

Space Charge Fields in BaTiO<sub>3</sub>

S. TRIEBWASSER

*Research Laboratory, International Business Machines Corporation, Poughkeepsie, New York*

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Experiments have been performed in which observations have been made in the presence of dc electric field bias of: (1) hysteresis loops, (2) double loops above the Curie point, (3) Kerr electro-optic effect above the Curie point and (4) capacitance above the Curie point in BaTiO<sub>3</sub>. The purpose of these experiments was to determine field distributions in the crystal under an externally applied difference of potential and to examine the extent to which space-charge surface layers cancel the bulk field. The first three types of observations were qualitative in nature. The biased hysteresis loops show normal behavior with no evidence of field cancellation. Double loop measurements show some evidence of field cancellation, while Kerr electro-optic measurements show strongly nonuniform fields indicating anomalous space-charge fields. The measurements of capacitance above the Curie point indicate that surface layers build up in the presence of a dc field, and these surface layers have a capacitance that varies with applied voltage. The results can be understood qualitatively if simple Schottky exhaustion barriers are assumed at the two metal electrode-crystal contacts. Observed asymmetries with respect to the applied fields are discussed.

## I. INTRODUCTION

A VARIETY of experimental results have been reported in which the existence of charged surface layers has been invoked to explain anomalies in the behavior of BaTiO<sub>3</sub> crystals: 1. Observation of a tetragonal surface layer above the Curie point,<sup>1</sup> 2. Thickness dependence of the coercive field,<sup>2</sup> 3. Frequency dependence of the dielectric constant during switching,<sup>3</sup> 4. Asymmetric pyroelectric effect,<sup>4</sup> and 5. Asymmetrical hysteresis loops.<sup>5</sup>

In addition, it has been reported in conversations among experimentalists in the field that application of dc electric fields to BaTiO<sub>3</sub> for an extended time results (at least in some samples) in surface layer space-charge accumulations. The charged layers, in turn, shield the bulk so that no macroscopic electric field exists in the body of the crystal even though there is a difference of potential across two opposite faces.

The experiments reported here were directed at acquiring an understanding of the character of such charged layers and their method of generation. Two favorable properties of BaTiO<sub>3</sub> were used to this end: (a) the highly nonlinear character of the dielectric constant of a ferroelectric exhibited in the temperature region just above the ferroelectric to paraelectric transition temperature; (b) the large Kerr electro-optic effect exhibited by BaTiO<sub>3</sub> in the same temperature region.

## II. PRELIMINARY EXPERIMENTS

Several preliminary experiments were performed to examine the question of the stability of BaTiO<sub>3</sub> under dc electric fields. The first experiment involved the application of a 60 cps switching sine wave superposed

on a dc electric field. Figure 1 shows the hysteresis loops observed under these circumstances. The purpose of the experiment was to ascertain whether or not there would be any drift in time of the apparent switching voltages (associated with each reversal direction) as would be expected if the dc field were to be cancelled by some surface space-charge field. Over time intervals of the order of twenty-four hours no such effect was observed. This is strange in the light of subsequent experiments above the Curie point which showed marked effects in times of the order of minutes.

The second experiment was performed above the Curie point in the double loop region.<sup>6</sup> In this case a biased ac signal was again applied so that the voltage swing was  $V_0 \pm V_1$  where  $V_1$  is greater than the bias voltage,  $V_0$ , and  $(V_0 + V_1)$  was large enough to drive the crystal from the paraelectric to the ferroelectric state but  $(V_0 - V_1)$  was not. The polarization vs electric field characteristics that result are shown in Fig. 2. Again, several crystals were cycled under these conditions for periods of about twenty-four hours with no apparent change in the observed curves. Others showed a shift of apparent transition field with a time constant

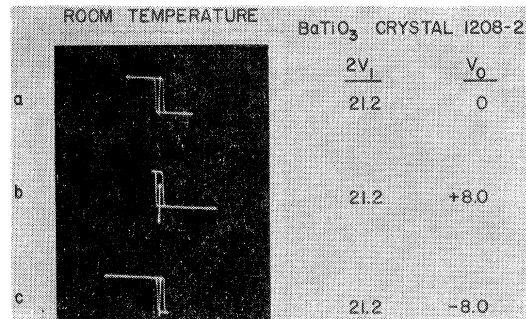


FIG. 1. Biased hysteresis loops. Crystal is 0.008 cm thick. Field is given by  $\bar{V} = V_0 + V_1 \cos(120\pi t)$ . Vertical line in loop represents  $V = 0$  point.

<sup>6</sup> W. J. Merz, Phys. Rev. **91**, 513 (1953).

<sup>1</sup> W. Kaenzig, Phys. Rev. **98**, 549 (1955).

<sup>2</sup> W. J. Merz, J. Appl. Phys. **27**, 938 (1956).

<sup>3</sup> M. E. Drougard, H. L. Funk, and D. R. Young, J. Appl. Phys. **25**, 1166 (1954); and private communication.

<sup>4</sup> A. G. Chynoweth, Phys. Rev. **102**, 705 (1956).

<sup>5</sup> Kugasu, Husimi, and Kataoka, J. Phys. Soc. (Japan) **12**, 432 (1957).

of the order of three minutes. Note that photographs (b) and (c) are taken under identical conditions except that the signs of  $V_0$  are opposite to each other. The kind of asymmetry of behavior demonstrated in these two traces showed up in most crystals examined, but was most pronounced when  $V_1 \leq V_0$ . In fact, when  $V_0$  was set equal to the average value required for the induced transitions from the ferroelectric state to the paraelectric state and back and  $V_1$  was made less than  $V_0$ , cycling between states (ferroelectric and paraelectric) ceased.

A third experiment was performed similar to those reported by Meyerhofer<sup>7</sup> and Koelsch.<sup>8</sup> The induced birefringence above the Curie point was observed under dc conditions. The reason such an experiment is of interest is that this phenomenon (quadratic Kerr electro-optic effect) represents the only means we have of arriving at a direct map of the macroscopic field distribution in the solid. This comes about because the induced birefringence is proportional to the square of the

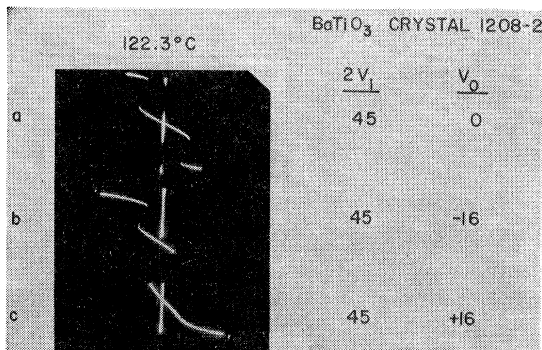


FIG. 2. Double loop traces.  $P$  vs  $V$ . Crystal 0.008 cm thick, temperature = 122.3°C,  $V = V_0 + V_1 \cos(120\pi t)$ . Vertical line represents  $V = 0$  point.

local electric field. In the absence of an applied field the crystal appears completely dark. Figure 3 shows typical results when a dc field is applied. In this case (as contrasted with the first two experiments described above) the electric field was applied in a (100) direction in the *plane* of a crystal plate rather than through the plate. The nominal field applied was considerably smaller than that required to induce a transition to the ferroelectric state. The two photographs differ only in the sign of field applied. The conclusion drawn from these latter experiments is that the field distributions cannot be understood unless the existence of gross inhomogeneities are assumed to exist in the crystals. This technique appeared quite interesting and would be most fruitful if applied to studies of imperfections.

The qualitative conclusions that were drawn from the three experiments discussed above are: (a) Contrary to our initial expectations the space-charge fields induced

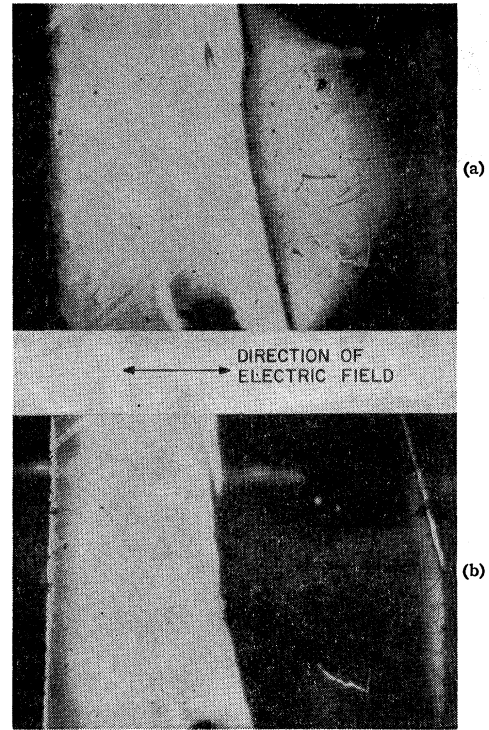


FIG. 3. Induced birefringence pattern in BaTiO<sub>3</sub> crystal at 125°C. Crystal is 0.3 cm × 0.3 cm × 0.016 cm with electrodes on 0.3 × 0.016 face. Field = 1500 v/cm. (a) and (b) differ only with respect to field direction.

by the application of a dc field to BaTiO<sub>3</sub> are not of such a nature as to *completely* cancel the applied field in the bulk. Evidence of some cancellation was observed. In general, behavior is not symmetric with respect to the direction of the applied field. (b) The actual field distributions as judged from the Kerr electro-optic effect can be quite complex. In unfavorable geometries induced strain patterns confuse the interpretation, but this technique can be quite useful in studying macroscopic field configurations.

### III. OPTICAL EXPERIMENTS

Further optical experiments were performed to study changes with time of field distribution in BaTiO<sub>3</sub> after the application of a step function of voltage. The geometry used was similar to that described in connection with Fig. 3. Again, the crystal was taken above its Curie point (into the cubic phase) and viewed between crossed polaroids. In the absence of fields the crystals appear dark (as expected). Upon application of an electric field, the induced birefringence causes a uniform brightness. In the course of time, space-charge fields build up and partially cancel the applied field so that the brightness decreases. As indicated above, the brightness pattern represents a map of the macroscopic field distribution.

Figure 4 shows typical results. It is noted that

<sup>7</sup> D. Meyerhofer, Phys. Rev. **112**, 413 (1958).

<sup>8</sup> A. C. Koelsch, IRE Natl. Conv. Record **5**, Part 3, 169 (1957).

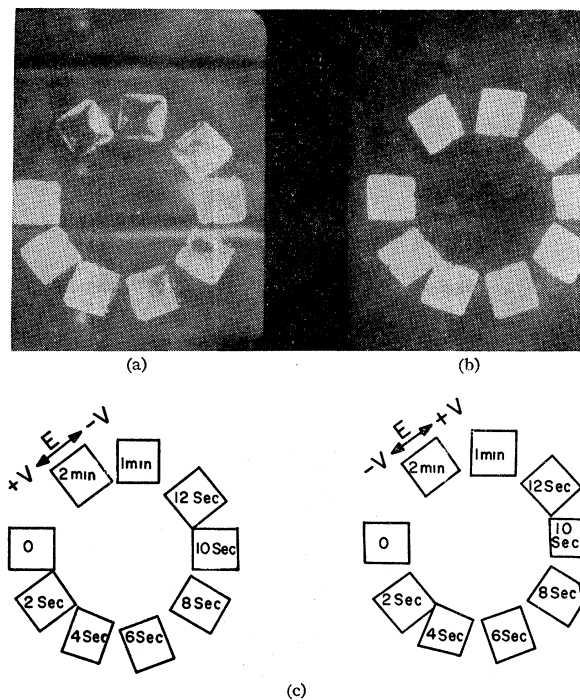


FIG. 4. Induced birefringence in BaTiO<sub>3</sub> crystal at 126°C. Crystal is 0.1 cm×0.1 cm×0.025 cm with electrodes on 0.1×0.025 face. Field is 2000 v/cm. Counting clockwise from gap, the first photograph is taken at instant field is applied and the next six at two second intervals. The 8th and 9th photographs are taken after 1 and 2 minute intervals, respectively. (a) and (b) differ only with respect to field direction.

initially, with either sign of field, the birefringence appears to be fairly uniform. In time, as evidenced by the birefringence pattern, the field distribution changes. As seen in the photographs and as observed in virtually every experiment performed in connection with this work, a decided asymmetry with respect to the direction of the field is evident. With the field in either direction, a noticeable decrease in bulk birefringence is observed; however, the effect is decidedly stronger with one

direction of applied field compared with the other. The time constants associated with the space-charge build-up appears to be of the order of one or two minutes as determined from visual observation of the time during which noticeable changes are occurring.

As indicated in Fig. 4, the field applied to the crystal was 2000 v/cm. It was observed that for the condition of Fig. 4(a) the current was  $3.6 \times 10^{-8}$  amp, and Fig. 4(b),  $2.6 \times 10^{-8}$  amp, indicating that the apparent resistance was higher when the field in the bulk was reduced due to space-charge effects. This is consistent with an assumption of the formation of a broad barrier.

Since the field applied was approximately one half of that necessary to induce the tetragonal phase,<sup>8</sup> it is to be expected that for the case of Fig. 4(b) the field in some region would be large enough to induce such a transition. Such a region was not seen within the resolution of the photograph of Fig. 4. However, in some of the experiments performed such regions were observed, typically, for example, in the vicinity of the dark-bright boundary in Fig. 3(a).

These experiments demonstrated the build-up in time of space-charge resulting in the reduction in the bulk of the samples of externally applied fields; the method could be adapted for actual quantitative field distribution plotting by measuring birefringence as a function of position on the crystal.

#### IV. NONLINEAR DIELECTRIC BEHAVIOR

The dielectric constant of a ferroelectric in the temperature region just above the Curie point represents a sensitive indicator of the electric field in the ferroelectric. This has been discussed by several authors<sup>9</sup> and calculated in some detail for BaTiO<sub>3</sub> and tri-glycine sulfate by this author.<sup>10</sup> If a dc field is applied to a ferroelectric in this temperature region, one of a number of effects might be observed. The most obvious behavior to be expected would be that of an ideal crystal, namely the field would be uniform and given by the applied voltage divided by the distance between elec-

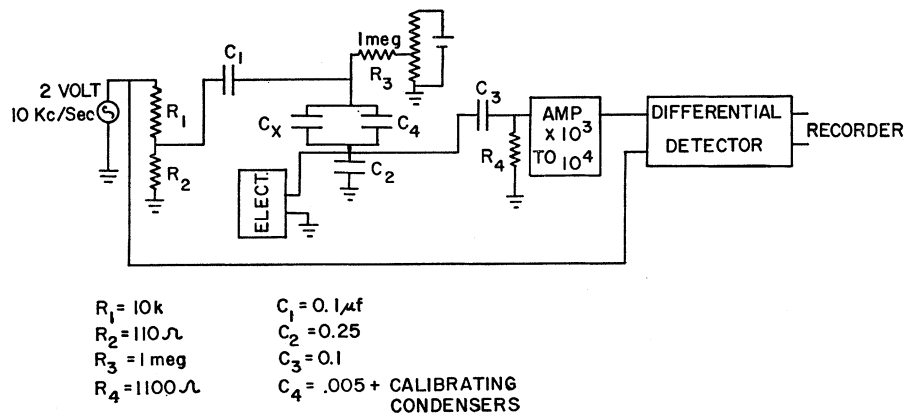


FIG. 5. Circuit used to record capacitance vs dc bias.

<sup>9</sup> M. E. Drougard, R. Landauer, and D. R. Young, Phys. Rev. **98**, 1010 (1955).

<sup>10</sup> S. Triebwasser, IBM J. Research Develop. **2**, 212 (1958).

trodes (in the case of plane parallel electrodes). The measured  $\epsilon$  would then be that calculated from a free energy function of the form<sup>11</sup>

$$F(P, T) = AP^2 + BP^4 + CP^6 + \dots, \quad (1)$$

and the relations:

$$E = \partial F / \partial P = 2AP + 4BP^3 + 6CP^5, \quad (2)$$

and

$$4\pi/(\epsilon - 1) = \partial E / \partial P = 2A + 12BP^2 + 30CP^4, \quad (3)$$

where  $P$  is the polarization,  $E$  the electric field, and  $A$ ,  $B$ ,  $C$  the expansion coefficients. Experiments performed on tri-glycine sulfate demonstrated that this method of analysis gives excellent quantitative agreement with the experimental results.<sup>10</sup>

A second possibility is that immediately after the application of the dc field (in the form of a step function),  $\epsilon$  would change to the value predicted by Eqs. (2) and (3), but, as space-charge appeared near the electrodes, the field in the bulk would be reduced to zero and  $\epsilon$  would be restored to the unbiased value. The apparent  $\epsilon$  would then be somewhat smaller than the unbiased value due to the fact that the equivalent circuit would include one or two surface layer capacitors in series with that represented by the bulk. The effective bulk thickness would be reduced by a negligible amount unless the sample is extremely thin, but the lower dielectric constant (as discussed below) of the space-charge layer(s) would more than compensate for this. Since the thickness of such layer(s) would be a function of the applied voltage, the apparent high frequency dielectric constant after equilibrium has been reached would depend on this voltage.

Actually it is found that both of these effects occur, namely: variation of dielectric constant with field as predicted by Eqs. (2) and (3), and finally, deviation from theory due to build-up of surface layer capacitance.

### Experimental Results

The samples on which the measurements reported below were made were prepared by etching a "butterfly wing" of BaTiO<sub>3</sub><sup>12</sup> in hot phosphoric acid and then evaporating Ag, Au, or Pt on the two major faces. From this large plate small rectangles were cut in such a manner that the two major opposite (100) faces were completely electroded.

Capacitance, as measured with very small rf signal, was recorded as a function of time after a step function of voltage was applied to the sample. The circuitry used is shown in Fig. 5. Typical traces taken from the recorder are shown in Fig. 6. The horizontal axis represents time while the vertical axis is capacitance. Sensitivity is as indicated in the diagram. The response time of the recorder was approximately two seconds.

Figure 7 shows the equilibrium values of the dielectric

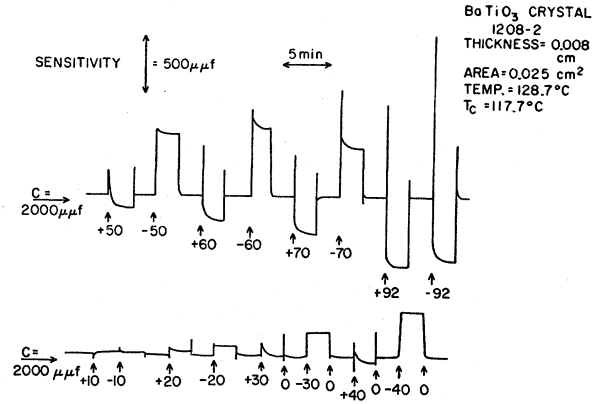


FIG. 6. Capacitance vs time. The small arrows indicate the time at which the indicated voltage was applied. Capacitance is plotted vertically and time horizontally.

constant plotted as a function of electric field together with the theoretical curves calculated from Eqs. (2) and (3). A comparison of Fig. 6 and Fig. 7 reveals that for the voltages at which theory and experiment agree in Fig. 7, the final value of capacitance is reached immediately (Fig. 6). The fields for which a noticeable drift with time appears in Fig. 6 coincide with those for which deviation from theory occur in Fig. 7.

### V. DISCUSSION

The results reported can be explained at least qualitatively by assuming the existence of Schottky barriers<sup>13</sup> at the two metal-crystal interfaces. The thickness and height of such barriers depends on the type and concentration of impurities, the dielectric constants, and of course, the band structure of BaTiO<sub>3</sub>. Because of the lack of detailed information on these properties, a detailed model cannot be presented which could be supported by a large body of experimental results. However, the semiquantitative analysis given below shows

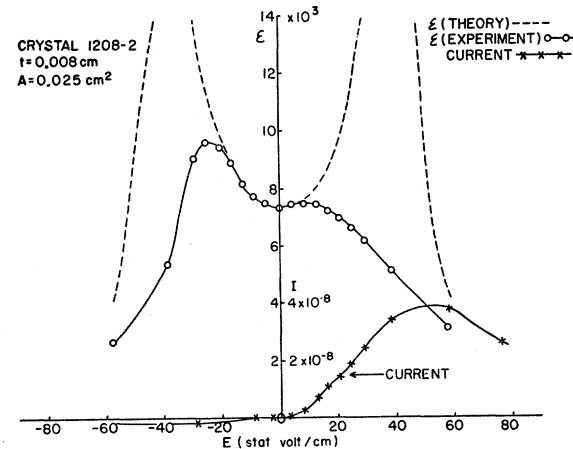


FIG. 7. Apparent  $\epsilon$  vs applied field and current vs applied field.

<sup>11</sup> See A. F. Devonshire, *Phil. Mag.* **40**, 1040 (1949).

<sup>12</sup> J. P. Remeika, *J. Am. Chem. Soc.* **76**, 940 (1954).

<sup>13</sup> See, for example, H. K. Henisch, *Rectifying Semiconductor Contacts* (Clarendon Press, Oxford, 1957), Sec. 7.3.4.

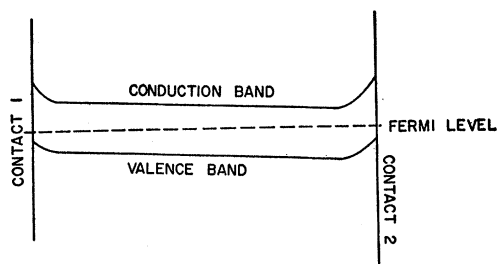


FIG. 8. Suggested band structure of BaTiO<sub>3</sub> crystal showing unequal barriers.

that the simplest of models is reasonably consistent with the data reported here.

From the results published by Remeika<sup>12</sup> and Nishioka et al.,<sup>14</sup> BaTiO<sub>3</sub> as grown by the former's technique is an *n* type material with a concentration of the order of 10<sup>19</sup> donors per cm<sup>3</sup>. The latter's work indicates an activation energy ~0.7 eV. Since the effective density of states in the conduction band itself is ~10<sup>19</sup>/cm<sup>3</sup> one would expect BaTiO<sub>3</sub> to have a density of carriers close to that expected in intrinsic Ge. If the electron mobility were of the order of magnitude exhibited by electrons in Ge, BaTiO<sub>3</sub> would have a bulk conductivity considerably higher than that actually observed. The fact that this is not the case probably indicates, as is to be expected, that the electron mobility is quite small.

With such large donor concentrations, it is to be expected that the position of the Fermi level would be quite sensitive to the value of that concentration. Furthermore, from the various pieces of evidence in the literature,<sup>4,5,15</sup> a considerable impurity concentration gradient is not unexpected. That is to say, the crystals are grown by a cooling process of BaTiO<sub>3</sub> in a KF flux in a Pt crucible. In general, the solubility of donor impurities (whatever their form) will be temperature dependent. Correlation has been observed between the actual inside and outside of so-called "butterfly wings" and the direction of the asymmetry of hysteresis loops.<sup>5</sup> From these arguments it can be assumed that space-charge surface layers exist (a reasonable assumption for any insulator-metal contact) and these layers are not identical at the two electrodes. A possible band structure is illustrated in Fig. 8. This is consistent with that hypothesized by Chynoweth<sup>15</sup> to explain his results. Figure 9 shows how the potential distribution will behave for zero applied volts and potentials applied in each direction after equilibrium is established.

From the results on nonlinear dielectric measurements by Drougard et al.<sup>9</sup> and our results at small fields, it can be concluded that no appreciable field normally exists in the bulk materials before the application of an external field. If this were not the case, a linear rather than quadratic dependence of  $\epsilon$  on field would appear

<sup>14</sup> A. Nishioka, K. Sekikawa, and M. Owaki, J. Phys. Soc. (Japan) **11**, 180 (1956).

<sup>15</sup> A. G. Chynoweth, J. Appl. Phys. **30**, 280 (1959).

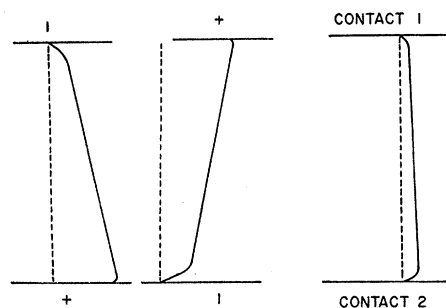


FIG. 9. Suggested potential distributions with no applied field and fields applied as shown.

for small fields. We were able to induce linear dependence in some samples, but the results depended in a variety of ways on the previous history of these samples.

Coming back to the band model represented by Fig. 8, it is clear that electric fields generated by the exhaustion barriers shown at the two contacts can be quite large. As first proposed by Kaenzig,<sup>1</sup> such fields can be large enough to induce the tetragonal phase well above the Curie point, or at least a large measure of saturation of polarization. In these circumstances the dielectric constant will be closer to that of the tetragonal phase (~200) than the cubic phase (10<sup>4</sup> near the Curie point). In order to estimate the effect of such a layer on the measured capacitance of the samples, a more detailed model is necessary. Kaenzig actually discusses a surface ionic vacancy model, but to be consistent with the model discussed in connection with Fig. 8, we shall assume a simple Schottky<sup>13</sup> exhaustion barrier. In this case the layer thickness is given by

$$\lambda = (V_B \epsilon / 2\pi N_D e)^{1/2}, \quad (4)$$

where  $V_B$  is the barrier height,  $\epsilon$  the dielectric constant,  $N_D$  the donor concentration and  $e$  the charge on the electron. If we take  $V \sim 1$  volt,  $N_D \sim 10^{19}$ /cm<sup>3</sup>, and  $\epsilon = 200$ , then the barrier thickness turns out to be  $5 \times 10^{-6}$  cm. Such a layer would have a capacitive impedance of about  $\frac{1}{10}$  that exhibited by the bulk crystal. When an external field is applied to the crystal,  $V_B$  must be replaced by  $(V_B + V_{ext}')$  where  $V_{ext}'$  is the additional voltage drop across the barrier caused by the application of the external field. The exact manner in which  $V_{ext}'$  is related to  $V_{ext}$  (the applied voltage) is a nontrivial problem which depends for its solution on details of the model which are unknown. Qualitatively, it can be expected that as the external field is increased,  $V_{ext}'$  will increase causing the barrier at one of the contacts to broaden to the point where the capacitive impedance of that barrier competes strongly with the bulk impedance. When this happens, the measured impedance will be higher than theoretical, or the measured capacitance will be lower and the experimental points in Fig. 7 fall below the theoretical ones.

The asymmetry in the current-voltage characteristic was observed to some degree in every crystal examined.

The general behavior of the current is consistent with a model of tunneling of carriers through the reversed biased barrier. Such a barrier becomes broadened, but sharply peaked. Again, it is required that the barriers be of different height in order to explain the difference in behavior with difference in sign of voltage.

It is to be noted in comparing Figs. 6 and 7 that for values of dc bias for which the equilibrium value of  $\epsilon$  coincides with the theoretical value in Fig. 7, the equilibrium value is reached instantaneously as shown in Fig. 6. Where experimental points in Fig. 7 fall below theoretical values, there is an observable delay between application of electric field and final value of capacitance taken from Fig. 6. This corresponds to the fact that time is required for space-charge layers to build up. Initially (in time) after application of the electric field, the normal dielectric constant is observed. As the space-charge layer broadens, the observed capacitance (or apparent  $\epsilon$ ) decreases.

## VI. CONCLUSION

Unraveling the assorted anomalous effects that have been reported in BaTiO<sub>3</sub> which have been attributed to

some form of surface layer has not led to any consistent picture of the layer except in the most qualitative terms. The results reported here are representative of measurements made on a number of crystals in which evidence of space-charge layers whose thickness is voltage dependent may be seen. The detailed quantitative interpretation of results is hampered by complicated electric field distributions and our general ignorance of BaTiO<sub>3</sub> surfaces. On the other hand, the observations indicate that further work in which impurity concentrations and dielectric constant are varied (the latter by varying temperature) as well as thickness of samples would be worthwhile in trying to understand details of surface layers and fields in insulators and BaTiO<sub>3</sub> in particular.

It is felt that measurements of this kind will shed light on the general problem of mapping of space-charge regions in insulators. It is clear that experiments that actually probe such regions are difficult to imagine on the general insulating material. The special property that makes a ferroelectric amenable to such probing is the sensitivity it exhibits to reasonably attainable fields in its Kerr effect and dielectric constant.

## Effect of the Lattice on Dielectric Properties of an Electron Gas\*

DAVID S. FALK†

*Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts*

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A system of  $N$  electrons in the presence of a rigid periodic background of positive charge is considered. Following Martin and Schwinger, an inverse dielectric operator,  $\mathcal{K}'$ , is introduced. An approximate equation which takes into account the long-range nature of the Coulomb field is derived for  $\mathcal{K}'$ . A representation is used where  $\mathcal{K}'$  is a matrix with rows and columns labeled by vectors of the reciprocal lattice. Poles and zeros in the dielectric operator are found to be manifestations of Bragg's law. Assuming these to be the major effect of the lattice, the equation for  $\mathcal{K}'$  is solved. The result is examined in the weak-binding limit and seen here, except at the Bragg reflections, to agree with that of Nozières and Pines. Finally the ground-state energy of the system is exhibited.

## INTRODUCTION

A GOOD deal of attention has been devoted lately to the properties of a gas of interacting electrons in the presence of a uniform background of positive charge. Much of the interest in this system stems from the hope that it will, for some applications, be a good model for the electrons in a solid. Now in a solid, of course, the positive charge is not uniformly distributed but rather concentrated on ions which vibrate about equilibrium positions that form a lattice structure containing various kinds of defects. The existence of

the lattice as well as the defects in it and the vibrations about it may be expected to have some effect on the dielectric properties of the electron gas. It is the purpose of this paper to investigate the effect of the lattice, ignoring defects and vibrations, in order to see where the free electron gas results are valid and where and how they must be modified because of the presence of the lattice. To this end the inverse dielectric operator of Martin and Schwinger will be considered here and techniques suited to the weak but long-ranged nature of the Coulomb field will be used to derive an approximate equation for it. This equation will be solved and the effect of the lattice on this operator and hence on, say, the ground-state energy of the system, will be seen.

Consider a system of volume  $\Omega$  containing  $N$  electrons

\* The work reported in this paper was performed at Lincoln Laboratory, a center for research operated by Massachusetts Institute of Technology with the joint support of the U. S. Army, Navy, and Air Force.

† Now at the University of Washington, Seattle, Washington.

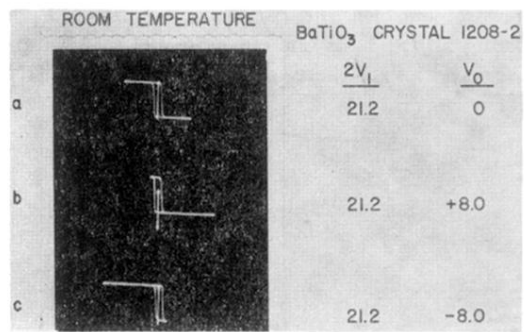


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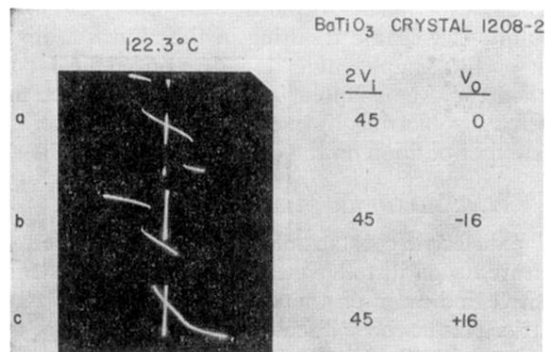


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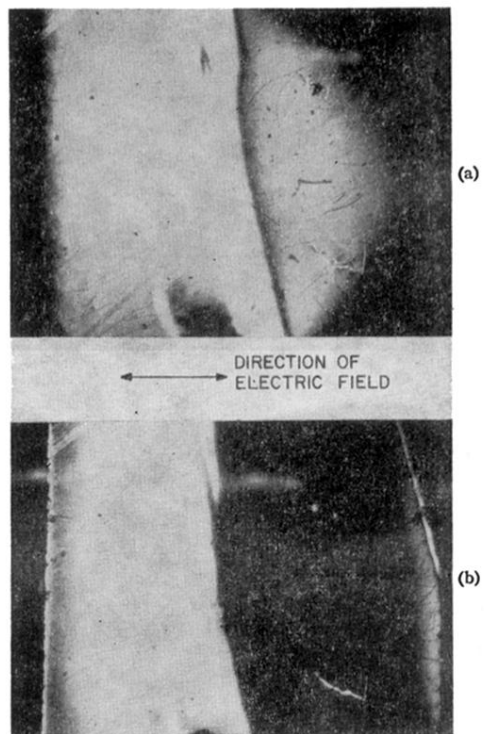
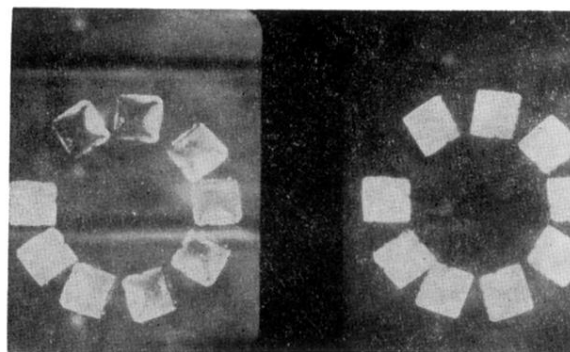
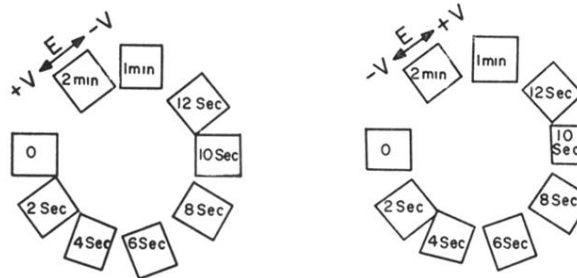


FIG. 3. Induced birefringence pattern in BaTiO<sub>3</sub> crystal at 125°C. Crystal is 0.3 cm×0.3 cm×0.016 cm with electrodes on 0.3×0.016 face. Field=1500 v/cm. (a) and (b) differ only with respect to field direction.



(a)

(b)



(c)

FIG. 4. Induced birefringence in  $\text{BaTiO}_3$  crystal at  $126^\circ\text{C}$ . Crystal is  $0.1\text{ cm} \times 0.1\text{ cm} \times 0.025\text{ cm}$  with electrodes on  $0.1 \times 0.025$  face. Field is  $2000\text{ v/cm}$ . Counting clockwise from gap, the first photograph is taken at instant field is applied and the next six at two second intervals. The 8th and 9th photographs are taken after 1 and 2 minute intervals, respectively. (a) and (b) differ only with respect to field direction.