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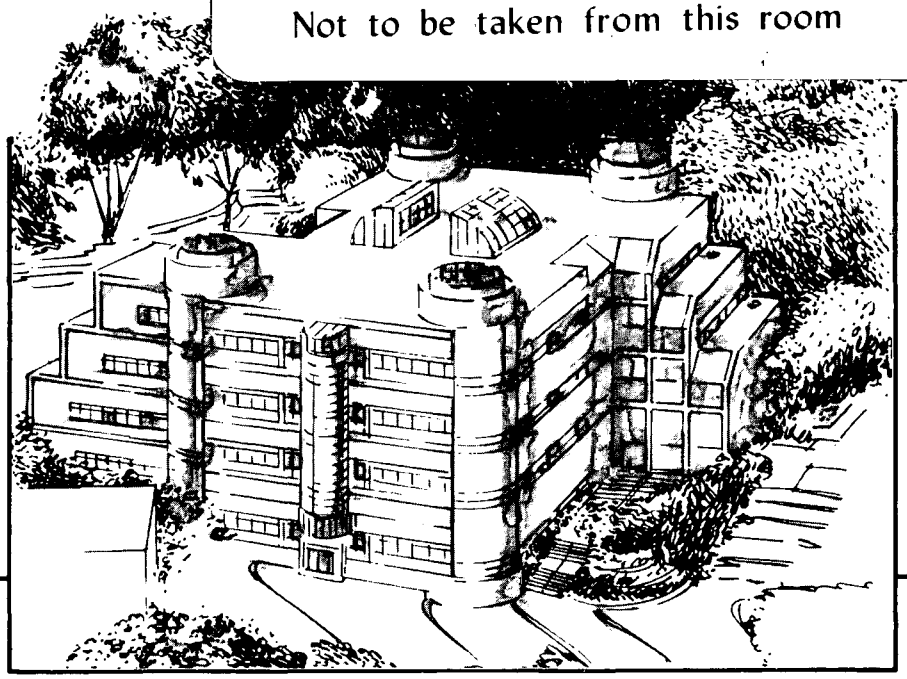

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THERMAL BOUNDARY RESISTANCE FOR $YBa_2Cu_3O_{7-\delta}$ FILMS*

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Abstract

We have made direct measurements of the thermal boundary resistance, R_{bd}, between high quality epitaxial films of $YBa_2Cu_3O_{7-\delta}$ and a variety of substrates with and without buffer layers. The boundary resistance was deduced from measurements of the electrical resistance changes in three parallel strips of $YBa_2Cu_3O_{7-\delta}$ when one was electrically heated. Our measurements indicate that R_{bd} is weakly dependent on temperature and, for all the measured samples, has a value of $0.8\text{-}1.4\times10^{-3}$ Kcm^2/W between 90 K and 200 K, which is a factor of ≈80 larger than the prediction of the acoustic mismatch model at 100 K. The thermal response time of ≈1 ns which results from the heat capacity of the film and R_{bd} is observed in many experiments with pulsed laser sources.

Since the discovery of high T_c superconductivity, much attention has been focused on the response of thin superconducting films to pulsed radiation from visible to far infrared wavelengths. Observations of a variety of response times from femtoseconds to milliseconds with different temperature dependences have caused some confusion about the response mechanisms. Several studies identify a bolometric response, due to heating of the film relative to distant parts of the substrate.[1-3] Others have proposed mechanisms relating to vortex motion,[4,5] phase slips,[6] and non-equilibrium superconductivity[5,7] to account for what is claimed to be a nonbolometric response. Any attempt to model the thermal response, and hence to test the hypothesis of a nonbolometric mechanism, requires a quantitative knowledge of the thermal contact between the film and the substrate.[8] This information has not generally been available.

The thermal boundary resistance R_{bd} is defined as the ratio of the temperature difference ΔT across the interface to the power per unit area $\dot{Q}/A$ flowing across it,

$$R_{bd} = \Delta T A/\dot{Q}. \qquad (1)$$

At temperatures below ≈30 K, the acoustic mismatch model has been very successful in explaining boundary resistances.[9] The main points are that at low temperatures the phonon wavelengths are long (≈400 Å at 1 K for the dominant phonon mode)[10] compared with the characteristic thickness of the interface region so that the bulk material properties and an ideal interface can be used to characterize the heat flow. Phonons are reflected from this interface due to a mismatch in the acoustic impedance across the boundary. The standard analysis yields

$$R_{bd} = B/T^3, \quad (2)$$

where T is the temperature and B is a materials-dependent parameter which depends on integrals over the transmission probabilities of the various acoustic phonon modes and their densities of states. We calculate[11] $B \approx 17\ K^4cm^2/W$ for $YBa_2Cu_3O_{7-\delta}$ on sapphire, a value which is similar to that of other metal/dielectric interfaces.

At higher temperatures, the phonon wavelengths are shorter (≈4 Å at 100 K)[10] and hence the energy transport is very sensitive to interface imperfections and irregularities. As a result the measured R_{bd} does not continue to decrease as T^{-3}, but approaches a constant value at high temperatures. At 100 K R_{bd} can exceed the model predictions by several orders of magnitude. Low temperature physicists are generally familiar with thermal boundary resistances. They play an important role in steady state experiments because they become important compared with bulk thermal conductances in many experimental situations. At higher temperatures, boundary resistances can often be neglected, but not when fast phenomena in thin films are involved.

Our measurements of the thermal boundary resistance use the technique reported by Swartz and Pohl.[12] The experimental geometry, shown in Fig. 1, consisted of three closely spaced, long, narrow strips of $YBa_2Cu_3O_{7-\delta}$ which were patterned using standard photolithographic techniques. A silver fan-out pattern at each end of the strips led to current and potential pads. The dc electrical resistance of each strip was measured as a function of temperature from 90 K to 200 K using a calibrated diode thermometer.[13] The resistances of strips 2 and 3 were measured with a small enough bias that the

temperatures T_2 and T_3 deduced for each strip were essentially equal to the temperatures of the substrate directly underneath the strips. A relatively large bias current was passed through strip 1 causing its temperature T_1 to rise significantly. Both T_1 and the dissipated power $\dot{Q}$ were measured. The temperature T_1' of the substrate beneath strip 1 was calculated from a two dimensional solution of the time independent heat equation which is a good approximation to the experimental geometry.[14]

$$T_1' = T + \dot{Q}(2L\pi\kappa)^{-1}[(x-1)^2 ln(x-1) + (x+1)^2 ln(x+1) - 2x^2 lnx]. \qquad (3)$$

Here $\dot{Q}/L$ is the power per unit length dissipated in strip 1, T is the temperature of the substrate under one of the other strips, κ is the thermal conductivity of the substrate, and x is the ratio of the strip separation s to strip width d. Both T_1' and κ were deduced by using Eq. 3 for the two cases $T=T_2$, $x=s/d$ and $T=T_3$, $x=2s/d$ as is shown in Fig. 1. The boundary resistance was then obtained from Eq. 1 where $\Delta T=T_1-T_1'$.

Epitaxial buffer layers which consisted of 100-500 Å of $SrTiO_3$, $LaAlO_3$, $CaTiO_3$, or MgO, were deposited by laser ablation on $(1\bar{1}02)$ sapphire substrates after which ≈2000 Å epitaxial films of $YBa_2Cu_3O_{7-\delta}$ were deposited *in situ* by laser ablation using deposition parameters that have been discussed elsewhere.[15] The $YBa_2Cu_3O_{7-\delta}$ films on (100) lanthanum aluminate were deposited by the same method but without buffer layers. The films were subsequently ion milled in another chamber to remove ≈500 Å of the $YBa_2Cu_3O_{7-\delta}$ surface and ≈2500 Å of silver was ion beam sputtered *in situ*. The contact pads were patterned using standard photolithography and the silver was etched in a solution of $1H_2O_2$:$1NH_4O_3$:$2CH_3O_4$. The $YBa_2Cu_3O_{7-\delta}$ strips were then similarly defined and etched in ≈0.5% phosphoric acid and

oxygen annealed at 500 °C for 5 minutes. The resulting structure consisted of three strips, each 12 μm wide, 300 μm long and separated by 24 μm. Typical resistances above the resistive transition were ≈250 Ω. The transition widths of several Kelvin were not broadened by patterning. The substrate was mounted on a computer-controlled temperature regulated stage. Low thermal conductance manganin leads were attached with silver epoxy. The experiment was repeated for various values of power $\dot{Q}$. In addition, the effect of thermoelectric voltage offsets was checked by reversing the current direction in strip 1. The thermal conductivity κ deduced for the sapphire substrates agreed with the values reported in the literature.[16] The thermal conductivity of lanthanum aluminate was found to decrease from 0.32 W/cmK at 100 K to 0.23 W/cmK at 170 K. To the best of our knowledge, this information is not available elsewhere.

In Fig. 2 we show the measured temperature dependence of R_{bd} for the various samples. Also shown is the prediction of the acoustic mismatch model for $YBa_2Cu_3O_{7-\delta}$ on sapphire, neglecting the effect of the buffer layers. The thermal boundary resistances of samples a) and b), which had two buffer layers on sapphire, are weakly dependent on temperature while sample c), which had only one buffer layer on sapphire has a smaller R_{bd} which decreases slightly with increasing temperature. The thermal boundary resistance of sample d), which had a lanthanum aluminate substrate without buffer layers, is similar in value to the previous samples, implying that the buffer layers have only a modest effect on R_{bd}. The larger scatter in the R_{bd} data for sample d) is due to the lower thermal conductivity of lanthanum aluminate compared to sapphire.

For our samples the measured values of R_{bd} varied from 0.8×10^{-3} to

1.4×10^{-3} Kcm^2/W, which is almost two orders of magnitude larger than the prediction of the acoustic mismatch model at 100 K. The thermal time constant, $\tau = C/G$ resulting from the heat capacity C of the film and the boundary conductance $G = A/R_{bd}$ for a film of area A, is several nanoseconds for a 500 Å thick film and for our measured values of R_{bd}. The relative insensitivity of R_{bd} to temperature is a common observation in the high temperature regime,[12] where the boundary region is thick compared to characteristic phonon wavelengths. The actual value of R_{bd} depends on the surface quality of the interfaces, as well as the specific details of the deposition. Transmission electron micrographs of these films are not available, but those for similar high quality epitaxial films show a variety of kinds of disorder at the interface which propagate ≈ 100 Å into the film. In addition, disordered layers of order 10 Å thick are frequently seen at interfaces.[17] The thermal conductance of this layer is not known, but would have to be of order $\approx 10^{-5}$ W/cmK to account for R_{bd}. Typical thermal conductivities of polycrystalline $YBa_2Cu_3O_{7-\delta}$ are ≈0.03 W/cmK at 100 K,[18] which are too low to account for R_{bd}. Carr et al.[1] measured the response of epitaxial films of $YBa_2Cu_3O_{7-\delta}$ on MgO to pulsed, broadband infrared radiation, as a function of thickness. They inferred a value of $R_{bd} \approx 1.1 \times 10^{-3}$ Kcm^2/W at 90 K, which is within our measured range. Marshal et al. inferred a value of $R_{bd} \approx 2.5 \times 10^{-3}$ Kcm^2/W for epitaxial films of $YBa_2Cu_3O_{7-\delta}$ on MgO at 300 K using the optical-transient grating method.[19]

Typical measurements of the response of thin $YBa_2Cu_3O_{7-\delta}$ films to pulsed laser radiation use a narrow $YBa_2Cu_3O_{7-\delta}$ bridge deposited on a dielectric substrate, the bridge is biased at a constant current and the voltage change due to the incident radiation is monitored. A speed of response on the order of several ns is commonly reported.[1,5,6,7,20,21] In addition, several

authors have reported a discrepancy between the temperature dependence of the observed response and that of dR(T)/dT,[4,5] where R(T) is the temperature dependent resistance of the film. We now consider whether the measured values for R_{bd} are able to account for these observations. The most thorough thermal analysis of such an experiment is that of Flick et al,[8] who analyzed the data of Frenkel et al.[5] In the absence of specific data they modeled R_{bd} with the acoustic mismatch model using B=40 K^4cm^2/W. They concluded that the temperature profile through the film is essentially flat even for pulses as short as 150 ps, and that most of the temperature drop occurs across the film/substrate interface rather than through the substrate. These conclusions are still valid for the larger values of R_{bd} that we measure. Realizing that the acoustic mismatch model underestimates R_{bd} above ≈ 60 K, Flick et al. calculate the effect of replacing it by a constant value of 0.2×10^{-3} Kcm^2/W, which was measured for a Rh:Fe/sapphire interface.[12] While the overall agreement between the calculations and the data was improved, the observed response could not be explained by the thermal hypothesis. Hence they concluded that the discrepancy is due to a nonbolometric mechanism. The values of R_{bd} that we have measured are a factor ≈ 7 larger than the largest values assumed by Flick et al. Consequently, the agreement between the analysis and the data will improve, thus weakening the evidence for a nonbolometric response on the ns time scale.

The various response times that are observed should be understood in the context of a hierarchy of relaxation processes which can take place after a sudden deposition of energy in a high T_c film. Although many details of these processes are not clear, the following phenomena probably occur. In general, hot quasiparticles relax to large numbers of "gap edge" quasiparticles and phonons of comparable energy in hundreds of femtoseconds.[22,23] "Gap

edge" quasiparticles cannot recombine until energy leaves the excited region. Depending on the size of this region, the energy can dissipate in picoseconds by diffusion of quasiparticles and phonons through the film or, as shown in this work, phonons can carry energy across the boundary in nanoseconds. Heat spreads out into the substrate in microseconds,[24] and into the heat sink in milliseconds. The complexity of these essentially thermal processes is a serious impediment to the unambiguous identification of non-thermal detection mechanisms.

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Figure Captions

Fig. 1. Experimental configuration. The interface studied lies beneath the left strip, which is biased with a relatively high current I.

Fig. 2. Thermal boundary resistance for thin films of $YBa_2Cu_3O_{7-\delta}$ on sapphire with, a) 100 Å $SrTiO_3$/100 Å MgO, b) 200 Å $LaAlO_3$/100 Å $CaTiO_3$, and c) 500 Å $CaTiO_3$ buffer layers. The dashed line is the prediction of the acoustic mismatch model (AMM) for $YBa_2Cu_3O_{7-\delta}$ on sapphire without buffer layers. The thermal boundary resistance for $YBa_2Cu_3O_{7-\delta}$ on lanthanum aluminate is shown in d).

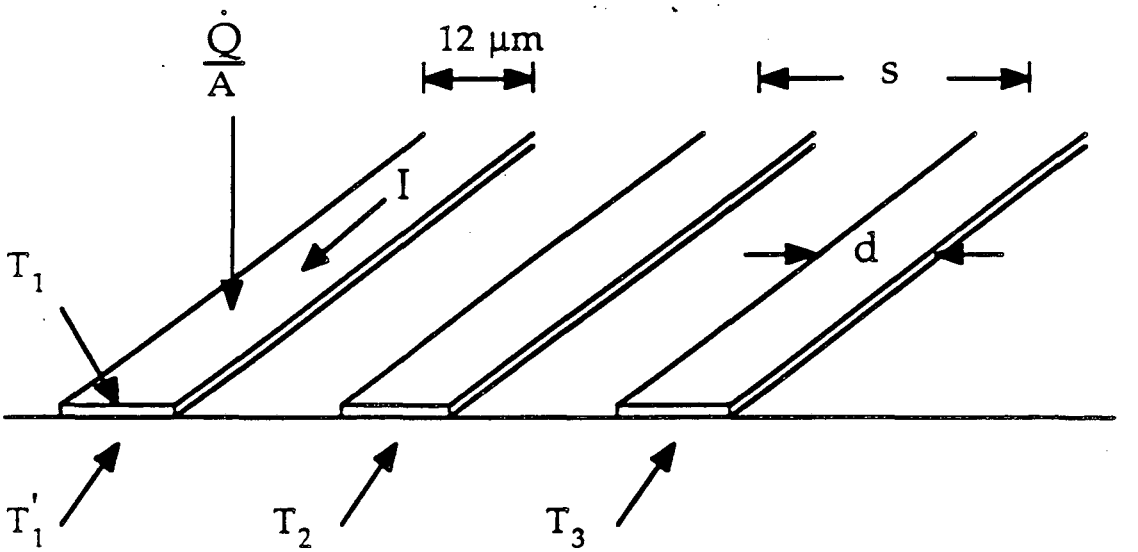

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FIGURE 1

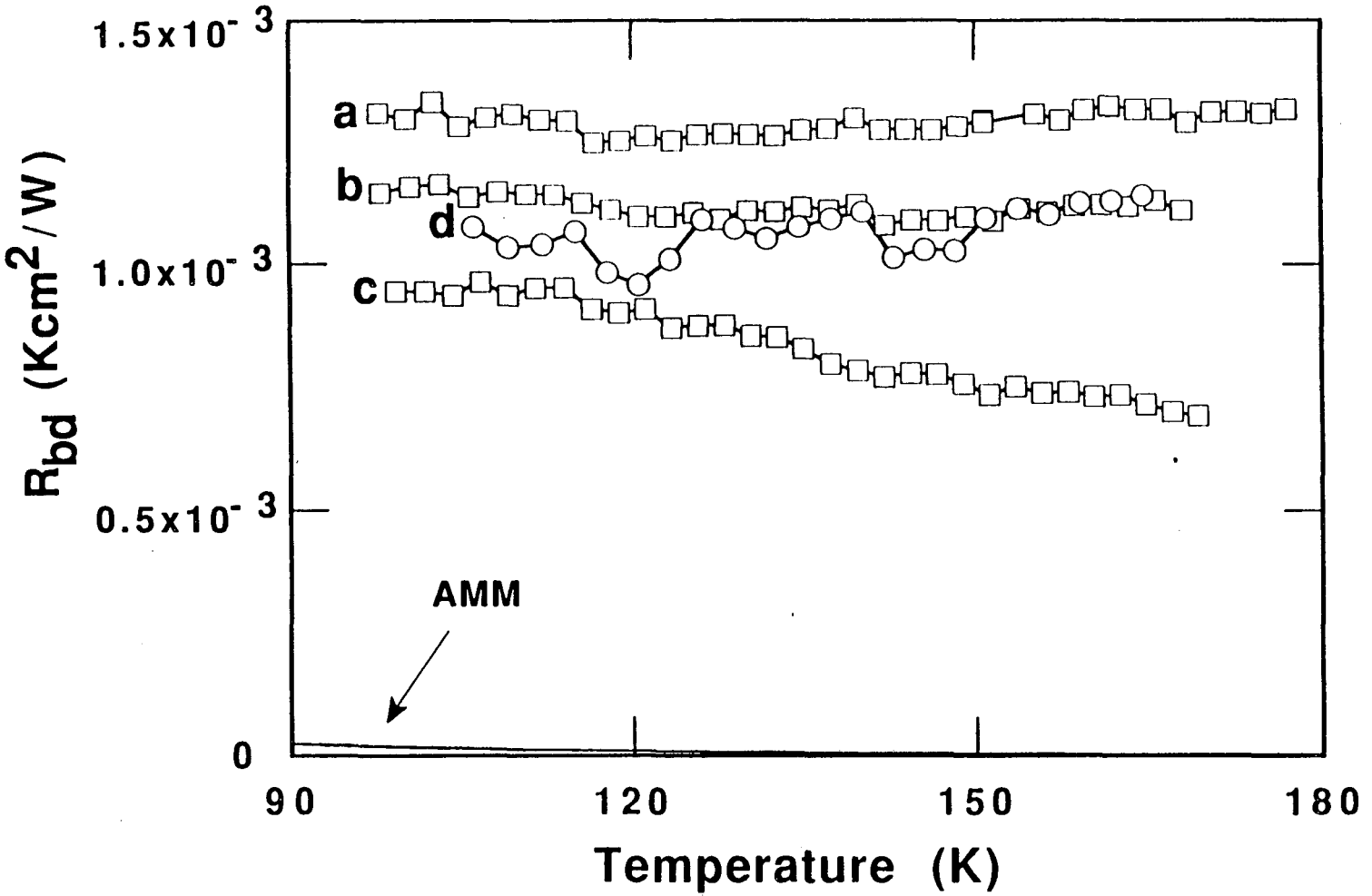

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FIGURE 2

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