A TECHNIQUE FOR STUDYING SMALL DEFORMATIONS OF THE CRYSTAL LATTICE BASED ON THE SHADOW EFFECT

B. G. AKHMETOVA, Yu. M. PLETS, and A. F. TULINOV

Nuclear Physics Institute, Moscow State University

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The sensitivity of proton patterns to small deformations of the unit cell of a crystal is investigated. A $BaTiO_3$ single crystal was used as the target. The change due to the transition from the cubic to the tetragonal phase has been determined.

1. THE methods employed to study the structure of crystals are at present based on the diffraction of various types of radiation (x rays, neutrons, and electrons). At the same time, it has been shown in recent years that there exists a group of phenomena connected with the interaction of fast charged particles (protons, deuterons, etc.) which exhibit practically no diffraction but are nevertheless sensitive to the structure of the lattice. This sensitivity can be observed rather clearly on "proton patterns" (or, more generally, "ion patterns") which represent images obtained on photographic plates of the angular distributions of heavy particles scattered by single-crystal samples.[1,2] The proton pattern contains a system of spots and lines with a lowered blackening density ("shadows") along the directions of the axes and crystal planes respectively. The fact that the traces of the planes appear on the plates in the form of thin straight lines results in great advantages for investigating by this method the geometric characteristics of the unit cell, and in particular of its small changes under the influence of various factors. We describe below the results of an experiment confirming this statement.

2. The object of investigation was a ferroelectric crystal of barium titanate ($BaTiO_3$). It is well known that at a temperature above 120°C this crystal has the perovskite structure (Fig. 1) with a lattice constant of 4 Å. On lowering the temperature the symmetry of the crystal changes as a result of a second-order phase transition (at T = 120°C) from the cubic to the tetragonal phase. The transition is accompanied by an increase of the lattice constant in the direction of the spontaneous polarization (c = 4.036 Å) and by a corresponding decrease perpendicular to it (a = 3.992 Å). It is seen that the spontaneous deformation c/a in this case amounts to about 1%. The purpose of the experiment was to observe this deformation with the aid of proton patterns.

It follows from simple geometric considerations that it is most convenient to set up an experiment in which the spontaneous polarization axis of the target crystal is normal to the plane of the photographic plate. It can then be expected that the shadows corresponding to planes with relatively low indices will appear on the plate as shown qualitatively on Fig. 2. The shadows belonging to the cubic phase are shown as solid lines, those of the tetragonal—by dashed lines. The shadows in Fig. 1 are constructed on the basis of the geometrical model of the lattice. The thickness of the lines corresponds qualitatively to the packing density of the planes. It is seen from Fig. 2 that the phase transition leads to a shift on the proton pattern of lines which do not pass through the center (the ⟨001⟩ axis). The deformation of the unit cell can be determined from these shifts.

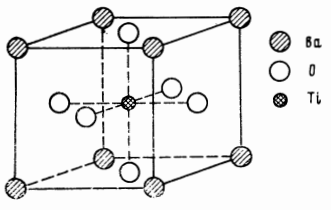

FIG. 1. The unit cell of barium titanate ($BaTiO_3$).

3. The measurements were carried out with a beam of protons accelerated with the aid of the NIIYaF MGU [Scientific Research Institute for Nuclear Physics, Moscow State University] cascade generator up to 500 keV. The experiment is shown schematically on Fig. 3. The collimator system made it possible to obtain a beam whose diameter in the region of the target did not exceed 0.3 mm. The single-crystal sample of $BaTiO_3$ was mounted in the center of a vacuum cham-

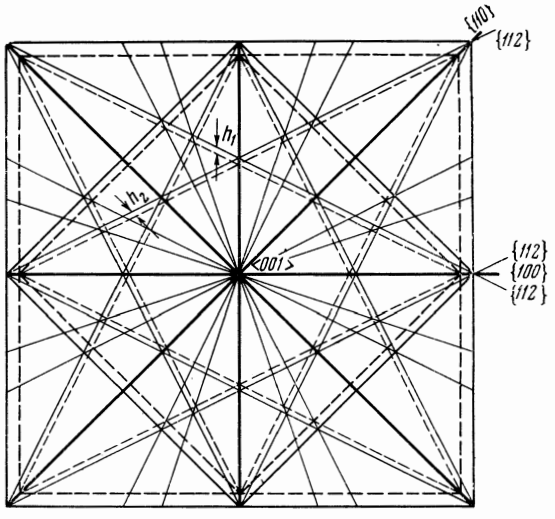

FIG. 2. Schematic distribution of lines of intersection of the crystallographic planes of the barium titanate lattice with the plane of the photographic plate. The continuous lines correspond to the cubic phase (T > 120°C), the dashed lines – to the tetragonal phase (T < 120°C).

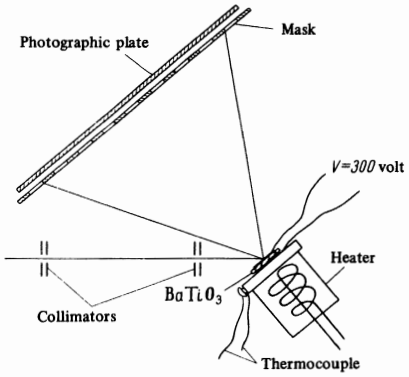

FIG. 3. Schematic diagram of the experiment.

ber on a holder which could simultaneously be used as a heater. The temperature of the sample was measured with a thermocouple. The scattered protons were registered with a photographic plate of the type MR which was placed within the vacuum chamber 65 mm from the target. The irradiated sample was oriented in such a way that its ⟨100⟩ axis was normal to the surface of the photographic plate and the crystallographic {110} plane coincided with the horizontal plane. The proton pattern obtained under those conditions at T = 150°C is shown in Fig. 4. It is seen that lines which do not pass through the center of symmetry, in particular those corresponding to planes of the {112} type (see Fig. 2), appear rather clearly. It was the shift of these lines which was considered.

4. Measurements with the tetragonal phase were carried out at room temperature. It is well known that as a rule in this case the $BaTiO_3$ single crystal consists of a large number of domains with different directions of the spontaneous polarization. In our experiment it was essential to use a single-domain crystal. The single domain was produced by keeping the crystal both before and during the irradiation in an external field with an intensity of 15–20 kV/cm. For this purpose both sides of the single-crystal sample cut normal to the ⟨100⟩ axis were sputtered with aluminum in the form of thin films. A voltage of 300 V was applied to these aluminum electrodes. A control analysis carried out optically confirmed that under these conditions the sample consisted of a single domain; the polarization axis coincided with the direction of the normal of the platelet.

5. Because the shift of the {112} lines in the transition from one phase to the other is extremely small, it was useful to obtain both proton patterns on the same plate. To this end we placed in front of the photographic plate a screen with vertical slits of the form shown in Fig. 5. The screen could be horizontally displaced by the width of the slits. The crystal was irradiated with the screen in the two positions at room temperature and at T = 150–200°C respectively. The results of such double irradiation were obtained for two different crystal orientations. In one case the trace of the {100} plane was horizontal and in the other—the trace of the {110}. In the first case we determined the shift of h_1 (see Fig. 2) and in the second case that of h_2. As an example we show on Fig. 6 the experimentally obtained double proton pattern referring to the second case. Here even bands correspond to the tetragonal and odd bands to the cubic phase.

The shadows obtained on the photographic plates were analyzed with a MF-4 microphotometer. Figure 7 shows, as an example, the variation of the blackening density on traversing one of the lines of the protons pattern. As a coordinate marking the position of the center of the straight-line shadow we used the point of intersection of the lines tangent to the inner slopes of the curve. The error amounted to ~0.05 mm. With the aid of such measurements we compiled two-dimensional maps of points belonging to the four {112} pairs of lines. Because it is impossible to show these maps on

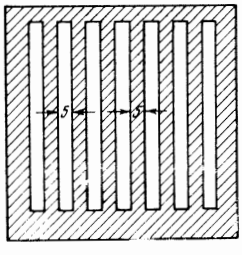

FIG. 5. Multislit screen.

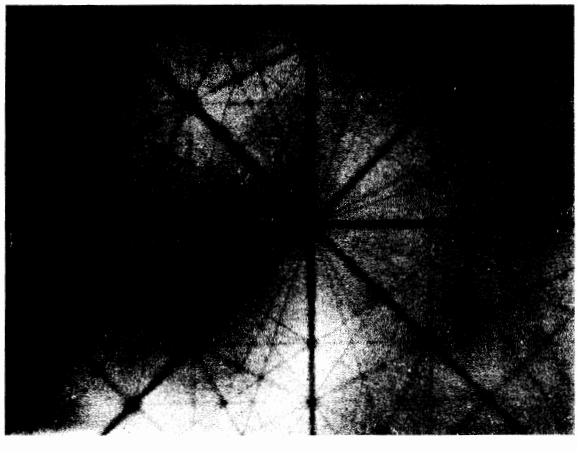

FIG. 4. Proton pattern of $BaTiO_3$. The ⟨110⟩ axis points from the center of the photographic plate; the shadow of the {110} plane is horizontal.

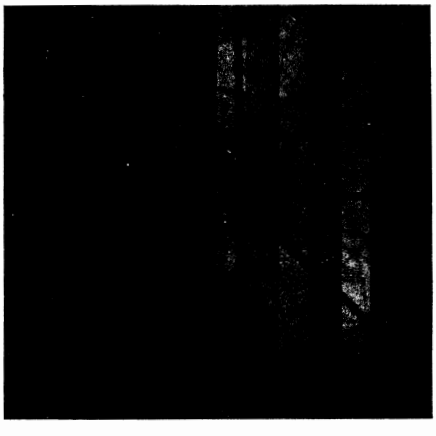

FIG. 6. Proton pattern of $BaTiO_3$ obtained with the aid of the multislit screen by the double exposure method. The even (light) bands correspond to the cubic and the odd (darker) ones – to the tetragonal phase.

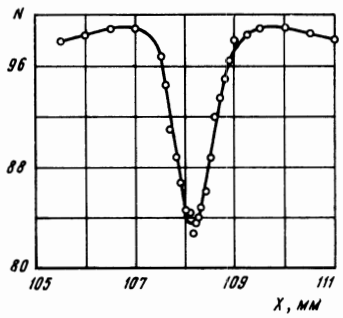

FIG. 7. The dependence of the blackening density of the photographic emulsion (in relative units) on the coordinate of the photometered section (according to the microphotometer scale) on crossing the shadow of the {110} plane.

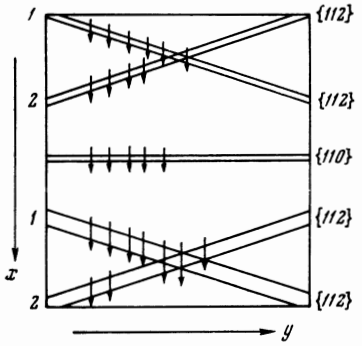

FIG. 8. Schematic location of the straight lines drawn through the experimental points (the line splitting is magnified tenfold). The arrows show the positions and directions of the photometry.

a small scale, we restrict ourselves to presenting a single purely illustrative diagram (see Fig. 8). On it arrows show the regions and directions of the photometering. The straight lines are drawn through the points obtained with the absolute value of the shifts on the diagram being magnified tenfold. Along with {112} lines we plotted on the maps the central horizontal lines. The fact that in the case of the horizontal lines we also observed splitting attests to the presence of some accidental shift unconnected with the phase transition. The absolute value of the shift amounted to 0.12 ± 0.04 mm. One must obviously use this value as a correction to the splitting of the lower and upper lines on the diagram. After carrying out this procedure the splitting of the upper and lower lines turned out to be equal within experimental error. For the case illustrated in the diagram (horizontal {110} axis) this splitting was h = 0.33 ± 0.06 mm. A calculation based on a geometrical model with c/a = 1.01 yields a value of h of 0.30 mm. It is seen that there is agreement within the limits of the measurement error. An analogous result was also obtained for the case when the {100} plane was horizontal.

In conclusion it should be noted that in this paper the authors have restricted themselves solely to the observation of the fact that the linear shadows shift. The next step will be to increase the accuracy of the measurements. This is in the first place connected with an increase in the quality of the photographic plates, since only then will it be possible to realize in full the strong point of the shadow technique which allows one to use as large a number of points as desired for constructing the straight lines.

[1] A. F. Tulinov, B. G. Akhmetova, A. A. Puzanov, and A. A. Bednyakov, ZhETF Pis. Red. 2, 48 (1965) [JETP Lett. 2, 30 (1965)].

[2] R. Behrisch, Canad. J. Phys. 46, 527 (1968).

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