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Acoustic microscopy of ferroelectric ceramics

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Ferroelectric ceramics have been observed in an acoustic microscope. The images demonstrate the ability of the microscope to image elastic properties and to penetrate opaque layers.

1. Acoustic microscopy

The advantages of the acoustic microscope [1] lie first in the distinctive nature of the contrast, which arises from the mechanical and elastic properties of the object, and second in the ability of ultrasonic waves to penetrate material which is opaque to light and electrons. Both of these features are employed in the images which are described here, and both are areas of important future development.

The images presented here show (a) a comparison of optical and acoustic pictures of flaws in a transparent ceramic, (b) grain structure in a polished but unetched surface, (c) contrast under an opaque surface film.

2. Materials

Two transparent ferroelectric ceramics were studied. The first was barium–lead–sodium–niobate (BPNN), of composition $(Ba_{1-x}Pb_x)_2NaNb_5O_{15}$, made by sintering in oxygen.

The second was lead–lanthium–zirconate–titanate (PLZT), first made by Haertling and coworkers in 1970 [2]. The general chemical formula is $Pb_{1-x}La_x(Zr_yTi_z)_{1-0.25x}O_3$, where $x \simeq 0.04$. The specimen was prepared by sintering using hot pressing in an oxygen environment. It was then cut, ground and polished, and gold electrodes were evaporated onto the surface. It contains a much lower density of defects than the BPNN.

3. Defects

The BPNN material contained a relatively high density of defects, which appear in the micrographs in Fig. 1. Fig. 1a is an optical picture. The defects which are in sharp focus lie on the surface; they have been exposed by polishing. Because the ceramic is transparent defects in the bulk are also visible, the deeper they lie the less well focused they appear. Fig. 1b is an acoustic image with the microscope focused on the surface. The microscope was operated at 730 MHz, giving a wavelength in water of 2 μm, so that the acoustic resolution approaches that obtained optically. The surface breaking flaws appear equally clearly in both images. There are also some features which appear bright in Fig. 3b, these may be flaws lying just below the surface so that a reflection from the flaw sums with the reflection from the surface.

Much of the interest under this heading lies in what can be seen as the microscope is focused below the surface. This is shown in Fig. 1c and 1d; The specimen is moved 5 μm towards the lens relative to its position in Fig. 1c. A flaw becomes apparent in Fig. 1d (marked with an arrow) which is not visible at the surface.

Because this ceramic is transparent, it is possible to compare optical and acoustic images of subsurface defects. This enables experience and confidence to be gained for the more important task of imaging subsurface flaws in opaque materials. The lens used in the present study has been designed for imaging surface features; other lenses are now being constructed which will be more suitable for imaging beneath surfaces, in

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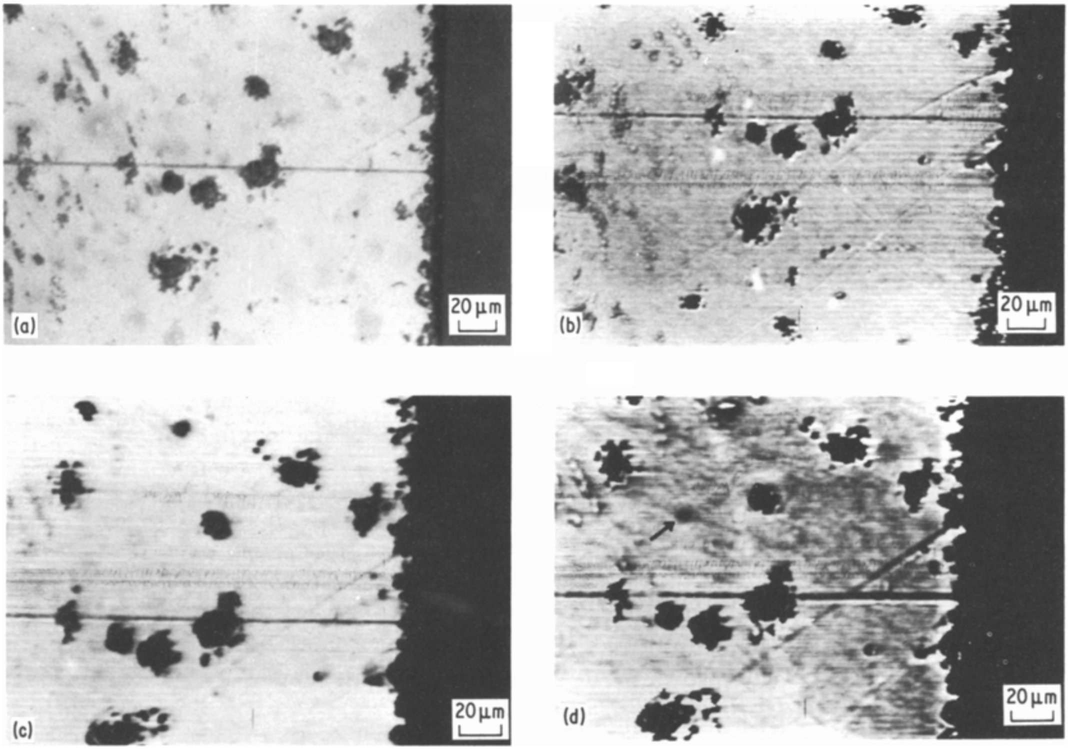

Figure 1 BPNN surface : (a) optical, (b) to (d) acoustic. The material is transparent; in the optical picture the less well-focused defects lie below the surface (the total specimen thickness is 0.17 mm). In the acoustic picture (b) some defects appear as bright areas. For (d) the specimen was moved 5 µm towards the surface relative to its position in (c). The image display system used for the acoustic pictures is a temporary one. Fine horizontal lines and a slight crazing of portions of the screen which appear on every picture are spurious and should be disregarded.

order to exploit the ability of the microscope to image inside solids.

4. Grain structure

If the surface of a polycrystalline material is examined in the acoustic microscope, different crystallographic orientations give rise to different intensities of reflected signals; this has been previously observed for metals and alloys [3]. In Fig. 2 the grain structure is revealed in an acoustic micrograph of a high-quality PLZT surface. The grain size is shown to be approximately 8 µm.

The acoustic images are quite different from optical images of grain structure. Since the ceramic is transparent, no features appear on the surface of an unflawed polished specimen. The grain boundaries may be revealed by etching with a reagent which dissolves them preferentially, but the contrast is then between grain and grain boundary, rather than between grains themselves. In acoustic images, however, it is the difference in orientation which gives rise to contrast between grains. The elastic properties of single crystals of PLZT and of related ceramics have never been measured, because of the difficulty of obtaining a crystal large enough to be measured using conventional apparatus. Acoustic microscopy offers a means of studying the elastic properties of individual grains.

An approximate estimate may be made from the

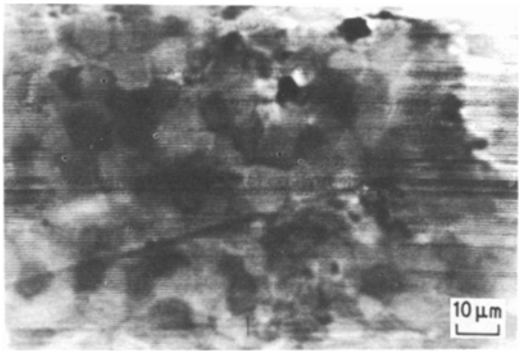

Figure 2 PLZT surface, showing grain structure. There is a gold film, 0.1 µm thick, on the specimen in the top right-hand corner of the picture.

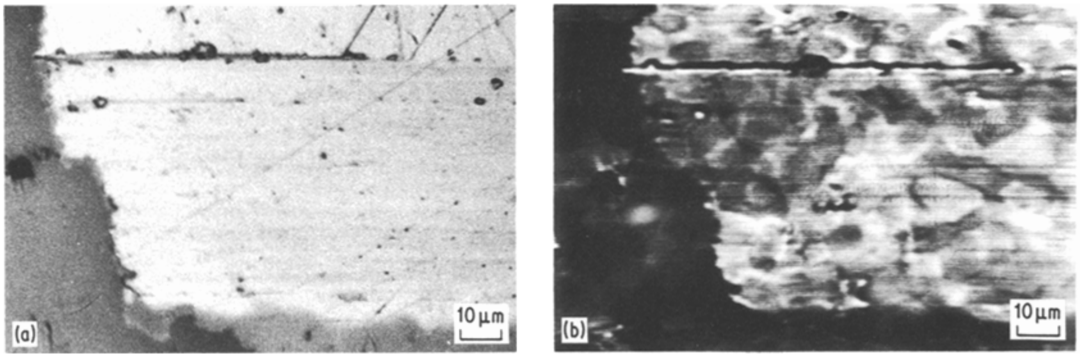

Figure 3 PLZT surface; (a) optical, (b) acoustic. The optical image shows a brighter gold film, 0.1 μm thick, on a darker ceramic surface.

present studies. The coefficient of reflection from a boundary between two materials of impedance Z_1 and Z_2, respectively, is

$$R = \frac{Z_2 - Z_1}{Z_2 + Z_1}.$$

The impedance is $Z = \rho v$ where ρ is density and v is the velocity of sound (in this case of longitudinal waves). For water $Z = 1.53$ Mrayl, and for polycrystalline PZT (the closest material to PLZT for which elastic properties have been measured) the isotropic average impedance is approximately 31 Mrayl*. In measurements of the PLZT surface the maximum difference in the amplitude of reflections from different grains was $\Delta R/R = 0.02$. (The contrast in Figs 2 and 3b has been enhanced electronically.) From the above formula, if $Z_2 \gg Z_1$, then

$$R \simeq 1 - \frac{2Z_1}{Z_2}$$

and

$$\Delta R \simeq \frac{2Z_1}{Z_2^2}\Delta Z_2.$$

Hence the variation in impedance deduced from the observed variation in reflections is 20%, and since density is independent of orientation this corresponds to a change in longitudinal wave velocity of 20% for the extremes of orientation.

The above approximate analysis was applied for normally incident ultrasonic waves. It may be extended using established methods [4, 5] to account for waves incident over the range of angles (in the present case 0°–60°) permitted by the aperture of the lens. If techniques can then be developed to give angular resolution of the intensity of reflected waves, together with spatial resolution, then it may be possible to determine elastic properties, including crystallographic orientation, of individual grains. This would be a significant extension of the progress that has already been made in this direction [5–7].

5. Opaque films

Acoustic images may be obtained through materials which are opaque to light and to electron beams. The lighter region in the top right corner of Fig. 2 is a gold film 0.1 μm thick, and grains can be observed through this film.

Comparison between optical and acoustic pictures may be made in Fig. 3. The optical image shows a gold film (bright) on the ceramic (darker); a gold film on the underneath surface is also visible through the ceramic near the bottom of the picture; the gold is again 0.1 μm thick, and the total specimen thickness is 0.25 mm. The optical image is otherwise featureless except for surface scratches, etc. In the acoustic image, surface defects are also apparent, but there is in addition considerable information about the grain structure, similar to that in Fig. 2.

In addition to being transparent to acoustic waves, thin surface films may even sometimes enhance imaging of surfaces, when, for example, they are of intermediate impedance and quarter-wave thickness. This effect may be analysed using conventional impedance arguments. In the present

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*Megarayls (Mrayl) are an S.I. unit of acoustic impedance, which is the ratio (— traction)/(particle displacement velocity). 1 rayl ≡ 1 kg m^{-2} sec^{-1}.

case the gold film is much less than one wavelength thick and the effect is small; gold has an impedance of 61 Mrayl, which is higher than that of PLZT, and, as can be seen, the film slightly increases the brightness of the reflection from the surface.

6. Conclusions

The ability of the acoustic microscope to image elastic properties of ceramics, such as flaws and grain structure, has been demonstrated. The microscope is able to image through surface layers which are opaque to light and electrons. Important areas of future development are to increase the ability of the microscope to image defects and microstructures in the bulk of a material, and to enable quantitative measurements of surface elastic properties to be made.

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