Investigation of the Dependence of the Dielectric Constant and the Tangent of the Dielectric Loss Angle of Barium Titanate on the Strength of a High Frequency Electric Field

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A method of measurement of dielectric constants and the tangent of the dielectric loss angle of ferroelectrics in strong high frequency (10^6 cps) fields is described. The results of measurements of the dependence of capacity and the tangent of the dielectric loss angle on the strength of electric field at various temperatures are presented and evaluated for a barium titanate sample.

1. INTRODUCTION

AT the present time the nonlinear properties of ferroelectrics are widely used in various radio installations[1].

To utilize a ferroelectric material in frequency and amplitude modulation equipment as well as in capacitative amplifiers one must know the dependence of the dielectric constant and of the tangent of the dielectric loss angle on the intensity of high frequency electric field. Thus far, the nonlinear properties of ferroelectrics are insufficiently known at high frequencies because of the considerable experimental difficulties encountered in measurements of dielectric constants and the tangent of the dielectric loss angle in strong electric fields of high frequency. These difficulties are caused by the heating of samples due to large dielectric losses. Since ϵ and $\tan\delta$ of ferroelectrics strongly depend on temperature, it is necessary to maintain the temperature of the sample constant during the investigation of their dependence on the strength of the electric field.

Kambe[2] has used the pulse technique to study the hysteresis loops of barium titanate in strong high frequency fields at various temperatures. From the analysis of the hysteresis loops the authors concluded that the maximum polarization decreases with increasing frequency. Only qualitative conclusions are possible in the cited work about the dielectric losses. Zavelskii[3] has also observed a decrease of the dielectric constant with a decrease in the duration of the charging of condenser made of $BaTiO_3$ and an increase of the dielectric constant with an increase in the electric field intensity.

The experimental technique used by Kambe[2] and Zavelskii[3] is not convenient for the measurement of the smooth variation $\epsilon = f(E)$ and $\tan\delta = \varphi(E)$. The investigation of the mentioned dependences at high frequency is important both for the theory

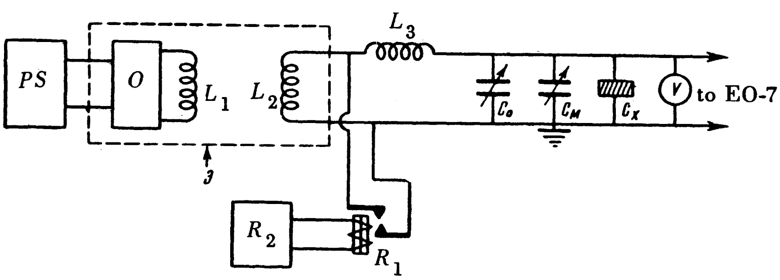

FIG. 1. Block diagram of the experimental apparatus. *PS*-stabilized power supply; *O*-Oscillator; L_1-induction coil of the oscillator; L_2-coupling coil; L_3-induction coil of the measuring circuit; C_0-standard condenser; C_M-modulating condenser; C_x-condenser containing the dielectric under investigation; *V*-vacuum tube voltmeter of the type VKS-7 (used with a voltage divider); R_1-relay; R_2-a system of control relays.

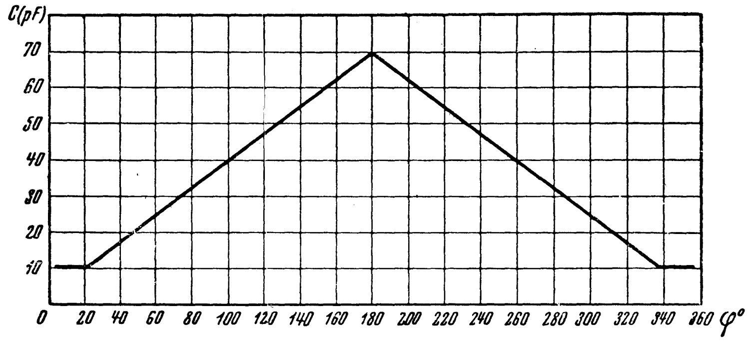

FIG. 2. The dependence of the capacity of the modulating condenser on the angle of rotation of its rotor.

of the ferroelectric phenomena and for its technical utilization. Hence, it is necessary to develop a method of measurement of the continuous dependence of $\epsilon = f(E)$ and $\tan\delta = \varphi(E)$ at high frequency. Such is the purpose of the present work.

EXPERIMENTAL TECHNIQUE

The basis of employed means of determination of ϵ and $\tan\delta$ is the resonance method of varying capacitance in which changes were introduced to permit measurements on ferroelectric samples in strong fields of high frequency. The block diagram of the apparatus is given in Fig. 1.

The special feature of the method employed is a modulation of the natural resonance frequency of the measuring circuit. This was accomplished by means of the modulation capacitor C_M. The axis of the capacitor was attached to the rotor of

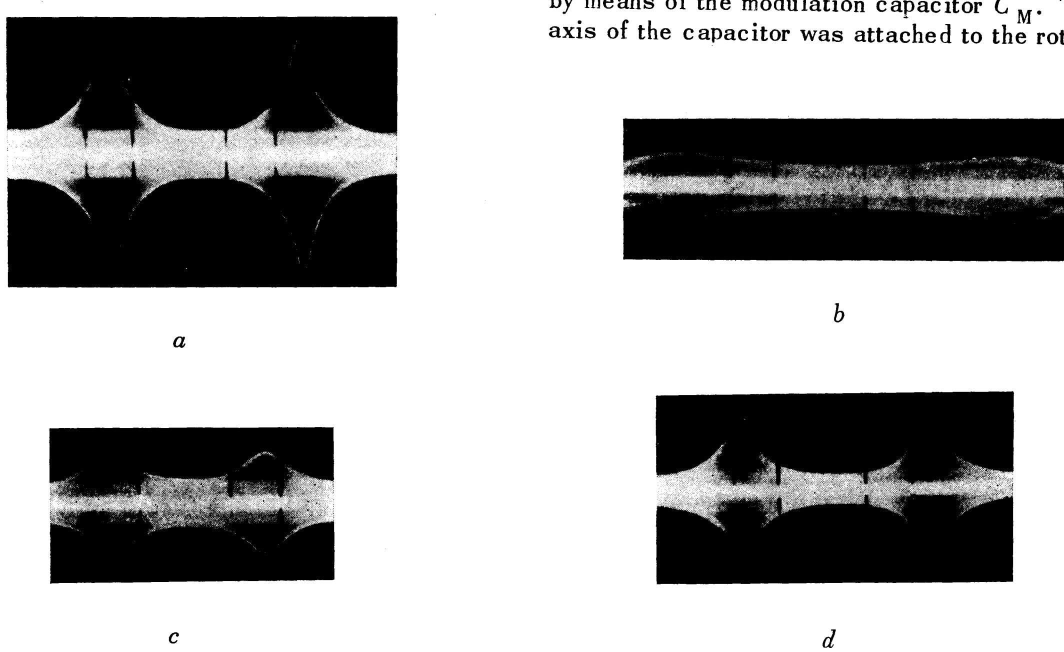

FIG. 3. Oscillograms of the resonance curves: *a*-without a sample; *b*-for a sample of $BaTiO_3$ ($T = 82°$); *c*-for a sample of $BaTiO_3$ ($T = 167°$); *d*-for a KTK type condenser.

an electrical motor rotating with an angular velocity of 1500 rpm. The dependence of the capacity of the modulating condenser on the angle of rotation is given in Fig. 2. At a particular value of the total capacitance a resonance takes place in the measuring circuit, the resonance capacitance occuring twice during the time of a single revolution of the capacitor C_M. Two resonance curves are then presented on the screen of the oscilloscope (Fig. 3). With a change of capacitance in the measuring circuit the separation between the resonance maxima changes. Therefore, it is possible to evaluate the change of capacity of the investigated sample from the separation of the two resonance peaks. The oscilloscope screen is first calibrated by means of a standard condenser C_0.

The tangent of the dielectric loss angle calculated from the formulas of Bogoroditskii[4] and Skanavi[5] is:

$$\tan \delta = \frac{\Delta c}{2} \frac{(U - U_x)}{U_x \cdot C_x}, \qquad (1)$$

where U is the resonance intensity in the measuring circuit without a sample, C_x is the capacitance of the sample, U_x is the resonance intensity in the measuring circuit with a sample, Δc the so-called distortion of the contour: $\Delta c = C_0'' - C_0'$ where C_0' and C_0'' are the values of the capactiance in the measuring circuit without a sample corresponding to an intensity $\sqrt{2}$ times smaller than the resonance intensity (taken on both sides of the resonance curve peak).

The values Δc, U and U_x are determined from the resonance curves. For this purpose the oscilloscope screen was calibrated for measurements of intensity by a vacuum tube voltmeter V. The determination of Δc directly from the resonance curve assured a greater accuracy than a measurement of that quantity by variation of the circuit capacity by the condenser C_0.

From the above it follows that for a determination of the dependence $\epsilon = f(E)$ and $\tan\delta = \varphi(E)$ it is necessary to photograph the resonance curves without and with the sample in the circuit at various intensities of the electric field. In order to prevent the heating of the ferroelectric samples due to dielectric losses the voltage was applied to the sample for the period of time $t = 0.1$ sec, during which the photographs were taken. For such a duration of the electric field the sample practically did not heat at all (it is also necessary to take into account that the time of action of the full resonance intensity on the sample is still shorter).

As is evident from the diagram of Fig. 1, the emf induced in the coupling coil L_2 by the oscillator 0 is passed through the measuring circuit only if the relay R_1 is open. The opening of the relay R_1 for the period of time $t = 0.1$ sec was made by a system of control relays R_2.

To stabilize the presentation of the resonance curves on the oscilloscope screen, synchronization by pulses of constant intensity was employed. The pulses were introduced on the external synchronization terminals of the EO-7 oscilloscope from the terminals of a storage battery through contacts that were periodically broken by a cam attached to the axis of the motor driving the modulating condenser.

The analysis of errors has shown that the maximum calculated error in measurements by our apparatus did not exceed 2% for the capacity and

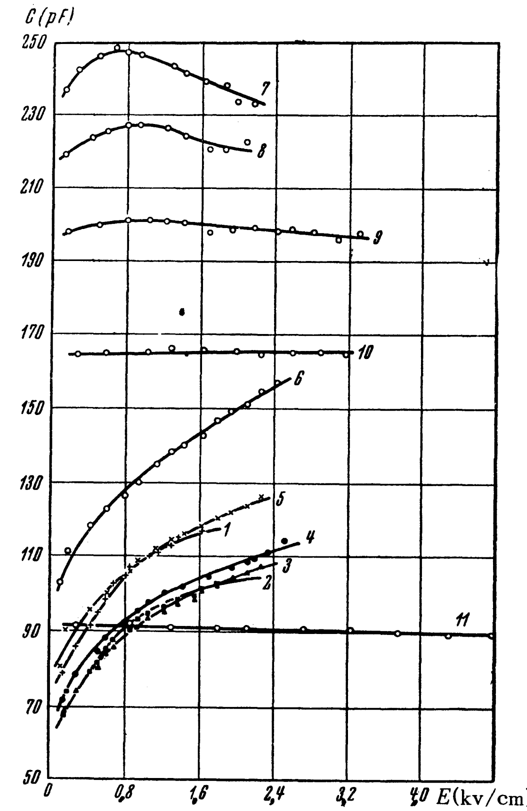

FIG. 4. The dependence of capacity of sample of $BaTiO_3$ on the electric field intensity at high frequency and various temperatures: 1 - $T = 19°$; 2 - $t = 38°$; 3 - $t = 60°$; 4 - $t = 82°$; 5 - $t = 98°$; 6 - $t = 116°$; 7 - $t = 123°$; 8 - $t = 127°$; 9 - $t = 132°$; 10 - $t = 142°$; 11 - $t = 167°$.

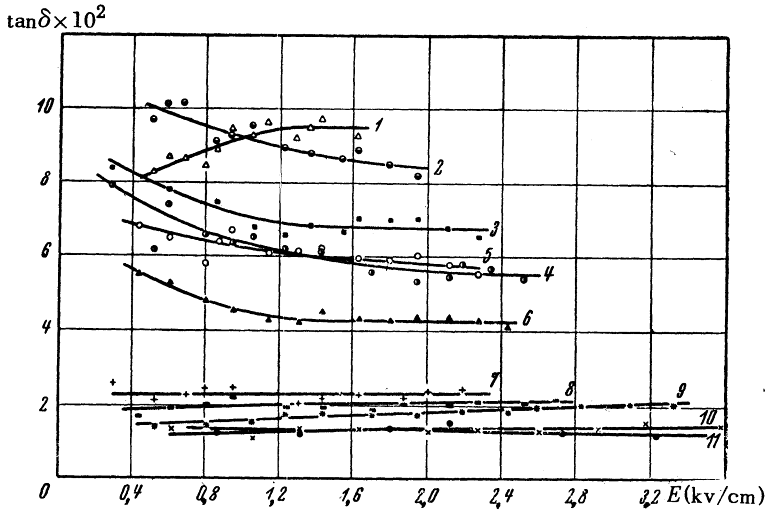

FIG. 5. The dependence of the tangent of the dielectric loss angle of $BaTiO_3$ on the intensity of high frequency electric field for various temperatures: *1* - $t = 19°$; *2* - $t = 38°$; *3* - $t = 60°$; *4* - $t = 82°$; *5* - $t = 98°$; *6* - $t = 116°$; *7* - $t = 123°$; *8* - $t = 127°$; *9* - $t = 132°$; *10* - $t = 142°$; *11* - $t = 167°$.

25% for the tangent of the dielectric loss angle. This is entirely satisfactory for the resonance methods.

EXPERIMENTAL RESULTS AND THEIR EVALUATION

By means of the above discussed method the dependence on the electric field intensity of the capacity and of the tangent of the dielectric loss angle of a sample of polycrystalline $BaTiO_3$ was investigated at a frequency 10^6 cps and at various temperatures (Figs. 4 and 5). On the basis of this data the curves for the temperature dependence of the capacity and of $\tan\delta$ were prepared for several values of the electric field intensity (Fig. 6). To verify the employed technique further we measured the dependence of $C = f(E)$ and $\tan\delta = \varphi(E)$ for ceramic condensers of the KTK type (Fig. 7).

The results of measurements show that at a high frequency and strong fields one observes a nonlinear dependence $C = f(E)$ characteristic for the ferroelectric region of $BaTiO_3$ and the capacity-temperature curve has a sharply defined maximum. The temperature dependence of $\tan\delta$ in strong fields of high frequency is similar to the analogous dependence in weak fields: in the region of a sharp increase of ϵ the value of $\tan\delta$ sharply decreases. Saturation in the curve $C = f(E)$ was observed for the given field intensities only for temperatures lying in the region of the Curie point, and with an approach to the Curie point the maximum of the curve $C = f(E)$ shifts to the region of the weaker fields.

A comparison of the values of the dielectric constant of $BaTiO_3$ at high frequency and strong fields with the dielectric constant of samples of the same batch measured at frequency 50 cps at the same values of field intensity and temperature shows that the dielectric constant is smaller at high frequency than at 50 cps. From a similar comparison it also follows that the nonlinear properties of barium titanate are less pronounced at high frequencies. As is evident from Fig. 5, the tangent of the dielectric loss angle depends on the electric field intensity weakly at high frequency. The fact that the nonlinear properties of barium titanate manifest themselves most significantly in the region of the Curie point can be explained qualitatively by saying that in the temperature range the rotation of the electrical moments of the spontaneous polarization domains is made easier by the action of an external field. The decrease in the value of the dielectric constant and weaker manifestation of the nonlinear properties at a high frequency in comparison with the corresponding data for a frequency of 50 cps are apparently due to the fact that at a high frequency the duration of the action of the electric field within one half period decrease relative to the time needed

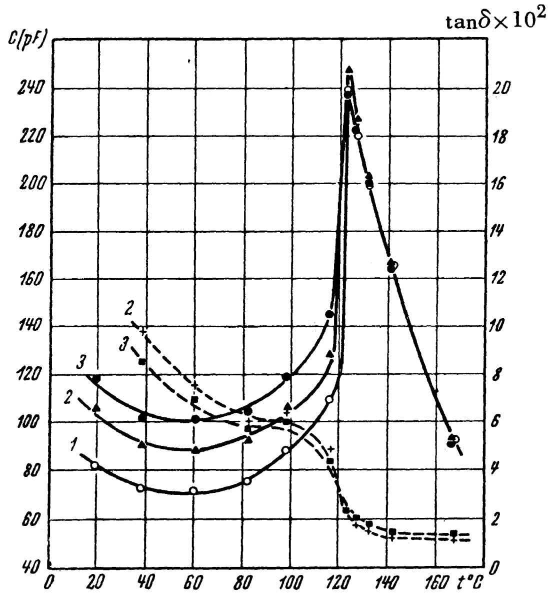

FIG. 6. The dependence of the capacity (solid curves) and tanδ (dotted curves) of $BaTiO_3$ sample on temperature at high frequency and for various field intensities: *1* - $E = 0.2$; *2* - $E = 0.8$; *3* - $E = 1.7$ kv/cm.

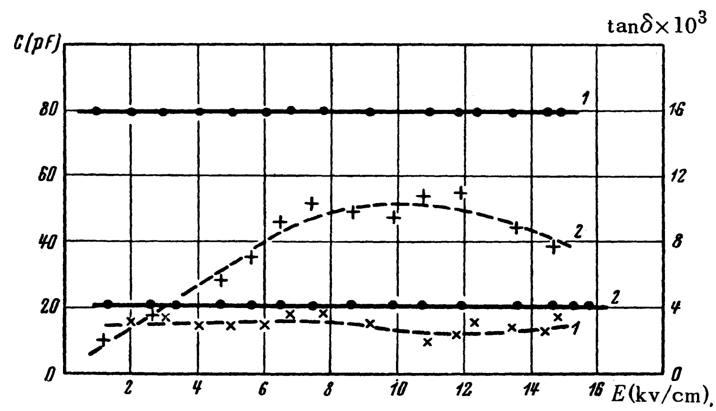

FIG. 7. The dependence of capacity (solid curves) and tanδ (dotted curves) of ceramic condensers on field strength of high frequency: *1* - KTK, orange, 82 ± 10% *μμf*; *2* - KTK, red, 20 ±5% *μμf*.

to establish polarization. (This is in the nature of polarization determined by rotation of the domain moments.)

CONCLUSIONS

In the studied range of electric field intensities:

1. The nonlinear properties of barium titanate are less pronounced at a high frequency (10^6 cps) than at a low frequency (50 cps).
2. The most significant nonlinear properties of barium titanate at high frequency take place in the temperature range close to the Curie point.
3. The tangent of the dielectric loss angle of barium titanate at high frequency depends weakly on the intensity of the electrical field.

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