As to the value of the complete effective cross section for the (dn) reaction with the Be⁹ nucleus, there are no experimental data in the literature, and therefore, for the time being, Eq. (16) cannot be verified for the complete effective cross section.

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SOVIET PHYSICS JETP

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MAY, 1957

Dielectric Properties of Bismuth Titanate

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G. I. SKANAVI AND A. I. DEMESHINA P. N. Lebedev Physical Institute, Academy of Sciences, USSR (Submitted to JETP editor February 4, 1956) J. Exptl. Theoret. Phys. (U.S.S.R.) 31, 565-568 (October, 1956)

It is shown that the titanates of bismuth possess high dielectric constants ($\epsilon = 70-120$) and comparatively large positive temperature coefficients (TC $\epsilon = 130-550 \times 10^{-6}$) which are evidently brought about by a combination of a favorable internal field and a high ionic polarizability. The formation process of one of the barium titanates is investigated, and the temperature and frequency dependencies of ϵ and tan δ of various bismuth polytitanates have been measured.

THE titanates of metals of the second group of the periodic table have an elevated dielectric constant which increases upon increase in the atomic weight of the metal¹. In this case, for nonpiezoelectric titanates, as well as for many other ceramic dielectrics, this rule holds: the higher the dielectric constant, the more negative the temperature coefficient $(1/\epsilon)(d\epsilon/dT)^2$. For small dielectric constants the quantity $(1/\epsilon)(d\epsilon/dT)$ has a positive sign. For most dielectric constants, $(1/\epsilon)(d\epsilon/dT)$ has a comparatively large negative value. As shown earlier³, the high dielectric constant of nonpiezoelectric titanates of barium and rutile (TiO₂) is brought about by a combination of high electronic polarization and a favorable polarization of the internal field produced by the ionic displacement. The ionic polariz-ability in this case is not large. The electronic polarizability does not depend on the temperature but the electronic polarizability per unit volume decreases with increase in temperature at the expense of a decrease in the number of polarized particles per unit volume in thermal expansion. The ionic polarizability increases with increase in temperature, since the elastic coupling is weakened. In dielectrics with high dielectric constant, the

electronic polarizability appreciably exceeds the ionic; therefore, the dielectric constant decreases with increase in temperature $[(1/\epsilon)(d\epsilon/dT) < 0]$. The positive temperature coefficient of ϵ points to the predominance of the ionic polarizability.

In the present work, an attempt was made to realize, in a polycrystalline, nonpiezoelectric dielecrtic, such conditions under which a high dielectric constant would be combined with a comparatively large positive temperature coefficient. An appreciable ionic polarization takes place in glasses, the dielectric constant of which increases sufficiently rapidly with increase in temperature; therefore, the presence of glassy layers in a polycrystalline dielectric ought to facilitate an increase of ϵ with temperature. On the other hand, it is necessary that relaxation processes connected with inelastic dislocations of the ions not take place in this glassy layer and in defective places in the crystalline lattice, for this would, in turn, lead to greater loss. In this connection, the glassy layer must contain only heavy ions, possessing small mobility.

Starting from these considerations, we investigated the possibility of the formation of a combination of titanium dioxide and bismuth trioxide. Bismuth trioxide possesses weakly basic properties

Molecular ratio of the components TiO2/Bi2O3	Temperature of annealing, ^o C	$\epsilon, f=2mc, t=20^{\circ}$	$\tan \delta, f=2mc, t=20^{\circ}$	$\begin{array}{c c} TC \epsilon \times 10^{6}, f = 2mc & M \\ TC \epsilon \times 10^{6}, f = 2mc & th \\ th & the interval \\ t = 20-80^{0} & 7 \end{array}$	olecular ratio o te components FiO ₂ /Bi ₂ O ₃	f Temperature of annealing, ^o C	ϵ , $f=2mc$, $t=20^{\circ}$	$t \operatorname{an} \hat{\partial}_{t} f = 2mc,$ $t = 20^{\circ}$	$TC \in \times 10^6$, $f = 2mc$ in the interval $t = 20-80^\circ$
$\begin{array}{c} 222.3:1\\ 13.7:1\\ 8.8:1\\ 8.8:1\\ 3:1\\ 3:1 \end{array}$	1240 1215 1215 1215 1220	92 88 98 88 80 88 80 88 80 88	0.0015 0.0034 0.0034 0.0039 0.0039 0.0031 0.0028 0.0028		1, 2: 1 1, 7: 1 1, 45: 1 1: 1	1185 1170 1150 1060	113 112 107 121	$\begin{array}{c} 0,0054\\ 0.0040\\ 0.0023\\ 0.0024\end{array}$	++++++530 +510

TABLE I

which appear both in solutions and at high temperature. Therefore, it can probably enter into a reaction at high temperature with titanium dioxide, which possesses weakly acidic properties, giving a compound of a type of bismuth polytitanate. Bismuth trioxide has a comparatively low melting point (about 900°). At this temperature, titanium dioxide undergoes a transition from the crystalline form of anatase into a crystalline modification of rutile and has an increased reaction capability. Therefore, we can expect that, at a temperature of about 900°, a reaction begins between TiO₂ present in the solid phase, and Bi₂O₃, present in the liquid phase. As a result of this reaction, polycrystals could be formed with appreciable glassy layers which contain the heavy bismuth ions and also defects in the lattice.

Thermographic analysis of the process of the formation of the titanates of bismuth confirms the assumptions made above. At $T = 900^{\circ}$ an endothermic peak occurs, corresponding to the lique-faction of bismuth trioxide. A sharp endothermic peak in the differential temperature curve at 1230° shows the intense reaction which leads to the formation of one of the bismuth titanates corresponding to a given ratio of components. X-rays of the bismuth titanates also point to their chemical individuality.

Measurement of the dielectric constant and the power factor at high frequencies and for different polytitanates of bismuth shows that for an appreciable change in the ratio of components, the dielectric constant changes slightly and lies within the limits 70-120. The power factor has a higher value ($\tan \delta \approx 0.002$ -0.006). The temperature coefficient of the dielectric constant, in agreement with theoretical assumptions, was shown to be positive for many bismuth titanates. The results of the measurements of ϵ , $\tan \delta$ and TC $\epsilon = (1/\epsilon)(d\epsilon/dT)$ are plotted in Table I for a number of bismuth titanates.

Figures 1 and 2 show curves for the dependence of ϵ and tan δ on temperature and frequency for one of the bismuth titanates. It is seen that ϵ increases appreciably and almost linearly with temperature. For certain ratios of TiO₂/Bi₂O₃ (of those plotted in Table I) the temperature coefficient of ϵ undergoes a transition from positive to negative values. As specially arranged experiments have shown, in this region of concentration, TC ϵ always depends on the annealing history. The thermogram of Fig. 3 bears witness to this fact. It is evident therein that at a temperature of 1230° a sharp

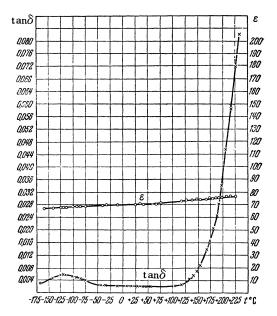


FIG. 1. Dependence of ϵ and $\tan \delta$ on the temperature for f = 700 kc for one of the bismuth titanates.

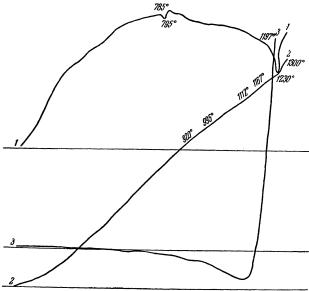


FIG. 3. Thermogram of the sintering of one of the bismuth titanates: l = difference in temperature of the specimen and the etalon; 2 = temperature of the furnace; 3 = shrinkage of the specimen.

change in the structure takes place. This is accompanied by an emission of heat (a phase change of first order) over a very narrow range of temperatures. If the temperature is $1-2^{\circ}$ less than the transition temperature (1230°), then TC ϵ has a large positive value; for the temperature 1230°

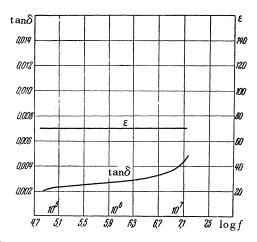


FIG. 2. Dependence of ϵ and $\tan \delta$ on the frequency for one of the bismuth titanates.

and higher, TC ϵ has a large negative value.

In this transition, an appreciable increase takes place in the dielectric constant, and there is some decrease in the power factor. The experimental data which characterize the effect of the temperature annealing on the dielectric properties of one of the bismuth titanates are shown in Table II.

In conclusion, it is interesting to note that the discovery of such a sharp change in TC ϵ and the dielectric constant is connected with the structure change which is noted in the thermogram (a sharp endothermic peak), but at the same time there is no effect on the Debye-gram. It is possible that in this transition a crystallization of the glass layer takes place. The problem of the change of the crystallographic structure in this case requires further x-ray investigation.

	TABLE II. Change of dielectric properties of one of the
_	bismuth titanates under the action of temperature anneal-
	ing.

Temperature of annealing, ^o C	$f = \frac{2}{2} mc,$ $t = 20^{\circ}$	$tan \delta,$ f = 2 mc, $t = 20^{\circ}$	$TC \epsilon \times 10^{6}$ for $f = 2 mc$, in the interval $t = 20-80^{\circ}$
1210 1220 1225 1230 1245	83 78 81 83 94	$\begin{array}{c} 0.0032\\ 0.0038\\ 0.0045\\ 0.0027\\ 0.0022 \end{array}$	$+360 \\ +360 \\ +160 \\ -430 \\ -480$

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SOVIET PHYSICS JETP

VOLUME 4, NUMBER 4

120

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On Quasiclassical Quantization

V. S. KUDRIAVTSEV

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A method of calculating the energy levels in a quasiclassical quantization is presented for the one-dimensional case. The value of the levels is obtained in the form of in expansion π . Under certain assumptions on the form of the potential energy U(x), this expansion can be obtained in a general form. Computations are carried out for a potential energy having a minimum and rising on either side of the minimum, i.e., of an oscillator type.

A S is well known, in the quasiclassical method for the solution of the problems of quantum mechanics, the wave equation ψ is written in the form

$$\Psi = e^{i\sigma/\hbar}.$$
 (1)

Making this formal substitution in the Schrödinger equation

$$\Delta \psi + (2\mu/\hbar^2) (E - U) = 0, \qquad (2)$$

we obtain an equation for the function σ :

$$(\nabla^{\sigma})^{2} + (\hbar/i) \Delta^{\sigma} = 2\mu (E - U), \qquad (3)$$

The formal solution of Eq. (3) is written in the form of a power series in π :

$$\sigma = \sigma_0 + (\hbar/i) \sigma_1 + (\hbar/i)^2 \sigma_2 + \dots \qquad (4)$$

Substituting (4) in (3), we ultimately get, for the one-dimensional case,

$$\begin{aligned} \sigma_{0}^{'} &= p; \ \sigma_{1}^{'} = -p'/2p; \ \sigma_{2}^{'} = p''/4p^{2} - 3p'^{2}/8p^{3}; \ ^{(5)} \\ \sigma_{3}^{'} &= -p'''/8p^{3} + 3p''p'/4p^{4} - 3p'^{3}/4p^{5}; \\ \sigma_{4}^{'} &= p^{(IV)}/16p^{4} - 5p'''p'/8p^{5} \\ &- 13p''^{2}/32p^{5} + 99p''p'^{2}/32p^{6} \\ &- 297p'^{4}/128p^{7}; \ldots, \end{aligned}$$

where $p = \sqrt{2\mu(E - U)}$ is the classical momentum.

For real p, the quantities $\sigma'_0, \sigma'_1, \ldots$ and $\sigma_0, \sigma_1, \ldots$ are real, and the quantity σ can be uniquely divided into

two components which define the phase and modulus of the wave function:

³ G. I. Skanavi, J. Exptl. Theoret. Phys. (U.S.S.R.) 5,

$$\psi = \exp \{\sigma_1 - \hbar^2 \sigma_3 + \hbar^4 \sigma_5 - \ldots\}$$

$$\times \exp \{i (\overline{\sigma_0} / \hbar - \hbar \sigma_2 + \hbar^3 \sigma_4 - \ldots)\}.$$
(6)

Another linearly independent solution of the Schrödinger equation is obtained by substituting $i \rightarrow -i$ in Eq. (6). For imaginary p, all the expressions in the exponent are real.

Let x = a be a turning point, i.e., U(a) = E. Let us find the phase of the wave function for x > a, considering that, in this region, E > U(x), and in the region x < a, E < U(x), and the modulus of the wave function decreases with decreasing x. Solving the Schrödinger equation exactly in the neighborhood of the turning point, where the potential energy can be approximated by a linear function of the coordinate x, and joining the exact solution with the quasiclassical one, we obtain an expression for the phase, as is usually done. The exact solution of the Schrödinger equation with a linear potential which satisfies the conditions set forth above has the form (except for a constant multiplier)

$$\dot{\Psi} = \begin{cases} V_{\bar{[\xi]}[I_{-1/s}\left(\frac{2}{3}\,|\xi|^{s/2}\right) + I_{1/s}\left(\frac{2}{3}\,|\xi|^{s/2}\right)], \ x < 0; \\ V_{\bar{\xi}}^{-} \left[J_{-1/s}\left(\frac{2}{3}\,\xi^{s/2}\right) + J_{1/s}\left(\frac{2}{3}\,\xi^{s/2}\right)\right], \ x > 0, \\ \tilde{\xi} = \alpha x/\hbar^{s/2}, \ \alpha = \sqrt{2\mu \left(-\partial U/\partial x\right)_a}. \end{cases}$$