Plateaus Observed in the Field Profile of Thermal Conductivity in the Superconductor $Bi_2Sr_2CaCu_2O_8$

K. Krishana, N. P. Ong, Q. Li, G. D. Gu, N. Koshizuka

Quasi-particles (QPs) are excitations of the superconducting state. The behavior of QPs in $Bi_2Sr_2CaCu_2O_8$ in a magnetic field was probed by measurement of the thermal conductivity κ. An anomaly in κ was observed at low temperatures. At a transition field H_k, κ displayed a sharp break in slope, followed by a plateau region in which it ceased to change with increasing field. The nonanalytic nature of the break at H_k suggests a phase transition of the condensate to a state in which the QP current is zero (the system remains superconducting). Detailed measurements of the new regime are presented, and implications for the QPs and the superconducting state are discussed.

In a superconductor, the pairing of electrons leads to a collective state called the condensate. The ability of the condensate to carry a dissipationless supercurrent and to shield its interior from external fields endows the superconductor with its best-known properties. At finite temperature T, some of the electrons are excited from the condensate into quasi-particle (QP) states, which are states of the low-lying excitations of the superconductor. The properties of this "normal gas" of particles and how they change in a field are of great interest, especially in the cuprate superconductors, in which the superconducting properties are highly anisotropic and the gap symmetry is d-wave, as shown by recent experiments (1–3). In conventional s-wave superconductors such as Sn and Nb, the energy gap Δ is nonzero everywhere on the Fermi surface (FS). For T significantly lower than Δ/k_B, the population of QPs is exponentially small (k_B is Boltzmann's constant). By contrast, in a d-wave superconductor, Δ is zero along lines of nodes on the FS. A sizeable population of QPs remains at cryogenic temperatures. This residual population may be used as a probe to address issues such as whether a magnetic field changes the d-wave state in unexpected ways. Hence, apart from their intrinsic interest, QPs can reveal subtle changes in the condensate and the gap symmetry. However, because of the excellent shunting capability of the condensate, QPs are difficult to probe by electrical means, and many of their low-temperature properties in the cuprates remain unexplored.

Unlike the condensate, QPs respond strongly to a temperature gradient, and thus thermal conductivity (κ) experiments may provide a direct window on QP behavior. This promise has not been fully realized because it is difficult to isolate the QP current from the much larger phonon current. The response of the two currents to an applied field is also not well understood. A distinctive feature of the cuprates is that the in-plane thermal conductivity $\kappa \equiv \kappa_{xx}$ (measured with the gradient $-\nabla T \|_x$ in the plane) exhibits a large anomalous enhancement (4, 5) below the critical temperature T_c. Experiments (6, 7) show that the anomaly is readily suppressed by the magnetic field H. The origin of the anomaly, its field sensitivity, and the response of the phonon current are open questions.

We report here a series of high-resolution measurements of κ in the cuprate $Bi_2Sr_2CaCu_2O_8$ (Bi 2212) that reveal a surprising feature of the QP heat current. At low temperatures, κ exhibits a nonanalytic break in slope at a transition field H_k. The break is followed by a plateau that extends to our highest field. Thus, at fields below H_k, the QPs carry a fraction of the heat current, whereas above H_k, their contribution vanishes (although the sample remains superconducting). The most direct interpretation is that the magnetic field induces a phase transition in the condensate to a new state in which the QP population is negligibly small. The new results raise the question whether the d-wave state in the cuprates is inherently unstable at low T in a relatively weak field.

The measurement of κ in intense fields at our level of resolution (three parts in 10^4) precludes the use of thermocouples to measure $-\nabla T$ directly. Unacceptably large errors are introduced by the field sensitivity of the thermocouple (which exceeds a few percent at 10 K in a 1-T field) and may be altered by extraneous factors such as strain and thermal cycling). Instead, we developed a bridge-balancing technique in which the gradients in the sample and in a reference (a nylon bar) are maintained by separate heaters. The thermocouple (chromel constantan) serves only as a null detector to compare the two gradients. At present, the bridge-balancing procedure allows pointwise measurements at discrete field values only. This technique is tedious but is capable of high resolution and is unaffected by changes in the sensor sensitivity (8).

Figure 1A shows the T dependence of κ_{xx} in sample 1 in zero field. Below T_c (92 K), we observe a broad anomaly in κ that peaks near 65 K. An applied field $\mathbf{H} \parallel \mathbf{c}$ readily suppresses the anomaly. Figure 1B displays the fractional change in κ with H at temperatures above 30 K. At each T, the initial decrease in κ with field is very rapid, which suggests a cusplike behavior in weak fields. The usual explanation of the field-induced changes is that both QPs and phonons are increasingly scattered by the growing density of vortices. From the present experiment, we believe that this model needs revision.

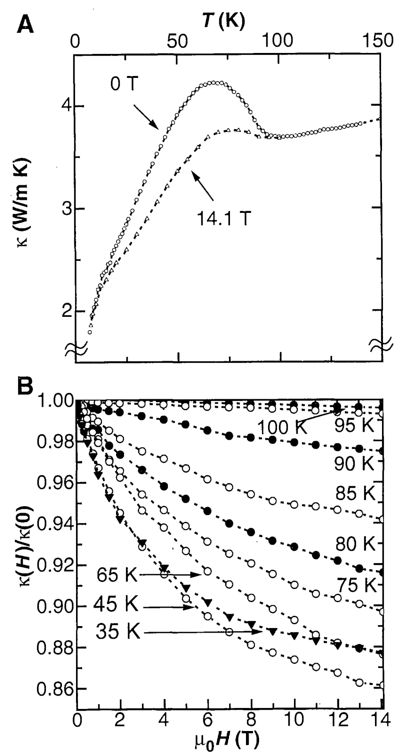

Fig. 1. (**A**) The T dependence of the in-plane thermal conductivity κ in $Bi_2Sr_2CaCu_2O_8$ at H = 0 and 14.1 T (sample 1). (**B**) The H dependence of κ at T above 25 K ($\mathbf{H} \parallel \mathbf{c}$; broken lines are guides to the eye). As T decreases below 90 K, the fractional change increases rapidly. The H dependence flattens noticeably at 35 K. (μ_0 is the vacuum permeability.)

K. Krishana and N. P. Ong, Joseph Henry Laboratories of Physics, Princeton University, Princeton, NJ 08544, USA.
Q. Li, Materials Science Division, Brookhaven National Laboratory, Post Office Box 5000, Upton, NY 11973, USA.
G. D. Gu, School of Physics, University of New South Wales, Sydney 2052, Australia.
N. Koshizuka, Superconductivity Research Laboratory, International Superconductivity Technology Center, 10-13, Shinonome 1-chrome, Koto-ku, Tokyo 135, Japan.

New features appear at lower temperatures. At 35 K, the rate of decrease weakens noticeably at high fields. Below 20 K, an anomaly becomes apparent. The decrease in κ is suddenly interrupted at a "kink" field H_k, above which it ceases to change (Fig. 2). The field H_k clearly divides the response into two parts: a low-field region in which κ falls steeply with field, and a high-field plateau region in which it is field-independent to our resolution (although still T-dependent). In the top three curves in Fig. 2 (scale expanded), the slight positive slope of the background is caused by the magnetoresistance (MR) of the heaters (8). The transition field H_k is strongly T-dependent, decreasing from 5 T at 20 K to about 0.6 T at 6 K. Below 10 K, the nonanalytic nature of the kink is apparent. Clearly, κ assumes its plateau value suddenly, instead of approaching it smoothly as in an exponential decay. In sample 2 (Fig. 3), we extended the measurements to 14 T; similar values for H_k were observed at 10, 15, and 20 K.

For later discussion, we isolate the field-dependent part of κ relative to its value κ_p at the plateau by defining $\Delta\kappa(B, T) \equiv \kappa(B, T) - \kappa_p(T)$ (B is the magnetic field). Figure 4A shows the field dependence of $\Delta\kappa(B, T)$ at selected temperatures. With decreasing T, its zero-field value $\Delta\kappa(0, T)$ decreases nominally as T^n, with $n = 1.9 \pm 0.2$ (open circles in Fig. 4B). The threshold field H_k follows a power-law dependence with the same exponent within our uncertainty (closed circles). Because their T dependences are similar, $\Delta\kappa(0, T)$ and H_k decrease proportionately with decreasing T, as shown in Fig. 4A. The nonanalytic nature of the kink suggests that H_k is a transition line in the phase diagram of the QPs. The small value of H_k at low temperatures is remarkable. It is highly unlikely that H_k is related to various phase transition lines in the vortex phase diagram in Bi 2212. Its magnitude and T dependence are incompatible with the irreversibility line, which increases very steeply with decreasing T to values in excess of 10 T near 14 K. It is also incompatible with the vortex-solid melting line (9), which saturates at 400 Oe below 40 K. The lower critical field H_{c1}(~100 Oe) is even smaller.

The insensitivity of κ to field in the plateau region imposes two independent constraints on the heat current. In a superconductor, the total conductivity is the sum of the QP (electronic) term κ_e and the phonon term κ_{ph}, namely $\kappa = \kappa_e + \kappa_{ph}$. Because a change in either component will violate the condition $\partial\kappa/\partial H = 0$, our results imply that $\partial\kappa_e/\partial H$ and $\partial\kappa_{ph}/\partial H$ are separately zero above H_k.

The most direct interpretation of the constraint $\partial\kappa_e/\partial H = 0$ is that the QP conductivity κ_e is zero in the plateau region. The observed κ arises entirely from the phonons, which flow as a large, field-independent background current. Hence, at each field value, the quantity $\Delta\kappa(B, T)$ is just κ_e.

We may roughly estimate the zero-field QP thermal current. In the theory of Bardeen, Rickayzen, and Tewordt (10, 11), the electronic conductivity is expressed as

$$\kappa_e = \Sigma_k (E_k v_k \cos\theta)^2 (T\Gamma)^{-1}(-\partial f_k^0/\partial E_k) \tag{1}$$

where $E_{\mathbf{k}} = \sqrt{[\epsilon_{\mathbf{k}}^2 + \Delta(\mathbf{k})^2]}$ is the QP energy in state $\mathbf{k}$, $v_{\mathbf{k}} \cos\theta$ the component of its group velocity parallel to $-\nabla T$, Γ the relaxation rate, and $f_{\mathbf{k}}^0$ the Fermi-Dirac distribution ($\epsilon_{\mathbf{k}}$ is the normal state energy). With the d-wave gap given by $\Delta(\mathbf{k}) = \Delta_0(\cos k_x a - \cos k_y a)$, we find for a two-dimensional (2D) superconductor at low T the simple result (12)

$$\kappa^{2D} = 4\eta(k_B^2 T/\hbar)(k_B T/\hbar\Gamma) \tag{2}$$

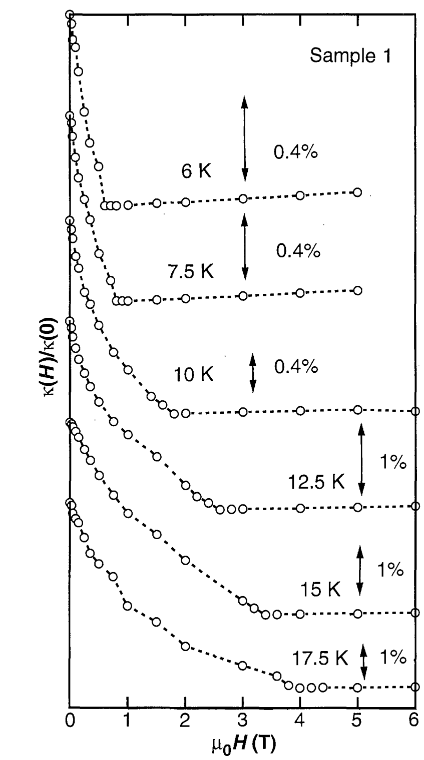

Fig. 2. The H dependence of κ in sample 1 below 20 K (**H** ∥ **c**). Starting at 20 K, κ ceases to change with field above a threshold field H_k. The slight positive slope above H_k in the expanded scale (top three curves) reflects the weak MR of the thin-film heaters. The break at H_k is sharp, suggestive of a transition in the condensate. Above 22.5 K, the kink is severely rounded by thermal broadening.

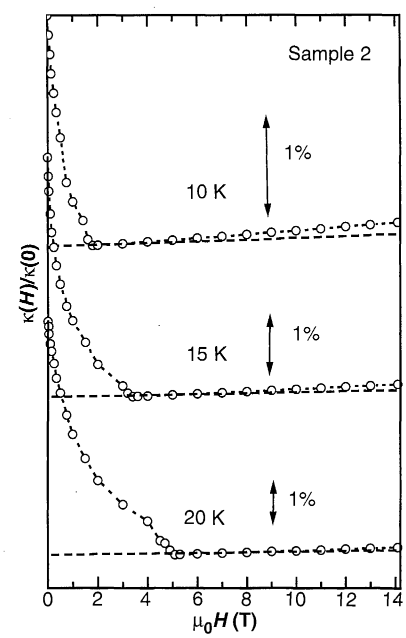

Fig. 3. The H dependence of κ in sample 2, showing the plateau region up to 14 T. The broken line shows the MR of the heaters (8). The slight shoulder just below H_k (5 T) in the curve at 20 K is observed in some curves but not in others (see also curve at 17.5 K in Fig. 2). More densely spaced data are needed to determine whether this is an intrinsic feature.

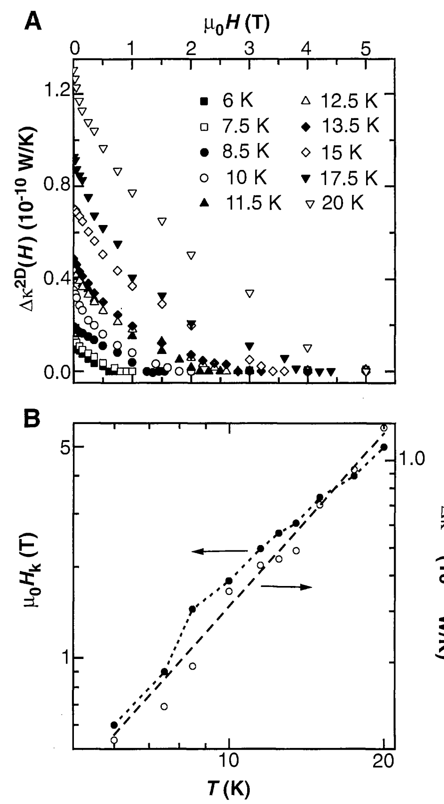

Fig. 4. (**A**) The H dependence of $\Delta\kappa^{2D}(B, T) \equiv [\kappa(B, T) - \kappa_p]s/2$ in sample 1 (the bilayer spacing s = 15.6 Å). (**B**) T dependence of $\Delta\kappa^{2D}(B, T)$ and the field H_k in log scale. The broken straight line is a fit to Eq. 2 with $1/\Gamma$ = 0.38 ps.

where $\eta = (3/2\pi) \int_0^\infty dz\, z^2(e^z + 1)^{-1} \sim 0.86$. The additional factor k_BT reflects the linear density of states at the nodes (3). Thus, the QP current in the clean d-wave superconductor varies as T^2 in the limit $T \rightarrow 0$, which is consistent with Fig. 4B. Equation 2 provides a good fit to the measured κ^{2D} with a relaxation time $1/\Gamma = 0.38$ ps.

These comparisons show that it is reasonable to identify the total field-induced change $\Delta\kappa(H_k, T)$ with the value of κ_e in zero field. However, the inference that $\kappa_e = 0$ in the plateau region presents a challenge to our understanding of the QP state. The simplest way to have $\kappa_e = 0$ is to assume that the QP density n_{QP} vanishes (above H_k). This could arise from a gap Δ_H that opens gradually with field, leading to an exponential decay, namely $n_{QP} \sim \exp(-\Delta_H/k_BT)$ (Δ_H is distinct from the superconducting gap).

However, the observed behavior of κ is incompatible with an exponential decrease in κ_e. As discussed above, the break at H_k is nonanalytic. It is strongly suggestive of a phase transition between distinct states of the condensate: A large gap appears abruptly at the field H_k. A number of possibilities come to mind. One proposal (*13*) is that the field may induce an abrupt change of gap parameter symmetry, from a simple d wave to a complex order parameter (such as $d_{x^2-y^2} + id_{xy}$ or $d_{x^2-y^2} + is$) that reflects the breaking of time-reversal invariance in the field. A complex gap parameter implies that the superconducting gap is non-zero everywhere on the FS. In the new phase, the removal of the nodes abruptly drives the QP population, as well as κ_e, to zero. This scenario, if valid, implies that the superconducting state in the cuprates may have other unexpected properties.

We have also repeated the measurements with **H** in-plane and perpendicular to $-\nabla T$. The behavior is qualitatively different from the behavior reported here and is much weaker in field sensitivity. This difference implies that the changes at H_k are predominantly an orbital effect of the field (as opposed to the Zeeman term involving the spins).

Finally, we comment on the constraint on the phonon current. The condition $\partial\kappa_{ph}/\partial H = 0$ means that vortices in Bi 2212 do not scatter phonons above H_k, despite the rather long phonon mean free path ℓ_{ph} (*14*). Insofar as the behavior at H_k is associated with the QPs, we may extend this conclusion to fields below H_k as well. The absence of phonon scattering by vortices stands in striking contrast to the situation in classical Bardeen-Cooper-Schrieffer superconductors (*15*), where vortices are strong scatterers of phonons.

REFERENCES AND NOTES

1. C. C. Tsuei *et al.*, *Phys. Rev. Lett.* **73**, 593 (1994).
2. D. A. Bonn *et al.*, *ibid.* **68**, 2390 (1992); D. A. Bonn *et al.*, *Phys. Rev. B* **47**, 11314 (1993).
3. K. A. Moler *et al.*, *Phys. Rev. Lett* **73**, 2744 (1994).
4. A. Jezowski *et al.*, *Phys. Lett. A* **122**, 431 (1987); C. Uher and A. B. Kaiser, *Phys. Rev. B* **36**, 5680 (1987).
5. S. Hagen, Z. Z. Wang, N. P. Ong, *Phys. Rev. B* **40**, 9389 (1989).
6. R. C. Yu, M. B. Salamon, J. P. Lu, W. C. Lee, *Phys. Rev. Lett.* **68**, 1431 (1992).
7. K. Krishana, J. M. Harris, N. P. Ong, *ibid.* **75**, 3529 (1995).
8. At our highest resolution, several background effects are observable. The most significant arises from the weak MR of the thin-film resistors, which is about one part in 10^3 at 14 T. This produces an extraneous, slowly rising background (versus H) when the voltage across the heater is used to monitor the heater current at the sample. The measurements are not affected by a putative field gradient between the sample and the reference. With our field homogeneity (one part in 10^4 over 1 mm), the maximum thermocouple distortion in a field of 1 T is 100 times smaller than our resolution. The two crystals investigated were cut from a boule grown by the traveling-solvent floating-zone technique. Optical microscopy showed the crystal to be single-domained (the structural modulations all lie in one direction).
9. E. Zeldov *et al.*, *Nature* **375**, 373 (1995).
10. J. Bardeen, G. Rickayzen, L. Tewordt, *Phys. Rev.* **113**, 982 (1959).
11. L. P. Kadanoff and P. C. Martin, *ibid.*, **124**, 670 (1961).
12. We are grateful to R. B. Laughlin for help in computing κ_e.
13. P. W. Anderson and F. D. M. Haldane, private communication.
14. From the heat capacity c_v at low T and the relation $\kappa_{ph} = c_v\ell_{ph}v_s/3$, we obtain a lower bound for ℓ_{ph} of 1200 Å at 10 K (the sound velocity $v_s < 5 \times 10^3$ m/s). Thus, scattering of phonons by vortex cores should be easily observed (the vortex lattice spacing is 450 Å at 1 T). $c_v \sim 10.8$ mJ/cm^3 K at 10 K [see A. Junod *et al.*, *Physica C* **162–164**, 480 (1989)].
15. W. F. Vinen, E. M. Forgan, C. E. Gough, M. J. Hood, *Physica* **55**, 94 (1971).

16. We have benefitted from discussions with P. W. Anderson, H. Fukuyama, F. D. M. Haldane, R. B. Laughlin, and P. A. Lee. Research at Princeton is supported by the U.S. Office of Naval Research (contract N00014-90-J-1013) and NSF (DMR 94-00362). Q.L. was supported by the U.S. Department of Energy, Division of Materials Science, contract DE-AC02-76CH00016.

18 March 1997; accepted 5 May 1997

Nuclear Spectroscopy in Single Quantum Dots: Nanoscopic Raman Scattering and Nuclear Magnetic Resonance

D. Gammon,* S. W. Brown,† E. S. Snow, T. A. Kennedy, D. S. Katzer, D. Park

Resonant Raman and nuclear magnetic resonance spectroscopies from single gallium arsenide quantum dots are demonstrated. The nuclei were probed through changes in the optical spectra of the quantum dot exciton arising from exciton-nuclear interactions. This approach allowed the application of optical spectroscopy with its extremely high sensitivity and selectivity. The experiments had a lateral spatial resolution of about 10 nanometers and probe a volume that was five orders of magnitude smaller than that of previous semiconductor nuclear spectroscopic studies.

Semiconductors are typically modeled in terms of almost independent electronic and nuclear systems with coupling through relatively weak electron-nuclear interactions. The electronic system can be studied at optical frequencies through sensitive optical spectroscopies with tunable lasers and highly sensitive detectors. Recently there has been significant progress in the spectroscopic study of very small semiconductor samples, culminating with the detailed study of individual quantum dots (QDs), which are solid-state crystalline structures so small that their electronic wave function is completely localized and their energy spectrum is fully quantized (*1–6*). It has been shown that the optical linewidths of single QDs are orders of magnitude narrower than those observed in ensemble measurements. In fact, the linewidths can approach the natural linewidths expected from radiative lifetimes (*3*). With such narrow lines in the optical spectra, single QD spectroscopy has led, for example, to the direct observation of fine structure (*4*), hyperfine shifts (*5*), and spectral wandering (*6*). Single QD spectroscopy has followed the earlier examples of atomic and, especially, single molecule spectroscopies (*7*). To date, however, only the electronic spectra of single QDs have been measured. Here we report spectroscopic measurements of the nuclear system in a single QD. The capability to do nuclear spectroscopy on a scale of 10 nm in single QDs could provide sensitive measurements of local strain and chemical composition, issues of great importance for semiconductor nanostructures.

Naval Research Laboratory, Washington, DC 20375-5347, USA.

*To whom correspondence should be addressed. E-mail: gammon@bloch.nrl.navy.mil

†Present address: National Institute of Standards and Technology, Gaithersburg, MD 20899, USA.