SCATTERING OF SURFACE ACOUSTIC WAVES AND HEAT TRANSFER THROUGH THE NONUNIFORM INTERFACE *

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(Получена 8 июля 1993 г. Принята к печати 9 июля 1993 г.)

Excitation of a thermal and acoustic transient grating along the nonuniform interface between a thin $YB_2Cu_3O_{7-x}$ film and its substrate is discussed. For the nonuniform interface, a ratio of the coherent acoustic to the thermal component is small, indicating the poor interface quality. Interface nonuniformity also manifests itself in the thermal relaxation kinetics. When the grating period is close to a size of the interface roughness, the coherent acoustic mode does not propagate, because of the strong scattering.

I. For a perfect contact between two solids, energy fransfer through the interface has to be limited by an acoustic mismatch between the two contacting materials [1], and heat transfer through the interface can be described by the boundary condition

$$k\frac{dT}{dz} = A\Delta T \tag{1}$$

which implies that heat is transferred through the interface with the «velocity» A, k is thermal diffusivity, ΔT is temperature jump on the interface. The numeric value of A, by order of magnitude, equals to the sound velocity: $A \sim v_s \sim k/\lambda_{ph}$, where λ_{ph} is phonon mean free path. However, because of the mismatch of the lattice structures, solid—solid interfaces are usually nonuniform [2]. We shall show that for a nonuniform interface, in the bulk of a thick sample, diffusion is still one-dimensional, with the boundary condition (1) where $A \sim k/d$, and d is the average size of the nonuniformity. In the case of a large ratio d/λ_{ph}, the value of A for the nonuniform interface may be several orders of magnitude less than the sound velocity.

To visualize the interface nonuniformity, the technique of photothermal excitation and probe [3], and the method of acoustic microscopy [4] were previously used. In this article, we discuss an alternative approach for the interface quality evaluation, where the thermal and acoustic excitations are combined. The approach is based on application of the improved version [5] of the transient grating (TG) technique [6]. In this technique, two coherent picosecond laser beams are crossed in the film. The third (probe) laser pulse is diffracted by TG in the direction specified by the photon energy and momentum conservation laws. By changing the probe delay time, one can define the relaxation kinetics of the excited media. The advantage of this approach is that both thermal and acoustic gratings are produced simultaneously. If an acoustic wave is excited along the interface, it is scattered by local random

* Хотя данная статья непосредственно касается ВТСП материалов, редколлегия сочла возможным опубликовать ее в журнале «Физика и техника полупроводников», поскольку рассматриваемые в ней вопросы могут оказаться актуальными и для полупроводников.

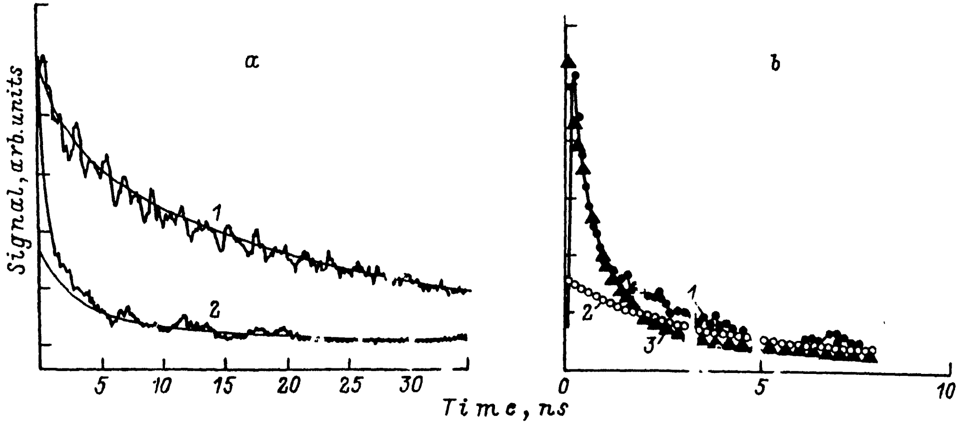

Fig. 1. *a* — transmitted (*1*) and reflected (*2*) diffracted signal for the transient grating induced in 350 nm thick $YB_2Cu_3O_{7-x}$ film on MgO substrate ($T = 300$ K), together with numerical fitting (smooth curves) [9]. *b* — initial portion of the reflection signal (*1*), fitted in the model of thermal barrier (*2*) and for ideal interface, $T = 0$ (*3*).

nonuniformities. For strong enough scattering, a component of the acoustic wave with the wave vector, equal to the excitation wave vector should disappear which is equivalent to the localization effect, discussed earlier for the other types of one- and two-dimensional waves [7]. Results of TG decay as well as the ratio of thermal and acoustic components excited in $YB_2Cu_3O_{7-x}$ thin films on different substrates are discussed below.

II. Lately, several articles were devoted to the problem of heat transfer through the interface between $YB_2Cu_3O_{7-x}$ thin films and different substrates [8–12]. For the TG experiments [8–10], $YB_2Cu_3O_{7-x}$ films were magnetron sputtered [13] on the optically polished substrates, with the *c*-axis normal to the substrate surface, and random *ab* orientation of the microcrystals in the film plane. In these experiments, the bulk and interface thermal and acoustic parameters (diffusivity $D_{ab} = 0.023$ cm^2/s in the film plane, diffusivity $D_c = 0.0028$ cm^2/s in *c*-direction, dispersion relations and damping of the surface acoustic waves) were measured. It was also found that the barrier for heat transfer defined by the parameter A in (1) is very high, almost two orders of magnitude higher than the acoustic mismatch model predicts. The measured value for A was ~8 m/s, instead of the theoretical estimate $\sim 10^3$ m/s (all numbers are for $T = 300$ K). Both reflection and transmission TG geometries [6, 14], with the grating excitation from surface and interface sides, were examined, and the whole experimental set including measurements for the free surface could be satisfactory explained [9, 10]. The only exception was reflection data from the interface which are especially sensitive to the quality of intimate contact between film and substrate. For the purpose of current discussion, Fig. 1 presents the results that only partially could be described in [9]. Curve *1* of Fig. 1, *a* represents the intensity of transmitted diffracted wave, and curve *2* — the intensity of the reflected diffracted wave, for the excitation from the interface side. Smooth lines show the quality of the fitting procedure using the one-dimensional diffusion equation with adjustable parameters k and A.

In the transmission geometry, the functional dependence of the transmitted diffracted wave amplitude is [5]

$$a_t \propto \int_0^\infty \Delta\varepsilon\,(x')\,dx', \tag{2}$$

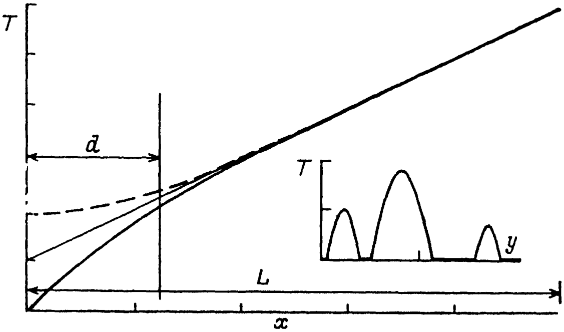

Fig. 2. Perturbation of steady-state linear temperature profile on absorbing black areas (solid line) and non-absorbing white areas (dashed line). Insert: steady-state temperature distribution on the interface: zero temperature on black areas and sinusoidal temperature profile on white areas.

where $\Delta\varepsilon\ (x)$ is the perturbation of a dielectric constant ε. Fitting the data Fig. 1, *a* with a one-dimensional model provides measurement of the barrier constant A. Results of these measurements agree with the measurements of the other authors [11, 12]. However, Fig. 1 shows that this model fails to explain both the small numerical value of A and the behavior of the reflected signal in the interface-excited geometry. Comparison of reflection and transmission data shows that only the long-time component of the reflected signal could be properly fitted.

In the reflection geometry, the signal amplitude a_r is [5]

$$a_r \propto \int_0^\infty \exp\left(2i\sqrt{\varepsilon}\ \frac{\omega\cos\theta}{c}\ x'\right)\Delta\varepsilon\ (x')\ dx'. \tag{3}$$

Reflected wave is formed inside a narrow layer $\sim c/\omega\sqrt{\varepsilon}$ close to the interface. Hence, the reflected signal is especially sensitive to the distribution $A\ (y,\ z)$ on the interface. One can see that the initial part of the curve *2* shows the fast temperature drop on the time scale $\theta < 1$ ns, which could not be fitted with any reasonable set of parameters A and k. In the same time, that initial stage of thermal relaxation could be described if one suggests that the thermal contact between the layer and substrate is perfect. Using formula for temperature kinetics with the perfect contact [15] ($T = 0$ on the interface),

$$\Delta T\ (x,\ \ t) = \frac{1}{2\sqrt{\pi kt}} \int_0^\infty \left\{\exp\left[-\frac{(x-\xi)^2}{4\ kt}\right] - \exp\left[-\frac{(x+\xi)^2}{4kt}\right]\right\} \exp\left(-\Omega\xi\right) d\xi \tag{4}$$

together with (3), for absorption depth $\Omega = 66$ nm [9], one obtains a result shown by the triangles in Fig. 1, *b*. With the boundary condition (1) incorporating the acoustic mismatch model the numerical results are practically indistinguisable from (4). Hence, the initial part of thermal relaxation shows a fast temperature drop in the narrow layer close to the interface (10% of the sample thickness).

To explain the experimental results, it is natural to suggest that the interface is nonuniform. Consider a film of thickness L, grown on a substrate with infinite thermal conductivity (the substrate temperature $T_s = 0$) characterized by nonuniform thermal and acoustic contact [16], with «black» background that have perfect contact and «white» spots that have no thermal and acoustic contact with the substrate. The interface plane ($x = 0$) is characterized by varying thermal conductivity both in the y- and z-directions with average size $\sim d$ for both black and white areas. The boundary conditions for heat transfer are (Fig. 2): $dT/dx = 0$ on the white areas, and $T = 0$ on the black areas (the acoustic mismatch is neglected). In the steady-state, the diffusion equation is converted to the Laplace equation

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = 0. \tag{5}$$

Close to the interface, for $x, y, z < d$, the solution of (5) is

$$T \sim \exp\left[i\,(k_x x + k_y y + k_z z)\right],$$

with the temperature distribution on the white spot

$$T \propto T_i \sin\left(\pi \frac{y - Y_i}{Y_{i+1} - Y_1}\right) \sin\left(\pi \frac{z - Z_i}{Z_{i+1} - Z_i}\right), \tag{6}$$

for $Y_{i+1} < y < Y_i$, $Z_{i+1} < z < Z_i$, and $T = 0$ on the black area, e. g. for $Y_{i+2} < y < Y_{i+1}$, $Z_{i+2} < z < Z_{i+1}$, (see insert in Fig. 2). The wave number k_x is imaginary, with the modulus equal to $\sim\pi/d$ which means that on depth d from the interface, the perturbation caused by the nonuniformity disappears (Fig. 2). The average temperature on the interface T_{av} is of the same order of magnitude as temperature on the depth d. If $d \ll L$, for $x \gg d$ the solution of (5) is a linear function of x

$$T \sim T_0 x/L, \tag{7}$$

T_0 — steady state temperature on the sample free surface. For $x \sim d$, the temperature is

$$T \sim T_{av} \sim T_0 d/L. \tag{8}$$

Substitution of (7) and (8) in (5) yields the effective speed of the heat flow $A \sim k/d$. In contrast to one-dimensional diffusion, where A would have been defined by the phonon mean free path, the experimentally defined A for a nonuniform interface might be less than the sound velocity by several orders of magnitude because of the temperature field nonuniformity. Specifics of the nonuniform interface manifests itself only on a short time scale $\theta \sim d^2/k$, being responsible for the three-dimensional diffusion in a thin layer $\sim d$ close to the interface. The lower limit for the white spot size following from this measurement is $\gg 1\ \mu m$. For the case d—L, computer calculations are needed.

III. Let us now discuss the acoustic component of the TG signal. When picosecond laser pulses are used to excite the TG fast thermal expansion locally accelerates the elastic media and creates two counter-propagating acoustic waves [17] with the wave vector $2\pi/FS$ where FS is the grating fringe spacing. For $YB_2Cu_3O_{7-x}$ films, the major part of the signal acoustic component was identified as a surface wave [8]. For long wavelength ($FS \gg L$) this wave is almost a Rayleigh wave of the substrate, for $FS \ll L$ it is almost a Rayleigh wave of the film. The initial portion of data, containing both the static (thermal expansion) and acoustic components is presented in Fig. 3. One can see that, even for a large fringe spacing, the acoustic modulation is less than ~15%. At small FS, the oscillating part decreases and practically disappears. In the same time, no significant acoustic damping was observed in the temperature range (17÷300) K.[1]

To our knowledge, only the thermal expansion component $\partial\varepsilon/\partial\rho$ is responsible for the signal amplitude [8, 9]. As follows from the virial theorem [18], the kinetic

[1] The TG experiment on $YB_2Cu_3O_{7-x}$ films was initially proposed to observe temperature dependence of the acoustic damping below the superconducting transition temperature [8].

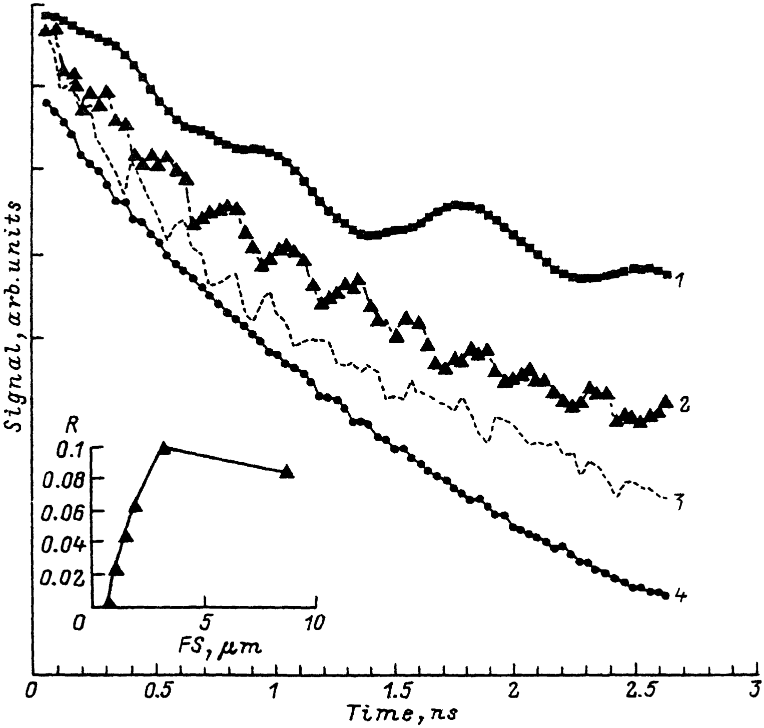

Fig. 3. Acoustic and thermal excitation components for 80 nm thick $YB_2Cu_3O_{7-x}$ film on MgO substrate (300 K) for fringe spacing, *μm*: *1* — 8.6, *2* — 1.4, *3* — 0.94, *4* — 0.68. Insert: acoustic to thermal component ratio *R* via fringe spacing.

energy of the acoustic mode, for the laser pulse duration θ short compared to the acoustic period τ, $\theta \ll \tau$, and negligible losses, equals the potential energy of the thermally expanded material. Experimentally, this excitation would result in 100% signal modulation: once a period the acoustic contraction virtually compensates for the thermal expansion. This effect was observed in many TG experimets [17, 19–21]. To explain the experimental data Fig. 3 one must assume that, beyond the coherent acoustic mode, the other modes, different from plane waves with the wave vector $2\pi/FS$, are excited. In the model of a nonuniform interface, one would expect that black and white areas act incoherently in the process of acoustic wave excitation. Plane waves are not the eigenmodes for the wave equation describing a nonuniform system,

$$\rho \frac{\partial^2 u}{\partial t^2} - m \frac{\partial^2 u}{\partial y^2} = \gamma \frac{\partial (\Delta T)}{\partial y},$$

where density ρ, elasticity m or a combination of the elastic modules γ are random functions of coordinates. Ratio of the acoustic mode energy to the energy of thermal expansion (equal to the total vibrational energy) is a measure of the interface nonuniformity.

By changing the fringe spacing, one changes the conditions for the acoustic wave excitation on the nonuniform interface. For large *FS*, the acoustic wavelength is large compared to the interface «roughness» *d*, and the interface is practically uniform. The short pulse creates local vibrations which are added almost coherently, such that the entire kinetic energy is proportional to the TG length. For $FS \sim d$, all the local vibrations acquire different phases, and the resulting amplitude is, roughly speaking, proportional to the square root of TG length. To accumulate the difference in phases $\sim\pi$, a time interval τ is required such that $\tau \sim 1/\Delta\omega$, where $\Delta\omega$ is the spectral width of the acoustic spectrum. If the excited frequency is less than the typical interface mode frequency, this phase shift will be accumulated during the first acoustic period. Disappearance of the acoustic component is similar to the «edge of mobility», or localization of the acoustic mode. However, the observed behavior is different from the standard localization experiments [7, 22], where continuous excitation and detection of a given frequency signal is produced, rather than a short pulse of specific wave vector.[2] When *FS* approaches the average size of the interface spot, all the local vibrations acquire different phases, and the acoustic component amplitude decreases, allowing the estimation of the average size of the interface scatterers (Fig. 3), which in this case is $\sim(0.7 \div 1)$ μm. These numbers are similar to the estimate of the black and white spot sizes obtained from the heat transfer measurements.

In principle, acoustic scattering might be caused by the grain boundaries or other imperfections. However, different films grown on MgO substrates showed similar acoustic modulation (~0.1), and films grown on $SrTiO_3$, showed [16] almost twice stronger modulation (~0.2). That correlates with the fact of much better intimate contact of $YB_2Cu_3O_{7-x}$ film with $SrTiO_3$ than with MgO substrate [13].

In conclusion, TG excitation of a system with nonuniform interface allows for observation of acoustic wave scattering and specifics of heat transfer. This type of characterization is especially useful for transparent substrates and substantially nonuniform thermal contacts, such as $YB_2Cu_3O_{7-x}$ films on different substrates or diamond films on silicon. Thin semiconductor films, paints on dielectric surfaces and thin biological objects are other obvious candidates for the future research.

The author is grateful to C. Herring, A. Efros and B. Shklowsky for valuable discussions.

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