Universal Heat Conduction in YBa$_2$Cu$_3$O$_{6.9}$

Louis Taillefer, Benoît Lussier$^*$ and Robert Gagnon

Department of Physics, McGill University, Montréal, Québec, Canada H3A 2T8

Kamran Behnia and Hervé Aubin

Laboratoire de Physique des Solides (CNRS), Université Paris-Sud, 91405 Orsay, France

The thermal conductivity of YBa$_2$Cu$_3$O$_{6.9}$ was measured at low temperatures in untwinned single crystals with concentrations of Zn impurities from 0 to 3% of Cu. A linear term $\kappa_0/T = 0.19$ mW K$^{-2}$ cm$^{-1}$ is clearly resolved as $T \rightarrow 0$, and found to be virtually independent of Zn concentration. The existence of this residual normal fluid strongly validates the basic theory of transport in unconventional superconductors. Moreover, the observed universal behavior is in quantitative agreement with calculations for a gap function of d-wave symmetry.

PACS numbers: 74.25.Fy, 74.72.Bk

The theory of quasiparticle transport in unconventional superconductors, developed over the last decade, has remained largely untested. A novel feature that arises when the superconducting gap function has nodes for certain crystal directions is the existence of quasiparticles at $T = 0$. This residual normal fluid is a consequence of impurity scattering, even for low concentrations of nonmagnetic impurities (see [1,2], and references therein). Its presence, which should dominate the conduction of magnetic impurities (see [1,2], and references therein). When the superconducting gap function has nodes for certain pairing states, with appropriate gap topology and symmetry, an appealing phenomenon is predicted to occur: quasiparticle transport should be independent of scattering rate as $T \rightarrow 0$. This universal limit, first pointed out by Lee [3] for the case of a d-wave gap in two dimensions, is the result of a compensation between the growth in normal fluid density with increasing impurity concentration and the concomitant reduction in mean free path.

In this Letter, we report the first observation of universal transport in a superconductor. Our study of heat conduction in the high-$T_C$ cuprate YBa$_2$Cu$_3$O$_{6.9}$ provides a solid validation of the basic theory of transport in unconventional superconductors and insight into the nature of impurity scattering in the cuprates. It also supports strongly an identification of the gap function as having d-wave symmetry.

The thermal conductivity $\kappa(T)$ was measured between 0.05 and 1 K, for a current along the a axis of five single crystals: four untwinned crystals of YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_{6.9}$ and one crystal of YBa$_2$Cu$_3$O$_{6.0}$. The latter was obtained by full deoxygenation via annealing at 800 °C in helium gas for 64 h; it is insulating, with $\rho_a(100$ K) = 42.7 $\Omega$ m. $x$ is the nominal concentration of Zn, achieved by mixing in ZnO powder at the start of the growth process in the atomic ratio Zn:Cu=1.5$x$ : 1 $-$ x, for $x$ = 0, 0.006, 0.02 and 0.03. The experimental technique and the sample preparation are described elsewhere [4,5]. The resistive $T_C$ is given in Table I. The uncertainty on the geometric factor is at most $\pm$ 10%, 10%, 20%, 5% and 10% for the $x$ = 0 (“pure”), 0.6%, 2%, 3% and deoxygenated (“deoxygen”) samples, respectively.

The a-axis resistivity is linear in temperature above 130 K [4], and a fit to $A + BT$ yields the values in Table I. Zn substitution has two effects: it reduces $T_c$ and it increases $\alpha$. At low concentration, both effects are linear, and $dT_c/d\alpha = \lambda $ 0.5 K / $\mu\Omega$ cm, in agreement with data on twinned crystals (e.g. [4]). Concentrations of Zn from 0 to 3% correspond to a large range of scattering rates, but to a modest level of pair-breaking: adding 3% Zn suppresses $T_c$ by only 20%. Given that the inelastic scattering term $B$ is independent of $x$, the impurity scattering rate $\Gamma = 1/(2\tau_0)$ may be estimated via the residual resistivity $\rho_0 = m^*/ne^2\tau_0$:

$$\Gamma(\omega) = (\omega^2/8\pi)[\rho_0(x = 0) + A(x) - A(0)]$$

The sample data on twinned crystals (e.g. [6]). Concentrations of Zn from 0 to 3% correspond to a large range of scattering rates, but to a modest level of pair-breaking: adding 3% Zn suppresses $T_c$ by only 20%. Given that the inelastic scattering term $B$ is independent of $x$, the impurity scattering rate $\Gamma = 1/(2\tau_0)$ may be estimated via the residual resistivity $\rho_0 = m^*/ne^2\tau_0$:

$$\Gamma(\omega) = (\omega^2/8\pi)[\rho_0(x = 0) + A(x) - A(0)]$$

| $x$ (%) | $T_c$ (K) | $\rho_a$ (\$\mu\Omega$ cm) | $\rho_a$ (\$\mu\Omega$ cm$^{-1}$) | $\Gamma(\omega)/T_c$ (\$h/k_B$) |
|--------|----------|-----------------------------|-------------------------------|--------------------------------|
| pure   | 93.6     | -14.3                       | 0.95                          | < 0.014                        |
| 0.6    | 89.2     | -6.0                        | 1.00                          | 0.13                           |
| 2      | 80.0     | 12.9                        | 0.94                          | 0.4                            |
| 3      | 74.6     | 22.9                        | 1.07                          | 0.54                           |
TABLE II. Parameters used in fitting $\kappa/T$ to $a + bT^2$, where $a = \kappa_0/T$ is the electronic residual linear term and $b = \kappa_{ph}/T^3$ is the asymptotic phonon $T^3$ term. $\bar{\omega}$ is the mean sample width and $\Lambda_0$ is calculated from Eq. (2) using $\beta = 0.3 - 0.4$ mJ/K$^2$ mole and $\langle v_{ph} \rangle = 4000$ m/s.

| sample | $\bar{\omega}$ (µm) | $\kappa_0/T$ (mW K$^{-1}$ cm) | $\kappa_{ph}/T^3$ (mW K$^{-4}$ cm$^{-1}$) | $\sqrt{\pi}\Lambda_0/2\bar{\omega}$ |
|--------|-----------------|------------------|------------------|------------------|
| pure   | 252             | 0.19 ± 0.03      | 17 ± 2           | 1.2 - 1.8        |
| 0.6%   | 242             | 0.17 ± 0.04      | 11 ± 2           | 0.8 - 1.0        |
| 2%     | 177             | 0.25 ± 0.07      | 7 ± 3            | 0.7 - 0.9        |
| 3%     | 238             | 0.20 ± 0.05      | 8 ± 3            | 0.6 - 0.8        |
| deox   | 315             | 0.00 ± 0.01      | 14 ± 2           | 0.8 - 1.0        |
Having established the existence of a residual normal fluid in YBa$_2$Cu$_3$O$_{6.9}$, the next question is that of universality. This is addressed by looking at concentrations of Zn such that $\Gamma$ ranges from $<0.014$ up to $0.54\ T_c$. The thermal conductivity of YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_{6.9}$ is shown in Fig. 2, where it is apparent that $\kappa$ is unaffected by the variation in $\Gamma$ at $\approx 0.1$ K, where the heat is carried predominantly by quasiparticles (cf. Fig. 1). In other words, transport by the residual normal fluid is universal.

The $T \to 0$ limit of $\kappa/T$ is obtained from a fit to $a + bT^2$ limited to $T < 150$ mK, as applied earlier, which yields the values for $a = \kappa_0/T$ and $b$ listed in Table II. Note that the ratio $\Lambda_0/(2\tilde{w}/\sqrt{\pi}) \approx 1$ for all crystals, proving that the asymptotic phonon regime was reached in all cases. (The somewhat larger ratio for the pure sample is intriguing – further work is needed to elucidate this.)

As seen from a plot of $\kappa_0/T$ versus $\Gamma$, shown in Fig. 3, these values are consistent with a universal linear term of $0.19$ mW K$^{-2}$ cm$^{-1}$. Note, however, that the error bars on the values of $a$ and $b$ in Table II are fairly large, because they combine uncertainties on the geometric factors (largest for the rather short 2% sample) and on the fit, which is limited to a small temperature range (smallest for the 3% sample). One way of eliminating the uncertainty on the geometric factor is to use the resistivity data obtained with the same contacts. Indeed, by fixing $B = 1.03$ $\mu$T cm$^{-1}$ for all samples, thereby imposing the reasonable constraint that the inelastic scattering is not affected by small levels of Zn, one can correct $\kappa_0/T$ by multiplying it by $B(x)/1.03$. This yields the following corrected values: $0.17 \pm 0.01, 0.17 \pm 0.02, 0.23 \pm 0.02$ and $0.21 \pm 0.04$ mW K$^{-2}$ cm$^{-1}$, for $x = 0, 0.6, 2$ and 3%, respectively. These are plotted in the inset of Fig. 3 versus

the similarly corrected $\Gamma_\rho$. The corrected plot with its smaller error bars no longer allows for a constant linear term: there is a small but definite upward slope, with a minimum growth of 30% over the range of $\Gamma_\rho$ and a maximum growth of 55%. From this we conclude that while the residual linear term is universal, in the sense that a 10-fold increase in $\Gamma$ (from $0.014$ to $0.13\ T_c$ in going from $x = 0$ to 0.6%) leaves $\kappa_0/T$ unchanged, at larger $\Gamma$ there is a slight increase, reaching approximately 40% at $\Gamma/T_c \approx 0.6$.
A quantitative comparison with the theory reinforces this conclusion. In the simplified case of the standard d-wave gap $\Delta \cos(2\phi)$, $S = 2\Delta_0$. Using available estimates of $h\omega_p$ and the gap maximum $\Delta_0$, respectively equal to 1.3 eV [8] and 20 meV [19], one gets

$$\frac{\kappa_{00}}{T} = 0.09 \text{ mWK}^{-2}\text{cm}^{-1}$$  \hspace{1cm} (4)

which is remarkably close to the measured value of 0.19 mWK$^{-2}$ cm$^{-1}$. Given that the real gap will have more structure than a simple $\cos(2\phi)$ dependence, the factor 2 discrepancy suggests that it actually rises from the node half as fast as in the simple model. This in no way detracts from the conclusion that a (generalized) d-wave gap is in excellent quantitative agreement with the universal heat conduction observed in YBa$_2$Cu$_3$O$_{6.9}$.

The second point to consider in a comparison with the theory is the fact that universality is only achieved when $h\gamma << \Delta_0$, where $h\gamma$ is the bandwidth of impurity bound states responsible for the zero-energy excitations [15]. The bandwidth grows with $\Gamma$ in a way which depends very strongly on the scattering phase shift $\delta_0$. It is largest in the limit of unitarity scattering, $\delta_0 = \pi/2$, where $h\gamma \sim \sqrt{\pi \Delta_0 \hbar \Gamma}/2$ [5]. For the pure and 3% samples, with $\Gamma_\rho/T_{c0} = 0.014$ and 0.54, this gives $h\gamma/\Delta_0 \simeq 0.1$ and 0.6, respectively (for $\Delta_0 \simeq 20$ meV = 2.5 $k_B T_{c0}$). Thus we expect deviations from universality for the samples with high Zn doping. Quantitatively, the dependence of $\kappa_{00}/T$ on $\Gamma$ was calculated by Sun and Maki [14], who find a monotonic increase, which gets to be a factor 1.9 at $\Gamma/T_{c0} = 0.54$ (see also Ref. 17). Such a large increase is incompatible with the data (see Fig. 3). On the other hand, a 40% growth in the residual linear term, consistent with the data, would agree with the calculation if $\Gamma \simeq \Gamma_\rho/2$, namely 0.3 $T_{c0}$ for the 3% sample instead of 0.54 $T_{c0}$. Interestingly, this is the $\Gamma$ one deduces self-consistently from the theory [2] based on the measured $T_c$ suppression. Note, however, that accounting for a smaller $\Gamma$ in terms of a smaller “effective” $\omega_0^2$ in Eq. (1) leads to an even smaller $\kappa_{00}/T$ from Eq. (3).

These minor discrepancies notwithstanding, one of the main implications of the good agreement with the theory is that impurity scattering in the cuprates is well-described by a phase shift very close to $\pi/2$ [15], something which has been assumed often but rarely verified. Nonetheless, a proper interpretation of the data should include the possibility of a small departure from the unitarity limit [17]. This would lower $\gamma$, making it easier to satisfy the condition $h\gamma << \Delta_0$, and possibly to account for the weak variation in $\kappa_{00}/T$.

The present results have implications for other properties of YBa$_2$Cu$_3$O$_{6.9}$, such as specific heat [11,12] and microwave conductivity [2]. It has not yet been possible to probe the residual normal fluid reliably via these properties, but the upper bounds imposed by the data so far are consistent with the behavior predicted on the basis of the thermal conductivity data reported here.

In summary, we presented the first observation of universality in the transport properties of a superconductor. A residual linear term $\kappa_0/T = 0.19$ mWK$^{-2}$ cm$^{-1}$ is clearly resolved in the thermal conductivity of YBa$_2$Cu$_3$O$_{6.9}$ and attributed to electronic carriers. The observation of this residual normal fluid is a powerful validation of the basic theory of impurity scattering in unconventional superconductors. The fact that $\kappa_0/T$ is universal, i.e. virtually unaffected by changes in the impurity scattering rate, strongly confirms the gap has having d-wave symmetry. However, from the magnitude of $\kappa_0/T$, it appears that the gap rises more slowly at the nodes than described by the standard function $\Delta \cos(2\phi)$ with $\Delta_0 = 2.5 k_B T_{c0}$.

We thank John Berlinsky, Brett Ellman, Matthias Graf, Peter Hirschfeld and Catherine Kallin for stimulating discussions. This work was funded by NSERC of Canada, FCAR of Québec and the Canadian Institute for Advanced Research. L.T. acknowledges the support of the Alfred P. Sloan Foundation.

---

* Present address: CRTBT-CNRS, 38042 Grenoble, France.

[1] P. Hirschfeld et al., Solid State Commun. 59, 111 (1986).
[2] S. Schmitt-Rink et al., Phys. Rev. Lett. 57, 2575 (1986).
[3] P.A. Lee, Phys. Rev. Lett. 71, 1887 (1993).
[4] Robert Gagnon et al., Phys. Rev. Lett. 78, 1976 (1997).
[5] Robert Gagnon et al., Phys. Rev. B 50, 3458 (1994).
[6] Y. Fukuzumi et al., Phys. Rev. Lett. 76, 684 (1996).
[7] D.A. Bonn et al., Phys. Rev. B 50, 4051 (1994).
[8] D.N. Basov et al., Phys. Rev. Lett. 74, 598 (1995).
[9] P.D. Thacher, Phys. Rev. 156, 975 (1967).
[10] S. Shamoto et al., Phys. Rev. B 48, 13 817 (1993).
[11] R.A. Fisher et al., Physica C 252, 237 (1995).
[12] Kathryn A. Moler et al., Phys. Rev. B 55, 3954 (1997).
[13] J. Dominec, Supercond. Sci. Tech. 5, 153 (1993).
[14] Y. Sun and K. Maki, Europhys. Lett. 32, 355 (1995).
[15] M.J. Graf et al., Phys. Rev. B 53, 15147 (1996).
[16] M.R. Norman and P.J. Hirschfeld, Phys. Rev. B 53, 5706 (1996).
[17] P.J. Hirschfeld and W.O. Putikka, Phys. Rev. Lett. 77, 3909 (1996).
[18] Citrad Uher, J. Supercond. 3, 337 (1990).
[19] I. Maggio-Aprile et al., Phys. Rev. Lett. 75, 2754 (1995).
[20] Y. Sun and K. Maki, Phys. Rev. B 51, 6059 (1995).
[21] Kuan Zhang et al., Phys. Rev. Lett. 73, 2484 (1994).