure similar to the Zeeman coupling, \( \sigma^{(2)}(\omega) \approx \frac{1}{\Delta_s^2} \), appears for \( A_s \neq 0 \). On the other hand, the conductivity smoothly goes to zero at \( \omega = \Delta \) for \( A_s = 0 \); in this case, \( \sigma^{(2)}(\omega) \approx \delta \omega^2 \) at the lower edge. Therefore, the magnetostriction effect also contributes to the spin current with a characteristic behavior at the lower edge \( \omega \sim \Delta \). Further details are presented in the supplementary information \([57]\).

Relaxation time dependence — The \( \tau \) dependence of a light-induced current often reflects its microscopic mechanism. For instance, in the study of photovoltaic effect, shift current does not depend on \( \tau \) while the injection current is linearly proportional to \( \tau [26, 30] \). The numerical results of \( \sigma^{(2)}(\omega) \) for different \( \tau \) are shown in Figs. 3(d)-(f); each figure shows the results for (d) asymmetric exchange, (e) Zeeman, and (f) magnetostriction couplings. All results are calculated using \( L = 2^{14} \) sites with periodic boundary conditions. The result shows that the photo spin current is insensitive against the value of \( \tau \). Therefore, the spin current is robust against the suppression of the relaxation time. This behavior is similar to the shift current in electronic photogalvanic effects.

Discussion — In this work, we explored the generation of spin current using nonlinear response. To this end, we considered simple but realistic quantum spin chains with three different types of couplings between spins and electromagnetic field: Inverse DM, Zeeman, and magnetostriction couplings. The spin current generated by all three mechanisms is independent of relaxation time of the magnetic excitation. However, our simple model shows the spin current appears from different microscopic processes compared with the relaxation-time-independent electronic photocurrent (shift current) \([29, 38, 56]\). This feature is crucial for magnets as the total number of the bands are much less than the electronic bands. Therefore, our proposal for the spin current is generally expected in simple magnetic structures.

Another interesting feature is the anisotropy of the spin current. In our model, the spin current by inverse DM and magnetostriction couplings can be switched by rotating the electric field; the field along \( y \) axis gives inverse DM component while \( x \) gives the magnetostriction. Similarly, Zeeman coupling contributes when the magnetic field along \( z \) axis. This anisotropy in the microscopic mechanism is reflected in the frequency dependence. Experimentally, the observation of the anisotropy distinguishes the microscopic mechanism of the spin current.

We also stress that the mechanism of generating spin current differs from spin pumping \([23, 25]\). Unlike the spin pumping, all three mechanisms we considered preserves the spin angular momentum along \( z \) axis. Therefore, in contrast to the spin pumping, no angular momentum is supplied from the electromagnetic waves. The conservation decidedly shows that the spin current studied here is by the nontrivial motion of magnetic excitations.

In the last, we estimate the order of the spin current for each of the contributions. A typical value of exchange interactions are used for the estimate: \( J \sim 10^2 k_B J, \ \delta \sim 0.1, \) and \( h_s \sim \mu_B J \). The relative permittivity \( \epsilon_r = 10 \) is assumed. The excitation gap for these values reads \( \Delta_g = \Delta_s^2 \sim 1\text{meV} \). Therefore, the frequency of the light
is assumed to be $\omega \sim \Delta / \hbar \sim 1$ terahertz. Using these values, we compute the strength of the oscillating electric field required for the spin current density $J_{sc}^{(0)} = 10^{-16}$ $\text{J/cm}^2$. Here $J_{sc}^{(0)}$ is an expected, typical value of the spin current observed in a recent experiment of the spin Seebeck effect for a quasi-1D magnet Sr$_2$CuO$_3$.$^57$.$^62$. For the inverse DM coupling, the magnitude of the electric polarization $p \sim 10^{-31}$ $\text{Cm}$ and $p_s \sim 10^{-32}$ $\text{Cm}$ are used $^{63,64}$. A bulk solid of the spin chains aligned with $a_0 = 4\AA$ distance gives the nonlinear conductivity $\sigma_{3D}^{(2)} \equiv \sigma^{(2)}/a_0^2 \sim 10^{-25}$ $\text{A}^2 \text{s}^2/\text{m}^2 \text{kg}$. Therefore, $E_{\nu} = \nu I(0)/\sigma_{3D}^{(2)}/2 \sim 10^{5}$ $\text{V/cm}$ is required so that the spin current approaches the value of $J_{sc}^{(0)}$. Similarly, in the case of Zeeman coupling, the staggered moment $\eta_s \sim 0.1\mu_B$ $I/T$ gives $\sigma_{3D}^{(2)} \sim 10^{-9}$ $I/T^2\text{m}^2$. This requires $B \sim 10^{-2}$ T (or $E \sim 10^4$ V/cm) to induce $J_{sc}^{(0)}$. In the last, the magnetostriiction coupling with $A \sim A_s \sim 10^{-28}$ $\text{Jm/V}$ $^{57}$ gives $\sigma_{3D}^{(2)} \sim 10^{-19}$ $\text{A}^2\text{s}^2/\text{m}^2\text{kg}$ and $E \sim 10^2$ V/cm. Therefore, the spin current generated by all three mechanisms should be observable in experiments.

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