Scanning-acoustic-microscopy study of the microstructure of a superconducting ceramic

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(Submitted 26 January 1988)
Pis'ma Zh. Eksp. Teor. Fiz. **47**, No. 5, 267–270 (10 March 1988)

Acoustic images of the surface and of the surface layer of the Y-Ba-Cu-O yttrium ceramic have been obtained with a resolution $\sim 1\ \mu$m. An interpretation of the images is offered. The velocity of a Rayleigh wave at the surface of an individual crystallite is estimated by quantitative acoustic-microscopy methods.

Scanning acoustic microscopy has been used in recent years to measure the local elastic properties and other physical characteristics of a variety of entities.[1-3] Images have been obtained of both the surfaces and surface layers of materials, including optically opaque materials, at depths up to several hundred microns. The resolution on the acoustic images has ranged from 20 μm to 0.5 μm, depending on the particular ultrasonic frequency used. Local values of the sound velocity and attenuation and also the local anisotropy of samples are being measured at the same resolution.[4]

Methods of acoustic microscopy are of major interest for studying high-temperature superconducting materials, since these methods offer the possibility of studying elastic and other physical properties of the sample as a whole as well as the properties of the individual crystallites which make up the sample. These methods make it possible to study the degradation and aging processes, to establish a correlation between the elastic behavior of crystallite and structural and phase transitions in them in measurements at different temperatures, and so forth.

As a first step we have analyzed the microstructure of the Y-Ba-Cu-O yttrium ceramic. The experiments were carried out on a reflection acoustic microscope at frequencies in the range 1–1.3 GHz (at a resolution $\sim 1\ \mu$m) and 0.2 GHz (resolution $\sim 7\ \mu$m).

We studied samples with both polished and natural surfaces. The measurements were carried out at room temperature ($T = 20\,^\circ$C). As the immersion fluid we used water (density $\rho = 1$ g/cm^3, sound velocity $c = 1.5$ km/s). Figure 1 shows acoustic images of the structure of a polished surface of a sample (Fig. 1a) and also an image obtained when the acoustic lens was moved 1.6 μm closer to the object (Fig. 1b), with a resolution $\sim 1\ \mu$m. We can clearly see the microstructure of the ceramic: the individual crystallites and the pores between them. The regions occupied by crystallites are the relatively bright regions, and the pore regions are the dark regions. When the focal point of the acoustic lens as displaced to a depth of $(1–2)\lambda$ (λ is the acoustic wavelength), the images of the crystallites remained essentially the same; the only change was the appearance of permanent contours on the crystallites, as a result of interference effects. On the other hand, moving the focus improves the resolution of the

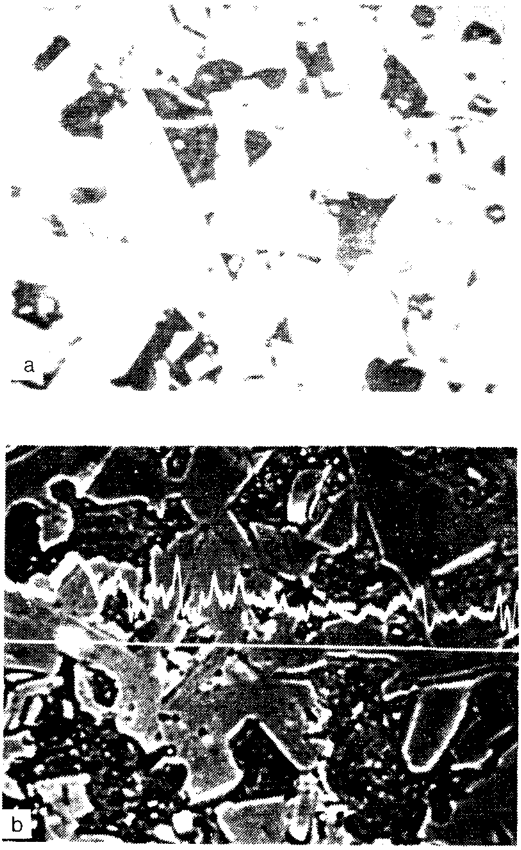

FIG. 1. Acoustic images of a polished sample of an yttrium ceramic at the frequency $f = 1.3$ GHz (the resolution is ~ 1 μm; the field of view is 100×80 μm). a: The surface ($z = 0$). Bright regions—Crystallites; dark regions—pores. b: Image obtained when the lens was moved 1.6 μm closer to the object ($z = -1.6$). The output signal during scanning in one dimension is superimposed on the acoustic images.

internal structure of a pore, which is apparently associated with the presence of finely dispersed material in the pore.

The contrast of an acoustic image is determined by the particular features of the local reflection of sound. Working from the data of Ref. 5 on the integral density and the integral sound velocities in the yttrium ceramic ($\rho = 6$ g/cm^3, $c_L = 4.2$ km/s, and $c_T = 2.4$ km/s), we can determine the nature of the reflection of focused sound at the interface of water with an individual crystallite. A focused beam is a set of ultrasonic rays within an aperture angle $\theta = 50°$. For rays incident normally, the reflection coefficient R is close to $1{:}R = (\rho_T c_L - \rho c)/(\rho_T c_L + \rho c) = 0.9$. It is relatively insensitive to variations in the parameter values either at the surface of an individual crystallite or as we go from one crystallite to another. The angular dependence of the reflection coefficient is quite weak up to a critical angle θ_T, determined by the transverse sound velocity in the sample. $[\theta_T = \sin^{-1}(c/c_T)]$. For the yttrium ceramic, the critical angle $\theta_T = 39°$ falls within the aperture of the acoustic lens. Rays incident at angles close to the critical angle excite a discharging Rayleigh wave, which, being reradiated, is received by the lens. However, the effect of the Rayleigh wave on the formation of the output signal (V) of the microscope is important only if the z-surface of the object is displaced significantly away from the focus along the direction toward the lens. In our case, the Rayleigh wave begins to have an effect at $z > 2$–$3\ \mu$m. Consequently, the significant value of the reflection coefficient when the surface on the sample is inside the focus causes the crystallite regions to be seen as bright regions with sharp boundaries.

The acoustic images of the regions between the crystallites (the so-called pores) are formed as a result of the reflection of a focused ultrasonic beam from the boundary of the immersion fluid which penetrates the pore to a certain depth. In principle, such a boundary is an ideal reflector, but the presence of a finely dispersed powder of the initial material of the ceramic in the pore leads to an effective scattering of the ultrasound and to a sharp reduction of the output signal from the acoustic microscope. Another factor tending to reduce the signal during the formation of the image of a pore is the occurrence of repeated reflections from the inner faces of a pore. As a result, the reflection from the pore regions is considerably weaker than the reflection from the crystallites, and these regions are seen as dark regions on the acoustic photographs. The backscattering during diffraction of the focused beam by small particles of the material forms an image of the internal structure of a pore (Fig. 1).

Our interpretation of the bright and dark regions on the acoustic images is confirmed by measurements of the behavior of the output signal V as a function of the displacement of the z-surface of the sample with respect to the focus (Fig. 2). In the case of a crystallite (curve 1 in Fig. 2) the output signal is formed as the result of an interference of the specularly reflected rays with rays which have been reradiated from the surface by the Rayleigh wave. The profile of this signal along the coordinate has the form of damped regular oscillations with a period determined by the velocity (c_R) of the Rayleigh wave at the surface of the crystallite.[1] Using the expression[6]

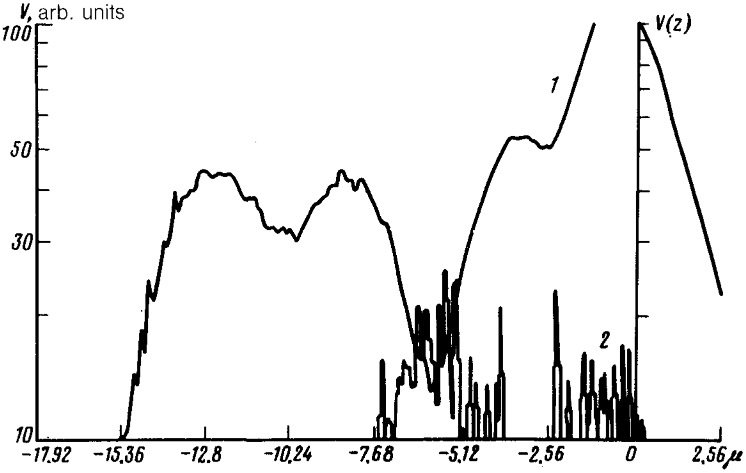

FIG. 2. $V(z)$ curves. 1—Crystallite region; 2—pore region (the frequency is $f = 1$ GHz).

$$\Delta z = \frac{\lambda_0}{2\left(1-\sqrt{1-\frac{c^2}{c_R^2}}\right)}$$

for the distance (Δz) between the maxima of the $V(z)$ curve, where λ_0 is the wavelength in water, we estimated $c_R = 2.3$ km/s, is close to the value calculated for c_R from the values of c_L and c_T found by integral methods in Ref. 5.

The behavior of the output signal in the vicinity of a pore is fundamentally different (curve 2 in Fig. 2). The signal takes the form of random oscillations, and its amplitude is substantially smaller. The oscillations do not have a characteristic period. The output signal cannot be interpreted on the basis of a model of the reflection of converging acoustic beams from plane surfaces. This behavior is apparently due to a scattering of sound by irregularities with dimensions smaller than or on the order of the acoustic wavelength.

These results demonstrate the promising outlook for the use of acoustic microscopy for studying the microstructure of high-temperature superconducting ceramics and for measuring the local elastic properties of materials of this sort.

We wish to thank L. A. Chernozatonskiĭ and V. A. Zaĭts for a useful discussion of these results.

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Translated by Dave Parsons