Gigantic enhancement of spin Seebeck effect by phonon drag

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We investigate both theoretically and experimentally a gigantic enhancement of the spin Seebeck effect in a prototypical magnet LaY$_2$Fe$_5$O$_{12}$ at low temperatures. Our theoretical analysis sheds light on the important role of phonons; the spin Seebeck effect is enormously enhanced by nonequilibrium phonons that drag the low-lying spin excitations. We further argue that this scenario gives a clue to understand the observation of the spin Seebeck effect that is unaccompanied by a global spin current, and predict that the substrate condition affects the observed signal.

When a temperature gradient is applied to a ferromagnet, a force is induced acting on electrons’ spin to drive spin currents. This phenomenon termed the spin Seebeck effect (SSE) has recently drawn tremendous attention as a new source of spin currents needed for future spin-based electronics. SSE is now established as an universal aspect of ferromagnetic materials as it has been observed in a variety of materials ranging from a metallic ferromagnet Ni$_1$Fe$_{19}$ and a semiconducting ferromagnet GaMnAs to an insulating magnet LaY$_2$Fe$_5$O$_{12}$. Besides its impact on the technological application, SSE offers a number of new topics on the interplay of heat and spin currents and it triggered the emergence of the new field named “spin caloritronics” in the rapidly-growing spintronics community.

A mystery concerning SSE was how conduction electrons can sustain the spin voltage over so long range of several millimeters in spite of the short conduction electron’s spin-flip diffusion length $\lambda_{sf}$, which is typically of several tens nanometers. This problem has recently been resolved by a series of experiments on spin currents using magnetic insulators. A recent experiment on the electric signal transmission through a magnetic insulator highlights the role of the low-lying magnetic excitation of localized spins, i.e., magnons, by demonstrating that magnons transmit the spin current over a long distance of several millimeters. A subsequent experiment on SSE for a magnetic insulator LaY$_2$Fe$_5$O$_{12}$ confirmed that the magnon-based scenario can explain the SSE experiment at room temperature, since the length scale associated with magnons $\gg \lambda_{sf}$. However, a new issue on SSE was brought by a very recent experiment on a ferromagnetic semiconductor GaMnAs, where it was demonstrated, by cutting the magnetic coupling in GaMnAs while keeping the thermal contact, that SSE can be observed even in the absence of global spin current flowing through GaMnAs. Obviously, the scenario of magnon-mediated SSE fails to explain the experiment, showing that the full understanding of SSE has not yet been reached.

For a deep understanding of the physics behind SSE, we here explore the low-temperature behavior of SSE in an insulating magnet LaY$_2$Fe$_5$O$_{12}$. Figure 1 shows a schematic illustration of our device structure. An in-plane external magnetic field $H$ and a uniform temperature gradient $\nabla T$ were applied along the $z$ direction [see FIG. 1(a)]. The $\nabla T$ generates a spin voltage across the LaY$_2$Fe$_5$O$_{12}$/Pt interface, and injects (ejects) a spin current $I_s$ into (from) the Pt wire. In the Pt wire, a part of the injected/ejected $I_s$ is converted into a charge voltage through the so-called inverse spin-Hall effect (ISHE):

$$V_{\text{ISHE}} = \Theta_H |e|I_s(\rho/w),$$

where $|e|$, $\Theta_H$, $\rho$ and $w$ are the absolute value of electron

![FIG. 1. (Color online) Gigantic enhancement of SSE in LaY$_2$Fe$_5$O$_{12}$ at low temperatures. (a) Schematic illustration of the LaY$_2$Fe$_5$O$_{12}$/Pt sample and the temperature profile along the $z$ direction. Here $H$ denotes an external magnetic field (with magnitude $H$). The sample comprises a LaY$_2$Fe$_5$O$_{12}$ film with $8 \times 4$ mm$^2$ rectangular shape and two separated Pt wires with the width $w$ attached to the LaY$_2$Fe$_5$O$_{12}$ surface at the interval of 5.6 mm. (b) $T$ dependence of $V/\Delta T$ at $H = 100$ Oe.](https://example.com/fig1.png)
FIG. 2. (Color online) Diagrammatic representation of the thermal spin injection process. (a) Magnon-mediated SSE. Here the system is composed of ferromagnet (F, in the experiment La$_2$Fe$_3$O$_{12}$) and nonmagnetic metals (N, in the experiment Pt), which are divided into three temperature domains of $F_1/N_1$, $F_2/N_2$, and $F_3/N_3$ with their local temperatures of $T_1$, $T_2$, and $T_3$. The thin solid lines with arrows (bold lines without arrows) are electron (magnon) propagators. Here, $J_{sd}$ ($J_{xx}$) is the strength of the s-d coupling at the F/N interface (the exchange coupling in F). (b) Phonon-dragged SSE where the dashed lines are phonon propagators. The process $P_1$ injects the spin current with the same magnitude as (but opposite sign to) the process $P_2$ due to the relation $T_1 - T_2 = -(T_3 - T_2)$, while no spin current is injected into $N_2$ because of the cancellation between the two relevant processes $P_3$ and $P'_3$. Here, $\Omega_0 = \sqrt{K_{ph}/M_{ion}}$ with the ion mass $M_{ion}$ and the elastic constant $K_{ph}$ in F. (c) Calculated spatial dependence of the spin current injected into $N_i$ ($i = 1, 2, 3$).

FIG. 3. (Color online) Comparison of experimental and theoretical SSE signal. Solid circles: experimental spin Seebeck data for La$_2$Fe$_3$O$_{12}$ (Dotted line is a guide to the eye). The solid curve: calculated $T$-dependence of $V_{SHE}$ due to the sum of the phonon-dragged SSE and the magnon-mediated SSE. The dashed curve: calculated $T$-dependence of $V_{SHE}$ due only to the magnon-mediated SSE. We have assumed $T$-dependent $\Theta_H$ and $\alpha$, and used $T_D = 565$ K and $T_M = 560$ K. The data are normalized by its value at 50 K except that the result for magnon-mediated SSE is plotted to reproduce the room-temperature signal. In set: experimental thermal conductivity $\kappa$ for Y$_3$Fe$_5$O$_{12}$ taken from Ref. 23 (solid circles) and the result of the fit (solid curve) using Eq. 1.
is proportional to the experimentally-detectable electric voltage via ISHE [Eq. (1)]. Following Ref. 11, the spin current $I_{s}^{\text{mag}}(N_1)$ injected into $N_1$ due to the magnon-mediated SSE is calculated as

$$I_{s}^{\text{mag}}(N_1)/\Delta T = \left(\frac{P}{\alpha}\right) \int_{0}^{T_M/T} dv \frac{(T/T_M)^2 s^2}{4 \sinh^2(v/2)} ,$$

(2)

where $\alpha$ is the Gilbert damping constant, $T_M$ is the characteristic temperature corresponding to the magnon high-energy cutoff, and $P$ is a nearly-temperature-independent coefficient. Equation (2) means that the magnon-mediated SSE cannot explain the low-$T$ enhancement of the signal (the dashed curve in FIG. 3).

Now we proceed to a detailed analysis of the $T$-dependence of SSE in terms of the phonon-drag mechanism. The Feynman diagram for the phonon-drag process in the present situation is shown in FIG. 2(b), where the phonons feel the temperature difference between $F_1$ and $F_2$, and drag magnons through the magnon-phonon interaction. Since the nonequilibrium phonons affect the magnon dynamics, this process injects spin current into $N_1$. The important point is that the spin current $I_{s}^{\text{drag}}$ injected in this process becomes proportional to the phonon lifetime $\tau_{\text{ph}}$ as

$$I_{s}^{\text{drag}}(N_1)/\Delta T = P'\tau_{\text{ph}}B_1B_2 ,$$

(3)

where $B_1 = (T/T_D)^{5/2} \int_0^{T_D/T} dv \frac{v^6}{\sinh^2(u/2)}$ and $B_2 = (T/T_D)^{5/2} \left( \frac{B_0 T_M}{k_B \tau_{\text{ph}} s^2} \right)^{3/2} \int_0^{T_M/T} dv \frac{v^2}{\sinh^2(v/2)}$ with the Debye temperature $T_D$, and $P'$ is a nearly-temperature-independent coefficient. Since $\tau_{\text{ph}}$ in a high-purity specimen is known to increase steeply at low $T$ because of the rapid suppression of umklapp scattering, it leads to the drastic enhancement of the phonon-dragged SSE. In our analysis, the $T$-dependence of $\tau_{\text{ph}}$ is extracted from the thermal conductivity data for Y$_3$Fe$_5$O$_{12}$ (see the inset of FIG. 3) using

$$\kappa(T) = (1/3)\nu_{\text{ph}}^2 C_{\text{ph}}(T) \tau_{\text{ph}}(T) ,$$

(4)

where $\nu_{\text{ph}}$ is the phonon velocity, and $C_{\text{ph}}(T) = 9N_Dk_B(T/T_D)^3 \int_0^{T_D/T} dv \frac{v^3}{\sinh^2(v/2)}$ is the phonon specific heat with the number of phonon modes $N_D$. After getting the information on $\tau_{\text{ph}}(T)$, we calculate the $T$-dependence of $V_{\text{ISHE}}$ resulting from the phonon-dragged SSE. The result, plotted in FIG. 3 (the solid curve), shows an excellent description of the low-$T$ enhancement of SSE. Our analysis demonstrates that the phonon-drag mechanism is of crucial importance to understand SSE below the room temperature.

Finally, we show in FIG. 4 our interpretation on the observation of SSE that is unaccompanied by a global spin current, where the heat is carried by phonons through the nonmagnetic substrate while the spin is injected locally at the $F/N$ interface. This interpretation is reinforced when we recall that the magnitude of the spin Seebeck signal is enhanced with decreasing $T$ even well below the Curie temperature, whose tendency is consistent with the phonon-drag mechanism as is seen in FIG. 3. Furthermore, the fact that the experiment was done below the room temperature supports the phonon-drag-based scenario, since the phonon-drag process becomes more effective at low $T$ as emphasized in the previous paragraph. All these considerations strongly support that the SSE experiment for GaMnAs can be interpreted in terms of the phonon-drag mechanism, and results in a prediction that the substrate condition affects the observed signal. Our demonstration opens a new route to control spin currents by means of phonons and stimulates further progresses in spin caloritronics.

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\[ I_s(N_1, t) = \frac{2\gamma_H}{N_F N \hbar} \Re C_{k,q}^< (t, t), \] (S1)

where \( N_F (N_N) \) is the number of lattice sites in \( F (N) \), \( S_0 \) is the size of the localized spins in \( F \), and \( J_{sd}^k+q \) is the Fourier transform of the \( s-d \) interaction at the \( F/N \) interface. Here, \( C_{k,q}^< (t, t') = -i(\alpha_q^+ (t') s_k^-(t)) \) measures the correlation between the magnon operator \( \alpha_q^+ \) and the spin-density operator \( s_k^-(t) \). Note that the time dependence of \( I_s(N_1, t) \) vanishes in the steady state and it is hereafter discarded. Introducing the frequency representation \( C_{k,q}^< (t-t') = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} C_{k,q}^< (\omega) e^{-i\omega(t-t')} \) and adopting the representation \( [2] \) \( \hat{C} = \begin{pmatrix} C_R & C_K \\ 0 & C_A \end{pmatrix} \) as well as using the relation \( C^< = \frac{1}{2} (C_K - C_R + C_A) \), we obtain

\[ I_s(N_1) = \sum_{q,k} \frac{2\gamma_H^{k-q} \sqrt{N_0}}{\sqrt{2N_F N \hbar}} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Re C_{k,q}^< (\omega), \] (S2)

for the spin current \( I_s(N_1) \) in the steady state.

When we introduce a renormalized magnon propagator \( \delta \hat{X}_q (\omega) \), the interface correlation \( \hat{C} \) appearing in Eq. (S2) is generally expressed as

\[ \hat{C}_{k,q}^< (\omega) = \frac{J_{sd}^{k-q}}{\sqrt{N_N N_F \hbar}} \chi_k^-(\omega) \delta \hat{X}_q (\omega), \] (S3)

where \( \chi_k^-(\omega) \) is the spin-density propagator satisfying the local equilibrium condition:

\[ \chi_k^-(\omega) = \chi_k^+(\omega)^*; \quad \chi_k^+(\omega) = 2i \Im \chi_k^R (\omega) \coth \left( \frac{\hbar \omega}{2\gamma_H} \right). \] (S4)

Here the retarded component of \( \chi_k^+(\omega) \) is given by \( \chi_k^R (\omega) = \chi_N / (1 + \lambda_d^2 k^2 - i\omega \tau_d) \) [S3] where \( \chi_N, \lambda_d, \) and \( \tau_d \) are the paramagnetic susceptibility, the spin diffusion length, and spin relaxation time, respectively.

We now consider the phonon-dragged SSE [the process \( P_3 \) shown in FIG. 2 (b)]. The renormalized magnon propagator \( \delta \hat{X}_q (\omega) \) in the present case is given by

\[ \delta \hat{X}_q (\omega) = \hat{X}_q (\omega) \tilde{\Sigma}_q (\omega) \hat{X}_q (\omega), \] (S5)

where \( \hat{X}_q (\omega) = \left( X_q^R (\omega), X_q^K (\omega) \right) \) is the bare magnon propagator satisfying the equilibrium condition:

\[ X_q^A (\omega) = \left( X_q^K (\omega) \right)^*; \quad X_q^K (\omega) = 2i \Im X_q^R (\omega) \coth \left( \frac{\hbar \omega}{2\gamma_H} \right). \] (S6)

Here, the retarded component is given by \( X_q^R (\omega) = (\omega - i\omega_q + i\omega)^{-1} \), where \( \omega_q = \gamma H_0 + \omega_q \) is the magnon frequency for uniform mode \( \gamma H_0 \) and exchange mode

SUPPLEMENTAL MATERIAL

1. Experimental details

The single-crystal LaY2Fe5O12 (111) film with the thickness of 3.9 μm was grown on a Ga3Ga5O12 (111) substrate by liquid phase epitaxy. The 15-nm-thick Pt wires were then sputtered in an Ar atmosphere. The length and width of each Pt wire are 4 mm and 0.1 mm, respectively. The temperatures of the lower- and higher-temperature ends of the sample were respectively stabilized to \( T \) and \( T + \Delta T \), where \( T \) was controlled in the range of 300-50 K by means of a closed-cycle helium refrigerator.

Following Ref. [1], the spin current \( I_s(N_1, t) \) injected into the nonmagnetic metal \( N_1 \) \((i = 1, 2, 3)\) is calculated as

\[ I_s(N_1, t) = -\sum_{q,k} \frac{2\gamma_H^{k-q} \sqrt{N_0}}{\sqrt{2N_F N \hbar}} \Re C_{k,q}^< (t, t), \] (S1)

where \( N_F (N_N) \) is the number of lattice sites in \( F (N) \), \( S_0 \) is the size of the localized spins in \( F \), and \( J_{sd}^{k+q} \) is the Fourier transform of the \( s-d \) interaction at the \( F/N \) interface. Here, \( C_{k,q}^< (t, t') = -i(\alpha_q^+ (t') s_k^-(t)) \) measures the correlation between the magnon operator \( \alpha_q^+ \) and the spin-density operator \( s_k^-(t) \). Note that the time dependence of \( I_s(N_1, t) \) vanishes in the steady state and it is hereafter discarded. Introducing the frequency representation \( C_{k,q}^< (t-t') = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} C_{k,q}^< (\omega) e^{-i\omega(t-t')} \) and adopting the representation \([2]\) \( \hat{C} = \begin{pmatrix} C_R & C_K \\ 0 & C_A \end{pmatrix} \) as well as using the relation \( C^< = \frac{1}{2} (C_K - C_R + C_A) \), we obtain

\[ I_s(N_1) = \sum_{q,k} \frac{2\gamma_H^{k-q} \sqrt{N_0}}{\sqrt{2N_F N \hbar}} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Re C_{k,q}^< (\omega), \] (S2)

for the spin current \( I_s(N_1) \) in the steady state.

When we introduce a renormalized magnon propagator \( \delta \hat{X}_q (\omega) \), the interface correlation \( \hat{C} \) appearing in Eq. (S2) is generally expressed as

\[ \hat{C}_{k,q}^< (\omega) = \frac{J_{sd}^{k-q}}{\sqrt{N_N N_F \hbar}} \chi_k^-(\omega) \delta \hat{X}_q (\omega), \] (S3)

where \( \chi_k^-(\omega) \) is the spin-density propagator satisfying the local equilibrium condition:

\[ \chi_k^-(\omega) = \chi_k^+(\omega)^*; \quad \chi_k^+(\omega) = 2i \Im \chi_k^R (\omega) \coth \left( \frac{\hbar \omega}{2\gamma_H} \right). \] (S4)

Here the retarded component of \( \chi_k^+(\omega) \) is given by \( \chi_k^R (\omega) = \chi_N / (1 + \lambda_d^2 k^2 - i\omega \tau_d) \) [S3] where \( \chi_N, \lambda_d, \) and \( \tau_d \) are the paramagnetic susceptibility, the spin diffusion length, and spin relaxation time, respectively.

We now consider the phonon-dragged SSE [the process \( P_3 \) shown in FIG. 2 (b)]. The renormalized magnon propagator \( \delta \hat{X}_q (\omega) \) in the present case is given by

\[ \delta \hat{X}_q (\omega) = \hat{X}_q (\omega) \tilde{\Sigma}_q (\omega) \hat{X}_q (\omega), \] (S5)

where \( \hat{X}_q (\omega) = \left( X_q^R (\omega), X_q^K (\omega) \right) \) is the bare magnon propagator satisfying the equilibrium condition:

\[ X_q^A (\omega) = \left( X_q^K (\omega) \right)^*; \quad X_q^K (\omega) = 2i \Im X_q^R (\omega) \coth \left( \frac{\hbar \omega}{2\gamma_H} \right). \] (S6)

Here, the retarded component is given by \( X_q^R (\omega) = (\omega - i\omega_q + i\omega)^{-1} \), where \( \omega_q = \gamma H_0 + \omega_q \) is the magnon frequency for uniform mode \( \gamma H_0 \) and exchange mode
while $\omega_q = D_{ex}q^2/\hbar$. In Eq. \[[S5]\], the selfenergy $\Sigma$ due to phonons is given by

$$\hat{\Sigma}_{q}(\omega) = \frac{i}{2N_F} \sum_{\mathbf{K}} \left( \frac{\Gamma_{\mathbf{K}q}}{\hbar} \right)^2 \int \delta D^R(\nu) \hat{X}_{q-}(-\tau_1) + \delta D^A(\nu) \hat{X}_{q-}(+\tau_1) \right],$$

(S7)

where $\Gamma_{\mathbf{K}q} = \bar{\omega}_q \sqrt{\frac{\hbar}{2M_{ion}v_{ph}}}$ is the magnon-phonon interaction vertex with $\nu_\mathbf{K}$, $v_{ph}$ and $M_{ion}$ being the phonon frequency, phonon velocity and the ion mass, $\tau$ is the Pauli matrix in the Keldysh space, and we have introduced the shorthand notations $\omega_- = \omega - \nu$, $q_- = q - \mathbf{K}$, and $\int_\nu = \int_{-\infty}^{\infty} \frac{d\nu}{2\pi}$. In Eq. \((S7)\), the full phonon propagator $\delta \hat{D}_\mathbf{K}$ (the whole of the phonon lines for $P_2$ in FIG. 2 (b)) is written as \[[S4]\]

$$\delta \hat{D}_\mathbf{K}(\nu) = \delta \hat{D}^{l-eq}_\mathbf{K}(\nu) + \delta \hat{D}^{n-eq}_\mathbf{K}(\nu).$$

(S8)

Here, $\delta \hat{D}^{l-eq}(\omega)$ is the local-equilibrium propagator satisfying the local-equilibrium conditions $\delta D^{l-eq,R}(\nu) = [\delta D^{l-eq,R}(\nu)]^*$ and $\delta D^{l-eq,K}(\nu) = [\delta D^{l-eq,K}(\nu)]^*$ with its retarded component given by

$$\delta D^{l-eq,R}(\nu) = \frac{\Omega_0^2}{N_F(\Lambda/a_S)} \sum_{\mathbf{K}'} [\hat{D}^{K}_\mathbf{K}(\nu)]^2 \hat{D}^{R}_\mathbf{K}(\nu),$$

(S9)

while $\delta \hat{D}^{n-eq} = \begin{pmatrix} 0 & \delta D^{n-eq,K} \\ \delta D^{n-eq,R} & 0 \end{pmatrix}$ is the nonequilibrium propagator with its Keldysh component given by

$$\delta D^{n-eq,K}(\nu) = \frac{\Omega_0^2}{N_F(\Lambda/a_S)} \sum_{\mathbf{K}'} \frac{[\hat{D}^{K}_\mathbf{K}(\nu) - \hat{D}^{A}_\mathbf{K}(\nu)]^2}{\left[ \text{coth}(\frac{\hbar \nu}{2k_B T_1}) - \text{coth}(\frac{\hbar \nu}{2k_B T_2}) \right]},$$

(S10)

In the above equations, $\Omega_0 = \sqrt{K_{ph}/M_{ion}}$ with the elastic constant $K_{ph}$ in $F$, and $\hat{D}_\mathbf{K}(\nu) = \begin{pmatrix} D^R_\mathbf{K}(\nu) & D^K_\mathbf{K}(\nu) \\ 0 & D^A_\mathbf{K}(\nu) \end{pmatrix}$ is the bare phonon propagator satisfying the equilibrium condition:

$$D^R_\mathbf{K}(\nu) = [D^R_\mathbf{K}(\nu)]^*; \quad D^K_\mathbf{K}(\nu) = 2i \text{Im} D^R_\mathbf{K}(\nu) \text{coth}(\frac{\hbar \nu}{2k_B T_1}),$$

(S11)

with its retarded component and the phonon lifetime given by $D^R_\mathbf{K}(\nu) = (\nu - \nu_\mathbf{K} + i/\tau_{ph})^{-1} - (\nu + \nu_\mathbf{K} + i/\tau_{ph})^{-1}$ and $\tau_{ph}$.

Now substituting these expressions into equation \[[S5]\] and use Eq. \[[S2]\], the spin current injected into $N_1$ by the phonon-drag process is calculated as

$$I^\text{drag}_s(N_1) = \frac{R_{\tau}}{N_N N_F} \sum_{k,q,\mathbf{K}} \int d\nu' \text{tr}(\nu') \left[ \hat{S}^{\nu'}_{\mathbf{K}q} \Gamma_{\mathbf{K}q}(\nu') \right] \cdot \left[ \text{coth}(\frac{\hbar \nu'}{2k_B T_1}) - \text{coth}(\frac{\hbar \nu'}{2k_B T_2}) \right],$$

(S12)

where $R = \sqrt{2(J_{sd}^2 S_0) \Omega_0^2 N_{int}(a_S/\Lambda) \tau_{ph}/(4\pi^3 \hbar^4 \nu_D^4)}$ with $\nu_D = v_{ph}/a_S$, and

$$A_{k,q}(\nu) = \int \text{Im} \chi^R_k(\omega) \text{Im} X^R_q(\omega) X^R_q(\omega) \left[ \text{coth}(\frac{\hbar \nu}{2k_B T_1}) - \text{coth}(\frac{\hbar \nu}{2k_B T_2}) \right].$$

(S13)

By setting $T = T_2$, $\Delta T = T_1 - T_2$, and after some algebra, we obtain Eq. (3) in the main text.

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