Propagation of nonequilibrium phonons in aluminum-oxide ceramics fabricated by cold isostatic pressing

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Propagation of slightly nonequilibrium phonons in aluminum-oxide ceramics fabricated by cold isostatic pressing has been studied. Assuming that phonon propagation in ceramic grains is ballistic, we have analyzed characteristics of the phonon scattering and drawn some conclusions about the nature of grain boundaries.
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In the previous publication[1] we proposed a diffusion model for propagation of slightly nonequilibrium phonons at liquid-helium temperatures in one-phase ceramic materials, taking into account their penetration through grain boundaries. We found out that it was possible to draw conclusions about the nature of phonon scattering on grain boundaries and their structure from the maximum bolometer output as a function of temperature measured in the thermal-pulse mode.[2]

In this work we have studied one modification of single-phase ceramics based on corundum (Al_2O_3), which is extensively used in industry as a stable abrasive material and is fabricated by isostatic pressing under pressures of up to 400 MPa and subsequent annealing at 1900 °C for one hour. This fabrication technique allows one to get rid of thin amorphous layers and states with higher impurity concentration between grains, which are usually produced when plastifiers are added or owing to the impurity diffusion, possibly unintentional, towards grain boundaries during the long synthesis time. Therefore we expected to detect a difference between the modes of thermal-pulse propagation in this material and in traditional corundum ceramics[3], and to obtain information about the nature of grain boundaries. The density of the ceramics fabricated using this technique was 3.72 g/cm^3, which is about 95% of the density of the Al_2O_3 single crystal.

We took microphotographs of samples of studied ceramics in order to obtain qualitative characteristics of the grains and boundaries between them. A typical photograph is shown in Fig. 1. The major fraction of the grains in this ceramics are close-packed, and most of them look like crystallites. A statistical analysis of grains yielded the average grain dimension $R_0=(2.3\pm0.8)\times10^{-4}$ cm.

More details about the structure of grain boundaries can be seen on transmission pictures recorded by a scanning electron microscope. Our data have indicated that more than 90% of grain boundaries are well ordered (see Fig. 2a, where the typical separation between atoms is about 0.3–0.4 nm), and the width of the disordered (amorphous) phase between grains is less than 100 Å (Fig. 2b).

Corundum ceramics can be considered as a model material in our research. The phonon free path l in Al_2O_3 at liquid-helium temperature is large (more than 1 cm in single crystals), which is considerably larger than the typical ceramic grain dimension $R\cong10^{-3}$ cm, and the condition $l\gg R$ is indispensable in one version of the theory,[1] which is based on the assumptions about ballistic phonon propagation within one grain and its reflection from the grain surface or transmission across a grain boundary.

The experimental technique was described in detail in the earlier publication.[2] Let us recall that a gold film is deposited in vacuum on one side of a ceramic plate; this film is heated by a current pulse and acts as a source of nonequilibrium phonons. A tin bolometer shaped as a meandering strip occupying an area of 0.3×0.25 mm^2 is fabricated on the opposite side of the sample. The studied temperature range was 1.7–3.8 K. The thermal power released in the heater is sufficiently small that $\Delta T\ll T_0$ holds, and in the analysis of experimental data the temperature of the injected phonons could be taken equal to that of the ambient.

A set of curves illustrating the thermal pulse propagation in samples of different lengths is shown in Fig. 3. We have analyzed them in order to prove that the phonon propagation in the material is controlled by diffusion, i.e., the theoretical model is adequate to describe the experimental data. The delay of the phonon signal maximum detected by the bolometer is proportional to L^2, where L is the dimension of the tested sample along the direction of phonon propagation (see the insert in Fig. 3), which indicates that phonons spread across the sample diffusely. The shapes of the curves in Fig. 3 for longer delays, when the signal amplitude S was expected to be proportional to $t^{-1/2}$ (the configuration of our experiments was planar), could not be accounted for in these terms. In reality we found $S\propto t^{-n}$, where $n\cong0.2$–0.25, i.e., the signal dropped off with time more slowly than predicted by the diffusive law. This fact should be taken into account in analysis and interpretation of experimental data.

Curves of thermal pulse propagation at several temperatures are given in Fig. 4. The fact that the curves are close to those of integrated thermal pulses indicates that the signal shape depends weakly on temperature. In our analysis of experimental data we differentiated the curves, determined the points of zero derivative, i.e., t_m, and plotted this param-

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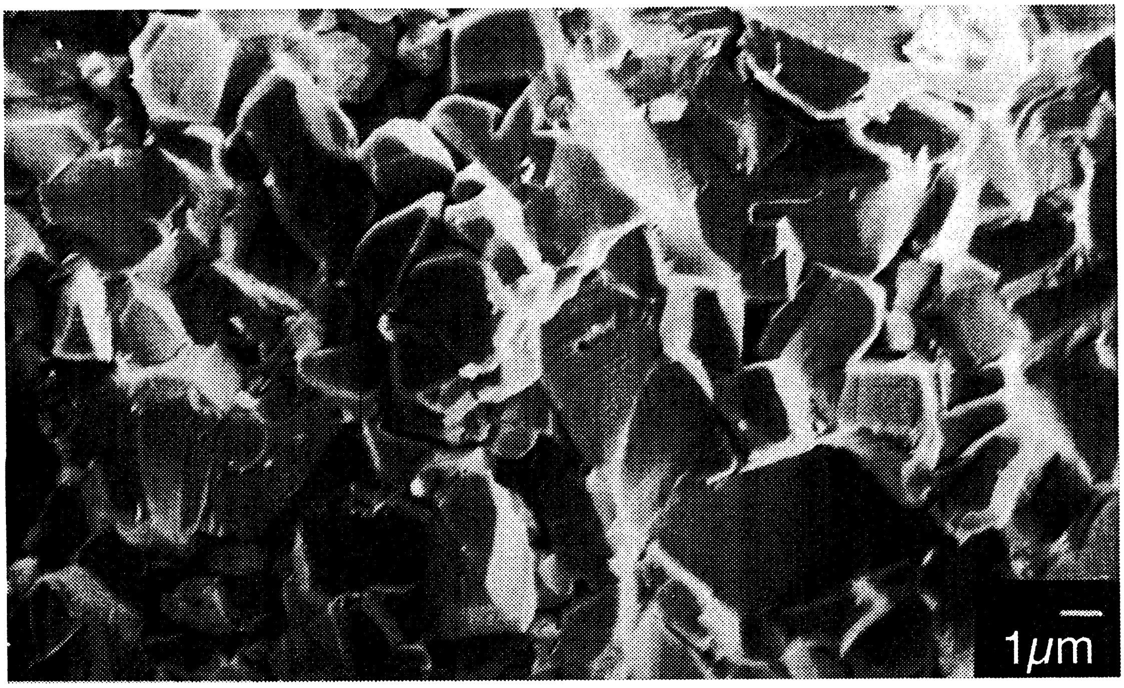


FIG. 1. Electronic micrograph of cleaved surface of an Al_2O_3 ceramic sample fabricated by cold isostatic pressing.

eter as a function of temperature. In the case of the curves of Fig. 4 and in several other samples t_m slightly increased as the temperature deceased. As we have mentioned above, the signal drops more slowly for $t > t_m$. This leads to broadening of the detected peak, and when the data are accumulated using a computer, its maximum is determined with an uncertainty of about 20%. Within this error t_m may be considered temperature–independent. This is possible[1] if the passage of phonons across grain boundaries is controlled by acoustic impedance matching, which, generally speaking, should be expected in samples with a highly-ordered structure of grain boundaries (Figs. 2a and 2b).

Let us give some estimates relevant to our experimental data. The time of the bolometer signal maximum[1] is determined by the relation

$$t_m \propto L^2 / D_{\mathrm{eff}}, \tag{1}$$

where D_{eff} is the effective diffusion factor. The phonon propagation within a grain is considered ballistic, and the lifetime within the grain is t_0, which implies the effective diffusion factor

$$D_{\mathrm{eff}} \propto R^2 / t_0, \tag{2}$$

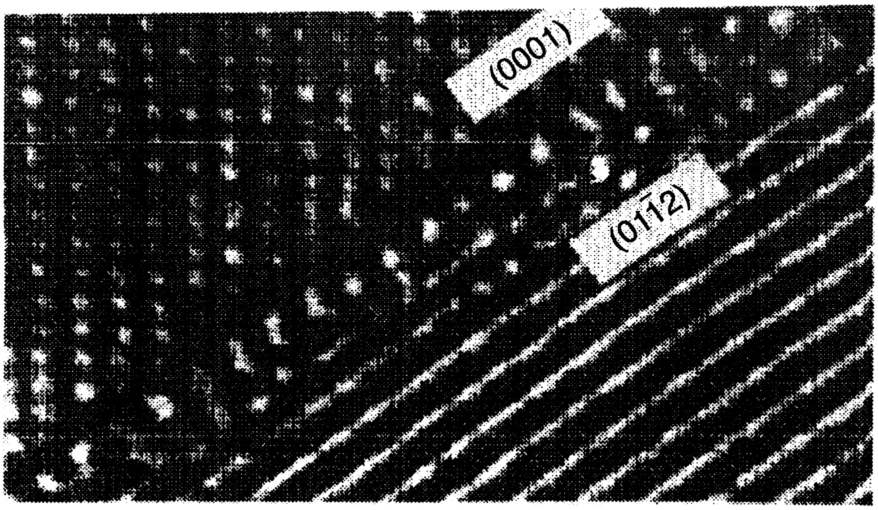
where R_0 is the average ceramic grain dimension. The time t_0 can be estimated as

$$t_0 \propto \frac{RS}{V_s \Sigma f_\omega}. \tag{3}$$

Here V_s is the average phonon velocity, S is the grain surface area, Σ is the total area of contacts (junctions) with neighboring grains, and f_ω is the transmission probability for a phonon with a frequency ω across a contact surface. In the acoustic impedance matching model, the transmission probability f is independent of the phonon frequency (wavelength).

Using the curves of Fig. 4, we derive from Eq. (1) an estimate for the effective diffusion factor $D_{\mathrm{eff}} \cong 18\ \mathrm{cm}^2/\mathrm{s}$, which is two orders of magnitude smaller than, for example, in yttrium-aluminum garnets doped with rare-earth ions, which act as scattering centers.[4] Given the average phonon velocity of 7.25×10^5 cm/s, the combination of Eqs. (1) and (2) yields the value of the parameter $(S/\Sigma) f_\omega^{-1}$, which measures the effect of grain boundaries on the phonon diffusion, approximately equal to 9.

Let us estimate $(S/\Sigma) f_\omega^{-1}$ numerically. Various models of the pore configuration in the ceramics with a given "weight" porosity of about 5% yield $(S/\Sigma)_{\max} \cong 1.1$, and we find the phonon transmission probability $f_\omega \cong 1.2 \times 10^{-1}$, i.e., only about 12% of the phonons incident on a grain boundary pass to the next grain. How can this value be interpreted? What model should be applied to the grain boundary? According to the data of Figs. 2a and 2b, the thickness of the spacer between grains with regular crystalline structure is much less than the wavelength $\lambda_{\mathrm{ph}} \cong 500$–$700$ Å of the dominant group of phonons (see Ref. 5) in the emitted thermal pulse. A thin and irregular interface between grains results in phonon scattering similar to that from fluctuations of atomic mass or elastic constants, as was first noted by Steg and Klemens.[6] According to some estimates,[1)] the probabilities of small-angle scattering (passage across an interface) and backscattering (reflection from a boundary) are approximately equal because all grains are composed of the same material, so the density of state, which is an important parameter in scattering, is also equal in different grains. There-



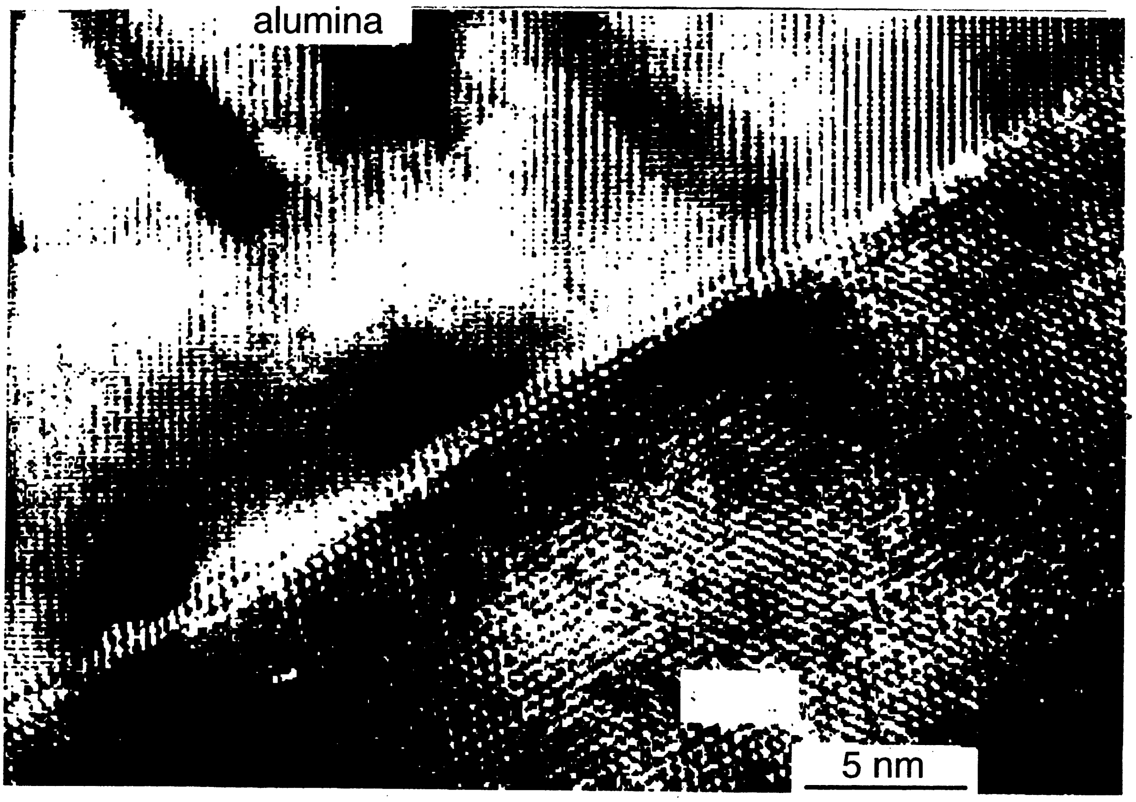


FIG. 2. (a) Transmission scanning electron microscope picture of a grain junction, in which single atoms can be seen; (b) transmission electron microscope pattern of a grain boundary with a poorer ordering of atoms in the interface, when atoms are slightly displaced, but the interface thickness is smaller than 100 Å.

fore the probability of phonon transmission across the boundary is $f_1 \cong 0.5$.

A phonon (longitudinal or transverse) passing across an interface enters a grain with a different orientation of crystal axes (Figs. 2a and 2b). A calculation of the probability of phonon transmission across an acoustic boundary between two arbitrarily oriented crystallites was undertaken by Mel'nikov *et al.*,[7] who obtained $f_2 \cong 0.8-0.9$.

In the analysis of phonon propagation across grain boundaries, conversion of phonon modes, namely the generation of surface waves, which may be quite important in ceramic materials, is usually ignored. If the irregularity dimension L in the interface plane (we follow the notation of Ref. 8) is smaller than the wavelength of the dominant pho-

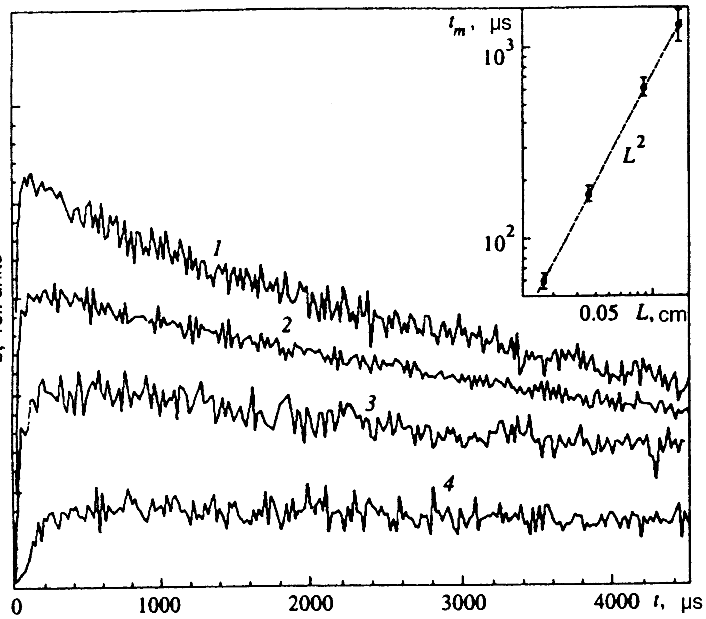


FIG. 3. Signals from a bolometer versus time at $T=3.8$ K and (*1*) $L=0.25$, (*2*) 0.45, (*3*) 0.86, and (*4*) 1.34 μm. The insert shows the time t_m as a function of the sample length.

non group generated by a thermal pulse, each irregularity acts as a source of surface waves. In the case of periodic surface roughness and low phonon frequency the problem was solved by Gulayev and Plesskii,[9] and the result was tested in experiments.[10] If the source is point-like ($L \ll \lambda_{ph}$), about 70% of its power is transformed to surface waves,[11] i.e., the generation efficiency is very high, and in our notation $f_3 \approx 0.3$.

The total probability of phonon transmission across an interface between two grains is the product of transmission probabilities on each stage and is equal to $f = f_1 f_2 f_3 \approx 0.12$–$0.15$, which is in a good agreement with measurements.

The real ceramic structure is, certainly, different from the present model in terms of both grain shapes and the structures of the junction pads between them. The estimates derived from this model, however, agree with the experimental curves, which provides a fair justification of the model.

In conclusion, we should note that our measurements of thermal-pulse propagation in aluminum-oxide ceramics provide evidence that boundary conditions between ceramic grains are close to those of acoustic impedance matching for thermal phonons, i.e., they do not contain large inclusions or structural defects.

We have suggested a model of short-range irregularities on the interface between grains as an efficient converter of three-dimensional phonons to surface waves and vice versa. As a result, the propagation of a thermal flow across a ceramic sample becomes slower and the trailing edge of the signal is broadened. The efficiency of an irregularity on an interface as a source of surface acoustic phonons should increase with decreasing temperature because the condition $L \ll \lambda_{ph}$ will be valid for a large fraction of phonons. One may expect that the signal maximum should shift to the bigger values, and the trailing edge flattens as the temperature drops.

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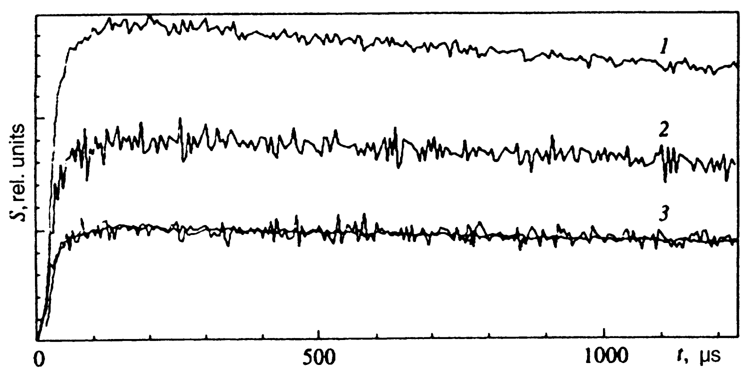

FIG. 4. Signals from a phonon detector in a sample with $L=0.045$ cm: (*1*) $T=3.8$ K; (*2*) $T=3.4$ K; (*3*) superposition of curves recorded at $T=3.8$ and 3.0 K.

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