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Large magnetothermal effect and spin-phonon coupling in a parent insulating cuprate $Pr_{1.3}La_{0.7}CuO_4$

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The magnetic-field (H) dependence of the thermal conductivity κ of $Pr_{1.3}La_{0.7}CuO_4$ is found to show a pronounced minimum for in-plane fields at low temperature, which is best attributed to the scattering of phonons by free spins that are seen by a Schottky-type specific heat and a Curie-Weiss susceptibility. Besides pointing to a strong spin-phonon coupling in cuprates, the present result demonstrates that the H dependence of the phonon heat transport should not be naively neglected when discussing the $\kappa(H)$ behavior of cuprates, since the Schottky anomaly is ubiquitously found in cuprates at any doping.

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I. INTRODUCTION

To elucidate the nature of the superconducting ground state, the quasiparticle (QP) transport properties are widely studied in high-T_c cuprates at low temperature (T), and the discussions are often based on the magnetic-field (H) dependence of the thermal conductivity κ.[1–7] The most striking findings include a "plateau" in the $\kappa(H)$ isotherm at low temperature,[1–3] an increase in the QP population due to the supercurrents around vortices,[4] and the magnetic-field-induced QP localization in underdoped cuprates.[5–7] A common assumption in these previous works is that the phononic thermal conductivity κ_p is *independent* of H and all the H dependence of κ is of electronic origin. This assumption may seem reasonable, because the vortex scattering of phonons (which is important in conventional superconductors) somehow becomes insignificant at low temperature in cuprates.[8] However, phonons could interact also with magnetic excitations and be scattered, which would introduce some H dependence in κ_p. Therefore, it is important to elucidate how the low-T phonon heat transport is affected by magnetic excitations in cuprates, particularly in the superconducting doping regime. Remember, even though antiferromagnetic (AF) magnon excitations are likely to be irrelevant in the superconducting regime, Schottky anomalies in the specific heat, which are usually attributed to excitations of localized free spins, are commonly found in superconducting cuprates.[9–15]

To gain insight into the possible H dependence of κ_p, it is useful to study parent insulating cuprates, where it is expected that the electronic contribution is absent and κ_p can be unambiguously studied; however, it has been discussed that in AF insulating materials the magnon excitations can also act as heat carriers or scatterers,[16–19] which brings complications to the analysis of the $\kappa(H)$ data of insulating samples. In particular, Jin *et al.*[20] recently reported a large magnetic-field-induced enhancement of κ in Nd_2CuO_4 (NCO), a parent insulating cuprate in the tetragonal T' phase, and they proposed that the increase in κ gives evidence for the magnon heat transport becoming active due to the closing of a magnon gap in the magnetic field. Note that in NCO the magnons for the Cu spins should not contribute to carrying heat because of a large (~5 meV) gap, and thus it must be the Nd magnons that are relevant;[21] the existence of the Nd magnon contribution to the heat transport has been backed up by a more recent study by Li *et al.*[21] Considering these findings, to unambiguously investigate the H dependence of κ_p, we turn to the $Pr_{1.3}La_{0.7}CuO_4$ (PLCO) system where there is essentially no magnetic contribution from the rare-earth ions. Although it was found that a small magnetic moment is induced in Pr^{3+} ions (whose ground state is singlet and would be effectively nonmagnetic),[22,23] in $Pr_{1.3}La_{0.7}CuO_4$ as much as 35% of the Pr^{3+} sites are diluted by nonmagnetic La^{3+} ions and thus the Pr magnons, even if exist, should be strongly damped and give negligible contribution to the heat transport. Also, the single crystals of PLCO were found to show very clean phonon heat transport (e.g., κ_p of PLCO is up to five times larger at 20 K compared to La_2CuO_4)[24] so that exotic scattering mechanisms of phonons are expected to be rather easily seen in PLCO without being masked by scatterings by disorder.

In this paper, we report detailed thermal conductivity, specific heat, and magnetic susceptibility measurements of high-quality PLCO single crystals. At low temperature, the $\kappa(H)$ curve shows a striking dip feature, which is different from previous data on similar T'-phase compounds, Pr_2CuO_4 (PCO) and NCO.[20,25] Our result on $\kappa(H)$ indicates that excitations of free spins, evidenced by the specific heat and the magnetic susceptibility data, are responsible for the peculiar H dependence of the phonon heat transport. Since the Schottky-type specific heat that is indicative of the existence of free spins is found almost ubiquitously in high-T_c cuprates,[9–15] the present result suggests that the phonon heat transport can be H dependent at low temperature in a wide range of cuprate samples and one should be careful upon discussing the QP properties based on $\kappa(H)$ behavior. Physically, the present result points to a strong spin-phonon coupling in cuprates, which might bear intriguing implications on the mechanism of high-T_c superconductivity.

II. EXPERIMENTS

High-quality $Pr_{1.3}La_{0.7}CuO_4$ single crystals are grown by the traveling-solvent floating-zone technique in flowing oxygen.[24] The partial substitution of La for Pr not only stabilizes the crystal growth[24,26] but also disturbs the possible Pr magnons and makes them irrelevant as heat carriers, which is

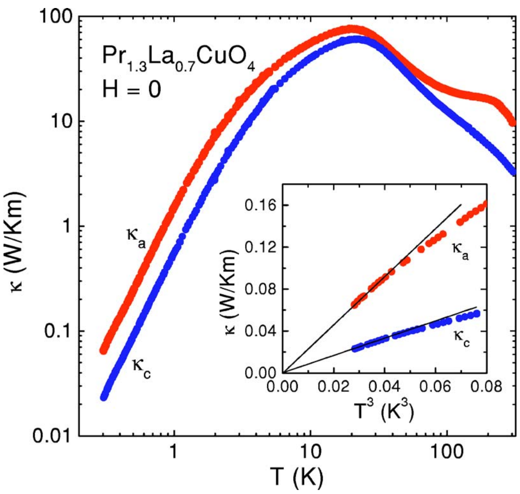

FIG. 1. (Color online) Temperature dependences of κ_a and κ_c of PLCO single crystals in zero field. Inset: κ vs T^3 plot of the low-T data below 0.43 K. Thin solid lines show the T^3 dependence expected in the boundary-scattering regime.

useful for the purpose of this work. For the heat transport measurements, the crystals are cut into rectangular platelets with a typical size of $2.5\times0.5\times0.1$ mm^3 (where the edges are made to be parallel to the crystallographic axes within 1°) and annealed in flowing Ar to remove excess oxygen.[24]

The a-axis and c-axis thermal conductivities (κ_a and κ_c) are measured by a steady-state technique in a ^{3}He refrigerator.[5] Two RuO_2 chip sensors are used for the "top" and "bottom" thermometers on the sample. For the magnetic-field dependent $\kappa(H)$ measurements, the base temperature of heat sink is precisely controlled by an ac resistance bridge (Linear Research LR-700) within 0.01% accuracy with another RuO_2 thermometer or a Cernox thermometer (for above 10 K), and the magnetic-field dependence of the thermometers were carefully calibrated beforehand using a $SrTiO_3$ capacitance sensor and a high-resolution capacitance bridge (Andeen Hagerling 2500A), which allow one to keep the temperature unchanged with a very high precision (±0.1 mK at 0.3 K and ±1 mK above 1 K) during the magnetic field sweeps. The relative uncertainty of the $\kappa(H)$ measurement was confirmed to be typically less than 1%. Specific heat and magnetic susceptibility measurements are carried out using a physical properties measurement system and a superconducting quantum interference device (SQUID) magnetometer (Quantum Design), respectively.

III. RESULTS AND DISCUSSIONS

A. Thermal conductivity

Figure 1 shows the temperature dependences of κ_a and κ_c of PLCO in zero field down to 0.30 K, where the data above 4 K are a reproduction from Ref. 24. As already discussed, one can assume that both κ_a and κ_c show purely phononic thermal conductivity in this insulating compound, and the large magnitude of the phonon peak evidences a high quality of our PLCO crystals.[24] The inset of Fig. 1 shows that the

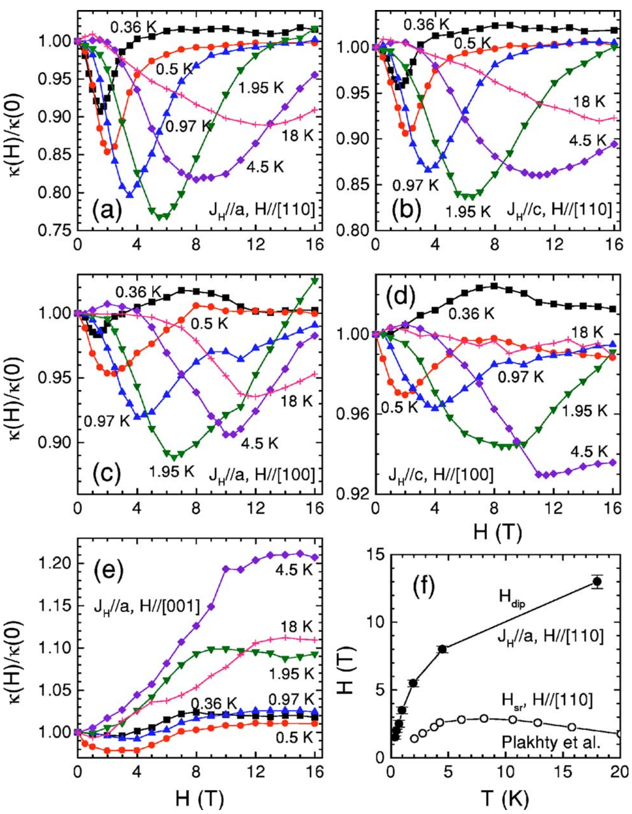

FIG. 2. (Color online) (a)–(e) Magnetic field dependences of κ in PLCO single crystals for various configurations of the heat current J_H and the magnetic field. (f) Comparison of H_{dip} [where the $\kappa(H)$ curve shows a minimum] for $J_H \| a$, $H\|[110]$ to the spin-reorientation field H_{sr} for the same field direction (Ref. 29).

boundary-scattering regime of phonons (where $\kappa_p \sim T^3$) is achieved below ~0.35 K.

Figure 2 shows our main result, the peculiar magnetic-field dependences of κ at temperatures from 0.36 to 18 K, for several different configurations of heat current and magnetic field. Notably, a pronounced dip in the $\kappa(H)/\kappa(0)$ curves is observed for in-plane fields at low tempratures. At first sight, this may seem a typical $\kappa(H)$ behavior in a spin-flop system where magnons act as phonon scatterers:[27] The zero-field κ is purely phononic, because there are no low-energy magnons due to the anisotropy gap in the magnon dispersion; when the magnetic field is applied, the Zeeman energy causes the magnon excitations to become gapless at the spin-flop transition field H_{sf}, but the gap opens again at higher field; consequently, the magnon scattering of phonons is the strongest at H_{sf} where the magnons are the most populated, and causes a dip in the $\kappa(H)$ curve.[27]

In the T'-phase compounds of NCO, PCO, and PLCO, the magnetic structures are essentially identical and spin-flop (or spin-reorientation) transition of the long-range-ordered Cu spins has been studied by the neutron scattering:[22,23,28–33] The Cu spins form a noncollinear structure with the spin easy axes of [100] and [010] in zero field; depending on the magnetic-field direction, a first-order transition (for $H\|[100]$) or a continuous spin-reorientation transition (for $H\|[110]$) is induced, and in both cases the high-field state has a collinear spin structure. If one tries to interpret our $\kappa(H)$ data in terms of this well-studied spin reorientation, several problems become apparent: Most notably, the characteristic field for the spin reorientation, H_{sr}, is known to decrease upon heating, while the "dip" field, H_{dip}, where the $\kappa(H)$ curve shows a minimum, increases upon heating. We show quantitative comparison between H_{sr} and H_{dip} for $H \| [110]$ in Fig. 2(f), where it becomes clear that H_{dip} is not likely to be related to the spin reorientation. Also, H_{dip} is observed to be very close for $H \| [100]$ and $H \| [110]$, while H_{sr} is much larger for $H \| [100]$ than for $H \| [110]$.[23,30,33] Furthermore, the H dependence of κ_c is very similar to that of κ_a, which is difficult to understand in the spin-flop scenario because the c-axis phonons can hardly couple with the magnons of the two-dimensional spin system. Therefore, the peculiar $\kappa(H)$ behavior in PLCO is clearly *not* due to the magnon scattering of phonons associated with the spin reorientation.

It is known from previous neutron experiments[22,23] that the exchange field of the ordered Cu spins induces a small ($\sim 0.1\ \mu_B$) ordered moment on Pr^{3+} ions. One may expect that the Pr magnons should contribute to the $\kappa(H)$ behavior. Since the weak Pr moments are induced by the interaction between Cu and Pr ions, the Pr magnons, if exist and play a role (as either heat carriers or scatterers) in determining the $\kappa(H)$ behavior, should affect $\kappa(H)$ in a way that corresponds to the spin reorientation transition. Therefore, the lack of correspondence between H_{dip} and H_{sr} means that not only Cu magnons but also Pr magnons are irrelevant to the observed $\kappa(H)$ behavior, which is likely due to the considerable dilution of the Pr sites by nonmagnetic La^{3+} ions.

B. Specific heat

To investigate the actual scatterers of phonons and the origin of the peculiar $\kappa(H)$ behavior, the specific heat of PLCO is measured at low temperature using a large single crystal ($\sim$34 mg, Ar annealed) from the same batch. As shown in Fig. 3, there is a pronounced H-dependent enhancement, whose temperature and field dependences are fully consistent with the two-level Schottky anomaly due to free $s=1/2$ moments[9,15]

$$C_{Schottky}(T,H) = n\left(\frac{g\mu_B H}{k_B T}\right)^2 \frac{e^{g\mu_B H/k_B T}}{(1+e^{g\mu_B H/k_B T})^2}, \qquad (1)$$

where g is the Landé factor. The concentration of free spins is n/R, where R is the universal gas constant. In our data, the peak position of the Schottky anomaly, T_{peak}, increases linearly with H [see Fig. 3(d)], which precisely follows Eq. (1) and allows us to determine the g factor. Moreover, fitting of the data to Eq. (1) [see Fig. 3(c)] gives the concentration of the free spins to be $\sim$1%, which is too large to be associated with impurities and suggests the existence of some intrinsic mechanism to produce free moments.

C. Magnetic susceptibility

Another evidence for the existence of free moments in PLCO is the magnetic susceptibility (χ) data. Figure 4 shows the temperature dependences of χ for $H \| a$ and $H \| c$ (χ_a and

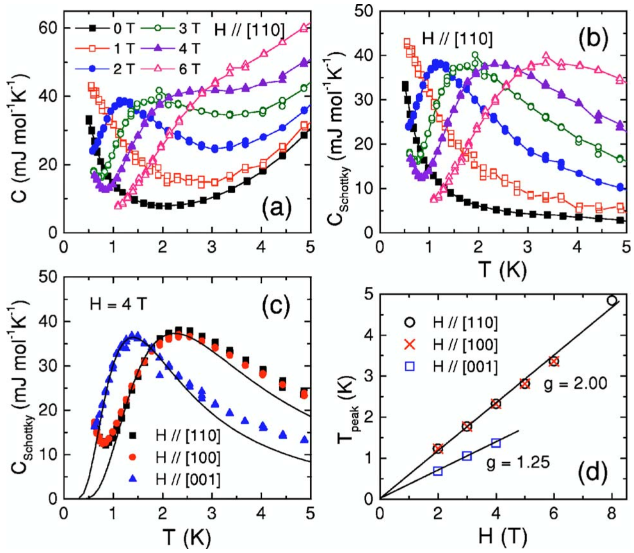

FIG. 3. (Color online) (a) Temperature dependences of the specific heat of a PLCO single crystal in various magnetic fields along [110]. (b) Schottky anomaly for $H \| [110]$, obtained by subtracting the phonon contribution from the raw data in panel (a). (c) Schottky anomaly at 4 T for different orientations of H, together with the theoretical curves expected for the Schottky anomaly from 1% of $s=1/2$ moments with the anisotropic Landé factor $g=2$ (for [100] and [110]) and $g=1.25$ (for [001]). (d) Magnetic field dependences of the peak temperature in the Schottky anomaly for the three directions.

χ_c) in 0.5 T field. Our data are essentially consistent with those for PCO single crystals,[34] including the low-T upturn that is observed in both χ_a and χ_c. As theoretically discussed by Aharony's group,[35] the magnetic susceptibility of R_2CuO_4 (R=Nd, Pr, or Sm) is governed by the magnetic

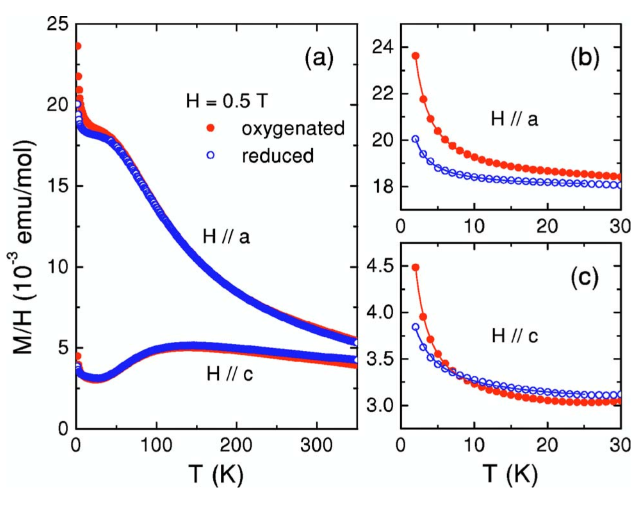

FIG. 4. (Color online) (a) Temperature dependences of susceptibility χ_a and χ_c of a PLCO crystal in oxygenated (as-grown) and reduced (Ar annealed) states. Panels (b) and (c) magnify the low-T part; solid lines show fittings with the formula $\chi(T) = \chi_0 + C/(T-\theta)$, where the first and second terms are T-independent background and additional Curie-Weiss contribution, respectively; the C parameter for the reduced sample is consistent with $\sim$1% of $s=1/2$ free spins with $g=2$ ($g=1.25$) for $H \| a$ ($\| c$).

states of rare-earth ions. The ground multiplet of Pr^{3+} ion is $^3H_4$ in the tetragonal crystalline electric field (CEF), so that Pr^{3+} ions have a singlet ground state with a well separated ($\sim$18 meV) first-excited state.[35] Therefore, the low-T susceptibility is predicted to be T independent,[35] and the observed low-T upturn, which is well described by the Curie-Weiss law, must come from some free moments not considered in the mean-field theory. Note that the sensitivity of the low-T upturn in χ to the oxygen treatment, shown in Fig. 4, suggests that the free moments do not originate from impurity ions, but are produced by some mechanism inherent to PLCO. Candidates for such a mechanism include an appearance of Pr^{4+} ions in some particular crystal environment or some "free" Cu^{2+} spins possibly produced in the AF domain boundaries.[36,37] Incidentally, the analyses of the Schottky anomaly and the Curie-Weiss contribution are consistent with the same anisotropic g factor, which gives confidence that the two phenomena are of the same origin.

D. Phonon scattering by paramagnetic moments

Whatever the origin of the free moments, they are clearly correlated with the H dependence of κ for in-plane fields. Although there has been no report in the cuprate context, it has been known that phonons can be scattered by free paramagnetic moments that form a two-level system,[38] where the energy levels of the two states split in magnetic fields by ΔE due to the Zeeman energy. In this situation, phonons are typically scattered in the following way: A phonon with energy ΔE excites the lower-level state of a magnetic moment to the higher-level state and is absorbed by a direct process; then, another phonon with the same energy (but with a different wave vector) is subsequently emitted by the higher-level state as it relaxes. Since the phonon spectrum has a (broad) maximum at $\sim 4k_BT$, this "paramagnetic scattering" of phonons is the strongest when the Zeeman energy ΔE is equal to $4k_BT$. Naturally, this causes a dip at H_{dip} in the $\kappa(H)$ isotherm with H_{dip} roughly proportional to T,[38] which is actually the case in our data below 2 K [see Fig. 2(f)]. Note that the free spins themselves do not carry heat because there is no dispersive collective excitations due to the negligibly small interactions between these diluted spins.

In passing, we note that the peculiar $\kappa(H)$ behavior in NCO (Ref. 20) may also be understood in terms of the paramagnetic scattering of phonons without employing the magnon heat transport, at least for $T > 1.5$ K: It has been discussed that the doublet ground state of the Nd^{3+} ions in NCO is split slightly ($\sim$3 K) in zero field due to the interaction with Cu spins,[35] which causes the Nd^{3+} ions to form a two-level system; hence, the phonons may well be scattered by paramagnetic Nd^{3+} ions at $T > 1.5$ K, since the Nd ions show Néel order only below $\sim$1.5 K,[35] and a sufficiently large H can remove the paramagnetic scatterings and enhances κ significantly.[38] Thus, although the magnons are likely to be carrying heat below $\sim$1.5 K,[21] their role at higher temperature had better be scrutinized.

We should point out that we do not currently have a good understanding for the enhancement of κ with $H \| c$ shown in Fig. 2(e), but it may be that for this field direction some additional heat carriers are induced and add a major contribution on top of $\kappa_p(H)$. One possibility is a remaining contribution of Pr magnons, although as much as 35% of the Pr^{3+} sites are diluted by nonmagnetic La^{3+} ions in PLCO. Remember that La-free PCO single crystals has been reported to exhibit a large enhancement of κ with H,[25] which might share a common origin to the present behavior shown in Fig. 2(e).

Although the interaction between phonons and the free spins has been known for a long time,[38] such phenomenon and its impact on the heat transport have never been revealed for cuprate materials. The present results demonstrate that the phonon heat transport can be H dependent at low temperature in high-T_c cuprates whenever excitations of free spins (that cause a Schottky anomaly in the specific heat) are present. The effect of the paramagnetic scattering is particularly pronounced in PLCO, probably because in this parent compound the phononic heat transport is very clean and also the Schottky anomaly is large. In the superconducting doping regime of cuprates, while the Schottky anomaly is almost ubiquitously observed,[9–15] its contribution is much smaller than that in PLCO and also the phonons are additionally scattered by doped carriers, both of which cause the contribution of the paramagnetic scattering to be reduced. Nevertheless, it is likely that some fraction of the H dependence of κ is due to the paramagnetic scattering of phonons even in superconducting samples, and therefore an analysis of $\kappa(H)$ should be done very carefully. Note that, while *quantitative analyses* of $\kappa(H)$ would be difficult in light of the paramagnetic scattering of phonons, *qualitative comparisons* of the $\kappa(H)$ behavior for different dopings[5,6] would still be legitimate, because the Schottky anomaly is observed across the whole doping range without any clear doping dependence.

Last, the present result suggests, together with the curious magnetic shape-memory effect in $La_{2-x}Sr_xCuO_4$,[39] that the spin-phonon coupling is rather strong in cuprates. The significance of the spin-phonon coupling in *doped* cuprates, although still remains to be verified, is rather naturally expected based on the behavior of the parent compound. Because of this coupling, AF fluctuations and phonons may couple in an unusual way in these materials and this could be the reason why the mechanism of high-T_c superconductivity is so difficult to understand. In this regard, it is useful to note that possible roles of the spin-phonon coupling in the peculiar electronic state of doped cuprates have been theoretically discussed.[40,41]

IV. SUMMARY

We measure the magnetic-field dependence of the thermal conductivity of a parent insulating cuprate $Pr_{1.3}La_{0.7}CuO_4$ to elucidate the possible field dependence of phonon heat transport in cuprates. A striking dip feature is found in $\kappa(H)$ isotherms for in-plane fields at low temperature. The dip field is found to increase linearly with T at low temperature, which is indicative of the phonon scattering by free spins. Both the low-T specific heat and the magnetic susceptibility are measured to give supplemental evidence for the existence of free spins. Since the Schottky-type specific heat that signifies the existence of free spins is found ubiquitously in high-T_c cuprates in a wide doping range, the present result suggests that the H dependence of the phonon heat transport should not be naively neglected, particularly when one tries to extract the QP information from the quantitative analyses of $\kappa(H)$. In addition, the present result points to a strong spin-phonon coupling in high-T_c cuprates.

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