Low-temperature heat transport of Nd_2CuO_4: Roles of Nd magnons and spin-structure transitions

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We report the magnetic-field dependence of thermal conductivity (κ) of an insulating cuprate Nd_2CuO_4 at very low temperatures down to 0.3 K. It is found that, apart from the paramagnetic moments scattering on phonons, the Nd^{3+} magnons can act as either heat carriers or phonon scatterers, which strongly depends on the long-range antiferromagnetic transition and the field-induced transitions of spin structure. In particular, the Nd^{3+} magnons can effectively transport heat in the spin-flopped state of the Nd^{3+} sublattice. However, both the magnon transport and the magnetic scattering are quenched at very high fields. The spin reorientations under the in-plane field can be conjectured from the detailed field dependence of κ.

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I. INTRODUCTION

The insulating parent compounds of cuprate superconductors are known to have an antiferromagnetic (AF) order of Cu^{2+} spins. Because of the quasi-two-dimensionality of the Cu^{2+} spin structure, these materials can show rather strong magnon heat transport at relatively high temperatures.[1–6] However, the magnon transport seems to be negligible at low temperatures, probably due to the anisotropy gap of the spin spectrum. The external magnetic field can strongly suppress or close the gap at some critical fields, usually accompanied with the spin-flop transitions. It was therefore expected to observe the magnon heat transport in a magnetic field.[6] In the last several years, a few works on this topic have been performed on parent compounds of electron-doped cuprate superconductors $Pr_{1.3}La_{0.7}CuO_4$ (PLCO) and Nd_2CuO_4 (NCO).[5–7] The low-T heat transport of PLCO seemed to be quite simple and indicated that some paramagnetic moments such as "free" Cu^{2+} spins can scatter phonons and lead to non-negligible magnetic-field dependence of phononic thermal conductivity.[5] It is notable that the PLCO data did not indicate the possibility of magnon heat transport, although the spin-flop transition can happen for not strong in-plane fields. On the other hand, the NCO results are more complicated because of the complexity of magnetism caused by the Nd^{3+} ions.[6,7]

It is known that, in NCO, the Cu^{2+} spins order antiferromagnetically below $T_N = 245 \sim 255$ K with a noncollinear magnetic structure,[8–14] that is, the Cu spins in the same CuO_2 plane are antiferromagnetically ordered and those in the adjacent planes along the c axis are perpendicular to each other. The moment of Nd^{3+} ion is enhanced drastically with lowering temperature, and a long-range AF order develops below ∼1.5 K with the same noncollinear spin structure as Cu^{2+} spins.[11,14,15] When the magnetic field is applied in the CuO_2 plane, the Cu^{2+} spins can reorientate and enter a spin-flopped state.[10–12,16–20] The transition fields are reported to be 4.5 and 0.75 T for $H \parallel a$ and $H \parallel [110]$, respectively.[10,19] In addition, the strong coupling between Nd^{3+} spins and Cu^{2+} spins, which was found to be about 4 T,[13] drives Nd^{3+} spins to rotate together with Cu^{2+} spins, keeping their relative orientation unchanged, and forms a three-layer unit consisting of one Cu^{2+} layer sandwiched between two Nd^{3+} layers as a whole. For this reason, the Nd^{3+} spins are generally considered to change together with Cu^{2+} sublattice under the influence of magnetic field. However, at low temperatures (<1.5 K), when the AF order of Nd^{3+} spins is formed, the magnetic structures and field-induced transitions of the Nd^{3+} sublattice are still unclear since all the earlier neutron scattering in the applied field were carried out at relatively high temperatures.

Two earlier studies on the low-T thermal conductivity of NCO have revealed strong magnetic-field dependence of κ, which was believed to be associated with the spin flop of Cu^{2+} spins.[6,7] An additional conduction channel of Nd^{3+} magnons was supposed to be the reason for the enhancement of κ in high fields, considering the reorientation of Nd^{3+} spins under the Nd^{3+}-Cu^{2+} interaction. However, there are some quite unclear issues calling for more careful investigations. First, it is apparently questionable in Ref. 6 to attribute the large increase of κ above some transition fields to the Nd^{3+} magnons at temperatures above 2 K, where the long-range ordering of Nd^{3+} spins has been lost. In particular, the field-induced enhancement of κ is even much larger at 5 K than that at 2 K.[6] In that work, more direct evidence for Nd^{3+} magnon transport, which should be detected below the Néel transition of Nd^{3+} spins, had not been investigated. Another work performing measurements down to millikelvin temperatures, however, did not report the magnetic-field-dependent data,[7] which is indispensable for showing the relationship between heat transport and the transition of magnetic structure. Second, although the low-energy magnons can be significantly excited at the spin-flop transition and may contribute to carrying heat, because of the closure of the gap in magnon spectra, further increasing the magnetic field will always shift the magnon dispersion upward and finally suppress the low-energy magnon excitations.[21] So, it is odd to assume that the magnon heat transport induced by the spin-flop transition can be still active in the high-field limit.[6,7] Third, it was assumed that the Nd^{3+} spin lattice should rotate together with the Cu^{2+} sublattice in the in-plane field. However, it is notable that the Nd^{3+}-Nd^{3+} interaction can be enhanced significantly at very low temperatures due to the increased magnitude of the Nd^{3+} moment, which means that the Nd^{3+} sublattice could reorientate independently. In this paper, we study the temperature and magnetic-field dependences of κ of high-quality NCO single crystals in great detail to investigate the role of magnons in the low-T heat transport. Our results on $\kappa(H)$ isotherms demonstrate that the phonon conductivity is strongly dependent on the magnetic field, and the Nd^{3+} magnon can play a dual role in the transport as the heat carriers or the phonon scatterers. To be exact, the Nd^{3+} magnons mainly scatter phonons at the critical region ($1 \sim 2$ K) of Néel transition in zero field, while they can significantly transport heat in the spin-flopped state induced by the in-plane field. Intriguingly, the drastic behaviors of $\kappa(H)$ under the in-plane field reveal the spin reorientation transitions of the Nd^{3+} sublattice at sub-Kelvin temperatures.

II. EXPERIMENTS

High-quality Nd_2CuO_4 single crystals are grown by using the slow cooling method with CuO_2 as a self-flux. The platelike samples for transport measurements are confirmed to grow along the *ab* plane by the x-ray back-reflection Laue photographs. The dimensions along the *c* axis of NCO crystals are so small that the measurement with heat current along the *c* axis is difficult to carry out. The thermal conductivity is therefore measured only along the *a* axis by using a conventional steady-state technique and two different processes: (i) using a "one heater, two thermometers" technique in a ^{3}He refrigerator and a 14-T magnet at a temperature region of 0.3–8 K; (ii) using a Chromel-Constantan thermocouple in a pulse-tube refrigerator for the zero-field data above 4 K.[22–24]

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of thermal conductivity of a NCO single crystal in zero field, together with data from some other groups.[6,7] The large peak at 20 K is apparently the so-called phonon peak in insulators.[25] It is known that the magnitude of the phonon peak is dominated by the phonon scattering by the crystal defects and impurities and is therefore a good characterization for the crystal quality.[25] It is clear that the phonon peak of our crystal is almost the same as the data obtained on a NCO crystal grown using the floating-zone method,[6] suggesting the high quality of our NCO crystal. Although the high-quality NCO had not been measured down to very low temperatures in the earlier work,[6] our crystal can alternatively demonstrate the intrinsic low-T heat transport of this compound. Below 10 K, the thermal conductivity decreases quickly with lowering temperature, like usual insulators, but there is an obvious variation in slope of the $\kappa(T)$ curve around 1.5 K, which is apparently related to the AF ordering of Nd^{3+} ions.[11,14,15] However, only from the zero-field data, it is not clear whether the slope change is caused by an enhanced magnon scattering on phonons at the critical region of AF phase transition or an appearance of the magnon heat transport below the transition. The magnetic-field dependence of κ is known to be useful to clarify this issue. Before analyzing the $\kappa(H)$ data, we can make an estimation of the phonon mean-free path (l) at very low temperatures, assuming that the magnons are not acting as heat carriers.

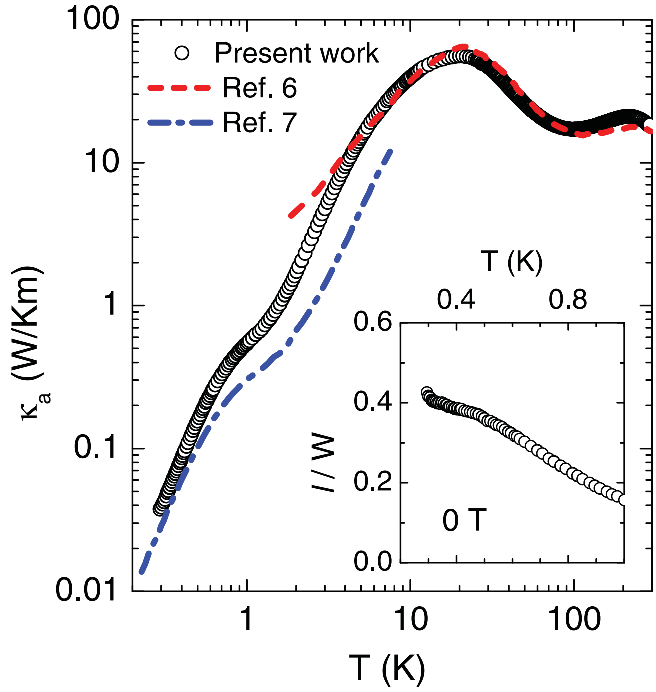

FIG. 1. (Color online) The *a*-axis thermal conductivity of Nd_2CuO_4 single crystal in the zero field, compared with the results reported by Jin *et al.* (Ref. 6) and Li *et al.* (Ref. 7). Inset: The ratio of the phonon mean-free path l to the averaged sample width W in the zero field.

Following a standard calculation,[26] l can be obtained from the kinetic formula $\kappa = \frac{1}{3}C\bar{v}l$, where $C = \beta T^3$ is the phonon specific heat and $\bar{v}$ is the averaged sound velocity, and both β and $\bar{v}$ are known experimentally for NCO.[27,28] The obtained l is compared with the averaged sample width W (=0.473 mm), which is taken to be $2/\sqrt{\pi}$ times the geometrical mean width $\bar{w}$,[26] and shown in the inset to Fig. 1. It can be seen that the ratio l/W is only about 0.4 at 0.3 K, which means that the microscopic scattering on phonons is still effective at this temperature region.[25,26] Note that, if the magnons can contribute to carrying heat at sub-Kelvin temperatures, the phonon conductivity must be smaller than the experimental data and the phonon mean-free path is even smaller than the calculated value.

The low-T thermal conductivity of NCO is further studied in the magnetic fields applied along the *a* axis, the [110] direction, and the *c* axis. For a low magnetic field along the *a* axis, a distinct steplike transition shows up in the $\kappa(T)$ curves below 1.5 K and shifts to lower temperature with increasing field, as indicated by the 1.5- and 2.5-T data in Fig. 2(a). This transition is not observable down to 0.3 K when the magnetic field is larger than 3 T. It is likely a field-induced AF-ferromagnetic (spin-polarized) transition of Nd^{3+} ions. In the strongest field (14 T) we applied, the low-T conductivities are increased and a nearly T^3 dependence is presented, which explicitly indicates that there is strong magnon-phonon scattering in the zero field and the scattering is almost smeared out in the high field. This can also be verified from the inset to Fig. 2(a) that the phonon mean-free path in the 14-T field, which is much larger than that in the zero field, increases with

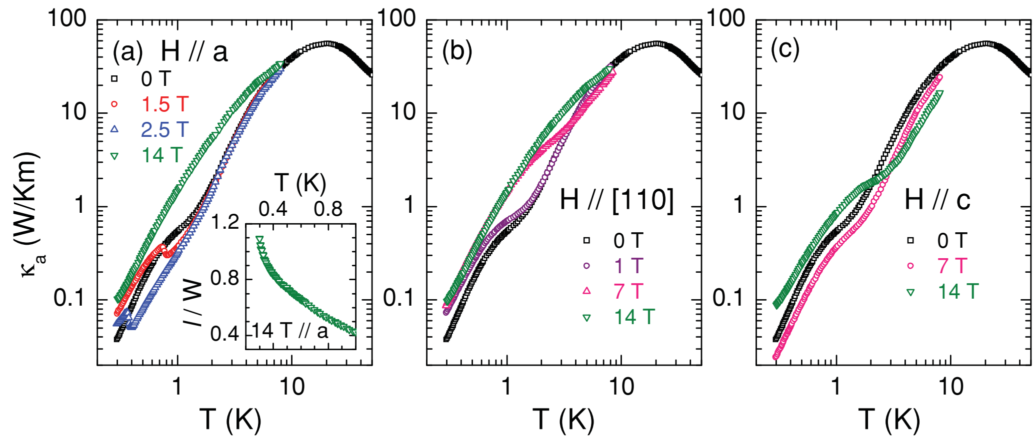

FIG. 2. (Color online) Temperature dependences of thermal conductivity in Nd_2CuO_4 with magnetic field applied along the (a) a axis, (b) [110] direction, and (c) c axis. Note that the $\kappa(T)$ curves show steplike transitions at 0.8 and 0.4 K for 1.5 and 2.5 T fields along the a axis, respectively. Inset to (a): The ratio of the phonon mean-free path l to the averaged sample width W in 14 T $\parallel$ a.

lowering temperature and approaches the averaged sample width at 0.3 K, demonstrating that the boundary scattering limit of phonons is nearly established in the 14-T field and at such low temperatures.[24,26] For the magnetic field applied along the [110] direction, a similar result to the a-axis-field case is that the 14-T field can also significantly enhance the low-T thermal conductivity; in particular, the thermal conductivity at the lowest temperature is almost the same as that in 14 T $\parallel$ a. However, there seems to be no AF-ferromagnetic transition of Nd^{3+} ions for $H \parallel [110]$, as shown in Fig. 2(b). When the c-axis field is applied, the thermal conductivities display the most complex dependence on temperature, as shown in Fig. 2(c).

The detailed dependences of thermal conductivity on the magnetic field are shown in Fig. 3. One can see from Figs. 3(e) and 3(f) that, as the c-axis field increases, κ is decreased first and then increased at high-enough field, which results in a broad "valley" in the low-T $\kappa(H)$ isotherms. Since both the Cu^{2+} spin structure and the Nd^{3+} spin structure are not changeable for $H \parallel c$,[29] this observation can only be attributed to the paramagnetic moments. In this regard, the shifting of the position of the $\kappa(H)$ minimum to higher field with increasing temperature is indeed compatible with the typical behaviors of the paramagnetic moments scattering on phonons.[5,30] In NCO, the "free" spins at very low temperatures can be either the spin vacancies or defects on the long-range-ordered Cu^{2+} spin lattice or those on the ordered Nd^{3+} spin lattice. It is known from the earlier studies on PLCO and $GdBaCo_2O_{5+x}$ that whether the high-field-limit conductivities are larger than the zero-field values is determined by the condition whether the magnetic ions have the zero-field energy splitting.[5,30] Since the low-T κ in the high-field limit is apparently larger than those in the zero field, it is likely that the "free" spins on the Nd^{3+} sites, the ground-state doublet of which can be split in the zero field,[11] rather than the Cu^{2+} free spins[5] are responsible for scattering phonons. In addition, it is clear that the decrease of κ in the 14-T field above 5 K, in contrast to the increase below 2 K, is simply because the field is not strong enough to remove the paramagnetic scattering.[30] Note that the paramagnetic moments scattering on phonons seems to be much more significant in NCO than that in PLCO.[5]

The phonon scattering by paramagnetic moments is known to be qualitatively isotropic for different field directions,[30] so the similar $\kappa(H)$ behaviors to those for $H \parallel c$ should also contribute to the $\kappa(H)$ data for $H \parallel ab$. In fact, at

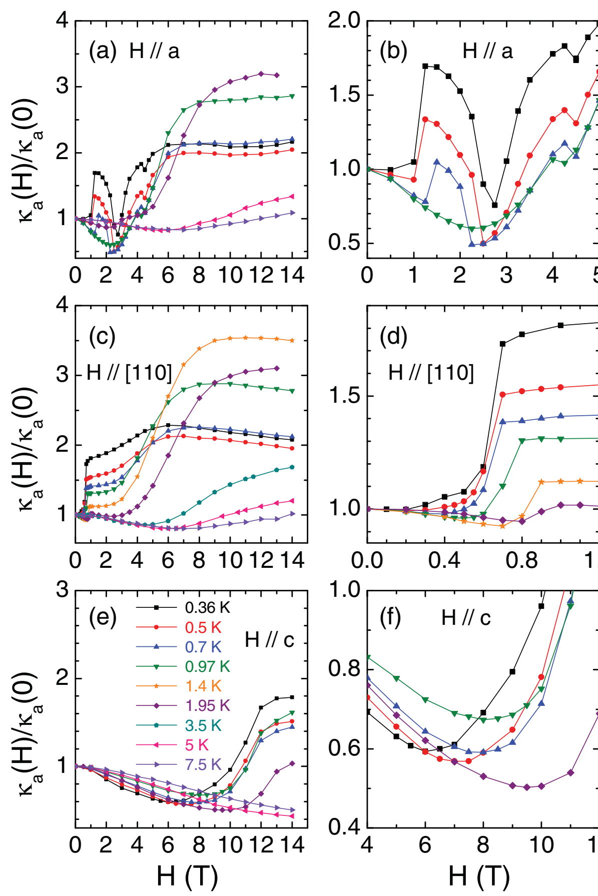

FIG. 3. (Color online) Magnetic-field dependencies of thermal conductivity of Nd_2CuO_4 single crystal with the field applied along the (a) a axis, (c) [110] direction, and (e) c axis. Panels (b), (d), and (f) zoom in the low-field plots of panels (a), (c), and (e), respectively.

relatively high temperatures above ∼2 K, the profiles of $\kappa(H)$ for $H \parallel a$ or [110] could still be understood in the picture of paramagnetic scattering, whereas at lower temperatures ($T < 1$ K), the $\kappa(H)$ isotherms display very different behaviors, which manifests that some other mechanisms are taking place. For $H \parallel [110]$, κ shows a steplike increase at ∼0.6 T and a weak field dependence above this transition field, as shown in Figs. 3(c) and 3(d). The situation for $H \parallel a$ is remarkably more complicated. As can be seen from Fig. 3(a), at sub-Kelvin temperatures, except for a sudden increase at ∼1 T, there is a sharp drop at ∼2.5 T. One possible mechanism for these complicated $\kappa(H)$ behaviors is naturally related to the transitions of spin structures induced by the in-plane field. It is known that the a-axis field and the [110] field cause the spin-flop transitions of the Cu^{2+} sublattice at ∼4.5 and 0.75 T, respectively.[10,19] Apparently, these reorientations have weak effects on the heat transport since there is only a very small dip in $\kappa(H)$ curves at 4.5 T for $H \parallel a$, as shown in Fig. 3(a), and the effect for $H \parallel [110]$ is not distinguishable from the strong increase of κ at 0.6 T. Therefore, the low-field anomalies of $\kappa(H)$ for $H \parallel a$ or [110] are most likely caused by the transitions of the Nd^{3+} sublattice, which has never been explored in experiments at sub-Kelvin temperatures.

The above $\kappa(H)$ behaviors can suggest the evolution of Nd^{3+} spin structure (at sub-Kelvin temperatures) for the in-plane magnetic fields, which are summarized in Fig. 4. In principle, whether the Nd sublattice rotates together with the Cu^{2+} spins depends on the competition between Nd^{3+}-Cu^{2+} and Nd^{3+}-Nd^{3+} interactions. As already known, NCO exhibits a noncollinear structure in the zero field, as shown in Fig. 4(a). With increasing field along the a axis, the Nd^{3+} ions are possible to make spin rotations if the field is large enough. As shown in Fig. 4(b), when $H > H_{r1}$(∼1 T), those Nd^{3+} spins pointing along the field direction could rotate by 90° in the ab plane, in other words, a spin-flop transition of Nd^{3+} ions happens. It is known that the low-energy magnons are usually well populated at the spin-flop transition due to the close of the magnon gap,[21] and meanwhile κ shows a sudden increase at the transition field. It is therefore natural to conclude that the Nd^{3+} magnons act as heat carriers at very low temperatures and hence make a positive contribution to the thermal conductivity. If the magnetic field increases further, the Nd^{3+} spins gradually turn to the direction of applied field and are finally polarized at $H = H_{r2}$ (∼2.5 T), as illustrated in Fig. 4(c), which corresponds to a dip in $\kappa(H)$.[21,23,24] After that, the thermal conductivity starts to recover due to the weakening of magnon scattering on phonons. At ∼4.5 T, the Cu^{2+} spin flop happens and the spin directions are switched to be perpendicular to the field. This field strength can destroy the intraunit Nd^{3+}-Cu^{2+} interaction, therefore, Nd^{3+} spins can no longer rotate together with Cu^{2+} spins and keep the alignment along the a axis, as shown in Fig. 4(d). Note that, in the high-field limit, the roles of magnons as either heat carriers or phonon scatterers are inactive and the plateau of $\kappa(H)$ also indicates that the paramagnetic scattering on phonons is smeared out, as the 14-T $\kappa(T)$ data indicate.

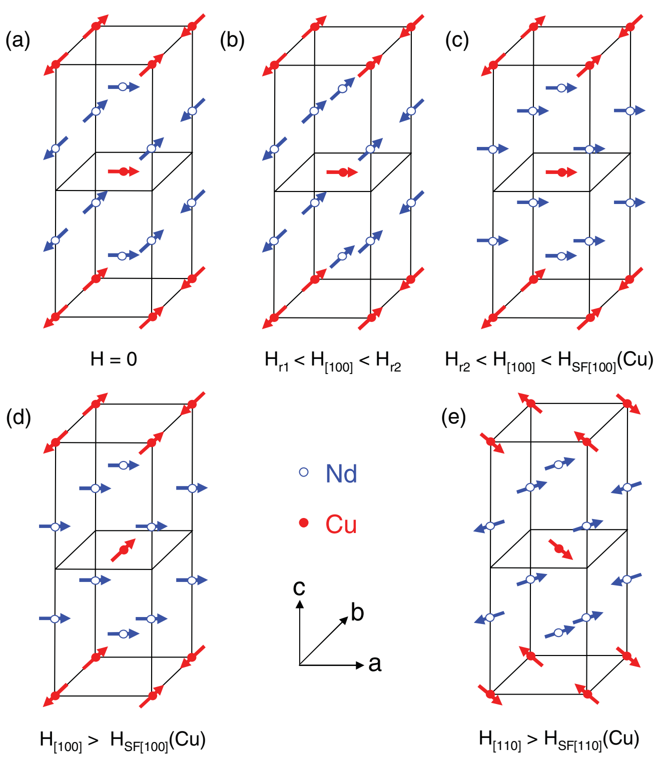

FIG. 4. (Color online) Very-low-temperature magnetic structures of Nd_2CuO_4 in the magnetic fields along two different in-plane directions. (a) Spin structure in the zero field. (b)–(d) Spin structures in the magnetic field along the a axis. H_{r1} and H_{r2} are the critical fields of the spin-flop and the spin-polarization transitions of the Nd^{3+} spins, respectively. (e) Spin structure in the magnetic field along the [110] direction. $H_{SF[100]}$(Cu) and $H_{SF[110]}$(Cu) are the spin-flop transition fields of the Cu^{2+} spins for $H \parallel a$ and [110], respectively.

In the case of $H \parallel [110]$, there is also a sharp increase of κ at ∼0.6 T and the magnitude of increase is almost the same as the increase at 1 T $\parallel a$. This strongly suggests a magnon heat-transport contribution appearing at this critical field, which is essentially the same as that for the Cu^{2+} spin reorientation (∼0.75 T in some former reports).[10,19] Since this field strength is much smaller than the Nd^{3+}-Cu^{2+} interaction, it is expected that the Nd^{3+} spins rotate together with the Cu^{2+} spins and thereby align along the field direction, as shown in Fig. 4(e). Similarly, the low-energy Nd^{3+} magnons are well populated at this spin-flop transition and are able to significantly transport heat. It is notable that there is no diplike feature in higher fields, which indicates that the Nd^{3+} spin polarization does not happen. This is supported by the $\kappa(T)$ in Fig. 2(b), where there is no AF-ferromagnetic transition behavior similar to those in Fig. 2(a).

It is useful to make a quantitative estimation on the Nd^{3+} magnon heat transport from the above data. For example, the steplike increases of κ at 0.36 K is about 0.7 times the zero-field value, which gives a magnon thermal conductivity of 0.0441 W/Km, for both $H \parallel a$ and $H \parallel [110]$. By taking a theoretical prediction of the velocity of Nd^{3+} magnons, ∼10 meVÅ(no experimental observation so far),[11] and assuming the ballistic transport of magnons at such low temperature,[7] we can obtain the magnon mean-free path of about 0.16 mm. This value is quite reasonable since it is in the same order of magnitude of the averaged sample width (0.473 mm).

With the above understandings on both the paramagnetic scattering and the magnon heat transport associated with spin-structure transitions, one can get a complete picture of the $\kappa(H)$

isotherms for the in-plane fields. At sub-Kelvin temperatures, the sharp increase and the "dip" at low fields along the a axis are mainly related to the magnon behaviors, while the high-field enhancement and the plateau feature are due to the disappearance of paramagnetic scattering. On the other hand, the step increase at low fields along [110] mainly results from the magnon heat transport, and the gradual increase of κ at higher field up to ∼6 T is likely due to the weakening of paramagnetic scattering; furthermore, the slow decrease of κ at a field above 6 T is related to the suppression of low-energy magnon excitations in very high fields.

In passing, it is worth pointing out that, at 1 ∼ 2 K, the thermal conductivity is most strongly recovered at very high in-plane field. It seems to be related to the temperature dependence of magnon excitations (Nd^{3+}); that is, at $T < 1$ K, the magnon population is negligibly small in zero field because of the magnon gap, while at 1 ∼ 2 K, the critical region of Nd^{3+} AF transition, the spin fluctuations are significant and scatter phonons rather strongly. So, in this temperature region, the zero-field phonon transport is most strongly damped by not only the paramagnetic moments but also the magnetic excitations and it can be remarkably recovered by applying high in-plane field.

It is intriguing to compare this work with the earlier ones. It has been concluded from the very-low-T thermal conductivity in the zero field and strong in-plane field, which is much higher than the Cu^{2+} spin-flop transition, that the high-field-induced enhancement of κ is a direct contribution from Nd^{3+} magnon heat transport.[7] The present data essentially support the capability of Nd^{3+} magnon transporting heat, but only in relatively low fields. Based on the detailed field dependence of κ, particularly for $H \parallel c$, it is able to be clarified that the enhancement of κ at very high fields is mainly due to the weakening of paramagnetic scattering on phonons. The reason that the earlier work did not notice the importance of paramagnetic scattering is that the $\kappa(T)$ data for $H \parallel c$ were taken only at 10 T for the comparison with the in-plane-field data.[7] It is a coincidence that, in this field, the thermal conductivities in the whole temperature range are not likely to be larger than the zero-field values, as shown in Fig. 3(e). Therefore, the present data provide a very important supplement to the earlier works and lead to a more accurate conclusion.

IV. CONCLUSIONS

In summary, we study the heat transport of a parent insulator of high-T_c superconductors Nd_2CuO_4 at low temperatures down to 0.3 K and in magnetic fields up to 14 T. It is found that, in zero field, the low-T thermal conductivity is purely phononic with rather strong scatterings from paramagnetic moments and Nd^{3+} magnetic excitations. In high magnetic field along either the c axis or the ab plane, the low-T thermal conductivity can be significantly enhanced because of the weakening of magnetic scattering. An interesting finding is the drastic changes of κ at low fields along the a axis or the [110] direction, which demonstrates the field-induced spin flop or spin polarization of Nd^{3+} spin lattice. At sub-Kelvin temperatures, the Nd^{3+} magnons can act as heat carriers in the spin-flopped state, however, their transport can exist only in some intermediate field regime and is suppressed by an succeeding spin-polarization transition for $H \parallel a$. In the field along the [110] direction, the magnon transport can be active in a broader field region but is also weakened in very high field. An evolution of the magnetic structure with the in-plane field can be suggested based on these field dependences of κ. One may note that, although the magnons can hardly affect the phonon heat transport for those superconducting samples, because of the disappearance of the long-range AF order, the paramagnetic moments can still effectively scatter phonons.

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