Anisotropic thermoelectric effect in Y–Ba–Cu–O thin films during time-varying laser heating

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A study has been made of the emf which arises in an anisotropic thin film during time-varying heating by a nanosecond-range laser pulse. The results of a theoretical analysis agree well with the fairly high voltage pulses (up to ~ 0.1 V) observed experimentally during the application of nanosecond laser pulses to Y–Ba–Cu–O films.

INTRODUCTION

Anomalously high pulsed voltages (~ 1 V) have been observed in *supposedly isotropic* conducting thin films (of molybdenum, tungsten, cadmium, etc.) when laser pulses were applied in several studies[1–4] over the past two decades.

The thermoelectric effect has been considered as a possible cause of these high emfs. That interpretation is suggested by the fact that these emfs are observed in films of materials for which the Seebeck coefficients α_{ij} are fairly high ($|\alpha_{ij}| \gtrsim 10$ μV/K; Ref. 5). However, an interpretation of this sort on the basis of the ordinary isotropic Seebeck effect ($\alpha_{ij} = \alpha_0 \delta_{ij}$) in a homogeneous medium runs into insurmountable difficulties. Specifically (Fig. 1), the potential difference measured between test contacts A and B on the film surface is $\Delta = \alpha_0 (T_A - T_B)$ in this case; i.e., a temperature difference $\sim 10^6$ K between the contacts would be required in order to observe $\Delta \approx 1$ V. Such a temperature difference is obviously impossible. It thus scarcely matters that the test contacts in the experiments of Refs. 1–4 were completely outside the illumination zone; i.e., the relation $T_A = T_B$ held, so the potential difference would have been $\Delta = 0$.

Several other, more or less exotic, versions of the thermoelectric effect have been accordingly considered as possible explanations. These other versions of the effect basically reduce to the following four.

First, there is the idea of a spatially nonuniform thermal emf, i.e., $\alpha_0(x) \neq$ const. The length scale for such a nonuniformity is assumed to be on the order of the fine grain structure of the film, i.e., on the order of d, as a rule. Simple estimates quickly show that a model of this sort could in principle lead to a potential difference $\Delta \lesssim \max\{\alpha_0\} T_0$, where T_0 is the maximum (along x) increment in the film temperature, even if the test contacts are at the same temperature. However, for the same reason as above, we would again need values $T_0 \approx 10^6$ K, so this model does not work.

Second,[3] it has been suggested that the heat evolution in the film is nonuniform, again with some length scale on the order of d for the variations along x, while the thermoelectric properties are assumed to be uniform along x. This nonuniformity of the heat evolution would lead to a small-scale nonuniformity of $T(x)$, with the same length scale. In this situation, the ordinary linear Seebeck effect would of course give us $\Delta = 0$, so the nonlinear Benedicks effect[5] is invoked: $E_x \propto \beta(\partial^3 T/\partial x^3)$. In this case, with a suitable $T(x)$ profile, one could reach values $\Delta \lesssim \beta T_0/d^2$, but the known values[3] of the Benedicks coefficient β are again too small to explain the experimental values of Δ.

The third possibility is a combination of the first two. Specifically, within the framework of the linear Seebeck theory, one assumes that $\alpha(x)$ and the heat evolution, i.e., $T(x)$, are simultaneously nonuniform. It is simple to estimate Δ in this case, by assuming a sinusoidal modulation

$$\alpha = \alpha_0 \sin \frac{2\pi x}{d},$$

$$T(x) = T_0 \cos\left(\frac{2\pi x}{d} + \varphi\right)$$

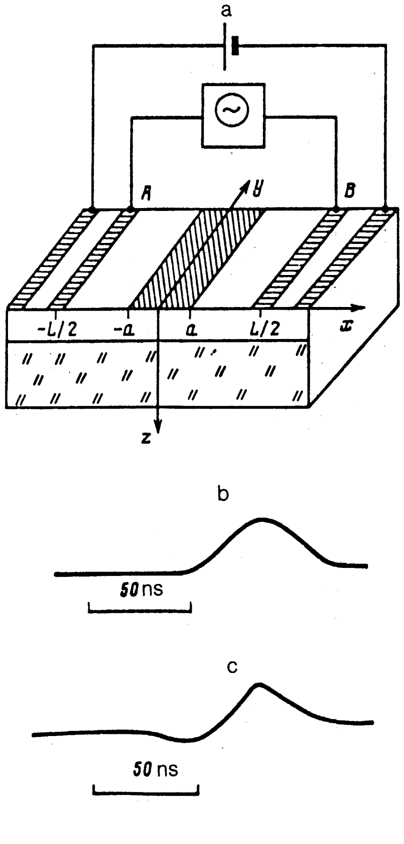

FIG. 1. a: Geometry of the illuminated film. The horizontal hatching shows the test strip contacts A and B and the contacts through which the bias current is applied. The vertical hatching shows the illuminated region on the film surface. b,c: Characteristic voltage pulses during illumination from the free surface of the film and through the substrate, respectively.

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(the nonzero averages of α and T over x under the condition $T_A = T_B$ do not contribute to Δ). We then find

$$\Delta \approx \alpha_0 T_0 \frac{2a}{d} \cos \varphi.$$

We see that this model is capable in principle of leading to sufficiently large values of Δ (if the parameter $2a/d$, where $2a$ is the width of the illuminated region, is sufficiently large, $\sim 10^3$). However, these values could be achieved only with $\varphi \approx 0$, i.e., only if there were a special phase relation between the spatial modulation of α and that of the optical absorption coefficient.

The fourth and final version, which has been formulated at a qualitative level in some papers by Von Gutfeld,[1,2] assumes an anisotropy of the Seebeck coefficients α_{ij}. Specifically, one assumes $\alpha_{xz} \neq 0$ in the geometry of Fig. 1. A residual stress in the film after the deposition and the particular morphological features of obliquely deposited films were discussed as possible physical reasons for this anisotropy in Refs. 1 and 2. In this case it is a simple matter to derive the estimate

$$\Delta \approx \alpha_{xz} \frac{T_0}{d} L$$

(L is the distance between the test contacts). This estimate is quantitatively identical to that of the third possibility. Note, however, that in this case (in contrast with the first three) the magnitude and sign of Δ are determined by the gradient of T *along z, not along x*, in the geometry of Fig. 1.

We have recently observed some voltage pulses much like those described above in thin films ($d \approx 300$ nm) of the compound $YBa_2Cu_3O_{7-\delta}$ deposited by laser deposition on (100)-cut strontium titanate.[6] Below we report a detailed experimental study of these voltage pulses and an interpretation of them based on a thermoelectric nature for the effect.

EXPERIMENTAL RESULTS

Figure 1a shows the layout used for the experimental observation of the laser-induced emf pulses in the films described above. The test samples were bridges of various widths, and the distance between the silver test contacts was ~ 1.5 mm. The thickness of the film grown by laser deposition on (100)-cut $SrTiO_3$ (Ref. 1) was $d \approx 300$ nm. Examination of a film under a polarizing microscope revealed a polycrystalline structure with a typical crystallite size on the order of d. The c axes were oriented along the normal to the substrate, while the **a** and **b** axes were oriented in the plane of the film, in an otherwise arbitrary way. The point of importance for our purposes is thus that the film is *isotropic* in the plane of the interface and should not have a *polar axis* in this plane.

A bridge of this sort was exposed to a single-mode pulse from the laser described above. The optical system formed a uniformly illuminated strip of variable width $2a$ on the surface of the film (Fig. 1a). The energy density during this exposure did not exceed 10 mJ/cm^2; this figure is below the threshold for so-called laser modification of films.[7,8] During an exposure of this type, a voltage pulse in the nanosecond range arises across the test contacts (Fig. 1b). This pulse is sent by means of a matched 50-Ω cable to a high-speed oscilloscope. This pulse has several interesting qualitative features.

In the first place, at room temperature the height of the signal is independent of whether a bias current is flowing and, if it is, on the direction of this current. The signal thus cannot be explained on the basis of any changes in the resistance of the film during the illumination.

Second, the height of the signal does not depend on the width of the strip, $2a$, at a fixed *total energy* of the light in the spot, as $2a$ is varied by a factor of more than 5. It is also independent of the *position* of the strip within the bridge region between the contacts.

Third, a study of the temporal envelope of the signal (Fig. 1b) shows that the rise time is essentially the same as that of the laser pulse, while the trailing edge has a length $\tau \sim 100$ ns, which is the same as the thermal relaxation time of a film of this thickness.[7,8]

Fourth, the temporal envelope of the signal undergoes substantial changes when the film is illuminated *from the substrate side* (Fig. 1c); these changes extend to a change in the *polarity* of the voltage during the laser pulse.

Figure 2a shows the amplitude of the electrical signal as a function of the exposure. We see that the dependence is very nearly linear up to exposure values on the order of 40 mJ/cm^2.

We thus have the established fact that a *polar* emf arises

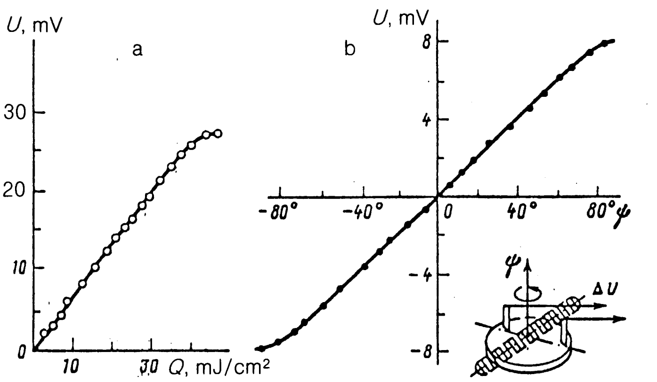

FIG. 2.

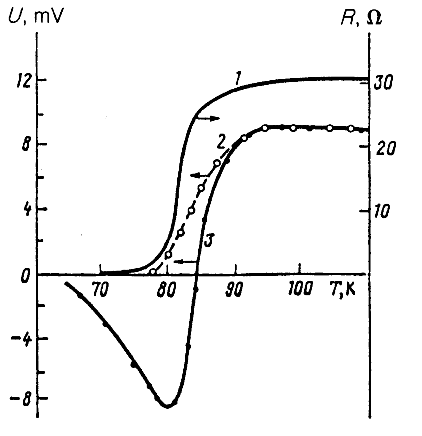

FIG. 3. *1*—Temperature dependence of the film resistance; *2*—$U(T)$ at a zero bias current; *3*—the same, at a bias current of 10 mA.

between two contacts held at *the same temperature*, under absolutely identical conditions, in the case in which the film is *apparently isotropic* in the plane of its surface. To further clarify the direction of the emf in the plane, we carried out the following measurements (see the inset in Fig. 2b). The positions of the contacts (mobile contacts in this case) were fixed with respect to the strip exposed to the light. The sample was rotated in the plane of its boundary. Figure 2b shows the signal amplitude as a function of the rotation angle ψ at a constant energy and a constant width $2a$ of the light strip. We see that there is a direction in which a potential difference does not arise; i.e., the photoinduced emf observed here is "linearly polarized" in the plane of the film. The amplitude of the observed signal does not depend on the polarization of the light in the case of normal incidence. The dependence of the signal on the angle of incidence α is smooth, accurately reproducing "Fresnel" curves

$$1 - R_{s,p}(\alpha),$$

where $R_{s,p}(\alpha)$ is the reflection coefficient for s or p polarization for the given angle α (Ref. 9). This result corresponds to the elementary fact that the signal is proportional to *the fraction of the laser energy which is absorbed.*

We turn now to an interpretation of the observed photoinduced emf. Since this test film is polycrystalline with randomly oriented **a** and **b** axes, we can rule out possible interpretations based on a photovoltaic emf (Ref. 10, for example). On the other hand, several experimental facts indicate that the effect is of a thermal nature. First, the length of the trailing edge of the voltage pulse is equal to the thermal relaxation time of the film. Second, the signal is proportional to the fraction of the energy which is absorbed, as follows from the dependence of the signal on the angle of incidence of the light. Further support for the idea of a thermal nature of the effect comes from the temperature dependence of the voltage (Fig. 3). We see that the effect disappears upon the transition to the superconducting state, as in Ref. 11.

Near T_c, in contrast with the situation at room temperature, the observed emf depends strongly on the presence and polarity of a bias current (Fig. 3). In other words, a significant bolometric component of the emf arises.

We immediately note that of the four versions discussed in the Introduction only the fourth—the model of an anisotropic thermal emf in the film—is in qualitative agreement with the experimental results. This conclusion follows from the experimental fact that the shapes of the emf pulses are sharply different when the film is illuminated from the side of the free surface and from the substrate side. According to this model, we have a qualitative explanation for the change in the polarity of the emf during the pulse when the film is illuminated from the substrate side: At the beginning of the pulse, while the thermal conductivity is still inconsequential, we have $\partial_z T > 0$ because of the gradient of the light intensity in the film. Later on, $\partial_z T$ becomes negative, because of heat transfer to the substrate, while the free surface of the film is thermally insulated.

The x distribution of the temperature is the same for both illumination layouts, so the first three models should lead to the same result for $\Delta(t)$ in the two illumination layouts, in contradiction of experiment. We will thus focus on a more detailed analysis of an anisotropic thermal emf in the film.

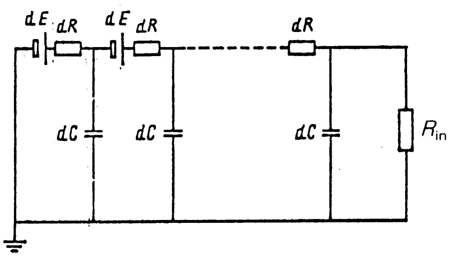

FIG. 4.

MODEL OF AN ANISOTROPIC THERMAL EMF

Let us assume that the Seebeck tensor is anisotropic in the geometry in Fig. 1a, with the principal axes of this tensor lying in the xz plane ($\alpha_{yx} = \alpha_{yz} = 0$). Experimentally, this situation corresponds to a maximum of Δ as a function of the azimuthal angle ψ in the plane of the film (Fig. 2b). For simplicity we assume that the electrical conductivity of the film is isotropic: $\sigma_{ik} = \sigma\delta_{ik}$. This assumption simplifies the calculations immensely, without causing any qualitative change in the results. The analog of Ohm's law is then

$$E_i = \sigma^{-1} j_i + \alpha_{ik}\partial_k T, \qquad (1)$$

where j is the current density. It is extremely difficult to derive an exact solution for the time-varying problem of finding the distribution of the field **E** in the film, even in the quasisteady approximation, since we would need to simultaneously solve wave equations in three media (the film, the substrate, and the air). The length scale of the field variations is $\lambda \simeq c/\tau_{\text{pulse}} \simeq 10^3$ cm, much greater than the actual size of the film. Accordingly, in solving the problem we were obliged to consider the boundary conditions at $x = \pm L/2$ also. Since there is no hope at all of finding an analytic solution for such a problem, the problem of finding the time-varying signal is actually solved (see the discussion below) by the standard methods of electric-circuit theory. In other words, the value of **E** [expression (1)] is averaged over the thickness of the film. However, there is a circumstance to bear in mind here: In the real experimental situation, the $\partial_z T(z)$ profile is fairly complex, to the point of having a change in sign on the interval $0 < z < d$. Correspondingly, the x component of **E** could in principle even change sign when we switch from the free surface of the film to the interface with the substrate. In other words, it may turn out that Δ is, for example, strongly dependent on the particular side of the film which has the test contacts. In this case, the "electric-circuit" averaging over the film thickness mentioned above would be meaningless.

Accordingly, we will start by making sure that the averaging is valid in the simpler example of the *steady-state* problem, $\partial_z T(z,x,t) = \partial_z T(z,x)$. In the geometry of Fig. 1a, we thus assume that the fields and the currents are in a steady state. The Poisson equation and the conditions that the boundaries of the film are impenetrable to the current are ($\mathbf{E} = \nabla\varphi$)

$$\Delta\varphi=\alpha_{ik}\partial_i\partial_k T,$$
$$\partial_z\varphi(x,\ 0)=0,\qquad \partial_z\varphi(x,\ d)=\alpha_{zz}\partial_z T(x,\ d), \tag{2}$$
$$\partial_x\varphi\left(-\frac{L}{2},z\right)=\partial_x\varphi\left(\frac{L}{2},z\right)=0.$$

Here we are using the fact that we have $\partial_z T(x,0)=0$ (the boundary is thermally insulated) and the fact that we have $\partial_z T=0$ at the boundaries $x=\pm L/2$, since these boundaries are outside the strip $-a<x<a$ used by the light. We also write $\operatorname{div} j=0$, and we assume that the problem is definitely two-dimensional, i.e., $\partial_y\equiv 0$. This is a reasonable assumption, since the edges of the illuminated strip are oriented along y, and we have $T(y)\equiv$const. Since the light intensity is uniform over the strip $-a<x<a$ in the actual experimental situation, we set

$$T(x,\ z,\ t)=T(z)\theta(a-x)\theta(a+x),$$

ignoring the small distance ($\sim d$) over which the boundary of the heated region is blurred by heat transfer along the film.

Now making the replacement $\varphi=\xi+\alpha_{zz}T$ in Eqs. (2), we find the following equation for ξ:

$$\nabla^2\xi=(\alpha_{xz}+\alpha_{zx})\frac{\partial T}{\partial z}[\delta(x+a)-\delta(x-a)],$$
$$\partial_z\xi(x,0)=\partial_z\xi(x,d)=\partial_x\xi\left(-\frac{L}{2},z\right)=\partial_x\xi\left(\frac{L}{2},z\right)=0. \tag{3}$$

On the right side of the Poisson equation we have ignored $\partial^2_{xx}T$ here in comparison with $\partial^2_{xz}T$, by virtue of the small value of the ratio d/a. We immediately note that we do not need to go over from ξ to φ in order to calculate Δ, since the test contacts are isothermal. We thus need to determine how strongly $\Delta(z)=\xi(L/2,z)-\xi(-L/2,z)$ depends on the value of z in the interval $0<z<d$. Boundary-value problem (3) can be solved easily by separation of variables:

$$\xi=\sum_{m=0}^{\infty}\cos\frac{\pi mz}{d}A_m(x).$$

For A_m we have

$$\frac{d^2A_m}{dx^2}-\left(\frac{\pi m}{d}\right)^2A_m=b_m[\delta(x+a)-\delta(x-a)],$$
$$\frac{dA_m}{dx}\left(\frac{L}{2}\right)=\frac{dA_m}{dx}\left(-\frac{L}{2}\right)=0, \tag{4}$$
$$b_m=\frac{2(\alpha_{xz}+\alpha_{zx})}{d}\int_0^d\frac{\partial T}{\partial z}\cos\frac{\pi mz}{d}dz.$$

Problem (4) can be solved by the standard methods for an ordinary differential equations (a particular solution of the inhomogeneous equation can be found by taking Fourier transforms in x). From the solution of this problem we easily find $\Delta(z)$ to be

$$\Delta(z)=\sum_{m=0}^{\infty}\Delta_m\cos\frac{\pi mz}{d}.$$

The expression for Δ_m is

$$\Delta_m=\frac{2b_m d}{\pi m}\exp\left[-\pi m\left(\frac{L-a}{d}\right)\right]\qquad m\neq 0,$$
$$\Delta_0=2ab_0=2a(\alpha_{xz}+\alpha_{zx})\frac{T(d)-T(0)}{d}. \tag{5}$$

It can be seen from (5) that the series $\Sigma\Delta_m\cos(\pi mz/d)$ converges very rapidly (exponentially). Furthermore, the correction corresponding to Δ_1 is already less than $10^{-30}\Delta_0$. We can thus restrict the discussion to the zeroth term. We find

$$\Delta(z)=a(\alpha_{xz}+\alpha_{zx})\frac{T(d)-T(0)}{d}\equiv\text{const}. \tag{6}$$

It can thus be concluded that, despite the change in the sign of $\partial T/\partial z$ on the interval $0<z<d$, we have $\Delta(z)\equiv$const in this interval, and the averaging over z which we mentioned above is completely legitimate. We also see that for this film thickness the magnitude of the anisotropic thermal emf is essentially independent of the particular $T(z)$ profile, being determined exclusively by the difference between the temperatures of the film boundaries.

We thus turn to a calculation of the time-varying signal corresponding to the experimental situation. Integrating the x component of expression (1) over z from 0 to d, we find

$$(S\sigma)^{-1}I\,dx=\langle E_x\rangle dx-\frac{(\alpha_{xz}+\alpha_{zx})}{2}\frac{T(d)-T(0)}{d}$$
$$\times\,\theta(a_2-x)\theta(a_1+x)dx. \tag{7}$$

Here S is the cross-section area of the film. The first term on the right side is none other than the work performed by the electric field in moving a unit charge a distance dx. In other words, dU is by definition the differential of the voltage. The second term is the work performed by external forces, i.e., the differential of the emf. Using $dx/\sigma S=dR$, where the right side is the resistance of a part of the film of length dx, we thus find the canonical form of the macroscopic Ohm's law:

$$IdR=-dU+dE,$$
$$dE=(\alpha_{xz}+\alpha_{zx})\frac{T(d)-T(0)}{d}dx. \tag{8}$$

A film substrate mounted on a grounded base (as in the experiments) can be represented by the equivalent circuit in Fig. 4, i.e., by a distributed RC line. This figure shows $dC=\varepsilon l dx/4\pi d_1$, where d_1 is the substrate thickness, and l is the width of the strip along the y direction. Since the signal is taken from the film by a 50-Ω cable matched with the oscilloscope input, the latter is equivalent to simply a resistance

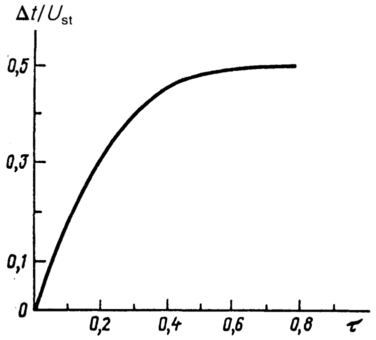

FIG. 5.

$R_{in} = 50\ \ \Omega$. When we supplement Eq. (8) with the condition which relates the current through an elementary "capacitor" to the voltage across it, we find the system of equations

$$-\frac{\partial U}{\partial x} = rI - G(x,t), \quad G = \frac{\partial E}{\partial x},$$
$$\frac{\partial I}{\partial x} = -C\frac{\partial U}{\partial t}. \tag{9}$$

Here r and C are the resistance and capacitance per unit length of the film along the x direction. Along with the boundary conditions $U(0,t) = 0$, $U(L,t) = R_{in}I(L,t)$, and $U(x,0) = 0$, Eqs. (9) lead to the following mixed problem:

$$\frac{\partial U}{\partial \tau} = \frac{\partial^2 U}{\partial \xi^2} - L\frac{\partial G}{\partial \xi},$$
$$U(0,\tau)=0, U(\xi,0)=0, \tag{10}$$
$$\frac{\partial U}{\partial \xi}(1,\tau) = -\frac{R}{R_{in}}U(1,\tau).$$

Here we have switched to the dimensionless variables $\xi = x/L$, $\tau = t/RC$. In addition, for convenience we have placed the origin for the x scale at test contact A (Fig. 1). Problem (10) can be solved easily by separation of variables. The solution is

$$U(x,t) = \sum_{n=1}^{\infty} A_n(t)\sin\lambda_n x,$$

where λ_n are the roots of the equation $\tan\lambda_n = (R/R_{in})\lambda_n$. The final expression for the observable signal $\Delta(t)$ is

$$\Delta(t) = U(1,t) = \int_0^t \Delta_{st}(\tau)F(t-\tau)d\tau. \tag{11}$$

Here $\Delta_{st}(t)$ is the steady-state solution in (6), in which we have substituted the instantaneous value $[T(d,t) - T(0,t)]$. The explicit expression for the function F is fairly complex:

$$F(t) = \sum_{n=1}^{\infty} \frac{2(\lambda_n^2+1)}{\lambda_n^2+2}\,\frac{\sin\lambda_n\xi_2 - \sin\lambda_n\xi_1}{\xi_2-\xi_1}\exp(-\lambda_n^2 t). \tag{12}$$

Here we have $\xi_{1,2} = a_{1,2}/L$, where $a_{1,2}$ are the coordinates of the boundaries of the illuminated strip. We see that series (12) converges exponentially rapidly at $t \neq 0$. At $t = 0$, in contrast, it diverges ($\lambda_n \approx -\pi/2 + \pi n$ at large n). This singularity at $t = 0$, which is quite natural (a δ-function singularity), unfortunately prevents us from approximating (12) by a few terms. To estimate the actual "instrumental function" of the film with the substrate, we thus restrict the discussion to the assumption $U_{st}(t) = \text{const}$ at $t > 0$. We then find

$$\Delta(t) = \sum_{n=1}^{\infty} \frac{2(\lambda_n^2+1)}{\lambda_n^2(\lambda_n^2+2)}\,\frac{\sin\lambda_n\xi_2 - \sin\lambda_n\xi_1}{\xi_2-\xi_1}[1-\exp(-\lambda_n^2 t)]\,U_{st}. \tag{13}$$

This series, which has the asymptotic behavior $(-1)^n/n^2$ at large values of n, converges quite rapidly—as $1/n^3$. Figure 5 shows the results of a numerical evaluation of the first four terms of this series (for $R/R_{in} = 1$, as in the experiments). We see that the time scale for the relaxation of $\Delta(t)$ to a steady-state value is $\tau^* \approx 0.3$. Under the actual experimental conditions ($d_1 = 1$ mm, $b = 3$ mm, $\varepsilon = 300$ for $SrTiO_3$, $R = 50\ \Omega$), we have $RC = 6$ ns. This result thus corresponds to a time $t^* \lesssim 2$ ns. We found the same width for the instrumental function in an indirect experimental determination of this function, by applying square voltage pulses 5 ns from a generator to the test contacts.

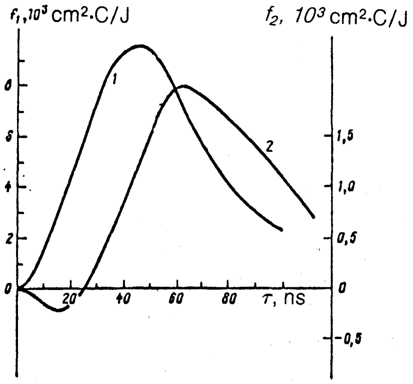

FIG. 6. *1*—Time evolution of f_1; *2*—that of f_2.

Looking ahead a bit, we note that the time scale for the variations of $U_{st}(t)$ is at least 15 ns, so we can set $\Delta_{st}(t) = \text{const}$ in (11). We find the approximate (but very accurate) result

$$\Delta = \frac{R_{in}}{R+R_{in}}\Delta_{st}(t) \approx \frac{1}{2}\Delta_{st}(t). \tag{14}$$

DISCUSSION OF EXPERIMENTAL RESULTS

In order to make the comparison with experiment, we are left with the task of finding an explicit expression for $\Delta_{st}(t)$, i.e., $\Delta T(t) = T(d,t) - T(0,t)$. To find this expression, we need to solve the standard problem of heat transfer in the film and the substrate, with a heat source $Q_0\exp(-\varkappa z)$ in the film, where $\varkappa = 2\cdot 10^5\ \text{cm}^{-1}$ is the absorption coefficient of the film. Unfortunately, solving a problem of this sort analytically[8] is extremely complicated. We have accordingly used the results of a numerical calculation of $T(z,t)$, reported in Ref. 8, for the same laser pulse length and the same film thickness as in the present experiments. We thus have

$$\Delta_{1,2}(t) = \frac{(\alpha_{xz}+\alpha_{zx})}{2}\frac{a}{d}Wf(t) = \frac{\alpha_{xz}+\alpha_{zx}}{2}\frac{Q}{bd}f_{1,2}(t). \tag{15}$$

Here W is the energy density of the incident light, Q is the total energy in the pulse, and $f_{1,2}(t)$ are universal functions for the experimental conditions. They are determined by the shape of the laser pulse and by the thermal constants of the film and the substrate, which we took from Ref. 7 (see Ref. 8). The subscripts 1 and 2 refer to the cases in which the light is incident from the side of the free surface of the film and from the substrate side, respectively. Figure 6, curves *1* and *2*, show results calculated for the functions f_1 and f_2, respectively. Comparison with experiment (compare curve *1* in Fig. 6 and Fig. 1b and curve *2* in Fig. 1 and Fig. 1c) reveals a good agreement between the calculated and experimental results. In particular, for the case in which the light is incident from the free surface of the film, there is a good agreement in the times at which the signal reaches its maximum value ($t \approx 40$ ns) and in the length of the trailing edge of the pulse. In the case in which the light is incident from the substrate side, we find a good agreement in terms of the calculated values of the point at which the signal reaches its minimum (at $t \approx 15$ ns), the zero value of $\Delta(t)$ (at $t \approx 25$ ns), and the signal maximum (at $t \approx 55$ ns, reckoned from the beginning of the laser pulse). The ratios $|\Delta_{\min}|/|\Delta_{\max}|$ for the experimental and calculated curves are also in satisfactory agreement.

Using the data in Fig. 6 (curve *1*) and Fig. 2, we can easily estimate the value of $\alpha_{xz} = \alpha_{zx}$ which we would need to find the best fit of the calculated results to the experimental data. This fit is achieved with $\alpha_{xz} \approx 1.5 \cdot 10^{-2} \mu V/K$. This value is smaller by a factor of about 30 than the diagonal components of the tensor α_{ik} for a $YBa_2Cu_3O_{7-\delta}$ single crystal, according to the measurements of Ref. 11. Such a small off-diagonal component α_{xz} could thus arise as the result of a deviation of the *c* axis of the film by only $\sim 2°$ in the *xz* plane during the deposition (Fig. 1a) from the normal to the substrate surface.

The good agreement between the experimental and calculated $\Delta(t)$ and $f_{1,2}(t)$ which we saw above, along with the reasonably small values of α_{xz}, as estimated from the experimental data, suggests that the laser-induced emf pulses observed here can be explained completely on the basis of an anisotropic Seebeck effect in Y–Ba–Cu–O films.

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