Quasiparticle Transport in the Vortex State of $YBa_2Cu_3O_{6.9}$

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The effect of vortices on quasiparticle transport in cuprate superconductors was investigated by measuring the low temperature thermal conductivity of $YBa_2Cu_3O_{6.9}$ in magnetic fields up to 8 T. The residual linear term (as $T \to 0$) is found to increase with field, directly reflecting the occupation of extended quasiparticle states. A study for different Zn impurity concentrations reveals good agreement with recent calculations for a d-wave superconductor. It also provides a quantitative measure of the gap near the nodes. [S0031-9007(99)08763-3]

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In unconventional superconductors, where the gap structure has nodes along certain directions, it was argued by Volovik [1] that the dominant effect of a magnetic field should be the Doppler shift of extended quasiparticle states due to the presence of a superfluid flow around each vortex. Reports of a $\sqrt{H}$ field dependence in the heat capacity of $YBa_2Cu_3O_{6.9}$ (YBCO) have generally been accepted as strong evidence for this effect [2]. However, a similar field dependence is also observed in an s-wave superconductor such as $NbSe_2$ [3], where it can only arise from localized states bound to the vortex core.

In this respect, it is interesting to look at heat conduction, to which only delocalized states contribute. So far, in all measurements performed above 5 K or so, the thermal conductivity κ is found to drop with field [4–6], much as it does in a clean type-II superconductor such as Nb [7], and eventually levels off to a roughly constant plateau [8]. In $Bi_2Sr_2CaCu_2O_8$ (BSCCO), the crossover from drop to plateau occurs abruptly, at a field whose magnitude increases with temperature [6]. Franz has recently proposed that the plateau is due to a compensation between the increasing occupation of extended quasiparticle states *à la Volovik* and a parallel increase in the scattering of quasiparticles by vortices [9]. While this may well be part of the explanation, it can hardly serve as a direct confirmation of the "Volovik effect." Neither can it explain the abruptness of the onset of the plateau. The situation remains quite puzzling, in part because of the large phonon background at these temperatures.

In this Letter, we present a study of heat transport which provides a solid experimental basis for a description of quasiparticle properties in terms of the field-induced Doppler shift of a d-wave spectrum due to the superflow around vortices. Furthermore, the good agreement found with recent calculations by Kübert and Hirschfeld [10] allows us to conclude that, in YBCO at low temperature, impurity scattering is close to the unitarity limit and vortex scattering is weak. Moreover, it provides a measure of the two parameters that govern the Dirac-like spectrum near the nodes, responsible for all low-energy properties.

We have measured the thermal conductivity of three optimally doped untwinned single crystals of $YBa_2(Cu_{1-x}Zn_x)_3O_{6.9}$ down to 0.07 K, with a current along the a axis of the basal plane and a magnetic field along the c axis. The nominal Zn concentrations are $x = 0$ (pure), 0.006, and 0.03, corresponding to $T_c = 93.6$, 89.2, and 74.6 K, respectively. The data were taken by sweeping the temperature at fixed field, and the field was changed at low temperature (below 1 K). Data taken by field cooling the samples gave the same results. Given that the field of full penetration is estimated to be about 1 T [11], the lowest field point was set at 2 T. The largest source of absolute error is the uncertainty in the geometric factor (approximately $\pm 10\%$); however, the relative error between different field runs on the same crystal is due only to the fitting procedure used to extract the electronic contribution (see below), which is less than 10%.

Figure 1 shows the measured thermal conductivity divided by temperature, κ/T, as a function of T^2 in fields up to 8 T. As discussed in Ref. [12], the only reliable way to separate the electronic contribution from the phonon background is to reach the T^3 regime for the phonon conductivity (below about 130 mK), from which a linear extrapolation to zero yields the purely electronic term at $T = 0$, labeled κ_0/T.

Zero field.—Let us first focus on the results at $H = 0$. A linear term is observed at $T = 0$, of comparable magnitude in all three crystals. From measurements on a number of pure (optimally doped) crystals of YBCO, we obtain an average value for $\mathbf{J} \parallel \mathbf{a}$ [13]:

$$\frac{\kappa_0}{T} = 0.14 \pm 0.03 \ \mathrm{mW\,K^{-2}\,cm^{-1}}. \qquad (1)$$

This residual conduction is associated with an impurity band whose width γ is a new energy scale relevant to all low-energy properties in superconductors with gap zeros [14]. γ grows with the impurity scattering rate Γ, in a way which depends strongly on whether impurities act as Born or resonant scatterers. In the unitarity limit, γ is maximum and given by $\gamma \simeq 0.61\sqrt{\hbar\Gamma\Delta_0}$ [18], where Δ_0 is the gap

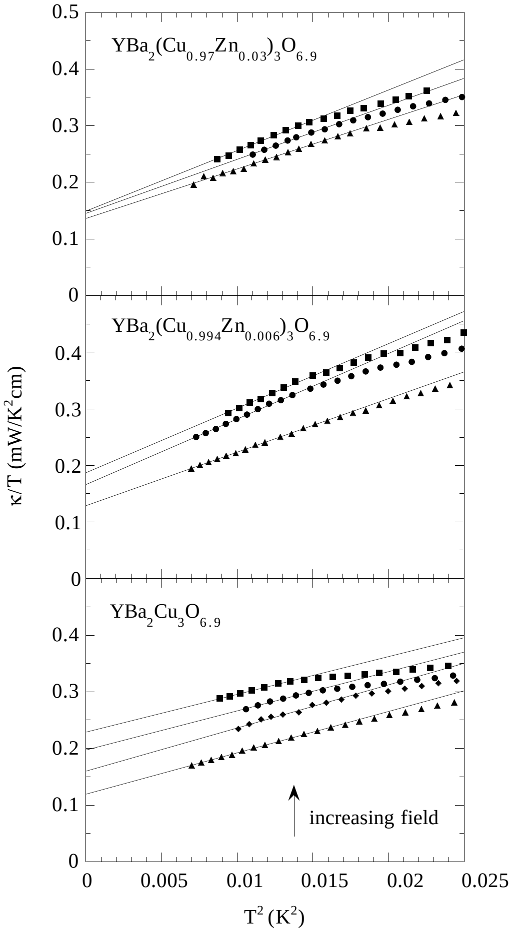

FIG. 1. Thermal conductivity divided by temperature vs T^2 of $YBa_2(Cu_{1-x}Zn_x)_3O_{6.9}$ in a magnetic field, for $H = 0$ (triangles), 2 (diamonds), 4 (circles), and 8 T (squares). The bottom panel corresponds to $x = 0$; middle, $x = 0.006$; top, $x = 0.03$. The lines are linear fits to the data below 130 mK.

maximum. This novel fluid, deep in the superconducting state, should carry heat and charge. Lee was the first to point out the *universal* character of that conduction as $T \to 0$ (and frequency $\omega \to 0$), for certain gap topologies [15]. For the $d_{x^2-y^2}$ gap, the conductance of a single CuO_2 plane is predicted to be $\sigma_{00} = \frac{e^2}{2\pi\hbar}\frac{2}{\pi}\frac{v_F}{v_2}$, where v_F and v_2 are the two parameters governing the Dirac-like spectrum near a node: $E(k) = \sqrt{\varepsilon_k^2 + \Delta_k^2} = \hbar\sqrt{v_F^2 k_1^2 + v_2^2 k_2^2}$. Here (k_1, k_2) defines a coordinate system whose origin is at the node, with k_1 normal to the Fermi surface and k_2 tangential; thus v_F is the Fermi velocity and v_2 is the slope of the gap, at the node. The conductivity is independent of impurity concentration as a consequence of the exact compensation between the decreasing scattering time $\tau \propto 1/\gamma$ and the growing residual quasiparticle density of states, $N(0) \propto \gamma/v_F v_2$, when impurities are added, i.e., $\sigma_{00} \propto N(0) v_F^2 \tau \propto v_F/v_2$.

Graf and co-workers showed that the residual normal fluid should obey the Wiedemann-Franz law at $T = 0$ [14], so that the thermal conductivity as $T \to 0$ in a d-wave superconductor should also be universal, and given by

$$\frac{\kappa_{00}}{T} = L_0\sigma_{00} = \frac{k_B^2}{3\hbar}\frac{v_F}{v_2} n\,, \qquad (2)$$

where n is the number of CuO_2 planes per meter stacked along the c axis and $L_0 = \frac{\pi^2}{3}(\frac{k_B}{e})^2$.

In recent measurements of $\kappa(T)$ in YBCO, the universal limit was observed [12]. It is also evident from the $H = 0$ curves in Fig. 1, noting that the scattering rate Γ in the 3% Zn crystal is some 20 to 40 times larger than it is in the pure crystal. Indeed, estimates from a combination of resistivity, microwave, and infrared measurements give $\hbar\Gamma/k_B T_{c0} = 0.014$, 0.13, and 0.54 for $x = 0$, 0.006, and 0.03, respectively [12]. Note also that the impurity bandwidth in the 3% Zn sample is a sizable fraction of the gap maximum, so that corrections to the universal limit are expected. Calculations by Sun and Maki give a 30% increase in κ_0/T for 20% T_c suppression [16], in good agreement with the observed slight increase (see Fig. 2).

For the bilayer structure of YBCO, $n = 2/c$ where $c = 11.7$ Å is the c-axis lattice constant. Combining this with Eqs. (1) and (2) yields

$$\frac{v_F}{v_2} = 14 \pm 3\,. \qquad (3)$$

Taking $v_F = 1.2 \times 10^7$ cm/s (see Ref. [17]), one gets $v_2 \simeq 1 \times 10^6$ cm/s, which corresponds to a slope of the gap at the node $d\Delta(\phi)/d\phi = \hbar k_F v_2 \simeq 380$ K, very close to that estimated using the simplest d-wave

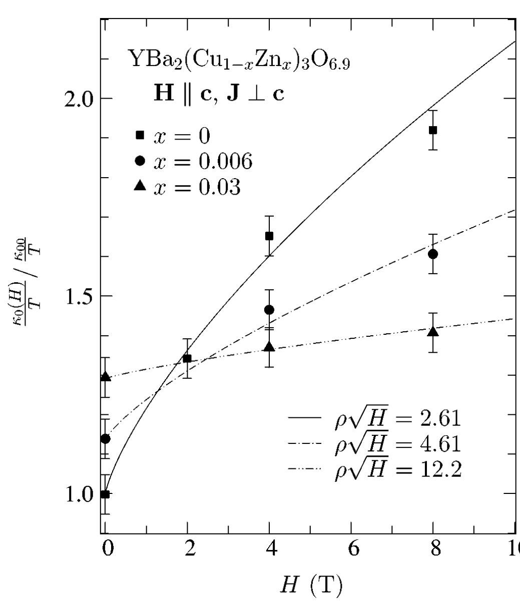

FIG. 2. Residual linear term $\kappa_0(H)/T$ normalized by κ_{00}/T as a function of applied field for pure (squares), 0.6% Zn (circles), and 3% Zn (triangles) samples. Fits to Eq. (4) for each crystal yield the values of ρ shown (in units of $T^{-1/2}$).

gap, $\Delta_0 \cos 2\phi$, in the weak-coupling limit: $2\Delta_0 = 2 \times 2.14 k_B T_c = 400$ K. The same ratio v_F/v_2 governs the linear drop in superfluid density with temperature when $\gamma \ll k_B T \ll \Delta_0$. In terms of penetration depth data, where $\lambda(T) = \lambda(0) + \delta\lambda(T)$, one has $\delta\lambda(T) = (\frac{c}{\omega_p}) 2 \ln 2 (\frac{k_n T}{\hbar k_F v_2})$ [17], where $\omega_p^2 = 2\pi^2 k_F v_2 \sigma_{00}$, so that $\delta\lambda/T = 4.8$ Å/K for the above values of v_F and v_2. This is in excellent agreement with the microwave measurements of Zhang and co-workers, who obtain $\delta\lambda_a/T = 4.7$ Å/K [18].

Vortex state.—As seen in Fig. 1, the effect of a magnetic field applied normal to the CuO_2 planes is to *increase* κ_a in YBCO. To ensure that only the electronic contribution is considered, the $T \to 0$ limit of κ/T is plotted as a function of field in Fig. 2, for each of the three crystals. The enhancement observed as $T \to 0$ is direct evidence for itinerant quasiparticle excitations *induced by the field*. This is in sharp contrast with the behavior found both in conventional superconductors [7] and in YBCO at higher temperatures [4,5,8], where low fields (compared to H_{c2}) *suppress* the conductivity.

Volovik has shown how even at $T = 0$ a small magnetic field can produce quasiparticles near the nodes of a d-wave gap [1]. In the presence of a superfluid flow, the quasiparticle spectrum is Doppler shifted to $E(\mathbf{k}, \mathbf{A}) = E(\mathbf{k}) - \frac{e}{c} \mathbf{v_k} \cdot \mathbf{A}$, where $\mathbf{v_k}$ is the normal state velocity and $\mathbf{A}$ is the vector potential [17]. In the case of vortices, the magnitude of the Doppler shift may be characterized by its average, E_H, obtained by integrating over a vortex-lattice unit cell (of radius R): $E_H = \frac{e}{c} \langle \mathbf{v_k} \cdot \mathbf{A} \rangle \sim \frac{1}{R^2} \int dr\, r v_F A \sim \frac{1}{R} \Phi_0 v_F \sim \sqrt{H}$, using $2R \simeq$ intervortex spacing $\sim 1/\sqrt{H}$ and the fact that $\Phi_0 = \oint \mathbf{A} \cdot d\ell$, with Φ_0 the flux quantum. Volovik first derived the $\sqrt{H}$ dependence and obtained the following field-induced density of states (DOS) for a d-wave gap in the clean limit [1]: $\delta N(0; H)/N_0 \simeq \sqrt{8/\pi}\, E_H/\Delta_0$, where N_0 is the DOS at the Fermi level, $E_H = a\hbar\sqrt{2/\pi}\, v_F \sqrt{H/\Phi_0}$, and a is a vortex-lattice parameter of order unity [19]. The effect is sizable even at low fields; in YBCO at 8 T, one expects $E_H \simeq 40$ K and $\delta N(0; H)/N_0 \simeq 30\%$ (for $a = 1$).

Comparison with theory.—Kübert and Hirschfeld have calculated the electronic thermal conductivity of a d-wave superconductor in a field, neglecting vortex scattering [10]. At $T = 0$, they obtain, for $\mathbf{H} \parallel \mathbf{c}$ and $\mathbf{J} \perp \mathbf{c}$,

$$\frac{\kappa(0; H)}{T} = \frac{\kappa_0}{T} \frac{\rho^2}{\rho\sqrt{1 + \rho^2} - \sinh^{-1}\rho}, \qquad (4)$$

where ρ is essentially the ratio of the two relevant energy scales, γ and E_H; in the dirty limit where $E_H < \gamma$, $\rho = \sqrt{6/\pi}\, \gamma/E_H \propto 1/\sqrt{H}$. A one-parameter fit of the data to Eq. (4) for each crystal is shown in Fig. 2. The first point to note is that the sublinear dependence on field is well reproduced. Perhaps more important is the fact that the magnitude of the response in all three cases is very much as expected. Indeed, the fits yield the following values for ρ, evaluated at 8 T: 0.92, 1.63, and 4.32 for $x = 0$, 0.006, and 0.03, respectively. These correspond to a ratio $\gamma/E_H = 0.67$, 1.18, and 3.12, which shows that none of the crystals is in the clean limit over the field range investigated. Treating impurity scattering in the unitarity limit, the scattering rate becomes $\hbar\Gamma/k_B T_{c0} = 0.02$, 0.07, and 0.5, respectively (taking $\Delta_0 = 2.14 k_B T_{c0}$), given that $E_H \simeq 20$ K at 8 T (assuming $v_F = 1 \times 10^7$ cm/s and $a = 1/2$). These values are in remarkable agreement with those given above as independent estimates based on the residual resistivity.

The same theoretical treatment has been applied to the specific heat. Kübert and Hirschfeld have obtained $N(0; H)/N_0 = \delta\gamma^*/\gamma_n = \sqrt{8/\pi}\, a\sqrt{H/H_{c2}}$, where $\delta\gamma^*$ is the increase in specific heat due to an applied field and γ_n is the normal state specific heat at the Fermi level [19]. We can reformulate this expression in terms of a and v_2 such that $\delta\gamma^* = \beta\sqrt{H}$, where $\beta = \frac{8k_B^2}{3\hbar} \frac{1}{\sqrt{\Phi_0}} \frac{a}{v_2}$. So now we have three equations for the three parameters a, v_F, and v_2. Using $\beta = 0.9$ mJ K^{-2} T$^{-1/2}$ mol^{-1} [2], we find $v_2 = 2.2a \times 10^6$ cm/s. From $\kappa_0(0; H)/T$, we have $a v_F = 5 \times 10^6$ cm/s. These two expressions, together with Eq. (3), yield $a = 0.40$. With this value of a, $v_2 = 0.9 \times 10^6$ cm/s and $v_F = 1.2 \times 10^7$ cm/s. All three values are very reasonable, so that the overall picture is quantitatively convincing, lending strong support to the basic mechanism of a Doppler shift by superflow around vortices. More generally, it validates the theory of transport in unconventional superconductors; in particular, the assumption that in these correlated electron systems scattering by impurities must be treated in the unitarity limit of strong, resonant scattering appears to be correct.

Other measurements.—Let us now compare our results with other κ measurements. Note first that previous low temperature measurements in YBCO were done with the field in the basal plane, where the Volovik effect is expected to be much smaller. The two studies were in disagreement, with a 50% increase in 8 T in one case [20], but no change detected in 6 T in the other [21]. At higher temperatures, the fact that κ decreases with field could come from a number of effects. Perhaps the most natural is vortex scattering, as invoked in the case of Nb. While this can apply to both quasiparticles and phonons, the fact that the largely electronic peak below T_c is almost completely suppressed in 10 T [4] suggests that the former suffer most of the impact. Franz has shown that a disordered vortex lattice in a d-wave superconductor can result in quasiparticles scattering off the superflow [9]. At low temperature, however, we find no indication of significant vortex scattering, in the sense that there is good agreement between calculations neglecting vortex scattering and our data on crystals for which the relative strength of impurity and vortex scattering must differ markedly from one crystal to the next. Further work is needed to arrive at a coherent description of quasiparticle transport in the vortex state which covers both regimes of behavior.

It is interesting to compare our results on YBCO with the corresponding results on BSCCO, obtained recently by Aubin and co-workers [22]. At temperatures below 0.7 K, an increase in κ/T with field is also found, again with a sublinear (roughly $\sqrt{H}$) dependence. What is striking is the *magnitude* of the response. The application of only 2 T increases κ/T by about 0.20 mW K^{-2} cm^{-1}. Now from our own measurements on pure, optimally doped single crystals of BSCCO, the (presumably universal) residual linear term is $\kappa_0/T = 0.18 \pm 0.03$ mW K^{-2} cm^{-1} [23]. This means that in BSCCO a field of 2 T causes the quasiparticle conduction to *double,* whereas it produces only a 35% increase in our pure YBCO crystal. In reality, the field dependence in BSCCO is much more than 3 times stronger, since the impurity scattering rate in the crystal used by Aubin and co-workers could be as much as 100 times larger. Indeed, its residual resistivity is 130 $\mu\Omega$ cm [24], compared with approximately 1 $\mu\Omega$ cm in our pure YBCO crystals [12]. These considerations lead us to conclude that the nature of defect scattering in these two (otherwise quite similar) materials is significantly different. Either the kind of defect found in nominally pure crystals is different or the impact that a given defect (e.g., impurity) has on the surrounding electron fluid is different. It will be interesting to see whether the current theoretical framework can account for the BSCCO data simply by adjusting the impurity phase shift, moving it away from the unitarity limit towards the Born limit.

In summary, we have measured the low temperature heat conduction by quasiparticles in the CuO_2 planes of YBCO as a function of magnetic field, for different impurity levels. In all cases, the residual linear term κ_0/T is found to increase with field strength, directly reflecting the additional population of extended quasiparticle states. The good agreement with calculations by Kübert and Hirschfeld for a d-wave superconductor allows us to draw the following conclusions: (1) The Volovik effect is fully verified, and it is the dominant mechanism behind the field dependence of transport in YBCO as $T \to 0$, for $\mathbf{H} \parallel \mathbf{c}$; (2) vortex scattering, invoked to explain the behavior at intermediate temperatures, is seemingly absent at low temperature; (3) the widespread assumption that impurities (or defects) can be treated as unitary scatterers in correlated electron systems appears to be verified in YBCO; (4) the nature of impurity scattering differs between BSCCO and YBCO.

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