Local and nonlocal spin Seebeck effect in lateral Pt-Cr$_2$O$_3$-Pt devices at low temperatures

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We have studied thermally driven magnon spin transport (spin Seebeck effect, SSE) in heterostructures of antiferromagnetic α-Cr$_2$O$_3$ and Pt at low temperatures. Monitoring the amplitude of the local and nonlocal SSE signals as a function of temperature, we found that both decrease with increasing temperature and disappear above 100 K and 20 K, respectively. Additionally, both SSE signals show a tendency to saturate at low temperatures. The nonlocal SSE signal decays exponentially for intermediate injector-detector separation, consistent with magnon spin current transport in the relaxation regime. We estimate the magnon relaxation length of our α-Cr$_2$O$_3$ films to be around 500 nm at 3 K. This short magnon relaxation length along with the strong temperature dependence of the SSE signal indicates that temperature-dependent inelastic magnon scattering processes play an important role in the intermediate range magnon transport. Our observation is relevant to low-dissipation antiferromagnetic magnon memory and logic devices involving thermal magnon generation and transport.

Recently, substantial scientific effort has focused on harnessing spin currents without Joule heating for low-dissipation information processing[1]. In magnetic materials, the spin current, (i.e., the directed propagation of spin angular momentum), can be either due to electron spin or to bosonic quasiparticle excitations of the magnetic order parameter called magnons. The realization of magnon spin currents in electrically insulating materials with large band gap is advantageous, since they prevent energy dissipation due to ohmic losses owing to the electronic motion. Moreover, the spin propagation length of electronic spin currents is relatively short, typically on the order of nanometers in most metals. Magnon spin currents, on the other hand, can travel distances up to several micrometers in magnetic insulators[2]. Magnons can be excited in magnetic insulators via numerous methods e.g., magnetically using microwave-frequency AC magnetic fields (coherent magnons in the GHz range), thermally via the spin Seebeck effect (incoherent magnons in the THz range)[3], and electrically using a low-frequency AC or DC electric current in a neighbouring heavy metal with large spin Hall angle, such as platinum[2].

Although the initial study of magnon spin currents was focused on ferromagnetic insulators, recently, antiferromagnetic insulators (AFI) have moved into the focus of magnonics research due to their abundance in nature, better scalability in nanodevices with minimal cross-talk, immunity against magnetic field perturbations, and orders of magnitude faster magnetization dynamics at the terahertz frequency range[4–7]. Magnon spin current transport has been demonstrated in many antiferromagnetic insulators, such as NiO[8, 9], CoO[10], α−Fe$_2$O$_3$, MnPS$_3$, MnF$_2$, and Cr$_2$O$_3$. Additionally, the absence of dipole-dipole interactions make axially symmetric AFI ideal materials for the realization of magnon Bose-Einstein condensates (BEC) [14, 15] and magnon spin superfluidity, i.e., a long-range propagating Goldstone mode arising from the spontaneous breaking of U(1) symmetry[16–18]. All these recent studies on magnon spin transport in antiferromagnets have lead to a new frontier research field called antiferromagnetic magnonics, as a subfield of spintronics.

One fascinating effect hinging on thermally excited magnon spin transport is the so-called spin Seebeck effect. The spin Seebeck effect refers to the generation of magnon spin currents by a temperature gradient applied across a magnetic material[3]. The SSE is manifested as an electric voltage in an adjacent heavy metal layer, in which the thermally driven spin current is converted into a charge current by the inverse spin Hall effect (ISHE)[10]. Originally it was assumed that AFI will not exhibit a finite SSE due to the lack of a net magnetization and the particular properties of magnon modes in AFI. More specifically, in a uniaxial AFI with two magnetic sublattices, there will be two degenerate magnon modes which produce spin current in opposite direction under a thermal gradient, such that the net spin current cancels out. However, the degeneracy of the two modes can be lifted by a magnetic field or even a spin flop transition to a ferromagnetic-like state, or by inversion symmetry breaking at an interface, resulting in finite SSE response[20, 21]. Such a magnetic field-induced SSE has been recently observed in various AFI like Cr$_2$O$_3$, MnF$_2$, and FeF$_2$. Moreover, indication of magnon spin superfluidity has been recently reported in Cr$_2$O$_3$. In this paper, we focus on local and nonlocal SSE experiments in antiferromagnetic α-Cr$_2$O$_3$ thin films for small injector-detector separation. We perform

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magnetic-field-orientation-dependent local and nonlocal SSE measurements by rotating an external magnetic field of constant magnitude in three orthogonal planes. By monitoring the amplitude of the local and nonlocal voltage modulation as a function of different parameters, such as the temperature and the spatial separation between the injector-detector Pt stripes, we probe the spin transport via antiferromagnetic magnons. From these data, we extract the magnon spin diffusion length ($l_m$) in the diffusive transport regime at low temperature. We show that although in the long-distance regime, superfluid spin transport might be realized, the intermediate-distance range is dominated by magnon diffusion, while the superfluid contribution is not detectable.

We use $\alpha$-Cr$_2$O$_3$ for our spin transport experiments, since this material is one of the best studied AFI with uniaxial anisotropy [26, 27]. The single crystal $\alpha$-Cr$_2$O$_3$ has hexagonal (corundum) crystal structure with R$ar{3}$c space group. The magnetic structure is dictated by layers of Cr$^{3+}$ ions (S=$3/2$) with an antiparallel sublattice spin alignment along the [0001] axis, such that in the (0001) plane, all Cr$^{3+}$ ions belong to the same antiferromagnetic sublattice. Previous spin transport experiments have shown that the transmission of the spin current in Cr$_2$O$_3$ depends on its Néel vector orientation[13, 28, 29]. The spin transmission is completely blocked below the Néel temperature ($T_N$), if Néel vector N of Cr$_2$O$_3$ (oriented along [0001] easy axis) is aligned perpendicular to the polarization of the injected spin current. Interestingly, Cr$_2$O$_3$ abruptly becomes a good spin conductor above $T_N$ [13].

The $\alpha$-Cr$_2$O$_3$ thin films were grown by magnetron sputter deposition (base pressure: 10$^{-7}$ mbar; Ar sputter pressure: 10$^{-3}$ mbar; deposition rate: 0.04 nm/s) of a pure Cr$_2$O$_3$ target material (robeko GmbH & Co. KG). To initiate the crystallization of the $\alpha$-Cr$_2$O$_3$, single-crystalline Al$_2$O$_3$ (0001) substrates (Crystec GmbH) were heated to 850 °C before the deposition. The $\alpha$-Cr$_2$O$_3$ thin films were deposited at 700 °C. The samples were capped in-situ with a sputtered 3-nm-thick Pt layer at lower temperatures of $\approx$100 °C (deposition rate 0.1 nm/s). The films were found to be of high crystallinity and atomically smooth with single lattice steps in height at the boundary between plateaus. Structural characterization reveal (0001)-oriented growth, ensuring an out-of-plane easy axis of the Néel vector for 200 nm thick films. From cross-sectional transmission electron microscopy (TEM) (see Fig. 1(a)), the films were found to be granular with about 50-nm-sized columnar grains with very high crystallinity within each grain [30].

Multiple lateral devices were fabricated on one and the same sample with electron beam lithography and Ar-ion milling. Figure 1(b) shows a schematic of a Pt-Cr$_2$O$_3$-Pt nonlocal device with two Pt stripes, one acting as injector, the other as detector. The length and width of the platinum stripes for all devices studied here are approximately $L = 145 \mu$m and $W = 500$ nm, respectively. We prepared a series of nonlocal Pt-Cr$_2$O$_3$-Pt devices with an edge-to-edge spacing $d_{nl}$ between the two Pt stripes varying from 0.4 to 4 $\mu$m. Local and nonlocal voltage measurements were performed using a quasi-DC method applying a DC current $I = 150$ $\mu$A through one of the Pt stripes and periodically reversing its direction. We measure both symmetric $V_{loc}^{\pm}$ measured at second harmonic voltages for both local (loc) and nonlocal (nl) measurement configurations. The antisymmetric voltage $V_{loc}^{\alpha}$ and symmetric voltage $V_{loc}^{\pm}$ are equivalent to the 1$^{st}$ harmonic voltage ($V^{1f}$) and second harmonic voltage ($V^{2f}$) measured with an AC lock-in technique, respectively[11, 31–35].

Driving a charge current through Pt can generate magnons in the $\alpha$-Cr$_2$O$_3$ via two different mechanisms, electrically and thermally[32, 36]. Due to the spin Hall effect inside Pt, a transverse spin current is generated orthogonal to the current direction, and a spin accumulation $\mu_s$ with spin polarization along the t-axis builds up at the Pt-Cr$_2$O$_3$ interface. When a magnetic field is applied, it can cause canting of the Cr$^{3+}$ moments in the two different magnetic sublattices, producing a canted moment $M_{cnt}$. Nonequilibrium magnon spin accumulation is created inside $\alpha$-Cr$_2$O$_3$ via exchange interaction at the interface, if the spin accumulation direction $\mu_s$ is not orthogonal to $M_{cnt}$. Additionally, Joule heating of the

![Figure 1](image-url)

**FIG. 1.** (a) Cross-sectional high-resolution TEM image of the Cr$_2$O$_3$-Pt bilayer film. A high crystalline quality of the $\alpha$-Cr$_2$O$_3$ individual grains and a sharp Cr$_2$O$_3$-Pt interface are evident. (b) Device schematics with local and nonlocal measurement configuration. An electric current $I$ is applied at one Pt strip, and voltages can be detected at the same strip (locally) or at the other strip (nonlocally). (c,d,e) Device schematics with different external magnetic field arrows. Rotation planes: (c) ip-rotation in the j – t plane with angle $\alpha = \angle jH$ (ip-rotation), (d) oopj-rotation in the t – n plane with angle $\beta = \angle nH$ (oopj-rotation), and (e) oopt-rotation in the n – j plane with angle $\gamma = \angle nH$ (opt-rotation).
A magnetic field of $\mu_0 H = 2 \, \text{T}$ was hereby used to induce a canted moment $M_{\text{cnt}}$ in $\alpha$-Cr$_2$O$_3$. (a) The local SSE voltage signal $V_{\text{loc}}^s$ shows a clear modulation only for $\alpha$ and $\beta$-rotations. (b) The same $\sin(\alpha, \beta)$ modulation with opposite sign is observed for the nonlocal SSE voltage signal $V_{\text{nl}}^s$. Please note, that an average voltage signal offset was subtracted.

We measured both the antisymmetric $V_{\text{loc(nl)}}^s$ and the symmetric $V_{\text{loc(nl)}}$ voltages in the local and nonlocal configuration, while the external magnetic field $H$ was rotated in three different orthogonal planes, as depicted in Fig. 1(c,d,e). The three orthogonal rotation planes of the external magnetic field $H$ are: (1) $\text{ip}$-rotation in the $j-t$ plane with angle $\alpha = \angle jH$ ($\alpha$-rotation), (2) $\text{oop}$-rotation in the $t-n$ plane with angle $\beta = \angle nH$ ($\beta$-rotation), and (3) $\text{oopt}$-rotation in the $n-j$ plane with angle $\gamma = \angle nH$ ($\gamma$-rotation). For the antisymmetric signal $V_{\text{loc(nl)}}^s$, no modulation was observed at any temperature and magnetic field. This can be rationalized considering that in our measurement configuration, the spin accumulation direction $\mu_s$ (in-plane) is perpendicular to the Néel vector $\mathbf{N}$ (out-of-plane). Therefore, the electrical excitation of magnons is inefficient\cite{13, 28}. In the following, we therefore focus only on the symmetric voltage $V_{\text{loc(nl)}}^s$ which includes the thermally generated SSE signal.

Fig. 2 summarizes the angular dependence of the local and nonlocal SSE voltage for all three rotation planes at 10 K. A constant, angle-independent voltage offset was subtracted from all the data. The angular dependence was measured applying a magnetic field $\mu_0 H = 2 \, \text{T}$, large enough to create a small canting of the Cr$^{3+}$ moments along the field direction. A clear modulation of the local voltage can be observed for the $\alpha$ and $\beta$-rotation planes in Fig. 2(a). The angular dependence follows a $\sin(\alpha, \beta)$ dependence. This agrees with the expected behavior, for the spin Seebeck effect\cite{37} and thus confirms the notion that the symmetric $V_{\text{loc}}^s$ arises due to magnons excited thermally. In contrast, if the magnons were generated electrically, one would expect a $\sin^2(\alpha, \beta)$ behavior\cite{32}. In a simple microscopic picture, we thus assume that if the magnetic field is applied perpendicular to the [0001] easy axis (along $\mathbf{n}$), a finite canting is induced, and consequently, a finite magnetization $M_{\text{cnt}}$ appears in $\alpha$-Cr$_2$O$_3$.

We measured the antisymmetric $V_{\text{loc(nl)}}^s$ and the symmetric $V_{\text{loc(nl)}}$ voltages in the local and nonlocal configuration, while the external magnetic field $H$ was rotated in three different orthogonal planes, as depicted in Fig. 1(c,d,e). The three orthogonal rotation planes of the external magnetic field $H$ are: (1) $\text{ip}$-rotation in the $j-t$ plane with angle $\alpha = \angle jH$ ($\alpha$-rotation), (2) $\text{oop}$-rotation in the $t-n$ plane with angle $\beta = \angle nH$ ($\beta$-rotation), and (3) $\text{oopt}$-rotation in the $n-j$ plane with angle $\gamma = \angle nH$ ($\gamma$-rotation). For the antisymmetric signal $V_{\text{loc(nl)}}^s$, no modulation was observed at any temperature and magnetic field. This can be rationalized considering that in our measurement configuration, the spin accumulation direction $\mu_s$ (in-plane) is perpendicular to the Néel vector $\mathbf{N}$ (out-of-plane). Therefore, the electrical excitation of magnons is inefficient\cite{13, 28}. In the following, we therefore focus only on the symmetric voltage $V_{\text{loc(nl)}}^s$ which includes the thermally generated SSE signal.

Fig. 2 summarizes the angular dependence of the local
FIG. 3. The angle dependence of (a) the local SSE signal $V_{loc}^s$ and (b) the nonlocal SSE signal $V_{nl}^s$ is shown here for several temperatures between 4 and 50 K. A clear increase of the amplitude of the thermal signal modulation is evident towards lower temperatures. An average offset voltage independent of the magnetic field direction was subtracted from the data.

FIG. 4. The amplitudes of the sino-type modulation in (a) the local and (b) the nonlocal SSE voltage as a function of reciprocal temperature. A clear increase of the signals towards lower temperatures is evident. Interestingly, the local signal $V_{loc}^s$ saturates already around 10 K, while the nonlocal signal $V_{nl}^s$ saturates only around 5 K. The measurements were done with an in-plane magnetic field ($\alpha$-rotation) $\mu_0 H = 2$ T.

Also, the modulation in the local SSE signal vanishes at around 100 K, whereas the nonlocal SSE signal disappears already above 20 K. This suggests that although the general trends are similar, the detailed mechanisms relevant for the local and nonlocal SSE signals might differ. This can be understood considering that the thermal magnon excitation which is important for the local SSE signal, depends only on the temperature gradient $\nabla T_n$ underneath the injector, while the nonlocal signal is caused by magnon transport reaching far beyond the thermal gradient generated by the injected electrode.

So far, the exact nature of the increase of the thermal signal at low temperatures is not fully resolved. In ref. [18], the low-temperature saturation in the nonlocal signal was attributed to a spin-superfluid ground state in the antiferromagnetic Cr$_2$O$_3$, resulting from spontaneous breaking of the uniaxial symmetry. However, a similar behavior was also observed in Pt-YIG-Pt lateral devices, where a spin-superfluid ground state cannot be realized[40]. Furthermore, in MnF$_2$-Pt bilayers, the temperature dependence of the SSE signal shows a magnetic-field-dependent peak around $T \approx 7$ K, which approximately matches the peak in thermal conductivity of MnF$_2$[24]. In the case of Cr$_2$O$_3$, a peak in thermal conductivity was observed for $T \approx 30$ K[41]. Taken together, the low-temperature SSE response in antiferromagnets is far from well understood and requires further investigation.

Finally, we measured the evolution of the nonlocal SSE signal as a function of the injector-detector separation $d_{nl}$, as shown in Fig. 5. The length scale for the magnon spin current can be estimated from this $\Delta V_{nl}^s$ vs $d_{nl}$ data considering a one-dimensional spin diffusion model which describes the decay as[32]

$$\Delta V_{nl}^s = C \exp \left(-\frac{d_{nl}}{l_m}\right),$$

where $l_m$ is the magnon spin diffusion length and $C$ is a constant independent of $d_{nl}$. We find this simple exponential decay of the signal (red line) fits best to the measured data, and we estimate $l_m = 500$ nm at 3 K. This is quite small compared to the magnon spin diffusion length in the ferrimagnetic insulator YIG, where $l_m = 40$ $\mu$m at $T = 3.5$ K was reported using a similar nonlocal method[39]. This is also quite small compared to the magnon spin diffusion length in other antiferromagnets like $\alpha$-Fe$_2$O$_3$ with $l_m = 9$ $\mu$m at 200 K[11]. In
previous spin transport experiments in Cr$_2$O$_3$ thin films, Yuan et al.[18] reported a very large $l_m = 16.3$ μm and assumed a spin-superfluid ground state to be realized in Cr$_2$O$_3$. However, short (< 10 nm) spin decay lengths through AFI were observed in vertical (longitudinal geometry) spin transport devices[42].

In magnon spin transport experiments in YIG, three distinct transport regimes have been identified. For very short distances ($d << l_m$), the magnon signals typically drop faster than the exponential decay (Eq. (1)) [36, 39]. Thereafter, for intermediate distances, an exponential decay is observed [32]. This regime is called the "exponential regime" or "relaxation regime". Beyond this intermediate regime, $\Delta V^S_{nl}$ shows a geometrical decay as $\Delta V^S_{nl} \sim \frac{1}{d^2}$ [39]. In this geometrical-decay regime, the signal is controlled by the temperature gradient $\nabla T$ present close to the detector, which also generates a magnon spin current that contributes to the measured nonlocal SSE signal. The good match of $\Delta V^S_{nl}$ to Eq.[1] shown in Fig. 5 indicates that the spin signal in our case is dominated by magnon relaxation rather than diffusive transport.

In case of a spin-superfluid ground state, the nonlocal signal was predicted to follow a decay of the form [16, 18]

$$\Delta V^S_{nl} \sim \frac{1}{d_{nl} + l_m}. \quad (2)$$

As evident from Fig. 5, in our case, the exponential decay given by Eq. (1) fits much better to the data than an algebraic decay given by Eq. (2).

The relatively short magnon relaxation length along with the strong temperature dependence observed in our experiment suggest that temperature-dependent inelastic magnon scattering processes play an important role in long-range magnon transport through antiferromagnets [21, 43]. Note that from our previous structural and magnetic characterization of α-Cr$_2$O$_3$ thin films, we have found that our films are of high crystalline quality but granular with a typical grain size of ~230 nm [30, 44, 45]. The similar scale $l_m = 38$ nm for sputtered YIG [48], $l_m = 140$ nm for pulsed-laser-deposited YIG [49] and $l_m = 9.4$ μm for liquid-phase-epitaxy YIG were observed [32].

In summary, we performed a detailed study of thermally excited magnon transport in antiferromagnetic α-Cr$_2$O$_3$ thin films using a nonlocal device geometry. No direct electronic excitation of magnons could be observed, while a clear voltage signal arising from thermally generated magnons was picked up in the symmetric voltage. We found that the thermally generated local and nonlocal voltages, measured while rotating a magnetic field of constant magnitude in three orthogonal planes, follow $\sin(\alpha)$ or $\sin(\beta)$ dependencies, as expected for the SSE. Temperature-dependent measurements showed that the SSE signals increase with decreasing temperature and saturate at very low temperatures. Finally, we estimated the length scale over which the thermally generated magnons diffuse by measuring the nonlocal SSE voltage signal as a function of the spatial separation of the injector and detector Pt stripes. We conclude that magnon transport in our α-Cr$_2$O$_3$ thin films is governed mainly by relaxation processes with a characteristic magnon spin diffusion length of $l_m = 500$ nm. The comparatively short magnon spin diffusion length and the strong temperature dependence of both the local and nonlocal SSE signals suggest that inelastic magnon scattering processes at the grain boundary or antiferromagnetic domain walls dominate the magnon transport at short distances in our samples. Our findings can inspire antiferromagnetic magnonic devices, such as non-volatile memory storage, logic gates, analog data processing, and quantum computing.

**ACKNOWLEDGMENTS**

We acknowledge financial support by the Deutsche Forschungsgemeinschaft via SFB 1143/C08, SPP 1538 (Project Nos. GO 944/4, TH1399/5 and MA5144/9-1, MA5144/24-1, MA5144/22-1 and Helmholtz Association of German Research Centres in the frame of the Helmholtz Innovation Lab “FlexiSens” (HIL-A04)). Furthermore, the use of the HZDR Ion Beam Center TEM facilities is gratefully acknowledged. We also thank T. Schönherr for technical support during ebl fabrication of devices.

**DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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