Thermal conductivity of polycrystalline $YBa_2Cu_4O_8$ from 10 to 300 K

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The thermal conductivity λ and electrical resistivity ρ were measured on two samples of the cuprate superconductor $YBa_2Cu_4O_8$ (Y124). The λ results pass through a maximum at about half of the superconducting transition temperature T_c. The peak λ value is much higher than the peak value for polycrystalline $YBa_2Cu_3O_{7-\delta}$, Y123, and is about equal to ab plane values for good melt processed Y123. In the normal state most of the λ is due to phonon transport λ_p and estimates of λ_p were obtained from the data by making a correction for electronic transport. Phonon-phonon and phonon-electron scattering limit the λ_p of both Y124 and Y123. In the two compounds, the phonon-phonon scattering is about equally strong, but electron-phonon scattering is considerably weaker in Y124. In the superconducting state, below the λ maximum at $T_c/2$, λ varies as $T^n(n\approx 1/2)$. This behavior has also been observed in Y123, but is not consistent with phonon scattering theory. The implications of this observation are discussed. [S0163-1829(98)08117-X]

INTRODUCTION

The high-temperature superconductor $YBa_2Cu_4O_8$ (Y124), was identified as a defect in bulk Y123,[1] and was later formed in thin films on $SrTiO_3$.[2–4] It was later shown[5–7] that the compound is stable over a wide range of temperature and oxygen pressures. Essentially phase-pure research samples have been synthesized in high-pressure oxygen at temperatures above 1000 °C and the results of many physical property studies have been published.[8–12] Data on the elastic modulus,[13] thermal expansion,[14] P_{O_2}-T stability range,[15] grain-boundary chemistry,[16] and weak-link behavior have been obtained in our laboratory. We also have collaborated on a study of oxygen diffusion in Y124.[17]

The results presented in this paper have both practical and theoretical significance. At present Bi-cuprate/Ag superconductors are being developed for $\sim$30 K conductor applications, but this superconductor is intrinsically unsuitable for liquid-nitrogen-cooled devices operating in magnetic fields.[18] This is not the case for Y124, since its intrinsic critical current values[19] are only slightly inferior to the results for Y123. Also, encouraging J_c results have recently been obtained on Ag-Y124 composite conductors.[20] Thermal conductivity λ is an important parameter for modeling conductor behavior during a quench.

The theoretical significance of the results lies in their relationship to the electron-phonon interaction and the nature of superconductivity in the cuprate superconductors. This is therefore an extension of many previous studies[21] of the thermal conductivity of the low-temperature metallic superconductors. The data have been compared to the results of an earlier study of the thermal conductivity of Y124,[22,23] λ results[24] for Y123, and a simplified theory of the phonon conductivity.

APPARATUS DESCRIPTION

The experimental device used to obtain the measurements was a modified version of the guarded absolute longitudinal heat-flow apparatus which has been operated at our laboratory for many years.[25] This method is well suited for measurements on small, high conductivity samples. If the sample conductivity is low, however, heat shunting errors can be very serious, especially at higher temperatures.[25] This is the case for the high-T_c compounds, where some of the reported thermal conductivity values[26] are too high and exhibit an anomalous temperature dependence. The method developed at ORNL has been used to make measurements on compounds whose thermal conductivities are lower than those of high-T_c oxides.[25]

The three principal modifications incorporated in the new apparatus were (1) silicon diodes were used for the temperature measurements instead of type E thermocouples, (2) flowing cold He gas was used as the heat sink, and (3) Ga-In-Sn eutectic alloy was extensively used for electrical and thermal contacts. The eutectic alloy was an important modification because it is very difficult to make electrical contacts to Y124. The two contacts on the sample were made by machining a 0.13 mm wide, 180° groove in the sample and wetting both the groove and a 0.13 mm diameter copper wire with eutectic alloy. Both ends of the wire were held in place with epoxy and the diode was attached to a platform mounted at one end of the wire.

The apparatus was tested by making a series of measurements on a standard, NIST SRM 1461.[27] The thermal conductivity of this austenitic stainless-steel alloy is higher than the values for high-T_c oxides, but lower thermal conductivity standards are not available. The experimental values tend to be a few percent low and scatter by about $\pm 2-3\%$. The error is probably associated with the determination of the effective spacing between the two sample temperature sen-

TABLE I. Characteristics of Y124 samples.

Sample number	Density (% theoretical)	T_c (K)	Measured electrical resistivity at 300 K (10^{-8} Ωm)
1	82.1	80	641
2	92.8	80	411

sors. The spacing between probes was typically 10–12 mm. A determinate error analysis indicated an uncertainty of about $\pm 3.5\%$.

SAMPLE CHARACTERISTICS

Thermal conductivity data were obtained on two Y124 samples and their characteristics are summarized in Table I. These samples were produced by sintering cold-pressed Y124 powder pellets in high-pressure oxygen at temperatures above 1000 °C. Details of materials synthesis have been reported previously.[15]

RESULTS

The thermal conductivity values for the two Y124 samples are shown in Fig. 1, which also shows the results of the earlier study.[22,23] The values for both samples were corrected for porosity by using Maxwell's equation.[28]

Four-probe dc resistivity data $\rho(T)$ for sample 2 are shown in Fig. 2. The resistivity of sample 1 was estimated from the data for sample 2 by assuming Mattheissen's rule applied and using the difference in room-temperature values for the two samples.

There are some qualitative comments which should be made. The λ reaches a maximum at about $T_c/2$ and at higher

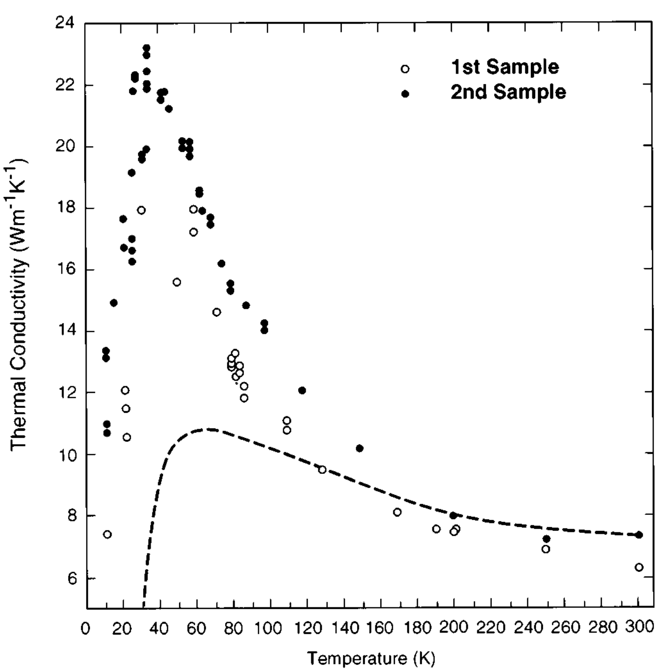

FIG. 1. Thermal conductivity values for two Y124 samples after correction to full density via Maxwell's equation. Data from Ref. 22 (dashed line).

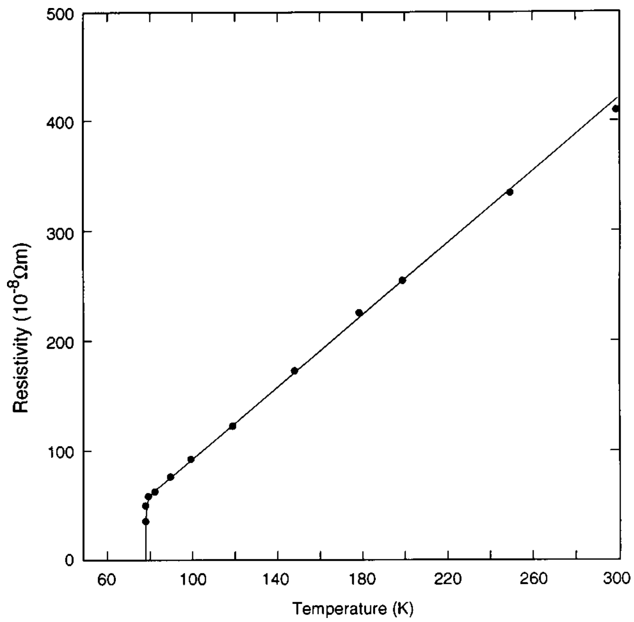

FIG. 2. Effect of temperature on the electrical resistivity of Y124 sample 2.

temperatures the values decrease. Yarbrough, Williams, and Shockley[24] obtained a similar result for Y123 and this variation is characteristic of a phonon conductor. Using the Sommerfeld-Lorenz number L_0 with the measured resistivity values yields an estimate of the electronic thermal conductivity. For the better sample (2) this estimate indicates that the electronic part of the thermal conductivity is about 30% of the measured room-temperature value. For Y123 the situation is similar.[24] This observation conflicts with some reports,[26] as discussed previously.

As in Y123, the λ does not undergo any drastic changes at T_c. For both Y124 and Y123 the conductivity maximum occurs at about $T_c/2$. It should be noted that the maximum λ value for randomly oriented Y124 is about a factor of 2 larger than the Y123 ab plane maximum reported by Hagen, Wang, and Ong.[29] The maximum λ value for Y124 is also only slightly smaller than the ab plane value[30] for good melt-processed Y123 samples.

As shown in Fig. 1, at 300 K the values of Andersson and co-workers[22,23] are about 10% higher than results obtained in this study. At lower temperatures the trend reverses and their peak thermal conductivity is only about 50% of the value obtained in this study. There is also a large difference between the room-temperature resistivities of our best sample (2) and the sample of Anderssen and Sundqvist.[22] The value for that sample was 630×10^{-8} Ωm.

DISCUSSION

In the normal state, Y124 appears to be a mixed conductor with significant λ contributions from both conduction electrons λ_e and phonons λ_p. The phonon contribution appears to be 3–4 times larger than λ_e and this discussion will emphasize a comparison of λ_p with theory.

The results of an earlier study[24] indicate that Y123 is also a mixed conductor. An analysis of the data also gives an indication that calculating λ_e from the measured ρ by using

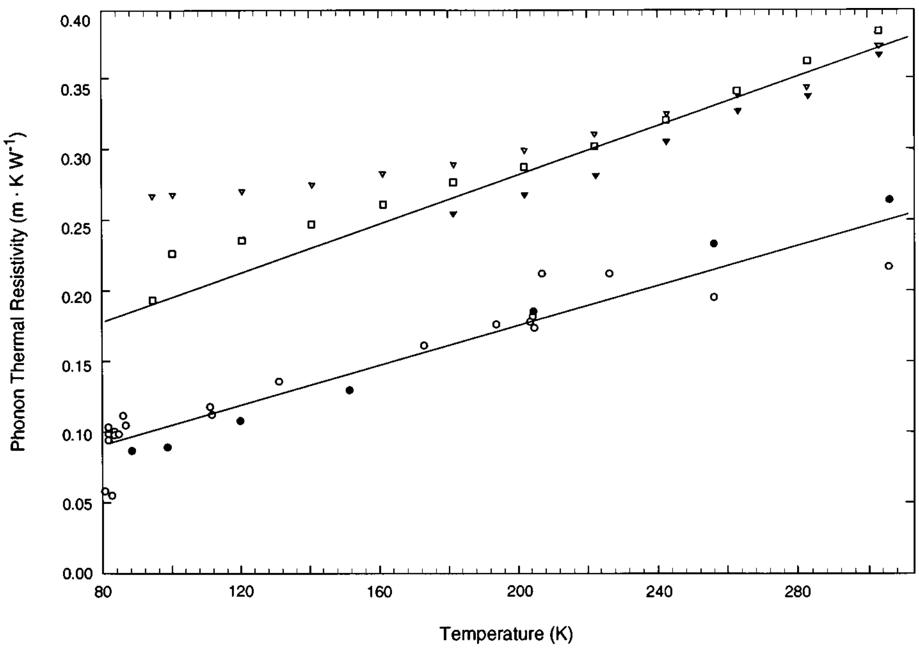

FIG. 3. Phonon resistivity values for two Y124 (○ sample 1, ● sample 2) samples and Y123 (▽,▼,□) samples from Ref. 24. The phonon resistivity values were derived from the measured values by correcting for porosity and electronic transport.

the Sommerfeld-Lorenz number L_0 may not be precisely correct. For Y123, small changes in the Ba content produce large ρ changes and measurements on three samples were analyzed[31] to yield experimental estimates of the Lorenz function. The results, $1.3L_0$ at 300 K and $1.5L_0$ at 200 K, require further verification but do suggest a similar uncertainty may apply to λ_e estimates for Y124.

The normal-state λ_p estimates for $T>T_c$ for Y124 and Y123 are shown in Fig. 3. The phonon thermal resistivity, λ_p^{-1} is determined by the net scattering rate:

$$\tau_p^{-1}=\tau_{ep}^{-1}+\tau_{pp}^{-1}+\tau_{ip}^{-1}+\tau_{bp}^{-1}. \quad (1)$$

Scattering by twin boundaries is neglected in Eq. (1) because Y124 does not undergo a phase change. The subscripts in Eq. (1) refer to electron, self, point-defect, and boundary scattering of the phonons. Point-defect and boundary scattering are both assumed to have negligible effects. Oxygen vacancies might cause phonon-point-defect scattering in both Y123 and Y124 but this seems less likely for Y124 because the oxygen content is essentially stoichiometric.[5,6] In fact, Fig. 4 shows evidence that phonon-point-defect scattering is also not very important in well oxygenated Y123. Low-temperature oxidation typically reduces the nonstoichiometry parameter δ to $\sim$0.05 and Fig. 4 (Ref. 26) shows that this degree of nonstoichiometry would produce an $\sim$8% reduction in λ at 100 K.

Boundary scattering has also been neglected in the analysis of the normal-state phonon conductivity. Calculations employing Callaway's model[32] indicate that boundary scattering is insignificant for $\sim$10 μm crystallites. This grain size was determined by light microscopy. At lower temperatures and for smaller crystallites the effect should be included. Data for Al_2O_3 showed[33] that a measurable reduction in λ could be obtained at ambient temperature and for grains as large as $\sim$1 μm. The Callaway formula does not predict this reduction and Klemens[34] has suggested that the effect is probably due to an enhanced contribution by long-wavelength longitudinal phonons.

The remaining two terms, electron-phonon and phonon-phonon determine the resistance to phonon transport in both compounds. As shown in Fig. 3, the resistive approximation gives a good description of both sets of data. Also, the linearity persists to fairly low temperatures which implies that the characteristic temperature appropriate for phonon transport is low.

The temperature dependence of the data measures the strength of phonon-phonon or self-scattering. The slopes of both sets of data are nearly equal, implying that the characteristic temperatures and Grüneisen constants for the two compounds are about equal. This is not surprising. Forcing numerical agreement with the Leibfried-Schlömann formula[35] requires a characteristic temperature of about 200

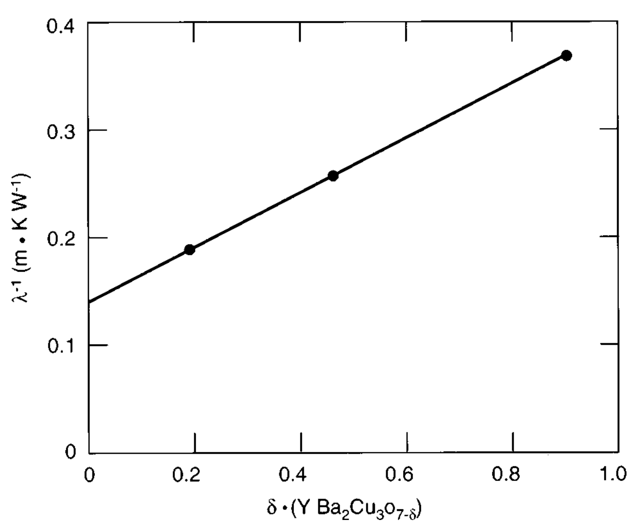

FIG. 4. Effect of oxygen vacancy concentration on the thermal conductivity of Y123 at 100 K. Results from Ref. 26.

K. For Y124, specific-heat measurements indicate a much higher Debye temperature, 550 K.[12] Andersson and Sundqvist[22] also reached this conclusion from a more elaborate analysis of their data.

The intercepts at $T=0$ for the two lines shown in Fig. 3 are presumed to represent the high-temperature limits of the resistance to phonon flow as determined by electron-phonon interactions W_∞. Again the result indicates a low characteristic temperature. For higher characteristic temperatures the $\lambda_p^{-1}-T$ values would deviate positively.

The variation of W_∞ for several metallic elements has been discussed by Butler and Williams[36] who showed that the variation of this scattering strength was about a factor of 10 larger than the interelement variation of the electrical resistivity and thermal conductivity. The electron-phonon thermal resistance values W_∞ for Y123 and Y124 are comparable to results obtained for metallic elements. The value for Y123 is about equal to W_∞ for Nb and the Y124 result is closer to the value for Mo.[36]

The theory shows that a minor part of the interelement variation arises from differences in phonon spectra and atomic sizes[36] and the major factors involved are the electron-phonon mass enhancement factor Λ and the density of states N_e:

$$W_\infty \propto \Lambda N_e \tag{2}$$

For Y123 and Y124 the phonon and atomic size factors should be essentially identical, but higher temperature (unenhanced) specific-heat measurements[11,12] indicate that N_e of Y124 is much lower than for Y123. The higher temperature data of Reeves *et al.* on Y123 (Ref. 37) are also consistent with electronic specific-heat coefficients for Y124 and Y123 obtained by Junod *et al.*[12] and Willis *et al.*[11] The mass enhancement factor for Y124 is unknown but a value can be estimated from the W_∞ values for Y124 and Y123 (Fig. 3), the average of the electronic specific-heat coefficients[11,12,37] and the Λ estimates for Y123.[37,38] Assuming Λ_{123} is about 2.5,[37,38] one obtains $\Lambda_{124}\approx 1.7$.

In the superconducting state, λ increases rapidly and reaches a maximum at about half the transition temperature. Maximum λ values for the three Y124 samples which have been studied vary considerably and Fig. 5 shows that there is a correlation between sample quality as measured by electrical resistivity and the maximum λ.

Hagen, Wang, and Ong[29] showed that the λ maximum at $\sim T_c/2$ is associated with the ab plane values of the thermal conductivity tensor and the origin of the maximum has been the subject of much discussion.[29,30,39–45] The two explanations which have been proposed are (1) enhancement of the phonon conductivity due to the disappearance of phonon scattering by electrons and (2) electronic energy transport at temperatures well below T_c. At present the electronic transport mechanism, which is based on d-wave superconductivity,[40,41] appears to be more generally accepted.

Although the data of Andersson and co-workers[22,23] on Y124 were analyzed on the dominant phonon model, our results at lower temperatures suggest that this picture is too simple. These data, which are shown in Fig. 6, vary roughly as $T^n(n\approx 1/2)$ and Cohn *et al.*[30] have reported that ab plane values for Y123 show similar behavior. Unfortunately no single-phonon scattering process is expected to vary with temperature in this fashion.[21,32] Mixing impurity and boundary scattering might produce the overall approximate $T^n(n\approx 1/2)$ variation; however, the λ values indicate strong scattering, while both impurity and boundary scattering are weak at low temperatures. Impurity scattering is particularly unlikely because there is little evidence that oxygen vacancies are present in Y124.

On the other hand, phonon transport must have some role because it dominates the normal-state properties, and the transition to the superconducting state should not add any

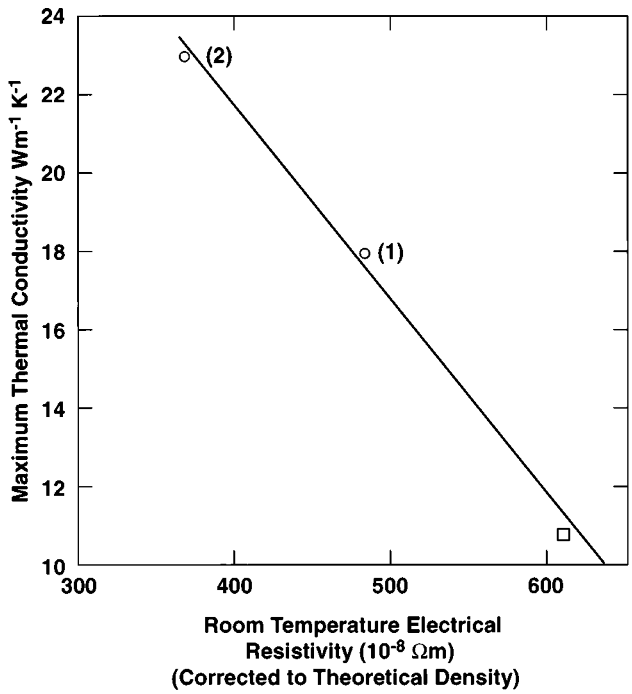

FIG. 5. Maximum thermal conductivity of Y124 is related to electrical resistivity. (□) Ref. 22.

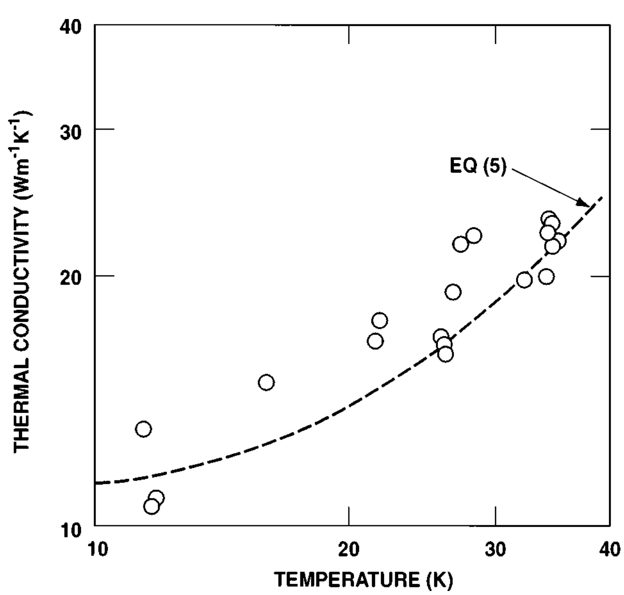

FIG. 6. Low-temperature thermal conductivity values for sample 2 vary as $T^n(n\approx 1/2)$ as shown on this log-log plot.

strong new scattering processes. An estimate based on L_0 indicates that the phonon component is about 80% of λ at 80 K, and both phonon-phonon and phonon-impurity scattering are expected to weaken as the temperature is lowered. It thus seems possible that although the maximum has an electronic origin, Y124 remains a mixed conductor in the superconducting state.

Indeed, recent experiments have revealed evidence for a finite density of electrons that, well below T_c, exist as ''normal fluid'' in a two-fluid picture. The specific evidence comes from studies of the London penetration depth $\Delta(T)$. Standard London theory[46] provides that

$$1/\Delta^2 = n(4\pi e^2/m^* c^2), \tag{3}$$

where $n(T)$ is the density of superconducting charge carriers at temperature T and m^* is their effective mass. Thus the normal fluid fraction is given by $[1-(n(T)/n(0)]$, which is unity at T_c. Far below T_c, this component vanishes exponentially for any pairing scheme with a finite gap at the Fermi surface, as with BCS theory. On the other hand, superconducting pairing schemes that are gapless or that have nodes in the energy gap, such as d-wave pairing, permit easy excitation of a normal fluid component. This means that $1/\Delta^2$ has a power-law dependence at low temperatures, rather than an exponentially activated dependence. In fact, Bonn *et al.*[47] deduced a power-law temperature dependence $n(T) \sim (1-t^2)$ covering the entire range $t = T/T_c < 1$ for single crystals of $YBa_2Cu_3O_{6.95}$. Other Y123 materials with different purities differ in detail,[48,49] but still give evidence for power-law behavior. Finally, a recent study of Hg cuprate $HgBa_2CuO_{4+\delta}$ superconductors[50] found that $1/\Delta^2 \sim n$ varied almost linearly with T down to at least $t = 0.3$; this implies that the normal fluid component also has a linear temperature variation over a large range. Taken together, these studies suggest that normal state, i.e., unpaired charge carriers, may be present in significant numbers below T_c and have a pronounced impact on the properties of the Y124 materials being investigated.

To test this hypothesis, the lower temperature data were compared to two simple models. Both are based on the assumption that unpaired charge carriers are present at temperatures well below T_c. The concentration of the unpaired carriers is assumed to be directly proportional to T below T_c, and the carriers can both transport energy and scatter phonons. The models assume metallic behavior as modified by a temperature-dependent carrier concentration.

For strong scattering—alloys—the expected λ variation is[51]

$$\lambda = \lambda_p + \lambda_e = a_1 T + \frac{L_0 T}{\rho} \ (\rho = \text{const}). \tag{4}$$

The temperature dependence of the carrier concentration modifies Eq. (4):

$$\lambda = \lambda_p + \lambda_e = a_2 + bT^2. \tag{5}$$

Similarly, for weak scattering—a pure metal—the expected behavior is[51–53]

$$\lambda = \lambda_p + \lambda_e = c_1 T^2 + d_1 T^{-2}, \tag{6}$$

which is modified to

$$\lambda = \lambda_p + \lambda_e = c_2 T + d_2 / T. \tag{7}$$

Both Eqs. (5) and (7) can generate an approximately $T^n (n \approx 1/2) - \lambda$ variation between 10 and 35 K and Eq. (5) is shown in Fig. 6. The strong scattering (5) approximation predicts roughly equal electron and phonon contributions. More precise λ data are required to test the adequacy of this approach.

Finally, it should be mentioned that these simple models suggest an experiment which could demonstrate the relative roles of electrons and phonons for Y123. Jin *et al.*[54] have shown that several rare-earth elements can partially replace yttrium in Y123. This replacement, which does not change T_c or the normal-state resistivity, should reduce the phonon component and make it possible to experimentally separate λ_e and λ_p.[36]

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