Doping dependence of phonon and quasiparticle heat transport of pure and Dy-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals

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The temperature and magnetic-field (H) dependences of thermal conductivity (κ) of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) are systematically measured for a broad doping range by using both pure Bi2212 single crystals with tuned oxygen contents and $Bi_2Sr_2Ca_{1-x}Dy_xCu_2O_{8+\delta}$ (Dy-Bi2212) single crystals with different Dy contents x. In the underdoped samples, the quasiparticle (QP) peak below T_c is strongly suppressed, indicating strong QP scattering by impurities or oxygen defects, whereas the phonon conductivity is enhanced in moderately Dy-doped samples and a phonon peak at 10 K is observed, which means Dy^{3+} ions not only introduce the impurities or point defects but also stabilize the crystal lattice. The subkelvin data show that the QP heat conductivity gradually decreases upon lowering the hole doping level. The magnetic-field dependence of κ at temperature above 5 K is mainly due to the QP scattering off vortices. While the underdoped pure Bi2212 show very weak field dependence of κ, the Dy-doped samples present an additional "dip-like" term of $\kappa(H)$ at low field, which is discussed to be related to the phonon scattering by free spins of Dy^{3+} ions. For nonsuperconducting Dy-Bi2212 samples with $x \simeq 0.50$, an interesting "plateau" feature shows up in the low-temperature $\kappa(H)$ isotherms with characteristic field at 1–2 T, for which we discuss the possible revlevance of magnon excitations.

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I. INTRODUCTION

Low-temperature heat transport has been extensively studied for high-T_c cuprates, since it not only provides the most straightforward information of quasiparticle (QP) transport properties and the peculiar electronic state but also shows rich physics such as the magnon heat transport of low-dimensional spin system, the phonon heat transport, and its interaction with peculiar spin and/or charge order. It was predicted that the heat transport of nodal QPs in a d-wave superconductor presents a "universal" behavior,[1–3] that is, the residual conductivity in the zero-T limit κ_0/T is independent of QP scattering rate. Physically, it results from the exact compensation between the decrease of scattering time and the increase of zero-energy density of states with increasing impurity density. In the standard self-consistent T-matrix approximation theory,[1] when the impurity bandwidth γ is in the universal limit $k_BT \ll \gamma \ll \Delta_0$ (Δ_0 is the maximum gap), κ_0/T is expressed as[2,3]

$$\frac{\kappa_0}{T} = \frac{k_B^2}{3\hbar}\frac{n}{c}\left(\frac{v_F}{v_2} + \frac{v_2}{v_F}\right) \simeq \frac{k_B^2}{3\hbar}\frac{n}{c}\frac{v_F}{v_2}, \tag{1}$$

with n the number of CuO_2 planes per unit cell, c the c-axis lattice constant, and v_F (v_2) the QP velocity normal (tangential) to the Fermi surface at the gap node.

The early measurements on nearly optimally doped $YBa_2Cu_3O_y$ (YBCO)[4] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212)[5,6] with various impurity scattering strengths were found to be supportive of the universal behavior. If valid, the universal nature is very useful because one can obtain Δ_0 from the bulk measurement of κ.[7,8] Sutherland *et al.*[7] analyzed their residual conductivity data of YBCO in the framework of the universal thermal conductivity and obtained the gap size Δ_0 from κ_0/T; based on this analysis, it was argued that the Δ_0 values coincide with those obtained from other spectroscopic measurements. Together with the similar analysis on the κ data of $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201) in a wide overdoping range,[8] a pure and simple d-wave superconducting state throughout the phase diagram was proposed. However, as more detailed and accurate data are accumulated, it has become obvious that there are cases that do not seem to obey the universal behavior, including the increase of κ_0/T with doping level confirmed in $La_{2-x}Sr_xCuO_4$ (LSCO),[7,9] YBCO,[7,10,11] and $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ (BSLCO)[12] and the varnishing residual thermal conductivity in an underdoped $YBa_2Cu_4O_8$ (Y124).[13] A possible explanation for these discrepancies was suggested based on a systematic study of Zn-doping effect on the residual thermal conductivity of several typical cuprate families.[14] By using exceptionally high-quality single-crystal samples and achieving very small experimental errors, it was found that the universal picture is not a precise description of the low-T QP transport properties,[14] whereas the electronic inhomogeneity was found to be the main reason for the failure of the classical d-wave theory.[15] It should be mentioned that there is a controversy over the data analysis of the very-low-T thermal conductivity, that is, whether the commonly observed T dependence of κ/T data below 1 K weaker than T^2 is due to the phonon specular reflection off the sample surface or simply because the phonon boundary scattering can only be achieved at temperature lower than

$\sim$130 mK.[4,7,11,14] The key question is whether the phonons can be scattered by something else except for the boundary at *very low temperatures*. An indirect but helpful result for this issue was recently obtained in another exotic oxide material $GdBaCo_2O_{5+x}$; the phonon scattering by free spins in this material was found to be very active in subkelvin temperatures, which makes the boundary scattering limit not achievable at temperature as low as 0.3 K.[16]

The magnetic-field dependence of low-T thermal conductivity is very informative for the QP transport properties. The pioneering work in nearly optimally doped Bi2212 discovered a striking "plateau" behavior of $\kappa(H)$ isotherms in the temperature range down to 4 K.[17] It was originally discussed that the plateau behavior is due to some kind of field-induced phase transition,[17,18] which, however, was questioned by a subsequent experimental observation that the κ value in the plateau depends on the history of applying the magnetic field.[19] It was then proposed that this behavior can be well understood by considering the competition between the QP scattering by vortices, which is predominant at higher temperature,[17,20–22] and the field-induced QP excitations, which is known as the "Volovik" effect that is more important at low temperature.[21,23] This explanation was supported by the $\kappa(H)$ measurements at subkelvin temperatures.[24,25] Furthermore, the in-plane anisotropic heat transport measurements done on Bi2212 single crystals discovered that the plateau behavior is only present for heat current along the b axis,[26] probably due to the distortion of $d_{x^2-y^2}$ pairing symmetry. Subsequently, more efforts were devoted for the studying of $\kappa(H)$ behaviors at subkelvin temperature for cuprate superconductors with different doping levels.[10,12,27,28] One interesting finding is the field-induced suppression of the very-low-T heat conductivity in the underdoped $La_{2-x}Sr_xCuO_4$ (LSCO), whereas the magnetic field naturally enhances the thermal conductivity of optimally doped and overdoped LSCO as expected from the Volovik effect.[27,28] The unusual behavior in the underdoped LSCO can be understood as the QP localization associated with the magnetic-field-induced spin or charge order.[27,29–33] Similar $\kappa(H)$ results have been reported for weakly doped YBCO and BSLCO.[10,12] Note that in all these previous studies, the field dependence of thermal conductivity was attributed only to the electron transport and the contribution of phonon heat conductivity was assumed to be independent of magnetic field. However, it was recently found that in an undoped compound $Pr_{1.3}La_{0.7}CuO_4$ (PLCO), dilute ($\sim$1%) free 1/2 spins (which are presumably created by a small oxygen nonstoichiometry) can strongly scatter phonons and produce a pronounced "dip" behavior in the $\kappa(H)$ isotherms at low temperatures.[34] Such an observation implies that the phonon heat transport could be sensitive to magnetic field in high-T_c cuprates, which are known to commonly possess some local magnetic moments.[35–41] Apparently, correctly understanding the phonon heat transport property becomes crucial, not only for its own research interests but also for its role in explaining the magnetic-field dependence of low-T thermal conductivity and the issue of boundary scattering limit, which are fundamental for capturing the QP transport properties.

In this work, we study the low-T thermal conductivity of high-quality Bi2212 single crystals at various dopings to probe both the QP heat transport and the phonon heat transport properties. It is worth mentioning that the doping dependence of the heat transport has been seldom studied for Bi2212.[14] Besides confirming the decrease of QP heat transport with decreasing doping by either tuning the oxygen content or doping Dy, we obtain several results: (i) Dy substitution for Ca stabilizes the crystal lattice and leads to the improvement of the phonon heat transport; (ii) the low-T phonon peak in $\kappa(T)$ curves is observed in appropriately Dy-doped samples; (iii) Dy doping introduces some magnetic-field dependence of phonon heat transport and a plateau phenomenon in $\kappa(H)$ isotherms is found in the nonsuperconducting Dy-doped samples.

II. EXPERIMENTS

High-quality pure $Bi_2Sr_2CaCu_2O_{8+\delta}$ and Dy-doped $[Bi_2Sr_2Ca_{1-x}Dy_xCu_2O_{8+\delta}]$ single crystals are grown by the floating zone method.[22,26] The pure crystals are carefully annealed at 400–800 °C and in different atmospheres to tune the oxygen content, e.g., the optimal doping with $T_c=95$ K is obtained by annealing at 800 °C in air. Most of the Dy-doped samples, if not specially mentioned, are annealed at the same condition as that of the optimal doping. We label the pure samples as UD70, OP95, OD70, etc., for showing their doping regimes and T_c values, and Dy-doped samples as Dy81, Dy45, etc., by their T_c values (the Dy doping effectively reduces the hole concentration and brings the samples into the underdoped regime). The actual Dy concentration x is determined by the inductively coupled plasma atomic-emission spectroscopy.

The thermal conductivity κ is measured along the a axis, perpendicular to the direction of the incommensurate modulation structure. The typical size of the sample is $2.5\times0.8\times0.03$ mm^3, where the longest and the shortest dimensions are along the a and c axes, respectively. The measurements are done by several different processes: using a conventional steady-state technique by using Chromel–Constantan thermocouple in a ^{4}He cryostat for $\kappa(T)$ data at "high" temperatures (2–150 K) and for $\kappa(H)$ data above 40 K; using a "one heater, two thermometers" technique with Cernox sensors in the ^{4}He cryostat for $\kappa(H)$ data at 5–30 K; using a one heater, two thermometers technique with RuO_2 sensors in a dilution refrigerator or a ^{3}He refrigerator for $\kappa(T)$ data at subkelvin temperatures and for $\kappa(H)$ data below 10 K. All the measurement techniques for κ have been described in our previous publications.[9,10,14,26,27,34] The error and irreproducibility of thermal conductivity data are typically less than 10%. Magnetization measurements are carried out using a Quantum Design superconducting quantum interference device magnetometer.

III. RESULTS AND DISCUSSION

A. Electrical transport and carrier concentration

Figure 1(a) shows the in-plane resistivity of several Dy-doped Bi2212 single crystals. The optimally doped sample (without Dy doping) shows a "negative" residual resistivity,[22] suggesting very high cleanliness of our crystals.

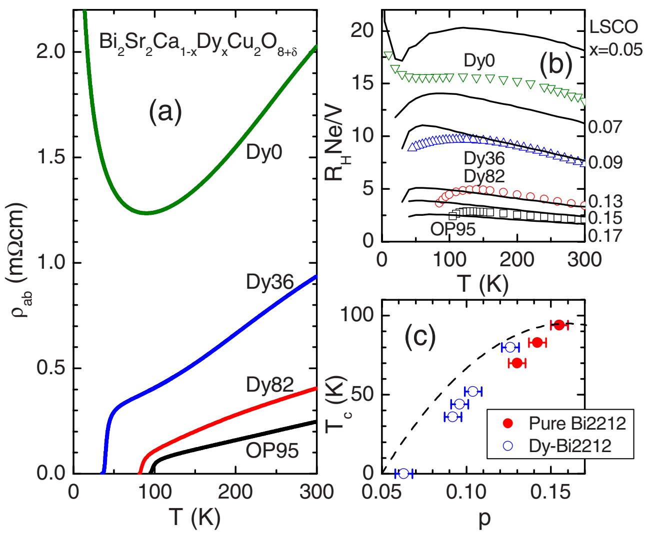

FIG. 1. (Color online) (a) In-plane resistivity of several pure and Dy-doped Bi2212 single crystals. (b) Renormalized Hall coefficient of these crystals. LSCO data (solid lines) from Ref. 44 are included for comparison. (c) T_c vs p for several pure and Dy-doped Bi2212 single crystals. Doping dependence of T_c does not follow the empirical formula $T_c/T_c^{max}=1-82.6(p-0.16)^2$, shown by the dashed line.

Upon doping Dy, the superconductivity is gradually suppressed and the increase of residual resistivity is rather weak, thus the main reason for the suppression of the superconductivity is the reduction of hole concentration when substituting Dy^{3+} ions for Ca^{2+} ions.

It is difficult to determine the carrier concentration in Bi2212 because the hole concentration per Cu, p, cannot be chemically determined like in LSCO. Ando *et al.* have shown that the magnitude of renormalized Hall coefficient $R_H Ne/V$, where N is the number of Cu atoms in a unit cell and V is the volume of the unit cell, can be used as a tool to identify the p value.[42] Figure 1(b) compares the $R_H Ne/V$ of the Dy-doped Bi2212 samples[43] with those of LSCO,[44] from which the p values can be easily estimated with rather small uncertainties. Figure 1(c) shows the obtained T_c vs p relation for several pure and Dy-doped Bi2212 single crystals.[43] Note that the doping dependence of T_c shows some deviations from the empirical formula $T_c/T_c^{max}=1-82.6(p-0.16)^2$,[45] which is often used for determining the hole concentration in Bi2212. In particular, the boundary between nonsuperconducting and superconducting phases turns out to be located at $p\approx 0.07$.

B. Temperature dependence of thermal conductivity at high temperature

Figure 2 shows the thermal conductivities of several pure and Dy-doped Bi2212 single crystals from 2 to 150 K. The optimally doped sample shows a well-known broad "QP peak" below T_c as well as a "knee" feature around 10 K, whose origin has never been clarified. The size of QP peak (the enhancement from the minimum near T_c to the peak is 1.1 W/K m or 25%) is among the largest in Bi2212 single crystals that have ever been studied,[17,22,26,46] confirming the high quality of the crystals. In pure samples, either increasing or decreasing the hole concentration results in the weakening of the QP peak, while the magnitude of total thermal conductivity monotonously decreases with decreasing the hole concentration. Furthermore, the peak suppression is much more significant in the underdoped samples than that in the overdoped samples, which is possibly related to the nanoscale inhomogeneity found by scanning tunneling microscopy experiments in the superconducting state of this compound.[47–49] This behavior is very similar to that in the Zn-doped Bi2212,[22] in which local suppression of the superconductivity leading to a patchy superconducting state is playing a role.[50] Upon doping Dy, the QP peak is also strongly suppressed but a new peak at 10 K emerges. Interestingly, in the Dy81 ($x=0.17$) sample, both the QP peak and the 10 K peak show up, which means that the 10 K peak is not directly related to the QP heat transport. This low-T peak is the strongest in the Dy45 ($x=0.30$) sample and survives in the nonsuperconducting sample (Dy0, $x=0.53$), which clearly indicates that it originates from the phonon heat transport.

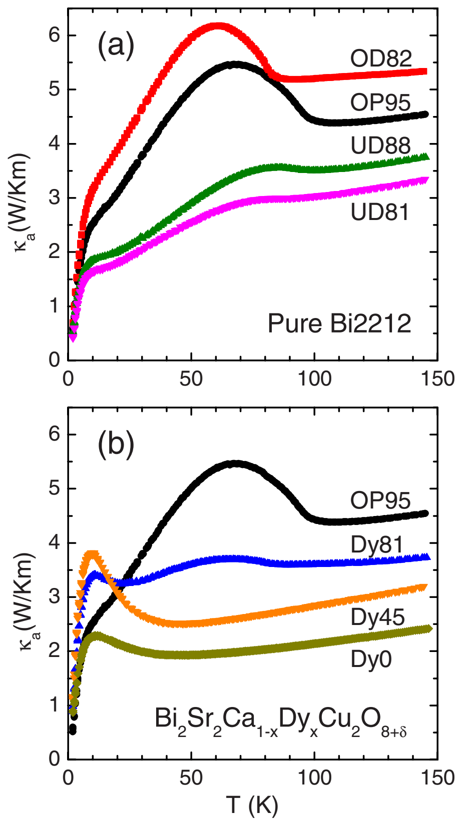

FIG. 2. (Color online) Temperature dependence of thermal conductivity of pure and Dy-doped Bi2212 single crystals. The Dy contents x for Dy81, Dy45, and Dy0 samples are 0.17, 0.30, and 0.53, respectively.

In the undoped compound of other cuprates, such as La_2CuO_4, $YBa_2Cu_3O_6$, Nd_2CuO_4, and $Pr_{1.3}La_{0.7}CuO_4$, the phonon heat transport behaves similarly to that of common insulators and shows a large phonon peak around 20 K, whose magnitude varies between $\sim$20 and

~80 W/K m.[34,51–54] However, it is known that Bi2212 system has much dirtier phonon heat transport than other cuprates,[46] probably due to the strong disorder of the crystal lattice caused by the excess oxygen and the incommensurate superlattice along the *b* axis. The incommensurate modulation had been discussed to be related to the extra oxygen locating in BiO double layers and the lattice mismatch between the BiO block and the CuO_2 layer.[55] In an earlier work, the thermal conductivity of Bi2212 parent material was studied in $Bi_2Sr_2YCu_2O_8$ and it was found that the phonon heat transport is too weak to give rise to the phonon peak (only a humplike feature observed at 20–30 K with the magnitude of κ smaller than 2 W/K m).[46] To our knowledge, the phonon peak was never observed before in Bi2212. It is not surprising that the phonon peak is located at much lower temperature than in other cuprates if the defect scattering is much stronger in Bi2212. The present result also indicates that a moderate Dy doping can enhance the phonon conductivity rather strongly despite that the atomic substitution always introduces additional impurity and/or defect scattering on phonons. Apparently, the crystal structure is somewhat stabilized by doping an appropriate amount of Dy. A possible scenario is that substituting Ca^{2+} ions with Dy^{3+} increases the oxygen content and results in the expansion of the BiO bolck, which is helpful for relaxing the lattice mismatch between the BiO block and CuO_2 plane.[55] Note that the low-*T* peak observed in the Dy-doped samples clarifies that the knee feature of $\kappa(T)$ in superconducting Bi2212 single crystals is due to the competition between the decrease of QP heat transport and the increase of phonon heat transport upon lowering temperature across 10 K. Furthermore, the phonon heat transport in pure Bi2212 is obviously very weak, and its magnitude at 10 K (where the phonon conductivity peaks) is smaller than or at most comparable to that in the nonsuperconducting Dy-Bi2212.

Similar to the pure Bi2212 crystals, the heat transport of Dy-Bi2212 is also sensitive to the oxygen content. After appropriate annealing to increase the oxygen content, for example, the $x=0.30$ crystal (Dy45 in Fig. 2) presents a parallel-shift-like enhancement of the thermal conductivity at first glance, as shown in Fig. 3. However, we should note that the thermal conductivity enhancement above T_c is mainly caused by the increase of the electron heat transport (which can be roughly estimated from the normal-state resistivity data using the Wiedemann–Franz law), while the enhancement of the 10 K peak should be mainly due to the improvement of phonon transport since the QP (or electron) heat transport in this temperature region is expected to be strongly damped in highly Dy-doped samples. It is likely that the oxygenating process reduces the number of oxygen vacancies in the crystal, which scatter phonons. The magnitude of the phonon peak at 10 K increases from 3.8 to 5.2 W/K m, but both curves do not show the QP peak although the superconducting temperature increases from 45 to 55 K.

C. Temperature dependence of thermal conductivity at subkelvin temperature

To further investigate the doping dependence of QP heat transport, the thermal conductivities of Bi2212 and Dy-

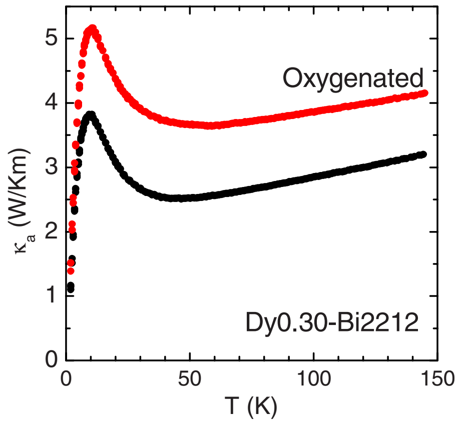

FIG. 3. (Color online) Thermal conductivity of Dy-doped Bi2212 ($x=0.30$) single crystal (annealed at 800 °C in air) and the same sample after oxygenating heat treatment (annealed at 500 °C in air).

doped Bi2212 single crystals are measured at very low temperatures down to 70 mK. Some of the data have been reported in a previous publication (see Figs. 3 and 4 in Ref. 14). It has been shown that in the lowest-*T* regime where phonons are in the boundary scattering limit, the QP term and the phonon term of thermal conductivity can be separated by fitting data (below ~130 mK) to

$$\frac{\kappa}{T} = \frac{\kappa_0}{T} + bT^2, \tag{2}$$

where the residual term κ_0/T is the QP contribution and bT^2 is the phonon contribution.[4,9,10,14] Note that the residual thermal conductivity of a *d*-wave superconductor was discussed to present a universal behavior, that is, it is independent of the QP scattering rate if the impurity bandwidth satisfies $k_BT \ll \gamma \ll \Delta_0$ (Δ_0 is the maximum gap).[2,3] In this picture, the QP heat conductivity (κ_e/T) of cuprate superconductor decreases with temperature and finally arrives at a certain value, which depends only on the doping level, at low enough temperature. However, our Bi2212 crystals clearly show much larger residual thermal conductivity compared to the samples used in earlier works.[5,6] Judging from the methods of growing single crystals and the normal-state resistivity data,[5,6] it is obvious that the difference in κ_0/T is strongly related to the cleanliness of the crystals, which actually challenges the validity of the universal behavior. Indeed, this issue was recently elucidated by a critical examination of the universal behavior in a well-controlled experiment;[14] it was found that the disorder or inhomogeneity of cuprates, which are especially strong in Bi2212, makes the universal behavior to be only a rough approximation. Nonetheless, the residual thermal conductivity universally shows a monotonous doping dependence in all cuprates.

Recently, a controversy arose for the data analysis of the very-low-*T* thermal conductivity of high-T_c cuprates; namely, whether the commonly observed *T* dependence of κ/T data below 1 K weaker than T^2 is due to the phonon specular reflection off the sample surface or simply because the phonon boundary scattering can only be achieved at temperatures lower than $\sim$130 mK.[4,7,11,14] In this regard, although Taillefer *et al.* was the first to propose the analysis of Eq. (2) and successfully described the very-low-T data of YBCO,[4] they have recently shifted to conjecture the existence of specular reflections in a wide temperature range and analyzed their data of YBCO and LSCO by fitting to

$$\frac{\kappa}{T} = \frac{\kappa_0}{T} + aT^{\gamma}, \tag{3}$$

where the second term with $\gamma < 2$ is purported to be the phonon conductivity in the specular scattering limit.[7] However, it was recently pointed out[56] that one can estimate the phonon mean-free path ℓ_p in the temperature range of interest, which shows that ℓ_p is too short for the specular-reflection scenario to be valid. Furthermore, the analysis involving Eq. (3) analysis is actually based on another assumption that the QP thermal conductivity is independent of temperature up to at least $\sim$1 K, which is inconsistent with an experimental finding from the same group[57] that in YBCO, the QP thermal conductivity at finite temperature increases quickly with temperature, following a T^3 dependence.[58] In this sense, the "good" fitting of Eq. (3) to some experimental data is not physically meaningful.

If one tries to analyze the data of Bi2212 using Eq. (3), it can be found that the low-T thermal conductivity actually follows

$$\frac{\kappa}{T} = \frac{\kappa_0}{T} + AT, \tag{4}$$

below 0.6 K down to nearly 100 mK, as shown in Fig. 4; here, the power of T in the second term is clearly too small to be attributed to the phonon specular reflection of phonons.[7] Note that the same temperature dependence of κ has recently been observed in $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201) system, whose doping levels are located in the overdoped regime.[8] Hawthorn *et al.* attributed the phonon conductivity of AT to the dominant electron-phonon scattering with high carrier concentration of Tl2201,[8] like the case in a metal, which actually means that the phonon boundary scattering limit *can never be established* in this system. Our present results, on the other hand, are observed mostly in the *underdoped* or *optimally doped* regions of Bi2212, indicating that the AT term may not be the simple phonon contribution. Considering both the much larger κ_0/T term of Bi2212 compared to other cuprates and the much weaker phonon transport seen in the high-T region (Fig. 2), it is reasonable to conclude that the T^2 dependence of the low-T thermal conductivity (above $\sim$100 mK) of Bi2212 is related to an amorphouslike phonon transport[59] together with a T^2-dependent QP heat conductivity. This is essentially consistent with a previous finding in Bi2212 by Nakamae *et al.*[5]

To determine the residual term κ_0/T at zero temperature by employing some extrapolation of the data at finite temperature, one should always keep in mind that the formula used for the extrapolation must have a physical reasoning, which assures that the same functional form holds throughout the range of the extrapolation (to 0 K). Therefore, although Eq. (4) can fit the Bi2212 data pretty well in a broad temperature range, this fitting *cannot* provide the precise determination of κ_0/T because the T^2-dependent QP heat conductivity does not hold at very low temperature ($\sim$100 mK or lower) when the temperature scale becomes smaller than the impurity bandwidth.[2,5] For this reason, it is still the most reliable way to obtain κ_0/T by fitting the lowest-T data to Eq. (2), as we did in Ref. 14.

In passing, it should be pointed out that the very-low-T data of overdoped Bi2212 show a significant downturn below 0.2 K, which is known to be due to the electron-phonon decoupling at very low temperatures,[60] which is often significant in the overdoped cuprates.[61] This phenomenon further demonstrates that the T^2 behavior of low-T thermal conductivity in this system does not have the same origin as that in Tl2201, which was discussed to be the strong electron-phonon scattering.[8]

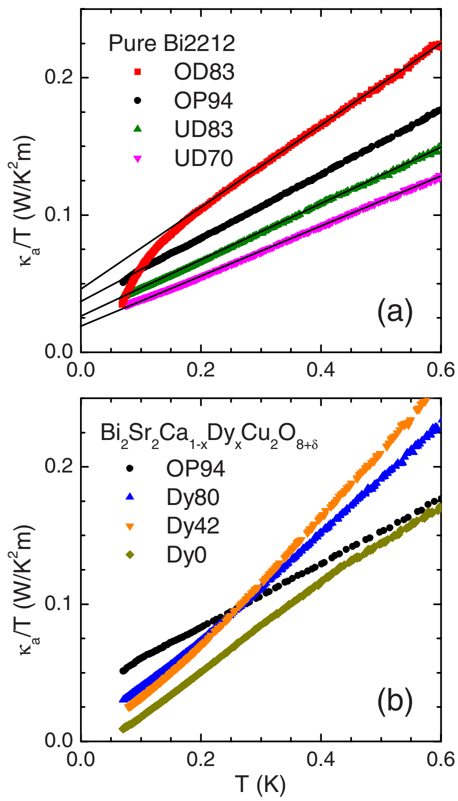

FIG. 4. (Color online) Low-T thermal conductivity of pure and several Dy-doped Bi2212 single crystals measured down to 70 mK, plotted as κ/T vs T. The Dy contents x for Dy80, Dy42, and Dy0 samples are 0.20, 0.34, and 0.51, respectively. Strong downturn of the low-T data of OD83 sample is caused by the electron-phonon decoupling.

D. Magnetic-field dependence of thermal conductivity

The magnetic-field dependence of both pure and Dy-doped Bi2212 crystals is studied in detail for $H \parallel c$. Figures 5(a) and 5(b) show the results of OP95 sample at 5–100 K, which are essentially consistent with previous studies.[17,22,26]

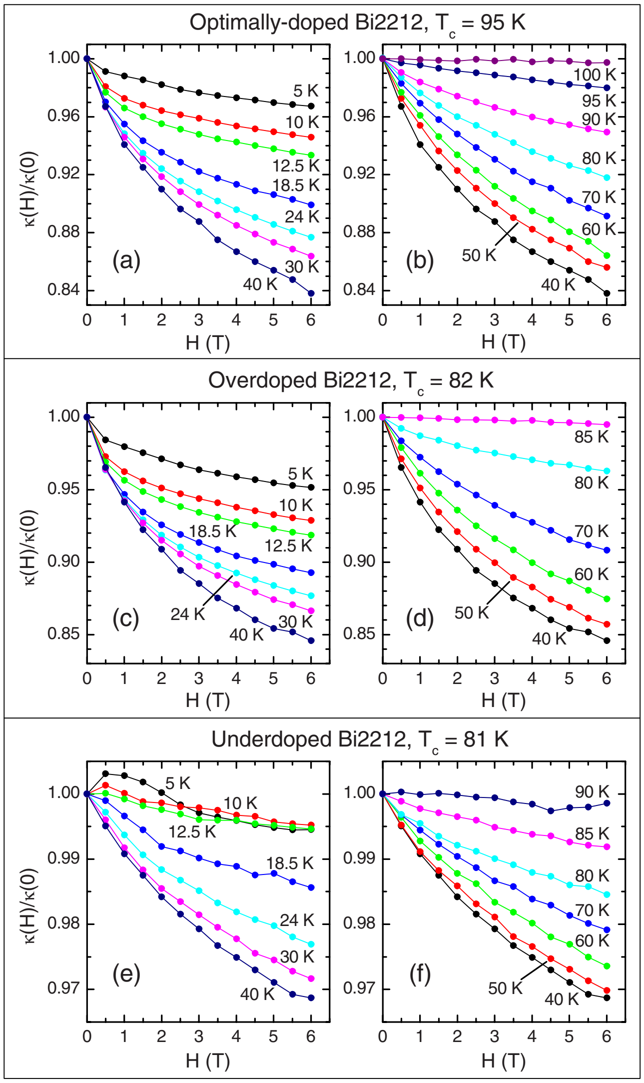

FIG. 5. (Color online) Magnetic-field dependences of the low-T thermal conductivity of the pure Bi2212 single crystals, whose doping levels cover from underdoped to overdoped regions. The field direction is along the c axis.

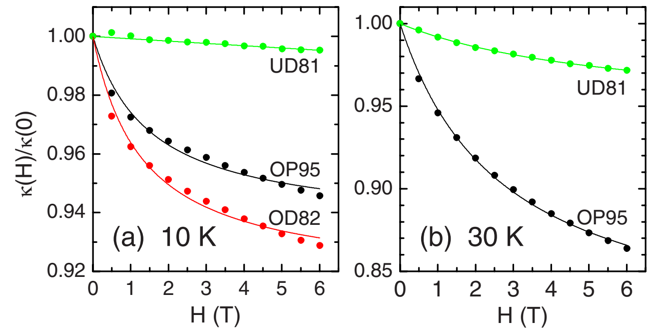

FIG. 6. (Color online) Representative $\kappa(H)$ isotherms of pure Bi2212 single crystals with different doping level and the corresponding theoretical fitting curves using Eq. (5).

Note that the famous plateau feature is absent in these $\kappa(H)$ isotherms since the thermal conductivity is always measured along the a axis in the present work. As shown in Fig. 6, the field dependence of κ can be well described by the simple relation,

$$\kappa(H,T) = \frac{k_e(T)}{1+p(T)H} + \kappa_{ph}(T), \tag{5}$$

where κ_e and κ_{ph} are the electronic thermal conductivity and the phononic thermal conductivity, respectively; here, the field dependence is attributed to the quasiparticle scattering off vortices.[22] Apparently, the amplitude of the field dependence of κ is dependent on the contribution of QPs to the total thermal conductivity. One should notice that the magnetic-field dependence of QP thermal conductivity in d-wave cuprates is determined by the competition between the decrease of heat conductivity due to the vortex scattering and the increase of heat conductivity due to the increase of the nodal QP excitations, the so-called Volovik effect.[20–25] The former effect is dominant at high temperature, whereas the latter one is more important at low temperature. Thus, the weakliness of the field dependence of κ at low temperatures could be due either to the decrease of QP portion of heat conductivity or to the Volovik effect, or both.

As shown in Figs. 5(c)–5(f), the field dependence of κ is also rather strong in the overdoped sample, almost the same as that in the optimally doped sample, whereas it is weakened by several times in the underdoped sample. Nevertheless, the $\kappa(H)$ data in these pure Bi2212 crystals at all temperatures studied are well described by Eq. (5) (see Fig. 6 for several representative data), which means the QP scattering mechanism does not change upon tuning the oxygen content. Apparently, the weakening of $\kappa(H)$ in underdoped sample is due to the suppression of the QP contribution to the heat conductivity, consistent with the conclusion from the temperature dependence of data.

Note that the above discussion is based on an assumption that the phonon thermal conductivity is independent of magnetic field, which, however, has recently been found to be not always valid for high-T_c cuprates. In a parent compound of high-T_c cuprates, PLCO, in which the phononic heat transport is very clean, diluted free spins associated with a small oxygen nonstoichiometry introduce strong magnetic-field dependence of phonon thermal conductivity through the phonon scattering off free spins.[34] Since the localized free spins are commonly found in superconducting cuprates, as is evidenced by the Schottky anomaly in the specific heat,[35–41] some fraction of the magnetic-field dependence of κ is likely due to the spin-phonon scattering. In the case of pure Bi2212, however, the effect of spin scattering on the phonon heat transport would be unimportant because the phonons are strongly scattered by the lattice disorder or defects.

The magnetic-field dependences of the thermal conductivity of Dy-doped samples with x=0.17, 0.30, and 0.51 are shown in Fig. 7. Similar to the case of Dy-free crystals, the field dependence of κ is gradually weakened with decreasing carrier concentration, which should also be due to the reduction of the electronic contribution to the heat conductivity. An interesting point is that the κ in Dy-Bi2212 samples show more complicated magnetic-field dependences. For x=0.17,

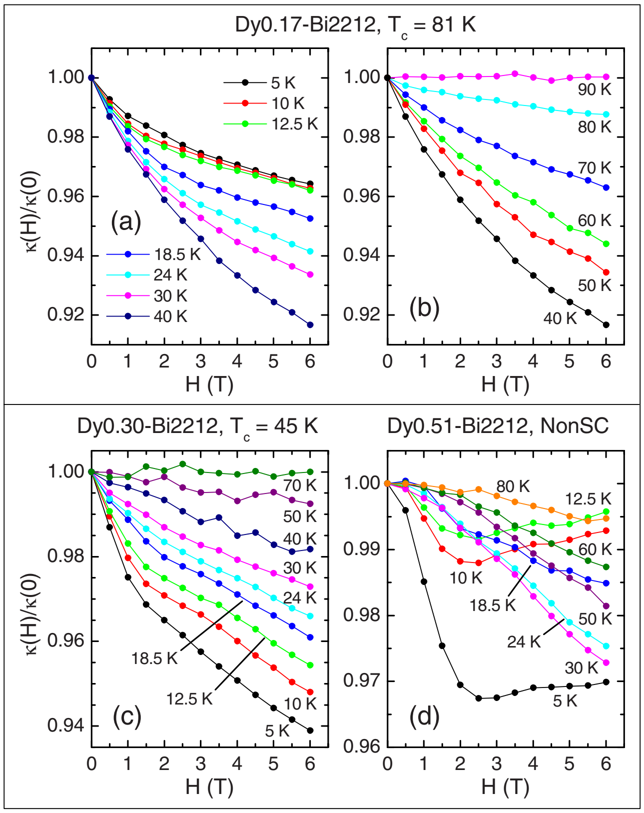

FIG. 7. (Color online) Magnetic-field dependences of the low-T thermal conductivity of the Dy-Bi2212 (x=0.17, 0.30, and 0.51) single crystals, which are superconducting at 81 K, 45 K, and non-superconducting, respectively.

the magnetic-field dependence of κ can still be well described by Eq. (5). On the other hand, for x=0.30, an S-shaped feature at 1–3 T shows up in the $\kappa(H)$ isotherms below 30 K; the fitting to these data using Eq. (5), as shown in Fig. 8, suggests that there is an additional diplike change of κ superimposed on the background of usual field dependence described by Eq. (5) and the position of the dip slightly moves to lower field with decreasing temperature. For the x=0.51 sample, in which the overall field dependence is much weaker, a steplike decrease of κ clearly occurs around 2 T at low temperatures.

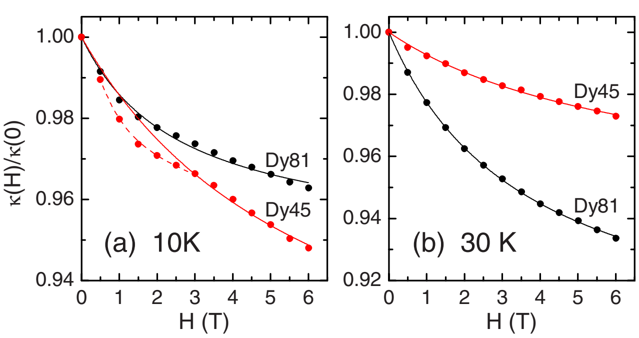

FIG. 8. (Color online) Representative $\kappa(H)$ isotherms of Dy-doped Bi2212 single crystals with x=0.17 and 0.30 and the corresponding theoretical fitting curves (solid lines) using Eq. (5). The dashed line indicates the deviation of experimental data from the theoretical curve.

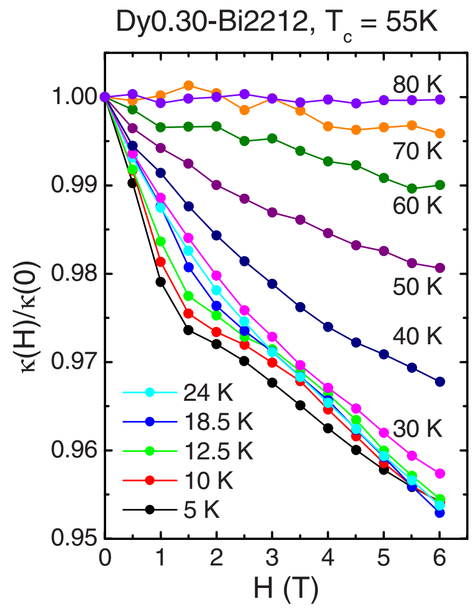

FIG. 9. (Color online) Magnetic-field dependences of the low-T thermal conductivity of x=0.30 sample after reannealed at 500 °C in air.

To prove whether the peculiar $\kappa(H)$ behavior for higher Dy dopings is caused by the oxygen defects, the field dependences of κ are also measured for the x=0.30 sample after reannealed at 500 °C in air, which increases the oxygen content and increases the T_c from 45 to 55 K (the temperature dependence of κ is shown in Fig. 3). It can be seen from Figs. 7(c) and 9 that the profiles of $\kappa(H)$ do not change so much with increasing oxygen content except that the diplike feature becomes more pronounced at low temperatures. The position of the dip is still located at 1–3 T. This indicates that the oxygen defects are not directly related to the physical origin of this peculiar feature.

Since both the quasiparticle transport and the oxygen defects are not likely to be producing the additional contribution to the field dependence of κ, it is natural to conclude that the Dy^{3+} ions are playing the key role. Apparently, the x =0.30 sample does not have any long-range order of Cu spins or Dy spins, so the Dy^{3+} ions are most likely affecting the heat transport as free spins. A common magnetic scattering on phonons is caused by free spins or paramagnetic moments through spin-phonon interaction.[59] In this case, the energy splitting of the spin states, which induces resonant scattering on phonons, is increased by the Zeeman effect. The phonon scattering off these spins is most effective in suppressing the heat transport when the energy splitting is equal to $\sim 3.8k_BT$, where the spectrum of phonon heat conductivity peaks;[16,59] therefore, the spin-phonon scattering usually generates a diplike feature in $\kappa(H)$ at this energy and the dip position shifts to higher fields with increasing temperature.[59] As we have mentioned, the effect of the magnetic phonon scattering on the heat transport has been known for a long time and has recently been found to affect the field

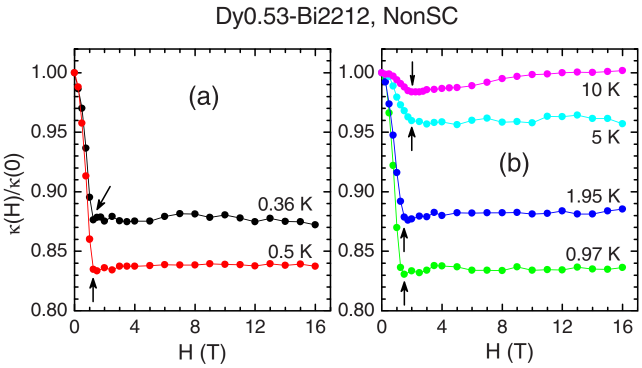

FIG. 10. (Color online) Magnetic-field dependences of the low-T thermal conductivity of the Dy-Bi2212 (x=0.53) single crystal down to 0.36 K. The magnetic field is applied along the c axis.

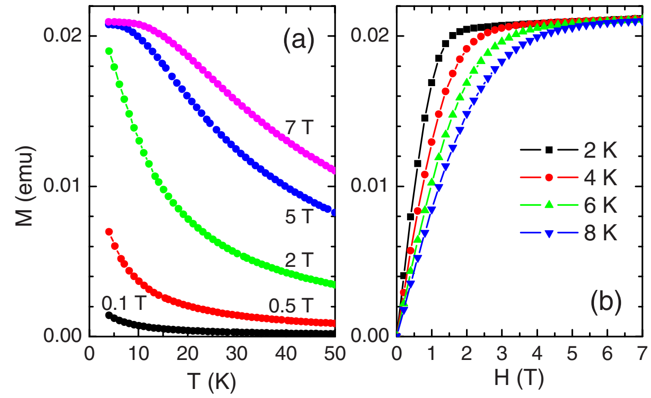

FIG. 11. (Color online) Temperature and field dependences of magnetization of a nonsuperconducting Dy-Bi2212 single crystal with x=0.53.

dependence of κ in cuprates.[34] While the magnetic scattering on phonons seems unimportant in pure Bi2212 single crystals, in which the phonon heat transport is strongly damped by defects and lattice disorders, it becomes much more pronounced upon doping Dy that not only brings free spins but also enhances the phonon heat transport.

As shown in Fig. 7(d), a steplike $\kappa(H)$ isotherm is observed at low temperatures in the x=0.51 sample. Is this also due to the phonon scattering by free spins? In this scenario, if the energy splitting is larger than $3.8k_BT$, the magnetic field further increases the energy splitting and simply weakens the scattering and produces a steplike *increase* of thermal conductivity.[16] Apparently, the steplike *decrease* of $\kappa(H)$ in Dy-Bi2212 is out of the expectation from the common spin-phonon scattering mechanism and suggests some other physical mechanism. Since the x=0.51 sample is nonsuperconducting with an insulating resistivity behavior at low temperatures, the steplike $\kappa(H)$ must also be irrelevant to the electron transport.

E. Plateau of low-temperature $\kappa(H)$ isotherms in Dy-Bi2212

In Fig. 7(d), the $\kappa(H)$ isotherm of x=0.51 sample at 5 K shows a steplike decrease at low field and κ becomes nearly independent of magnetic field above 2 T, a plateaulike behavior. We note that it is not clear whether this plateau behavior is the same as that observed in pure Bi2212 along the b axis[26] because the latter is believed to have its origin in the QP transport, while in the present case, the sample is an insulator. To investigate this intriguing behavior in Dy-doped Bi2212, the magnetic-field dependence of κ is studied in more detail at very low temperature down to 0.36 K and in high magnetic field up to 16 T (along the c axis). As shown in Fig. 10, the plateaulike behavior is well reproduced in another sample of x=0.53 at low temperatures. With lowering temperature, the steplike suppression of κ becomes more significant and the strongest (~16%) suppression is observed between 0.97 and 0.5 K. At even lower temperature of 0.36 K, the field-induced suppression of κ is getting weaker. The characteristic field, where the suppression completes or the plateau starts, is weakly dependent on the temperature, increases slightly from ~1 T at 0.36 K to ~2 T at 5 K. Somewhat intriguingly, this trend is the same as that found for the plateau in the superconducting Bi2212 samples.[17]

Figure 11 shows the temperature and field dependences of magnetization for an x=0.53 sample. It is clear that the magnetization data show rather conventional spin polarization of paramagnetic spins, which must be spins of Dy^{3+} ions. The characteristic field for spin polarization is about 2 T at 2 K and increases quickly with increasing temperature; this is rather different from the characteristic field of $\kappa(H)$ where the plateau appears. In this sense, the $\kappa(H)$ behavior does not appear to have direct relation with the Dy^{3+} free spins and their polarization, which is different from the case of the x=0.30 sample.

It is known that in rare-earth-or Y-doped Bi2212,[62] the antiferromagnetic (AF) order of Cu spins starts to be established when the superconductivity is completely suppressed and charge carriers are localized upon increasing the concentration of rare-earth ions. Therefore, one possible picture of the plateau behavior would be the magnon heat transport being affected by applied magnetic field: In a long-range-ordered spin system, the low-energy magnon excitations can act as heat carriers, but when the magnetic field drives the magnon states to sufficiently high energies, these magnons are no longer thermally excited and drop from heat conduction.[63] This causes a plateaulike $\kappa(H)$ behavior in magnetic systems, as has been observed in the yttrium iron garnet system.[63] However, one should keep in mind that the magnons are possibly involved only when the antiferromagnetic state has ungapped (or very weakly gapped) acoustic magnon branches, which needs to be confirmed by neutron measurements. More importantly, the very abrupt nature of the onset of the plateau observed at very low temperature [Fig. 10(a)], together with the rather weak temperature dependence of the magnetic field from which the plateau starts, seems to be difficult to be understood within this picture. Also, it is even not clear whether the plateau is only observable in nonsuperconducting Dy-doped samples. In other words, although the plateau feature is not present in the low-T $\kappa(H)$ curves for the a-axis Bi2212 samples in strong fields up to 16 T,[26] the present data could not clarify whether it is also absent in high magnetic fields for superconducting (and not AF ordered) Dy-Bi2212 with lower concentration of x. Detailed future study of this plateau phenomenon is clearly called for.

In passing, we note that the plateau behavior gradually weakens with increasing temperature, which is possibly due to the disappearance of the antiferromagnetic order. However, at even higher temperatures, the field dependence becomes again stronger above 18.5 K, as is shown in Fig. 7(d). The strongest H dependence is observed at 30 K, where the magnetic field of 6 T induces ~3% suppression of κ without any signature of saturation. This magnetic-field dependence of κ at high temperature is actually difficult to understand because if phonon transport is not affected by magnetic field, the only way to bring the field dependence of κ is through the electron channel. In this regard, the magnetoresistance measurements have shown that the in-plane resistivity changes by less than 0.1% in magnetic field ($\parallel c$) up to 14 T.[43] Such small change of resistivity cannot bring several percent change of thermal conductivity unless the Wiedemann–Franz law is strongly violated in this case.

IV. SUMMARY

Both the quasiparticle and phonon heat transport of $Bi_2Sr_2CaCu_2O_{8+\delta}$ are carefully studied for a broad doping range by using pure Bi2212 single crystals with tuned oxygen contents and $Bi_2Sr_2Ca_{1-x}Dy_xCu_2O_{8+\delta}$ single crystals with different Dy contents. It is found that the QP transport decreases with lowering doping by either decreasing oxygen content or increasing Dy content, whereas the phonon conductivity is enhanced in moderately Dy-doped samples where a phonon peak is observed at 10 K, which means that Dy^{3+} ions not only act as impurities or point defects but also stabilize the crystal lattice. The magnetic-field dependence of κ in pure Bi2212 at temperature above 5 K is mainly due to the QP scattering off vortices. In contrast, the Dy-doped samples (with $x=0.30$) present an additional diplike term of $\kappa(H)$ at low field, which is discussed to be related to the phonon scattering by free spins of Dy^{3+} ions. In nonsuperconducting Dy-Bi2212 samples with $x \simeq 0.50$, a plateaulike $\kappa(H)$ isotherm with characteristic field at 1–2 T is observed. We discuss that this plateau is possibly related to the magnon transport.

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