Transparency of a $YBa_2Cu_3O_7$-film/substrate interface for thermal phonons measured by means of voltage response to radiation

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The transparency of a film/substrate interface for thermal phonons was investigated for $YBa_2Cu_3O_7$ thin films deposited on MgO, Al_2O_3, $LaAlO_3$, $NdGaO_3$, and ZrO_2 substrates. Both voltage response to pulsed-visible and to continuously modulated far-infrared radiation show two regimes of heat escape from the film to the substrate. That one dominated by the thermal boundary resistance at the film/substrate interface provides an initial exponential decay of the response. The other one prevailing at longer times or smaller modulation frequencies causes much slower decay and is governed by phonon diffusion in the substrate. The transparency of the boundary for phonons incident from the film on the substrate and also from the substrate on the film was determined separately from the characteristic time of the exponential decay and from the time at which one regime was changed to the other. Taking into account the specific heat of optical phonons and the temperature dependence of the group velocity of acoustic phonons, we show that the body of experimental data agrees with acoustic mismatch theory rather than with the model that assumes strong diffusive scattering of phonons at the interface.

I. INTRODUCTION

Thermal boundary resistance at the interface between a high-temperature superconducting film and a substrate has recently attracted considerable attention. Much interest has sprung from numerous possible applications of the fast bolometric response of high-temperature superconducting films to electromagnetic radiation. Based on the response time scale, recent investigations clearly separated bolometric and nonequilibrium responses.[1-5] However, the physical mechanism of the phonon transport across the film/substrate interface is still under debate. This problem is not limited to the question of thermal contact between high-T_c film and substrate. Various solid-solid interfaces have also been intensively studied.[6,7] In this paper we present the results of systematic investigations of the phonon transparency of $YBa_2Cu_3O_7$-film/substrate interfaces that give an excellent tool to test models claiming to describe the phonon transport across the boundary. Thorough investigation of the thermal boundary resistance could also yield valuable information on the phonon system in high-temperature superconductors.

Two principal methods are used to determine the thermal boundary resistance R_{bd}. It can be deduced from the characteristic time of the voltage response to pulsed or chopped irradiation.[8] Originally, nanosecond response times were attributed to nonequilibrium effects in superconductors.[9] Later on it was demonstrated that the characteristic time of nonequilibrium response is essentially shorter.[1-3] Carr *et al.*[5] found that the nanosecond response time is proportional to the film thickness and, therefore, is rather caused by the thermal boundary resistance. In contrast, the heat diffusion in the film provides a characteristic response time proportional to the square root of the film thickness.[8,10] Response times for films on various substrates with or without buffer layers have been measured recently by many authors. The results reviewed in Ref. 8 indicate that the ratio of the response time to the film thickness slightly depends on the substrate material. Values of R_{bd} lie in the interval 8×10^{-4}–3×10^{-3} K cm^2 W^{-1}.

Direct measurements of the thermal boundary resistance have been performed by means of the stationary method.[8,11] In this approach the temperature of a film is controlled by its electrical resistance. Commonly, several strips on the same substrate are used. One of them is electrically heated while the others are used as thermometers monitoring substrate temperature. The temperature field in the substrate is then calculated to deduce the Kapitza temperature step at the interface between the heated strip and the substrate. The value of $R_{bd}\approx10^3$ K cm^2 W^{-1} appeared to be relatively independent of temperature.[12] For Er-Ba-Cu-O films on a MgO substrate, R_{bd} was found[8] to be about two order of magnitude larger than for $YBa_2Cu_3O_7$ films.[11] The main conclusion of all the aforementioned papers, excluding that of Bluzer,[4] is that experimental values of R_{bd} are too large to be explained by the acoustic mismatch theory.[13]

In this paper we further study[1] the fast bolometric response of $YBa_2Cu_3O_7$ thin films on various substrates to far-infrared and visible radiation. The observed transition from heat escape governed by the localized thermal boundary resistance to diffusion heat propagation in the substrate enables us to determine independently transmission coefficients for phonons crossing the boundary from the side of the film and from the side of the substrate. Taking into account a more realistic description of the phonon system in $YBa_2Cu_3O_7$ we obtain values of the transparency which do not contradict the acoustic mismatch theory.

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II. THEORETICAL CONCEPTS

It is well known that the diffusion model of phonon transport does not hold if the times of interest are short compared with the phonon scattering time τ_{ph} or if the characteristic distances are smaller than the phonon mean free path l_{ph}. An interface with the phonon transmission coefficient α modifies spatial and temporal scales as follows:

$$\tau_R = \frac{\tau_{ph}}{\alpha^2}, \quad l_R = \sqrt{D\tau_R} = \frac{l_{ph}}{\alpha}. \tag{1}$$

Here τ_R is the time a phonon, located within the distance l_{ph} from the interface, needs to cross the boundary, l_R is the diffusion length corresponding to τ_R, and D is the phonon diffusion coefficient. Referring to a film on a substrate, τ_R simply denotes the time after which a phonon, once having escaped from the film, may return. The diffusion model, involving the Fourier equations for the temperature in the film and in the substrate with the continuity condition at the interface, is applicable for times and distances large than τ_R and l_R respectively. Relaxation of the film temperature is then governed by phonon diffusion into the substrate. For smaller times and distances the regime of film cooling is determined by the thermal boundary resistance.

Let us consider the thermal-boundary-resistance regime in more detail. At liquid-nitrogen temperatures, the film thickness is commonly greater than $l_{ph,f}$, which causes diffusion motion of phonons in the film (hereafter, subscripts s and f refer to the substrate and to the film respectively). Nevertheless, for a film with thickness $d < l_{ph,f}/\alpha$ the temperature is uniform through the film thickness and the time of phonon escape from the film can be written in the form

$$\tau_{es}^{-1} = \frac{\sum_i c_{i,f} u_{i,f} \int_0^{\pi/2} \alpha_{i,f}(\theta) \cos\theta \sin\theta \, d\theta}{2d \sum_i c_{i,f}} = \alpha_{f-s} \frac{\sum_i c_{i,f} u_{i,f}}{4d \sum_i c_{i,f}}, \tag{2}$$

which is quite similar to those for ballistic-phonon propagation in the film ($d < l_{ph,f}$). Here the summation is performed over all phonon modes of the film, $c_{i,f}$ is the specific heat, $u_{i,f}$ is the average group velocity, and $\alpha_{i,f}(\theta)$ is the transparency of the boundary for a phonon incident on the boundary from the film at the angle θ. The last term in (2) is a definition of the average transparency coefficient α_{f-s} for phonons incident from the film on the substrate. In contrast to the definition $\tau_{es}^{-1} = \alpha_{f-s} u (4d)^{-1}$ valid at low temperatures (u is the speed of sound), Eq. (2) takes account of the group velocities of different phonon modes.

Return of nonequilibrium phonons from the substrate back to the film is negligible for times $t < \tau_R = \tau_{ph,s}/\alpha_{s-f}^2$ after the beginning of relaxation. Here α_{s-f} is the average transparency coefficient for phonons incident from the substrate on the film. Within this interval the temperature of the film relaxes exponentially with the characteristic time τ_{es} towards the equilibrium substrate temperature. The thermal boundary resistance R_{bd} may be then expressed as follows

$$R_{bd} = \frac{4}{\alpha_{f-s} \sum_i c_{i,f} u_{i,f}} = \frac{\tau_{es}}{d c_f}, \tag{3}$$

where c_f is the phonon specific heat of the film. To satisfy the balance of heat flow across the boundary, the average transparency coefficients α_{f-s} and α_{s-f} should be connected by the equation

$$\alpha_{f-s} \sum_i c_{i,f} u_{i,f} = \alpha_{s-f} \sum_i c_{i,s} u_{i,s}. \tag{4}$$

The above analysis [Eqs. (1)–(4)] is valid for thermal equilibrium as well as within the linear approximation for the temperature discontinuity at the interface $\Delta T \ll T$, where T is the equilibrium temperature. This condition is always fulfilled in our measurements.

In the stationary approach, the return of nonequilibrium phonons from the substrate can be neglected only for short and narrow strips with all in-plane dimensions smaller than $l_{ph,s}/\alpha_{s-f}$. Hence, for the film the substrate may be considered as a heat sink with the temperature measured at the distance $L > l_{ph,s}/\alpha_{s-f}$ from the film. To determine R_{bd} (or α) this configuration is preferable to that reported in Refs. 8,11, as it does not require calculations of the temperature field in the substrate.

III. EXPERIMENTAL PROCEDURE

Films were deposited by laser evaporation and magnetron sputtering on different substrates (MgO, Al_2O_3, $LaAlO_3$, $NdGaO_3$, and ZrO_2). Samples (see Table I) had transition temperatures from 86 to 89 K and represented microstrip transmission lines patterned within the 2-mm length into a structure of parallel strips separated by blanks with the same width. Constantly biased samples were heated up by pulsed or cw irradiation. Voltage change across the sample was measured as a function of real time or modulation frequency. As a pulsed radiation source we used a Nd-YAG laser (YAG is yttrium aluminum garnet) with the wavelength $\lambda = 0.63$ μm and a pulse duration of 80 ps. Transient voltage response was monitored with a temporal resolution of 100 ps. Modulation characteristics were measured with a cw low-pressure

TABLE I. Investigated samples and characteristic times τ_b of exponential transient decay; W is the strip width.

No.	Substrate	d (nm)	W (μm)	τ_b(90 K) (ns)
1	MgO	50±5	2	2.3±0.2
2	MgO	70±5	30	3.2±0.5
3	MgO	130±10	30	7.0±1
4	Al_2O_3	45±3	8	4.5±0.5
5	Al_2O_3	65±5	3	6.5±1
6	$LaAlO_3$	60	20	6.2±0.2
7	$NdGaO_3$	80	3	5.5±0.5
8	ZrO_2	50	4	75[a]

[a]Time at which the signal drops to half of its initial value.

CO_2 laser ($\lambda=10$ μm) and an acousto-optical modulator. Amplitude-modulation frequency up to $f\simeq 100$ MHz was available. Signals were monitored by a spectrum analyzer. Samples were mounted on a copper block at the bottom part of a sample holder inserted into a storage helium Dewar. Temperature was regulated by lowering the holder into the Dewar. Signals from the sample were recovered via semirigid 50-Ω coaxial cable. A bias-T mounted at the top of the holder allowed us to bias the sample and to monitor its dc resistance.

IV. EXPERIMENTAL RESULTS

Examples of the pulsed-response signals measured at $T=90$ K for films on different substrates are shown in Fig. 1. The characteristic times τ_b of the fast-signal decay obtained from the best exponential fit $\exp(-t/\tau_b)$ are listed in Table I. We chose the radiation energy density (less than 20 μJ cm^{-2} per pulse) and the bias current density (less than 10^4 A cm^{-2}) to remain within the linear regime, i.e., where characteristic times do not change with either radiation density or current density. The maximum temperature change of a film calculated from the signal amplitude and temperature dependence of the resistance never exceeded 0.2 K. For samples on $NdGaO_3$, $LaAlO_3$, and MgO substrates an exponential decay of the signal is followed by a slower tail corresponding to the power law $t^{-1/2}$. The crossover time from an exponential to a power-law approximation is 15±2 ns for $NdGaO_3$ (sample no. 7), 10±1 ns for $LaAlO_3$ (sample no. 6), and 65±20 ns for MgO (sample no. 3). These times are counted from the beginning of the exponential decay. For samples on Al_2O_3 substrates we did not find any power-law relaxation within the 100-ns time window [Fig. 1(a)]. In contrast, the film on the ZrO_2 substrate showed only slow power-law relaxation [Fig. 1(d)]. In Fig. 2, τ_b for sample no. 5 is plotted as a function of temperature, together with the transition curve measured

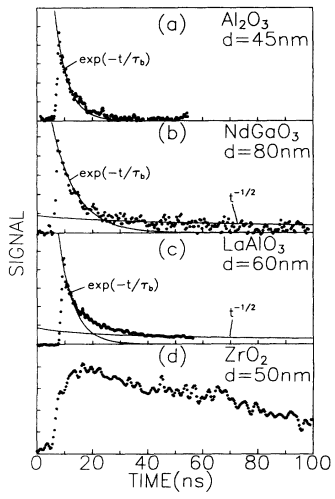

FIG. 1. Examples of photoresponse signals for several samples at $T=90$ K. Solid lines represent exponential and power-law approximations.

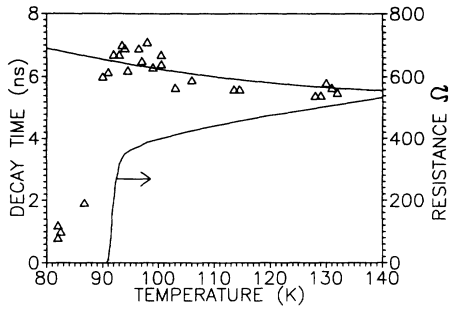

FIG. 2. Resistance and exponential decay time (open triangles) for sample no. 5 plotted as functions of temperature. The solid line is a guide to the eye for the $\tau_b(T)$ dependence above T_c.

at the same bias current. In the transition region and at higher temperatures for all our samples we did not find any changes in the shape of the response signals. Although τ_b slightly decreases with temperature, it drops drastically down to about 1 ns when the resistance becomes zero. This result is typical for all samples independent of substrate. We believe the drop in the decay time can be related to the superconducting state of a film. It is most likely connected with the kinetic inductance response described in Ref. 4. Therefore, in further discussion we use values of τ_b measured in the normal state just above the transition.

Data obtained with the modulation technique are shown in Fig. 3. For sample no. 5 on Al_2O_3 [Fig. 3(a)], there are three different regions in the modulation-frequency dependence of the response. At low frequencies the signal gradually decreases according to the law f^{-m} with the factor m between $\frac{1}{2}$ and $\frac{1}{3}$. After a plateau at intermediate frequencies, a knee appears, which is followed by $1/f$ decrease of the signal. We found the knee and the $1/f$ slope perfectly described by the single-time relaxation formula $U\propto[1+(2\pi f\tau)^2]^{-1/2}$. The characteristic times τ extracted from the fitting procedure are in accordance with the time τ_b obtained from pulse measurements. The crossover from f^{-m} behavior to the plateau at intermediate frequencies occurs at the frequency $f_0\approx 450$ kHz, which corresponds to the time $t=(2\pi f_0)^{-1}\approx 350$ ns. Sample no. 7 on the $NdGaO_3$ substrate has a rather smooth modulation characteristic [Fig. 3(b)]. Nevertheless, we found this curve coinciding

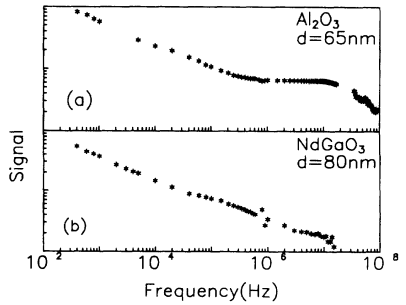

FIG. 3. Modulation-frequency dependencies of the response for samples nos. 5 and 7.

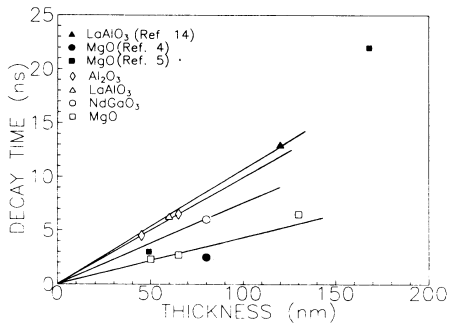

FIG. 4. Summary of exponential decay times for different film thicknesses and for different substrates. Solid lines indicate the linear fit $\tau_b \propto d$.

with the Fourier transformation of the decreasing part of the pulse signal [Fig. 1(b)]. In Fig. 4 we summarize results on decay times τ_b measured at $T=90$ K including published data for similar films.[4,5,14] It is seen that for films with $d<150$ nm on Al_2O_3, $LaAlO_3$, and MgO substrates τ_b is practically proportional to the film thickness.

V. DISCUSSION

In the following we will evaluate values of $\alpha_{f\text{-}s}$ and $\alpha_{s\text{-}f}$ in the framework of a simple phonon model, taking account of optical-phonon modes and the decrease of the group velocity of acoustic phonons at high temperatures. Exponential decay of signals (Fig. 1) with the characteristic time τ_b proportional to the film thickness (Fig. 4) gives strong evidence that at the initial stage of temperature relaxation for all substrates, excluding ZrO_2, the heat escape from the film is governed by the thermal boundary resistance of the film-substrate interface. Hence, τ_b can be identified with the time of phonon escape from the film. This allows us to calculate the thermal boundary resistance R_{bd} using Eq. (4). With $c_f=0.9$ J cm^{-3}K^{-1} (Ref. 15), we get values $R_{bd}(90\text{ K})$ which are listed in Table II. The weak temperature variation of τ_b above T_c leads to a temperature dependence of R_{bd} that is quite similar to that reported in Ref. 11.

To describe acoustic phonons in $YBa_2Cu_3O_4$ films we use the nearest-neighbor force model with three sine-shaped dispersion curves and with the cutoff frequency ν_0 corresponding to the Debye temperature $\Theta_f=500$ K.[12,16] For $T=90$ K the model gives the average group velocity of acoustic phonons, which is by a factor $a(T/\Theta_f)=0.49$ smaller than the speed of sound in $YBa_2Cu_3O_7$, $u_f=3.5\times10^5$ cm s^{-1}.[17] At this temperature some optical phonons are already active, but their group velocity is negligible compared to the group velocity of the acoustic phonons which are mostly responsible for heat transport across the boundary. Several research groups[12,16] have estimated the optical-phonon contribution to the $YBa_2Cu_3O_7$ specific heat c_f, varying between $0.35c_f$ and $0.6c_f$ at $T=T_c$. Using the average value $0.5c_f$ and the reduced group velocity we deduce from Eq. (2)

$$\alpha_{f\text{-}s}=\frac{8d}{u\tau_{es}a(T/\Theta_f)}\ . \tag{5}$$

Calculated with Eq. (5), values of $\alpha_{f\text{-}s}$ lie in the interval 0.04–0.1 for all our substrates (Table II).

We attribute the $t^{-1/2}$ tail of the response signals to diffusion phonon propagation in the substrate.[18] Consistently, the crossover time from the $\exp(t/\tau_b)$ to the $t^{-1/2}$ behavior [Fig. 1(b),(c)] is identified with the time τ_R of phonon return to the film. In the modulation curve shown in Fig. 3(a), this time corresponds to the inverse of the characteristic frequency at which the transition from the f^{-m} decrease to the plateau occurs. To obtain the transparency coefficients $\alpha_{s\text{-}f}$ from values of τ_R, we first evaluate phonon scattering times in our substrates $\tau_{ph,s}=3k_s[b(T/\Theta_s)c_s u_s^2]^{-1}$. The decrease of the group velocity of acoustic phonons is taken into account here by the factor $b(T/\theta_s)$, which is calculated in the framework of the model we used for acoustic phonons in the film. This coefficient simply represents the ratio of the average squared group velocity to the squared speed of sound. For substrates we neglect optical phonons and use acoustic-phonon cutoff frequencies ν_0 corresponding to their Debye temperatures. With the thermophysical properties listed in Table II we found $\tau_{ph,s}$ ranging from 1.4 ps for ZrO_2 to 230 ps for Al_2O_3. Using calculated values of $\tau_{ph,s}$ together with the equation $\tau_R=\tau_{ph,s}/\alpha_{s\text{-}f}^2$ we then obtain the values of $\alpha_{s\text{-}f}$ presented in Table II. Note that the extremely short phonon scattering time in ZrO_2 makes us unable to observe any exponential signal decay for sample no. 8.

According to Eq. (4) the ratio of transparency coefficients is

$$\zeta=\frac{\alpha_{f\text{-}s}}{\alpha_{s\text{-}f}}=\frac{c_s a(T/\Theta_s)u_s}{0.5c_f a(T/\Theta_f)u_f}\ , \tag{6}$$

TABLE II. Thermophysical properties, phonon scattering times, transparency coefficients, and thermal boundary resistance for different substrates at $T=90$ K.

Material	c_s (J cm^{-3}K^{-1})	k_s (W cm^{-1}K^{-1})	u_s (10^5 cm s^{-1})	$\tau_{ph,s}$ (ps)	$10^2\alpha_{f\text{-}s}$	$10^2\alpha_{s\text{-}f}$	R_{bd} (10^{-3} K cm^2 W^{-1})
MgO	0.53	3.4	5	160	9.6	5.0	0.53
Al_2O_3	0.39	6.4	7.9	230	4.7	2.5	1.1
$LaAlO_3$	0.4	0.35	3	42	4.5	5.9	1.15
$NdGaO_3$				42[a]	6.8	5.3[a]	0.76
ZrO_2	0.7	0.015	3	1.4			

[a]Values calculated with the thermophysical properties of $LaAlO_3$.

where the factor 0.5 in the denominator account for the specific heat of optical-phonon modes in the film. The thermophysical properties of $YBa_2Cu_3O_7$ are close to those of $LaAlO_3$ and probably of $NdGaO_3$ (Table II). Consequently, we calculate $\zeta=1.26$ in accordance with an approximate equality of the experimental values $\alpha_{f\text{-}s}$ and $\alpha_{s\text{-}f}$. For Al_2O_3 and MgO substrates, experimental values of $\alpha_{f\text{-}s}$ and $\alpha_{s\text{-}f}$ differ much more. This correlates with the calculated value $\zeta\simeq 2$.

To check the criterion of uniform temperature distribution through the thickness of a film we evaluate the characteristic length $l_{\mathrm{ph},f}/\alpha_{f\text{-}s}\approx 200$ nm, which appears to be large compared to d for all our samples. For the thicker films used by Carr *et al.*[5] there is a deviation from τ_b to d proportionality (Fig. 4).

Finally we estimate the thickness of an intermediate layer which may form the thermal boundary resistance. It has been supposed[5] that the thermal barrier has the thickness $d_b\simeq 10$ nm. The characteristic time of heat diffusion through such a barrier is $\tau\simeq d_b c_b R_{\mathrm{bd}}$ where c_b is the specific heat of the barrier material. Considering $c_b=c_f$ we deduce $\tau\simeq 0.9$ ns for $R_{\mathrm{bd}}=10^{-3}$ K cm^2 W^{-1}. During this time the temperature shift of a film would decrease by d_b/d of its initial value and, which is more important, the temperature decay would not be exponential. For our samples an exponential decay of the response is observed beginning from 0.1 ns after the maximum of the signal. This gives an upper limit of about 1 nm for the thickness of the intermediate layer.

The phonon model we use provides good agreement between the time of phonon scattering $\tau_{\mathrm{ph},f}$ calculated from the heat conductivity k_f of high-quality films and the phonon-electron interaction time $\tau_{\text{ph-}e}$ obtained from study of nonequilibrium photoresponse. For films with $k_f(T_c)\simeq 0.1\text{–}0.3$ W cm^{-1} K^{-1}, the well-pronounced peak in the $k_f(T)$ dependence below T_c is related to phonon-electron scattering due to electron coupling into Cooper pairs.[19] Consistently, the phonon scattering time due to phonon-electron interaction may be evaluated as $\tau_{\mathrm{ph},f}\simeq 3k_f[0.5c_f b(T/\Theta_f)u_f^2]^{-1}\simeq 20\text{–}60$ ps. For $YBa_2Cu_3O_7$ the factor $b(T/\Theta_f)\simeq 0.3$. On the other hand the time of phonon-electron interaction may be deduced from the characteristic time of the nonequilibrium photoresponse ($\simeq 1$ ps at $T=90$ K) that was attributed[1] to the electron-phonon interaction time $\tau_{e\text{-ph}}$. With the detailed energy-balance equation $c_{\mathrm{ph}}/\tau_{\text{ph-}e}=c_e/\tau_{e\text{-ph}}$, assuming $c_{\mathrm{ph}}/c_e\simeq 40$, we get $\tau_{\text{ph-}e}\simeq 40$ ps.

Thus the main mechanisms of the voltage response to radiation become clear in the context of the hierarchy of interaction times in the electron and phonon subsystems. The nonequilibrium response with the characteristic time $\tau_{e\text{-ph}}\simeq 1\text{–}3$ ps dominates for a time scale shorter than the phonon-electron scattering time[1–4] $\tau_{\text{ph-}e}\sim 50$ ps. As we have demonstrated above, an exponential bolometric response with the characteristic time τ_{es} prevails for the time interval $\tau_{\text{ph-}e}<t<\tau_R$. For longer times $t>\tau_R$, the bolometric response is governed by diffusion phonon transport in the substrate.

VI. CONCLUSION

Taking into account both optical-phonon modes and the decrease of the acoustic-phonon group velocity at high energies, we have obtained values of phonon transmission coefficients of $YBa_2Cu_3O_7$/substrate interfaces which were calculated in the linear approximation from the time of phonon escape from the film and from the time of phonon return to the film from the substrate. Our main result are:

(i) The transition from an exponential decay of the bolometric response to a power-law relaxation occurs at a time of phonon return to the film from the substrate that ranges from 350 ns for Al_2O_3 to ≈ 100 ps for ZrO_2.

(ii) The statement of a two-order-of-magnitude discrepancy between experimental results and predictions of the acoustic mismatch theory is likely not correct. Despite the fact that our temperature range $T\approx 90$ K lies at the border of the area of application of the acoustic mismatch theory, for the majority of substrates the acoustic mismatch theory[13,20] gives values of $\alpha_{f\text{-}s}$ which are only 1.5–4 times larger than those reported here ($\alpha_{f\text{-}s}\simeq 0.02\text{–}0.1$).

(iii) Our results suggest for all substrates $\alpha_{f\text{-}s}+\alpha_{s\text{-}f}\ll 1$, thus contradicting the diffusion mismatch model, which supposes this sum to be strictly equal to unity.[6]

(iv) The thickness of an intermediate layer which may provide the thermal boundary resistance at a film/substrate interface is not larger than 1 nm. This also gives evidence in support of the acoustic mismatch theory.

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