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Predicted rectification and negative differential spin Seebeck effect at magnetic interfaces

Jie Ren*

Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

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We study the nonequilibrium Seebeck spin transport across metal-magnetic insulator interfaces. The conjugate-converted thermal-spin transport is assisted by the exchange interaction at the interface, between conduction electrons in the metal lead and localized spins in the insulating magnet lead. We predict the rectification and negative differential spin Seebeck effect and resolve their microscopic mechanism, as a consequence of the strongly fluctuated electronic density of states in the metal lead. The rectification of spin Peltier effect is also discussed. The phenomena predicted here are relevant for designing efficient spin/magnon diode and transistor, which could play crucial roles in controlling energy and information in functional devices.

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Recently, spin Seebeck effect (SSE), a phenomenon that temperature bias can produce a spin current and an associated spin voltage, has been observed in magnetic metals,[1] semiconductors,[2,3] insulators,[4–7] and even nonmagnetic materials with spin-orbit coupling.[8] Since then, the SSE has ignited a new upsurge of research interest, because it acts as a new method facilitating the functional use of heat and opens a new possibility of spintronics,[9] magnonics,[10] and spin caloritronics.[11,12]

Of particular interest is the SSE in the insulating magnetic interface.[4–7] The reason is that different from spin-dependent Seebeck effect in metallic materials, SSE allows heat to generate a pure flow of spin angular momentum, a flow of spins *without* electron currents. This becomes obvious only after the observation of the SSE through magnetic insulator-metal interfaces.[4] Itinerant electrons are often problematic in the thermal design of devices, an issue that can be avoided by the SSE in magnetic insulator interfaces without conducting electron currents. It allows us to construct efficient thermoelectric devices upon new principles[13] and to realize nondissipative information and energy transfer in the absence of Joule heating.[14,15]

In this Rapid Communication, we uncover interesting phenomena of the SSE across metal-insulating magnet interface: the *rectification* and *negative differential* SSE, that is, reversing the thermal bias gives asymmetric spin currents and increasing thermal bias abnormally gives a decreasing spin current. The rectification of spin Peltier effect (SPE) is also discussed. We first demonstrate the absence of rectification and negative differential SSE in magnetic interfaces with *constant* electronic density of states (DOS) in the metallic lead. We then uncover that the *strongly fluctuated* electron DOS is the key to retaining these intriguing spin Seebeck properties. As examples, we demonstrate the nontrivial rectification and negative differential SSE in several typical cases with nonsmooth strongly fluctuated electron DOS. Our results readily render analytic interpretations and clear physical insights of the microscopic mechanism, which can provide further guidance for the optimization of the predicted effects in the future.

The rectification and negative differential electronic/phononic conductances have played fundamental roles in realizing functional electronic/phononic diodes and transistors that are building blocks of modern electronics/phononics.[16] By the same token, the predicted rectification and negative differential SSEs are also fundamental for constructing magnonic/spin caloritronic circuits with efficient spin Seebeck diodes and transistors. Therefore, we expect that our results would play crucial roles in spintronics,[9] magnonics,[10] and spin caloritronics,[12] and could have potential applications in controlling energy and information in low-dimensional nanodevices.[16]

The magnetic interface system is schematically illustrated in Fig. 1(a), similar to the setup of longitudinal spin Seebeck experiments.[5,6] The left metallic lead is described by the free electrons: $H_L = \sum_{k\sigma}(\varepsilon_{k\sigma} - \mu_\sigma)c^\dagger_{k\sigma}c_{k\sigma}, (\sigma = \uparrow, \downarrow)$, with possibly different spin-dependent chemical potentials μ_σ induced by spin accumulation.[17,18] The spin voltage $\Delta\mu_s = \mu_\downarrow - \mu_\uparrow$ can be measured by the Hanle method[3] or be converted into an electric voltage through the inverse spin Hall effect.[4] The right insulating magnetic lead can be described by a Heisenberg lattice $H_R = -J\sum_{\langle i,j\rangle}[\frac{1}{2}S^+_i S^-_j + \frac{1}{2}S^-_i S^+_j + S^z_i S^z_j]$, where $S^\pm_j$ is the raising (lowering) operator for the localized spin at site j, S^z_j is the spin operator of the z direction, and J denotes the exchange coupling strength. The spin operators are conveniently mapped into bosonic magnons by Holstein-Primakoff transformation:[19] $S^+_j = \sqrt{2S_0 - a^\dagger_j a_j}\,a_j$, $S^-_j = a^\dagger_j\sqrt{2S_0 - a^\dagger_j a_j}$, $S^z_j = S_0 - a^\dagger_j a_j$, where S_0 is the length of localized spins. At large spin limit or low temperatures ($\langle a^\dagger_j a_j\rangle \ll 2S_0$) we can approximate $S^-_j \approx \sqrt{2S_0}a^\dagger_j$ and $S^+_j \approx \sqrt{2S_0}a_j$. Clearly, the creation (annihilation) of a local magnon $a^\dagger_j(a_j)$ at site j corresponds to the lowering (rising) of the local spin component, i.e., excitation of magnons means that spins point less in the z direction and magnetization goes down. Therefore, after a Fourier transform into the momentum space, the right insulating magnetic lead is approximated by

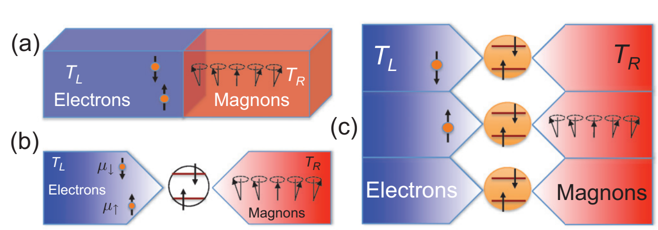

FIG. 1. (Color online) Schematic illustration of different setups of the metal-insulating magnetic interfaces.

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the free magnon gas: $H_R \approx \sum_q \hbar\omega_q a_q^\dagger a_q + \text{const}$, where the dispersion of ω_q depends on the lattice details.

Similar to Refs. 20–23, the interfacial electron-magnon interaction is described by the s-d exchange coupling:[24]

$$H_{sd} = -\sum_{k,q} J_q [S_q^- c_{k\uparrow}^\dagger c_{k+q\downarrow} + S_q^+ c_{k+q\downarrow}^\dagger c_{k\uparrow}], \tag{1}$$

where $S_q^- \approx \sqrt{2S_0}\, a_q^\dagger$, $S_q^+ \approx \sqrt{2S_0}\, a_q$ are in the momentum space and J_q denotes the effective exchange coupling at the interface. The first term $S_q^- c_{k\uparrow}^\dagger c_{k+q\downarrow}$ describes the magnon emission process into the right insulating magnet associated with scattering of a spin-down electron to a spin-up electron at the left metallic side of the interface. The second term $S_q^+ c_{k+q\downarrow}^\dagger c_{k\uparrow}$ describes the reversed process that a spin-up electron near the interface absorbs a magnon from the right side and excites to a spin-down state with spin flip. Note that the contribution of $-J_q S_q^z (c_{k\uparrow}^\dagger c_{k\uparrow} - c_{k\downarrow}^\dagger c_{k\downarrow})$ is customarily absorbed into H_L.

Considering each magnon carries a unit spin angular momentum of $-\hbar$ (associated with a magnetic moment), the magnonic spin current is equivalent to a spin-down current, which can be obtained by the Heisenberg equation $I_S = \frac{i}{\hbar}\langle [H_{sd}, \sum_q a_q^\dagger a_q]\rangle$. Also, the magnon carries energy so that the magnonic heat current can be calculated through $I_Q = \frac{i}{\hbar}\langle [H_{sd}, \sum_q \hbar\omega_q a_q^\dagger a_q]\rangle$. Following the approach as detailed in the Supplemental Material,[25] we obtain the spin and heat currents from left to right, respectively:

$$I_S = \frac{2S_0}{\hbar}\int_0^\infty d\omega F_R(\omega)\int_{-\infty}^{\infty} d\varepsilon \rho_L(\varepsilon)\mathcal{W}(\varepsilon,\omega), \tag{2}$$

$$I_Q = \frac{2S_0}{\hbar}\int_0^\infty d\omega F_R(\omega)\hbar\omega\int_{-\infty}^{\infty} d\varepsilon \rho_L(\varepsilon)\mathcal{W}(\varepsilon,\omega), \tag{3}$$

with

$$\mathcal{W}(\varepsilon,\omega) = f_{L\downarrow}(\varepsilon+\hbar\omega)[1-f_{L\uparrow}(\varepsilon)][1+N_R(\hbar\omega)] - f_{L\uparrow}(\varepsilon)[1-f_{L\downarrow}(\varepsilon+\hbar\omega)]N_R(\hbar\omega), \tag{4}$$

where $\rho_L(\varepsilon)$ denotes the electron DOS in the left metal side; $F_R(\omega)$ contains the magnon DOS and the electron-magnon coupling strength, which is reminiscent of Eliashberg function[24] in the field of electron-phonon scattering; $f_{L\sigma}(\varepsilon) = [e^{(\varepsilon-\mu_\sigma)/(k_B T_L)}+1]^{-1}$ is the electron distribution with spin σ in the left metal side that is equilibrium at temperature T_L; and $N_R(\hbar\omega) = [e^{\hbar\omega/(k_B T_R)}-1]^{-1}$ denotes the magnon distribution in the right magnetic insulator that is equilibrium at temperature T_R. Equation (3) is reminiscent of the thermal transport induced by interfacial electron-phonon coupling, studied in Ref. 26.

It is clear that the first product $f_{L\downarrow}(1-f_{L\uparrow})(1+N_R)$ in $\mathcal{W}(\varepsilon,\omega)$ describes the down-scattering rate of the occupied spin-down state with high energy $\varepsilon+\hbar\omega$ flipping to the empty spin-up state with low energy ε, accompanied by emitting a magnon with energy $\hbar\omega$ into the right magnet. The second product $f_{L\uparrow}(1-f_{L\downarrow})N_R$ reversely describes the up-scattering rate of the occupied low-energy spin-up state flipping to the empty high-energy spin-down state, with absorbing a magnon from the right magnet. These two spin-flip scattering processes, accompanied by the magnon emission/absorption, conserve not only the energy but also the spin angular momentum.

Following the procedure in the field of electron-phonon scattering, it is customary and legitimate to take a constant bulk electron DOS $\rho_L(\varepsilon) \approx C_L$, because for a good metal the electron DOS is flat and the integral over $d\varepsilon$ converges within a thermal energy $k_B T_L$ around the chemical potential.[24] Therefore, by applying the equality $\int d\varepsilon f_{L\downarrow}(\varepsilon+\hbar\omega)[1-f_{L\uparrow}(\varepsilon)] = (\hbar\omega-\Delta\mu_s)N_L(\hbar\omega-\Delta\mu_s)$, Eqs. (2) and ((3)) finally lead to the formulas

$$I_S = \frac{2S_0C_L}{\hbar}\int_0^\infty d\omega F_R(\omega)(\hbar\omega-\Delta\mu_s) \times [N_L(\hbar\omega-\Delta\mu_s)-N_R(\hbar\omega)]; \tag{5}$$

$$I_Q = \frac{2S_0C_L}{\hbar}\int_0^\infty d\omega F_R(\omega)\hbar\omega(\hbar\omega-\Delta\mu_s) \times [N_L(\hbar\omega-\Delta\mu_s)-N_R(\hbar\omega)]. \tag{6}$$

The expressions are familiar from earlier discussions with interfacial fermion-boson coupling.[24,26–28] We first examine the SPE that generates heat current from merely spin voltage, by taking $T_L = T_R$ but $\Delta\mu_s \neq 0$. Clearly, in the absence of thermal bias, the heat current I_Q is asymmetric under reversing the spin voltage $\Delta\mu_s \to -\Delta\mu_s$ so that *the rectification of SPE exists*. Similarly, the spin current I_S is also asymmetric under revering the spin voltage. Moreover, it is straightforward to prove that $\partial_{\Delta\mu_s} I_Q$ and $\partial_{\Delta\mu_s} I_S$ are both positive, thus *the negative differential SPE and spin conductance are absent*. By taking $\Delta\mu_s = 0$ but $T_L \neq T_R$, we then examine the SSE that generates spin current from merely thermal bias, which is of our central interest. In this way, Eqs. (5) and (6) reduce to Landauer-type formulas,

$$I_S = \frac{2S_0C_L}{\hbar}\int_0^\infty d\omega F_R(\omega)\hbar\omega\,[N_L(\omega)-N_R(\omega)]\,; \tag{7}$$

$$I_Q = \frac{2S_0C_L}{\hbar}\int_0^\infty d\omega F_R(\omega)(\hbar\omega)^2[N_L(\omega)-N_R(\omega)]. \tag{8}$$

Since the temperature dependence only manifests in the Bose-Einstein distributions N_L and N_R, one can readily prove that *in this magnetic interface with constant electron DOS we can never have the rectification and negative differential SSE*.

As we can see from the above derivations, the constant electron DOS is the key assumption that leads to the Landauer-type Eqs. (7) and (8). Therefore, *to obtain the nontrivial rectification and negative differential SSE, we need metallic materials with strongly fluctuated electron DOS*.[29]

For example, considering the typical Lorentzian-type DOS $\rho_L(\varepsilon) = \frac{1}{\pi}\frac{\Gamma}{(\varepsilon-\varepsilon_0)^2+\Gamma^2}$ where the half width Γ is small, the resonant peak of DOS becomes sharp at ε_0, so that Eq. (2) reduces to

$$I_S = \frac{2S_0}{\hbar}\int_0^\infty d\omega F_R(\omega)\mathcal{W}(\varepsilon_0,\omega). \tag{9}$$

It is straightforward to verify that now the rectification and negative differential SSE are present, as illustrated in Fig. 2. Clearly, the spin current profile is asymmetric under reversing thermal bias—so-called rectification of SSE, and when $|\Delta T| \gg 40$ K, the further increasing thermal bias gives an anomalously decreasing spin current—named negative differential SSE. These behaviors are qualitatively similar to

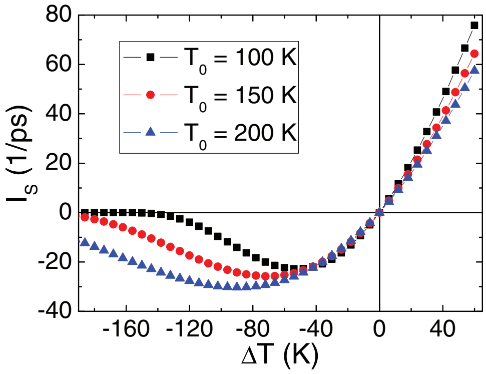

FIG. 2. (Color online) Rectification and negative differential spin Seebeck effect in macroscopic magnetic interface. Spin Seebeck current I_S as a function of the temperature bias ΔT with $T_{L,R} = T_0 \pm \Delta T/2$, for varying T_0 and Lorentzian peak positions: $T_0 = 100$ K, $\varepsilon_0 = 20$ meV; $T_0 = 150$ K, $\varepsilon_0 = 35$ meV; $T_0 = 200$ K, $\varepsilon_0 = 50$ meV. We set $S_0 = 16$, which is comparable with the experimental ones in typical magnetic insulators, cf. Refs. 21,30, and 31. $\mu_0 = 0$ and the Ohmic spectrum $F_R(\omega) = \alpha \frac{\omega}{\omega_c} e^{-\omega/\omega_c}$ is adopted, with $\alpha = 10$, $\omega_c = 50$ meV. Other choices of $F_R(\omega)$ will not change the results qualitatively.

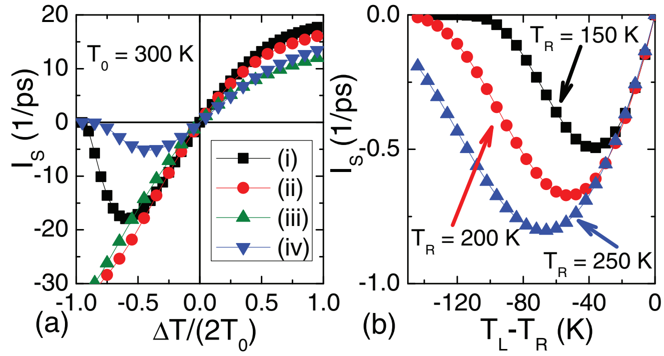

FIG. 3. (Color online) Rectification and negative differential spin Seebeck effect in nanoscale magnetic interface. (a) Magnon spin current I_S as a function of the normalized temperature bias $\Delta T/(2T_0)$ with $T_{L,R} = T_0 \pm \Delta T/2$, for varying $\varepsilon_{\uparrow,\downarrow}$: (i) $\varepsilon_\downarrow = 50$ meV, $\varepsilon_\uparrow = 10$ meV; (ii) $\varepsilon_\downarrow = 50$ meV, $\varepsilon_\uparrow = 0$; (iii) $\varepsilon_\downarrow = 50$ meV, $\varepsilon_\uparrow = -25$ meV; (iv) $\varepsilon_\downarrow = -25$ meV, $\varepsilon_\uparrow = -75$ meV. (b) Magnonic spin Seebeck current as a function of the temperature bias: for $T_R = 150$ K, $\varepsilon_\downarrow = 50$ meV, $\varepsilon_\uparrow = 30$ meV; For $T_R = 200$ K, $\varepsilon_\downarrow = 60$ meV, $\varepsilon_\uparrow = 40$ meV; For $T_R = 250$ K, $\varepsilon_\downarrow = 70$ meV, $\varepsilon_\uparrow = 50$ meV. We set $\Gamma_J = 2.5$ meV and $S_0 = 16$, which is comparable with the experimental ones in typical magnetic insulators; cf. Refs. 21,30, and 31.

those of Eq. (10) for a nanoscale magnetic interface as plotted in Fig. 3, which we will discuss in detail later. In fact, for the Pt/YIG junction used in the spin Seebeck measurement,[4,5] the electron DOS of the bulk Pt is strongly fluctuated and is indeed well described by Lorentzian shapes near the Fermi energy.[32,33]

Even for good metals with smooth DOS, when they are engineered into low-dimensional nanoscale, the electron DOS could become nonsmooth and vary strongly in energy due to the quantum confinement effect and other size-induced many-body interactions. Therefore, for low-dimensional nanoscale magnetic interfaces, we can also retain the intriguing properties of rectification and negative differential SSE. For example, the electronic states in one-dimensional (1D) and 2D tight-binding models possess sharp peaked DOS;[34] see also the strongly fluctuated DOS of carbon nanotubes.[35] In fact, although the good metal Au has a constant DOS near the Fermi energy for bulk gold,[32] when scaled down to the 1D gold chain, its electronic DOS is peaked as Lorentzian shapes.[36] We also note that the longitudinal SSE was recently measured in a thin-film (10 nm) Au/YIG (and Pt/YIG) interface junction.[6] In principle, we can further reduce the thickness of the metallic thin-film Au (or Pt) so that electrons will be confined in two dimensions and the DOS will be steplike functions. As such, we also expect to retain the rectification and negative differential SSE in such a setup when we are far from the small temperature bias regime. In fact, for the 2D free-electron gas, we can obtain its DOS as a step function $\rho_L(\varepsilon) = C_L \Theta(\varepsilon - \mu_0)$. As such, Eq. (2) reduces to $I_S = \frac{2S_0C_L}{\hbar} \int d\omega F_R(\omega) k_B T_L \ln \frac{2e^{\hbar\omega/(k_BT_L)}}{e^{\hbar\omega/(k_BT_L)}+1} [N_L(\omega) - N_R(\omega)]$. It is easy to check that this case retains the rectification and negative differential SSE, similar to the behaviors in Fig. 2.

Let us finally exemplify the rectification and negative differential SSE in a 0D interface as in the situation of quantum point contacts. As such, the electron states near the interface can be simplified as local states on a two-level quantum dot: $H_C = \varepsilon_\uparrow d_\uparrow^\dagger d_\uparrow + \varepsilon_\downarrow d_\downarrow^\dagger d_\downarrow$, where $d_\sigma^\dagger (d_\sigma)$ is the creation (annihilation) operator of the local electron with spin σ and energy ε_σ at the interface [see Fig. 1(b)]. The local electron is freely exchanged with the electron reservoir H_L through the coupling $V_L = \sum_{k\sigma} t_{k\sigma} c_{k\sigma}^\dagger d_\sigma +$ H.c. The exchange coupling to the right magnon reservoir H_R is described by[20–23] $V_R = -\sum_q J_q [S_q^z (d_\uparrow^\dagger d_\uparrow - d_\downarrow^\dagger d_\downarrow) + S_q^- d_\uparrow^\dagger d_\downarrow + S_q^+ d_\downarrow^\dagger d_\uparrow]$. It is clear that the first term just splits the two local energy levels of the interface so that the contribution can be absorbed into a renormalized ε_σ and we have the splitting $\varepsilon_\downarrow > \varepsilon_\uparrow$ generally. In practice, multiple point contacts can be sandwiched between metal and insulating magnets so that multiple transport channels will form in parallel to enhance the transport signal and efficiency [see Fig. 1(c)]. Without loss of generality, we only focus on the single channel case, as an in-principle demonstration of the rectification and negative differential SSE.

Following the same approach as above,[25] we obtain the spin current for the 0D interface system:

$$I_S = \frac{2S_0}{\hbar} \Gamma_J [f_{L\downarrow}(1 - f_{L\uparrow})(1 + N_R) - f_{L\uparrow}(1 - f_{L\downarrow})N_R], \tag{10}$$

where $f_{L\sigma} = [e^{(\varepsilon_\sigma - \mu_\sigma)/(k_BT_L)} + 1]^{-1}$, $N_R = [e^{(\varepsilon_\downarrow - \varepsilon_\uparrow)/(k_BT_R)} - 1]^{-1}$, and $\Gamma_J = 2\pi \sum_q J_q^2 \delta(\varepsilon_\downarrow - \varepsilon_\uparrow - \hbar\omega_q)$ can be regarded as a constant in the wide-band limit. The expression is similar to the spin current of the Lorentzian-DOS case in Eq. (9) and they share qualitatively the same behaviors. The mere difference is that instead of an integral over all magnon spectra there, the spin current here only contains a single-mode resonant transfer ($\hbar\omega_q = \varepsilon_\downarrow - \varepsilon_\uparrow$), which is imposed by the delta function $\delta(\varepsilon_\downarrow - \varepsilon_\uparrow - \hbar\omega_q)$.

The rectification of SSE is clearly displayed in Fig. 3(a) where the thermal-induced spin currents are asymmetric with respect to reversing the temperature bias. In particular, when the two levels are either both above the chemical potential $\varepsilon_\downarrow > \varepsilon_\uparrow > \mu_0$ [see case (i) in Fig. 3(a)] or both below $\mu_0 > \varepsilon_\downarrow > \varepsilon_\uparrow$ [see case (iv) in Fig. 3(a)], we can have the negative differential SSE: When increasing the temperature difference $|T_L - T_R| = |\Delta T|$, instead of observing an increasing spin current as in linear-response regime, we get a decreasing and even vanishing spin current from the right magnetic insulator to the left metallic lead. It is readily proven that when $\varepsilon_\downarrow \geqslant \mu_0 \geqslant \varepsilon_\uparrow$, $\partial I_S/\partial \Delta T$ is always positive so that the negative differential SSE is absent in this parameter regime, as shown by cases (ii) and (iii) in Fig. 3(a). Figure 3(b) shows that the negative differential SSE also exists for a wide range of temperatures.

The emergence of negative differential SSE can be reasoned as follows: when $T_R > T_L$, the thermal bias drives spin current from the right to the left. If near the linear-response regime, it is natural to have a positive differential SSE that lowering T_L will increase the thermal bias which in turn increases the spin current. If we further decrease T_L with assuming two levels are both above (below) the chemical potential, the two states will be both depleted (occupied), which in turn severely suppresses the magnon emission/absorption process that requires the concurrence of one occupied and one empty state. As a consequence, the Seebeck spin conductance will decrease although the thermal bias increases. When the conductance decreases faster than the increasing of the bias, the negative differential SSE emerges.

In summary, we have uncovered the rectification and negative differential SSE in the metal-insulating magnet interface, as a consequence of the strongly fluctuated electron DOS of the metallic lead. We then have exemplified the identification of these intriguing spin Seebeck properties in several typical cases. Since the magnon carries not only spin but also energy, we additionally possess the rectification and negative differential thermal conductance, as in dielectric phononics.[16]

Note that throughout the work, we have ensured the condition $2S_0 \gg \langle a^\dagger a\rangle$ so that the noninteracting magnon picture assumed in the theory is valid. In fact, in a following work we have shown that the high-order magnon-magnon interaction even becomes crucial for the manifestation of asymmetric and negative differential SSE in magnon tunneling junctions,[37] based on which the concept of a functional spin Seebeck transistor is illustrated. If considering the ferromagnetic-paramagnetic phase transition, the rectification and negative differential SSE will be enhanced. Our findings can also be readily generalized to the interfaces of metal-magnetic metal/semiconductor or the ferromagnetic metal-magnetic insulator interfaces. Since recent studies imply the important role of phonon drag in SSE,[8,38–41] taking account of the effect of nonequilibrium phonons on rectification and negative differential SSE would be an interesting topic. By integrating the phononics[16] with spintronics,[9] magnonics,[10] and spin caloritronics,[12] we expect to invigorate more opportunities to achieve the smart control of energy and information in low-dimensional nanodevices.

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*renjie@lanl.gov

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both the magnon DOS and the electron-magnon exchange couplings, the latter of which also depends on electron energy. By considering strong electronic-energy-dependent $F_R(\varepsilon,\omega)$ in some designed magnetic interfacial systems, we are able to have similar rectification and negative differential transport effects.

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