Heat conduction in low-dimensional quantum magnets

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Abstract. Transport properties provide important information about the mobility, elastic and inelastic of scattering of excitations in solids. Heat transport is well understood for phonons and electrons, but little is known about heat transport by magnetic excitations. Very recently, large and unusual magnetic heat conductivities were discovered in low-dimensional quantum magnets. This article summarizes experimental results for the magnetic thermal conductivity $\kappa_{\text{mag}}$ of several compounds which are good representations of different low-dimensional quantum spin models, i.e. arrangements of $S=1/2$ spins in the form of two-dimensional (2D) square lattices and one-dimensional (1D) structures such as chains and two-leg ladders. Remarkable properties of $\kappa_{\text{mag}}$ have been discovered: It often dwarfs the usual phonon thermal conductivity and allows the identification and analysis of different scattering mechanisms of the relevant magnetic excitations.

1 Introduction

Heat transport by magnetic excitations was originally predicted in 1936 [1]. However, it took almost 30 years until the first convincing experimental evidence for magnetic heat transport by classical spin waves was found in ferrimagnetic yttrium-iron-garnet (YIG) [2,3,4,5]. In principle, the analysis of this magnon heat conductivity should yield valuable information about the excitation and scattering of magnons (e.g. off defects, phonons, and electrons) as is the case for the well-understood phononic and electronic thermal conduction [6]. However, most of the early experiments on YIG and following experiments on other materials [7,8,9] were restricted to magnetically ordered phases at very low temperature ($T < 10$ K). The first signature of magnetic heat transport at higher temperatures ($T > 50$ K) was observed for the one-dimensional quantum antiferromagnet KCuF₃ [10]. However, only the recent theoretical prediction of dissipationless heat conduction in one-dimensional antiferromagnetic Heisenberg chains [11,12] and the discovery of huge magnetic contributions in the quantum spin ladder material Sr₁₄Cu₂₄O₄₁ [13,14,15] triggered intense research on the heat transport of low-dimensional quantum spin systems [11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27,28,29,30,31,32,33,34,35,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52,53,54,55,56,57,58,59,60,61,62,63,64,65]. Over the course of this work, more and more cases for low dimensional magnetic heat conduction were observed in various materials. Today, the clearest experimental examples of low dimensional magnetic heat conduction are found in copper oxide (cuprate) systems. The overview of the experimental research status on low dimensional magnetic heat conduction provided in this article therefore focuses on these compounds.

Particular examples from the plethora of possible spin structures in cuprate systems are spin arrangements in the geometrical form of chains, so-called two-leg ladders, and square lattices with a strong antiferromagnetic Heisenberg exchange ($J \approx 1500 - 2000$ K) between

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Fig. 1. Illustration of low-dimensional spin structures: (a) a spin chain, (b) a two-leg spin ladder, and (c) a two-dimensional square lattice. Arrows represent localized $S = 1/2$ spin and shaded bars symbolize strong antiferromagnetic exchange between them. (d) Schematic illustration of the underlying chemical building block giving rise to the spins and their interaction. Only the relevant Cu $3d_{x^2-y^2}$ and O $2p_x$ orbitals are indicated. Arrows represent the spins of the electrons involved.

nearest neighbor spins. Sketches of such spin arrangements are shown in Fig. 1a-c. These low-dimensional spin structures usually arise from similar low dimensional Cu-O structures, in which the antiferromagnetic exchange originates from straight Cu-O-Cu bonds (bonding angle: 180°) as depicted in Fig. 1d. All these systems are based on Cu$^{2+}$-ions and therefore represent $S = 1/2$ systems with a strong quantum nature.

Good examples for materials containing $S = 1/2$ Heisenberg chains as depicted in Fig. 1a are given by the compounds CaCu$_2$O$_3$, SrCuO$_2$ and Sr$_2$CuO$_3$, where straight Cu-O-Cu bonds and hence a strong antiferromagnetic exchange only exist along one particular crystallographic direction; the magnetic exchange perpendicular to this direction is much weaker [56,57]. In the (Sr, Ca, La)$_{14}$Cu$_{24}$O$_{41}$ family of compounds, parallel pairs of such chains are coupled to each other via bridging O-ions, producing in straight Cu-O-Cu bonds perpendicular to the chain direction. The resulting magnetic interchain coupling perpendicular to the chain direction $J_\perp$ is of a similar magnitude to the intrachain coupling, i.e. $J_\perp \approx J$ [58]. This spin ensemble is a so-called two-leg spin ladder, the ladder legs being formed by the two chains and the ladder rungs arising from the Cu-O-Cu bonds which connect the chains (cf. Fig. 1b). Following this concept, ladder structures with more legs can in principle be created by coupling more chains to the structure; eventually this would lead to a two dimensional Heisenberg antiferromagnet on a square lattice (2D-HAF) in the infinite limit. A good realization of a 2D-HAF with $S = 1/2$ is given by La$_2$CuO$_4$ and other related antiferromagnetic parent compounds of high-temperature superconductors.

The corresponding low-dimensional quantum spin models are characterised by very peculiar ground state properties and elementary excitations, which vary strongly from system to system. Spin-spin correlations of homogeneous spin chains, for example, are quasi-long range in the ground state and decay algebraically with distance between the spins [59]. The elementary excitations, so-called spinons, are gapless and carry a spin $S = 1/2$ [60]. In contrast to this, the ground state of a two-leg ladder is a spin liquid, i.e. the spin-spin correlations are short range and decay exponentially as a function of distance. The elementary excitations are $S = 1$ particles (usually called magnons or triplons) and possess a spin gap $\Delta$ ($\Delta/k_B \approx 400$ K in the case of the systems discussed here) [58]. Finally, the ground state of the 2D-HAF is a Néel state with strongly reduced sublattice magnetization, which only exists at temperature $T = 0$ [61]. In this case the elementary excitations are well described using a spin wave framework. However, it should be noted that alternative descriptions have been discussed [62,63,64].

In the case of hole doping, all these model systems yield interesting and exotic properties. A Luttinger liquid forms in hole-doped spin chains, i.e. electronic excitations decay into collective excitations of holes (holons) and spins (spinons). This phenomenon is usually called spin-charge separation. Radically different properties have been predicted for two-leg spin ladders; superconductivity competing with a charge ordered ground state is expected in this case [65,66]. Finally, hole doping has great importance in the case of the 2D-HAF as is evident from the high temperature superconductivity which is observed in such systems. Note that controlled hole doping of $S = 1/2$ chains and the observation of spin-charge separation signatures has not
yet been achieved experimentally, whereas charge ordering and superconductivity are prominent experimental features of hole-doped spin ladder and 2D-HAF materials.

Concerning heat transport, little is known for all these model systems. Often the attention in theoretical works is focussed on the possibility of ballistic magnetic heat transport in 1D-systems: in integrable models like the $XXZ$ Heisenberg spin chain the Hamiltonian and the thermal current operator commute, i.e. once a thermal current is established in such a system it will never decay $[11]$. In other words, the thermal resistance vanishes and the magnetic thermal conductivity $\kappa_{\text{mag}}$ diverges. While such surprising properties are well established for integrable spin models $[11]$, ballistic heat transport in non-integrable quasi 1D-systems (e.g. two-leg spin ladders) is currently a subject of intense discussion $[38,46,39,55]$. However, in real materials scattering processes involving defects and other quasiparticles such as phonons and charge carriers must play an important role and render $\kappa_{\text{mag}}$ finite in all cases $[49]$. The analysis of $\kappa_{\text{mag}}$ should hence provide further insight into the nature of these scattering processes and the dissipation of magnetic heat currents.

### 2 Experimental signatures of magnetic heat conduction

Fig. 2 shows experimental results $[14,15,17,22]$ for the thermal conductivity $\kappa$ of CaCu$_2$O$_3$, Sr$_{14}$Cu$_{24}$O$_{41}$ and La$_2$CuO$_4$ which are good experimental representations of $S = 1/2$ isotropic antiferromagnetic Heisenberg spin chains, two-leg spin ladders and the 2D-HAF, respectively. The experimental thermal transport properties of all these electrically insulating materials are intriguing: when $\kappa$ is measured perpendicular to the low dimensional structure ($\kappa_{\perp}$), i.e. a direction along which the magnetic coupling is negligible, the $T$-dependence of ordinary phonon thermal conductivity $[6]$ $\kappa_{\text{ph}}$ is found for all three materials: $\kappa_{\perp}$ exhibits a peak at low temperature $T \approx 20$ K, which is followed by a continuous decrease as $T$ rises further. Note the exception in CaCu$_2$O$_3$, where one component of $\kappa_{\perp}$ increases monotonically with rising $T$. Here, a strong suppression of $\kappa_{\text{ph}}$ due to disorder and possible contributions from optical phonons could give rise to the observed $T$-dependence $[23]$. The situation is completely different for $\kappa$ parallel to the low-dimensional structures, i.e. along the directions with large $J$ ($\kappa_{\parallel}$). Again, a phononic low-$T$ peak is observed. However, $\kappa_{\parallel}$ evolves very differently at higher $T$. At $T \gtrsim$ 75 K, $\kappa_{\parallel}$ strongly increases upon heating and exhibits a peak for Sr$_{14}$Cu$_{24}$O$_{41}$ and La$_2$CuO$_4$ at 140 K and 310 K, respectively, while for CaCu$_2$O$_3$ the increase continues up to

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1. Despite the spin ladder compound Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$ being intrinsically hole doped, electronic contributions to $\kappa$ are negligible.
2.1 Extraction of magnetic contributions

In order to obtain the magnitude and $T$-dependence of $\kappa_{\text{mag}}$, it is essential to accurately estimate $\kappa_{\text{ph}}$ and subtract it from the total $\kappa_{\parallel}$. In the case of $\text{La}_2\text{Cu}_4\text{O}_4$ and $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ this can be performed in a convenient way since the magnetic contributions are expected to be negligibly small in comparison to $\kappa_{\text{ph}}$ in the vicinity of the phononic peak. This is due to the expected $T$-dependence of $\kappa_{\text{mag}}$ being approximately $\propto T^2$ and $\propto \exp(-\Delta/T)$ for the 2D-HAF and the spin ladders, respectively. We may hence estimate $\kappa_{\parallel} \approx \kappa_{\text{ph}}$ at $T \lesssim 40$ K and to extrapolate the low temperature $\kappa_{\text{ph}}$ to higher $T$. The thus estimated $\kappa_{\text{ph}}$ is indicated in Fig. 2b and 2c as solid lines. For details of the procedure the reader is referred to Refs. [14] [15] [17]. In the case of $\text{CaCu}_2\text{O}_3$ this procedure is not applicable since $\kappa_{\text{mag}} \propto T$ is expected for a $S = 1/2$ Heisenberg chain hence providing significant contributions to $\kappa_{\parallel}$ even at low $T$. However, in the present case of $\text{CaCu}_2\text{O}_3$ the situation is quite fortunate, since $\kappa_{\parallel} \gg \kappa_{\perp} \approx \kappa_{\text{ph}} \approx \text{const}$ at $T \gtrsim 100$ K which allows the extraction of $\kappa_{\text{mag}}$ from the total $\kappa_{\parallel}$ at temperatures higher than 100 K simply by subtracting a constant value [23]. For all three cases the conjectured $\kappa_{\text{mag}}$ are shown in Fig. 3. The figure also shows $\kappa_{\text{mag}}$ of $\text{La}_5\text{Ca}_9\text{Cu}_{24}\text{O}_{41}$, which belongs to the same family of two-leg spin ladder compounds as $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ but differs by a lower content of charge carriers in the ladders (cf. discussion in Section 3.3).

3 Analysis of magnetic heat conductivity

We start our analysis by considering the qualitative $T$-dependence of $\kappa_{\text{mag}}$, which comprises a simple peak structure ($\text{La}_2\text{Cu}_4\text{O}_4$ and $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$) and a monotonic increase ($\text{CaCu}_2\text{O}_3$) in the highest temperature measured. In these three cases, the remarkable anisotropy of $\kappa$ is the qualitative evidence for large magnetic contributions to $\kappa_{\parallel}$, i.e. for magnetic heat conductivity $\kappa_{\text{mag}}$. A similar anisotropy is also present for $\kappa_{\parallel}$ of the spin chain materials ($\text{Sr}_{\text{Cu}}\text{O}_2$ and $\text{Sr}_2\text{Cu}_3\text{O}_4$ which led to the conclusion that spinon heat transport is also present in these materials [26] [20] [21]. However, as will be discussed in Section 3.4 a high-$T$ peak is absent in those cases which renders a quantitative analysis of phononic and magnetic contributions to $\kappa_{\parallel}$ more difficult. However, a clear separation of $\kappa_{\parallel}$ into magnetic and phononic parts ($\kappa_{\text{mag}}$ and $\kappa_{\text{ph}}$) is possible for the three cases shown in Fig. 2b where the strong features of $\kappa_{\text{mag}}$ appear at a much higher $T$-scale than the low-$T$ phonon peak. In the following sections we will therefore focus on these cases and examine what can be learned.
studied range \( T = 100-350 \) K, where the latter may be regarded as the low temperature edge of a peak. A peak structure is very common for the thermal conductivity \( \kappa \) of any kind of heat carrying particle, such as phonons or electrons \([6]\). In the following we will investigate whether the underlying physics can be applied to magnetic excitations as well. The basic physics which determines the \( T \)-dependence of \( \kappa \) can be inferred from the kinetic estimate

\[
\kappa = \frac{1}{d} \frac{1}{(2\pi)^d} \int c_k v_k l_k d\mathbf{k},
\]

(1)

with \( d \) the dimensionality of the considered system, \( c_k = \frac{d}{\Delta} \epsilon_k \eta_k \) the specific heat (\( \epsilon_k \) and \( \eta_k \) are the energy and the statistical occupation function of the mode \( \mathbf{k} \)), \( v_k \) the velocity and \( l_k \) the mean free path of a particle with wave vector \( \mathbf{k} \). At low \( T \) only a few particles are excited and contribute to the heat transport. Often scattering processes are rare and \( l_k \) is a slowly varying function of momentum in the relevant energy range. In this situation, the low-\( T \)-increase of \( \kappa \) is (a) characteristic of the excitation of the heat carrying particle (reflecting the \( T \)-dependence of the specific heat if \( v_k \) is momentum independent) and (b) proportional to the mean free path \( l \approx l_k \). At higher \( T \) the momentum-dependent scattering becomes more important and leads to a decrease of the mean free path and hence to a decrease of \( \kappa \). Normally this decrease is characteristic of the relevant scattering mechanisms and allows an advanced analysis.

The application of Eq. (1) for the case of 1D and 2D magnetic systems leads to the general result

\[
\kappa_{\text{mag}}(T) \propto l_{\text{mag}} f(T) ,
\]

(2)

where \( l_{\text{mag}}(T) \) is a general magnetic mean free path based on the approximation \( l_{\text{mag}} \equiv l_k \). For \( k_B T \ll J \) this assumption is justified because the heat carrying excitations exist in significant numbers only in the vicinity of the band minima, i.e., a very small fraction of the Brillouin zone. This requirement is always fulfilled for all systems discussed in this article since \( J \approx 1500-2000 \) K and the experimental data only extend over temperatures \( T < 350 \) K. The function \( f(T) \) depends on temperature in a manner which is characteristic of the considered spin system\([3]\).

In particular, for a gapless \( S = 1/2 \) Heisenberg chain Eq. (1) yields \([26]\)

\[
\kappa_{\text{mag}} = \frac{2n_s k_B^2}{\pi \hbar} l_{\text{mag}} T \int_0^{\frac{\Delta}{k_B T}} x^2 \frac{\exp(x)}{(\exp(x) + 1)^2} dx ,
\]

(3)

where \( n_s \) is a geometrical factor that counts the number of chains per unit area. At low temperatures \( k_B T \ll J \) the integral is only weakly temperature dependent and approaches the constant value \( \pi^2/6 \) for \( T \rightarrow 0 \). Note that the condition \( k_B T \ll J \) holds even at room temperature (up to \( T \lesssim 0.15J/k_B \approx 300 \) K), i.e., the experimental data always represent the low temperature behaviour of \( \kappa_{\text{mag}} \) \([26,23]\) and the upper boundary of the integral may be set to infinity. The corresponding expression for a gapped two-leg ladder is \([15]\)

\[
\kappa_{\text{mag}} = \frac{3n_s k_B^2}{\pi \hbar} l_{\text{mag}} T \int_{\frac{\Delta}{k_B T}}^{\infty} x^2 \frac{\exp(x)}{(\exp(x) + 3)^2} dx ,
\]

(4)

where \( n_s \) is now the number of ladders per unit area. At low temperatures \( k_B T \ll \Delta < J \) one might approximate \( f(T) \propto \exp(-\Delta/(k_B T)) \). Finally, for a 2D-HAF one finds, accounting for both magnon branches \([17]\),

\[
\kappa_{\text{mag}} = \sum_{i=1,2} \frac{n_p k_B^3}{4 \pi \hbar^2 v_0} l_{\text{mag}} T^2 \int_{\frac{\Delta_{i}}{k_B T}}^{\infty} x^2 \sqrt{x^2 - x_0^2} \frac{\exp(x)}{(\exp(x) - 1)^2} dx ,
\]

(5)

with the spin wave velocity \( v_0 \approx 1.287 \cdot 10^5 \) m/s \([67]\) and \( n_p = 2/c \) the number of planes per unit length along the \( c \)-axis, which is the direction perpendicular to the planes (\( c \) is the

\[\text{Footnote: For all three different types of systems the reader is referred to the original literature \([15,17,26,23]\) for the derivation of the respective } f(T). \text{ Within this article, we specify only the final results for } \kappa_{\text{mag}}.\]
corresponding lattice constant). The integral is temperature dependent via its lower boundary \( x_{0,1} = \Delta_i/(k_B T) \), where \( \Delta_1/k_B \approx 26 \text{ K} \) and \( \Delta_2/k_B \approx 58 \text{ K} \) account for the anisotropy gaps which arise in \( \text{La}_2\text{CuO}_4 \) \cite{15}. However, the temperature dependence is weak in the \( T \)-range where the experimental data are discussed and one can approximate \( \kappa_{\text{mag}} \propto T^2 \).

### 3.1 Low-temperature characteristics – thermal occupation

Interestingly, the experimental data shown in Fig. 5 exhibit extended regions at low \( T \) where a reasonable description of the data with Eq. 4 using a temperature independent mean free path \( l_{\text{mag}} \) is possible. The solid lines in Fig. 5 represent fits where \( l_{\text{mag}} \) is a free, temperature independent parameter. A remarkably good description is found for \( \kappa_{\text{mag}} \) of the spin chain \( \text{CaCu}_3\text{O}_3 \), as depicted in Fig. 5a. Here, \( \kappa_{\text{mag}} \) is excellently described by a simple linear increase over the large temperature range \( T \approx 100-300 \text{ K} \) and the fit yields \( l_{\text{mag}} = 22 \pm 5 \text{ Å} \) corresponding to about 5-6 lattice spacings.

For the spin ladder compounds \( (\text{Sr, Ca, La})_{14}\text{Cu}_{24}O_{41} \) the situation is somewhat different. As can be seen in Fig. 5b), the temperature range over which a good fit with Eq. 4 can be achieved is strongly reduced with respect to the previous case and apparently also depends on the composition of the material. In particular, the temperature interval where Eq. 4 describes \( \kappa_{\text{mag}} \) with a \( T \)-independent mean free path is 54-102 K for \( \text{Ca}_{9}\text{La}_{5}\text{Cu}_{24}O_{41} \) but only 61-91 K for \( \text{Sr}_{14}\text{Cu}_{24}O_{41} \). Nevertheless, restricted to these \( T \)-ranges the fit (with the spin gap \( \Delta \) and the mean free path \( l_{\text{mag}} \) as free fit parameters) yields similar results for both compounds. In particular, the value for the spin gap is found to be somewhat larger than but still in reasonable agreement with spin gap results from neutron scattering (\( \Delta/k_B = 418 \pm 15 \text{ K} \) and \( \Delta/k_B = 396 \pm 10 \text{ K} \) for \( \text{Ca}_{9}\text{La}_{5}\text{Cu}_{24}O_{41} \) and \( \text{Sr}_{14}\text{Cu}_{24}O_{41} \), respectively) \cite{10,72,21}. The magnetic mean free path is of similar magnitude in both materials (\( l_{\text{mag}} = 2980 \pm 110 \text{ Å} \) and \( l_{\text{mag}} = 2890 \pm 230 \text{ Å} \) respectively). It is important to note that for temperatures higher than the mentioned ranges, Eq. 4 completely fails to properly describe the experimental data with a \( T \)-independent \( l_{\text{mag}} \), as is also evident from Fig. 5b). However, as will be discussed in Section 3.3, a consistent picture arises if \( l_{\text{mag}} \) is allowed to become \( T \)-dependent at higher \( T \).

A similar observation is also made in the case of the 2D-HAF \( \text{La}_2\text{CuO}_4 \) (cf. Fig. 3c). Eq. 5 with \( l_{\text{mag}} \) as a free parameter yields a reasonable fit in the range 70-158 K, where \( l_{\text{mag}} \approx 558 \pm 140 \text{ Å} \) \cite{17}. Again, the theoretical model fails to account for the high temperature regime as long as \( l_{\text{mag}} \) remains \( T \)-independent.

### 3.2 Low temperature characteristics – scattering off defects

The actual meaning of the magnetic mean free path \( l_{\text{mag}} \) as a material parameter is a priori not clear. On one hand it is known from the analogous case of phonon heat transport, that at low temperature the phonon mean free path can become as large as the crystal dimensions, i.e. of the order of millimeters \cite{72}. However, the extracted values for \( l_{\text{mag}} \) are several orders of magnitude smaller than the dimensions of the crystals which have been studied in the experiments. It therefore appears natural to conclude that \( l_{\text{mag}} \) should reflect the density of static defects in the material. On the other hand, a magnetic mean free path of the order of up to \( \sim 1000 \) lattice spacings implies almost perfect crystallinity of the underlying material, which is astonishing in view of the large and complicated unit cell of, for example, \( \text{Ca}_{9}\text{La}_{5}\text{Cu}_{24}O_{41} \). Note that a much smaller general magnitude of \( l_{\text{mag}} \) has been suggested for this compound based on Exact Diagonalization calculations of the thermal Drude weight which implied ballistic heat transport in spin ladder systems \cite{35}. Such reduced values for \( l_{\text{mag}} \) could not be confirmed by more recent calculations which suggest a vanishing Drude weight \cite{39,16}.

\[3\] The slightly smaller values of \( l_{\text{mag}} \) and \( \Delta \) for \( \text{Ca}_{9}\text{La}_{5}\text{Cu}_{24}O_{41} \) as compared to previous results \cite{15} are a consequence of the usage of more accurate lattice parameters and an optimized fit-interval. It is stressed that these small corrections have no further consequences on the conclusions drawn in Ref. \cite{15}. 

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\(\text{mag} = 2890 \pm 110 \text{ Å} \), \(\text{mag} = 2980 \pm 110 \text{ Å} \), \(\text{mag} = 2890 \pm 230 \text{ Å} \) respectively. It is important to note that for temperatures higher than the mentioned ranges, Eq. 4 completely fails to properly describe the experimental data with a \( T \)-independent \( l_{\text{mag}} \), as is also evident from Fig. 5b). However, as will be discussed in Section 3.3, a consistent picture arises if \( l_{\text{mag}} \) is allowed to become \( T \)-dependent at higher \( T \).
A straightforward experimental method to elucidate the connection between $l_{\text{mag}}$ and the density of magnetic defects in the material is to measure the latter independently from heat transport using a different experimental technique. One possibility is to study the magnetic susceptibility $\chi(T)$ where one has to assume that paramagnetic moments, the concentration of which is deducible from $\chi$, are connected with defects within the magnetic structure and hence may have an effect on the magnetic heat transport. For CaCu$_2$O$_3$ the situation appears to be quite fortunate, since in a recent study Goiran et al. suggested a direct link between paramagnetic moments located off the 1D magnetic chain structures (detectable by $\chi$-measurements) and (possibly non-magnetic) defects within the chains [73]. From susceptibility measurements on CaCu$_2$O$_3$ (cf. Fig. 4) it was then possible to deduce an upper limit for the mean distance between defects within a chain which turned out to be a factor of 2-3 larger than the extracted $l_{\text{mag}}$ [23]. The same order of magnitude of both quantities indicates that these defects are the main scatterers for magnetic excitations within a chain. In the absence of a similar method for La$_2$Cu$_{1-z}$O$_4$ and (Sr, Ca, La)$_{14}$Cu$_{24}$O$_{41}$, an alternative approach was selected to compare $l_{\text{mag}}$ with known distances between intentionally doped defects in the material. Such defects can be induced by substituting a small amount of non-magnetic Zn$^{2+}$ for the magnetic Cu$^{2+}$-ions. This has been performed for both La$_2$Cu$_4$O$_4$ and Sr$_{14}$Cu$_{24}$O$_{41}$ for a series of different doping levels. As can be inferred from Fig. 4(b) and 4(c), once again $l_{\text{mag}}$ has the same order of magnitude as the mean distance between the defects, which is in these cases well defined by the mean distance between the Zn dopants [17][22]. This quantitatively confirms that a good analysis of $\kappa_{\text{mag}}$ in these low dimensional spin systems can be performed using a rather simple kinetic model.

3.3 Scattering processes at higher temperatures

We now turn briefly to the behavior of $\kappa_{\text{mag}}$ at high $T$ in order to elucidate the impact of temperature dependent scattering processes on the magnetic heat transport, i.e. scattering of magnetic excitations off other quasiparticles such as phonons, charge carriers or other magnetic excitations. Here we focus on heat transport in the spin ladder compounds, since these are susceptible to a variety of different types of doping. Most remarkable (apart from doping with non-magnetic impurities which has already been discussed) is certainly the possibility of hole-doping the ladders. Interestingly, the stoichiometric parent compound Sr$_{14}$Cu$_{24}$O$_{41}$ is already inherently doped with holes. These holes reside only partially in the two-leg ladder structures
the largest portion is located in chain substructures which are also present in the material beside the two-leg ladders. The holes are redistributed between the chain and ladder structures upon the isovalent substitution of Ca for Sr: with increasing Ca-content a significant increase of the hole concentration in the ladders is observed [74,75]. It is also possible to reduce the overall hole content in the material and thereby render the ladders virtually hole-free by replacing the divalent Sr or Ca by trivalent ions such as La [74,75].

A good example for the latter case of doping is the compound Ca$_2$La$_2$Cu$_2$O$_{4.5}$ whose magnetic thermal conductivity is shown in Fig. 3 in comparison with that of Sr$_{14}$Cu$_{24}$O$_{41}$ [18]. The effect of hole-doping on $\kappa_{\text{mag}}$ can immediately be observed in this figure. For $T \lesssim 100$ K the increase in $\kappa_{\text{mag}}$ with $T$ is almost identical for both compounds. Pronounced differences only occur at higher $T$: $\kappa_{\text{mag}}$ of La$_2$Ca$_9$Cu$_{24}$O$_{41}$ exhibits a large peak ($\sim 140$ W m$^{-1}$K$^{-1}$ at $\sim 180$ K) and stays very large even at room temperature ($\sim 100$ W m$^{-1}$K$^{-1}$). In contrast, the peak is much smaller in the case of Sr$_{14}$Cu$_{24}$O$_{41}$ ($\sim 75$ W m$^{-1}$K$^{-1}$ at $\sim 150$ K). Here $\kappa_{\text{mag}}$ decreases much more strongly at high $T$ and saturates at $\kappa_{\text{mag}} \approx 10$ W m$^{-1}$K$^{-1}$ for $T \gtrsim 240$ K.

It is straightforward to attribute the strong high-$T$ suppression of $\kappa_{\text{mag}}$ in Sr$_{14}$Cu$_{24}$O$_{41}$ compared with La$_2$Ca$_9$Cu$_{24}$O$_{41}$ to scattering of the magnons off holes, since the hole doping in this compound is the most relevant difference with respect to the undoped ladders of La$_2$Ca$_9$Cu$_{24}$O$_{41}$. Both $\kappa_{\text{mag}}$ curves are almost identical below a characteristic temperature $T_0 \approx 100$ K, confirming that this scattering mechanism becomes completely unimportant below $T_0$ and only reveals its full strength above a characteristic temperature $T^* \approx 240$ K.

The surprising temperature dependence of the strength of the magnon-hole scattering has been checked for robustness against changes of the hole content in the ladders, where $\kappa_{\text{mag}}$ of a series of Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$ single crystals has been investigated in detail [18]. The comparison with $\kappa_{\text{mag}}$ of the Ca-doped samples (see Fig. 5a) reveals that $T_0$ and $T^*$ are gradually shifted towards lower $T$: i.e., the temperature region where $\kappa_{\text{mag}}$ is suppressed extends and magnon-hole-scattering also becomes important at low $T$. At $x = 4, 5$ this region appears to becomes so wide that even the peak at low $T$ is suppressed. It was shown that the temperature dependence of $\kappa_{\text{mag}}$ is unambiguously correlated with a charge ordered state in the compound, where charge ordering sets in at $T \lesssim T^*$. More precisely, the charge ordering in the ladders is accompanied by a drastic enhancement of the magnon mean free path $l_{\text{mag}}$: the probability for magnon-hole scattering, which is close to unity for mobile holes, vanishes in the charge ordered state [18].

Turning to $\kappa_{\text{mag}}$ of La$_2$Ca$_9$Cu$_{24}$O$_{41}$, i.e. that of undoped ladders, it is a priori clear that the magnon hole scattering, which is dominant in Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$, cannot play any significant role. Nevertheless, the necessity to allow a formal $T$-dependence of $l_{\text{mag}}$ at $T > 100$ K indicates that further scattering processes are also relevant for this case. The only remaining possible scattering mechanism for magnons in this material are magnon-magnon or magnon-phonon scattering. In a more involved analysis the $T$-dependence of $l_{\text{mag}}$ in La$_2$Ca$_9$Cu$_{24}$O$_{41}$ has been calculated from the $\kappa_{\text{mag}}$ data using Eq. 4 by employing the previously extracted $\Delta = 418$ K. The resulting $l_{\text{mag}}(T)$ as shown in the main panel of Fig. 3b reflects the different $T$-regimes which govern $\kappa_{\text{mag}}$. For $T \lesssim 110$ K, $l_{\text{mag}}$ is $T$-independent with a mean value $l_0 = 2980$ Å which reflects the scattering of magnons off static defects. In order to describe the $T$-dependent $l_{\text{mag}}$ at higher $T$ it was assumed that all scattering mechanisms were independent of each other and Mattheisen’s rule applied: $l_{\text{mag}}^{-1} = l_0^{-1} + \gamma_{\text{ph}} d_{\text{ph}}^{-1} + \gamma_{\text{mag}} d_{\text{mag}}^{-1}$. Here $d_{\text{ph}}$ and $d_{\text{mag}}$ are the mean ”distances” of phonons and magnons respectively, as calculated from the particle densities with $\gamma_{\text{ph}}$ and $\gamma_{\text{mag}}$ the corresponding scattering probabilities. Since it is unclear as to what extent the separate scattering mechanisms contribute to $l_{\text{mag}}$, its behavior was analyzed based on the assumption that only one mechanism is active in addition to magnon-defect scattering.

The case of dominant magnon-phonon scattering was modelled by three energy-degenerate non-dispersive optical branches along the ladder direction, yielding $1/d_{\text{ph}} = 7.6 \times 10^9 \, \text{m}^{-1}$ with $\Delta_{\text{opt}}$ the optical gap (cf. Ref. [20] for details). The experimental $l_{\text{mag}}$ was then fitted with $l_{\text{mag}}^{-1} = l_0^{-1} + \gamma_{\text{ph}} l_{\text{ph}}$ using $\gamma_{\text{ph}}$ and $\Delta_{\text{opt}}$ as free parameters. The fit (solid line in Fig. 3b) describes the data fairly well. Remarkably, the value found for $\Delta_{\text{opt}} = 795$ K is of the same order of magnitude as the energy of the longitudinal Cu-O stretching mode which is involved in the two-magnon-plus-phonon absorption observed in optical spectroscopy [77,78].
Fig. 5. a) $\kappa_{\text{mag}}(T)$ of Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$ ($x = 0, 2, 3, 4, 5$). Top panel: Data for ($x = 0, 2, 3$) in comparison with $\kappa_{\text{mag}}$ of La$_2$Ca$_9$Cu$_{22}$O$_{41}$. Lower panel: enlarged representation for $x = 3, 4, 5$. From Ref. [18]. b) $l_{\text{mag}}$ of La$_2$Ca$_9$Cu$_{22}$O$_{41}$ as a function of $T$. The solid and broken lines represent fits of $l_{\text{mag}}$ accounting for magnon-phonon and magnon-magnon scattering respectively. From Ref. [20]. c) The thermal conductivity along the chains $\kappa_c$ of Sr$_{1-x}$Ca$_x$CuO$_2$ at $x = 0$ (○) and $x = 0.05$ (●). An estimation of $\kappa_{\text{mag}}$ of the doped material via $\kappa_{\text{mag}} \approx \kappa_{||} - \kappa_{\perp}$ ($\kappa_{\perp}$ is not shown) is indicated. Data from Ref. [21].

For the assumption of dominant magnon-magnon scattering, a less satisfactory agreement was obtained with $l_{\text{mag}}^{-1} = l_0^{-1} + \gamma_{\text{mag}} d_{\text{mag}}^{-1}$, where $1/d_{\text{mag}} = \frac{1}{\pi L} \int_0^\infty \frac{3}{\pi k T} \cdot \frac{3}{\pi \exp(\epsilon_k/2k T)} d\epsilon_k$ (broken line in Fig. 5b). $L$ is the lattice constant along the ladders and $\epsilon_k$ was taken from Johnston et al. for the case of isotropic ladder coupling [79], with $\epsilon_{k=\pi} = \Delta = 418$ K employed. The comparison between both fits suggests that scattering off optical phonons is dominant in this compound.

4 Other developments

Complementary to $\kappa_{\text{mag}}$ of the "dirty" spin chains of CaCu$_2$O$_3$, the spinon heat transport of the very "clean" spin chain materials Sr$_2$CuO$_3$ and SrCuO$_2$ has been investigated by A. V. Sologubenko and coworkers [25][26]. Despite the data giving very clear evidence for spinon heat conductivity in this compounds, the precise extraction of $\kappa_{\text{mag}}$ from the experimental data is quite involved, since the signature of $\kappa_{\text{mag}}$ appears as a shoulder in the high-$T$ edge of the phononic low-$T$ peak (see Fig. 5c) for the case of SrCuO$_2$). In their analysis, Sologubenko et al. suggest that $\kappa_{\text{mag}}$ exhibits a peak-like $T$-dependence which allows for further analysis [25][26]. In order to achieve a better extraction of $\kappa_{\text{mag}}$, P. Ribeiro et al. investigated the thermal conductivity of Sr$_{1-x}$Ca$_x$CuO$_2$ with $x = 0.05$ with the expectation that the Ca-impurities selectively suppress $\kappa_{\text{ph}}$ while $\kappa_{\text{mag}}$ remains unchanged [21]. However, as can be seen in Fig. 5c, the Ca-doping apparently leads to a complete suppression of the shoulder-like anomaly of the phonon peak and no signature of a peak structure remains (cf. Ref. [21] for details). Further studies are necessary to elucidate the origin of this intriguing observation.

5 Conclusion

From this summary of recent developments in the research on magnetic heat transport in quantum spin systems, it becomes evident that even though considerable progress has been made, this new thermal transport mechanism is still far from being thoroughly understood. We have seen that in some cases the magnetic heat conductivity can serve as a sensitive probe for magnetic excitations, which is a promising new approach to access the scattering mechanisms and dissipation of these excitations. However, many issues still remain to be resolved. For example, little is known about how $\kappa_{\text{mag}}$ evolves when the magnetic systems become less quantum in nature, i.e. when $S > 1/2$. Initial experiments and theoretical work have already addressed this topic [33][47][50][51] but the number of investigated materials of this type is still small. A
better understanding of scattering processes (non-magnetic vs. magnetic impurities, phonons), frustration, and the effect of an external magnetic field will also be required.

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