Density of normal carriers below T_c and thermal resistance of twin boundaries in $YBa_2Cu_3O_{7-x}$ single crystals

I. M. Fishman, G. S. Kino, and X. D. Wu
Ginzton Laboratory, Stanford University, Stanford, California 94305-4085
(Received 2 May 1994)

Photothermal measurements of diffusivity in $YBa_2Cu_3O_{7-x}$ have been made in regions only 10 μm in extent. The measured diffusivity within a single crystallite is much larger than that across a twin boundary. The results obtained are shown to be consistent with the microwave penetration depth measurements of normal electron density over a wide temperature range below the critical temperature. The results are not in good agreement with the BCS theory. It is shown that phonons are reflected at the twin boundaries, and thermal resistance of the boundary is approximately a linear function of temperature in the range $20 < T < 300$ K.

INTRODUCTION

Several recent publications have been devoted to the investigation of thermal conductivity[1–3] and thermal diffusivity[4–8] of high-T_c materials. The generally accepted viewpoint[1] is that, in perfect $YBa_2Cu_3O_{7-x}$ (YBCO) single crystals, phonons are primarily responsible for heat transfer. We have used a high-resolution (2-μm) photothermal measurement technique to characterize thermal diffusion inside a single domain and through the twin boundaries. We confirm that, within a single-crystal domain, phonon diffusion is the dominating mechanism for heat transfer, especially at low temperatures. However, the twin boundaries appear to behave almost like fully reflecting mirrors for the phonons.

Photothermal diffusion measurements in the *a-b* plane of single YBCO crystals show values of diffusivity at room temperature a factor of 2 larger, and show the increase in thermal diffusivity at low temperatures to be much larger than that measured by conventional methods. At low temperatures, diffusivity increases over two orders of magnitude, which is interpreted as an increase in the phonon mean free path due to decrease of density of the normal carriers. This result is in good agreement with the density variation obtained from microwave penetration depth measurements.

In our experiments, an argon laser beam of 514-nm wavelength, acousto-optically modulated by a Bragg cell, is focused to a 2-μm-diam spot on the sample. This pump beam periodically modulates the sample temperature and excites a thermal wave. A second beam from an infrared laser diode, 780 nm in wavelength, is focused on a spot a few microns away and probes the reflectivity change caused by the thermal wave. A lock-in amplifier is used to determine the phase and amplitude of the probe signal relative to that of the pump. The sample can be imaged through a microscope and observed on a video display to ensure that measurements are made within a single crystallite, or alternatively, across twin boundaries. Thermal diffusivity is calculated from the phase delay, the pump frequency, and the beam separation.[4–6,9] To measure the phase delay, we subtract the pump beam phase from the probe phase. The measurement system is described in more detail in Ref. 6.

SINGLE-CRYSTAL THERMAL DIFFUSIVITY

The $YBa_2Cu_3O_{7-x}$ crystals used in this study were twinned. Most of the crystallites were narrow strips several microns wide and 70–100 μm long. Several domains 15–18 μm wide were primarily used in the measurements.

It has been shown by several authors[1–3] that, below the critical temperature, the thermal conductivity of perfect YBCO single crystals increases by a factor of 1.5–2. This increase in thermal conductivity was attributed to a decrease of electron-phonon scattering, and the thermal conductivity maximum is shaped by a fast decrease of heat capacity at low temperatures. The photothermal technique measures directly the thermal diffusivity, rather than the thermal conductivity, in a very small sample volume, avoiding the averaging caused by the sample nonuniformity.[4–6,9] We shall show that this technique is advantageous for diffusivity measurements, since we observe far larger increases in the thermal diffusivity below T_c within a single crystallite than in previous work.

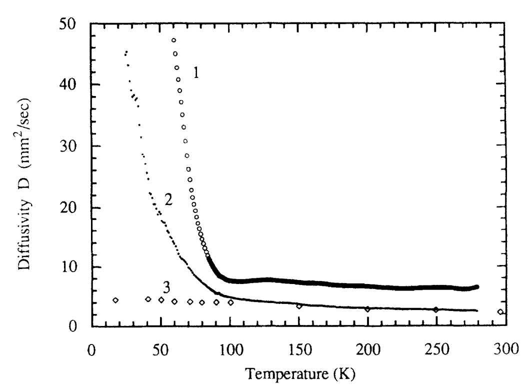

FIG. 1. Diffusivity as a function of temperature for a single YBCO domain (1), across the twin boundary (2), and for a thin film (3).

Results of the diffusivity measurements are shown in Fig. 1. The spacing between the excitation and the probe beams was 10 μm. Curve 1 presents the values measured inside a single YBCO domain, and curve 2 across the twin boundaries. For comparison, the diffusivity measurements for thin YBCO films[7,8] using the transient grating technique in a region 200 μm across are shown by curve 3. It will be seen that above T_c the measured thermal diffusivity is almost independent of temperature, and the thin-film diffusivity agrees with the measurement across twin boundaries. However, the diffusivity measured within a domain is approximately twice as large. Below T_c, the data for different samples differ dramatically. Within a single domain (curve 1), for $T<T_c$ the diffusivity increases sharply over two orders of magnitude in the temperature range from 90 to 30 K. This result implies that the thermal conductivity $\kappa=CD\rho$, where C is the heat capacity, and ρ is the density, reaches a maximum at $T=50$ K about six times its value at room temperature. For samples of lower quality, the enhancement is not as extreme, especially for the thin-film samples (curve 3), where the superconducting transition has almost no effect upon the diffusivity. The measurement across the twin boundaries (curve 2) is intermediate between curves 1 and 3. The twin boundary, though thin compared to the beam spacing, creates significant additional resistance for heat transfer. We shall discuss the origin of the boundary resistance in the next section.

In terms of the conventional model of phonon-dominated heat transfer, a possible explanation of the large increase in diffusivity below T_c is that the phonon mobility is dominated by electron-phonon scattering and that the diffusivity enhancement is mainly due to the decrease in the number density of normal electrons. To explore this idea, we have plotted our data as $1/D$ vs temperature (Fig. 2). It will be seen that the scattering mechanism limiting the diffusion above T_c almost disappears at low temperatures. Since $1/D$ is proportional to the scattering probability of the phonons, we assume that the magnitude of curve 1 in Fig. 2 is proportional to the free carrier density. To confirm this, we plot in Fig. 2 the normalized value of $1/D$ within a domain as a function of the reduced temperature T/T_c, together with estimates of the carrier density variation by microwave measurements of the penetration depth.[10] There is good agreement between the two sets of data, which implies that they both show the density variation of the normal carriers. For sake of comparison, the dashed curve in Fig. 2 represents the density of normal carriers computed from a local *s*-wave BCS approximation with $\Delta(0)=3.5kT_c$.[11] It will be seen that experimental results do not agree well with this version of the theory.

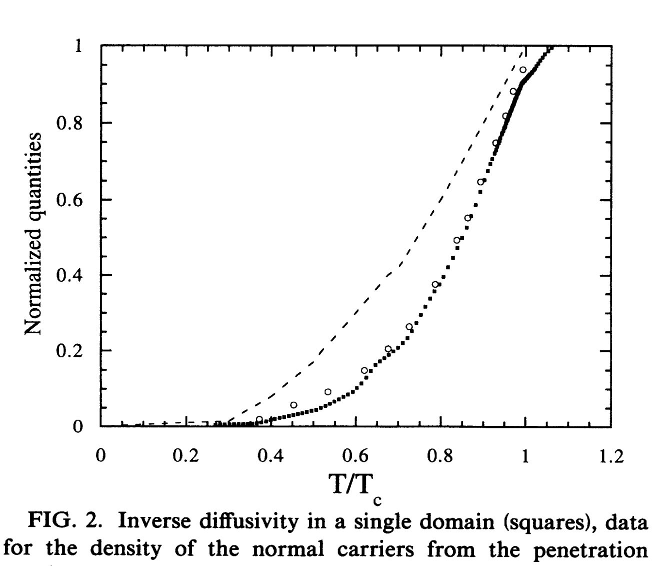

FIG. 2. Inverse diffusivity in a single domain (squares), data for the density of the normal carriers from the penetration depth measurements (Ref. 10) (open curves) and the BCS function (Ref. 11) for the density of the normal carriers (dashed curve).

To explain the weak temperature dependence of diffusivity in a thin film, we suggest that in thin films, an additional mechanism of phonon scattering, probably scattering by point defects, is dominant, which makes the diffusivity insensitive to the superconducting transition. Hence, our results are naturally explained by the traditional model of heat transfer in high-T_c materials as being primarily due to phonons.[1,12,13] If heat transfer was regarded as being due entirely to electrons,[2,14] our data would indicate that, below T_c, the electron mean free path would have to increase over five orders of magnitude to compensate for the rapidly decreasing normal electron density. This is not likely, although some increase of the electron mobility has been reported.[15] The use of diffisivity data rather than conductivity data makes the discussion more straightforward because the heat capacity is not involved and because the inverse of the diffusivity should be approximately proportional to the scattering rate.

BOUNDARY HEAT TRANSFER RESISTANCE

The temperature variation of thermal diffusivity across the domain boundary is similar to the single-crystal domain behavior; a slow temperature dependence is observed above T_c with a steep increase below T_c (Fig. 1). Since the measured diffusion constants are different in the two cases, the twin boundary between the two YBCO crystal domains must create a temperature jump at the boundary. This is demonstrated in Fig. 3, where the inverse diffusivity, which is proportional to the thermal resistance, is plotted against temperature (curve 1). For comparison, the data for the thermal resistance inside the domain is also shown (curve 2). It will be seen that the total resistance (curve 1) is a sum of the bulk resistance (curve 2) and an almost linear function (curve 3), which is naturally identified as the thermal boundary resistance. Suppose that κ, κ_{bo}, and $\bar{\kappa}$ are the bulk, boundary, and average thermal conductivities, respectively, and δ is the boundary thickness. In a one-dimensional model, if the distance between measuring points is $L\sim 10^{-3}$ cm, then the thermal resistance $\bar{R}(T)$ between the two points is

$$\bar{R}(T)=L/\bar{\kappa}=\delta/\kappa_{bo}+L/\kappa \ . \quad (1)$$

Taking $\delta\sim 10^{-7}$ cm, $\kappa\approx 15$ W/m K, and $\bar{\kappa}\approx 7.5$ W/m K, we find that $\kappa_{bo}\sim 10^{-3}$ W/m K, a value that is very small compared to κ.

We conclude that there is a step change in temperature

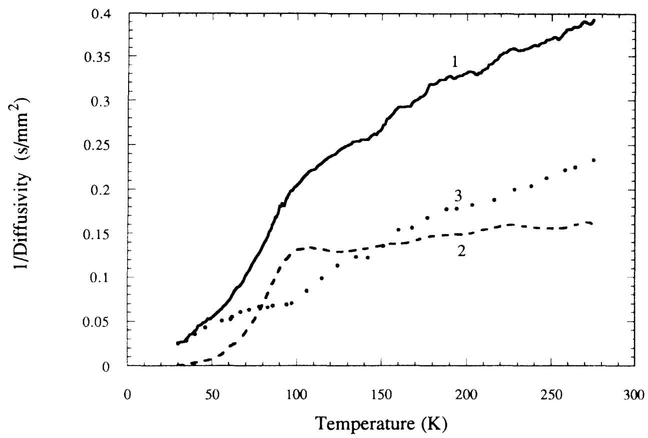

FIG. 3. Inverse diffusivity in a single domain (1) and across the boundary (2) as a function of temperature. Dotted line is a difference between the data sets (1) and (2), presenting the thermal boundary resistance.

at the boundary, and that twin boundaries create a significant obstacle for heat transfer. A similar effect of abnormal thermal resistance on the interface between the thin YBCO film and the MgO substrate was observed earlier in the transient grating measurements,[7,8] where the interface was characterized by a thermal resistance two orders of magnitude larger than the Kapitza jump. A possible explanation of the thermal resistance of the twin boundaries is participation of optical phonons in heat transfer and phonon reflection from the complex boundary structure.

It is instructive to compare data for the boundary thermal and electrical resistances.[16] The electrical resistivity of the boundary is known to exceed the bulk electrical resistivity by several orders of magnitude and to be a linear function of temperature. Since the large thermal resistance of the boundary implies that phonons are primarily reflected there, it is tempting to suggest that heat is transferred through the boundary by electrons. Such as assumption would be consistent with the fact that the normal electrical resistance tends to vary linearly with temperature.

However, electrical resistivity and thermal diffusivity measurements along and across the boundaries were not conducted simultaneously on the same samples, and the only reliable conclusion is that the boundary presents both thermal and electrical resistance. For better understanding of transport through the boundary (and the use of the Wiedemann-Franz Law), further experiments are needed.

CONCLUSION

We observed large thermal diffusivity at low temperatures in a single YBCO domain and across a twin boundary. These measurements became possible because of the application and development of the thermal wave technique, which is capable of probing small volumes of material. It has been shown that diffusivity measurements below the superconducting transition temperature make it possible to determine the temperature dependence of the density of normal carriers. The analysis shows that, over a wide temperature range, this measurement of density agrees with the penetration depth measurements. The predictions for density variation of normal electrons with temperature of the *s*-wave BCS model do not agree well with our experimental results. It is also confirmed that, in agreement with the existing viewpoint, heat is transferred by the phonons inside a YBCO domain. However, a twin boundary creates a high thermal barrier for the phonons.

ACKNOWLEDGMENT

This work was supported under the Department of Energy under Contract No. DOE DE-FG-3-90ER14157.

[1] C. Uber, J. Supercond. **3**, 337 (1990).

[2] R. C. Yu, M. B. Salamon, Jian Ping Lu, and W. C. Lee, Phys. Rev. Lett. **69**, 1431 (1992).

[3] J. L. Cohn, E. F. Skelton, S. A. Wolf, J. Z. Liu, and R. N. Shelton, Phys. Rev. B **45**, 13 144 (1992).

[4] J. T. Fanton, D. B. Mitzi, A. Kapitulnik, B. T. Khuri-Yakub, and G. S. Kino, Appl. Phys. Lett. **55**, 598 (1989).

[5] X. D. Wu, J. G. Fanton, G. S. Kino, S. Ryu, D. B. Mitzi, and A. Kapitulnik, *Photoacoustic and Photothermal Phenomena II*, Proceedings of the 6th International Topical Meeting, Baltimore, MD (Springer-Verlag, Berlin, 1990), pp. 202–4.

[6] X. D. Wu, G. S. Kino, J. T. Fanton, and A. Kapitulnik, Rev. Sci. Instrum. **64**, 3321 (1993).

[7] C. D. Marshall, I. M. Fishman, and M. D. Fayer, Phys. Rev. B **43**, 2696 (1991).

[8] C. D. Marshall, I. M. Fishman, R. C. Dorfman, C. B. Eom, and M. D. Fayer, Phys. Rev. B **45**, 10 009 (1992).

[9] A. Rosencwaig, J. Opsal, W. L. Smith, and D. L. Willenborg, Appl. Phys. Lett. **46**, 1013 (1985).

[10] See, for example, E. J. Nicol and J. P. Carbotte, Phys. Rev. B **43**, 1158 (1991), and references therein.

[11] M. Tinkham, *Introduction to Superconductivity* (McGraw Hill, New York, 1975).

[12] L. Tewordt and Th. Wolkhausen, Solid State Commun. **70**, 839 (1989).

[13] L. Tewordt and Th. Wolkhausen, Solid State Commun. **75**, 515 (1990).

[14] A. S. Alexandrov and N. F. Mott, Phys. Rev. Lett. **71**, 1075 (1993).

[15] D. A. Bonn, P. Dosanjh, R. Liang, and W. N. Hardy, Phys. Rev. Lett. **68**, 2390 (1992).

[16] T. A. Friedmann, J. P. Rice, John Giapintzakis, and D. M. Ginsberg, Phys. Rev. B **39**, 4258 (1988); T. A. Friedmann, M. W. Rabin, J. Giapintzakis, J. P. Rice, and D. M. Ginsberg, *ibid.* **42**, 6217 (1990).