Violation of the magnonic Wiedemann-Franz law in the strong nonlinear regime

Kouki Nakata,[1] Yuichi Ohnuma,[2] and Se Kwon Kim[3]

[1]*Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan*

[2]*Research Center for Advanced Science and Technology (RCAST), The University of Tokyo, Meguro, Tokyo 153-8904, Japan*

[3]*Department of Physics, Korea Advanced Institute of Science and Technology, Daejeon 34141, Republic of Korea*

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The celebrated Wiedemann-Franz (WF) law, which governs the relation between charge and heat transport traces back to the experimental discovery in 1853 by Wiedemann and Franz. Despite the fundamental difference of the quantum-statistical properties between fermions and bosons, the linear-in-T behavior of the WF law at low temperatures has recently been found to be the universal property by the discovery of the WF law for magnon transport. However, the WF law is for the linear response, and whether or not the universal law is valid even in the nonlinear regime of Bose systems remains an open issue. Here we provide a solution to this fundamental challenge. We show that the ratio of the thermal to spin transport coefficient of magnons in topologically trivial insulating magnets exhibits a different behavior from the linear response and the universal law breaks down in the strong nonlinear regime. This finding is within experimental reach with current device and measurement technologies. Our discovery is the key ingredient in magnon-based spintronics, in the evaluation of the figure of merit for thermomagnetic conversion elements of spintronics devices.

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I. INTRODUCTION

The research on thermoelectric properties of materials started more than two centuries ago and it has a long history. The celebrated Wiedemann-Franz (WF) law, which dictates the linear relation between charge and heat transport traces back to the experimental discovery in 1853 by Wiedemann and Franz [1] that the ratio of the thermal κ to electrical conductivity σ of several metals reduces to approximately the same value for a fixed temperature. Lorenz established that this ratio is linear in the absolute temperature T and the proportionality constant takes a material-independent value [2]. Using quantum theory on solids, Sommerfeld appropriately derived the universal constant, dubbed the Lorenz number $\mathcal{L}_\mathrm{e}$, which is independent of any material parameters [3]. Thus the WF law has been formulated [4,5] at low temperature in the form of

$$\frac{\kappa}{\sigma} \overset{\rightarrow}{=} \mathcal{L}_\mathrm{e} T,$$

$$\mathcal{L}_\mathrm{e} := \frac{\pi^2}{3}\left(\frac{k_\mathrm{B}}{e}\right)^2,$$

where e is the elementary charge and k_B the Boltzmann constant. This universal law characterizes the figure of merit for thermoelectric conversion elements and has been playing a central role in electronics.

Toward efficient transmission of information that goes beyond what is offered by conventional electronics, the last two decades have seen a rapid development of spintronics, aiming at utilizing another degrees of freedom of electrons, spins, by means of spin transport [6,7]. For this holy grail, it is desirable to formulate the spin analog of the WF law because the law is expected to be a promising building block in spintronics, in the evaluation of the figure of merit for thermomagnetic conversion elements of spintronics devices.[1] Then we have established the magnon analog of the WF law, namely, the WF law for magnon transport, in ferromagnets and antiferromagnets [8–10]. Magnons are bosonic magnetic excitations, i.e., the quantized spin-waves. Since magnons carry the spin angular momentum, spin currents are generated in the absence of charge currents when magnons propagate in insulating magnets. Thus the WF law for magnon transport, dubbed the magnonic WF law, is defined as the ratio of the thermal conductivity K_1 within the linear response regime to spin conductivity G of magnons. In the bulk of topologically trivial insulating magnets at sufficiently low temperatures compared to the magnon energy gap, the ratio reduces to [9]

$$\frac{K_1}{G} \overset{\rightarrow}{=} \mathcal{L}_1 T, \tag{1a}$$

$$\mathcal{L}_1 := \frac{5}{2}\left(\frac{k_\mathrm{B}}{g\mu_\mathrm{B}}\right)^2, \tag{1b}$$

where g is the g factor of the constituent spins and μ_B the Bohr magneton. The thermomagnetic ratio is linear in temperature. In analogy to charge transport in metals, we refer to this behavior as the magnonic WF law. The constant $\mathcal{L}_1$ analogous to the Lorenz number, i.e., the magnetic Lorenz number of

[1]We refer to the spin analog of the figure of merit for thermoelectric conversion elements as that for thermomagnetic ones Z_s, i.e., $Z_\mathrm{s} := \mathcal{S}^2 G/K$, where $\mathcal{S}$, G, and K are the spin Seebeck coefficient, the spin conductivity, and the thermal conductivity of magnons, respectively.

magnons, is independent of any material parameters except the g factor, which is material specific. The role of the charge e is played by $g\mu_{\rm B}$.

Magnons are bosonic excitations, while electrons are fermions. Still, remarkably, the magnonic WF law exhibits the same linear-in-T behavior at low temperatures as the one for electronic transport despite the fact that the quantum-statistical properties of bosons and fermions are fundamentally different, particularly in the low temperature regime where quantum effects dominate. In that sense, the linear-in-T behavior of the WF law is found to be the universal property.

As spintronics technologies develop, there has recently been a growing interest in the properties of the nonlinear response [11–19]. However, the existing theory for the magnonic WF law is intended to be applied only for the linear response regime. Whether or not the universal law is valid even in the nonlinear regime remains an open issue. In this paper, we provide a solution to this fundamental challenge by using the Boltzmann equation. This is the main aim of this paper.

This paper is organized as follows. In Sec. II we investigate longitudinal thermal transport of magnons in the bulk of insulating magnets, and study the validity and violation of the magnonic WF law in the nonlinear response regime. Then, we remark on several issues in Sec. III. Finally, we give an estimate for the experimental feasibility in Sec. IV and summarize in Sec. V. Technical details are described in the Appendix.

II. NONLINEAR THERMAL TRANSPORT

We consider longitudinal transport of magnons in the bulk of a topologically trivial three-dimensional insulating magnet,[2] subjected to a temperature gradient, where the magnon of the energy dispersion relation $\epsilon_{\mathbf{k}} = Dk^2 + \Delta$ with the group velocity $\mathbf{v}_{\mathbf{k}} = \partial\epsilon_{\mathbf{k}}/(\partial\hbar\mathbf{k})$ carries the spin angular momentum -1 in units of the reduced Planck constant $\hbar$: In which $k := |\mathbf{k}|$ denotes the magnitude of the wavenumber $\mathbf{k} = (k_x, k_y, k_z)$, D represents the spin stiffness constant, and Δ is the magnon energy gap, e.g., due to an external magnetic field, a spin anisotropy, etc. Throughout this paper assuming that the magnon energy gap takes a nonzero value $\Delta \neq 0$ and that the nonequilibrium Bose distribution function of magnons $f_{\mathbf{k}}$ is described by the Boltzmann equation within the quasiparticle approximation, we study magnon transport at low temperatures using a relaxation time approximation.[3] Note that if one assumed a magnon energy dispersion including the k-linear term, nonreciprocal responses [20], e.g., the Doppler shift of spin-waves [21], could arise in certain materials with broken inversion symmetry, which is outside the scope of this paper: See Ref. [22] for the generation of the magnon nonreciprocity in the absence of a finite energy gap, i.e., nonreciprocal transport of gapless spin-waves.

The applied temperature gradient $\nabla T = (\text{const.})$ induces a magnonic spin current along the longitudinal direction, which leads to an accumulation of magnons at the boundaries and builds up a nonuniform magnetization in the magnet. This magnetization gradient plays a role of an effective magnetic field gradient ∇B, which works as the gradient of a nonequilibrium spin chemical potential [23–26], and drives magnon currents [9,27–29]. Therefore, magnon transport subjected to the temperature gradient along the x axis $\partial_x T = (\text{const.})$ is characterized including the nonlinear response by the longitudinal transport coefficient L_{ij} for $i \in \mathbb{N}$ and $j \in \mathbb{N}$ as

$$\begin{pmatrix} j_x^{\rm s} \\ j_x^{\rm E} \end{pmatrix} = \begin{pmatrix} L_{11} & L_{12} & L_{13} & L_{14} & L_{15} & L_{16} \\ L_{21} & L_{22} & L_{23} & L_{24} & L_{25} & L_{26} \end{pmatrix} \times \begin{pmatrix} \partial_x B \\ -\frac{\partial_x T}{T} \\ (\partial_x B)^2 \\ -\left(\frac{\partial_x T}{T}\right)(\partial_x B) \\ \left(\frac{\partial_x T}{T}\right)^2 \\ -\left(\frac{\partial_x T}{T}\right)^3 \end{pmatrix}, \tag{2}$$

where the spin current density $\mathbf{j}^{\rm s} = (j_x^{\rm s}, j_y^{\rm s}, j_z^{\rm s})$ and the energy current density $\mathbf{j}^{\rm E} = (j_x^{\rm E}, j_y^{\rm E}, j_z^{\rm E})$ are defined as [9]

$$\mathbf{j}^{\rm s} := -\int \frac{d^3\mathbf{k}}{(2\pi)^3} g\mu_{\rm B} \mathbf{v}_{\mathbf{k}} f_{\mathbf{k}}, \tag{3a}$$

$$\mathbf{j}^{\rm E} := \int \frac{d^3\mathbf{k}}{(2\pi)^3} \epsilon_{\mathbf{k}} \mathbf{v}_{\mathbf{k}} f_{\mathbf{k}}, \tag{3b}$$

respectively. The transport coefficient L_{11} is identified with the spin conductivity of magnons [28] $G := L_{11}$. In contrast to the junction system [8,30], the second-order response vanishes[4] in the bulk of topologically trivial insulating magnets due to the property of the odd function in k_x:[3]

$$L_{13} = L_{14} = L_{15} = L_{23} = L_{24} = L_{25} = 0. \tag{4}$$

Note that since the third-order response to the temperature gradient $O((\partial_x T)^3)$ can become dominant at sufficiently low temperatures compared to the magnon gap,[3] we neglect the other third-order terms such as $O((\partial_x T)^2(\partial_x B)^1)$, $O((\partial_x T)^1(\partial_x B)^2)$, and $O((\partial_x B)^3)$.

In analogy to charge transport [5] and using Eq. (2), we formulate thermal transport of magnons in the nonlinear response regime. Under the applied temperature gradient, the magnonic spin current is generated and this leads to an accumulation of magnons at the boundaries. Consequently, the nonuniform magnetization is developed and this effective magnetic field gradient $\partial_x B$ generates a counter-current of magnons. Then, the system reaches a stationary state such that in- and out-flowing magnonic spin currents balance each other $j_x^{\rm s} = 0$, which results in $\partial_x B = \partial_x B^*$ with

$$\partial_x B^* := \frac{L_{12}}{L_{11}} \frac{\partial_x T}{T} + \frac{L_{16}}{L_{11}} \left(\frac{\partial_x T}{T}\right)^3. \tag{5}$$

Thus the thermal conductivity is measured. This effective magnetic field gradient in the new quasiequilibrium state $\partial_x B^*$ brings the nonequilibrium spin chemical potential [23], being

[2] We refer to the magnet where Berry curvatures are zero as the topologically trivial magnet.

[3] See the Appendix for details.

[4] This result changes in general if one assumes a magnon energy dispersion with the k-linear term.

peculiar to the system out of equilibrium, and contributes to the thermal conductivity associated with the heat current density [9,23], $j_x^Q = L_{21}\partial_x B^* - L_{22}\partial_x T/T - L_{26}(\partial_x T/T)^3$, as

$$j_x^Q = -K_1\partial_x T - K_2(\partial_x T)^2 - K_3(\partial_x T)^3, \tag{6}$$

where

$$K_1 := \frac{1}{T}\left(L_{22} - \frac{L_{12}L_{21}}{L_{11}}\right), \tag{7a}$$

$$K_2 = 0, \tag{7b}$$

$$K_3 := \frac{1}{T^3}\left(L_{26} - \frac{L_{16}L_{21}}{L_{11}}\right), \tag{7c}$$

and K_1 represents the thermal conductivity in the linear response regime and $K_{2(3)}$ is the thermal transport coefficient of the second-order (third-order) nonlinear response. Note that in the stationary state under the applied temperature gradient, the heat current density j_x^Q is different from the energy current density j_x^{E} [9,23,31],

$$j_x^Q \neq j_x^{\mathrm{E}}, \tag{8}$$

in that

$$j_x^Q = L_{21}\partial_x B^* - L_{22}\frac{\partial_x T}{T} - L_{26}\left(\frac{\partial_x T}{T}\right)^3, \tag{9a}$$

$$j_x^{\mathrm{E}} = 0 - L_{22}\frac{\partial_x T}{T} - L_{26}\left(\frac{\partial_x T}{T}\right)^3. \tag{9b}$$

We remark that if one wrongly omits the contribution of $\partial_x B^*$ associated with the counter-current and identifies j_x^{E} as the heat current density in theoretical calculation, the ratio of the thermal to spin conductivity would not obey the magnonic WF law, breaking the linear-in-T behavior, even in the linear response regime [9]. Note that for thermal transport of electrons in metals, the contribution of the counter-current is strongly suppressed by the sharp Fermi surface of fermions at temperatures $k_{\mathrm{B}}T$, which is much smaller than the Fermi energy even at room temperature. This is the crucial difference in the thermal conductivity between magnons and electrons, i.e., bosons and fermions, respectively.

We evaluate the thermal transport coefficient of the third-order nonlinear response K_3. The Onsager relation holds $L_{12} = L_{21}$ and at low temperatures $k_{\mathrm{B}}T \ll \Delta$, it reduces to [9] $L_{12}/L_{11} = L_{21}/L_{11} \overset{\rightarrow}{=} -\Delta/(g\mu_{\mathrm{B}})$. Thus, the thermal transport coefficient of the third-order nonlinear response at low temperature is recast into

$$K_3 \overset{\rightarrow}{=} \frac{1}{T^3}\left(L_{26} + \frac{\Delta}{g\mu_{\mathrm{B}}}L_{16}\right). \tag{10}$$

A straightforward calculation using the Boltzmann equation provides the transport coefficients of the nonlinear response at low temperatures as[3]

$$L_{16} \overset{\rightarrow}{=} -g\mu_{\mathrm{B}}F\mathrm{e}^{-b}\left[\frac{\Delta^3}{(\beta D)^{7/2}}A_3 + \frac{3D\Delta^2}{(\beta D)^{9/2}}A_4 + O(T^{11/2})\right], \tag{11a}$$

$$L_{26} \overset{\rightarrow}{=} F\mathrm{e}^{-b}\left[\frac{\Delta^4}{(\beta D)^{7/2}}A_3 + \frac{4D\Delta^3}{(\beta D)^{9/2}}A_4 + O(T^{11/2})\right], \tag{11b}$$

where $A_n := \sqrt{\pi}(2n)!/[2^{2n+1}(n!)]$ is the Gaussian integral for $n \in \mathbb{N}$, $F := (2D/\hbar)^4[\tau^3\beta^3/(10\pi^2)]$, the inverse temperature $\beta := 1/(k_{\mathrm{B}}T)$, $b := \beta\Delta$, and the relaxation time τ. Note that at low temperatures the relaxation time takes a constant value of being temperature-independent: At sufficiently low temperatures, the effect of magnon-magnon interactions and that of phonons are negligibly small, and impurity scattering makes a major contribution to the relaxation. Under the assumption that impurities are dilute and scattering is elastic and spatially isotopic with the impurity potential localized in space, the relaxation time at low temperatures reduces to [31] $\tau \overset{\rightarrow}{=} \hbar/(2\alpha\Delta)$, where α is the Gilbert damping constant. Since the Gilbert damping constant is little influenced by temperature [32] (i.e., the temperature dependence is negligibly small), the relaxation time at low temperatures takes a constant value of being temperature-independent.

At low temperatures the thermal transport coefficient of the third-order nonlinear response reduces to

$$K_3 \overset{\rightarrow}{=} \frac{1}{T^3}F\mathrm{e}^{-b}\frac{D\Delta^3}{(\beta D)^{9/2}}A_4, \tag{12}$$

and the spin conductivity to $G \overset{\rightarrow}{=} (g\mu_{\mathrm{B}})^2\mathrm{e}^{-b}\tau(k_{\mathrm{B}}T)^{3/2}/(4\pi^{3/2}\hbar^2\sqrt{D})$. Thus, we find at low temperatures $k_{\mathrm{B}}T \ll \Delta$ that the ratio of the thermal transport coefficient of the third-order nonlinear response to the spin conductivity is given as

$$\frac{K_3}{G} \overset{\rightarrow}{=} \mathcal{L}_3\frac{1}{T^3}, \tag{13a}$$

$$\mathcal{L}_3 := \frac{32\tau^2 D\Delta^3 A_4}{5\sqrt{\pi}\hbar^2(g\mu_{\mathrm{B}})^2}. \tag{13b}$$

The thermomagnetic ratio in the nonlinear regime is proportional to $1/T^3$ [Eq. (13a)]. This is in contrast to the one in the linear response regime K_1/G, which exhibits the linear-in-T behavior [Eq. (1a)]. The proportionality constant $\mathcal{L}_3$, which is independent of temperature, is less universal than the magnetic Lorenz number $\mathcal{L}_1$ [Eq. (1b)] in that the constant depends on other material parameters as well as the g factor, such as the spin exchange interaction, the spin anisotropy, the spin length, the lattice constant, etc. Note that each transport coefficient does not diverge even at low temperature $T \overset{\rightarrow}{=} 0$ as $K_3 \propto \mathrm{e}^{-b}/T^{3/2} \overset{\rightarrow}{=} 0$ and $G \propto T^{3/2}\mathrm{e}^{-b} \overset{\rightarrow}{=} 0$.

Finally, we discuss the validity and violation of the magnonic WF law in the nonlinear response regime. The magnonic WF law is originally for the linear response [Eq. (1a)]. Instead of the thermal conductivity K_1, it is recast in terms of the heat current density $j_x^Q = -K_1\partial_x T$ as $j_x^Q/G = -[\mathcal{L}_1(\partial_x T)]T$, which states that the ratio of the heat current density to the magnonic spin conductivity G is linear in temperature for a fixed temperature gradient $(\partial_x T) = (\text{const.})$. This is the magnonic WF law in terms of the heat current density. Since the heat current density includes the nonlinear response [Eq. (6)], it can be concluded that the magnonic WF law does hold even in the nonlinear response regime if the ratio of the heat current density to the magnonic spin conductivity G exhibits the linear-in-T behavior for the fixed temperature gradient: This is the criterion for the magnonic WF law in the nonlinear response regime.

From Eq. (6) the ratio of the heat current density including the nonlinear response to the magnonic spin conductivity G

becomes

$$\frac{j_x^Q}{G} = -\frac{K_1}{G}(\partial_x T) - \frac{K_2}{G}(\partial_x T)^2 - \frac{K_3}{G}(\partial_x T)^3. \quad (14)$$

Since the thermal transport coefficient of the second-order nonlinear response vanishes $K_2 = 0$ [Eq. (7b)], the magnonic WF law holds even in the nonlinear regime if the ratio of the thermal transport coefficient of the third-order nonlinear response to the spin conductivity, K_3/G, exhibits the linear-in-T behavior. However, we find from Eq. (13a) that the ratio K_3/G is proportional to $1/T^3$ and does not exhibit the linear-in-T behavior. Thus, it is concluded that the magnonic WF law violates in the nonlinear regime.

We remark that the magnonic WF law breaks down in the strong nonlinear regime where the third-order nonlinear response to the temperature gradient $O((\partial_x T)^3)$ becomes relevant by the large temperature gradient. It is not until the third-order response contributes that the law violates. Since the thermal transport coefficient of the second-order nonlinear response vanishes $K_2 = 0$, the universal law remains valid even in the nonlinear regime where the temperature gradient is large but not enough for the third-order response to become relevant: We refer to this region as the weak nonlinear regime for convenience. In conclusion, in the bulk of topologically trivial insulating magnets, the magnonic WF law remains valid even in the weak nonlinear regime but breaks down in the strong nonlinear regime.

III. DISCUSSION

In contrast to the bulk of topologically trivial materials studied in this paper [Eq. (4)], the second-order nonlinear response does not vanish in junction systems [8,30], including quantum dot systems, and it contributes to thermal transport. Therefore, in the quantum dot system [33,34] the WF law for electronic transport violates in the weak nonlinear regime due to the second-order response. Thus, we find that in the bulk of topologically trivial materials the WF law is more robust against the nonlinear effect compared with in junction systems, in that the law breaks down in the strong nonlinear regime for the bulk of topologically trivial materials, while it violates in the weak nonlinear regime for junction systems.

Note that throughout this paper, we focus on longitudinal thermal transport in the bulk of topologically trivial insulating magnets where Berry curvatures are zero, and find that the WF law breaks down in the strong nonlinear regime. In the bulk of topological materials[5], however, the WF law for electronic Hall transport violates in the weak nonlinear regime due to the second-order response arising from nonzero Berry curvatures [18]. Thus, it is concluded that in the bulk of topologically trivial materials the WF law is more robust against the nonlinear effect compared with in the bulk of topological materials.

In this paper, we have studied magnon transport in the nonlinear response regime under the assumption that the energy dispersion of magnons is gapped and parabolic in terms of k. We remark that the second-order nonlinear response [Eq. (4)] does not vanish in general if one assumes a magnon energy dispersion including the k-linear term. In that case, the magnon nonreciprocity [20], e.g., the spin-wave Doppler shift [21], could arise in certain materials with broken inversion symmetry. See Ref. [22] for nonreciprocal transport in a gapless spin-wave system, i.e., the magnon nonreciprocity in the absence of a finite energy gap.

In general, there exists a spin anisotropy in magnets, which causes the magnon energy gap. Therefore, toward the development of various functions of spintronics devices, it is of importance to establish the fundamental principle of magnon transport even in the gapped systems. Hence, we studied the gapped magnonic systems. In the gapped system, the magnonic WF law holds only at low temperature $k_B T \ll \Delta$ and the linear-in-T behavior violates at higher temperatures even within the linear response [8,9]. Therefore, in this paper focusing on the gapped magnonic system at such low temperatures, we have studied the effect of the nonlinear response on the magnonic WF law (i.e., the linear-in-T behavior). We also remark that if the magnon gap is much smaller than the thermal energy, then there would be a large number of low-energy magnons and therefore frequent interactions between magnons. In this case, we would need to consider the magnon-magnon interaction to capture the transport properties well. In this paper, we focused on the systems with the magnon gap larger than the thermal energy in part so that the magnon-magnon interactions can be neglected. Studying the effect of magnon-magnon interactions is beyond the scope of our current paper. For these reasons, in this paper, we have focused on the gapped magnonic system at low temperatures. Still, it will be of significance to develop our work into the gapless magnon mode that possesses appropriate symmetry and include the magnon-magnon interaction. We leave the advanced study for future work.

By manipulating the applied magnetic field, magnon and phonon thermal conductivities can be distinguished experimentally [35] because the former does depend strongly on the field whereas the latter does not. In fact, in Ref. [35], it has been experimentally shown that thermal conductivities of phonons and magnons in a magnetic insulator can be separately characterized at low temperatures $T \leqslant O(1)$K. For this reason, we focus only on magnon thermal conductivity in our paper.

IV. ESTIMATES FOR EXPERIMENTS

In the bulk of topologically trivial insulating magnets, the heat current density [Eq. (6)] consists of the linear response $K_1 \partial_x T$ and the third-order nonlinear response $K_3(\partial_x T)^3$. When the large temperature gradient is applied enough that the third-order nonlinear response begins to contribute in that the ratio of the nonlinear to linear response amounts to $1/10$, $K_3(\partial_x T)^3/(K_1 \partial_x T) = 1/10$, we identify it with the strong nonlinear regime. Thus, for observation of the violation of the magnonic WF law in the bulk of insulating magnets, the temperature gradient needs to reach $|\partial_x T| = \sqrt{(1/10)(K_1/K_3)} =: \mathcal{T}$, where $K_1/K_3 = 25\sqrt{\pi}\hbar^2 k_B^2 T^4/(64\tau^2 D\Delta^3 A_4)$. The criterion for observation of the violation is whether or not the temperature gradient exceeds the value of $\mathcal{T}$.

For an estimate, we assume the following experimental parameter values [36,37] for Cr_2O_3: $D = 10$ meV(nm)^2, $\Delta \sim$

[5]See Ref. [50] for topological magnon systems.

4 meV, $\alpha = O(10^{-3})$, and $T = 4$ K. This results in $\mathcal{T} = O(10)$ K/mm. In addition, since the value of $\mathcal{T}$ is proportional to the Gilbert damping constant and it takes [32,38] $\alpha = O(10^{-4})$ or even $\alpha < O(10^{-4})$ for YIG depending on the shape, it is roughly estimated as $\mathcal{T} = O(1)$ K/mm or even $\mathcal{T} < O(1)$ K/mm for YIG, respectively. In both cases, those temperature gradients are experimentally realizable [39].

Observation of long-distance transport of spin-wave spin currents [40] and measurement of the nonequilibrium spin chemical potential [25], the magnonic spin conductivity [24,41], and the thermal conductivity [42] have been reported. References [43–45] develop magnonics technologies at low temperatures. Given these estimates, we expect that observation of the magnonic WF law and the violation, while being challenging, seem within experimental reach with current device and measurement techniques. The key is to decrease the overall temperature while the temperature gradient is maintained constant.

V. SUMMARY

Focusing on longitudinal transport of magnons at low temperatures in the bulk of topologically trivial insulating magnets, we have studied the validity and have found the violation of the magnonic Wiedemann-Franz law in the nonlinear response regime. In terms of the heat current density, the magnonic Wiedemann-Franz law is recast into the form that the ratio of the heat current density to the magnonic spin conductivity is linear in temperature for a fixed temperature gradient. Then we have shown that the universality of the Wiedemann-Franz law, the linear-in-T behavior, breaks down in the strong nonlinear regime. In contrast to the linear response, the ratio of the thermal transport coefficient of the third-order nonlinear response to the spin conductivity is proportional to $1/T^3$, and the proportionality constant is less universal in that it depends on other material parameters as well as the g factor. Thus, the universal law violates in the strong nonlinear regime where the third-order nonlinear response becomes relevant by the large temperature gradient. Since the second-order nonlinear response vanishes in the bulk of topologically trivial insulating magnets, the magnonic Wiedemann-Franz law remains valid even in the weak nonlinear regime but breaks down in the strong nonlinear regime. Those findings are within experimental reach with current device and measurement technologies. Toward efficient transmission of information that goes beyond what is offered by conventional electronics, our discovery is a promising building block in magnon-based spintronics, in the evaluation of the figure of merit for thermomagnetic conversion elements of spintronics devices.

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APPENDIX: TRANSPORT COEFFICIENTS OF MAGNONS IN THE NONLINEAR RESPONSE REGIME

In this Appendix, we provide some details of the straightforward calculation for the transport coefficient L_{ij} of the nonlinear response in the bulk of topologically trivial insulating magnets, and make a few remarks. Under the relaxation time approximation, the Boltzmann equation of the quasiparticle approximation describes the transport property of a steady state in terms of time as [23,27–29,46,47]

$$\left(\mathbf{v}_\mathbf{k}\cdot\nabla T\frac{\partial}{\partial T}-g\mu_\mathrm{B}\mathbf{v}_\mathbf{k}\cdot\nabla B\frac{\partial}{\partial\epsilon_\mathbf{k}}\right)f_\mathbf{k}=-\frac{f_\mathbf{k}-f_\mathbf{k}^0}{\tau}, \tag{A1}$$

where τ is the relaxation time, the Bose distribution function of magnons out of equilibrium $f_\mathbf{k}$, and the one in equilibrium $f_\mathbf{k}^0 := (\mathrm{e}^{\beta\epsilon_\mathbf{k}}-1)^{-1}$. Defining the deviation from equilibrium $g_\mathbf{k} := f_\mathbf{k}-f_\mathbf{k}^0$, it is described as

$$g_\mathbf{k}=-\tau\left(\mathbf{v}_\mathbf{k}\cdot\nabla T\frac{\partial}{\partial T}-g\mu_\mathrm{B}\mathbf{v}_\mathbf{k}\cdot\nabla B\frac{\partial}{\partial\epsilon_\mathbf{k}}\right)\left(f_\mathbf{k}^0+g_\mathbf{k}\right). \tag{A2}$$

Using the method of successive substitution for $g_\mathbf{k}$, the deviation up to $O(\tau^3)$ is given as

$$g_\mathbf{k} = g_1+g_2+g_3+O(\tau^4), \tag{A3a}$$

$$g_1 := -\tau\left(\mathbf{v}_\mathbf{k}\cdot\nabla T\frac{\partial}{\partial T}-g\mu_\mathrm{B}\mathbf{v}_\mathbf{k}\cdot\nabla B\frac{\partial}{\partial\epsilon_\mathbf{k}}\right)f_\mathbf{k}^0, \tag{A3b}$$

$$g_2 := \tau^2\left(\mathbf{v}_\mathbf{k}\cdot\nabla T\frac{\partial}{\partial T}-g\mu_\mathrm{B}\mathbf{v}_\mathbf{k}\cdot\nabla B\frac{\partial}{\partial\epsilon_\mathbf{k}}\right)^2 f_\mathbf{k}^0, \tag{A3c}$$

$$g_3 := -\tau^3\left(\mathbf{v}_\mathbf{k}\cdot\nabla T\frac{\partial}{\partial T}-g\mu_\mathrm{B}\mathbf{v}_\mathbf{k}\cdot\nabla B\frac{\partial}{\partial\epsilon_\mathbf{k}}\right)^3 f_\mathbf{k}^0, \tag{A3d}$$

where $g_1 = O(\tau)$, $g_2 = O(\tau^2)$, and $g_3 = O(\tau^3)$. From $\partial/(\partial T) = -(\epsilon_\mathbf{k}/T)[\partial/(\partial\epsilon_\mathbf{k})]$, the component g_3 is recast into

$$g_3=\tau^3\left(\mathbf{v}_\mathbf{k}\cdot\nabla T\frac{\epsilon_\mathbf{k}}{T}+g\mu_\mathrm{B}\mathbf{v}_\mathbf{k}\cdot\nabla B\right)^3\frac{\partial^3 f_\mathbf{k}^0}{\partial\epsilon_\mathbf{k}{}^3}. \tag{A4}$$

Focusing on sufficiently low temperatures compared to the magnon gap and thus assuming $\Delta/(k_\mathrm{B}T) \gg g\mu_\mathrm{B}\partial_x B/(k_\mathrm{B}\partial_x T)$, the third-order response to the temperature gradient $O((\nabla T)^3)$ becomes dominant. This condition can be met when the system has a sufficiently large magnetic anisotropy and the resulting magnon gap, such as $\Delta \sim 4$ meV $= O(10)$ T for [9,36,37] Cr_2O_3, $\Delta \sim 2$ meV $= O(10)$ T for [48] $SrRuO_3$, and $\Delta \sim 0.4$ meV $= O(1)$ T for [49] CrI_3. Then, the component reduces to

$$\begin{aligned} g_3 \overset{\rightarrow}{=} & -\tau^3(\mathbf{v}_\mathbf{k}\cdot\nabla T)^3\frac{\partial^3 f_\mathbf{k}^0}{\partial T^3}+O((\nabla T)^2(\nabla B)^1) \\ & +O((\nabla T)^1(\nabla B)^2)+O((\nabla B)^3), \end{aligned} \tag{A5}$$

and we neglect the other third-order terms such as $O((\nabla T)^2(\nabla B)^1)$, $O((\nabla T)^1(\nabla B)^2)$, and $O((\nabla B)^3)$. Using the relation at low temperatures $k_{\rm B}T \ll \Delta$,

$$\frac{\partial^3 f^0_{\mathbf{k}}}{\partial T^3} \overset{\rightarrow}{=} \mathrm{e}^{-\beta\epsilon_{\mathbf{k}}}\frac{(\beta\epsilon_{\mathbf{k}})^3}{T^3}, \tag{A6}$$

we obtain the component as

$$g_3 \overset{\rightarrow}{=} -\tau^3(\mathbf{v}_{\mathbf{k}}\cdot\nabla T)^3\mathrm{e}^{-\beta\epsilon_{\mathbf{k}}}\frac{(\beta\epsilon_{\mathbf{k}})^3}{T^3}. \tag{A7}$$

Assuming magnons, which carry the spin angular momentum -1 in units of $\hbar$, we define the spin current density $\mathbf{j}^{\rm s}$ and the energy current density $\mathbf{j}^{\rm E}$ as

$$\mathbf{j}^{\rm s} = -\int \frac{d^3\mathbf{k}}{(2\pi)^3} g\mu_{\rm B}\mathbf{v}_{\mathbf{k}} g_{\mathbf{k}}, \tag{A8a}$$

$$\mathbf{j}^{\rm E} = \int \frac{d^3\mathbf{k}}{(2\pi)^3} \epsilon_{\mathbf{k}}\mathbf{v}_{\mathbf{k}} g_{\mathbf{k}}. \tag{A8b}$$

Substituting the function $g_{\mathbf{k}} = g_1 + g_2 + g_3$ into each current density and performing the Gaussian integrals, we obtain the transport coefficient of the nonlinear response at low temperatures as

$$L_{13} = L_{14} = L_{15} = L_{23} = L_{24} = L_{25} = 0, \tag{A9a}$$

$$L_{16} \overset{\rightarrow}{=} -g\mu_{\rm B}F\mathrm{e}^{-b}\left[\frac{D^3}{(\beta D)^{13/2}}A_6 + \frac{3D^2\Delta}{(\beta D)^{11/2}}A_5 + \frac{3D\Delta^2}{(\beta D)^{9/2}}A_4 + \frac{\Delta^3}{(\beta D)^{7/2}}A_3\right], \tag{A9b}$$

$$L_{26} \overset{\rightarrow}{=} F\mathrm{e}^{-b}\left[\frac{D^4}{(\beta D)^{15/2}}A_7 + \frac{4D^3\Delta}{(\beta D)^{13/2}}A_6 + \frac{6D^2\Delta^2}{(\beta D)^{11/2}}A_5 + \frac{4D\Delta^3}{(\beta D)^{9/2}}A_4 + \frac{\Delta^4}{(\beta D)^{7/2}}A_3\right], \tag{A9c}$$

$$\frac{L_{26}}{L_{16}} \overset{\rightarrow}{=} -\frac{\Delta}{g\mu_{\rm B}}, \tag{A9d}$$

where $A_n := \sqrt{\pi}(2n)!/[2^{2n+1}(n!)]$ for $n \in \mathbb{N}$, $F := (2D/\hbar)^4[\tau^3\beta^3/(10\pi^2)]$, and $b := \beta\Delta$. The second-order response vanishes due to the property of the odd function, and this results in $K_2 = 0$. See Ref. [9] for the transport coefficient of the linear response, where the Onsager relation holds $L_{12} = L_{21}$. At low temperatures, it reduces to [9] $L_{12}/L_{11} = L_{21}/L_{11} \overset{\rightarrow}{=} -\Delta/(g\mu_{\rm B})$. Therefore, the thermal transport coefficient of the third-order nonlinear response K_3 at low temperature is given as

$$K_3 \overset{\rightarrow}{=} \frac{1}{T^3}\left(L_{26} + \frac{\Delta}{g\mu_{\rm B}}L_{16}\right). \tag{A10}$$

Finally, substituting L_{26} and L_{16} into Eq. (A10) we obtain the thermal transport coefficient of the third-order nonlinear response K_3 at low temperatures in the main text.

Next, we remark on the relaxation time. The relaxation time is different from the lifetime of magnons in general. Those are distinct quantities. However, under the assumption that impurities are dilute and scattering is elastic and spatially isotopic with the impurity potential localized in space (i.e., the relaxation time depends solely on the magnitude of the wavenumber), the relaxation time coincides with the lifetime. At low temperatures $k_{\rm B}T \ll \Delta$, the relaxation time reduces to [31]

$$\tau \overset{\rightarrow}{=} \frac{1}{2\alpha}\frac{\hbar}{\Delta}, \tag{A11}$$

where α is the Gilbert damping constant. Since the Gilbert damping constant is little influenced by temperature [32] (i.e., the temperature dependence is negligibly small), it is concluded that at low temperatures the relaxation time takes a constant value of being temperature-independent. Note that at sufficiently low temperatures, the effect of magnon-magnon interactions and that of phonons are negligibly small, and impurity scattering makes a major contribution to the relaxation. See Ref. [31] for details.

Lastly, we comment on the Boltzmann equation. Throughout this paper, we study magnon transport using the conventional Boltzmann equation of the quasiparticle approximation. From the viewpoint of quantum field theory, under the assumption that the variation of the center-of-mass coordinates is slow compared with that of the relative coordinates, the quantum kinetic equation of the lowest order gradient approximation becomes the quantum Boltzmann equation, which reduces to the conventional Boltzmann equation in the limit of the quasiparticle approximation [46,47]. Therefore, the criterion for magnon transport to be described by the conventional Boltzmann equation is whether or not the quasiparticle approximation is applicable to the system. From Ref. [31], this results in the condition $\Delta \gg \hbar/(2\tau)$. Since $\hbar/(2\tau) \overset{\rightarrow}{=} \alpha\Delta$ at low temperatures and $\alpha \leqslant O(10^{-3})$ for insulating magnets [32,38], the condition is satisfied. Thus, it is concluded that magnon transport we study in this paper is described by the conventional Boltzmann equation.

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