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

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(Received 9 November 2018; revised manuscript received 4 February 2019; published 15 May 2019)

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.

DOI: 10.1103/PhysRevLett.122.197702

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]. Spintronics using magnetic insulators is also studied, which have several advantages over the metallic materials; magnetic excitations in the insulators typically have a longer lifetime and no Ohmic loss. The magnetic states and excitations of these insulators 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 the high potential of the electromagnetic wave in controlling the magnetic states and opened a subfield utilizing lights, called optospintronics [9,22].

In contrast, the manipulation of the spin current carried by magnetic excitations is limited to ferromagnets (spin pumping) [23–25]. On the other hand, other magnetic states (antiferromagnetic, spiral, spin liquid states, etc.) potentially have different advantages such as faster response [22]. One issue, however, lies in moving the magnetic excitations; the magnetic excitations do not accelerate or drift by the electromagnetic field because they are chargeless. This problem is potentially solved by utilizing the nonlinear response of magnetic insulators [Figs. 1(a) and 1(b)]. In the nonlinear optics of noncentrosymmetric electron systems [26–28], a nontrivial dynamics of electrons during the transition process induces a "shift" of the particle position [29–32]. Recent experiments investigating this mechanism find the current propagates faster than the quasiparticle velocity [33,34]. In addition, it is insensitive to the quasiparticle 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, we here 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

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

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 or terahertz regime, h is the uniform magnetic field

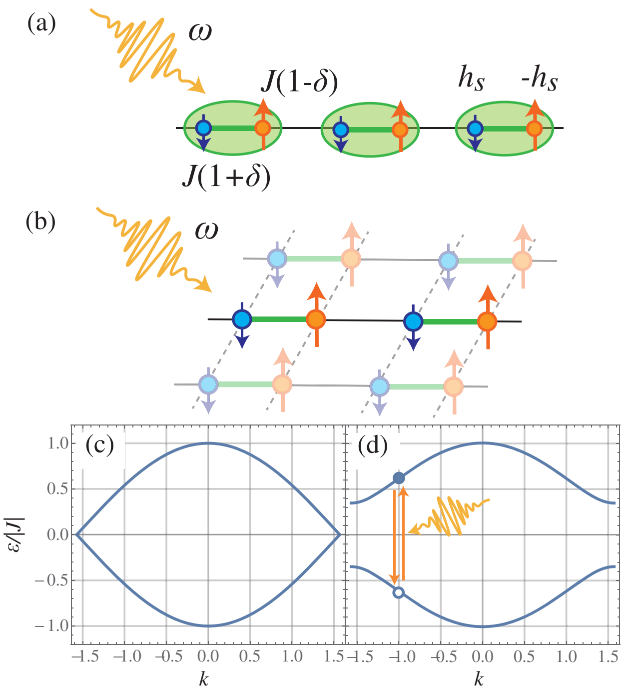

FIG. 1. Noncentrosymmetric spin chains considered in this work. Schematic picture of (a) a dimerized spin chain and (b) an antiferromagnet of weakly coupled spin chains in a staggered magnetic field. The model consists of two magnetic atoms with different g factor and alternating bonds. Band structure of Jordan-Wigner fermions for (c) $\delta = h_s/J = 0$, and (d) $\delta = 1/3$, and $h_s/J = 1/10$.

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 dimerized XY spin chains with two alternating ions [Fig. 1(a)]. In this case, the staggered magnetic field h_s reflects 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 $T = 0$ in which the Ising interaction $J_z S_i^z S_{i+1}^z$ is treated via the mean-field approximation $S_i^z = \langle S_i \rangle + (S_i^z - \langle S_i \rangle)$. For the Néel ordered state, the field $(-1)^i h_s$ is the sum of the external staggered field and the mean field $J_z \langle S_i \rangle = (-1)^i J_z M_s$ (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 interchain coupling by a mean-field theory [44–47] gives an effective one-dimensional model, Eq. (1). Namely, in this system, the staggered field h_s is renormalized by the interchain Néel order. Note that the dimerization parameter δ 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 $c_i \equiv e^{-i\pi \sum_{j=1}^{i-1} S_j^+ S_j^-} S_i^-$ and $c_i^\dagger \equiv S_i^+ e^{i\pi \sum_{j=1}^{i-1} S_j^+ S_j^-}$, Eq. (1) is fermionized as

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

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 the ith site. Figures 1(c) and 1(d) show the band structure of the JW fermions. The model has a band gap $\Delta_{\pi/2} \equiv 2\sqrt{J^2\delta^2 + h_s^2}$ for $|\delta| < 1$ [Fig. 1(d)], while the gap is $\Delta_0 \equiv 2\sqrt{J^2 + h_s^2}$ if $|\delta| > 1$. The model is gapless only if $h_s = \delta = 0$ [Fig. 1(c)]. Therefore, the ground state is robust against h as long as $h < \Delta/2$, where $\Delta \equiv \min(\Delta_0, \Delta_{\pi/2})$. We focus on the weak h 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

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

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]:

$$H_{\mathrm{iDM}} = E_y(t) \sum_i [p + (-1)^i p_s] (\boldsymbol{S}_i \times \boldsymbol{S}_{i+1})^z. \tag{4}$$

Here, the chain is along the x axis, $p \mp p_s$ 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 ω (typically, gigahertz or terahertz). 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

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

where $\epsilon_\alpha(k)$ is the eigenenergy of an αth-band state with momentum k ($|\alpha k\rangle$), $f_\alpha(k) \equiv (1 + e^{\epsilon_\alpha(k)/(k_B T)})^{-1}$ is the fermion distribution for $|\alpha k\rangle$, τ is the relaxation time of JW fermions, $J_{\alpha\beta}(k) \equiv \langle\alpha k|J_{sc}|\beta k\rangle$, and $B_{\alpha\beta}(k) \equiv \langle\alpha k|H_{\mathrm{iDM}}|\beta k\rangle$. In electronic systems, this formula well explains the experimentally observed nonlinear conductivities [57]. Hereafter, we consider the $T=0$ case of the model in Eq. (2). The conduction and valence bands [Figs. 1(c) and 1(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

$$\mathrm{Re}[\sigma^{(2)}(\omega)] = \frac{1}{\pi}\mathrm{Re}\left\{\sum_k \frac{B_{+-}(k)J_{-+}(k)[B_{--}(k)-B_{++}(k)]}{\omega^2-[\varepsilon_{+,k}-\varepsilon_{-,k}-i/(2\tau)]^2}\right\}, \quad (6)$$

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

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

$$\mathrm{Re}[\sigma^{(2)}(\omega)] = \mathrm{sgn}(1-\delta^2)\frac{h_s(p_s-p\delta)(p-p_s\delta)}{2\pi\omega^2J^2(1-\delta^2)^2} \times\sqrt{(\omega^2-\Delta_{\pi/2}^2)(\Delta_0^2-\omega^2)}, \quad (7)$$

when $\Delta\le\omega\le W\equiv\max(\Delta_0,\Delta_{\pi/2})$. On the other hand, no spin current appears for a frequency $\omega<\Delta$ or $W<\omega$, which implies that an interband 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 the following relations: $\sigma^{(2)}(\omega;\delta,h_s,p_s)=-\sigma^{(2)}(\omega;-\delta,h_s,-p_s)$ and $\sigma^{(2)}(\omega;\delta,h_s,p_s)=-\sigma^{(2)}(\omega;\delta,-h_s,p_s)$ [58]. Hence, the lowest order terms in the symmetry-breaking parameters are proportional to $h_s\delta$ or h_sp_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 general, 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 Δ, i.e., close to the lowest frequency with nonzero $\mathrm{Re}[\sigma^{(2)}(\omega)]$. The asymptotic form of $\mathrm{Re}[\sigma^{(2)}(\omega)]$ reads $\sigma^{(2)}(\omega)\propto\sqrt{\delta\omega}$, where $\delta\omega=\omega-\Delta$ [58]. 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

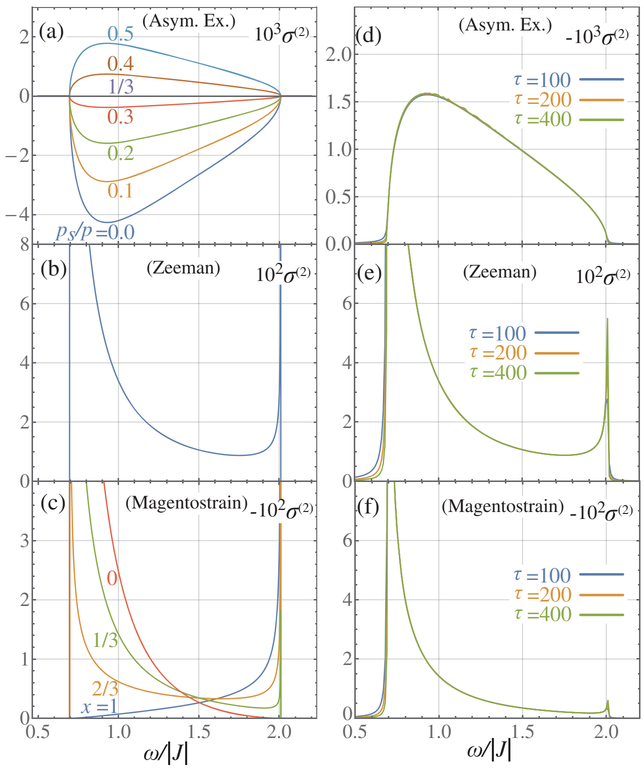

FIG. 2. Frequency dependence of the nonlinear conductivity $\sigma^{(2)}(\omega)$ for $J=1$, $\delta=1/3$, and $h_s=1/10$. Panels (a)–(c) are the results for $\tau\to\infty$ with (a) inverse Dzyaloshinskii-Moriya coupling with $p=1$, (b) Zeeman coupling, and (c) magnetostriction effect with $A=x$ and $A_s=1-x$. Different lines in (a) and (c) are the results for different ratio of parameters p_s/p and A/B, respectively. Panels (d)–(f) show the τ dependence of (d) inverse DM coupling with $p=1$ and $p_s=0.2$, (e) Zeeman coupling, and (f) magnetostriction effect with $A=1/3$ and $A_s=2/3$.

$$\sigma^{(2)}(\omega)=\frac{1}{8}\frac{\mathrm{Im}[g(k_0+k_\omega)]}{\varepsilon_+(k_0+k_\omega)}\rho[\varepsilon_+(k_0+k_\omega)], \quad (8)$$

where $\rho(\varepsilon)$ is the density of states (DOS) and $k_\omega>0$ is a wave number 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 $k_\omega\to0$ when $\delta\omega\to0$. The asymptotic form $g(k_0+k_\omega)\propto k_\omega^n$ makes $\sigma^{(2)}(\omega)\propto\delta\omega^{(n-1)/2}$ through the relations $\delta\omega\propto k_\omega^2$ and $\rho\propto1/\sqrt{\delta\omega}$. For the present case, $g(k_0+k_\omega)\propto k_\omega^2$ 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, a 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

$$H_{\mathrm{Z}} = -B(t)\sum_i [\eta - (-1)^i \eta_s] S_i^z. \tag{9}$$

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 $B_{\alpha\beta}(k) \to \langle \alpha k | H_{\mathrm{Z}} | \beta k \rangle$. The result reads

$$\sigma^{(2)}(\omega) = \frac{8\mathrm{sgn}(1-\delta^2)\delta J^4 h_s \eta_s^2}{\pi\omega^2 \sqrt{(\omega^2 - \Delta_{\pi/2}^2)(\Delta_0^2 - \omega^2)}}, \tag{10}$$

at $T = 0$ and $\tau \to \infty$. The photocurrent depends on the staggered magnetic field η_s and not on η. This follows from the form of the two-band equation in Eq. (6). Naively, three terms appear for H_{Z}, which are proportional to η_s^2, $\eta\eta_s$, and η^2. However, the $\eta\eta_s$ term has $B_{\alpha\beta}(k) = \eta \hat{1}_{\alpha\beta}$ for one of the two $B_{\alpha\beta}(k)$'s in Eq. (6) [$B_{+-}(k)$ or $B_{--}(k) - B_{++}(k)$]. As $B_{+-}(k) = B_{--}(k) - B_{++}(k) = 0$ for $B_{\alpha\beta}(k) = \eta \hat{1}_{\alpha\beta}$, the $\eta\eta_s$ term vanishes. Similarly, the η^2 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 $\omega = \Delta$. Figure 2(b) shows the result of $\sigma^{(2)}$ for $\eta_s = 1$. The conductivity shows a divergence; the asymptotic form is $\sigma^{(2)}(\omega) \propto 1/\sqrt{\delta\omega}$ [58]. 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 $\omega \searrow \Delta$. The substitution of $g(k)$ into Eq. (8) gives the asymptotic form $\sigma^{(2)} \propto \rho(k_\omega) \propto \delta\omega^{-1/2}$. Hence, the divergence reflects the structure of the DOS.

We also note that our models discussed here strictly conserve the angular momentum. The optical switching [66] and generation [67] of angular momentum in electronic systems were recently studied in metallic systems. Here, the spin-orbit interaction plays a crucial role, which transfers the angular momentum from the lattice (atoms). In contrast, our model strictly conserves the total angular momentum for S^z; the spin current is generated without generating the angular momentum. This is analogous to the photovoltaic effect where the electric current is induced without generating an electric charge.

Magnetostriction effect.—Magnetostriction effect also leads to a coupling between local exchange interaction and an external electromagnetic field [54,55,68–71]; the Hamiltonian reads

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

Here, A and A_s 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_s) is the magnetostriction effect for J ($J\delta$).

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

$$\begin{aligned}\sigma^{(2)}(\omega) = &-\frac{\mathrm{sgn}(1-\delta^2)h_s}{4\pi\omega^2 J^2(1-\delta^2)^2\sqrt{(\omega^2 - \Delta_{\pi/2}^2)(\Delta_0^2 - \omega^2)}} \\ &\times \{A(\Delta_{\pi/2}^2 - \omega^2) + A_s\delta(\omega^2 - \Delta_0^2)\} \\ &\times \{A_s(\Delta_0^2 - \omega^2) + A\delta(\omega^2 - \Delta_{\pi/2}^2)\}. \end{aligned} \tag{12}$$

Figure 2(c) shows the ω 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_s. When $\delta^2 < 1$, a divergent structure similar to the Zeeman coupling, $\sigma^{(2)}(\omega) \approx (1/\sqrt{\delta\omega})$, 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, $\sigma^{(2)}(\omega) \approx \delta\omega^{3/2}$ 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 Supplemental Material [58].

Relaxation time dependence.—The τ 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 τ, while the injection current is linearly proportional to τ [26,30]. The numerical results of $\sigma^{(2)}(\omega)$ for different τ are shown in Figs. 2(d)–2(f); each panel shows the results for [Fig. 2(d)] asymmetric exchange, [Fig. 2(e)] Zeeman, and [Fig. 2(f)] magnetostriction couplings. All results are calculated using $L = 2^{14}$ site chains with periodic boundary condition. The result shows that the photo-spin current is insensitive against the value of τ. 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, which is related to the shift of the center of the mass during the optical transition [58].

The robustness against τ is potentially beneficial for applications. The energy scale for the magnetic excitations in magnetic insulators is smaller than the electron excitations in semiconductors. Therefore, a change in the temperature affects the magnetic excitations more seriously. In particular, the effect of thermal fluctuations often appears as the decrease of the lifetime. Therefore, the insensitiveness of $\sigma^{(2)}(\omega)$ to τ implies the weak effect of heating to the conversion efficiency.

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 the 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) [29,30,56]. This feature is crucial for magnets as the total number of the bands is 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 the y axis gives the inverse DM component while x gives the magnetostriction. Similarly, Zeeman coupling contributes when the magnetic field is along the 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 preserve the spin angular momentum along the 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.

There are several experimental setups for observing the spin current of our mechanism. The most straightforward setup is to use an isolated magnet without any attached leads. In this setup, the spin current accumulates the angular momentum to the edges of the sample. In our mechanism, the sign of angular momentum accumulated to one edge is the opposite of that to the other edge because the photovoltaic effect carries the angular momentum of one end to the opposite end. Therefore, observation of the accumulated angular momentum by Kerr effect or Faraday rotation should provide evidence for the flow of angular momentum.

Alternatively, the spin current should be measurable using the inverse spin Hall effect [72–74]. In contrast to the spin pumping case [23,24], however, the attachment of two leads ("source" and "drain") is useful for our mechanism because our mechanism does not generate angular momentum in the magnets. The angular momentum flows from one lead (source) to the other lead (drain) if our mechanism is dominant. [58]. In contrast, the spin currents in the two leads both flow outward in spin pumping or spin Seebeck effect [75,76]. Hence, the two-lead setup should distinguish different mechanisms.

Finally, we compute the strength of an ac electromagnetic field required for an observable spin current. The spinon spin current is already realized in the experiment which is generated by the spin Seebeck effect [76]; we here use the magnitude of the observed spinon current as the reference. Using the above results, the required fields are estimated as $E \sim 10^5$, 10^4, and 10^2 V/cm for the cases of inverse DM, Zeeman, and magnetostriction couplings, respectively [58]. Therefore, the spin current generated by all three couplings should be observable in experiments.

The authors thank R. Matsunaga for fruitful discussion. This work was supported by JSPS KAKENHI (Grants No. JP16H06717, No. JP18H03676, No. JP18H04222, No. JP26103006, No. JP17K05513, and No. JP15H02117), ImPACT Program of Council for Science, Technology and Innovation (Cabinet office, Government of Japan), CREST JST (Grant No. JPMJCR16F1), and Grant-in-Aid for Scientific Research on Innovative Area "Nano Spin Conversion Science" (Grant No. 17H05174).

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