Title

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Supplementary Material:
Spin Seebeck Effect near the Antiferromagnetic Spin-Flop Transition

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Theoretical formalism.—We calculate the spin currents, $\boldsymbol{J}_l = (\hbar g_l^{\uparrow\downarrow}/4\pi)\,\boldsymbol{l}\times\partial_t\boldsymbol{l}$ and $\boldsymbol{J}_m = (\hbar g_m^{\uparrow\downarrow}/4\pi)\,\boldsymbol{m}\times\partial_t\boldsymbol{m}$, by averaging over thermal fluctuations of the magnetic variables. The latter can be obtained from the symmetrized fluctuation-dissipation theorem:

$$\langle\delta\phi_i\delta\phi_j\rangle = \frac{i\hbar}{2}\int\frac{d^3k}{(2\pi)^3}\left[\chi_{ji}^*(\mathbf{k},\omega)-\chi_{ij}(\mathbf{k},\omega)\right]N(\omega), \tag{1}$$

where $\delta\phi_i$ stands for a Cartesian component of $\boldsymbol{l}$ or $\boldsymbol{m}$ and χ_{ij} is the corresponding linear-response function. $N(\omega)\equiv n_{\mathrm{BE}}(\omega)+1/2$ accounts for thermal fluctuations associated with occupied modes, according to the Bose-Einstein distribution function n_{BE}, with $1/2$ reflecting the zero-point motion[1]. The dynamic susceptibility tensor is defined by $\delta\phi_i = \chi_{ij}\xi_j$, for the field ξ_j thermodynamically conjugate to ϕ_j. Our system is driven according to the energy density $E(B,t) = E(B) - \boldsymbol{m}\cdot\boldsymbol{h}(t) - \boldsymbol{l}\cdot\boldsymbol{g}(t)$, where $\boldsymbol{g}$ and $\boldsymbol{h}$ are conjugate to $\boldsymbol{l}$ and $\boldsymbol{m}$, respectively. The off-diagonal components of the Néel response $\chi_{ij}^{(l)}$ thus determine the Néel pumping as $\langle\boldsymbol{l}\times\partial\boldsymbol{l}/\partial t\rangle_k \to i\omega\epsilon^{ijk}\langle l_i l_j\rangle$ (in terms of the Levi-Civita tensor ϵ^{ijk}, and upon the Fourier transform), and similarly for the magnetic response, $\chi_{ij}^{(m)}$.

For convenience, we reproduce the dispersions here,

$$\omega_{1k},\omega_{2k} = \mp\gamma B + \sqrt{(\gamma B_c)^2+(ck)^2}, \tag{2a}$$

$$\omega_{3k} = ck, \quad \omega_{4k} = \sqrt{\gamma^2B^2-\gamma^2B_c^2+(ck)^2}, \tag{2b}$$

where $c = s^{-1}\sqrt{A/\chi}$ is the speed of the large-k AF spin waves. The components of χ_{ij} contributing to spin currents in I are

$$\chi_{xy}^{(l)} = -\frac{i}{4s^2\chi\omega_{0k}}\left(\frac{1}{\omega-\omega_{1k}+i\epsilon}-\frac{1}{\omega+\omega_{1k}+i\epsilon}-\frac{1}{\omega-\omega_{2k}+i\epsilon}+\frac{1}{\omega+\omega_{2k}+i\epsilon}\right), \tag{3a}$$

$$\chi_{xy}^{(m)} = \chi^2K_1^2\chi_{xy}^{(l)}, \tag{3b}$$

where $\omega_{0k} = \sqrt{(\gamma B_c)^2+(ck)^2}$. According to Eq. (3a), the fluctuations perpendicular to $\boldsymbol{l}_{0,\mathrm{I}} = \hat{\mathbf{z}}$ at ω_{1k} and ω_{2k} produce opposite contributions to the spin currents. The magnetic fluctuations in I in, e.g. $\mathrm{Cr_2O_3}$, are a factor $(\chi K_1)^2\sim10^{-7}$ smaller than the Néel fluctuations and will be neglected. $\delta\boldsymbol{m}$ is elliptically polarized in the ω_{4k} mode, with magnetic fluctuations producing a spin current according to

$$\chi_{xy}^{(m)} = \frac{i\gamma\chi B}{2}\left(\frac{1}{\omega-\omega_{4k}+i\epsilon}-\frac{1}{\omega+\omega_{4k}+i\epsilon}\right). \tag{4}$$

ω_3 is linearly polarized in $\delta\boldsymbol{l}$ and $\delta\boldsymbol{m}$ so it does not produce spin currents in the nonlinear-σ model discussed in the main text. However, there may still be small Néel fluctuations which are not captured by this model. One contribution arises if we relax the nonlinear constraint $\delta\boldsymbol{l}^2 = 1$, allowing for an additional term $\boldsymbol{m}\times\delta E/\delta\boldsymbol{l}$ in the equation of motion for $\boldsymbol{l}$. Explicitly, the Euler-Lagrange equations for the Lagrangian density $\mathcal{L}(\boldsymbol{l},\boldsymbol{m}) = s\boldsymbol{m}\cdot(\boldsymbol{l}\times\partial\boldsymbol{l}/\partial t) - E$ now are

$$s\frac{\partial\boldsymbol{l}}{\partial t} = -\boldsymbol{H}_m\times\boldsymbol{l} - \boldsymbol{H}_l\times\boldsymbol{m}, \tag{5a}$$

$$s\frac{\partial\boldsymbol{m}}{\partial t} = -\boldsymbol{H}_m\times\boldsymbol{m} - \boldsymbol{H}_l\times\boldsymbol{l}, \tag{5b}$$

where $\boldsymbol{H}_l\equiv-\delta E/\delta\boldsymbol{l}$ and $\boldsymbol{H}_m\equiv-\delta E/\delta\boldsymbol{m}$ are the effective fields. When we consider linear excitations about the same ground states as before, the only change is then that ω_{3k} develops small elliptical polarization in $\delta\boldsymbol{l}$. This produces a Néel spin current parallel to the field with similar magnitude to the ω_{4k} magnetic spin current,

$$\chi_{xy}^{(l)} = \frac{i\gamma\chi B}{2}\left(\frac{1}{\omega-\omega_{3k}+i\epsilon}-\frac{1}{\omega+\omega_{3k}+i\epsilon}\right). \tag{6}$$

Since it pumps at $g_l^{\uparrow\downarrow}\lesssim g_m^{\uparrow\downarrow}$, we discard this contribution to SSE from our analysis.

The magnitude of ϵ arises from dissipation. Dissipation is included by extending the Euler-Lagrange equations of motion with dissipative forces $\partial\mathcal{F}/\partial\dot{\boldsymbol{m}}$ and $\partial\mathcal{F}/\partial\dot{\boldsymbol{l}}$ from the Rayleigh dissipation functional $\mathcal{F} = \alpha\dot{\boldsymbol{l}}^2/2 + \widetilde{\alpha}\dot{\boldsymbol{m}}^2/2$, parametrized by Gilbert damping constants α and $\widetilde{\alpha}$. By including it in our calculation of χ_{ij}, we end up with Lorentzians centered at these poles, whose widths are determined by bulk Gilbert damping and the effective damping due to interfacial spin pumping[2,3]. When these resonance modes' quality factors are large, however, their spectral weight is sharp and may be simply integrated over. We assume this is the case, allowing us to neglect dissipation and simply use infinitesimal ϵ.

Evaluation of Seebeck coefficients.— We can now evaluate the Seebeck coefficients, $S = \partial_T(J_l + J_m)$. The spin currents are calculated by inserting Eqs. (3a) below SF and (4) above SF into (1), and integrating over the Brillouin zone. Since the spin currents are even in ω, the bounds of integration over ω may be changed from (-∞, ∞) to (0, ∞) with the spin current expression multiplied by a factor of two. Then, only the positive poles contribute.

This reproduces the results for S_{I} and S_{II} in the main text. For temperatures $T \ll T_N$, thermal occupation of magnons with momentum near the Brillouin zone boundary is exponentially suppressed, so we can extend the upper limit of integration to ∞. Furthermore, we can evaluate S_{I} and S_{II} analytically when $k_B T \gg \hbar\gamma B_c$:

$$S_{\mathrm{I}} \approx \frac{g_l^{\uparrow\downarrow}\gamma B k_B^2 T}{2\pi^3 c^3 \chi s^2}\int_0^\infty dx\; x^2 e^x n_{\mathrm{BE}}^2(x) \propto g_l^{\uparrow\downarrow} BT, \tag{7}$$

$$S_{\mathrm{II}} \approx \frac{g_m^{\uparrow\downarrow}\gamma \chi B k_B^4 T^3}{4\pi^3 c^3 \hbar^2}\int_0^\infty dx\; x^4 e^x n_{\mathrm{BE}}^2(x) \propto g_m^{\uparrow\downarrow} BT^3, \tag{8}$$

where x is dimensionless and the integrals are convergent, simply evaluating to numbers. Eqs. (7), (8) are used to plot the theoretical curves in Fig. 2 and 3 of the main text. In evaluating the Seebeck coefficients as a function of temperature, we have neglected slow temperature dependencies in the energetic constants A, χ, and K, which is valid when $T \ll T_n$.

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