Theory of Thermal Conductivity in $YBa_2Cu_3O_{7-\delta}$

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We calculate the electronic thermal conductivity in a *d*-wave superconductor, including both the effect of impurity scattering and inelastic scattering by antiferromagnetic spin fluctuations. We argue that phonons dominate heat transport near T_c, but that electrons are responsible for most of the peak observed in clean samples, The peak position is predicted to vary nonmonotonically with disorder, in good agreement with experiments on $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$. [S0031-9007(96)01535-9]

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Analysis of transport experiments in the superconducting state of the high-temperature cuprate superconductors has already provided the most compelling evidence for electronic pairing in these materials. The collapse of the quasiparticle relaxation rate below T_c, as observed in optical [1,2] and microwave [3] measurements, is not observed in classic superconductors, and is most naturally interpreted in terms of a gapping of the spectral density of electronic excitations responsible for inelastic scattering just above T_c. This collapse is now understood to be responsible, e.g., for the peak at intermediate temperatures in the microwave conductivity of $YBa_2Cu_2O_{6.95}$ [3].

Thermal conductivity measurements provide information on order parameter symmetry and quasiparticle relaxation and have the advantage that they are bulk probes not subject to extrinsic surface effects which have hampered the interpretation of the low-*T* microwave conductivity [3]. They have the disadvantage that the electronic contribution to the heat current must be separated from the phononic one. As pointed out by Yu *et al.* [4], the similarity between the microwave conductivity peak and measurements of the thermal conductivity $\kappa(T)$ in $YBa_2Cu_3O_{7-\delta}$ single crystals suggests that at least part of the thermal conductivity peak should be due to the electronic thermal conductivity $\kappa_{\rm el}(T)$, in contrast to earlier analyses of this peak in terms of a phonon conductivity $\kappa_{\rm ph}(T)$ alone [5,6]. In the work of Yu *et al.,* the phononic mean free path $\ell_{\rm ph}$ is assumed to vary only weakly with $T < T_c$, whereas in the Peacor *et al.* approach [5], $\ell_{\rm ph}$ is assumed to be dominated by phonon-electron relaxation, leading via the pair correlations in the electronic system to an exponential behavior below T_c. In Ref. [5] it is assumed that $\kappa_{\rm ph} \gg \kappa_{\rm el}$ over the entire temperature range, whereas Yu *et al.* deduce $\kappa_{\rm ph}(T_c) \simeq 2 - 3\kappa_{\rm el}(T_c)$ using the measured $\sigma_1(T_c)$ on similar quality crystals, and assuming the Wiedemann-Franz law $\kappa_{\rm el} = L_0 T \sigma_1$ with the free electron Lorenz number L_0.

In this paper, we adopt a theoretical model of electronic transport in a *d*-wave superconductor limited by impurity and spin fluctuation scattering which has proven successful in describing many of the systematics of microwave measurements, and apply it to calculate the electronic thermal conductivity. We analyze experiments on Zn doped $YBa_2Cu_3O_{7-\delta}$ to argue that (a) phonons do in fact dominate heat conduction at T_c; (b) a peak in $\kappa_{\rm ph}$ does indeed occur at about 20–25 K; (c) electronic conduction does nevertheless provide most of the peak in clean samples; and (d) the temperature at which the peak in $\kappa(T)$ occurs may vary nonmonotonically with disorder. Conclusions (a)–(c) agree with a recent thermal Hall conduction measurement and analysis by Krishana *et al.* [7]. Conclusion (d) agrees well with recent data on $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ over a wide range of Zn concentrations [8]. We further focus on the very low-temperature behavior of the thermal conductivity in the *d*-wave model, discussed recently in considerable detail by Graf *et al.* [9]. In particular, we analyze the disorder and phase shift dependence of the "universal" linear-*T* term in κ predicted recently by several groups [9–11]. Since at very low temperatures the phonon mean free path has saturated, this contribution is a direct reflection of electronic correlations and relaxation.

Electronic thermal conductivity.—The electronic thermal conductivity κ for an unconventional superconductor [12–15] is evaluated using a Kubo formula for the heat-current response as in the original treatment for an *s*-wave superconductor [16]. In this Letter we particularly wish to study the $d_{x^2-y^2}$ pair state thought to provide a good description of optimally doped $YBa_2Cu_3O_{7-\delta}$ [17,18]. For simplicity we work in what follows with the approximate order parameter $\Delta_k = \Delta_0 \cos(2\phi)$ over a circular Fermi surface to describe *ab* plane transport. The impurity-averaged matrix (Nambu) electron propagator in such a state is given by

$$\mathbf{g}(\mathbf{k},\omega) = \frac{\tilde{\omega}\boldsymbol{\tau}^0 + \xi_{\mathbf{k}}\boldsymbol{\tau}^3 + i\Delta_{\mathbf{k}}\boldsymbol{\sigma}^2\boldsymbol{\tau}^1}{\tilde{\omega}^2 - \xi_{\mathbf{k}}^2 - |\Delta_{\mathbf{k}}|^2}, \qquad (1)$$

where $\boldsymbol{\sigma}^i$ and $\boldsymbol{\tau}^i$ are the Pauli matrices in spin and particle-hole space, respectively. Here we have already exploited the assumed particle-hole symmetry of the normal state, as well as the symmetries of the gap functions which lead to vanishing renormalizations for both the order parameter and the single-particle energies. In this case, only self-energy contributions to the frequency ω, namely, $\tilde{\omega} = \omega - \Sigma_0$ need to be included [14]. The self-energy Σ_0 due to the elastic impurity scattering is treated in a self-consistent t-matrix approximation and is given by $\Sigma_0 = \Gamma G_0/(c^2 - G_0^2)$, where $\Gamma = n_i n/\pi N_0$ is the normal state unitarity limit scattering rate depending on the concentration of defects n_i, the electron density n, and the density of states at the Fermi level N_0. The quantity $c \equiv \cot \delta_0$ parameterizes the scattering strength of an individual impurity through the s-wave phase shift δ_0. In this work we consider only near-unitarity limit scattering $c \simeq 0$ since it is clear that weak scattering will lead to a weak temperature dependence inconsistent with experiment for the states in question. The integrated propagator is $G_0 = (1/2\pi N_0) \sum_{\mathbf{k}} \mathrm{Tr}\{\boldsymbol{\tau}^0 \mathbf{g}(\mathbf{k}, \omega)\}$. The equation for the self-energies is then solved self-consistently together with the gap equation $\Delta_{\mathbf{k}} = -T \sum_{\omega_n} \sum_{\mathbf{k}'} V_{\mathbf{k}\mathbf{k}'} (1/2)\, \mathrm{Tr}\{\boldsymbol{\tau}^1 \mathbf{g}(\mathbf{k}', \omega_n)\}$.

The above approximation is insufficient to describe transport at temperatures close to T_c, where inelastic scattering is known to dominate. As in Refs. [19,20], we adopt a model of scattering by antiferromagnetic spin fluctuations based on an RPA treatment of the Hubbard model with parameters chosen to reproduce normal state NMR and resistivity data in $YBa_2Cu_3O_{7-\delta}$ [21]. The relaxation rate due to spin fluctuations $1/\tau_{\rm in}$ is quasilinear in temperature above T_c and falls as $\sim T^3$ in the superconducting state due to (a) a crossover to Fermi liquid behavior below a spin fluctuation scale and (b) an additional factor of T due to the restriction of relevant quasiparticle momenta to the vicinity of the d-wave order parameter nodes [21]. To include inelastic scattering in the model in a crude way, we make the replacement $\Sigma_0 \rightarrow \Sigma_0 - i/2\tau_{\rm in}$. Although we have adopted a particular microscopic model, we emphasize that the two features (a) and (b) noted may be common to many models of inelastic relaxation, which will then lead to qualitatively similar results.

The bare heat current response is now given by a convolution of the Green's function g with itself at zero external frequency and wave vector weighted with the bare heat current vertex $\omega \mathbf{v}_{F\mathbf{k}} \boldsymbol{\tau}^3$ [16]. Impurity scattering vertex corrections to current-current correlation functions at $q = 0$ have been shown to vanish identically for even parity states ($\Delta_{\mathbf{k}} = \Delta_{-\mathbf{k}}$) [14]. For the diagonal thermal conductivity tensor one obtains

$$\frac{\kappa_{\rm el}^{i}(T)/T}{\kappa_{\rm el}^{N,i}(T_c)/T_c} = \frac{6}{\pi^2} \int_0^\infty d\omega \left(\frac{\omega}{T}\right)^2 \left(\frac{-\partial f}{\partial \omega}\right) K_i(\omega, T)\,, \tag{2}$$

$$K_i(\omega, T) = \frac{\Gamma_{\rm tot}(T_c)}{\tilde{\omega}' \tilde{\omega}''} \mathrm{Re} \left\langle \hat{\mathbf{k}}_i^2 \cdot \frac{\tilde{\omega}^2 + |\tilde{\omega}|^2 - 2|\Delta_{\mathbf{k}}|^2}{\sqrt{\tilde{\omega}^2 - |\Delta_{\mathbf{k}}|^2}} \right\rangle_{\hat{\mathbf{k}}}, \tag{3}$$

where $\tilde{\omega}'$ and $\tilde{\omega}''$ are the real and imaginary parts of $\tilde{\omega}$, f is the Fermi function, and $\Gamma_{\rm tot} \equiv \Gamma + 1/2\tau_{\rm in}$ is the total quasiparticle scattering rate. We have numerically evaluated Eqs. (2) and (3), and show results in Fig. 1. In the clean limit, the combination of the relaxation rate collapsing with decreasing temperature due to gapping of the spin fluctuation spectral density and the rapidly decreasing number of quasiparticles at low T leads to a peak in the thermal conductivity very similar to that found for the electrical conductivity [3,19]. As impurities are added, the collapse of the inelastic scattering rate is cut off at progressively higher and higher energy scales, such that the peak moves to higher temperatures and simultaneously weakens. A preliminary report on these findings is contained in Ref. [22].

Phonon thermal conductivity.—Were the phonon and electron thermal conductivities comparable at T_c, the electronic peak in clean samples would, according to Fig. 1, be very large, leading to ratios $\kappa(T_{\rm peak})/\kappa(T_c)$ much larger than the observed range of about 1.8–2.4 [4,7,8,22]. We therefore expect on theoretical grounds that the phononic conductivity at T_c is several times larger than the electronic conductivity [22]. This conclusion was reached independently by Krishana *et al.* [7], who performed a semiclassical Boltzmann type analysis of their measurements of the diagonal and off-diagonal components of the tensor κ_{ij} in a magnetic field, assuming as the origin of the off-diagonal terms skew-scattering of quasiparticles off the vortex lattice. The diagonal phonon conductivity $\kappa_{\rm ph}$ was then extracted from the data under the assumption that only electrons were skew scattered. Here we adopt the $\kappa_{\rm ph}$ as shown in Fig. 2 following the analysis of Ref. [7], using a value of the transverse scattering cross section such that $\kappa(T_{\rm peak}) - \kappa(T_c)$ is primarily of electronic origin [7]. We use the form of $\kappa_{\rm ph}$ shown for all calculations at any impurity concentrations, thereby neglecting the weak effect of point defects on

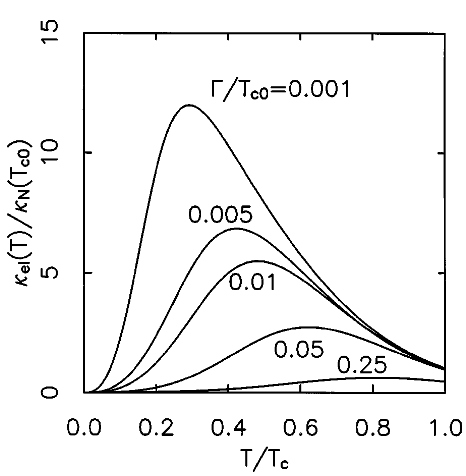

FIG. 1. Normalized electronic thermal conductivity κ/κ_N for normalized impurity scattering rate $\Gamma/T_{c0} = 0.001$, 0.005, 0.01, 0.05, and 0.25, $\Delta_0/T_c = 3$ and $1/\tau_{\rm in}(T_c) = T_c$.

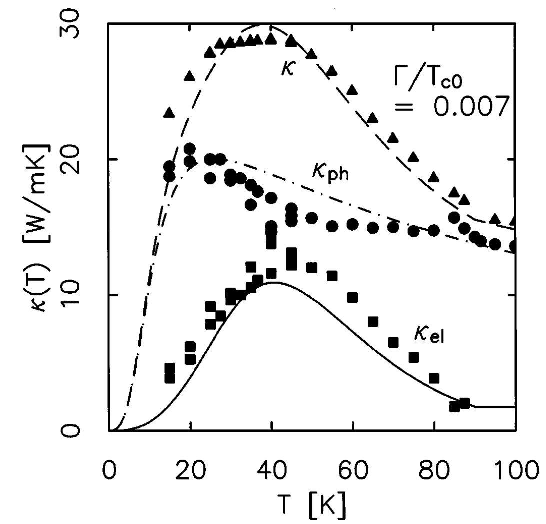

FIG. 2. Solid line: electronic thermal conductivity κ_{el}; dash-dotted line: phononic thermal conductivity κ_{ph}; dashed line: κ vs T/T_c for $\Gamma/T_{c0} = 0.007$. Data are from Ref. [7].

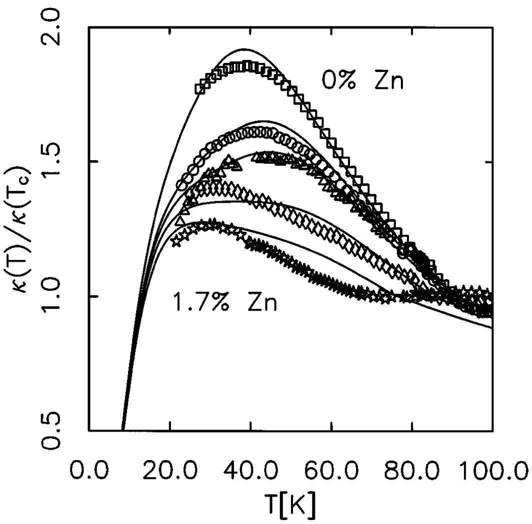

FIG. 3. Total thermal conductivity κ vs T[K]. Symbols are data from Ref. [8], Zn concentration 0.0%, 0.1%, 0.2%, 0.7%, and 1.7%. Solid lines: theoretical κ for $\Gamma/T_{c0} = 0.007 + 0.12(\mathrm{Zn})$.

the long wavelength phonon mean free path $\tau_{\mathrm{point}}^{-1} \sim \omega^4$. The overall scale of κ_{ph} is set by its value at T_c, which is found to be roughly seven times $\kappa_{\mathrm{el}}(T_c)$.

Zn substitution: high temperatures.—In Fig. 2, we plot the theoretical $\kappa(T)$ obtained by adding $\kappa_{\mathrm{ph}}(T)$, determined as above, to $\kappa_{\mathrm{el}}(T)$, with disorder parameter Γ chosen to give rough agreement with the peak height and position of the nominally clean sample of Ref. [7]. We note that the quasiparticle mean free path extracted from nominally pure crystal data in Ref. [7], is actually similar to the mean free path in the 0.15% Zn sample of Ref. [3], making the assignment $\Gamma/T_c = 0.007$ consistent with earlier analysis [20]. In Fig. 3 we now show results for the total thermal conductivity with systematic Zn doping compared to data of Ref. [8] on twinned crystals of $\mathrm{YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}}$. Note that the impurity scattering parameters Γ for the various curves were chosen to reflect Zn concentrations of 0.1%, 0.2%, 0.7%, and 1.7% using the identification of a contribution to the scattering rate $\Gamma/T_{c0} = 0.12$ per 1% Zn. This is a factor of 2 larger than the contribution extracted from comparisons with microwave data in Ref. [20], but consistent with the error bars cited in that work. We have also added in all cases a residual scattering rate of $\Gamma/T_c = 0.007$ consistent with the nominally pure sample as in Fig. 2, apparently representative of oxygen defects in the near-optimally doped samples. With this assignment, there are no further free parameters in the theory.

We note first that the position of the peak in temperature is nonmonotonic in both the data and the theory. In the current theory this occurs because, while the electronic peak initially dominates and moves upward in T with disorder, the phonon peak at 20–25 K eventually becomes more important as the scale of κ_{el} is reduced by disorder. We expect that continued Zn doping will lead to saturation and a peak position fixed at 20–25 K when κ_{el} disappears, leaving the (roughly impurity independent) κ_{ph} of Fig. 2.

Wiedemann-Franz and anisotropy ratios.—Measured values of the ratio $\kappa(T_c)/T_c\sigma(T_c)$ are typically $(1.0\text{–}1.5) \times 10^{-7}\ \mathrm{W\Omega/K^2}$. If we take $R \equiv \kappa_{\mathrm{ph}}(T_c)/\kappa_{\mathrm{el}}(T_c) = 7$ as above, we find for the Lorenz number $L = \kappa_{\mathrm{el}}(T_c)/T\sigma(T_c) = (1.2\text{–}2.0) \times 10^{-8}\ \mathrm{W\Omega/K^2}$. Given that electron correlations can change this ratio substantially, this is quite close to the free electron value of $L_0 = 2.44 \times 10^{-8}\ \mathrm{W\Omega/K^2}$. The measured anisotropy ratio in the plane, $\kappa_b(T_c)/\kappa_a(T_c)$, is 1.2–1.3 for clean untwinned crystals [4,8]. If we assume that all anisotropy arises from the electronic component, and using $R \simeq 7$ once again, we find $\kappa_{b,\mathrm{el}}(T_c)/\kappa_{a,\mathrm{el}}(T_c) = 2.6\text{–}3.4$. This is close to the measured electrical conductivity ratio of $\sigma_b/\sigma_a \simeq 2.4$ reported by Zhang *et al.* [23].

Zn substitution; low temperatures.—Direct information on electronic properties may be obtained by working at very low temperatures, such that the phonon contribution, which should fall as T^3 when the mean free path saturates due to boundary scattering, is negligible. In the resonant scattering limit considered here, a significant linear-T electronic contribution $\kappa_{\mathrm{el}} \simeq aT$ should dominate the phonon conductivity in a d-wave superconductor. The thermal conductivity in this limit is "universal" in the sense that the prefactor a is independent of the impurity scattering rate to leading order [9–11], in analogy to the limiting $T = 0, \omega \to 0$ conductivity $\sigma_{00} \equiv ne^2/m\pi\Delta_0$ [24]. As pointed out by Graf *et al.*, the Wiedemann-Franz law is obeyed exactly at $T = 0$ in the clean limit, i.e., $a \to L_0\sigma_{00}$.

While σ_{00} is difficult to measure due to its small size and to possible extrinsic contributions alluded to above,

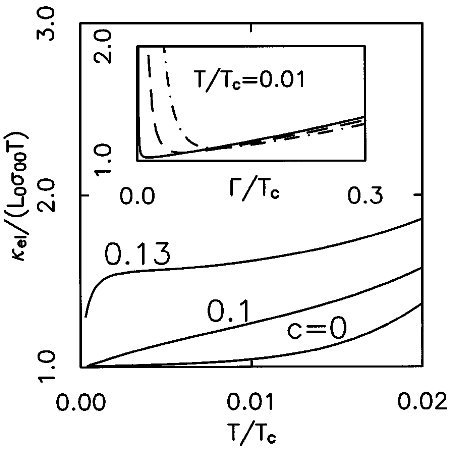

FIG. 4. Normalized electronic thermal conductivity $\kappa_{\mathrm{el}}/L_0\sigma_{00}T$ vs T/T_c for varying electronic scattering phase shifts, $c = 0, 0.1, 0.13$, $\Gamma/T_c = 0.007$. Inset: $\kappa_{\mathrm{el}}/L_0\sigma_{00}T$ vs Γ/T_c for fixed $T = 0.01T_c$ for $c = 0$ (solid line); $c = 0.1$ (dashed line); and $c = 0.3$ (dash-dotted line).

the linear term in $\kappa(T)$ should be clearly visible, and its magnitude and impurity dependence sensitive tests of the "dirty d-wave" model.

The exact expression for the limiting value of κ_{el}/T for $T \ll T_c$ is [9,10] $a = L_0\sigma_{00}k'\mathbf{E}(k')$, where $k' = \sqrt{\Delta_0/(\Delta_0^2 + \gamma^2)}$, $\mathbf{E}$ is a complete elliptic integral of the 2nd kind, and $\gamma = -\mathrm{Im}\,\Sigma_0(\omega = 0)$. In the unitarity limit $c = 0$, the $\kappa_{\mathrm{el}} = aT$ relation holds over a temperature range $T \lesssim \sqrt{\Gamma\Delta_0}$, whereas away from this point the range of validity quickly vanishes. A quasilinear behavior may nonetheless be observed; for example, in the opposite limit, limit $c \gg 1$, we have $\kappa_{\mathrm{el}}(T)/T \simeq [\kappa_{\mathrm{el}}(T_c)/T_c][2\Gamma\tau(T_c)]^{-1}$ above an exponentially small crossover scale $\sim\Delta_0 \exp -\Delta_0(1 + c^2)/\Gamma$. To illustrate the possible range of behavior we plot in Fig. 4 κ_{el}/T at $T/T_c = 0.01$ for various values of c close to the unitarity limit, and as a function of Zn concentration. The main point we wish to make here is that it is possible that small deviations from the unitarity limit may lead at low T to behavior quite different from that predicted for unitarity limit scattering.

In conclusion, we have shown that dirty d-wave theory provides a good account of the systematic behavior of the thermal conductivity in $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$. In particular, it allows for a simple understanding of the size and position of the intermediate temperature peak as a function of disorder. The analysis is consistent with earlier studies of the microwave conductivity, enabling semiquantitative predictions. On this basis we have provided strong evidence for a phononic conductivity significantly larger than its electronic counterpart near T_c. The electronic component is nevertheless responsible for the peak in clean samples, but disappears with percent level Zn doping. Finally, we have discussed the low-T limiting behavior of κ, and pointed out that small deviations from the unitarity scattering limit can give rise to quite nonuniversal results.

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