Theory of Huge Thermoelectric Effect Based on Magnon Drag Mechanism: Application to Thin-Film Heusler Alloy

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To understand the unexpectedly high thermoelectric performance observed in the thin-film Heusler alloy Fe2V0.8W0.2Al, we study the magnon drag effect, generated by the tungsten-based impurity band, as a possible source of this enhancement, in analogy to the phonon drag observed in FeSb2. Assuming that the thin-film Heusler alloy has a conduction band integrating with the impurity band, originated by the tungsten substitution, we derive the electrical conductivity \( \sigma \) and the Seebeck coefficient \( S \), for the present alloy, is not sufficiently understood. The contribution of the magnon drag to the Seebeck coefficient has not yet been taken into account. In addition to recent experimental reports, revealing an enhancement of the Seebeck coefficient of various systems through magnetic interactions [4, 6, 8], it has recently been experimentally demonstrated that spin fluctuation enhances the Seebeck coefficient of a doped itinerant ferromagnetic Fe2VAl system [5].

The temperature dependencies of the electrical resistivity and Seebeck coefficient observed in this thin-film Heusler alloy are very similar to those in FeSb2: FeSb2 shows a huge Seebeck coefficient at low temperatures (\( T \sim 10 \text{ K} \)), and at the same temperature, the electrical resistivity changes its temperature dependence to the semiconducting behavior as the temperature decreases [11]. The origin of this huge Seebeck effect observed in FeSb2 has been suggested being caused by a phonon drag, in which acoustic phonons couple with large effective mass electrons in an impurity band [12–14]. From the analogy with FeSb2, the origin of huge Seebeck effect observed in the thin-film Heusler alloy is supposed to be magnon drag related in the context of an impurity band and the conduction band with a large effective electron masses.

The contribution of the magnon drag to the Seebeck effect has been studied experimentally [15–18] and theoretically [19–23] from the 1960s. However, it appears that the magnon drag, related with an impurity band such as for the present alloy, is not sufficiently understood.

In this letter, we study the magnon drag effect with an impurity state to clarify the origin of the huge See-
beck coefficient and PF observed in the thin-film Heusler alloy. Firstly, since the electronic state of the thin-film Heusler alloy has not been entirely understood yet, we assume an electronic state from the viewpoint of a dimensional reduction. Extending the phonon drag theory studied in FeSb2 [14] to the thin-film Heusler alloy, we study the temperature dependence of the electrical resistivity, Seebeck coefficient, and PF related with such an impurity state. We then compare the obtained theoretical results with experimental results to understand the origin of huge thermoelectric effect observed in the thin-film Heusler alloy.

Schematic picture of electronic states.—Firstly, we deduce the electronic state of the thin-film Heusler alloy based on the electronic state of bulk Fe2VAl. Figure 1(a) shows a schematic picture of the electronic state of the bulk Fe2VAl near the Fermi level. It is found [9, 24, 25] that this electronic state is a typical semimetallic state. In the thin-film, it is expected that the bandwidth decreases due to the dimensional reduction. Therefore, we suggest that a band gap appears by the lower dimension in the thin-film Fe2VAl (Fig. 1(b)). When vanadium (V) is replaced by tungsten (W) in this thin-film, it is natural to expect that impurity states appear near the bottom of the conduction band, because the energy level of 5d electrons in W is lower than the 3d energy level in V. Figure 1(c) shows a schematic picture of the electronic state of a) bulk Fe2VAl, b) thin-film Fe2VAl, and c) thin film Fe2(V,W)Al.

![Schematic picture of the electronic state of a) bulk Fe2VAl, b) thin-film Fe2VAl, and c) thin film Fe2(V,W)Al.](image)

FIG. 1. Schematic picture on the electronic state of a) bulk Fe2VAl, b) thin-film Fe2VAl, and c) thin film Fe2(V,W)Al.

In this letter, we study the electrical and thermal transports on the basis of this electronic state shown in Fig. 1(c).

Model Hamiltonian and formulation of Electric and Thermal transports.—To study the magnon drag based on the electronic state shown in Fig. 1(c), we use a following model Hamiltonian [14, 21–23].

\[ H = H_0 + H_W + H_{\text{mag}} + H_{e-\text{mag}}, \]  

where \( H_0 \), \( H_W \), \( H_{\text{mag}} \) and \( H_{e-\text{mag}} \) are Hamiltonians for a ferromagnetic conduction band, W sites, a ferromagnetic magnon, and an electron-magnon interaction, respectively. These Hamiltonians are given as

\[ H_0 = \sum_{k,\sigma} (\epsilon_{k\sigma} - \mu) c_{k\sigma}^\dagger c_{k\sigma}, \quad H_W = V_0 \sum_{i} c_{i\sigma}^\dagger c_{i\sigma}, \quad H_{\text{mag}} = \sum_q h_\omega q^b_{q\uparrow} b_{q\downarrow}, \quad \text{and} \quad H_{e-\text{mag}} = -\frac{1}{\sqrt{N}} \sum_{q,\sigma} \sum_{\mathbf{k}} b_{q\uparrow} c_{\mathbf{k}\uparrow} c_{\mathbf{k}+\mathbf{q}\sigma} + b_{q\downarrow}^\dagger c_{\mathbf{k}\downarrow} c_{\mathbf{k}+\mathbf{q}\sigma}, \]

where \( \epsilon_{k\sigma} \) or \( c_{i\sigma} \) (\( c_{i\sigma}^\dagger \) or \( c_{i\sigma}^\dagger \)) is an annihilation (creation) operator of an electron with the wave number \( \mathbf{k} \) on the \( i \)-th site and spin \( \sigma = \uparrow \downarrow \); \( b_{q\uparrow} \) (\( b_{q\downarrow}^\dagger \)) is an annihilation (creation) operator of a magnon with wave vector \( \mathbf{q} \). \( \epsilon_{k\sigma} \) or \( c_{i\sigma} \) is the energy dispersion in the ferromagnetic state, \( \mu \) is a chemical potential, \( V_0 \) is the strength of a random impurity potential, (\( i \)) is the position of impurities, and \( h_\omega q \) is the energy dispersion of ferromagnetic magnons given by \( h_\omega q = Dq^2 \), where \( D \) is the spin wave stiffness constant. Finally, \( I = J\sqrt{V} \) is the strength of the electron-magnon interaction, where \( J \) and \( V \) are the coupling constant between electron and magnon, and the volume of unit cell, respectively. In this letter, we use the following simple energy dispersion: \( \epsilon_{k\sigma} = \frac{\hbar^2 k^2}{2m^*} - \Delta \), and \( c_{k\sigma} = \frac{\hbar^2 k^2}{2m^*} \), where \( m^* \) is the effective mass of conduction electrons, \( \Delta \) is the energy difference between up spin and down spin electrons to express the ferromagnetic state, which corresponds to d orbitals of Iron (Fe) in FeV0.8W0.2Al [9]. We assume that \( D \) is independent of temperature for simplicity. Because the Fermi energy is located near the bottom of the conduction band or in the impurity band as shown in Fig. 1(c), the valence band is neglected although it will contribute at high temperatures.

To treat the random potential of the W site, we use a self-consistent t-matrix approximation [14, 26–29]. As discussed in Ref [14], we define the retarded Green’s function of electron with spin \( \sigma \) as

\[ G_R^{\sigma}(k, \epsilon) = \frac{1}{\epsilon - \epsilon_{k\sigma} - \Sigma_R^{\sigma}(\epsilon)}, \]

where by the self-consistent t-matrix approximation, a retarded self-energy, \( \Sigma_R^{\sigma}(\epsilon) \), is given as \( \Sigma_R^{\sigma}(\epsilon) = \frac{n_1 V_0}{1 - \frac{\pi}{2} \int_{-\infty}^{\infty} d\epsilon' \Sigma_R^{\sigma}(\epsilon')} \). Here, \( n_1 \) is the concentration of W sites. The density of state (DOS) is obtained by \( D_\sigma(\epsilon) = D_0 \text{Im}[\gamma_\sigma] \), where \( D_0 = \frac{\sqrt{\mu^2 + \epsilon^2}}{2\pi^2} \). Here, \( \epsilon \) is determined by solving the cubic equation: \( y_\sigma^3 - 2y_\sigma + (\frac{\mu^2 + \Delta^2}{\epsilon B}) y_\sigma - \nu = 0 \). We assume that the binding energy of a single W impurity for down spin (up spin) as a first step. It should be noted that the principles calculation shows no spin splitting in 5d orbitals of W [9].

The Fermi energy \( (E_F) \) and the temperature dependence of the chemical potential are determined self-consistently by \( \sum_\sigma \int_{-\infty}^{\infty} f(\epsilon) D_\sigma(\epsilon) d\epsilon = \sum_\sigma \int_{E_F}^{\infty} D_\sigma(\epsilon) d\epsilon = n_1 \), where \( f(\epsilon) \) is the Fermi distribution function defined by \( f(\epsilon) = 1/(e^{\beta(\epsilon-E_F)} + 1) \).

The electrical current \( (J_e) \) and the heat current due to electrons \( (J_{e,\text{ele}}) \), and the heat current due to ferromagnetic magnons \( (J_{e,\text{mag}}) \) are defined as \( J_e = \sum_\sigma \int_{-\infty}^{\infty} f(\epsilon) \epsilon d\epsilon \) and \( J_{e,\text{ele}} = \sum_\sigma \int_{-\infty}^{\infty} f(\epsilon) \epsilon d\epsilon \).
Under an electric field $E$ and temperature gradient $\nabla T$, the electrical current density $j$ is described in the linear response theory as $j = \sigma_{q} \frac{e}{m^*} \nabla T + j_{Q}^{\text{mag}}$, where $\sigma_{q}$ is the electrical conductivity and $j_{Q}^{\text{mag}}$ is the heat current due to magnons.

The transport coefficient $L_{ij}$ due to the electrical currents and $L_{ij}^{\text{ele}}$ owing to the electrical current and the heat current due to electrons are [31–33]:

$$L_{ij} = \lim_{\omega \to 0} \frac{\Phi_{ij}(\omega + i\delta) - \Phi_{ij}(0)}{i\omega + i\delta},$$

where $\omega$ is a frequency of the external field. In the present case, $L_{12}$ contains two components due to $j_{Q}^{\text{ele}}$ and $j_{Q}^{\text{mag}}$, which we refer to $L_{12}^{\text{ele}}$ and $L_{12}^{\text{mag}}$, respectively.

The transport coefficient $L_{11}$ due to the electrical current and $L_{12}^{\text{ele}}$ owing to the electrical current and the heat current due to electrons are [33]

$$L_{11} = \int d\epsilon ( - \frac{\partial f(\epsilon)}{\partial \epsilon} ) \sigma(\epsilon),$$

$$L_{12}^{\text{ele}} = \frac{1}{e} \int d\epsilon ( - \frac{\partial f(\epsilon)}{\partial \epsilon} ) (\epsilon - \mu) \sigma(\epsilon),$$

where $\sigma(\epsilon)$ is the function of electrical conductivity, depending on $\epsilon$. The relaxation time of electrons is included in $\sigma(\epsilon)$. When we use a Green’s function, which is obtained in Eq. (2), $\sigma(\epsilon)$ is given by

$$\sigma(\epsilon) = \sum_{\sigma} \frac{e^{2} \sqrt{m^{*}}}{12\pi^{2} h^{2}} \left( \sqrt{x_{\sigma}^{2} + \Gamma_{\sigma}(\epsilon)^{2}} + x_{\sigma} \right)^{2},$$

where $x_{\sigma} = \epsilon + \Delta \delta_{\tau} - \text{Re} \Sigma_{\tau}^{R}(\epsilon)$ and $\Gamma_{\sigma}(\epsilon) = -\text{Im} \Sigma_{\tau}^{R}(\epsilon)$, respectively. It has to be noted that we consider only the effect of the random potential, given by the self-consistent t-matrix approximation, and neglect the effect of relaxation due to the electron-magnon interaction in the calculation of the electrical conductivity $L_{11}$.

Next, we study the correlation function between the electrical current and the heat current of magnons defined as $\Phi_{12}(\tau) = \frac{1}{\tau} \langle [T_{\tau} j_{1} (\tau)] j_{Q}^{\text{mag}}(0) \rangle$, where $\tau$ is an imaginary time and $T_{\tau}$ denotes the imaginary time ordering operator [22, 23]. By the second order perturbation on the exchange interaction based on the Green’s function of electrons, Eq. (2), the correlation function due to the magnon drag is obtained as

$$\Phi_{12}^{\text{drag}}(\omega) = i\omega \frac{L^{2} e^{2} (m^{*})^{2}}{4\pi^{2} h \Gamma_{\text{mag}}(T)} \int_{0}^{\epsilon_{\text{cut}}} d\epsilon_{q} \frac{\beta_{e} \beta_{q} e^{\beta_{q}}}{(e^{\beta_{q}} - 1)^{2}} \times \int dx f(x) \left[ \frac{L_{1}^{+}}{\Gamma_{x}(x + \epsilon_{q})} - \frac{L_{2}^{+}}{\Gamma_{x}(x)} - \frac{L_{-}^{-}}{\Gamma_{x}(x)} + \frac{L_{-}^{-}}{\Gamma_{x}(x - \epsilon_{q})} \right],$$

where $\Gamma_{\text{mag}}(T)$ and $\epsilon_{\text{cut}}$ are the temperature dependent magnon relaxation rate, and an energy cutoff of magnons, respectively; $L_{1}^{\pm} = \epsilon_{q} \pm \alpha \epsilon - \Delta - \text{Re} \Sigma_{\tau}^{R}(x + \epsilon_{q}) + \text{Re} \Sigma_{\tau}^{R}(x - \epsilon_{q})$ and $L_{2}^{\pm} = \epsilon_{q} \pm \alpha \epsilon - \Delta + \text{Re} \Sigma_{\tau}^{R}(x - \epsilon_{q}) - \text{Re} \Sigma_{\tau}^{R}(x)$. In the supplemental material, we show the derivation of Eq. (7) in detail. Using Eq. (3), the thermoelectric conductivity due to the magnon drag, $L_{12}^{\text{drag}}$, is obtained. The vertex corrections, which are neglected in this letter for simplicity, have been discussed in ref. [23].

**Numerical Results.**— Figure 2(a) shows the density of states for $\tilde{\nu} \equiv \nu/\nu_{0} = 1.1, 2$ and 4. We set $\Delta/\epsilon_{B} = 0.5$. For $\tilde{\nu} = 2$ and 4, the impurity band hybridizes the conduction band naturally, while for $\tilde{\nu} = 1.1$, the impurity band only slightly touches with the conduction band. The Fermi energy is located in $E_{F}/\epsilon_{B} \simeq -1.20$ for $\tilde{\nu} = 1.1$ , $-1.16$ for $\tilde{\nu} = 2$, and $-1.07$ for $\tilde{\nu} = 4.0$. It should be noted that the chemical potential does not show a drastic temperature dependence.

Figure 2(b) shows the temperature dependent electrical resistivity ($\rho = 1/L_{11}$) for $\tilde{\nu} = 1.1, 2$ and 4. Here, $\rho_{0} = 12\pi^{2} h^{2}/e^{2} \sqrt{m^{*}} \epsilon_{B}$. It has to be noted that we introduce the dimensionless phenomenological parameter $\gamma$ to consider additional contributions of the valleys and other unspecified processes to the electrical conductivity. As shown in Fig. 2(b), the resistivity increases gradually, as the temperature decreases from high temperatures, while around $k_{B} T/\epsilon_{B} \simeq 0.5$, the resistivity drastically increases; the resistivity becomes constant at low temperatures. This behavior is a result of the impurity band. We also conclude that the constant resistivity value at low temperatures depends on the impurity concentration.
Next, let us discuss the Seebeck coefficient due to the heat current of electrons, i.e. $S_{\text{ele}} = L_{12}^\text{ele}/T L_{11}$ and the Seebeck coefficient due to the magnon drag, i.e. $S_{\text{drag}} = L_{12}^\text{drag}/T L_{11}$. The inset of Figure 2(b) shows the temperature dependent Seebeck coefficients $S_{\text{ele}}$ for $\nu = 1.1$, 2 and 4. As the impurity concentration decreases, the Seebeck coefficient increases, while the Seebeck coefficient does not show a peak structure. Figure 2(c) shows the temperature dependent term $S_{\text{drag}}$, for $\nu = 1.1$, 2 and 4 and $\alpha = 1.0$. We assume a temperature dependent magnon relaxation rate, $\Gamma_{\text{mag}} = (h/2\tau_0)T$, where $\tau_0$ is a constant. The factor $S_0$ is defined by $S_0 = I^2 (m^*)^{3/2} k_B^2 \tau_0/2e\pi\hbar^2 \sqrt{\epsilon_B}$. Note that the Seebeck coefficient does not depend on $\gamma$. As shown in Fig.2(c), the Seebeck coefficient increases as the impurity concentration decreases. We also find that a peak structure of the temperature dependent Seebeck coefficient appears around $k_B T/\epsilon_B \simeq 0.5$ for $\nu/\nu_0 = 1.1$, 0.6 for $\nu/\nu_0 = 2$ and 0.7 for $\nu/\nu_0 = 4$. Figs. 2(d) show the temperature dependent power factor due to the magnon drag, $PF_{\text{drag}}$, where we define $PF_0 = S_0^2/\rho_0$. The $PF_{\text{drag}}$ traces closely the temperature dependent Seebeck coefficient as shown in Figs. 2(c) regarding several impurity concentrations, while we find that the peak temperature of $PF_{\text{drag}}$ is higher than that of Seebeck coefficient, because of the distinct decrease of the electrical resistivity. It should be noted that the temperature dependences of $S_{\text{drag}}$ and $PF_{\text{drag}}$ are insensitive to $\alpha$, while these values strongly depend on $\alpha$ (See the supplemental material).

Discussion: Comparison with experiments.— Here, we compare the obtained theoretical results with the experimental results of the thin-film Heusler alloy. Since there are no experimental data on theoretical parameters, we have chosen a set of the reasonable values: $\epsilon_B/k_B = 300 K$, $m^*/m_0 = 10$, $J/k_B = 1000 K$, $V = 10^{-27} m^3$, and $\gamma = 10$. It should be noted that the large effective mass is due to the large density of states of the conduction band as shown from the first principles DFT calculations [9]; then, the impurity concentration ($n_i$) is of the order of $10^{27} m^{-3}$ for $\nu = 1 \sim 4$, which is consistent with the concentration of W in Fe$_2$VAl. Here, we set the life time of magnon as $\tau = \tau_0/T \sim 10^{-14} s$ at $T = 300 K$. This value is reasonable for a ferromagnetic metal [34].

Using these parameters, $\nu/\nu_0 = 4$ and $\alpha = 1.0$, the temperature dependent electrical resistivity and Seebeck coefficient due to the magnon drag, as well as the power factor are displayed in Fig. 3. We find that the electrical resistivity attains $\rho \simeq 1000 \mu\Omega cm$ at $T \sim 300 K$; we also find that the Seebeck coefficient due to the magnon drag exhibits a peak structure, with $S_{\text{max}} \sim -500 \mu V/K$ at $T \sim 300 K$. The power factor reaches $PF \sim 60 \text{mW/m K}^2$ around $T \sim 400 K$. Since these theoretical results are similar to the experimental results, we presume that the origin of the huge Seebeck coefficient and the large $PF$ observed experimentally for the thin-film Heusler alloy is likely due to a magnon drag, related to the tungsten-based impurity band.

Finally, we comment on the life time of magnons. In this letter, we used a simple temperature dependent life time of magnons. However, the life time is expected to be very complicated in a real material, because it is derived from many kinds of scattering mechanisms such as impurity scattering, magnon-electron, magnon-magnon, magnon-phonon interactions related with (without) the Umklapp process, and so on. The understanding of these microscopic mechanisms for the life time of magnon is a future problem.

Conclusion.— We studied the origin of the large Seebeck coefficient and unprecedented large $PF$ observed in the thin-film Heusler alloy FeV$_{0.8}$W$_{0.2}$Al on the basis of the linear response theory. Assuming that this thin-film alloy has a conduction band integrating with the impurity band originated from the W substitution, and by extending the microscopic phonon drag theory observed in FeSb$_2$, we derived $L_{11}$ based on the self-consistent $t$-matrix approximation and $L_{12}$ due to the magnon drag. As a result, we found that the theoretical results of the Seebeck coefficient and $PF$ are in agreement with the experimental ones. Therefore, we concluded that the origin of these striking thermoelectric properties is likely due to the magnon drag related with the W-based impurity band.

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Supplemental Materials

1: DERIVATION OF EQ. (7)

In this supplemental material, we derive eq. (7) on the basis of the linear response theory. By the second order perturbation of \( H_{\text{e-mag}} \), the lowest order correlation function is given as

\[
\Phi_{12}(q = 0, \tau, 0) = \frac{1}{2V} \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \langle T_\tau [H_{\text{e-mag}}(\tau_1)H_{\text{e-mag}}(\tau_2)j_\tau(\tau)j_Q^{\text{mag}}(0)] \rangle. \tag{S1}
\]

By Wick’s theorem, \( \langle T_\tau [\cdots] \rangle \) is expressed as

\[
\langle T_\tau [H_{\text{e-mag}}(\tau_1)H_{\text{e-mag}}(\tau_2)j_\tau(\tau)j_Q^{\text{mag}}(0)] \rangle = \frac{I^2}{V} \sum_{k,q} \frac{\epsilon_h k_x}{m} \frac{2D^2 q_x q_y}{\hbar} \times [G_\uparrow(k, \tau - \tau_1)G_\downarrow(k + q, \tau_1 - \tau_2)G_\uparrow(k, \tau_2 - \tau)D(q, -\tau_1)D(q, \tau_2) + G_\downarrow(k, \tau_1 - \tau)G_\uparrow(k, \tau - \tau_2)G_\downarrow(k - q, \tau_2 - \tau)D(q, -\tau_1)D(q, \tau_2) + G_\uparrow(k - q, \tau_1 - \tau_2)G_\downarrow(k, \tau_2 - \tau)G_\downarrow(k, \tau - \tau_1)D(q, \tau_1)D(q, -\tau_2) + G_\downarrow(k + q, \tau_2 - \tau_1)G_\uparrow(k, \tau_1 - \tau)G_\downarrow(k, \tau - \tau_2)D(q, \tau_1)D(q, -\tau_2) ], \tag{S2}
\]

where the Green’s functions for electron and magnon are defined as

\[
G_\sigma(k, \tau) = \langle T_\tau [c_{k\sigma}(\tau)c_{k\sigma}^\dagger(0)] \rangle, \tag{S3}
\]

\[
D(q, \tau) = \langle T_\tau [b_q(\tau)b_q^\dagger(0)] \rangle. \tag{S4}
\]

By the Fourier transformations of

\[
\Phi_{12}(i\omega_\lambda) = \int d\tau e^{i\lambda \lambda \tau} \Phi_{12}(\tau), \tag{S5}
\]

\[
G_\sigma(k, \tau) = \frac{1}{\beta} \sum_n e^{-\epsilon_m \tau} G_\sigma(k, \tau), \tag{S6}
\]

\[
D(q, \tau) = \frac{1}{\beta} \sum_m e^{-i\omega_m \tau} D(q, \tau), \tag{S7}
\]

where \( \omega_\lambda, \epsilon_n \) and \( \omega_m \) are the Matsubara frequencies with \( \omega_\lambda = 2\lambda \pi k_B T, \epsilon_n = (2n + 1)\pi k_B T, \) and \( \omega_m = 2m \pi k_B T \) with \( \lambda, n \) and \( m \) being the integers, the correlation function is given as

\[
\Phi_{12}(i\omega_\lambda) = \frac{I^2}{2V^2} \sum_{k,q} \frac{\epsilon_h k_x}{m} \frac{2D^2 q_x q_y}{\hbar} \frac{2}{\beta^2} [G_\uparrow(k, i\epsilon_n)G_\downarrow(k + q, i\epsilon_n + i\omega_m + i\omega_\lambda)G_\uparrow(k, i\epsilon_n + i\omega_\lambda) + G_\downarrow(k, i\epsilon_n)G_\uparrow(k - q, i\epsilon_n - i\omega_m)G_\downarrow(k, i\epsilon_n + i\omega_\lambda)] D(q, i\omega_m)D(q, i\omega_m + i\omega_\lambda). \tag{S8}
\]

Here, the thermal Green’s functions for electron and magnon are defined as

\[
G_\sigma(k, i\epsilon_n) = \frac{1}{i\epsilon_n - (\epsilon_{k,\sigma} - \mu) - \text{sgn}(\epsilon_n)\Sigma(\epsilon_n)}, \tag{S9}
\]

\[
D(q, i\omega_m) = \frac{1}{i\omega_m - h\omega_q - \text{sgn}(\omega_m)\Gamma_{\text{mag}}(T)}. \tag{S10}
\]

When \( \omega_\lambda > 0 \) and we use the region of \( \omega_m < 0 \), by take the sum on the integers of \( n \) and \( m \), the correlation function is calculated as

\[
\Phi_{12}(\omega_\lambda) = \frac{I^2}{2V^2} \sum_{k,q} \frac{\epsilon_h k_x}{m} \frac{2D^2 q_x q_y}{\hbar} \int_{-\infty}^{\infty} \frac{n(y)}{2\pi i} \left[ -[F_A(k, q, y, i\omega_\lambda) + F_B(k, q, y, i\omega_\lambda)] D^A(q, y)D^R(q, y + i\omega_\lambda) + [F_A(k, q, y - i\omega_\lambda, i\omega_\lambda) + F_B(k, q, y - i\omega_\lambda, i\omega_\lambda)] D^A(q, y - i\omega_\lambda)D^R(q, y) \right]. \tag{S11}
\]
where \( n(y) = 1/(e^{βy} - 1) \) and \( R(A) \) indicates the retarded (advanced) Green’s function. \( F_A(k, q, y, iω_n) \) and \( F_B(k, q, y, iω_n) \) are

\[
F_A(k, q, y, iω_n) = -\int_{-∞}^{∞} \frac{f(x)}{2πi} dx \left[ G^R(\mathbf{k}, x) - G^A(\mathbf{k}, x) \right] G^R_\perp(\mathbf{k} + q, x + iω_n + y) G^R_\perp(\mathbf{k}, x + iω_n)
+ G^A_\perp(\mathbf{k}, x - y - iω_n) G^R(\mathbf{k} + q, x) - G^A(\mathbf{k} + q, x) \right] G^R_\perp(\mathbf{k}, x - y) \right] + G^A_\perp(\mathbf{k}, x - iω_n) G^A(\mathbf{k} + q, x + y) \left[ G^R_\perp(\mathbf{k}, x) - G^A_\perp(\mathbf{k}, x) \right]. \tag{S12}
\]

\[
F_B(k, q, y, iω_n) = -\int_{-∞}^{∞} \frac{f(x)}{2πi} dx \left[ G^R(\mathbf{k}, x - y - iω_n) G^R(\mathbf{k} - q, x - y) G^R(\mathbf{k}, x - iω_n)
+ G^A_\perp(\mathbf{k} + q, x + y) \left[ G^R_\perp(\mathbf{k} - q, x) - G^A_\perp(\mathbf{k} - q, x) \right] G^R_\perp(\mathbf{k}, x + y + iω_n)
+ G^A_\perp(\mathbf{k}, x - iω_n) G^A(\mathbf{k} - q, x - y - iω_n) \left[ G^R_\perp(\mathbf{k}, x) - G^A_\perp(\mathbf{k}, x) \right]. \tag{S13}
\]

where \( f(x) = 1/(e^{β(x-μ)} + 1) \) and \( G^R(\mathbf{k}, x) \) is given in Eq. (??), and \( G^A = (G^R)^* \). It should be noted that we transformed the variable \( x + iμ \to x \).

Using the analytic continuation \( (iω_n → hω + iδ) \), and \( D^RDA \simeq πδ(y - hω_n)/Γ_{mag} \), the correlation function becomes

\[
Φ_{12}(hω + iδ) = \frac{I^2}{2V^2} \sum_{k, q} \frac{e^{ikhk_x}}{m} \frac{2D^2q^2q_x}{h^4} \frac{2}{2πi} \Gamma_{mag}(T) (e^{βhω_n} - 1)^2 \left[ A(\mathbf{k}, q, +, hω_q) + A(\mathbf{k}, -q, -, -hω_q) \right], \tag{S14}
\]

where \( A(\mathbf{k}, q, σ, hω_q) \) is given as

\[
A(\mathbf{k}, q, σ, hω_q) = -\int_{-∞}^{∞} \frac{f(x)}{2πi} dx \left[ G^R_σ(\mathbf{k}, x) - G^A_σ(\mathbf{k}, x) \right] G^R_σ(\mathbf{k} + q, x + hω_q) G^R_σ(\mathbf{k}, x)
+ G^A_σ(\mathbf{k}, x - hω_q) \left[ G^R_σ(\mathbf{k} + q, x) - G^A_σ(\mathbf{k} + q, x) \right] G^R_σ(\mathbf{k}, x - hω_q)
+ G^A_σ(\mathbf{k}, x - iω_n) G^A_σ(\mathbf{k} - q, x - y - iω_n) \left[ G^R_σ(\mathbf{k}, x) - G^A_σ(\mathbf{k}, x) \right]. \tag{S15}
\]

where \( σ = (+(-) \) corresponds to \( ↑ (↓) \).

When we neglect \( G^R G^R G^R \) and \( G^A G^A G^A \), and we use the following approximations

\[
G^R_σ(\mathbf{k}, x) G^A_σ(\mathbf{k}, x) = \frac{π}{Γ_σ(x)} δ(x - ϵ_κσ - ReΣ^R_σ(x)), \tag{S16}
\]

\[
G^R_σ(\mathbf{k}, x) - G^A_σ(\mathbf{k}, x) = -2πi δ(x - ϵ_κσ - ReΣ^R_σ(x)), \tag{S17}
\]

\( A(\mathbf{k}, q, σ, hω_q) \) is approximated as

\[
A(\mathbf{k}, q, σ, hω_q) \simeq -\int_{-∞}^{∞} f(x) dx \left[ \frac{π}{Γ_σ(x)} δ(x - ϵ_κσ - ReΣ^R_σ(x)) δ(x + hω_q - ϵ_{k+q,-σ} - ReΣ^R_{-σ}(x + hω_q))
- \frac{π}{Γ_σ(x - hω_q - ϵ_κσ - ReΣ^R_{-σ}(x - hω_q)) δ(x - ϵ_{k+q,-σ} - ReΣ^R_{-σ}(x))} \right]. \tag{S18}
\]

To perform the integrals on \( k, q, x \), we set a new coordinate as shown in Fig. S1. In this coordinate, \( q_x \) and \( k_x \) are

\[
q_x = q \cos θ \cos Φ + q \sin θ \sin Φ \cos Ω, \tag{S19}
\]

\[
k_x = k \cos θ. \tag{S20}
\]

By integrating on \( k, θ, φ, Θ, Ω \), the correlation function becomes Eq. (7).

II: TEMPERATURE DEPENDENCE OF SEEBECK COEFFICIENT AND POWER FACTOR DUE TO THE MAGNON DRAG: THE EFFECT OF \( α \)

Figure S2 shows the temperature dependent Seebeck coefficient and power factor due to the magnon drag. The amplitude of \( α \) is set as \( α = 0.5 \) for (a) and (d), \( α = 1.0 \) for (b) and (d), and \( α = 5 \) for (c) and (e), respectively. We find that the temperature dependences of Seebeck coefficient and power factor are insensitive to \( α \), while the amplitude of Seebeck coefficient and power factor strongly depend on \( α \): As \( α \) increases, the Seebeck coefficient and the power factor increase.
FIG. S1. Coordination to perform the integrals. $\mathbf{q}_\perp$ and $\mathbf{x}_\perp$ are the components of $\mathbf{q}$ and $\mathbf{x}$ perpendicular to $k$.

FIG. S2. Temperature dependent Seebeck coefficient and power factor due to the magnon drag. We set the parameters as $\alpha = 0.5$ for (a) and (d), $\alpha = 1.0$ for (b) and (d), and $\alpha = 5$ for (c) and (e).