Heat propagation in high T_c films investigated by optical response measurements

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(Received 2 March 1992; accepted for publication 13 June 1992)

The optical response of granular Tl-Ba-Ca-Cu-O films has been used to investigate thermal properties of the films. An analysis of the response using a heat transfer model yields a thermal diffusivity $D=10^{-3}$ cm^2/s at 150 K which rises to 6×10^{-3} cm^2/s at a temperature of 30 K and allows for an estimation of the boundary resistance $R_{bd}\approx10^{-3}$ K cm^2/W between film and substrate. The dependence of the response time on film thickness obtained from the heat transfer model is compared with published data indicating that in many experiments the observed response is mainly bolometric in origin.

Since it has become possible to prepare high T_c superconducting films, much attention has been focused on their optical response, where films are irradiated with pulsed radiation (visible to far infrared) and the transient resistance change of the films is measured. Response times on a remarkably large time scale between milliseconds and picoseconds have been found meanwhile, and were attributed to a variety of different physical mechanisms.[1–14] Irradiation induced heat production within the films leads to a bolometric response due to the temperature dependent film resistance. In this case the response time is governed by film cooling due to heat diffusion into the substrate. It is the purpose of this letter to demonstrate, that an analysis of the resistance response of a Tl-Ba-Ca-Cu-O film can be used to investigate the thermal diffusivity of the film as well as the thermal boundary resistance of the film-substrate interface. Furthermore, a comparison of published data with response times obtained from the heat transfer model allows a clear separation of bolometric and nonbolometric responses.

The measurements were carried out on polycrystalline Tl-Ba-Ca-Cu-O films grown on MgO substrates by a laser ablation technique.[15] The films were patterned by means of an excimer laser into 200-μm-wide stripes with contact pads on both ends. Thin copper wires were attached using silver epoxy paint and the stripes were biased using a constant current source (see the inset of Fig. 2). The samples were mounted in a temperature variable optical cryostat and irradiated with short pulses from an atmospheric pressure CO_2 laser (pulse duration $\approx$80 ns, wavelength $\approx$10 μm).

The dependence of the resistance R on temperature T of a film of 1 μm thickness is shown in Fig. 1 for several bias currents. A sufficiently high current leads to nonzero dR/dT even at low temperatures, allowing measurements of the bolometric signal well below T_c. Response measurements at two temperatures are shown in Fig. 2 for this film. With a laser pulse energy density $\approx$3 mJ/cm^2 the maximum resistance change was several ohms at 120 K indicating an overall temperature change of the film of the order of 5 K. At 50 K, due to the smaller value of dR/dT, the resistance change was 0.5 Ω. Comparison of the measurements clearly shows a shorter resistance recovery time at low temperature due to faster heat diffusion.

By use of a heat transfer model thermal properties of the high T_c material can be extracted from the response measurements. As an approximate description of our experiment we adapt a model of heat diffusion in a thin slab[16] with thermal diffusivity D and thickness d. The film is assumed to be thermally isolated at the laser heated film surface and to have a thermal boundary resistance $R_{bd}=1/H_{bd}$ to the substrate, where H_{bd} is the boundary conductance. Relatively high values $R_{bd}\approx10^{-3}$ K cm^2/W have been found recently[1,17] for $YBa_2Cu_3O_{7-\delta}$ films on several substrates. The substrate temperature in the above model may be assumed to be constant because the thermal conductivity of the substrate material is much higher than that of the high T_c film.

Assuming instantaneous surface heating by the laser source due to absorption of energy E per unit area within a penetration depth δ of the radiation the temperature rise in the film at time t and distance x from the heated surface is[16]

$$T(x,t)=\frac{2E}{C}\sum_{n=1}^{\infty}\exp(-\alpha_n^2Dt)\frac{(h^2+\alpha_n^2)\cos\alpha_n x}{(h^2+\alpha_n^2)d+h}\times\int_0^d\frac{1}{\delta}\exp(-x/\delta)\cos\alpha_n x\,dx. \quad (1)$$

In this equation, α_n are the roots of

$$\alpha\tan(\alpha d)=h, \quad (2)$$

where $h=H_{bd}/k$, k is the thermal conductivity, and C the specific heat per unit volume of the film.

In the fits to the experimental data we used $E_p=3$ mJ/cm^2, $\delta=100$ nm, an absorption of 30% leading to $E=0.3\,E_p\approx1$ mJ/cm^2 and values of C from Junod *et al.*[18]

From the calculated temperatures [Eq. (1)] and the measured temperature dependence of the resistivity, the time dependent total film resistance $R(t)$ is obtained by modeling the film as m layers with temperature $T_i(t)$ and resistance $R_i(t)$, $i=1,\ldots,m$. A value of 20 for m has been found to give sufficient accuracy. We note that for thick films with thermal resistance $R_f=d/k>R_{bd}$ the boundary resistance has a negligible influence and can be omitted,

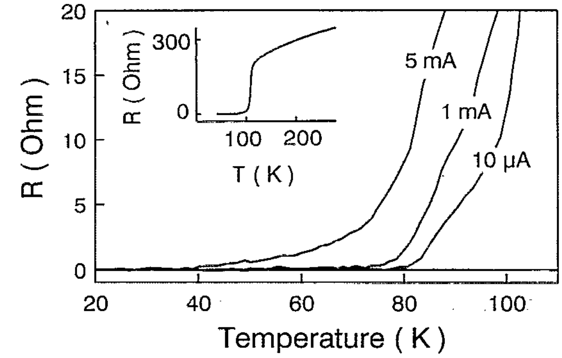

FIG. 1. Resistance of a Tl-Ba-Ca-Cu-O film of 1-μm thickness vs temperature (inset). By increased bias currents, nonzero dR/dT can be obtained at low temperatures.

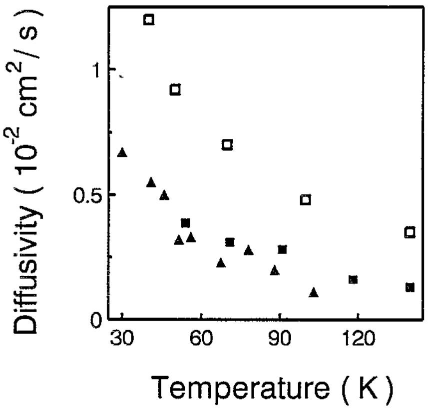

FIG. 3. Diffusivity obtained from fits $R(t)$ to the experimental curves for a thick film (d=1 μm, full squares) and for a thin film (d=400 nm, full triangles). Values for sintered $Tl_2Ba_2Ca_2Cu_3O_{10}$ are shown for comparison (open squares).

i.e., $H_{bd}=\infty$ and the diffusivity D is directly obtained choosing the best fit $R(t)$ to the response curve (Fig. 2). If D is known, a fit of $R(t)$ to a response measurement on a thin film can be used for an estimation of R_{bd}.

In Fig. 3, the diffusivity D, obtained from fits to response measurements on films with thickness $d=1$ μm and d=400 nm, from low to high temperatures, are shown. A consistent description $D(T)$ for both films is found with $R_{bd}=(1\pm0.5)\times10^{-3}$ K cm^2/W which is similar to values obtained in recent measurements on $YBa_2Cu_3O_{7-\delta}$ films.[1,17] Values of $D(T)$ calculated from thermal conductivity and specific heat measurements on sintered Tl-Ba-Ca-Cu-O samples[18,19] are also shown, for comparison. Measurements on sintered $YBa_2Cu_3O_{7-\delta}$ samples show diffusivities larger by a factor 3 to 5, depending on sample.[20]

Thus, from the heat transfer model the characteristic response time for bolometric response can be deduced. Qualitatively, for thick films the response time is due to the diffusion time $\tau\sim d^2/D$ for heat propagation through the film. For thin films a nearly homogeneous temperature distribution through the entire film is reached quickly and the film cooling is governed by heat flow through the film-substrate interface, leading to a response time proportional to the film heat capacity, i.e., $\tau=R_{bd}\ C\ d$. The diffusion time $\tau(d)$ has been calculated from the heat transfer model and the temperature dependent film resistivity using typical values for the diffusivity ($D=10^{-2}$ cm^2/s) and the thermal conductivity (k=0.02 W/cm K). This curve (see Fig. 4) shows the expected behavior $\tau\propto d$ for thin films ($R_{bd}>R_f$) and $\tau\propto d^2$ for thick films. In addition, values τ_{exp} taken from published optical response experiments (Table I) are displayed. We suggest that in experiments where τ_{exp} is near our calculated curve a mainly bolometric response is observed. On the other hand, the fast response observed on thick films using far infrared radiation is clearly of nonbolometric origin. Recently, a fast response has also been observed for relatively thin films, with visible to near IR radiation,[6,10] where response times in the psec time regime have been observed. Such a timescale may indicate a nonbolometric response. We would like to point out, however, that a fast response ($\tau\sim$200 ps) followed by

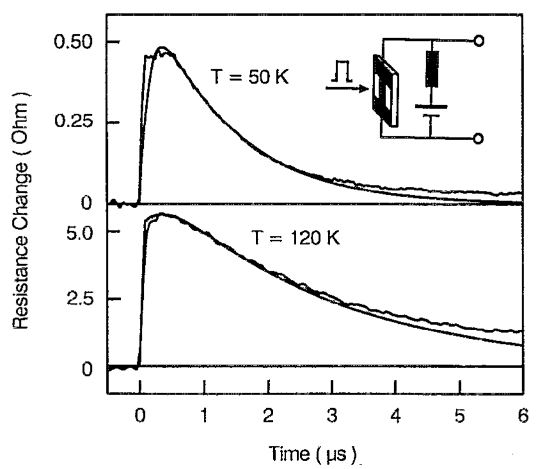

FIG. 2. Experimental arrangement (inset) and optical response at two temperatures, together with calculated curves $R(t)$ from the heat transfer model.

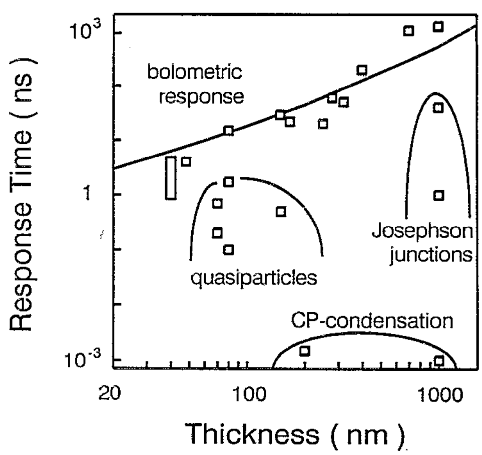

FIG. 4. Calculated response time $\tau(d)$ (line) in comparison with published response time data (squares). The experiments are grouped for the eye according to the suggested response mechanisms. Processes in the picosecond time domain were attributed to Cooperpair (CP) condensation.

TABLE I. Summary of experimental response times.

Material	Thickness (nm)	λ (μm)	τ_{exp}	Reference
YBCO	40	1	1–4 ns	7
YBCO	48, 168, 320	1–100	4, 22, 50 ns	1
YBCO	70	1	0.2, 0.7 ns	10
YBCO	80	0.53	~15 ns	5
YBCO	80	0.66	~100 ps, 1.7 ns	6
BPBO	150	1–8	0.5 ns	9
TBCCO	150	0.53	~30 ns	14
YBCO	200	0.63	0.3, 1.5 ps	12
YBCO	250	0.58	~20 ns	2
YBCO	280	1	~60 ns	3
YBCO	700	1	~1 μs	8
YBCO	1000	500	40 ns	4
TBCCO	1000	0.61	0.5, ~1 ps	13
TBCCO	1000	385	~1 ns	11
TBCCO	400, 1000	10	200 ns, 1.2 μs	This work

a slow decay of the signal (~ns) for a 50 nm film can easily be obtained within our model if the temperature dependence of the resistance is highly nonlinear, e.g., if the film surface is heated up to T_c.

In summary, we have analyzed optical response measurements with a heat transfer model to extract the thermal diffusivity of high T_c superconducting films and to estimate the boundary resistance between film and substrate. The model yields a classification scheme for pulsed response experiments with respect to the character of the response.

The work was supported by the Deutsche Forschungsgemeinschaft (H.L.), the Bundesministerium für Forschung und Technologie, and the Bayerischer Forschungsverbund Hochtemperatursupraleitung (FORSUPRA) (K.F.R.), and the European Communities (W.P.).

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