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Magnon diffusion lengths in bulk and thin film Fe<sub>3</sub>O<sub>4</sub> for spin Seebeck applicationsG. Venkat <sup>1,\*</sup> C. D. W. Cox <sup>1</sup> D. Voneshen <sup>2</sup> A. J. Caruana <sup>2</sup> A. Piovano,<sup>3</sup> M. D. Cropper,<sup>1</sup> and K. Morrison <sup>1</sup><sup>1</sup>*Department of Physics, Loughborough University, Loughborough, LE11 3TU, United Kingdom*<sup>2</sup>*ISIS Neutron and Muon Source, Didcot, Oxfordshire, OX11 0QX, United Kingdom*<sup>3</sup>*Institut Laue-Langevin, 6 rue Jules Horowitz, 38042 Grenoble Cedex 9, France*

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The magnon diffusion length (MDL) is understood to play a major role in the thickness dependence of spin Seebeck effect (SSE) voltages in Fe<sub>3</sub>O<sub>4</sub>/Pt thin films. Here we extract the MDL in an Fe<sub>3</sub>O<sub>4</sub> single crystal using inelastic neutron scattering (INS) and in Fe<sub>3</sub>O<sub>4</sub>/Pt thin films using accurate heat flux SSE and static magnetization measurements. The INS MDLs were  $34 \pm 8$  nm at 300 K and  $27 \pm 6$  nm at 50 K. The SSE MDLs decreased with temperature ( $19 \pm 2$  nm at 300 K and  $13 \pm 4$  nm at 50 K), but were markedly smaller. Whilst the bulk MDL is expected to be an upper limit of the MDL in thin films, we show that the film magnetization must be considered to obtain MDLs from SSE measurements. This study highlights the importance of disentangling the role of various effects in SSE measurements which is crucial in increasing the efficiencies of thermomagnetic energy harvesting devices.

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The spin Seebeck effect (SSE) was demonstrated in 2008 by Uchida *et al.* and has since sparked a lot of interest for potential thermoelectric applications [1]. In the longitudinal configuration [2], a spin current is generated along an applied temperature gradient which is normal to the magnetization. As spin currents cannot be detected easily, they are usually converted to charge currents in an adjacent metallic layer with high spin-orbit interaction (e.g., Pt) by the inverse spin Hall effect (ISHE) [2]. This discovery has paved the way for spin caloritronics that includes other thermomagnetic effects such as the spin Peltier and Nernst effects [3,4]. While the efficiency of conventional thermoelectrics is limited by the interplay of thermal ( $\kappa$ ) and electric ( $\sigma$ ) conductivities [5], the SSE is a candidate for improving the performance by allowing for the decoupling of  $\kappa$  and  $\sigma$  in the heavy metal and magnetic material [6]. The SSE is also a viable source of spin currents for spintronic applications [7,8].

Although the SSE was initially demonstrated in a metallic ferromagnet [1], it was later seen in the insulating ferrimagnet yttrium iron garnet (YIG) [9], which has become the benchmark material for SSE studies [2]. There have also been studies of the SSE in half metals such as Heusler alloys [10,11] and Fe<sub>3</sub>O<sub>4</sub>, which are particularly relevant to spintronic applications due to the theoretically proposed 100% spin polarization at the Fermi level [12]. As the oldest known magnetic

material, Fe<sub>3</sub>O<sub>4</sub> has many technological applications [13] and has been attracting attention for SSE devices [14–17].

In an insulator, the SSE is driven by magnon propagation [18] as opposed to being driven by spin polarized conduction electrons in metallic systems [3]. In the former case and for low conductivity magnets like Fe<sub>3</sub>O<sub>4</sub>, an important length scale for quantifying the maximum SSE efficiency is the magnon diffusion length (MDL)  $\Lambda$  [19]. Quantitative knowledge of  $\Lambda$  is also useful for studying fundamentally new physics such as magnon Bose-Einstein condensates [20]. Kehlberger *et al.* extracted  $\Lambda_{\text{YIG}} = 90\text{--}140$  nm from SSE measurements in YIG thin films, using an exponential model [21]. However, microwave measurements on YIG have found significantly larger MDLs  $\sim 400$  nm [22]. In MgO/Fe<sub>3</sub>O<sub>4</sub>/Pt thin films, the MDL was extracted from SSE measurements using the linear response theory (LRT) [23] and found to be  $\Lambda_{\text{Fe}_3\text{O}_4} = 17$  nm at 300 K, increasing to 40 nm at 70 K [14]. Since Fe<sub>3</sub>O<sub>4</sub> seems to show a temperature dependence of the MDL affecting the spintronic efficiency and there is some discrepancy in the MDLs obtained (for YIG), a careful direct measurement of the MDL of Fe<sub>3</sub>O<sub>4</sub> is required.

Another aspect of the SSE is its dependence on the saturation magnetization  $M_s$ . A decrease in SSE in bulk single-crystal YIG/Pt at 175 K has been correlated with an increase in magnetic surface anisotropy [24]. A similar decrease in the SSE in both bulk and thin films of YIG/Pt at 300 K at low magnetic fields has been attributed to the perpendicular magnetic anisotropy and its effect on the ISHE [25]. Thus, for comparing the SSE in different systems, we need to decouple the ISHE from the SSE and study the static magnetization and magnon propagation contributions separately.

In this paper, we measure the magnon dispersion and group velocity in single-crystal Fe<sub>3</sub>O<sub>4</sub> using inelastic neutron scattering (INS). Subsequently, we fit integrated energy cuts of the magnon dispersion to simulated scattered neutron intensity

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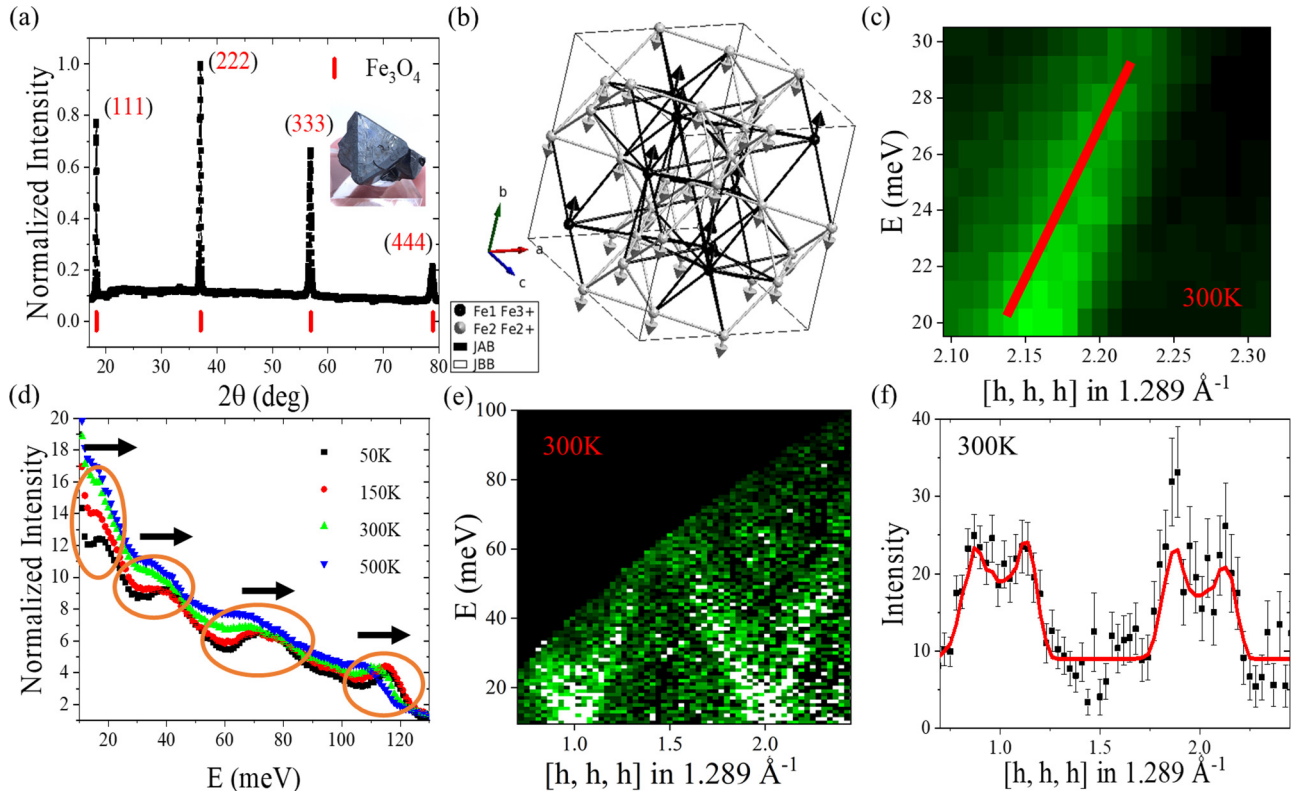


FIG. 1. (a) XRD spectrum of the single crystal showing the [111] orientation. The red dashes mark the positions of the [111] (and harmonics) obtained from the ICSD database (ICSD: 26410). The inset shows a portion of the single crystal used for the measurement. (b) The magnetic structure of  $\text{Fe}_3\text{O}_4$  showing the different magnetic sites, antiferromagnetic exchange  $J_{AB}$  (light grey lines) and ferromagnetic exchange  $J_{BB}$  (black lines) interactions. (c) Magnon dispersion of  $\text{Fe}_3\text{O}_4$  along the [222] direction at 300 K measured using TAS on IN8 at the ILL, Grenoble. The red line is a guide used to extract the group velocity. (d) Energy variation of scattered neutron intensity obtained from MAPS, integrated over all reciprocal space directions. The arrows indicate the blue shift of the magnon modes (marked by ellipses) with decreasing temperature (accompanied by narrowing of the spectra). (e) Magnon dispersion of  $\text{Fe}_3\text{O}_4$  along the [HHH] direction at 300 K measured using TOF spectroscopy on MAPS at ISIS, UK. The almost linear variation of the dispersion is due to the antiferromagnetic exchange  $J_{AB}$  dominating in  $\text{Fe}_3\text{O}_4$ . (f) A line cut along  $\mathbf{q}$  in Fig. 1(e) and integrated in energy from 20–30 meV. This was fit (shown in red) to the simulated scattered neutron intensity for the  $\text{Fe}_3\text{O}_4$  magnetic structure and used to extract magnon linewidths.

profiles to obtain magnon linewidths and diffusion lengths at 300 and 50 K. We then report SSE measurements on  $\text{Fe}_3\text{O}_4$  thin films at the same temperatures normalized to the heat flux  $J_Q = Q/A$  (where  $Q$  is the heat passing through the sample and  $A$  is the sample area) passing through the film. We also measure the variation of the in-plane magnetization with  $\text{Fe}_3\text{O}_4$  thickness and show that it mirrors the trend of the SSE coefficient. We proceed to normalize the SSE coefficient with the magnetization of the films to remove the static magnetization contribution to the measured SSE (introduced via the ISHE) and extract MDLs from the SSE measurements by fitting to the LRT [14] and exponential models [21]. We observe a decrease of the  $\text{Fe}_3\text{O}_4$  MDL as the temperature is decreased, contrary to what was previously reported [14]. In addition, the MDL measured by INS is larger than that determined from SSE measurements, which could be a difference between bulk and thin film MDL values.

We initially used triple axis spectroscopy (TAS) using the IN8 instrument at the ILL, Grenoble to measure the magnon dispersion at 300 K. The measurement was performed on a 5-g naturally grown [111] oriented single crystal

of  $\text{Fe}_3\text{O}_4$ , which was obtained from the Three Peaks mine in Utah, USA and is shown in the inset of Fig. 1(a). The XRD pattern in Fig. 1(a) shows the [111] orientation of the crystal. Since the magnon dispersion in  $\text{Fe}_3\text{O}_4$  is isotropic along [001], [110], and [111] directions [26], measurement along the [111] is representative. Figure 1(b) shows the cubic inverse spinel structure of  $\text{Fe}_3\text{O}_4$  with two different symmetry sites for the Fe ions: tetrahedral A sites occupied by  $\text{Fe}^{3+}$  ions and octahedral B sites populated by a random distribution of  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions. The antiferromagnetic exchange coupling between the ferromagnetic A and B sublattices results in the overall magnetic structure being ferrimagnetic. The magnetic exchange is well approximated by considering the antiferromagnetic exchange constant  $J_{AB}$  and the ferromagnetic exchange constant of the B sublattice  $J_{BB}$  [27]. Figure 1(c) shows the magnon dispersion along the [222] direction for the acoustic mode at 300 K from IN8 from which we obtained a magnon group velocity of  $(14.63 \pm 0.80) \times 10^3$  m/s.

We then used time-of-flight (TOF) INS spectroscopy (on MAPS at the ISIS Neutron and Muon Source) to obtain the magnon dispersion at 50, 150, 300, and 500 K on the same

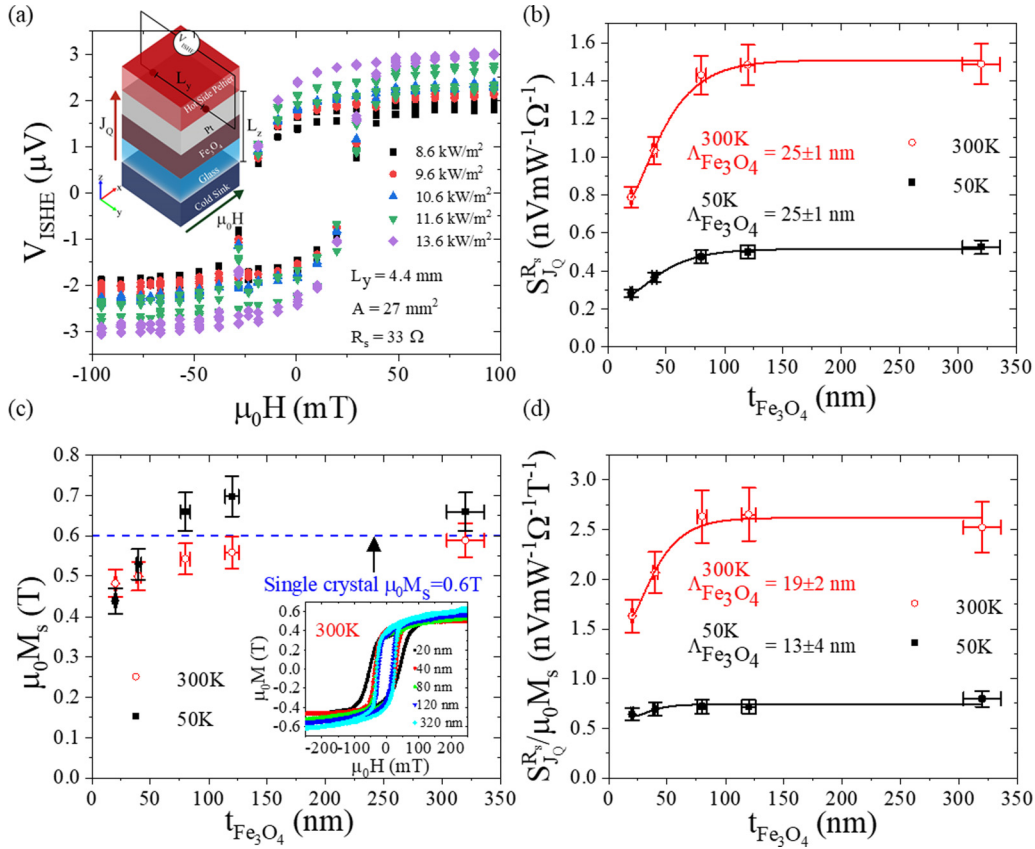


FIG. 2. (a) Example of the raw SSE voltage,  $V_{\text{ISHE}}$ , as a function of applied field  $\mu_0 H$  for different heat fluxes  $J_Q$  at 300 K for the 80 nm film. The inset shows the schematic of the SSE measurement. (b) Variation of  $J_Q$  and sheet resistance normalized SSE coefficient  $S_{J_Q}^{R_s}$  with  $t_{\text{Fe}_3\text{O}_4}$  at 50 and 300 K. The solid lines are fits obtained using Eq. (3). (c) The variation of the saturation magnetic flux density  $\mu_0 M_s$  with  $t_{\text{Fe}_3\text{O}_4}$  at 50 and 300 K. The dashed line marks the single-crystal value of  $\mu_0 M_s$  for  $\text{Fe}_3\text{O}_4$ . The inset shows the variation of the magnetic flux density  $\mu_0 M$  with  $\mu_0 H$  for the films of different  $t_{\text{Fe}_3\text{O}_4}$  at 300 K. A linear diamagnetic background has been subtracted from the datasets. (d) Variation of  $S_{J_Q}^{R_s}/\mu_0 M_s$  with  $t_{\text{Fe}_3\text{O}_4}$  at 50 and 300 K. The solid lines are fits obtained using Eq. (3).

sample. Figure 1(d) shows the scattered neutron intensity (integrated over  $\mathbf{q} \in (-0.2, 0.2)$  r.l.u. in all directions in reciprocal space), as a function of temperature. The detected magnon modes occur as broad peaks in the spectrum and are marked by ellipses. A pronounced blue shift and decrease in magnon linewidth with temperature can be seen, which agrees well with simulated magnon spectrum in other ferrimagnetic systems such as YIG [28]. Figure 1(e) shows the magnon dispersion along the [HHH] direction for the acoustic mode at 300 K. The dispersion slope gave a group velocity of  $v_g^{300\text{ K}} = (13.97 \pm 1.82) \times 10^3$  m/s at 300 K. This is in good agreement with the group velocity extracted from the TAS IN8 measurement.

Subsequently, four cuts along the  $\mathbf{q}$  axis in Fig. 1(e) and integrated along energy (from 20–60 meV in four 10 meV ranges) were simultaneously fit (using the Tobyfit routine [29]) to the simulated scattered neutron intensity for the  $\text{Fe}_3\text{O}_4$  magnetic structure (obtained using SPINW [30]). The fits, shown for the 20–30 meV cut in Fig. 1(f), gave an antiferromagnetic exchange constant of  $J_{\text{AB}} = -4.49 \pm 0.01$  meV and a ferromagnetic exchange constant of  $J_{\text{BB}} = 0.670 \pm 0.002$  meV, which agree well with literature [27]. We also extracted a linewidth of  $\Gamma^{300\text{ K}} = 0.27 \pm 0.05$  meV, which corresponded to a magnon lifetime of  $\tau^{300\text{ K}} = 2.44 \pm 0.45$  ps

at 300 K. Using  $v_g^{300\text{ K}}$ , we obtained a magnon diffusion length of  $\Lambda_{\text{Fe}_3\text{O}_4}^{300\text{ K}} = v_g^{300\text{ K}} \tau^{300\text{ K}} = 34.1 \pm 7.7$  nm. Similarly, the magnon dispersion at 50 K, gave us  $\Lambda_{\text{Fe}_3\text{O}_4}^{50\text{ K}} = 27.2 \pm 5.7$  nm. We thus observe that  $\Lambda_{\text{Fe}_3\text{O}_4}$  is equal within error at 50 and 300 K. We regard these values obtained from bulk single-crystal measurements as upper limits on the MDL in  $\text{Fe}_3\text{O}_4$ .

We now describe the SSE measurements. The samples investigated in this study were  $5 \times 5$  mm<sup>2</sup>  $\text{SiO}_2/\text{Fe}_3\text{O}_4/\text{Pt}$  thin films grown by pulsed laser deposition, with thicknesses  $t_{\text{Fe}_3\text{O}_4} = 20, 40, 80, 120,$  and  $320$  nm and  $t_{\text{Pt}} = 3\text{--}7.5$  nm with a preferential [111] texture [6]. More details of the sample characterization (including x-ray diffraction, reflectivity and fitting with GENX [31]) can be found in the SI [32] and in Ref. [6], while details of the SSE measurement setup is part of a manuscript which is under preparation [33]. Figure 2(a) shows example SSE measurements for the 80-nm film, where the raw voltage,  $V_{\text{ISHE}}$ , as the magnetic field  $\mu_0 H$  was varied is plotted for different applied (and measured)  $J_Q$ . A schematic of the measurement configuration is also shown as an inset. The increase in  $V_{\text{ISHE}}$  with  $J_Q$  is due to the generated spin current being proportional to the temperature gradient,  $\nabla T$ , across the  $\text{Fe}_3\text{O}_4$  layer.

Since the SSE is usually associated with  $\nabla T$ , the SSE coefficient is often defined (with units of V/K) as [34,35]

$$S_{\nabla T} = \frac{V_{\text{ISHE}} L_z}{L_y \Delta T}, \quad (1)$$

where  $L_y$  is the contact separation and  $L_z$  is the sample stack thickness. However, for thin films, it was shown by Sola *et al.* [36] that the thermal resistance between the sample and the hot and cold baths makes  $S_{\nabla T}$  unreliable [35]. In addition, the limitation of  $\nabla T$  measurements sampling the temperature difference across the substrate and active layers can result in stark differences between samples where the substrate thermal conductivity varies [6]. Instead, the SSE coefficient can be defined in terms of the heat flux  $J_Q$  (with units of Vm/W) as [36]

$$S_{J_Q} = \frac{V_{\text{ISHE}}}{L_y J_Q}, \quad (2)$$

where the direct relationship between  $J_Q$  and the temperature difference across the active layer is utilized. We thus consider  $J_Q$  normalization to define the SSE coefficient in this paper and note that  $S_{\nabla T}$  can be obtained from  $S_{J_Q}$  by multiplying with the thermal conductivity of the magnetic layer.

An artefact that can contribute to the measured SSE response is the anomalous Nernst effect (ANE). The ANE has two possible contributions: (a) proximity magnetism induced in the Pt layer [37–39] and (b) the finite (but low) electrical conductivity of  $\text{Fe}_3\text{O}_4$ . Since the Pt thickness in our films was larger than 3 nm, the proximity ANE was considered negligible [39,40]. The heat flux normalized ANE coefficient of a bare 80-nm-thick  $\text{SiO}_2/\text{Fe}_3\text{O}_4$  film was measured to be  $15.9 \pm 2.8 \text{ nVmW}^{-1}$  at 300 K. The resistance of the bare film was  $756 \Omega$  while that of an 80-nm  $\text{Fe}_3\text{O}_4/5 \text{ nm Pt}$  bilayer was  $50 \Omega$ . Using the approach in [14], we obtained a corrected ANE response of  $0.98 \pm 0.17 \text{ nVmW}^{-1}$ , which is  $<2.5\%$  of the SSE signal reported here. We therefore ignore the ANE. This is supported by measurements of Anadon *et al.* who found a negligible ANE contribution to the SSE signal ( $\sim 7\%$ ) [14]. It is also worth noting that there is no ANE from  $\text{Fe}_3\text{O}_4$  at 50 K as it is electrically insulating [41].

Since the SSE signal is dependent on the minor variations in the Pt layer thickness, we further normalize  $S_{J_Q}$  by the sheet resistance of the Pt layer  $R_s$  (measured using the four-probe technique and given in the SI) and define  $S_{J_Q}^{R_s} = \frac{S_{J_Q}}{R_s}$ . The variation of  $S_{J_Q}^{R_s}$  with  $t_{\text{Fe}_3\text{O}_4}$  is shown in Fig. 2(b) at 50 and 300 K. The decrease in SSE coefficient with temperature is consistent with previous reports [14].

We fit the variation of  $S_{J_Q}^{R_s}$  with  $t_{\text{Fe}_3\text{O}_4}$  to that predicted by the linear response theory (for direct comparison with [14]) given by [14,23]

$$S_{J_Q}^{R_s} \propto \frac{(1 - \text{sech}(\delta_{\text{FM}}))(1 - \text{sech}(\delta_{\text{NM}}))}{(\tanh(\delta_{\text{NM}}) + F_s)(\tanh(\delta_{\text{FM}}) + F_m) - G_s G_m}, \quad (3)$$

where  $\delta_{\text{FM}} = \frac{t_{\text{Fe}_3\text{O}_4}}{\Lambda_{\text{Fe}_3\text{O}_4}}$ ,  $\delta_{\text{NM}} = \frac{t_{\text{Pt}}}{\Lambda_{\text{Pt}}}$ ,  $\Lambda_{\text{Pt}}$  is the spin diffusion length of Pt,  $F_s$ ,  $F_m$ ,  $G_s$  and  $G_m$  are material constants. The fits were performed for  $F_m$  and  $\Lambda_{\text{Fe}_3\text{O}_4}$  with  $F_s = G_s = 5$ ,  $G_m = 1$ ,  $t_{\text{Pt}} = 5 \text{ nm}$ , and  $\Lambda_{\text{Pt}} = 7.7 \text{ nm}$  [42] and are shown in Fig. 2(b).

We obtained  $\Lambda_{\text{Fe}_3\text{O}_4}^{300\text{K}} = 25.2 \pm 1.5 \text{ nm}$  and  $\Lambda_{\text{Fe}_3\text{O}_4}^{50\text{K}} = 24.6 \pm 1.2 \text{ nm}$ , which are equal within error. This would suggest that there is no change in the MDL with temperature, in contrast with previous measurements [14].

However, consideration of the in-plane magnetization should also be made due to its impact on the ISHE. The ISHE generated electric field is

$$\mathbf{E}_{\text{ISHE}} \propto \mathbf{J}_s \times \boldsymbol{\sigma}, \quad (4)$$

where  $\mathbf{J}_s$  is the spin current generated along the applied heat flux, in the direction of the surface normal.  $\boldsymbol{\sigma}$  is the spin polarization of the conduction electrons in Pt, which is along the direction of the  $\text{Fe}_3\text{O}_4$  layer magnetization  $\mathbf{M}$  [25]. We measured the in-plane saturation magnetic flux density  $\mu_0 M_s$ , at both 50 and 300 K as a function of  $t_{\text{Fe}_3\text{O}_4}$  [shown in Fig. 2(c)]. We observed a variation similar to that of  $S_{J_Q}^{R_s}$  and thus must be accounted for to extract the thickness dependence of the spin current  $\mathbf{J}_s$ . The inset of Fig. 2(c) shows the variation of the magnetic flux density  $\mu_0 M$  with applied field at 300 K, showing the typical hysteretic behavior. A linear diamagnetic background has been subtracted from the datasets. The single-crystal value of  $\mu_0 M_s$  at both 50 and 300 K is  $\approx 0.6 \text{ T}$  [43] and we can see that for  $t_{\text{Fe}_3\text{O}_4} > 40 \text{ nm}$ ,  $\mu_0 M_s$  for the thin films approach this value. We observe a decrease in  $\mu_0 M_s$  for lower  $t_{\text{Fe}_3\text{O}_4}$  and expect that a decrease in  $t_{\text{Fe}_3\text{O}_4}$  causes an out-of-plane canting of the magnetization and thus a decrease in the in-plane magnetization [44].

To account for the thickness dependence of  $\boldsymbol{\sigma}$  introduced via the ISHE and  $\mu_0 M_s$  in  $S_{J_Q}^{R_s}$ , we monitor the variation of  $\frac{S_{J_Q}^{R_s}}{\mu_0 M_s}$  with  $t_{\text{Fe}_3\text{O}_4}$  [shown in Fig. 2(d)], which from Eq. (4) is proportional to  $\mathbf{J}_s$ . We observe that while a decreasing trend for lower  $t_{\text{Fe}_3\text{O}_4}$  is still evident at 300 K, it is considerably reduced at 50 K, indicating that there is a larger contribution from  $\mu_0 M_s$  to the SSE response at this temperature. We also fit these trends (of the variation of  $\frac{S_{J_Q}^{R_s}}{\mu_0 M_s}$  with  $t_{\text{Fe}_3\text{O}_4}$ ) to that predicted by Eq. (3) [also shown in Fig. 2(d)] and obtained  $\Lambda_{\text{Fe}_3\text{O}_4}^{300\text{K}} = 19.2 \pm 2.3 \text{ nm}$  and  $\Lambda_{\text{Fe}_3\text{O}_4}^{50\text{K}} = 12.6 \pm 4.0 \text{ nm}$ . These values show the same trend as the decreased MDL at low temperatures observed in YIG thin films which has been attributed to the reduction in the magnon thermal velocity with decrease in temperature [45]. We believe that these are accurate estimates of the MDLs in  $\text{Fe}_3\text{O}_4$  thin films after accounting for the effect of the decrease in  $\boldsymbol{\sigma}$  with decreasing film thickness. We have also fit the thickness variation of  $\frac{S_{J_Q}^{R_s}}{\mu_0 M_s}$  to the exponential model described in Refs. [21,46] (more details can be found in the SI) and obtain similar values for  $\Lambda_{\text{Fe}_3\text{O}_4}$  (within error).

The MDL values estimated from thin film SSE measurements are around half the values estimated from bulk single-crystal INS measurements. This could be a result of defects or due to the presence of a minor  $\alpha$ -Fe phase (seen in the XRD spectrum in Fig. S3 in Ref. [32]) in the thin films, which can cause higher magnon scattering/damping and a lowering of the MDL. Nevertheless, INS measurements provide an upper limit on the expected MDL at 300 and 50 K. We also note that we find no evidence of the increase in MDL with decreasing temperature, which was reported in Ref. [14], from both INS and SSE estimates. We expect that some of the

trends observed in Ref. [14] are skewed by the uncertainties introduced by considering  $S_{VT}$  as the SSE coefficient.

In conclusion, we have measured the magnon dispersion in a single crystal of  $\text{Fe}_3\text{O}_4$  using inelastic neutron scattering as a function of temperature and observe that the extracted MDLs  $\Lambda_{\text{Fe}_3\text{O}_4}^{300\text{ K}} = 34.1 \pm 7.7$  nm and  $\Lambda_{\text{Fe}_3\text{O}_4}^{50\text{ K}} = 27.2 \pm 5.7$  nm. We also measured the heat flux normalized SSE in  $\text{SiO}_2/\text{Fe}_3\text{O}_4/\text{Pt}$  thin films and normalize by the static magnetization contribution to the SSE (introduced via the ISHE) before determining the MDLs  $\Lambda_{\text{Fe}_3\text{O}_4}^{300\text{ K}} = 19.2 \pm 2.3$  nm and  $\Lambda_{\text{Fe}_3\text{O}_4}^{50\text{ K}} = 12.6 \pm 4.0$  nm, which are smaller than that obtained from INS measurements. We hope that these studies will highlight the importance of disentangling the role of various effects in SSE measurements as well as motivate further studies to relate the SSE to the variation in  $\mu_0 M_s$  and MDL in magnetic thin films.

All supporting data will be made available via the Loughborough data repository [47]. The MAPS data is available in Ref. [48]. The IN8 data is available in Ref. [49].

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