Pseudospin transport in the $J_{\text{eff}} = 1/2$ antiferromagnet Sr$_2$IrO$_4$

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Abstract – Spin transport by itinerant electrons and collective excitations of localized spins with small relaxation rates is of eminent interest for both fundamental research and applications. Spin-orbit coupling (SOC) is not only considered a crucial origin for spin relaxation in spin transport, it recently emerged as the source of novel quantum phases such as topological insulators or SOC-induced Mott insulators with $J_{\text{eff}} = 1/2$ pseudospins. Here we show that emergent pseudospin excitations in Sr$_2$IrO$_4$ give rise to significant heat transport despite this compound being a strong SOC-induced Mott insulator. The analysis of the heat conductivity reveals boundary-limited relaxation of the pseudospin excitations at low temperature. However, the relaxation rate dramatically increases upon heating towards room temperature due to thermally activated scattering off phonons. The comparison of this result with findings for cuprate analogs with $S = 1/2$ spin excitations suggests a radically stronger coupling of the $J_{\text{eff}} = 1/2$ pseudospin excitations to the lattice.

Introduction. – The magnetic heat conductivity is generally considered an important tool to probe quantum spin and even topological excitations [1]. In the past years, this particular sensitivity has been exploited extensively for probing the elementary spin excitations in many different $S = 1/2$ low-dimensional quantum magnets [2–11]. For materials with a strong SOC and $J_{\text{eff}} = 1/2$ pseudospins [12] it remains however unclear whether heat transport can be used to probe the pseudospin excitations at all, because of the pseudospin relaxation is unknown. Successful experiments on strong SOC materials are lacking, apart from pioneering attempts [13].

Iridium oxide compounds play a prominent role among the SOC-induced Mott insulators because, among others, square, honeycomb, and hyper-kagome lattice types [14–18] are known as host for $J_{\text{eff}} = 1/2$ pseudospin quantum phases. One of the up to the present best studied iridate materials is the compound Sr$_2$IrO$_4$ which is a prototype SOC-induced Mott insulator [15] with localized electrons on the Ir$^{4+}$ ions in a $J_{\text{eff}} = 1/2$ state. The material possesses a square lattice of Ir$^{4+}$ ions formed by corner-sharing IrO$_2$ plaquettes, where adjacent IrO$_2$-planes are separated by SrO layers [19], very similar structure of La$_2$CuO$_4$. A strong antiferromagnetic exchange of the order $J \sim 0.06$ eV couples the $J_{\text{eff}} = 1/2$ pseudospins giving rise to two-dimensional (2D) magnetic excitations as is revealed by resonant inelastic x-ray scattering (RIXS) [20]. Sr$_2$IrO$_4$ orders long-range antiferromagnetically at $T_N \approx 224$ K, where a weak ferromagnetic moment occurs due to canting of the Ir$_6$ octahedra [12,16,21].

In order to probe the significance of $J_{\text{eff}} = 1/2$ pseudospin heat transport we performed heat conductivity measurements on Sr$_2$IrO$_4$. Apart from being a prototypical SOC-induced Mott insulator, as already mentioned, this material is particularly well suited for such studies because single crystals are available. Our most striking finding is clear-cut evidence for magnetic in-plane heat transport in this $J_{\text{eff}} = 1/2$ compound which derives from a highly unusual temperature dependence of the heat conductivity that is absent for the out-of-plane direction. Furthermore, our analysis in terms of a Boltzmann-type approach reveals that the magnetic mean free path $l_{\text{mag}}$ at low temperature is limited by boundary scattering. Upon
heating to room temperature, $l_{\text{mag}}$ decreases by an order of magnitude, indicating a temperature-activated scattering due to strong magneto-elastic coupling.

**Sample preparation and experiment.** – The growth and characterization of single crystals of Sr$_2$IrO$_4$ have been described in ref. [14]. The crystal dimension in our experiment was $0.51 \times 0.87 \times 0.17$ mm$^3$ where the shortest edge is the crystallographic $c$-direction. The thermal conductivity $\kappa$ has been measured with a home-made device in a four-point configuration using a chip resistor as heater and a thermocouple to measure the temperature gradient parallel to the $ab$-plane [22]. Due to the limited size of the thin plate-like single crystal it was impossible to perform a four-point measurement along the $c$-direction. Nevertheless, a two-point measurement was possible. For our setup we carefully investigated the differences between the two- and four-point configuration. For the two-point configuration we found, that above $\sim150$ K the temperature dependence of the heat conductivity is reproduced correctly with the caveat of an uncertain absolute value. Therefore, we consider data from the two-point measurement only to search for anomalous temperature dependence in the vicinity of $T_N$.

**Results.** – Figure 1 shows the measured heat conductivity of Sr$_2$IrO$_4$ along the $ab$ and $c$ directions, $\kappa_{ab}$ and $\kappa_c$, respectively. From resistivity measurements [23,24] and applying the Wiedemann-Franz-law it follows that Sr$_2$IrO$_4$ has a low contribution of electrons to the heat conductivity which is three orders of magnitude lower than the measured $\kappa$. Therefore, we neglect the contribution of the electrons and consider the total heat conductivity in $ab$-direction as the sum of a conventional phononic and a potential magnetic contribution, whereas $\kappa_c$ is purely phononic.

The in-plane heat conductivity $\kappa_{ab}$ exhibits a peak at low temperature ($\sim12$ K), which is followed by a steep decrease that slightly levels off at around 75 K. At further increased temperature a broad step around $T_N$ is observed and the curve almost saturates close to room temperature. This temperature dependence is incompatible with canonical phononic heat conduction. In a simple approach the heat conductivity is proportional to the specific heat $c_v$, the velocity $v$, and the mean free path $l$ of the heat carriers, i.e.

$$\kappa \sim c_v v l. \quad (1)$$

In the case of phonons as heat carriers, the velocity and the mean free path are approximately constant at low temperature and $\kappa$ follows the temperature dependence of the specific heat. At higher temperatures, umklapp scattering becomes important which reduces the mean free path and thus leads to the observed low-temperature peak. This process depends on the number of excited phonons and leads to $l \propto 1/T$. Thus, at high temperatures where $c_v$ approaches the Dulong-Petit constant [24], the phononic heat conductivity is approximated by $\kappa_{\text{phon}} \propto 1/T$ [25].

The leveling off at $\sim75$ K and the broad step-like feature near $T_N$ are clearly inconsistent with this expected temperature dependence. Two completely different scenarios are conceivable for explaining this unexpected behavior. It is possible that enhanced scattering of phonons occurs due to critical magnetic fluctuations near $T_N$. In fact, a dip structure near $T_N$ is often found in antiferromagnetic materials like in MnO [26] or in CoF$_2$ [27]. However, such critical fluctuations are unlikely to affect $\kappa_{\text{phon}}$ near 75 K, i.e. far away from $T_N$. Moreover, the phonon scattering due to magnetic fluctuations typically affects the heat conductivity isotropically even in layered systems [28]. Therefore, we carefully inspected the heat conductivity parallel to the $c$-axis in the vicinity of $T_N$. At temperatures higher than $\sim200$ K, the $\kappa_c$ curve is absolutely featureless and fully described by the aforementioned $1/T$-law as is indicated in the figure. Thus, we can clearly rule out a phononic origin of the anomalous behavior in $\kappa_{ab}$. On the other hand, the anomalous behavior can also arise from a 2D magnetic heat conductivity within the IrO$_2$-layers which adds to $\kappa_{\text{phon}}$, i.e., $\kappa_{ab}$ results from the sum of phononic and magnetic contributions while $\kappa_c$ is purely phononic. Indeed, such magnetic heat transport is frequently observed in low-dimensional $S = 1/2$ quantum magnets [2–11]. Thus, we conclude that in Sr$_2$IrO$_4$ a magnetic contribution to the heat conductivity is present in $\kappa_{ab}$.
Having established this main experimental finding, namely the first observation of magnetic heat conductivity in a $J_{\text{eff}} = 1/2$ system, we move on to its quantitative analysis by extracting the magnetic mean free path. A phenomenological $T^{-1}$ approach is used to model the phononic heat conductivity at high temperatures (cf. the solid line in fig. 1). To estimate the uncertainty of the phononic background in $\kappa_{\text{ph}}$, we performed extreme phononic fits for determining lower and upper bounds as is indicated by the shaded area\(^1\).

In the temperature range between $T_N$ and $\sim 50$ K the measured $\kappa$ exceeds the expected phononic heat conductivity remarkably, corroborating our conclusion of a significant $\kappa_{\text{mag}}$. We subtracted the phononic fit from the measured $\kappa$ and obtain $\kappa_{\text{mag}}$ as shown in fig. 2. $\kappa_{\text{mag}}$ increases with decreasing temperature up to $\sim 150$ K with a maximum value of about 1.8 W/Km, and decreases for higher temperatures. If one considers the coarse generic behavior of heat conductivity given by eq. (1), the low-temperature increase arises from the thermal occupation of pseudospin excitations (termed magnons hereafter). The peak at 150 K and the high-temperature decrease of $\kappa_{\text{mag}}$ cannot be related to the maximum of the specific heat of 2D spin excitations, as the latter is expected constant in a 2D $S = 1/2$ Heisenberg model, i.e. at much higher temperatures than considered here. Instead, the decrease must be primarily related to a temperature dependence of the mean free path since the average magnon velocity is unlikely to change strongly at the rather low temperatures ($T < J/k_B$, with $J = 0.06$ eV [20,31,32]) considered here.

For investigating our result for $\kappa_{\text{mag}}$ further, we follow an approach that has previously been used to analyze $\kappa_{\text{mag}}$ of the $S = 1/2$ analog La$_2$CuO$_4$ [9], i.e., we extend the simple kinetic description of eq. (1) by accounting for possible momentum dependences in two dimensions [9,11], i.e. $\kappa \propto \int \epsilon_k v_k l_k d\mathbf{k}$, with $\epsilon_k = \epsilon(k)$ the specific heat ($\epsilon(k)$ and $v_k$ are the energy and the Bose occupation function of the mode $k$), $v_k$ the velocity and $l_k$ the mean free path of a magnon with wave vector $\mathbf{k}$.

In Sr$_2$IrO$_4$, the dispersion $\epsilon_k$ is a steeply increasing function with band maxima at $\epsilon > 100$ meV = $k_B$ . 1160 K [20]. Thus, at the low temperatures considered here, primarily modes with small momenta are relevant for the heat transport. For simplicity, we therefore assume a temperature-independent mean free path $l_{\text{mag}} = l_k$. We approximate the dispersion $\epsilon_k = \epsilon_k = \sqrt{\Delta^2 + (hv_0k)^2}$ [9], where $\Delta = 0.83$ meV is the experimental anisotropy gap revealed by ESR [33] and $v_0 = (6.25 \pm 0.4) \cdot 10^5$ m/s the small-k magnon velocity extracted from the RIXS single magnon dispersion [20]. The heat conductivity of the magnons can then be estimated in a two-dimensional

\[^1\text{Note that the lower bound found from a } 1/(T - T_0) \text{ fit resembles the high-temperature behavior found by a fit to the Callaway model [29].}\]
The strong magnon-phonon scattering evident in our data reveals a qualitative difference of the pseudospin heat transport of Sr$_2$IrO$_4$ and the spin heat transport of the almost isostructural and thus closely related $S = 1/2$-system La$_2$CuO$_4$ [9], as is inferred from a direct comparison, cf. fig. 4. Both the spin wave velocity of La$_2$CuO$_4$ [43] and the low-temperature magnon mean free path ($\sim 56$ nm) of the sample considered in the figure [9] are approximately twice as large as those of our Sr$_2$IrO$_4$.

**Discussion.** It is instructive to compare the mean free path with the spin-spin correlation length $\xi$ measured by resonant x-ray diffusive scattering [36], see inset of fig. 3. This quantity is a conceivable natural upper limit for $l_{\text{mag}}$. In the long-range ordered phase below $T_N$, the spin-spin correlation length is infinitely large and thus unimportant for the magnetic heat transport. Above the $T_N$, the system becomes truly two-dimensional, and the magnetic structure is then defined by the correlation length $\xi$ [42], i.e., the system becomes non-paramagnetic but stays antiferromagnetically correlated at length scales $\lesssim \xi$. With increasing temperature, $\xi(T)$ decreases rapidly but in Sr$_2$IrO$_4$ it remains still more than an order of magnitude larger than the determined $l_{\text{mag}}$. Therefore, it plays only a minor role in limiting the magnetic heat conductivity above $T_N$, if any. This suggests that the seeming anomaly in $\kappa_{\text{ab}}$ around $T_N$ (cf. fig. 1) is mostly unrelated to the onset of magnetic ordering but rather connected with the growing importance of temperature-activated magnon scattering. Note, however, that a faint change of slope is discernible in the semilogarithmic representation of $l_{\text{mag}}$ shown in the inset of fig. 3.

The shaded area marks the range spanned by the lower and upper bound for the phononic heat conductivity. At low temperatures the constant magnetic mean free path is found to be $l_0 = (32 \pm 10)$ nm. The dashed line is the phenomenological fit to $l_{\text{mag}}(T)$ by eq. (3) with $T^* = 482$ K and $A_S = 1.4 \cdot 10^7$ K$^{-1}$m$^{-1}$. The inset shows the magnetic mean free path together with the spin-spin correlation length $\xi$ (open squares) determined by resonant x-ray diffusive scattering [36] on a semilogarithmic scale.

The strong decrease at elevated temperatures clearly signals the onset of a temperature-activated scattering process. We assume that both the low-temperature boundary scattering and the temperature-activated process are independent of each other. Following Matthiessen’s rule, the mean free path can then be written as

$$l_{\text{mag}}^{-1} = l_0^{-1} + \left(\frac{\exp(T^*/T)}{A_S T}\right)^{-1},$$

where the second term is an empirical formula for temperature-activated scattering in magnetic heat transport that has been successfully used in one-dimensional systems [4,6,38,39] (with $T^*$ the characteristic energy scale of the temperature-activated scattering process and $A_S$ a proportionality factor). As can be seen in the figure, this formula fits the experimental $l_{\text{mag}}(T)$ quite well. The fit yields $T^* \sim 480$ K (see footnote 2) which should be considered as a very coarse estimate of the energy of

$^2$We estimate the uncertainty of the fit result by separately fitting the upper and lower bounds of the grey-shaded area, which yield $T^* = 450$ K with $A_S = 0.8 \cdot 10^7$ K$^{-1}$m$^{-1}$ and $T^* = 555$ K with $A_S = 5.2 \cdot 10^7 \cdot 10^9$ K$^{-1}$m$^{-1}$.
sample. Considering these parameters and eq. (2), the almost identical low-temperature increase of both curves at $T \lesssim 100$ K can be understood as the consequence of dominating magnon-boundary scattering in both cases. However, upon further increasing $T$, a strong suppression of $\kappa_{\text{mag}}$ of Sr$_2$IrO$_4$ occurs while that of La$_2$CuO$_4$ continues to increase up to room temperature. Apparently, the magnon-phonon scattering in Sr$_2$IrO$_4$ is dramatically stronger than that in La$_2$CuO$_4$, despite similar phonon spectra in both compounds [31,40,44]. This unambiguously evidences a peculiar and particularly strong nature of the magneto-elastic coupling in Sr$_2$IrO$_4$, arising from the large SOC and the resulting entanglement of spin and orbital degrees of freedom [12].

**Conclusion.** – In summary, our data provide the first experimental result for low-dimensional $J_{\text{eff}} = 1/2$ pseudospin heat transport in an iridate compound. Our data show that the magnetic heat conductivity remains a valuable tool to probe the generation and the scattering of magnetic excitations also in these systems. At low temperatures $T \lesssim 100$ K, the magnetic heat transport $\kappa_{\text{mag}}$ is dominated by magnon scattering off static boundaries and thus comparable with that of 2D $S = 1/2$ systems. However, at higher temperatures unusual strong magnon-phonon scattering becomes increasingly important, highlighting the peculiar nature of the pseudospin moments and excitations.

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