Spin Seebeck effect in Cu_{2}OSeO_{3}: Test of bulk magnon spin current theory

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Spin-Seebeck Effect in Cu_2OSeO_3: Test of Bulk Magnon Spin-Current Theory

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We report measurements of the low-temperature ($T \leq 15$ K) longitudinal spin-Seebeck coefficient (S_{LSSE}) in bulk single-crystals of the helimagnetic insulator Cu_2OSeO_3 with Pt contacts. Simultaneous measurement of both S_{LSSE} and the magnon thermal conductivity (κ_m) demonstrates their correlation and allows for quantitative and favorable comparison to bulk magnon spin-current theory.

Magnon transport and energy exchange between magnons and phonons are central to the growing fields of spin caloritronics [1] and magnon spintronics [2]. Crucial to potential applications is the conversion of thermally-driven spin currents in a magnetic insulator to an electrical signal via the inverse-spin Hall effect in a heavy-metal thin film in interfacial contact – the spin-Seebeck effect. Considerable experimental and theoretical development has focused on studies of Pt contacts to the insulating ferrimagnet yttrium-iron garnet (YIG).

Magnon spin-current theory for the bulk spin-Seebeck effect [3–5] implies a direct relationship between the longitudinal spin-Seebeck coefficient (S_{LSSE}) and magnon thermal conductivity (κ_m). Quantitative tests of this relationship have not been possible in any material because κ_m is not typically large enough or easily separable from the lattice thermal conductivity. Though κ_m has been determined for YIG at low temperature in applied magnetic field [6–8], it is not clearly correlated with S_{LSSE} (e.g. their maxima appear to occur at very different temperatures [9, 10]).

Recent studies [11] demonstrated that Cu_2OSeO_3, a helimagnetic insulator with $T_C = 58$ K, harbors the largest known κ_m for any ferro- or ferri-magnetic insulator, with a maximum $\kappa_m \sim 60-80$ W/mK at $T \simeq 5-6$ K. Here we report on measurements of S_{LSSE} in 10-nm Pt/bulk single-crystal Cu_2OSeO_3 heterostructures with which κ_m, measured simultaneously, is well correlated. The data, which include interfacial spin-mixing conductances varying by more than an order of magnitude, are in quantitative agreement with the predictions of bulk spin-current theory.

Cu_2OSeO_3 comprises a three-dimensional distorted pyrochlore (approximately fcc) lattice of corner-sharing Cu tetrahedra [12, 13]. Strong magnetic interactions within tetrahedra lead to a 3-up-1-down, spin $S = 1$ magnetic state [14, 15] with weaker interactions between tetrahedra leading to their ferromagnetic ordering below $T_C \simeq 58$ K. At low temperatures [18] the low-field state [inset, Fig. 1 (a)] includes multiple helical (H) domains (aligned along the $\langle 100 \rangle$ easy-axis directions) wherein atomic spins rotate within a plane perpendicular to the helical axis with a wavelength $\lambda_h \simeq 62$ nm. At $H \gtrsim 10-25$ mT (depending on field orientation) the helices of individual domains rotate along the field to form a single-domain, conical phase (C). For $H \gtrsim 50-75$ mT the ferrimagnetic, fully-polarized (FP) state emerges.

Phase pure, single crystals of Cu_2OSeO_3 were grown by chemical vapor transport as described elsewhere [11, 19]. Specimens were cut from single-crystal ingots, oriented by x-ray diffraction, and polished into thin parallelepipeds. A two-thermometer, one-heater method was employed to measure the spin-Seebeck effect (using 25 μm diam. Au wires) and thermal conductivity simultaneously. A sputtered Pt film (10-nm thick) was deposited onto the heater end of the crystal and isolated from the heater with varnish. Further details on the measurements, crystal polishing/etching [20, 21], and properties of the Pt films are discussed in the Supplemental Material [22].

We focus in this work on data for three specimens, all with heat flow along the [111] direction and magnetic field along $[1\bar{1}0]$ [inset, Fig. 1 (a)]. Crystal 1 is the same crystal ($5 \times 1.10 \times 0.26$ mm^3) for which thermal conductivity data were reported in Ref. 11. This crystal was subsequently cut, polished (new cross-sectional area $A = 0.86 \times 0.20$ mm^2), prepared with a fresh Pt film, and remeasured. This second data set is the primary focus of the narrative since it is most extensive and because its SSE signal was a factor of 4-5 larger than during the first experiment. The two specimens are distinguished by their transverse dimensions, $\ell_0 = 0.60$ mm and 0.47 mm, respectively, where $\ell_0 \equiv 2\sqrt{A/\pi}$. A second crystal with $\ell_0 = 0.31$ mm ($A = 0.70 \times 0.11$ mm^2; crystal 5 from Ref. 11) was also studied. Data for the $\ell_0 = 0.60$ mm and $\ell_0 = 0.31$ mm specimens are included in Fig. 3 and more extensive data in Fig.'s S4 and S5 [22]. Given the high thermal conductivity of Cu_2OSeO_3 [11] and desire to maximize length along the inverse spin-Hall field ($[11\bar{2}]$), long, thin parallelepiped specimens were necessary, leading to large demagnetization factors ($N \sim 0.75$) and some nonuniformity of the applied field; we report external field values here. Extensive prior measurements of $M(H)$ and $\kappa(H)$ on crystals [11, 23] with small N re-

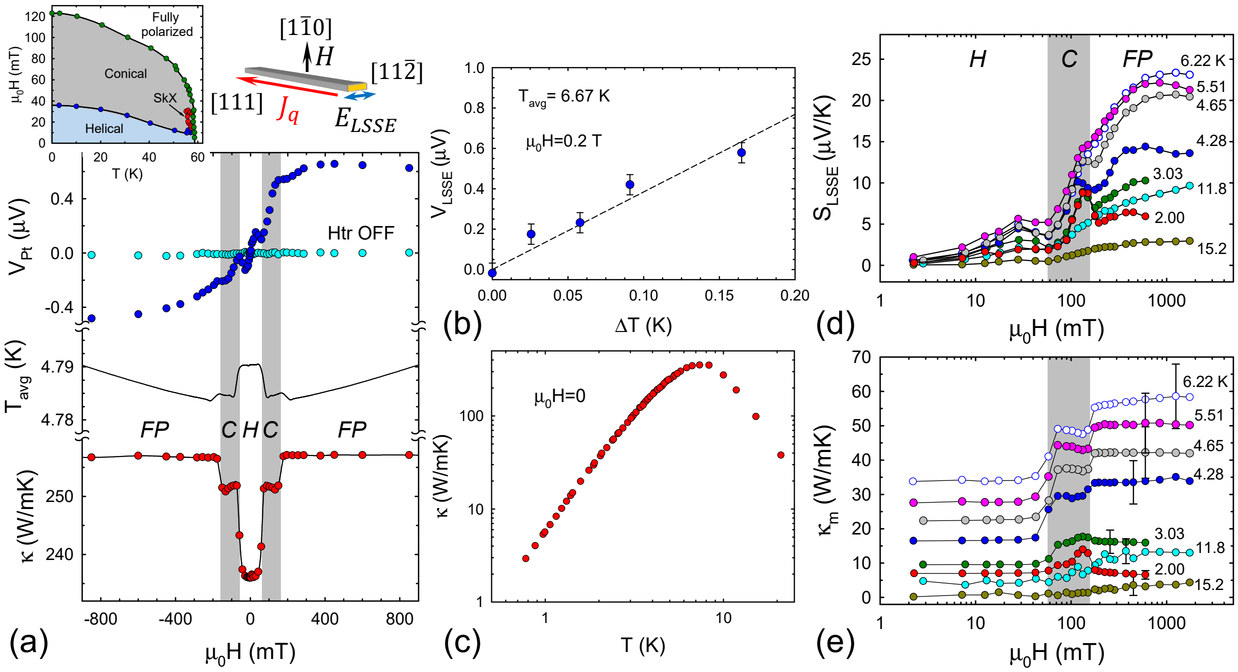

FIG. 1. (a) From top to bottom: Spin-Seebeck voltage (for heater on and off), average temperature, and thermal conductivity *vs.* applied field at $T_{avg} = 4.79$ K. Left inset: magnetic phase diagram (adapted from Ref. 18), right inset: orientation of heat flow and fields, (b) SSE voltage *vs.* ΔT at 6.67 K, (c) zero-field $\kappa(T)$, (d) SSE coefficient and (e) magnon thermal conductivity *vs.* applied field for various temperatures. Error bars are discussed in the text and in Ref. 11. The shading in (a), (d), (e) distinguishes the conical (C) spin phase from helical (H) and fully-polarized (FP) phases at lower and higher field, respectively.

veal consistent, coincident signatures of the spin-phase transitions that are employed here to identify the phase boundaries from $\kappa(H)$ data.

Prior work demonstrates that field dependent changes in κ are entirely magnonic [11, 23]. Separation of κ_L and κ_m is possible for $T \lesssim 1.2$ K where the high-field condition $E_H \gg k_B T$ is met ($E_H = g\mu_B H$, the Zeeman energy) and thus spin-wave excitations are depopulated (gapped). The mean-free paths for both phonons and magnons are comparable to ℓ_0 at $T \lesssim 2$ K. At higher T where field suppression of κ_m is incomplete, Callaway model fitting is employed to estimate κ_L, with κ_m computed by subtraction [11, 22].

Figure 1 (a) shows the field dependence of, from top to bottom, the Pt film voltage (with a constant offset voltage subtracted), average specimen temperature, and thermal conductivity at $T_{avg} = 4.79$ K. The null Pt voltage during the same sweep with heater off is also shown. Figure 1 (b) confirms linearity in ΔT of the anti-symmetrized spin-Seebeck voltage, $V_{LSSE} = [V_{Pt}(H) - V_{Pt}(-H)]/2$, and Fig. 1 (c) shows the zero-field $\kappa(T)$. $\kappa(H)$ exhibits a step-like increase at the H-C spin-phase transition, a plateau within the C phase, and another step-like increase at the C-FP phase boundary. Similar features for various orientations of heat flow and applied field have been reported in prior studies [11, 23].

Figure 1 (d) shows the longitudinal spin-Seebeck coefficient as a function of field at selected temperatures, $S_{LSSE} = (V_{LSSE}/\Delta T)(l/w)$, where l is the distance between thermometers along the heat flow and w is the length of the Pt film (approximately the specimen width). Figure 1 (e) shows the magnon thermal conductivity computed by subtracting a field-independent κ_L. The error bars reflect uncertainties in estimating κ_L from the model fitting and are largest at $T \sim 7$ K where $\kappa(T)$ has its maximum [11, 22]. Similar $S_{LSSE}(H)$ and $\kappa_m(H)$ data for the other specimens are presented in Fig.'s S4, S5 [22].

$S_{LSSE}(H)$ exhibits a small maximum at fields below the H-C phase transition, presumably associated with partial reorientation of the three $\langle 100 \rangle$-oriented helical domains, established in zero-field cooling. A sharp increase in $S_{LSSE}(H)$ characterizes the transition to the conical phase, following the increase in $\kappa_m(H)$. At $T \gtrsim 6$ K, $S_{LSSE}(H)$ increases smoothly through the C-FP transition, and saturates or declines in magnitude within the FP phase. For lower T an inflection appears at the C-FP transition and a step-like decrease emerges, becoming more prominent at the lowest T. This latter feature coincides with a step-like decrease in $\kappa_m(H)$) (see data for 3.03 K, 2.00 K), and thus can be attributed to the effects of a larger spin gap (estimated in analysis below as $\Delta \sim 0.3$ meV [22]) within the FP phase (the spin gap in the conical phase is quite small [28], ~ 12 μeV). A frac-

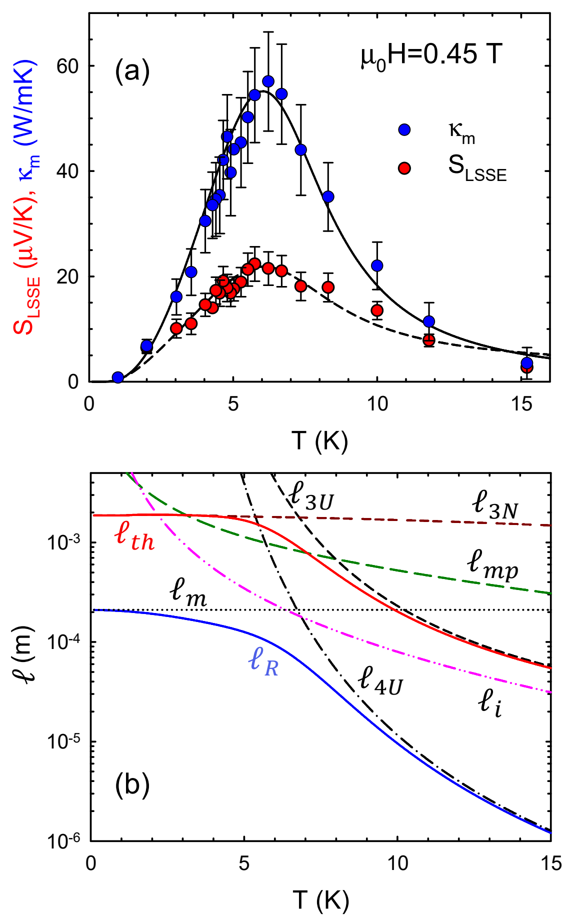

FIG. 2. (a) $\kappa_m(T)$ and $S_{LSSE}(T)$ in the fully-polarized phase at $\mu_0 H = 0.45$ T. Error bars for κ_m are described in the text, and for S_{LSSE} are dictated by uncertainty in the geometric factor (20%). The solid curve is computed from Eq. (1) and the dashed curve from Eq. (2) with $\tau_{th} = (\tau_{3N}^{-1} + \tau_{mp}^{-1} + \tau_{3U}^{-1})^{-1}$ (see text). (b) thermally averaged scattering lengths computed from the model of Ref. 27; see also the Supplemental Material [22].

tion of the thermal magnons thus become gapped as the field increases through the *C-FP* transition, effectively removing their contribution to κ_m. That the effects of the spin gap opening are evidenced in S_{LSSE} at higher T than for κ_m suggests that *subthermal* magnons contribute some weight to the spin-Seebeck effect, as has been proposed to understand the field-induced suppression of the SSE in YIG [9, 29, 30].

The most significant observation from Fig.'s 1 (d) and (e) and principal result of this work, is the clear correspondence between κ_m and S_{LSSE}; Fig. 2 (a) illustrates this correspondence in T at fixed field $\mu_0 H = 0.45$ T, within the *FP* phase where S_{LSSE} is near its maximum value. As a first test of theory, we demonstrate that the same magnon relaxation rate, employed in prior work to model $\kappa_m(T)$ for crystal 1 and other similar crystals [11], also describes $S_{LSSE}(T)$.

Inelastic neutron scattering studies [24] indicate a single spin wave branch relevant to magnon transport at low T in Cu_2OSeO_3 that is well described by an isotropic dispersion [25], $E = \Delta + g\mu_B H + \hbar\omega_{ZB}\left[1 - \cos(\pi q)\right]$, with $\hbar\omega_{ZB} = 4.55$ meV and $q = k/k_m$ the reduced wavenumber (k_m is the maximum wavenumber). The magnon thermal conductivity and spin-Seebeck coefficient from Boltzmann theory can be written as [3–5],

$$\kappa_m = \frac{k_B}{6\pi^2}\tau_R B_{21}, \tag{1}$$

$$S_{LSSE} = R_N \lambda_N \frac{2e}{\hbar}\theta_{SH}\left(\tau_m \tau_{th}\right)^{1/2} \frac{B_{11}C_2}{\left(B_{10}C_1\right)^{1/2}} F g_{eff}^{\uparrow\downarrow}, \tag{2}$$

where B_{ij} and C_k are the integrals,

$$B_{ij} = \int_0^1 dq q^2 v_m^2 \frac{x^i \left(e^x\right)^j}{\left(e^x - 1\right)^2}, \quad C_k = \int_0^1 dq q^2 \frac{x^k}{\left(e^x - 1\right)},$$

$$F = \frac{\hbar\gamma k_B k_m^2 \omega_{ZB}}{4\pi M_S 2\pi\sqrt{3}},$$

$v_m = (1/\hbar)dE/dq$ is the magnon velocity, $x = E/k_B T$, R_N, $\lambda_N = 3.7$ nm and $\theta_{SH} = 0.05$ are the Pt film resistance, spin-diffusion length and spin-Hall angle [5], $\gamma = 1.82 \times 10^{11}$ $T^{-1}s^{-1}$ is the gyromagnetic ratio [26], and $4\pi M_S \simeq 1.15 \times 10^5$ A/m is the saturation magnetization [18]. The integrals are performed over a spherical Brillouin zone with $(4/3)\pi k_m^3 = (2\pi/a)^3$. We employ thermally-averaged scattering times for which the momentum dependence has already been integrated out.

The transport relaxation rate (τ_R^{-1}) incorporates magnon-magnon Umklapp ($3U$, $4U$), magnon-impurity (i), and magnon-boundary (b) scattering, $\tau_R^{-1} = \tau_{3U}^{-1} + \tau_{4U}^{-1} + \tau_i^{-1} + \tau_b^{-1}$, computed for an isotropic Heisenberg model with quadratic magnon dispersion [27]. The expressions rely on four parameters, two of which are fixed by the value of the lattice constant and T_C [11, 22]. The strength of impurity and boundary terms are set by the non-magnetic impurity concentration (c) and magnetic domain size $\ell_m \leq \ell_0$ ($\tau_b = \ell_m/\langle v_m\rangle$, with $\langle v_m\rangle$

TABLE I. Magnon scattering and spin-Seebeck parameters.

Specimen	ℓ_0 (mm)	c(ppm)	ℓ_m (mm)	R_N (Ω)	$g_{eff}^{\uparrow\downarrow}(10^{15}m^{-2})$
crystal 1	0.60	22	0.30	467	2.45
"	0.47	22	0.21	120	39.3
crystal 2	0.31	44	0.18	293	1.27

the momentum-averaged magnon velocity [22]). The latter, employed here as a fitting parameter, was determined directly in Ref. 11 from the $\kappa_m \propto T^2$ behavior observed within the C phase at low T as ~ 0.30 mm for the specimen with $\ell_0 = 0.60$ mm. The solid curve in Fig. 2 (a) demonstrates good agreement with κ_m using the T-dependent scattering lengths ($\ell_j = \langle v_m \rangle \tau_j$) shown in Fig. 2 (b). Similar quality fitting curves for the other specimens are shown in Fig.'s S4 and S5 [22]; Table I summarizes the parameters.

Two relaxation times are distinguished in Eq. (2) for the SSE coefficient, characterizing scattering that conserves (does not conserve) magnon number, τ_m (τ_{th}) [31]; the magnon diffusion length and the SSE signal are proportional to $\sqrt{\tau_m \tau_{th}}$, where $\tau_m \ll \tau_{th}$. We take $\tau_m = \tau_R$, as τ_R is dominated by magnon conserving processes given that $\tau_{3U} \ll \tau_{4U}$ [Fig. 2 (b)]. Note that magnon-phonon interactions (characterized by τ_{mp}), which do not conserve magnon number (2-magnon, 1 phonon interactions are predominant), are weak in the low-T regime relevant here [27, 32] and play little role in κ_m provided there is sufficient coupling to ensure energy from the heater (coupling only to phonons) enters the magnon system. The criterion for this [33], $\tau_{mp} \gtrsim \ell_0/v_{ph}$ ($v_{ph} \simeq 2$ km/s is the phonon velocity [11, 22]), is satisfied [11] using τ_{mp} estimated from the intrinsic ferromagnetic resonance linewidth [26] [Fig. 2 (b)]. With $\tau_m = \tau_R$ fixed by fitting to $\kappa_m(T)$ and $\tau_{th}^{-1} = \tau_{mp}^{-1} + \tau_{3N}^{-1} + \tau_{3U}^{-1}$, $\mathrm{g}_{eff}^{\uparrow\downarrow}$ was adjusted to produce good agreement with S_{LSSE} [dashed curve in Fig. 2 (a) and Fig.'s S4, S5 for the other specimens].

The T dependence arising from the relaxation times for S_{LSSE} differs from that for κ_m by the factor $(\tau_{th}/\tau_R)^{1/2}$, which is weakly T dependent over the investigated range [Fig. S6 (a)]. This observation motivates a more fundamental test of the theory, independent of the relaxation times – Eq.'s (1) and (2) predict the two transport coefficients to be directly related through their integral expressions. A *sublinear* relationship between S_{LSSE} and κ_m for all specimens emerges when the spin-Seebeck coefficients are re-scaled by plotting βS_{LSSE} against κ_m [Fig. 3 (a)], where β is the ratio of $R_N \mathrm{g}_{eff}^{\uparrow\downarrow}$ for the $\ell_0 =$ 0.47 mm specimen to that for the others: $\beta = 4.1$ (12.7) for $\ell_0 = 0.60$ (0.31) mm specimens. Figure 3 (b) demonstrates that a power-law relation, $(S_{LSSE})^n \propto \kappa_m$ provides a good description of the data with $n = 1.15$ providing the best fit [inset, Fig. 3 (b)]. In Fig. S6 (b) we demonstrate that the integrals follow the relationship, $B_{11}C_2/(B_{10}C_1)^{1/2} \propto (B_{21})^{0.852}$ over most of the T range, yielding $n = (1/0.852) \simeq 1.17$ in excellent agreement with the data.

In summary, the unprecedentedly large magnon thermal conductivity of Cu_2OSeO_3 and simultaneous measurement of spin-Seebeck coefficient have allowed for new quantitative tests affirming bulk magnon spin-current theory. These results, heretofore inaccessible for any other material, highlight this compound as a model system for the study of magnon interactions and their role in the transport of spin and heat.

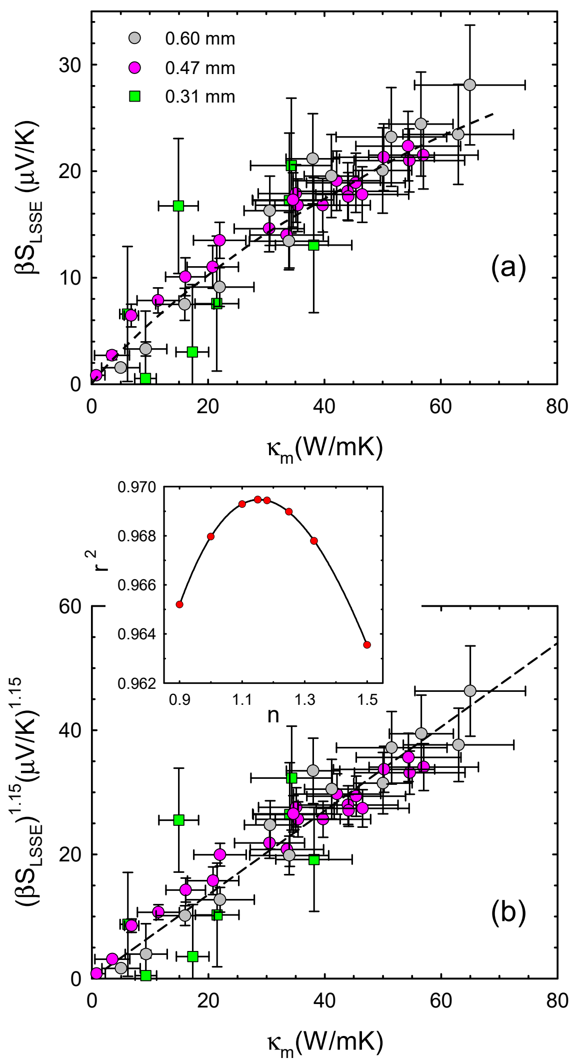

FIG. 3. Correlation between S_{LSSE} and κ_m in the fully-polarized phase ($\mu_0 H = 0.45$ T) for all three specimens on linear (a) and power-law (b) scaling. Data for crystal 1 with $\ell_0 = 0.60$ mm and crystal 2 ($\ell_0 = 0.31$ mm) have been re-scaled by their values of $R_N \mathrm{g}_{eff}^{\uparrow\downarrow}$ (Table I) to match that of crystal 1 with $\ell_0 = 0.47$ mm as described in the text. The dashed line in (a) is a guide and in (b) a linear least-squares fit. The inset shows the quality of the power-law fit to be maximized for $n = 1.15$.

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