

## Magnon heat transport in $(\text{Sr,Ca,La})_{14}\text{Cu}_{24}\text{O}_{41}$

C. Hess,<sup>1,2</sup> C. Baumann,<sup>1,2</sup> U. Ammerahl,<sup>1</sup> B. Büchner,<sup>2,1</sup> F. Heidrich-Meisner,<sup>3</sup> W. Brenig,<sup>3</sup> and A. Revcolevschi<sup>4</sup>

<sup>1</sup>*II. Physikalisches Institut, Universität zu Köln, 50937 Köln, Germany*

<sup>2</sup>*II. Physikalisches Institut, RWTH Aachen, 52056 Aachen, Germany*

<sup>3</sup>*Institut für Theoretische Physik, Technische Universität Braunschweig, 38106 Braunschweig, Germany*

<sup>4</sup>*Laboratoire de Physico-Chimie, Université Paris-Sud, 91405 Orsay, France*

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We have measured the thermal heat conductivity  $\kappa$  of the compounds  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  and  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  containing doped and undoped spin ladders, respectively. We find a huge anisotropy of both, the size and the temperature dependence of  $\kappa$  that we interpret in terms of a very large heat conductivity due to the magnetic excitations of the one-dimensional spin ladders. This magnon heat conductivity decreases with increasing hole doping of the ladders. Magnon heat transport is analyzed theoretically using a simple kinetic model. From this analysis we determine the spin gap and the temperature-dependent mean free path of the magnons, which measures about several thousand angstrom at low temperature. The relevance of several scattering channels for the magnon transport is discussed.

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Low-dimensional quantum magnets have attracted much attention in recent years among both experimental and theoretical physicists. On the one hand, these compounds serve as model systems for a comparison between experiment and theory, since exact solutions or numerical treatments of the model Hamiltonians yield clear-cut predictions. On the other hand, unusual ground states and magnetic excitations are present, in particular, in quasi-one-dimensional spin systems. A prominent example is the quantum-disordered spin-liquid state with a spin gap that is well established in frustrated and dimerized spin chains and in spin ladders.<sup>1-8</sup>

Usually the magnetic excitations of these spin systems are experimentally studied by measuring spectroscopic or thermodynamic quantities that reveal information on the magnetic ground state and the excitation spectra as a function of energy and momentum. In principle, dispersive magnetic excitations should also contribute to a transport property, i.e., the thermal heat conductivity  $\kappa$ . The experimental investigation of the magnon heat transport could give interesting complementary information on the magnetic excitations, such as dissipation and scattering of magnons, similar as the study of electronic transport properties does in metals.

Recently, several studies of thermal heat conductivity in low-dimensional spin systems have been performed and magnetic contributions to  $\kappa$  are discussed in one-dimensional spin systems such as, e.g.,  $\text{CuGeO}_3$ ,<sup>9-11</sup>  $\text{Sr}_2\text{CuO}_3$ ,<sup>12</sup> and  $\text{SrCuO}_2$  (Ref. 13) as well as in two-dimensional cuprates.<sup>14,15</sup> However, in many compounds the interpretation of the data is controversial, since an unambiguous discrimination of different contributions to  $\kappa$  is difficult or even impossible.<sup>16</sup> Very convincing experimental evidence for a magnon heat transport has been presented by Sologubenko *et al.* for  $(\text{Sr,Ca})_{14}\text{Cu}_{24}\text{O}_{41}$ .<sup>17</sup> The crystal structure of this compound contains two quasi-one-dimensional magnetic subsystems along the  $c$  axis. One subsystem is a sheetlike arrangement of  $\text{Cu}_2\text{O}_3$  two-leg ladders, where the  $\text{Cu } S = \frac{1}{2}$  spins are strongly coupled via a Cu-O-Cu superexchange, while the other subsystem is an array of  $\text{CuO}_2$   $S = \frac{1}{2}$  spin chains with weak magnetic interactions. While even the stoichiometric

compound  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  is hole doped the holes are predominantly located in the chains.<sup>18,19</sup> Changing the composition, i.e., the Sr, Ca, La content in  $(\text{Sr,Ca,La})_{14}\text{Cu}_{24}\text{O}_{41}$ , alters concentration and distribution of the holes. This results in drastic changes of the magnetic properties of the chains, whereas a large spin gap in the ladders of the order of 400 K is observed in all compounds.<sup>20-23</sup> However, the charge transport is determined by the holes in the ladders, which are already present in  $(\text{Sr,Ca})_{14}\text{Cu}_{24}\text{O}_{41}$ . Undoped ladders are only found in systems containing a large amount of trivalent ions, e.g., La.

In this paper we report on the thermal conductivity of  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  and  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  parallel and perpendicular to the chain/ladder direction. In the undoped ladders of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  the magnon contribution is very large and exceeds the phonon contribution by nearly two orders of magnitude. A simple approach to describe the energy transport due to magnetic excitations in the ladders is presented.

We have grown single crystals of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  and  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  by the traveling solvent floating zone method.<sup>24,25</sup> Using a standard steady-state method measurements of  $\kappa$  have been performed on pieces cut along the principal axes with a typical length of 2 mm along the measuring direction and of about 0.5 mm for the two other directions. The thermal gradient has been determined by measuring the temperature difference  $\Delta T$  between the junctions of a differential Au/Fe-Chromel thermocouple. The junctions of this thermocouple have been glued onto the sample using GE varnish.<sup>26</sup>  $\Delta T$  varied between 0.5% and 2% of the absolute temperature that has been stabilized for each data point. The magnetic-field dependence of the thermoelectric power of our Au/Fe-Chromel thermocouple has been carefully determined using a  $\text{SiO}_2$  sample as a nonmagnetic reference.

Figure 1 presents  $\kappa$  of  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  as a function of temperature  $T$  measured along the three crystallographic axes ( $\kappa_a, \kappa_b, \kappa_c$ ). A striking anisotropy of both, absolute value and temperature dependence of the thermal conductivity is apparent. Only for the  $b$  axis we find the qualitative behavior

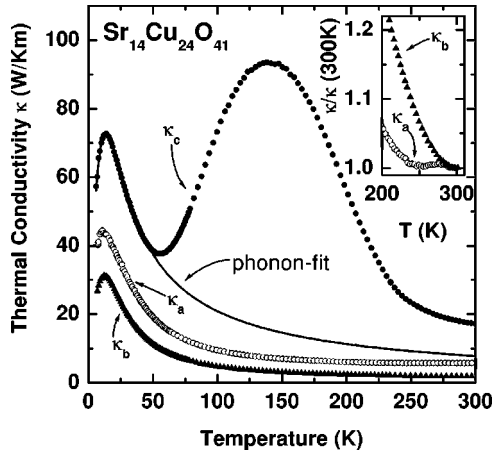


FIG. 1. Anisotropic thermal conductivity  $\kappa_a$ ,  $\kappa_b$ , and  $\kappa_c$  of  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  as a function of temperature. Inset:  $\kappa_a$  and  $\kappa_b$  normalized to the value at 300 K.

expected for phonon heat transport. At low  $T$  the occupation of phonon states implies an increase of the heat conductivity, whereas  $\kappa_b$  decreases at high  $T$  due to increased phonon scattering.  $\kappa_a$  deviates slightly from this usual phonon thermal conductivity of insulators (see inset of Fig. 1): At high temperature we find a slight increase with increasing  $T$ . This small increase of  $\kappa_a$  might be related to the complexity of the phonon spectrum and/or unusual scattering processes possibly related to the anomalous  $T$  dependence of the lattice constants.<sup>27</sup> We note that small deviations from the  $T$  dependence expected for the phonon heat transport as we observe for  $\kappa_a$  in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  are rather common for complex transition-metal oxides. It is still possible and meaningful to discuss this data in the framework of a phonon heat conductivity  $\kappa_{ph}$ .

This is impossible in the case of  $\kappa_c$ , i.e., for the heat transport along the chain/ladder direction in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ . In this case the phonon heat transport can only explain the data below  $\approx 40$  K where  $\kappa_c$  exhibits a low- $T$  maximum very similar to  $\kappa_a$  and  $\kappa_b$ . The absolute value is about 1.6 times larger than for  $\kappa_a$ . Since the velocity of sound and the elastic constants do not show a pronounced anisotropy,<sup>28</sup> one has to attribute the anisotropy of  $\kappa_{ph}$  to anisotropic phonon scattering. Such an anisotropy of the phonon mean free path is also observed in other transition-metal oxides<sup>14,29</sup> with low-dimensional structure elements.

The most striking observation is the behavior of  $\kappa_c$  for temperatures above  $\approx 40$  K. With increasing  $T$ ,  $\kappa_c$  increases strongly and a second very pronounced maximum of the heat conductivity occurs at  $T \approx 140$  K. Further increasing the temperature causes a sharp decrease of  $\kappa_c$  and for temperatures above 250 K the heat conductivity seems to saturate at a still rather large value of about 18 W/Km. Both the very large values of  $\kappa_c$  at intermediate  $T$  and the strange  $T$  dependence with the pronounced second maximum differ drastically from the usual  $\kappa_{ph}$ . We mention that our data for  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  agree qualitatively with those reported by Sologubenko *et al.*<sup>17</sup> There are, however, strong deviations from the data of Kudo *et al.*<sup>30</sup> who report much smaller  $\kappa$  for all lattice directions and  $T$  probably due to enhanced defect

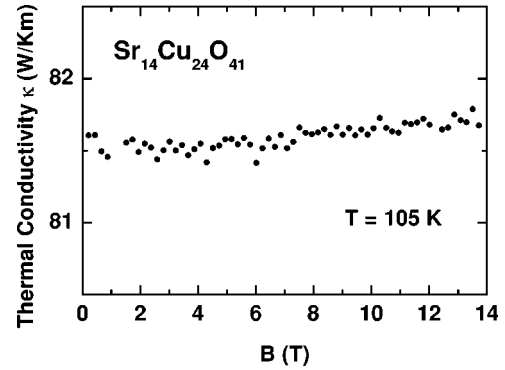


FIG. 2. Field dependence of  $\kappa_c$  at 105 K.

scattering in their samples. Comparing our data for  $\kappa_c$  quantitatively with those in Ref. 17 reveals a perfect agreement for the low- $T$  maximum while the second peak is about 30% larger in our sample. At present we are systematically studying different crystals in order to understand the origin of these quantitative deviations.

As discussed in Ref. 17 it is reasonable to attribute the high-temperature maximum of  $\kappa_c$  to magnetic excitations propagating along the spin ladders. The electronic heat transport as estimated from resistivity data and the Wiedemann-Franz law is negligible. The  $T$  dependence, i.e., the anisotropy of  $\kappa$  and the absolute value of  $\kappa_c$  cannot be explained by a phonon heat transport and the data for  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  allow also to exclude exotic electronic contributions as discussed below.

In order to clarify the origin of the anomalous contribution to  $\kappa_c$  we have investigated  $\kappa_c$  as a function of a magnetic field. We find only very small field dependencies. Representative data measured at  $T = 105$  K are displayed in Fig. 2. Up to our maximum field of  $B = 14$  T we find a slight increase of  $\kappa_c$  of about 0.3%. We stress that this does not contradict a magnetic origin of the anomalous  $\kappa_c$ . Theoretical estimates yield a small increase of about 1% as long as the spin gap is much larger than the Zeeman energy, as is the case for the spin ladders and the maximum applied field of  $B = 14$  T.<sup>31</sup>

A determination and analysis of this very unusual magnon contribution requires a separation of a phonon background. Unfortunately, it is impossible to determine unambiguously the phonon background for  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ . For example, at high  $T$  the absolute values and  $T$  dependence are unpredictable, which is clearly demonstrated by the qualitatively different behavior of  $\kappa_a$  and  $\kappa_b$ . Similar problems do exist at low  $T$ , since  $\kappa$  is determined by strongly anisotropic phonon scattering that cannot be determined from independent experimental data or from theory. This means that any determination of the additional magnetic contribution in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  leads to a large error for low  $T$  and high  $T$ , i.e., for temperatures where the total  $\kappa$  is not much larger than  $\kappa_{ph}$ . In order to estimate the phonon background we fit the data at low temperatures  $T \lesssim 40$  K with a Debye model and extrapolate the behavior up to 300 K. The result of this description of  $\kappa_{ph}$  is indicated by a solid line in Fig. 1, which is much smaller than the measured  $\kappa$  in the entire temperature range above 50 K.

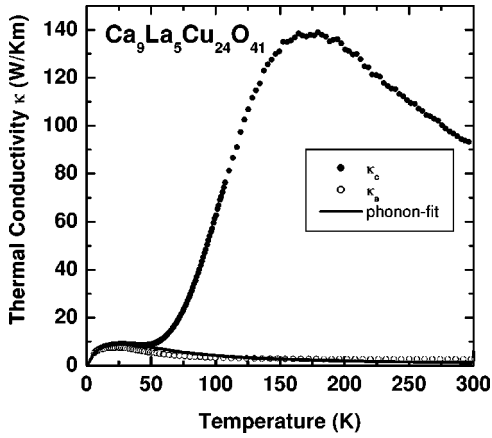


FIG. 3. Thermal conductivities of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  as a function of temperature measured along the  $a$  and  $c$  axes,  $\kappa_a$  and  $\kappa_c$ , respectively. The solid line represents an estimate of the phonon contribution to  $\kappa_c$ .

However, considering only the data for  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  one can question any nonphononic heat conductivity at  $T \gtrsim 250$  K. A much more reliable separation of phonon and magnon heat transport is possible in the case of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  whose thermal conductivities along the  $a$  and  $c$  axes are shown in Fig. 3. At low  $T$  the thermal heat conductivity of this compound that contains undoped spin ladders and only slightly doped spin chains<sup>18,19</sup> is much smaller than in stoichiometric  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  for both directions. It is straightforward to attribute this isotropic suppression of  $\kappa_{ph}$  at low  $T$  to structural defects. On the one hand, a lattice site is occupied by randomly distributed  $\text{Ca}^{2+}$  and  $\text{La}^{3+}$  ions with strongly different size, which is known to suppress  $\kappa_{ph}$ . On the other hand x-ray diffraction studies show structurally disordered chains in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$ . Surprisingly,  $\kappa_c$  strongly increases above 40 K in spite of the structural disorder. At intermediate and high  $T$   $\kappa_c$  is even larger than in stoichiometric  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  and the room-temperature value is comparable to that found in metals. In contrast to that a very small heat conductivity is found along the  $a$  axis in the entire temperature range. Reduced size and  $T$  dependence are typical for  $\kappa_{ph}$  of a compound with many structural defects. It is apparent from Fig. 3 that this strong damping of  $\kappa_{ph}$ , which is inferred from  $\kappa_a$  as well as from the low- $T$  behavior of  $\kappa_c$ , enables a reliable determination of the additional contributions to  $\kappa_c$  above  $\approx 40$  K.

Before we turn to a quantitative analysis of  $\kappa_{mag}$  we mention several conclusions that can be drawn from the qualitative behavior of  $\kappa$  in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  and, in particular, from the comparison to the findings in the stoichiometric compound. The suppression of  $\kappa_{ph}$  that coincides with an enhancement of  $\kappa_c$  at high  $T$  gives very strong evidence that there are two independent contributions to the heat transport. Moreover, some thinkable origins of the large additional contributions to  $\kappa_c$  in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  can be ruled out from the additional data. Excitations of the chains are irrelevant, since the magnetic and electronic properties of the heavily doped chains in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  showing charge order, spin gap, and dimerization are completely different from the long range

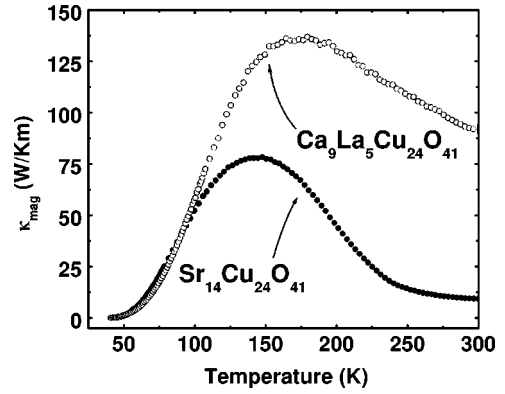


FIG. 4. Magnon thermal conductivities of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  and  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  as a function of temperature.

ordered chains in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$ .<sup>32,33</sup> For example, heat transport due to collective electronic excitations as suggested for the thermal conductivity of the charge-density wave compound  $\text{K}_{0.3}\text{MoO}_3$  (Ref. 34) could be relevant in the strongly doped charge ordering  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ , whereas it is certainly irrelevant in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  due to the strongly reduced hole content.

Similarly, the magnetic excitations in the chains cannot explain the anomalous  $\kappa_c$ . In  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  the magnetic excitations in the chains are well described by localized triplets on nearly noninteracting dimers.<sup>27,35</sup> These excitations do not contribute significantly to the thermal conductivity. The magnetic coupling of the Cu spins in the nearly undoped chains of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  is weakly ferromagnetic and below  $T \approx 10$  K long range magnetic order is observed.<sup>32,33</sup> The corresponding nearly gapless excitation spectrum might lead to magnon contributions to the thermal conductivity at low  $T$ . However, our data, which are nearly isotropic at low  $T$ , do not show any evidence for magnetic contributions at low  $T$ . In any case, the chain excitations are completely irrelevant for our discussion of the anomalous thermal conductivity at higher  $T$ .

Summarizing these arguments we have to state that the findings in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  strongly support the interpretation of the additional contribution to  $\kappa_c$  in terms of a magnon heat transport along the spin ladders. This magnon heat conductivity  $\kappa_{mag}$  is apparently not suppressed by structural disorder and, moreover, a smaller hole doping seems to enhance  $\kappa_{mag}$  at high  $T$ .

In Fig. 4 we show  $\kappa_{mag}$  for  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  and  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$ , which are derived by subtracting the Debye fits of the phonon contribution from the measured  $\kappa_c$ . For  $T \lesssim 100$  K absolute value and temperature dependencies of  $\kappa_{mag}$  are similar in the two compounds. However, pronounced differences occur at higher  $T$ . The magnetic heat conductivity on  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$   $\kappa_{mag}$  strongly decreases above  $\approx 150$  K. This decrease is much less pronounced and is found at higher temperature (above  $\approx 200$  K) in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  where we find a very large  $\kappa_{mag} \approx 100$  W/Km even at room temperature.

In order to analyze the magnon heat transport theoretically, we start from the simple kinetic expression

$$\kappa_{mag} = \frac{d}{dT} \sum_k v_k \epsilon_k n_k l_k, \quad (1)$$

where  $v_k$ ,  $\epsilon_k$ , and  $l_k$  denote velocity, energy, and mean free path of the magnetic excitations. Assuming a momentum-independent mean free path, i.e.,  $l_k \equiv l_{mag}$  and using the distribution function

$$n_k = \frac{3}{3 + e^{\epsilon_k/k_B T}} \quad (2)$$

for the triplet excitations yields the following expression for the magnon heat conductivity:

$$\kappa_{mag} = \frac{3Nl_{mag}}{\pi\hbar k_B T^2} \int_{\Delta_{ladder}}^{\epsilon_{max}} \frac{\exp(\epsilon/k_B T)}{[\exp(\epsilon/k_B T) + 3]^2} \epsilon^2 d\epsilon. \quad (3)$$

Here,  $N$  is number of ladders per unit area and  $\epsilon_{max}$  is the band maximum of the spin excitations, which is at approximately 200 meV in  $(\text{Sr,Ca,La})_{14}\text{Cu}_{24}\text{O}_{41}$ . Note, that the distribution function (2) is different from a Bose distribution. This difference is used to account, on average, for the hardcore constraint of no on-site double occupancy for the triplet excitations. While such a form of the distributions function would describe the occupation of a *local* triplet excitation exactly, it is only meant as an approximate phenomenology regarding the momentum space distribution — which is unknown for the present case. The primary motivation for this type of a triplet distribution is to suppress unphysically large triplet densities at higher temperatures. From Eq. (3) it is apparent that a particular form of the magnon dispersion does not enter the heat conductivity of a one-dimensional triplet gas within a kinetic description — an effect that has not been noticed in other studies.<sup>17</sup> In particular, the momentum or energy dependence of the magnon velocity  $v$ , which emerges from Eq. (1) plays no role in our approach. Note that the assumption of a  $k$ -independent mean free path implies a  $k$ -dependent scattering rate since  $v$  depends on the wave number. While this assumption seems reasonable for defect scattering more involved scenarios will apply, in particular, to inelastic scattering. Within our treatment the magnon heat conductivity for experimentally relevant temperatures  $T \ll \epsilon_{max}/k_B$  mainly depends on two parameters: the spin gap  $\Delta_{ladder}$  and the mean free path  $l_{mag}$ . We mention, that Eq. (3) differs from an expression used by Sologubenko *et al.*<sup>17</sup> for the heat conductivity of one-dimensional bosons not only by the distribution function (2) but also by an overall factor of 3 accounting for the triplet degeneracy.

For temperatures below 300 K,  $\kappa_{mag}$  does not depend significantly on  $\epsilon_{max}$  and in the following analysis of the data in the framework of Eq. (3) we use  $\epsilon_{max} = 200$  meV according to the literature.<sup>22,36</sup> It is impossible, to determine reasonable values of the two remaining parameters  $l_{mag}$  and  $\Delta_{ladder}$  by fitting the data with the expression in Eq. (3) without further assumptions. Therefore, we ignore possible temperature dependencies of the spin gap and, moreover, assume a constant, temperature-independent mean free path at low  $T$ , i.e., for a small number of thermally activated phonons and

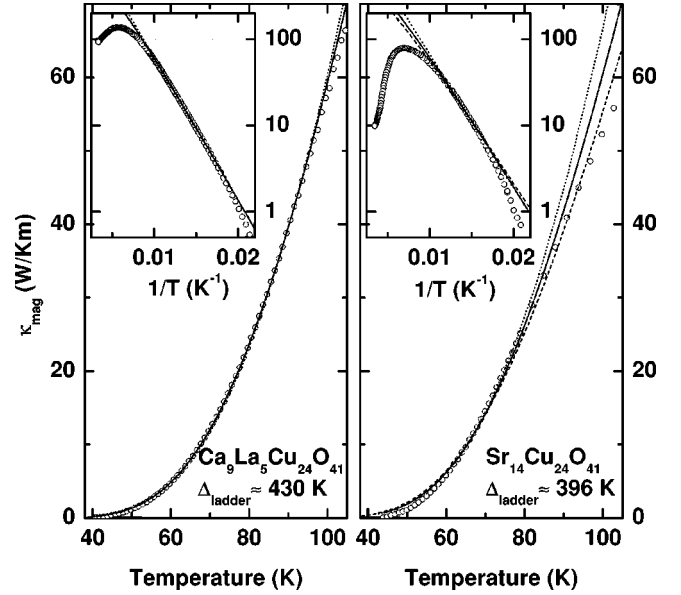


FIG. 5. Temperature dependence of the magnon thermal conductivities of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  (left) and  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  (right) at low temperatures in comparison with fit results to the data [dotted lines, simple activation; solid lines, one-dimensional (1D) model for  $\kappa$ ; broken lines (only  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ ), 1D model with fixed spin gap]. Insets: Arrhenius representation of  $\kappa$  and fit results.

magnons. Knowing the spin gap from this first step of the analysis it is possible to extract the temperature-dependent  $l_{mag}$  directly from the data.

Our assumption of a constant  $l_{mag}$  at low  $T$  implies that the heat conductivity is determined by the temperature-dependent activation of magnons in this temperature range. This is in agreement with the experimental data that roughly follow a simple activated behavior as displayed in Fig. 5. Note that Eq. (3) predicts deviations from the simple activation law and taking into account these corrections, slightly improves the description of the data as demonstrated in Fig. 5. The spin gaps of  $\Delta_{ladder} = 396$  K and  $\Delta_{ladder} = 430$  K we obtain are in fair agreement with the results from neutron scattering. We conclude that both, the temperature dependence of  $\kappa_{mag}$  and the absolute values of the spin gaps extracted from the data confirm our analysis. Comparing the left and the right columns of Fig. 5 reveals that the agreement between data and theory is much better for  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  than for  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ . In  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  the separation of  $\kappa_{mag}$  and  $\kappa_{ph}$  is reliable and the thermal conductivity for  $T \lesssim 100$  K is well described by our model with constant  $l_{mag}$ . In  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  a fair agreement with the theory at constant  $l_{mag}$  can only be obtained in a limited temperature range  $55 \text{ K} \lesssim T \lesssim 85 \text{ K}$ . Since the ambiguity in choosing a reasonable phonon background leads to appreciable uncertainties in the determined spin gap, we have also analyzed the data by fixing the spin gap to the value known from neutron scattering ( $\Delta_{ladder} = 377$  K).<sup>22</sup> In this case  $l_{mag}$  is the only fit parameter. The result is also shown in the right column of Fig. 5 (broken lines). Again we obtain a reasonable agreement between experiment and theory. At temperatures above 80 K, the agreement is even better than

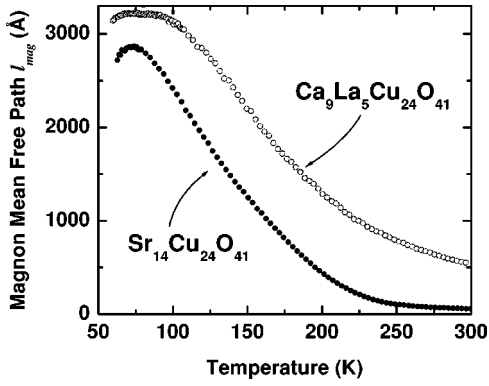


FIG. 6. Magnon mean free paths of  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  (open symbols) and  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  (full symbols) as a function of temperature.

obtained with our two-parameter fit. However, the deviations are slightly larger at low  $T$  where our assumption of a constant  $l_{\text{mag}}$  is more reasonable.

We conclude that the thermal conductivity does not allow one to determine the spin gap with extremely high precision due to the uncertain phonon background. However, it is apparent that our theoretical model and the assumption of a constant  $l_{\text{mag}}$  yield a good description of the data for reasonable values of the spin gap as long as one considers low  $T$ . The deviations occurring at higher  $T$ , i.e., for  $T \geq 100$  K, in both compounds signal a significant temperature dependence of the mean free path.

Based on the preceding analysis of the low-temperature behavior of  $\kappa_{\text{mag}}$  we determined the temperature dependence of the mean free path  $l_{\text{mag}}$  (see Fig. 6). At low temperatures we find a very large  $l_{\text{mag}}$  of several thousand angstrom. Within the experimental uncertainty due to the phonon background we cannot resolve differences of the low- $T$  saturation values of  $l_{\text{mag}}$  for the two compounds containing spin ladders with different hole content. There are, however, unambiguous and drastic differences at higher temperatures. In  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ , which contains spin ladders with a rather large hole doping, the magnon mean free path is much smaller and we find a rather strong temperature dependence between 85 K and 220 K. According to our analysis  $l_{\text{mag}}$  saturates for higher temperatures and at  $T = 300$  K the mean free path is about 60 Å. While the saturation of  $l_{\text{mag}}$  above 220 K is clearly extractable from our data, the absolute values of  $\kappa_{\text{mag}}$  and  $l_{\text{mag}}$  strongly depend on the choice of the background, i.e., the extrapolation of  $\kappa_{\text{ph}}$ , and, therefore, a much smaller  $l_{\text{mag}}$  at 300 K is also compatible with the raw data in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ . For the second crystal with nearly undoped spin ladders a very large  $l_{\text{mag}}$  at room temperature is out of question. Our analysis yields a magnetic mean free path larger than 500 Å at 300 K. Moreover, the overall temperature-dependent decrease of  $l_{\text{mag}}$  is less pronounced and deviations from the constant low- $T$  value start at higher temperatures than in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ . Further, the magnon mean free path in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$  is also distinctly larger than in other one-dimensional spin systems like the spin-chain compounds  $\text{SrCuO}_2$  and  $\text{Sr}_2\text{CuO}_3$ .<sup>12,13</sup>

Our results for  $l_{\text{mag}}$  in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  are in qualitative agreement with the findings reported by Sologubenko *et al.*<sup>17</sup>

There are, however, quantitative discrepancies due to the different theoretical description: for a given  $\kappa_{\text{mag}}$  our analysis leads to a smaller value of  $l_{\text{mag}}$ , mainly due to the more realistic distribution function for the magnetic excitations that we use in our approach [Eq. (2)]. Yet, the mean free paths that we extract from the analysis of our data are very large. In Ref. 17 it was speculated that holes are the main scatterers in the doped spin ladders of  $(\text{Sr,Ca})_{14}\text{Cu}_{24}\text{O}_{41}$ . This speculation is only partially supported by our results on a crystal with undoped spin ladders. At low  $T$ , our data do not show a significant difference between the two crystals indicating that the very large  $l_{\text{mag}}$  at low temperature does not depend strongly on the hole content of the ladders. It is likely that the maximum magnon mean free path is determined by defects. Note, however, that one has to discriminate between crystal defects and “magnetic defects,” since there is no correlation between the maximum  $l_{\text{mag}}$  and the maximum phonon mean free path. The latter is much smaller in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$ , which is obvious from the damping of  $\kappa_{\text{ph}}$  at low  $T$ .

In contrast to the results at low  $T$ , we do find a pronounced doping dependence of the magnon scattering at high  $T$ . The magnon mean free path is much smaller for the doped spin ladders of  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  and of the order of the mean hole distance in the ladders ( $\approx 25$  Å) at 300 K.<sup>19</sup> In the same crystal resistivity shows a pronounced change of its temperature dependence at  $T \approx 220$  K. This anomaly, which is frequently interpreted in terms of charge ordering, is absent in  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$ . Thus, one might argue that instead of the hole content, the hole mobility is crucial for the scattering of magnons, in order to explain the weak doping dependence of  $l_{\text{mag}}$  at low  $T$  and the strong differences at high  $T$ . Yet, holes and their mobility are not the only source of temperature-dependent magnon scattering as demonstrated by our data for the crystal with undoped spin ladders. In this crystal we find a less pronounced, but qualitatively similar decrease of  $l_{\text{mag}}$  for  $T > 100$  K. At room temperature the number of both, thermally excited magnons and phonons is already rather large. For example, the inverse density of magnons at 300 K as calculated for  $\Delta_{\text{ladder}} = 430$  K can be interpreted as the “mean distance” of magnons and is already one order of magnitude smaller than  $l_{\text{mag}}$ . Further experimental and theoretical studies are necessary in order to determine the importance of magnon-defect, magnon-phonon and magnon-magnon scattering.

In summary we have measured the thermal conductivity of hole doped and undoped spin ladders realized in the compounds  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  and  $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_{41}$ . In both cases we find a huge contribution to  $\kappa$  due to magnon heat transport. We have applied a simple kinetic approach to describe this contribution and have obtained the spin gaps of the ladders and the temperature dependence of the mean free path from our analysis. Obviously, at low  $T$ , scattering of magnons on holes is rather ineffective since in this temperature region  $\kappa_{\text{mag}}$  of doped ladders is almost identical to  $\kappa_{\text{mag}}$  of the undoped ladders. For both we find a very large mean free path of magnons of several thousand angstrom. At high  $T$ , increased mobility of holes causes a strong damping of magnon heat transport in the hole-doped ladders due to increased

magnon-hole scattering, whereas  $\kappa_{mag}$  in the undoped ladders only decreases slightly. This is reflected in the mean free path that in the former case at 300 K reduces to  $l_{mag} \approx 60$  Å that is in the same order of magnitude as the mean-hole distance ( $\approx 25$  Å). For undoped ladders  $l_{mag} \approx 500$  Å at 300 K. This large value is one order of magni-

tude larger than the inverse density of magnons at this temperature and might indicate only weak magnon-magnon scattering effects.

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