

Nonbolometric laser-induced voltage signals in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films at room temperature

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We report on detailed studies of laser-induced voltage signals observed in unbiased $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films at room temperature. The induced signals are measured as a function of incident energy, bias current, light polarization, and wavelength. The observed signals are remarkable because of their magnitude, up to 2 V across 4 Ω at 30 mJ/cm², and because they represent potentials which are forbidden by the known crystal symmetry of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ if two obvious explanations, pyroelectricity and off-diagonal thermoelectricity, are considered. The signal proves to be independent of the polarization of light as well as its wavelength for $\lambda = 1064, 532, \text{ and } 355 \text{ nm}$. Our measurements provide strong evidence that the observed signals do not have a bolometric origin. The signal amplitude is proportional to the magnitude of the incident energy and not its energy density. The time constant of the bolometric response, determined by applying a bias current, is different from that of the transient. The dependence of the time constant, τ_s , of the transient on the film thickness is not unique. For films with thicknesses below 4000 \AA , τ_s agrees with simulated thermal diffusion times. Thicker films appear to show time constants independent of thickness, in contrast with predictions for thermal diffusion. Although the reflectivity is a symmetric function of the angle of incidence about the normal, the voltage signals are not.

I. INTRODUCTION

High-temperature superconducting thin films have been suggested to be well suited as optical detectors. Braginski, Forrester, and Talvacchio¹ give a rather complete review of the current state of experimental and theoretical progress. Most of the investigative effort has been spent on bolometric response,²⁻⁴ although nonthermal optically induced signals were observed and studied as well.⁴⁻⁷ Generally, samples are tested at and below the superconducting transition temperature T_c . In most experiments a bias current I_b is applied and the response to incident light is recorded as a transient voltage $V_b(t)$ superimposed on the bias level V_b . In contrast, we observe a nonbolometric optically induced signal at room temperature in *unbiased* $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Y-based 1:2:3) thin films.

A thermal, i.e., bolometric response, is caused by the temperature rise ΔT of the sample due to the illumination and reflects the temperature dependence of the resistance $\partial R / \partial T$. The recorded signal $V_b(t) = I_b(\partial R / \partial T)\Delta T$ is proportional to ΔT , which is a function of the incident energy, and the heat capacity and thermal conductivity of the material itself. The characteristic decay time for the signal reflects the underlying thermal process and depends on the thermal diffusion time across the film which increases with the square of the film thickness d . This time dependence is expected as long as d is larger than the optical penetration length and the thermal contact resistance to the substrate is small compared with the thermal resistance of the film.

The room-temperature signal we observe is nonbolometric and is not associated with the superconducting properties of the sample. Thus far we have been un-

able to associate a conventional mechanism with the induced voltages. The observed voltage cannot result from pyro- or piezoelectric effects because of the presumed centrosymmetric crystal structure of Y-based 1:2:3. Thermoelectric effects resulting from the diagonal components of the Seebeck coefficient matrix are far too small to explain the observed magnitudes. Similar effects from the off-diagonal components are not allowed for symmetry greater than monoclinic and, therefore, should also be precluded by the presumed symmetry.⁸ (The possibility of local symmetry breaking is discussed below.)

As described in an earlier paper,⁸ the signal rises over the illumination period (approximately 10 ns) and decays with a time constant τ_s of approximately 20–100 ns depending on the film in question. The peak amplitude of the transient can reach up to 2 V across 4 Ω , induced with an illumination level of 30 mJ/cm², which does not alter the film morphology. Note that a current of substantial magnitude has to be flowing through the sample. If an external 5- Ω resistor is placed in parallel to the film resistance (5 Ω), the signal voltage decreases by a factor of 2, indicating a “real” current flow.⁸

Independent evidence of the symmetry-breaking nature of the effect was provided by Tate *et al.*⁹ and confirmed in our laboratory. When a film is illuminated from the front as well as from the back through the transparent substrate, nearly identical but sign-reversed voltage signals are observed.

We measured also the temperature dependence of the signal and find that the amplitude decreases linearly between room temperature and T_c . Below T_c no signal is observed at low light intensities. It can be recovered, however, when the film temperature is elevated above T_c using a more energetic light pulse and will remain until

some portion of the film cools down below T_c . We modeled the temperature profile of the film and found satisfactory agreement between the calculation and the time dependence of the signal.

In this paper we summarize the results of detailed studies measuring the induced signal as a function of incident energy, bias current, light polarization, and wavelength. In addition we measured the reflectivity of our films and the laser-induced voltages as a function of angle of incidence θ . Overall, we investigated films of various thicknesses ranging from 1000 to 10 000 Å. [In addition to our measurements on thin films we also observed the signal in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ (80-K compound) single crystals.]

The signal amplitude is found to be proportional to the magnitude of the incident energy E_L and not, as one might have expected, to the energy density E_{DL} . Conventional thermally activated processes are proportional to the incident energy density and can therefore be excluded as a cause for the signal.

The signal amplitude was recorded as a function of bias current and we find that the induced electric field and the bias superimpose linearly. We deconvoluted the bolometric response when a bias current was applied and find that the thermally induced voltage and the transient observed without bias have very different time dependences.

Changing the polarization and wavelength of the light has no effect on the observed signals. A simultaneous measurement of the reflectivity and the induced signal as a function of angle of incidence shows a surprising result. While the angular dependence of the reflectivity evolves symmetrically around normal incidence, the induced signal decreases linearly when the incident angle is varied from -55° to $+55^\circ$; i.e., the signal changes asymmetrically.

II. SAMPLE

Thin films of Y-based 1:2:3 are produced via laser ablation. The films are of rectangular shape with dimensions of $(3 \times 10)\text{-mm}^2$ on average. During deposition the substrate is maintained at approximately 750°C in an atmosphere of 200 mTorr of oxygen. Slow cooling in oxygen of ambient pressure produces films of excellent quality without need for further post processing. The surface quality is substantially improved by introducing a quartz rod between the target and the heater so that the plume of ablated material is blocked from reaching the substrate in a straight path.¹⁰ Scanning electron microscope studies show that the surface morphology is homogeneous on length scales between 10 and 1000 μm .

The film generally reproduce the crystallographic orientation of the employed substrates in the growth direction, i.e., (100) SrTiO_3 and LaAlO_3 produce films with the c axis oriented perpendicularly to the substrate (c oriented). Films deposited on (110) SrTiO_3 show a (110) or ab orientation. In either case x-ray spectra reveal highly preferred structural orientation. Superconducting transitions that are completed above 90 K are routinely obtained via four probe resistivity measure-

ments and the transition temperatures are confirmed in inductive tests as well. The transition width ΔT is of the order of 1–2 K. Room-temperature resistivities of 0.25 and 2.8 $\text{m}\Omega\text{cm}$ and corresponding resistivity ratios, $\rho(300\text{ K})/\rho(100\text{ K})$, of 2.5–3 and approximately 1.5 are obtained for c and ab textured films, respectively. Films of c orientation yield transport critical current densities of $3 \times 10^6\text{ A/cm}^2$ at 77 K and $2 \times 10^7\text{ A/cm}^2$ at 4.2 K.

Contacts to the films are made either by Ag pads sputtered on to a masked film and Ag wire attached with Ag paste to the pads or by connecting the Ag wire directly onto the film with Ag paste and firing the film at 500°C in 500 mTorr of oxygen for 10 min. Either method produces sturdy contacts of low resistance.

III. EXPERIMENT

Measurements were made using a pulsed Nd:YAG laser (where YAG denotes yttrium aluminum garnet) in the single shot mode. Most results were obtained using the second harmonic ($\lambda=532\text{ nm}$); however, for wavelength-dependent studies we used the fundamental (1065 nm) and the third harmonic (355 nm), as well. The laser pulse exhibits a full width at half maximum of approximately 10–20 ns and a rectangular beam profile with Gaussian width. The illuminating beam area was restricted by a mask of 1 mm in width and varying length. Signals were recorded with a 450-MHz oscilloscope terminated into 50 Ω , digitized via a “Frame grabber,” and stored in a personal computer.

Figure 1(a) shows the experimental setup. A fraction of the laser beam (approximately 10%) is split off and

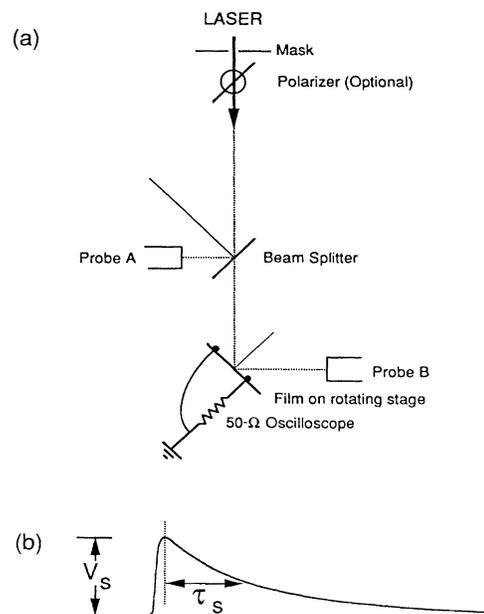


FIG. 1. (a) Experimental setup. (b) Typical laser-induced signal; V_s and τ_s represent signal amplitude and time constant, respectively.

detected by a pyroelectric probe with energy sensitivity ranging between $1 \mu\text{J}$ and 1J . The energy of each laser pulse is measured and recorded. The illuminated film is mounted on a rotating stage and the induced signal is fed via a coaxial cable into the 50Ω terminated scope. Simultaneously the reflectivity is measured using a second pyroelectric probe. It should be stressed that the observed signal is the difference between the transient and a featureless "baseline" obtained by screening the film from the laser pulse. Figure 1(b) shows a typical signal shape. V_s represents the maximum signal amplitude and τ_s the time constant defined as the interval during which the signal decays from its peak value to $V_s/2$.

IV. RESULTS

We measured the laser-induced signal as a function of bias current. The resistance of the film used in this experiment is approximately 5Ω and the temperature coefficient of the resistance between 100 and 300 K is constant and was determined to be $1.5 \times 10^{-2} \Omega/\text{K}$. The film has a thickness of $6500 \pm 300 \text{ \AA}$, which was verified via a profilometer scan across an acid etched pattern after the experiments were completed.

For this experiment we employed a contact arrangement similar to a four-wire potential measurement and used the potential contacts for the signal connections. In Fig. 2 we display the signal amplitude V_s as a function of bias current for incident energies of 4.5 and 9 mJ/cm^2 . In either case V_s varies linearly with the applied current, indicating that the bolometric response of the film superimposes on the transient.⁸ The linear increase of V_s with I_b corresponds to the initial change in the average inverse conductance of the film. We find $(1/\Delta R)^{-1}$ values of 0.24 and 0.47 Ω from Fig. 2 for incident energy densities of 4.5 and 8 mJ/cm^2 , respectively, in good agreement with our simulation results. Doubling the incident energy roughly doubles ΔR , which is expected as long as the average temperature change is small (see analysis).

The signal was monitored as a function of polarization and wavelength of light at normal incidence. Results are summarized in Figs. 3(a) and (b). We recorded the signal

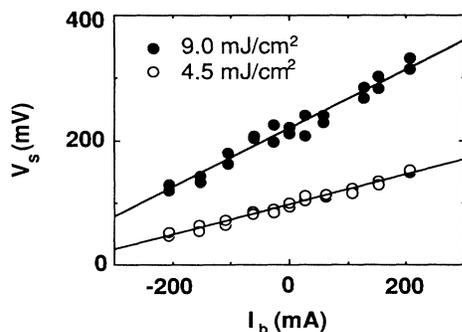


FIG. 2. Signal amplitude as a function of bias current I_b for incident energies of 4.5 and 9 mJ/cm^2 , as indicated. The signal amplitude depends linearly on the bias current with slopes of 0.24 and 0.47 Ω , respectively.

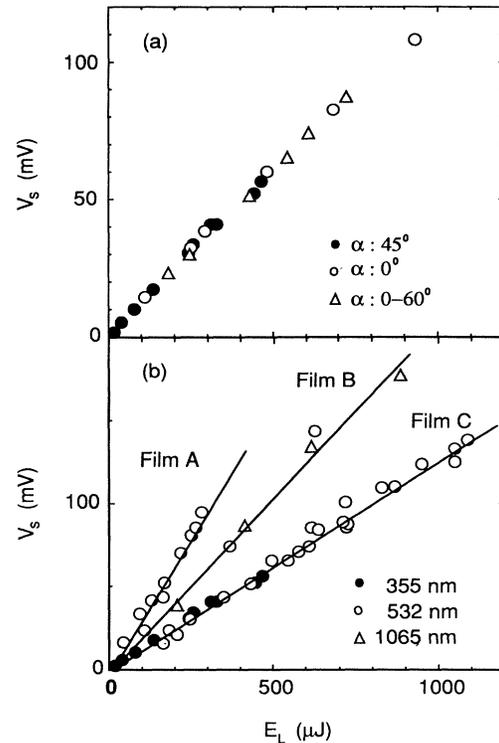


FIG. 3. (a) Signal amplitude for different angles of polarization as a function of incident energy. (b) Signal amplitude as a function of incident energy for various films using light of different wavelength.

as a function of incident energy at polarization angles α of 0° and 45° . Alternatively we kept the energy input constant and varied α from 0° to 60° , thereby changing the incident energy according to Malus's law. For all three measurements the incident energy proves to be the determining factor as the signal amplitude increases linearly with the energy input, as shown in Fig. 3(a). We also checked for a signal dependence on the rotational sense of circularly polarized light and find no difference to within 5% for left- and right-handed light.¹¹

In Fig. 3(b) we display the signal amplitude as a function of energy for three different films using wavelength of light as an implicit parameter. Films A, B, and C have thicknesses of 2500, 8000, and 6500 \AA , respectively. For individual films the signal is always linear in applied energy independent of wavelength. A possible threshold for the signal has to be below 10 μJ , the lowest energy we applied.

In all figures we intentionally graph the signal as a function of incident energy E_L and not as a function of energy density E_{DL} . We find that the signal amplitude stays unchanged when we vary the size of the illumination area between the potential probes by moving a focusing lens through its focal point while keeping the beam energy constant (Fig. 4). The area of the rectangular illumination spot was changed by a factor of approximately 100 from 0.125 to 12 mm^2 varying the incident energy

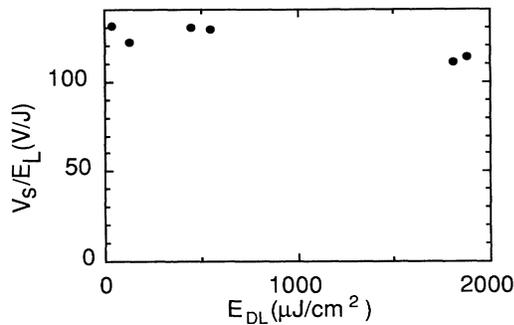


FIG. 4. Normalized signal amplitude as a function of energy density. The signal was normalized to the incident energy to eliminate variations due to fluctuations in the laser output.

density by as much. The distance from the potential probes to the closest illuminated area changed by a factor of 10. The large change in separation between illumination area and potential contact which lead to no change in signal voltage is inconsistent with planar thermal emf's.

As mentioned earlier, our setup permits simultaneous

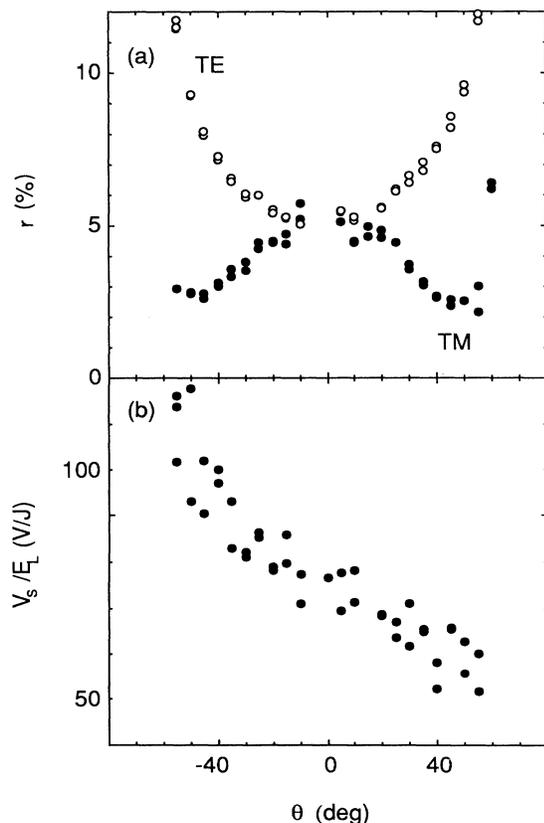


FIG. 5. (a) Reflectivity as a function of angle of incidence for TE- and TM-polarized light. r is corrected for light lost due to surface scattering as described in the text. (b) Signal amplitude normalized to the incident energy as a function of angle of incidence.

measurement of the reflectivity of the film, r , and the induced signal as a function of angle of incidence. We recorded the reflectivity (r) for light polarized perpendicular (TE) and parallel (TM) to the plane of incidence, as shown in Fig. 5(a). The small value of approximately 5% deduced for normal incidence is at the lower bound of published values which span a range from approximately 3% to 20%.¹²⁻¹⁶ The measured reflectivity, however, agrees rather well with other measurements on thin films.¹⁵ Corrections have been made for light scattered out of the solid angle of the detector (see below).

Most surprisingly, the angular dependence of the laser-induced signal does not show a symmetric behavior about normal incidence. In this measurement the film is rotated around an axis parallel to a line connecting the potential probes and perpendicular to the plane of incidence. The illuminated area is approximately 1×2 mm² with the long axis long the direction between the potential probes.

When the angle of incidence θ is changed from -55° to $+55^\circ$, the signal decreases linearly from 110 to 55 mV [Fig. 5(b)]. A comparison with the angular dependence of the reflectivity shows that the symmetric change in absorptance with angle of incidence cannot be the cause for the asymmetric decrease in signal. In addition, the change in absorptance ($1-r$) amounts to about 5%, much too small to account for a decrease in signal by a factor of 2.

Tate, Hilinski, and Foster¹⁷ reported an asymmetric variation of the signal amplitude with the angle of incidence in thin polycrystalline Y-based 1:2:3 bulk samples. In contrast to our measurements they do not observe a signal for light of normal incidence. It is not obvious, however, that signals observed in thin films and in polycrystalline samples have the same origin.

In most films we find that the direction of the laser-induced electric field coincides with the long axis of the film. In some films, however, the signal is observed only along the short transverse axis, while two films show the transient signal along both axes. In the latter case one direction shows a dominant response, approximately two times larger than along the other side. Although four signals were observed with the potential probes located in a square arrangement, the algebraic signs of the induced electric fields along the sides of the square are inconsistent with a single large current loop. Because polarity and amplitude of the induced voltage signal remain the same when measured along opposite sites of the square,⁸ the sum of the voltages around the loop does not add up to zero: $\sum_{\text{loop}} V_i \neq 0$. One current pattern consistent with this result are circulating current sheets traveling in one direction along the top of the film and the opposite direction along the bottom.

In Fig. 6 we show a compilation of the time constant τ_s versus film thickness d for all films which were tested. Although the data are scattered, τ_s shows a distinctly different dependence on the film thickness for d smaller and larger than 4000 Å. Below 4000 Å, τ_s increases with d , while for films with a thickness larger than 4000 Å the time constant appears to be independent of film thickness.

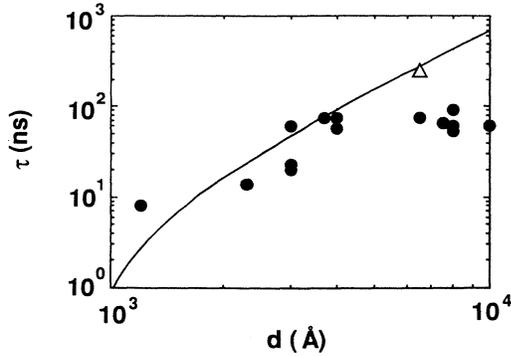


FIG. 6. Time constants τ_s displayed as a function of film thickness for various films on a log-log plot. The solid line simulated thermal diffusion times calculated as described in the text. The open triangle indicates the thermal diffusion time measured for a film with a thickness of 6500 Å.

V. ANALYSIS

A. Bias current

When a bias current is supplied the laser-induced signal will be a superposition of a bolometric response as well as the transient signal observed in the absence of a bias, assuming additive contributions:

$$\begin{aligned} V(t) &= R_0 I_b + (\partial R / \partial T) \Delta T(t) I_b + V_t(t) \\ &= V_b + V_b(t) + V_t(t). \end{aligned} \quad (1)$$

In the following discussion we will neglect the steady-state bias voltage $V_b = R_0 I_b$ ($R_0 = 5 \Omega$ at 300 K) since we define the signal as the difference between the baseline and the laser-induced transient. The average film temperature ΔT and V_t have different time dependences as shown below. Our calculations modeling the temperature of the film, however, show that after the initial temperature rise, the average film temperature remains constant to approximately 2% for approximately 40 ns as the heat travels across the film ($d \sim 6500 \text{ \AA}$, Film C). We will interpret our data by restricting the analysis to the peak value of the signal, $V_s \sim V(10 \text{ ns})$. In doing so, our analysis will be uncomplicated by heat-transfer effects at the film-substrate interface. The discussion of the time constants for films of various thicknesses, however, shows that the thermal contact resistance at that boundary is negligibly small (see below).

From Eq. (1) we expect a linear relationship between V_s and I_b with the slope being equal to $(\partial R / \partial T) \Delta T$. This relationship would be exact if the temperature change was uniform across the film. After the light energy has been absorbed in the front layer of the film, however, the temperature profile shows a maximum at the front surface and decreases towards the substrate which is assumed to be an infinite heat sink. The film can be thought of as a structure made up of thin layers or segments all of which contribute a finite resistance in parallel. The average resistance $\langle R \rangle = (\partial R / \partial T) \langle \Delta T \rangle$, where

$\langle \Delta T \rangle$ is the temperature averaged over all segments, does not equal the inverse average conductance, which is calculated by simulating a parallel network where each segment represents a temperature dependent resistance: $\langle 1/R \rangle^{-1} = (\partial R / \partial T) \langle \sum_{i=1}^n 1/T_i \rangle^{-1}$. The difference, however, is small as long as the induced change in average temperature remains small and approximate results can be quickly obtained using the average resistance and Eq. (1).

Doubling the incident energy should double the slope, i.e., $(\partial R / \partial T) \Delta T$, if the average resistance were the determining factor. Because the average conductance grows more slowly than $\langle R \rangle$ with increasing energy input, the ratio of the slopes is not exactly 2 but 1.96, and the intercepts of the two curves with the x axis do not coincide. The difference, however, is small, less than 4%, in agreement with the data.

For incident energy densities of 4.5 and 9 mJ/cm² used in the experiment we calculate an average temperature rise of 18 and 36 K using a specific heat of 2.7 J/cm³ K (Ref. 18) and a thermal conductivity along the *c* axis of 15 mW/cm K.¹⁹ The simulation yields inverse average conductances of 0.3 and 0.6 Ω for these two energy densities in fair agreement with the experimental values of 0.24 and 0.47 Ω (see Fig. 2).

The analysis of the peak voltage as a function of bias current establishes that the bolometric and the transient response superimpose linearly and additively. Further information is extracted by examining the full time dependence of the spectra. We deconvoluted the bolometric response of the film by taking the difference and sum between spectra obtained with $\pm 200 \text{ mA}$ of bias current [Fig. 7(a)]. From Eq. (1) we expect the difference spec-

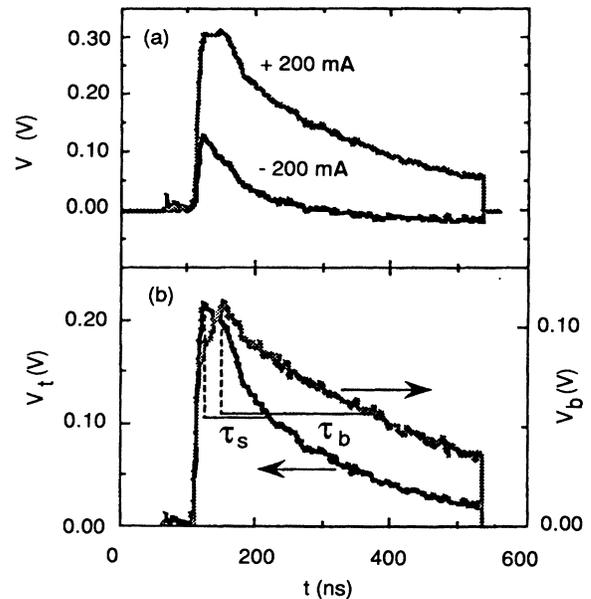


FIG. 7. (a) Recorded signals for bias currents of $\pm 200 \text{ mA}$, as indicated. (b) Transient and bolometric signals obtained by taking the sum and difference of the two spectra shown in (a). The time constant for the bolometric signal is approximately three times longer than for the transient response.

trum to show the thermal response only, while the sum spectrum displays the laser-induced transient only. We believe the deconvolution scheme works well because the transient thus obtained traces exactly a spectrum recorded without bias current. In Fig. 7(b) we compare the two deconvoluted components of the signal. The isolated bolometric spectrum does not have a maximum at approximately 10 ns, where the transient peaks, but reaches a peak at approximately 40 ns, which according to our simulation is the time for the heat pulse to transverse the sample. The initial increase for times between 10 and 40 ns occurs because the average film conductance continues to decrease during the time of heat transmission. The decay time constant of the bolometric response is roughly three times longer than that of the transient, 250 and 85 ns, respectively. Therefore the laser-induced transient does not have a time constant representative of the average temperature of the film and the mechanism is most likely not bolometric in nature.

Using the simulation routine with the quoted specific heat and thermal conductivity values as an input, we calculate the thermal diffusion time across films of different thicknesses ranging from 1000 to 10 000 Å. The diffusion time constant is defined as the time at which the average film temperature has fallen to half its maximum value. In our simulation we have assumed negligible thermal contact resistance at the film substrate interface. The values so obtained are least-squares fitted to a polynomial yielding $\tau_s \sim 1 - 7.9 \times 10^{-3}d + 7.8 \times 10^{-6}d^2$, where τ_s and d are given in nanoseconds and angstroms, respectively. The theoretical curve in Fig. 6 was drawn according to this expression. The calculated curve agrees with the one available experimental point: the thermal diffusion time of approximately 250 ns was determined for a film of 6500 Å thickness via the analysis of the bias current experiment.

B. Correction of reflectivity

When we measured the reflectivity κ , we found that its value depends on the distance between film and detector, i.e., the solid angle Ω accommodating the reflected light. We believe the observed variation in reflectivity as a function of distance stems from the surface roughness of the films. Indeed, scanning electron microscopy reveals rough surface features on a length scale of 0.5–1 μm , which is comparable to the wavelength we used.

To correct for the reflected light lost due to surface scattering, we measured $\kappa(\Omega)$ at constant $\theta=40^\circ$ and fit the data to a power law:

$$\kappa(\Omega) = A\Omega^y = 1.74\Omega^{0.16} \quad (2)$$

In the absence of scattering $\kappa(2\pi)$ represents the proper value of the reflectivity. It should be noted that our method of fitting the data implicitly offers a way of characterizing the surface roughness of films via the exponent y . We then correct the reflectivity data via

$$\kappa_c(\theta) = \kappa_M(\theta, \Omega)\kappa(2\pi)/\kappa(\Omega) \quad (3)$$

where $\kappa_M(\theta, \Omega)$ is the measured reflectivity and the ratio $\kappa(2\pi)/\kappa(\Omega)$ is calculated from Eq. (2).

Because of the smooth variation of the reflectivity as a function of incident angle over the limited range of θ available, we could not extract the index of refraction n and the extinction coefficient unambiguously. Nonetheless, the bounds we place on n and κ permit us to estimate the penetration depth as 700–1000 Å, in fair agreement with published results.¹² In our heat-flow simulations we use an average value of 800 Å.

VI. DISCUSSION

The signal does not depend on the polarization of the incident light. Therefore the symmetry breaking element is intrinsic to the film itself. The independence of the signal on the quantum energy over a range of 1.16–3.48 eV places an upper bound of approximately 1 eV on a threshold if an excitation process is considered. Furthermore, the signal is linear in the incident energy with 10 μJ being an upper bound for a possible threshold value.

The dependence of the signal on the magnitude of the incident energy and not on the incident energy density is significant. Thermal processes that depend on the energy density are thereby excluded as an explanation.

The dependence of the signal amplitude on the applied bias current shows that the bolometric and transient responses superimpose linearly. The signal when deconvoluted into a bolometric and a transient component shows clearly that the time constant of the thermal process is three times longer than for the induced transient signal. Under the assumption that the transient represents an integral film property, the difference in time constants makes it unlikely that the transient is caused by the average film heating.

The measured reflectivity for light polarized parallel and perpendicularly to the plane of incidence shows qualitatively the expected behavior. The low value for the reflectivity at normal incidence (approximately 5%) might be related to the surface roughness of the films investigated.

The asymmetry in signal amplitude as a function of angle of incidence is the most unexpected feature of this paper. We did not make a correction for the increased illumination area which varies as $1/\cos\theta$ with θ , as we find that the signal does not depend on the energy density but on the magnitude of the incident energy. Clearly the signal variation cannot be explained by the symmetric variation in reflectivity.

The difference in signal voltage versus incident energy for various films indicates that the signal depends on the local structure. The idea is supported by the fact that in films where there is a signal across the width as well as along the length of the film, no single closed current loop consistent with the direction of the induced electric fields could be established. Examining the films via a scanning electron microscope did not provide any clues although the surface morphology for *c*- and *ab*-oriented films is quite different.

As mentioned in Sec. IV, the time constant τ_s does not show a unique dependence on the film thickness. When we graph the simulated thermal diffusion times with the experimental data (Fig. 6), the distinction becomes even

more apparent. For films with thicknesses below 4000 Å data and simulation curve are compatible, while above 4000 Å data and calculation clearly disagree.

All data points lie either on the simulation curve or below it. If thermal resistance at the film substrate interface had an effect on the time constants the data would lie above the simulation curve which was calculated assuming zero contact resistance. Therefore the contact resistance at the film substrate boundary must be too small to effect the decay of the signal. The reliability of our simulation curve is easily checked by comparison with the experimentally determined thermal diffusion time determined from Fig. 7(b) and indicated by the open triangle in Fig. 6.

The disagreement between the simulation results of the thermal diffusion time across the sample and the measured time constants for films with thicknesses above 4000 Å clearly indicates that the laser-induced signal does not have a bolometric origin in thick films. For thinner films a bolometric mechanism cannot be excluded by the data. However, if only one mechanism is invoked as the cause of the induced signal for films of all thicknesses, an interpretation involving a bolometric origin has to be rejected.

VII. CONCLUSIONS

The symmetry-breaking mechanism, responsible for the laser-induced signal, is provided by the film and is most likely part of the local structure of the film itself. The proposed local structure must have a coherence which extends over several millimeters, corresponding to the separation between the potential probes, but not leading to any change in the x-ray diffraction spectrum.

Three independent observations permit us to exclude a single bolometric process as a cause for the induced transients. First, the signal amplitude is proportional to the magnitude of the incident energy and not its energy density. Second, the time constant of the bolometric response determined by applying a bias current is different from that of the transient. Third, the dependence of τ_s on the film thickness is not unique. If a single mechanism for the laser-induced signal independent of film thickness is assumed, then the time constant is not in agreement with predictions for thermal diffusion across the sample. All three observations exclude bolometric mechanisms; however, the signal could still be thermally initiated.

The asymmetry of the signal with respect to the rotation of the film around an axis perpendicular to the plane of incidence is unexpected and warrants further examination.

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