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SOME PRELIMINARY CONSIDERATIONS CONCERNING THE EXPECTED OVER-ALL EXPANSION OF BARIUM TITANATE CERAMIC

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ABSTRACT

A preliminary mathematical study is made of the maximum expected expansion of barium titanate ceramic. It is noted that there is insufficient physical evidence for a complete answer and some possible mathematical models are discussed and compared.

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SOME PRELIMINARY CONSIDERATIONS
CONCERNING THE EXPECTED OVER-ALL EXPANSION
OF BARIUM TITANATE CERAMIC

1. Introduction. In a study of the piezo-electric properties of Barium Titanate the question may be raised as to what is the maximum expected expansion of a mass of ceramic Barium Titanate during polarization. Some preliminary calculations toward answering this question will be given in what follows. It should be emphasized that the answer depends very strongly on considerations of the geometric relationships of the component parts of the ceramic. There seems to be no available information about this subject. This is discussed in paragraph 4.

What is actually done here is to set up a procedure which will give answers when more information is available and to calculate results for a few possible forms of models.

2. Physical Background. Without delving deeply into the physical properties of Barium Titanate we can summarize the facts we need as follows:

(a) Barium Titanate crystals are, basically, strictly rectangular like those of salt. Thus they have three natural, mutually perpendicular axes implicit in them.

(b) Under the proper physical treatment a Barium Titanate crystal will respond by contracting from proportional dimension 1 to $1 - \delta$ along two of the axes, and expanding to $1 + 2\delta$ along the third. Note that, up to a second order term in δ , the rectangular volume is preserved.

(c) The expansion is carried out in terms of domains, substructures of the crystal whose existence can be verified experimentally. Each domain contains a great number of "molecules" which expand and contract in unison along the axes. That is, the domain expands along one and contracts along two axes as a whole.

(d) δ is found by experiment to be 1/3%. That is $\delta = .003$.

(e) When a Barium Titanate crystal is then completely polarized, the expansion of every domain changes from axis to axis in order to conform as closely as possible to expansion in the direction of polarization. This causes the crystal as a whole to expand, since unpolarized the expansions of the several domains cancel one another. The net expansion of the crystal will be 2δ , because 2/3 of the domains expand an amount δ .

(f) A mass of ceramic Barium Titanate is made up of numerous finely powdered crystals pressed together. It will be assumed that the axes of these crystals are randomly oriented with respect to the direction of polarization.

3. Mathematical Treatment. In order to find the expected expansion of a mass of ceramic the mean expected proportional expansion of a single domain (or crystal, or "molecule") should be calculated. The mean of these means should give the proportional expansion of the ceramic itself.

This mean expected proportional expansion (hereafter abbreviated m.e.p.e.) is a function, clearly, of the angle ϕ between the direction of polarization and the axis most nearly parallel to that direction. We shall write this function

$$\text{m.e.p.e.} = \delta(\phi).$$

Then the expected ceramic expansion is given by the relation

$$\text{expansion} = \int_0^{\frac{\pi}{2}} \delta(\varphi) \rho(\varphi) d\varphi$$

where $\rho(\varphi)$ is the relative density of axes most nearly parallel to the direction of polarization.

The difficulty lies in finding a reasonable value for $\delta(\varphi)$. This is discussed in paragraph 4. The function $\rho(\varphi)$ can be calculated and turns out to be

$$\rho(\varphi) = \begin{cases} 3 \sin \varphi & 0 \leq \varphi \leq 45^\circ \\ 3 \sin \varphi (1 - \frac{4}{\pi} \cos^{-1} \cot \varphi) & 45^\circ \leq \varphi \leq \arctan \sqrt{2} \\ 0 & \arctan \sqrt{2} \leq \varphi \leq 90^\circ \end{cases}$$

See the Appendix for the derivation of this result. A graph is given in Figure 1. Certain features of this should be intuitively clear. First, if $\varphi \leq 45^\circ$ there is no possibility of any other axes being closer to the direction of polarization. Second, if $\varphi \geq \arctan \sqrt{2}$ one of the other axes must be closer, thus the division into three expression is quite natural.

In paragraph 5 values of $\int \delta(\varphi) \rho(\varphi) d\varphi$ will be given for some possible values of $\delta(\varphi)$.

4. Evaluation of $\delta(\varphi)$. We suppose we have a domain (or crystal or "molecule") embedded in a ceramic mass and oriented so that one axis (which we shall call the azimuth axis) makes an angle φ with the fixed direction of polarization. We ask what is the m.e.p.e. of the domain contributed to

the ceramic expansion in the fixed direction. A number of values are available.

(a) We might take the domain as a line. Then $2/3$ of the time the length of domain expands 3δ , since $1/3$ of the domains are already expanded along the azimuth axis. Then we take the contribution of m.e.p.e. to be the length times the cosine of φ . That is,

$$\delta(\varphi) = 2\delta \cos \varphi. \quad (a)$$

This has the advantage of being simple, and the disadvantages of being heuristically unconvincing, and moreover, of not taking on the limit $\delta(\varphi) = 0$ when $\tan \varphi = \sqrt{2}$. This last point arises when we consider that if all the axes are equally far from the fixed direction no expansion is possible. This last configuration must occur if $\tan \varphi = \sqrt{2}$.

(b) Answering this last objection we may take an expression of the form $A + B \cos \varphi$ and fit it to the values 1 for $\varphi = 0$ and 0 for $\tan \varphi = \sqrt{2}$. Then we get

$$\delta(\varphi) = 2\delta \frac{\sqrt{3} \cos \varphi - 1}{\sqrt{3} - 1} \quad (b)$$

This procedure seems quite arbitrary.

(c) We might consider the domain as a rectangle box and measure the expansion from the highest point to lowest point. This expansion is

$$3\delta \frac{a \cos \varphi - b \sin \varphi \cos \theta}{a \cos \varphi + b \sin \varphi \cos \theta + c \sin \varphi \sin \theta}$$

7

$\rho(\varphi)$

$$3 \sin \varphi$$

$$0 \leq \varphi \leq 45^\circ$$

$$\rho(\varphi) = 3 \sin \varphi (1 - \frac{1}{\sqrt{2}} \cos^{-1} \cot \varphi)$$

$$45^\circ \leq \varphi \leq \arctan \sqrt{2}$$

$$0$$

$$\arctan \sqrt{2} \leq \varphi \leq 90^\circ$$

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Figure 1

0 10° 20° 30° 40° 50° 54.7° 60°
Angle φ
=arctan $\sqrt{2}$

where a, b, c are the dimensions of the box and θ determines the rotation of the other axes about the azimuth axis. We multiply this by $2/3$ and average over all possible a, b, c, θ . The result is given approximately by

$$\begin{aligned} \delta(\varphi) &\sim 2\delta \left(1 - \frac{6}{\pi} \tan \varphi\right) \quad \text{for } \varphi \sim 0, \\ \delta(\varphi) &\sim \delta \tan(\beta - \varphi) \quad \text{for larger } \varphi \end{aligned} \quad (c)$$

where $\tan \beta = \sqrt{2}$.

(d) We might consider the unit of expansion (group of domains or crystal) as a prolate spheroid, in which case we have to the first order in δ .

$$\delta(\varphi) = 2\delta \cos 2\varphi. \quad (d)$$

This is peculiar in that $\delta(\varphi) < 0$ for $\varphi \geq 45^\circ$. It is possible that such is really the case as a little thought should convince one that it is possible that a large longitudinal contraction could more than counter-act an expansion along the azimuth.

(e) Finally without any great motivation we might take a linear expression in φ which vanishes at $\tan \varphi = \sqrt{2}$. That is

$$\delta(\varphi) = 2\delta \left(1 - \frac{\varphi}{\beta}\right) \quad (e)$$

where $\tan \beta = \sqrt{2}$.

Figure 2 gives comparative graphs of these five functions. The values of

$\int \delta(\varphi) \rho(\varphi)$
are tabulated in paragraph 5.

$\delta(\varphi)$

9

Figure 2

Angle φ

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All of these functions suffer from the defect that no account is taken of the fitting between crystals of domains. It is even likely that the effects of inexact fitting are as large as those of expansion. Unfortunately there seems to be no available experimental data on this point; so we have had, of necessity, to ignore it in our calculations. It is to be hoped that this will be corrected soon.

5. Expected Expansions. Using the expression

$$\rho(\varphi) = \begin{cases} 3 \sin \varphi & 0 \leq \varphi \leq 45^\circ \\ 3 \sin \varphi (1 - \frac{4}{\pi} \cos^{-1} \cot \varphi) & 45^\circ \leq \varphi \leq \arctan \sqrt{2} \end{cases}$$

we get the following values for

$$\Delta = \int \delta(\varphi) \rho(\varphi) d\varphi :$$

(a)	$3\sqrt{2} (1 - \frac{2}{\pi} \tan^{-1} \sqrt{2}) \delta$	1.66 δ	.56%
(b)	$\frac{1}{\sqrt{3} - 1} (3\sqrt{6} - 2 - \frac{6\sqrt{6}}{\pi} \tan^{-1} \sqrt{2}) \delta$	1.20 δ	.40%
(c)	evaluated numerically	.54 δ	.18%
(d)	$\frac{2}{3} [4 \frac{\sqrt{3}}{\pi} - 1] \delta$	.80 δ	.27%
(e)	evaluated numerically	.78 δ	.26%

By all available criteria (d) is the best value for $\delta(\varphi)$, that is

$$\delta(\varphi) = 2\delta \cos 2\varphi .$$

227 9

It is the simplest case mathematically and most convincing physically. Moreover, the other physically plausible model (c) gives a very similar answer.

Appendix: Value of $\rho(\varphi)$. Here the expression

$$\rho(\varphi) = \begin{cases} 3 \sin \varphi & 0 \leq \varphi \leq \beta \\ 3 \sin \varphi (1 - \frac{4}{\pi} \cos^{-1} \cot \varphi) & 45^\circ \leq \varphi \leq \beta \\ 0 & \beta \leq \varphi \leq 90^\circ \end{cases}$$

will be derived.

Let us restate the problem in the following terms: We are given a fixed direction. We consider cartesian coordinate systems oriented at random with respect to the fixed direction. In each coordinate system let $\alpha_x, \alpha_y, \alpha_z$ be the angles between the x, y, and z axes respectively and the fixed direction. Let α be the least of $\alpha_x, \alpha_y, \alpha_z$. Then consider $A(\varphi)$, the portion of the α 's which are less than φ . Then $\frac{d}{d\varphi} A(\varphi)$ is the density of α at φ , that is, the limit as $\Delta\varphi \rightarrow 0$ of the portion of α between φ and $\varphi + \Delta\varphi$ divided by $\Delta\varphi$. In other words

$$\rho(\varphi) = \frac{d}{d\varphi} A(\varphi).$$

Let us parameterize the coordinate axes as follows: Let $\xi = \alpha_x$. Let θ be the angle between the Y axis and the plane containing the X axis and the fixed direction. Then

$$\cos \alpha_y = \sin \xi \cos \theta$$

which follows by considering the right spherical triangle cut out of a

sphere about the origin by the planes $x = 0$ $z = 0$ and the plane defined above. Similarly

$$\cos \alpha_z = \sin \xi | \sin \theta |$$

Considering the fact that the X-axis may be oriented so that it intersects a sphere about the origin with surface area 1 at any point in the hemisphere about the fixed direction, we see that the portion of

$\alpha_x = \xi \leq \varphi$ is $1 - \cos \varphi$, the area of spherical segment about the fixed direction of opening φ . So, since $1 - \cos \varphi = \int_0^\varphi \sin \xi d\xi$ the density of such ξ as a function of ξ is $\sin \xi$.

Now we need to determine α and take the mean over-all possible orientation of the coordinates.

There are three possible cases (I) $\alpha = \alpha_x$ (II) $\alpha = \alpha_y$ (III) $\alpha = \alpha_z$. Case (I) holds if $\alpha_y \geq \alpha_x$ and $\alpha_z \geq \alpha_x$, that is $\cos \alpha_y \leq \cos \alpha_x$ and $\cos \alpha_z \leq \cos \alpha_x$, or $\sin \xi \cos \theta \leq \cos \xi$ and $\sin \xi | \sin \theta | \leq \cos \xi$. Similarly, case (II) holds if $\sin \xi \cos \theta \geq \cos \xi$ and $\sin \xi \cos \theta \geq \sin \xi | \sin \theta |$. The last is equivalent to $\cos \theta \geq | \sin \theta |$ or $| \theta | \leq \frac{\pi}{4}$ for $-\frac{\pi}{2} \leq \theta \leq \frac{\pi}{2}$. Case (III) holds if $\sin \xi | \sin \theta | \geq \cos \xi$ and $| \theta | \geq \frac{\pi}{4}$. Thus the five curves determined by (a) $\cos \xi = \sin \xi \cos \theta$ (b) $\cos \xi = \sin \xi | \sin \theta |$ (c) $| \theta | = \frac{\pi}{4}$ divide the values of ξ and θ into the areas where the cases occur. Figure 3a shows this relationship. At the points where the three cases come together (P_1 and P_2 in Figure 3a) $\cos \xi = \sin \xi \frac{1}{\sqrt{2}}$ or $\tan \xi = \sqrt{2}$. If we let $\beta = 54.7^\circ = \arctan \sqrt{2}$ then $\xi = \beta$ at P_1 and P_2 .

From the symmetry of the situation in Figure 3a we see we need only average over one quarter of the θ , let us say $0 \leq \theta \leq \frac{\pi}{4}$.

Figure 3a

Figure 3b

Figure 3c

Now let us evaluate $A(\varphi)$. In order that $\alpha \leq \varphi$ we need either (a) $\cos \varphi \leq \cos \xi$ in case I, or (b) $\cos \varphi \leq \sin \xi \cos \theta$ in case II, for our restricted range of θ . This determines two areas in our $\theta - \varphi$ diagram. This is shown in Figures 3b and 3c for two of the three essentially different situations depending on whether $\varphi \leq \frac{\pi}{4}$ or $\frac{\pi}{4} \leq \varphi \leq \beta$ or $\beta \leq \varphi$. In the last situation where $\beta \leq \varphi$ we have $\alpha \leq \varphi$ for all possible θ and φ so that

$$A(\varphi) = 1 \quad \varphi \geq \beta$$

Recalling that the density of ξ is $\sin \xi$, the area of the square $0 \leq \theta \leq \frac{\pi}{4}$, $0 \leq \xi \leq \frac{\pi}{2}$ is

$$\int_0^{\frac{\pi}{4}} \int_0^{\frac{\pi}{2}} \sin \xi \, d\xi \, d\theta = \frac{\pi}{4},$$

so that $\frac{4}{\pi}$ times the shaded areas in Figures 3b and 3c gives $A(\varphi)$.

First suppose $\varphi \leq \frac{\pi}{4}$ then

$$A(\varphi) = \frac{4}{\pi} \int_0^{\varphi} \int_0^{\frac{\pi}{4}} \sin \xi \, d\theta \, d\xi + \frac{4}{\pi} \int_{\frac{\pi}{2} - \varphi}^{\frac{\pi}{2}} \int_0^{\cos^{-1} \frac{\cos \varphi}{\sin \xi}} \sin \xi \, d\theta \, d\xi.$$

Integrating somewhat we get

$$A(\varphi) = 1 - \cos \varphi + \frac{4}{\pi} \int_{\frac{\pi}{2} - \varphi}^{\frac{\pi}{2}} \sin \xi \cos^{-1} \frac{\cos \varphi}{\sin \xi} \, d\xi.$$

In order to get $\rho(\varphi) = \frac{d}{d\varphi} A(\varphi)$ we differentiate and

$$\rho(\varphi) = \sin \varphi + \frac{4}{\pi} \cos \varphi \cos^{-1} \frac{\cos \varphi}{\cos \varphi} + \frac{4}{\pi} \sin \varphi \int_{\frac{\pi}{2} - \varphi}^{\frac{\pi}{2}} \frac{\sin \xi}{\sqrt{\sin^2 \xi - \cos^2 \varphi}} d\xi.$$

The integral is easily evaluated by the substitution $\cos \xi = U$ and we get, since $\cos^{-1} 1 = 0$

$$\rho(\varphi) = \sin \varphi \left(1 + \frac{4}{\pi} \int_{\xi = \frac{\pi}{2} - \varphi}^{\frac{\pi}{2}} \cos^{-1} \frac{\cos \xi}{\sin \varphi} d\xi \right)$$

$$\rho(\varphi) = \sin \varphi \left(1 + \frac{4}{\pi} \frac{\pi}{2} \right) = 3 \sin \varphi$$

The situation of Figure 3c is more complicated; we have

$$a(\varphi) = \frac{4}{\pi} \left[\int_0^{\varphi} \frac{\pi}{4} \sin \xi d\xi + \int_{\varphi}^{\sin^{-1}(\sqrt{2} \cos \varphi)} \sin \xi \cos^{-1} \frac{\cos \varphi}{\sin \xi} d\xi + \int_{\sin^{-1}(\sqrt{2} \cos \varphi)}^{\frac{\pi}{2}} \frac{\pi}{4} \sin \xi d\xi \right],$$

and differentiating

$$\rho(\varphi) = \sin \varphi - \frac{4}{\pi} \frac{\sqrt{2} \sin \varphi}{\sqrt{1-2 \cos^2 \varphi}} \cdot \sqrt{2} \cos \varphi \cos^{-1} \frac{1}{\sqrt{2}} - \frac{4}{\pi} \sin \varphi \cos^{-1} \frac{\cos \varphi}{\sin \varphi}$$

$$+ \frac{4}{\pi} \frac{\sqrt{2} \sin \varphi}{\sqrt{1-2 \cos^2 \varphi}} \cdot \frac{\pi}{4} \sqrt{2} \cos \varphi + \frac{4}{\pi} \sin \varphi \int_{\varphi}^{\sin^{-1}(\sqrt{2} \cos \varphi)} \frac{\sin \xi}{\sin^2 \xi - \cos^2 \varphi} d\xi$$

The integral is evaluated as above and

$$\rho(\varphi) = \sin \varphi \left(1 + \frac{4}{\pi} \int_{\xi=\varphi}^{\sin^{-1}(\sqrt{2} \cos \varphi)} \cos^{-1} \frac{\cos \xi}{\sin \varphi} - \frac{4}{\pi} \cos^{-1} \cot \varphi \right)$$

$$\rho(\varphi) = \sin \varphi \left(1 + \frac{4}{\pi} \cos^{-1} \frac{\sqrt{1-2 \cos^2 \varphi}}{\sin \varphi} - 2 \cdot \frac{4}{\pi} \cos^{-1} \cot \varphi \right),$$

$$\text{but } \cos^{-1} \frac{\sqrt{1-2 \cos^2 \varphi}}{\sin \varphi} = \frac{\pi}{2} - \sin^{-1} \frac{\sqrt{1-2 \cos^2 \varphi}}{\sin \varphi}$$

$$= \frac{\pi}{2} - \cos^{-1} \frac{\cos \varphi}{\sin \varphi} = \frac{\pi}{2} - \cos^{-1} \cot \varphi,$$

so that

$$\rho(\varphi) = 3 \sin \varphi \left(1 - \frac{4}{\pi} \cos^{-1} \cot \varphi \right).$$

Since for $\varphi \geq \beta$ $A(\varphi) = 1$ we see that $\rho(\varphi) = 0$ and combining we get

$$\rho(\varphi) = \begin{cases} 3 \sin \varphi & \varphi \leq \frac{\pi}{4} \\ 3 \sin \varphi \left(1 - \frac{4}{\pi} \cos^{-1} \cot \varphi \right) & \frac{\pi}{4} \leq \varphi \leq \beta \\ 0 & \beta \leq \varphi \leq \frac{\pi}{2} \end{cases}$$

which was what we were trying to show.

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