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ELECTRO-OPTICAL EFFECTS IN CRYSTALS

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THE optical properties of dielectric crystals, in particular, their refractive indices n_{ij}, change under the influence of an electric field. It is usual to represent changes in the refractive indices indirectly in terms of changes in the polarization constants a_{ij}, which are squares of the reciprocals of the refractive indices. In the simplest case of an isotropic medium, exhibiting only the electronic polarization, the polarization constant a, the refractive index n and the permittivity ϵ are related by

$$a = \frac{1}{n^2} = \frac{1}{\epsilon}. \tag{1}$$

In some crystals, the changes (increments) in the polarization constants are proportional to the first power of the electric field (polarization). In this case, we say that such a crystal exhibits the linear electro-optical effect. All dielectric crystals should, in principle, exhibit the quadratic electro-optical effect, i.e., the increments in the polarization constants should be proportional to the square of the electric field (polarization).*

Intensive investigations of the electro-optical effects have begun relatively recently. The foundations of the theory of the linear electro-optical effect were laid by Pöckels in 1894. [1] In the eighteen nineties, Pöckels carried out one of the first experimental investigations of this effect. However, for a long time there was little investigation of the electro-optical effect and its applications were very limited. The main reason for this was the need to apply relatively high electric voltages (tens and even hundreds of kilovolts) in order to obtain a perceptible effect. The relatively intensive investigations of electro-optics began about 15 years ago. They were primarily due to the discovery of strong electro-optical properties in crystals of KDP (KH_2PO_4) and some isomorphous compounds. The electro-optical coefficients of these crystals are more than an order of magnitude greater than the coefficients of linear dielectrics known up to that time. Moreover, the development of electro-optical investigations was stimulated recently by the growth of quantum radioelectronics. Electro-optical

crystals are now being used in devices employed in the modulation of laser radiation.

Investigations of the electro-optical properties of crystals are not only of practical but also of scientific importance. The electro-optical properties of crystals characterize in some way their polarizability. In the case of ferroelectrics, the spontaneous electro-optical effect, i.e., the change in the optical properties of a ferroelectric due to the appearance of the spontaneous polarization, is a highly important subject. The investigation of the spontaneous effect yields information on the role of the electronic polarization in the processes associated with the appearance of the spontaneous polarization because, as is known, the refractive indices of crystals basically represent their electronic polarizability. It should be stressed that the problem of the spontaneous polarization is of itself the most important in ferroelectricity. Investigations of the electro-optical properties of crystals are important also in the elucidation of the nature of the spontaneous polarization of linear pyroelectrics.

Up till now, quantitative measurements of the electro-optical properties have been carried out on a small number of crystals—about 20, including about ten ferroelectrics.

From the theoretical point of view, the problems of electro-optics are equivalent to the problems of the electronic polarizability of crystals. [62,63] * In spite of this, there are as yet practically no papers on the polarizability of crystals treated from the point of view of their electro-optical properties. The theoretical treatments of the electro-optical phenomena are usually limited to a thermodynamic description of the electro-optical effect.

The electro-optical properties of crystals now find wide practical applications. Some of these applications will be considered at the end of the present paper.

1. RELATIONSHIPS DESCRIBING THE ELECTRO-OPTICAL EFFECT

The optical properties of anisotropic media may be described by index ellipsoids (optical indicatrices), defined (in the principal system of coordinates) by the equation

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*The optical properties of crystals are described best in terms of refractive index ellipsoids. Using such a description, we may say (in graphical terms) that the electro-optical effects are the result of the "deformation" of these ellipsoids under the action of an electric field.

*We mean here the electro-optics in the visible part of the spectrum. In the infrared region, the electro-optical effect may obviously be associated also with the ionic polarizability of crystals.

$$\frac{x^2}{n_1^2} + \frac{y^2}{n_2^2} + \frac{z^2}{n_3^2} = 1. \qquad (2)$$

A triaxial ellipsoid describes the optical properties of crystals of lower symmetry systems (orthorhombic, monoclinic, triclinic); an ellipsoid of revolution describes crystals of intermediate symmetry systems (tetragonal, trigonal, and hexagonal); and a sphere describes the optical properties of cubic crystals.

Making the following substitutions in Eq. (2)

$$\frac{1}{n_1^2} = a_0^2, \qquad \frac{1}{n_2^2} = b_0^2, \qquad \frac{1}{n_0^2} = c_0^2,$$

we can write this equation in the form

$$a_0^2 x^2 + b_0^2 y^2 + c_0^2 z^2 = 1. \qquad (3)$$

In an arbitrary system of coordinates, Eq. (3) is written in the form

$$a_{11}^0 x^2 + a_{22}^0 y^2 + a_{33}^0 z^2 + 2a_{23}^0 yz + 2a_{31}^0 zx + 2a_{12}^0 xy = 1, \qquad (4)$$

where the index "0" represents the state of a crystal in the absence of an electric field. The coefficients a_{ik}^0, known as the polarization constants, are expressed in terms of the reciprocals of the principal values of the refractive indices a_0, b_0, c_0, using the well-known formulas for the transformation of the components of a polar tensor:

$$\begin{cases}
a_{11}^0 = c_{11}^2 a_0^2 + c_{21}^2 b_0^2 + c_{31}^2 c_0^2, \\
a_{22}^0 = c_{12}^2 a_0^2 + c_{22}^2 b_0^2 + c_{32}^2 c_0^2, \\
a_{33}^0 = c_{13}^2 a_0^2 + c_{23}^2 b_0^2 + c_{33}^2 c_0^2, \\
a_{23}^0 = c_{12}c_{13} a_0^2 + c_{22}c_{23} b_0^2 + c_{32}c_{33} c_0^2, \\
a_{31}^0 = c_{13}c_{11} a_0^2 + c_{23}c_{21} b_0^2 + c_{33}c_{31} c_0^2, \\
a_{12}^0 = c_{11}c_{12} a_0^2 + c_{21}c_{22} b_0^2 + c_{31}c_{32} c_0^2.
\end{cases} \qquad (5)$$

In the principal system of coordinates of the index ellipsoid [cf. Eqs. (3) and (4)], we have

$$a_{11}^0 = a_0^2, \qquad a_{22}^0 = b_0^2, \qquad a_{33}^0 = c_0^2, \qquad a_{23}^0 = a_{31}^0 = a_{12}^0 = 0.$$

Upon application of an electric field, the ellipsoid changes and, in general, its new principal axes do not coincide with the old axes. We shall denote the new polarization constants (allowing for the applied field) by the symbols a_{ik}, and the new reciprocals of the principal indices will be denoted by a*, b*, c*. These new polarization constants, expressed in terms of the reciprocals of the new principal indices, are written as follows in the old (initial) principal system of coordinates

$$\begin{cases}
a_{11} = c_{11}^2 a^{*2} + c_{21}^2 b^{*2} + c_{31}^2 c^{*2}, \\
a_{22} = c_{12}^2 a^{*2} + c_{22}^2 b^{*2} + c_{32}^2 c^{*2}, \\
a_{33} = c_{13}^2 a^{*2} + c_{23}^2 b^{*2} + c_{33}^2 c^{*2}, \\
a_{23} = c_{12}c_{13} a^{*2} + c_{22}c_{23} b^{*2} + c_{32}c_{33} c^{*2}, \\
a_{31} = c_{13}c_{11} a^{*2} + c_{23}c_{21} b^{*2} + c_{33}c_{31} c^{*2}, \\
a_{12} = c_{11}c_{12} a^{*2} + c_{21}c_{22} b^{*2} + c_{31}c_{32} c^{*2}.
\end{cases} \qquad (6)$$

The increments (changes) in the polarization constants, due to the application of an electric field, re-

ferred to the old system of coordinates, are [from Eqs. (5) and (6)]

$$\left.\begin{array}{ll}
a_{11} - a_0^2, & a_{23}, \\
a_{22} - b_0^2, & a_{31}, \\
a_{33} - c_0^2, & a_{12}.
\end{array}\right\} \qquad (7)$$

In an analytic description of the electro-optical effect one considers the relationship between the changes in the polarization constants and the electric field (polarization).

We shall introduce the notation:

$$\begin{array}{ll}
\Delta a_{11} = a_{11} - a_0^2, & \Delta a_{23} = a_{23}, \\
\Delta a_{22} = a_{22} - b_0^2, & \Delta a_{31} = a_{31}, \\
\Delta a_{33} = a_{33} - c_0^2, & \Delta a_{12} = a_{12}
\end{array}$$

$(\Delta a_{23} = \Delta a_{32}, \ \Delta a_{31} = \Delta a_{13}, \ \Delta a_{12} = \Delta a_{21}).$

Then, it follows from the foregoing treatment that the equations for the linear electro-optical effect will have the form

$$\Delta a_{ij} = r_{ijk} E_k, \qquad (8)$$

$$\Delta a_{ij} = m_{ijk} P_k, \qquad (9)$$

where r_{ijk} and m_{ijk} are the coefficients of the linear electro-optical effect, which are polar third-rank tensors, and E_k and P_k are, respectively, the electric field intensity and the polarization. *

In the matrix notation, using two indices for a third-rank tensor and introducing the notation

$$\begin{array}{ll}
\Delta a_1 = a_{11} - a_0^2, & \Delta a_4 = a_{23}, \\
\Delta a_2 = a_{22} - b_0^2, & \Delta a_5 = a_{31}, \\
\Delta a_3 = a_{33} - c_0^2, & \Delta a_6 = a_{12},
\end{array}$$

Eqs. (8) and (9) may be written in the form

$$\Delta a_i = r_{ij} E_j, \qquad (8a)$$

$$\Delta a_i = m_{ij} P_j. \qquad (9a)$$

The matrix form of the first of these equations is given by the table

	E_1	E_2	E_3
$a_{11}-a_0^2$	r_{11}	r_{12}	r_{13}
$a_{22}-b_0^2$	r_{21}	r_{22}	r_{23}
$a_{33}-c_0^2$	r_{31}	r_{32}	r_{33}
a_{23}	r_{41}	r_{42}	r_{43}
a_{31}	r_{51}	r_{52}	r_{53}
a_{12}	r_{61}	r_{62}	r_{63} .

$$(8b)$$

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*It is desirable to associate the electro-optical properties of crystals with the polarization (and not with the electric field) in the case of the spontaneous electro-optical effect in ferroelectrics. Under real conditions, due to the finite conductivity of a crystal and the surrounding medium, the equations $E_{dep} = 0$, $D = 4\pi P$ (where E_{dep} is the depolarizing field) govern the appearance of the spontaneous polarization in a ferroelectric. Precisely this relationship between D and P ($D/4\pi = P$) is assumed in Eq. (9). We note that under the conditions of the "induced" electro-optical effect in a ferroelectric (the effect induced by an external field), the equation $D/4\pi = P$ is also satisfied approximately because the relationship $\epsilon \approx 4\pi\alpha$ ($\epsilon \gg 1$) is satisfied approximately due to the high polarizability of ferroelectrics.

Table I. Matrix form of the electro-optical coefficients (r_{ij})
and (m_{ij}) of various crystals: component equal to zero;
● component not equal to zero; ${}^{O}_{●}\!-\!{}^{O}_{●}$ equal components.
●-o components equal in magnitude but opposite in
sign. The number of nonzero coefficients is given
parentheses after each matrix.

When this notation is used, the matrices of the co-
efficients r_{ij} and m_{ij} are fully equivalent to the ma-
trices of the piezoelectric coefficients n_{ij} and e_{ij}. [2]
The formal relationship between the electro-optical
(linear effect) and the piezoelectric phenomena, de-
scribed by third-rank tensors, extends even further:
in accordance with symmetry considerations, all piezo-
electric crystals should exhibit the linear electro-
optical effect. Hence, it follows that, for example, the
electro-optical properties should be exhibited by all
crystals which do not have a center of symmetry (with
the exception of acentric cubic class O). *

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*The notation of crystal classes follows Schönflies (cf. [²]).

The symmetry of crystals imposes certain restric-
tions on the matrices of the coefficients r_{ij} [cf.
Eq. (8b)] and m_{ij}. Some of these coefficients are the
same for several classes, others are equal to zero,
etc. Table I gives the matrices of the coefficients r_{ij}
of all the electro-optical crystal classes (linear effect).
The matrices of the coefficients m_{ij} are fully equivalent
to the matrices of the coefficients r_{ij}.

Since the linear electro-optical effect is possible
only in piezoelectric crystals, it follows that the linear
effect and the converse piezoelectric effect coexist.
This means that in the description of the electro-
optical effect we must allow for the piezo-optical effect:
the change in the optical constants due to the deform-

Table 1. (Cont'd)

Hexagonal system

Class C_6

(4)

Class C_{6v}

(3)

Class D_6

(1)

Class D_{3h}

(3)

Class D_{3h}

$m \perp x$

(1)

Class D_{3h}

$m \perp y$

(1)

Orthorhombic system

Class V

(3)

Class C_{2v}

(5)

Tetragonal system

Class C_4

(4)

Class S_4

(4)

Class D_4

(2)

Class C_{4v}

(3)

Class V_d

(2)

Cubic system

Classes T_d and T

(1)

ations caused by the converse piezoelectric effect. The change in the polarization constants when an electric field is applied, to a crystal, which is not associated with the converse piezoelectric effect (i.e., the piezo-optical effect), represents the true linear electro-optical effect. Thus, the true linear electro-optical effect is solely due to the direct influence of an electric field on the charge in a dielectric, which causes a redistribution of the density of the electron clouds of the particles in the dielectric. The false linear electro-optical effect represents the change in the polarization constants due to the converse piezoelectric effect (through the piezo-optical effect). The division of the electro-optical effect into the true and false effects is in many respects analogous to the division of the pyroelectric effect into the true and false effects. In contrast to the pyroelectric effect, the true electro-optical effect is considerably greater than the false electro-optical effect.

The equations for the electro-optical effect (both true and false taken together) have the form

$$\Delta a_{ij} = r_{ijk}E_k = r^*_{ijk}E_k + p_{ijlm}r_{lm},$$
$$\Delta a_{ij} = m_{ijk}P_k = m^*_{ijk}P_k + p_{ijlm}r_{lm}, \qquad (10)$$

where r^*_{ijk} and m^*_{ijk} are the coefficients of the true electro-optical effect, r_{lm} is the deformation tensor (second-rank tensor), p_{ijlm} is the tensor of the elasto-optical coefficients (fourth-rank tensor). We note that the false electro-optical effect [the second term on the right-hand side of equations denoted by (10)] may be also be written using a different piezo-optical coefficient, i.e., the coefficient π_{ijlm}. The tensors π and p are related by

$$p_{ijrs} = \pi_{ijkl}c_{klrs},$$
$$\pi_{ijkl} = p_{ijrs}s_{rskl}, \qquad (11)$$

where c is the elastic stiffness, and s is the compliance of a crystal.[2] The matrices of the tensor of the coefficients π are identical with the matrices of the electrostriction coefficients G and H.[3] The coefficients of the tensors p_{ijkl} and π_{ijkl} are commutative with respect to the inner indices i, j and k, l, but are not commutative with respect to index pairs.

Under experimental conditions, one measures the total effect, representing the true and false electro-optical effects. The false effect contribution is given by the second terms on the right-hand side of the equations denoted by (10). The false electro-optical effect may be regarded as a correction to the true effect. With this in mind, using the piezoelectric effect equations[2]

$$r_{lm} = d_{klm}E_k,$$
$$r_{lm} = g_{klm}P_k,$$

where d_{klm} and g_{klm} are the piezoelectric coefficients, Eq. (10) may be transformed into

$$\Delta a_{ij} = (r^*_{ijk} + p_{ijlm}d_{klm})\,E_k,$$
$$\Delta a_{ij} = (m^*_{ijk} + p_{ijlm}g_{klm})\,P_k. \qquad (12)$$

The products pd and pg in the above equations do indeed look like corrections to the coefficients of the true electro-optical effect, r* and m*. *

Vlokh and Zheludev[4] published a general treatment of the changes in the optical properties of crystals of all classes due to the linear electro-optical effect. These changes are given in Table II, which deals with the two cubic classes T_d and T when a field E is oriented along some of the most important directions. An examination of Table II and its analogs for other crystals, together with simple symmetry considerations, allow us to make some general remarks on the electro-optical properties of crystals.

Thus, in particular, the symmetry of the indicatrix of optically uniaxial crystals (crystals of intermediate symmetry, except crystals of the classes V_d and S_4) is not affected by the application of a field along a higher-order axis (optical axis): such crystals remain uniaxial. Moreover, an electric field applied along the [001] or [0001] directions does not change at all (in the linear approximation) the optical properties of crystals of some symmetry classes (D_4, D_3, C_{3h}, D_6, D_{3h}). The symmetry of the optical indicatrix of crystals of the classes V_d and S_4 is altered by a field applied along any direction (including the [001] direction) and such crystals transform from uniaxial to biaxial. A field along the [111] direction makes cubic crystals of the classes T and T_d optically uniaxial and a field along any other direction makes them optically biaxial.

In addition to the linear electro-optical effect, which occurs only in piezoelectric crystals, all dielectric crystals exhibit the quadratic electro-optical effect. In the latter effect, the changes in the polarization constants of crystals are proportional to the square of the electric field intensity (E^2) or the polarization (P^2) †

$$\Delta a_{ij} = L_{ijkl}E_kE_l,$$
$$\Delta a_{ij} = M_{ijkl}P_kP_l, \qquad (13)$$

where L and M are the coefficients of the quadratic electro-optical effect and are fourth-rank tensors. The coefficients of these tensors, like the electrostriction coefficients, are commutative with respect to the inner indices i, j and k, l, but are not commutative with respect to index pairs.

Using the notation employed in the present section

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*Sometimes the true electro-optical effect is called the "primary" effect and the false effect is called "secondary". Thus, the primary effect is observed in a crystal which cannot be deformed by the application of a field, while the total effect is observed in a crystal which can be deformed by a field (due to the piezoelectric effect). Hence, it follows that the secondary effect is equal to the difference between the total and primary effects:

$$r^* = r^r, \quad r = r^t, \quad r_{sec} = r - r^* = r^t - r^r.$$

†The meaning of P is the same as in Eq. (9).

Table II. Change in the optical properties of cubic crystals under the action of an electric field (linear electro-optical effect).

Initial symmetry class	Field direction	Symmetry class after application of a field		Indicatrix equation in the crystallographic system of coordinates x, y, z for the initial class	Canonical indicatrix equation in the principal system of x', y', z'	Angle between the crystallographic axes and the principal system of coordinates
		for T	for T_d			
T, T_d	$<001>$	$C_2(6)$	$C_{2v}(6)$	$a^2(x^2+y^2+z^2)$ $+2r_{41}E_z xy = 1$	$(a^2+r_{41}E_z^2)x'^2+(a^2 -r_{41}E_z)y'^2+a^2z'^2 =1$	$\xi_3 = \pm 45°$
	$<010>$	$C_2(6)$	$C_{2v}(6)$	$a^2(x^2+y^2+z^2)$ $+2r_{41}E_y zx = 1$	$(a^2+r_{41}E_y)x'^2+a^2y^2{}' +(a^2-r_{41}E_y)z'^2=1$	$\xi_2 = \pm 45°$
	$<100>$	$C_2(6)$	$C_{2v}(6)$	$a^2(x^2+y^2+z^2)$ $+2r_{41}E_x yz = 1$	$a^2x'^2+(a^2+r_{41}E_x)y'^2 +(a^2-r_{41}E_x)z'^2-1$	$\xi_1 = \pm 45°$
	$<111>$	$C_3(4)$	$C_{3v}(4)$	$a^2(x^2+y^2+z^2)$ $+2r_{41}(E_x yz$ $+E_y zx+E_z xy)$ $=1$		
	$<110>$	$C_1(12)$	$C_s(12)$	$a^2(x^2+y^2+z^2)$ $+2r_{41}(E_x yz$ $+E_y zx)=1$	$(a^2+r_{41}\sqrt{E_x^2+E_y^2})x'^2$ $+(a^2-r_{41}\sqrt{E_x^2+E_y^2})$ $\times y'^2+a^2z'^2=1$	$\xi_1=\xi_2=\pm 45°$

for the quantities Δa_{ij}, and going over to single indices in accordance with the notation $E_1E_1 = E_1^2$, $E_2E_2 = E_2^2$; $E_3E_3 = E_3^2$; E_3E_2, $E_2E_3 = E_4^2$; E_1E_3, $E_3E_1 = E_5^2$; E_1E_2, $E_2E_1 = E_6^2$, we obtain the following matrix form for the first of the two equations denoted by (13)

	E_1^2	E_2^2	E_3^2	E_4^2	E_5^2	E_6^2
Δa_1	L_{11}	L_{12}	L_{13}	L_{14}	L_{15}	L_{16}
Δa_2	L_{21}	L_{22}	L_{23}	L_{24}	L_{25}	L_{26}
Δa_3	L_{31}	L_{32}	L_{33}	L_{34}	L_{35}	L_{36}
Δa_4	L_{41}	L_{42}	L_{43}	L_{44}	L_{45}	L_{46}
Δa_5	L_{51}	L_{52}	L_{53}	L_{54}	L_{55}	L_{56}
Δa_6	L_{61}	L_{62}	L_{63}	L_{64}	L_{65}	L_{66} .

(14)

Similarly, we can write the second of the two equations (13). When our notation of the quantities Δa_{ij}, E_m, E_n, P_m, P_n is used, the matrices of the coefficients L_{pq} and M_{pq} are found to be completely identical with the matrices of the electrostriction coefficients G and H. [3]

The first of the two equations in (13) should be used in the description of the quadratic electro-optical effect due to the application of a field to a crystal, while the second equation is convenient in the description of the changes in the optical properties of ferroelectric crystals, associated with the appearance of (or the change in) the spontaneous polarization.

We can easily see that the quadratic effect can be also divided into the true and false effects. The true quadratic electro-optical effect represents the change in the polarization constants, proportional to the square of the applied field (polarization) due to the direct influence of an electric field (polarization) on the charges in a dielectric; the true quadratic electro-optical effect is not associated with the deformation of a crystal, which always occurs when a field is applied (or the polarization is altered), due to the

piezoelectric effect or the electrostriction. The false quadratic electro-optical effect represents the change in the polarization constants, proportional to the square of the electric field (polarization), due to the deformation of a crystal because of the electrostriction.

The true and false effects taken together are described by the relationships

$$\Delta a_{ij} = L_{ijkl}^* E_h E_l + p_{ijmn} r_{mn},$$
$$\Delta a_{ij} = M_{ijkl}^* P_h P_l + p_{ijmn} r_{mn},$$

(15)

which, by the use of the relationships (cf. [3])

$$r_{mn} = R_{mnkl} E_h E_l,$$
$$r_{mn} = Q_{mnkl} P_h P_l$$

(where R_{mnkl}, Q_{mnkl} are the electrostriction coefficients), may be written in the form

$$\Delta a_{ij} = (L_{ijkl}^* + p_{ijmn} R_{mnkl}) E_h E_l,$$
$$\Delta a_{ij} = (M_{ijkl}^* + p_{ijmn} Q_{mnkl}) P_h P_l.$$

(16)

This form of notation allows us to treat the false quadratic electro-optical effect, described by the products pR and pQ, as a correction to the coefficients of the true quadratic electro-optical effect, L* and M*.

The quadratic electro-optical effect (like, for example, electrostriction) is a second-order effect and, in general, gives rise to small (compared with the linear electro-optical effect) changes in the optical properties of dielectrics. However, for polar liquids— whose quadratic electro-optical effect is known as the Kerr effect[5] —and ferroelectrics, the quadratic effect (like the electrostriction) may be considerable. In the case of linear dielectric crystals, which exhibit the piezoelectric effect, the quadratic electro-optical effect may be neglected compared with the linear effect. We shall show that in an alternating electric

field of frequency f the optical properties of a crystal vary, due to the quadratic electro-optical effect, at a frequency 2f.

Investigations of the electro-optical properties of a crystal reduce, in the final analysis, to the determination of the change in the polarization constants of the crystal under the influence of an electric field (polarization). The changes in the polarization constants can be found most simply from increments in the refractive indices Δn and the angle of rotation χ of the optical indicatrix.

The quantities Δn and χ may be measured by various methods (refractometry, interferrometry, etc.). In real crystals, the problem may be reduced to the direct measurement not of these quantities but of quantities related to them, from which the electro-optical coefficients may be calculated. Such measurements are, as a rule, carried out in plane-polarized or circularly polarized light and they involve a visual or photometric determination of the varying intensity of the light transmitted by a system when an electric field is applied to a crystal. The electro-optical coefficients may be calculated from the value of the voltage necessary to produce a path difference of $\lambda/2$, the angle of rotation of the optical indicatrix under the influence of the field, and the angle of compensation, using various birefringent compensators.

Other methods are based on the measurement of the changes in the interference-conoscopic pattern under the influence of a field (measurement of the angle between optical axes, determination of the shift of interference bands). The electro-optical coefficients may also be determined by spectroscopic methods. One of these involves the measurement of the shift of interference bands in a continuous spectrum, obtained in a spectrograph by placing in front of its slit a birefringent plate of the crystal under investigation, located between crossed polarizers and in the maximum transmission position.

2. OPTICAL SYSTEMS USED TO INVESTIGATE THE ELECTRO-OPTICAL EFFECT [6,7]

Plane-parallel plates or parallelepipeds, cut from the investigated crystal, are used in the studies of the electro-optical effect. The cross section of the optical indicatrix, parallel to the plane of the plate, is, in general, an ellipse, whose semiaxes are equal to the refractive indices of two rays which are traveling at different velocities and whose light vectors vibrate along these axes. Under the action of a field, this cross section of the indicatrix changes (Fig. 1), i.e., the ellipse may, in general, rotate and its semiaxes—refractive indices—change (Fig. 1a). In special cases, this change may be one of the following three types: rotation of the ellipse (Fig. 1b), deformation of the ellipse (Fig. 1c), and finally, in the case of a circular cross section of the indicatrix (Fig. 1d), the circle
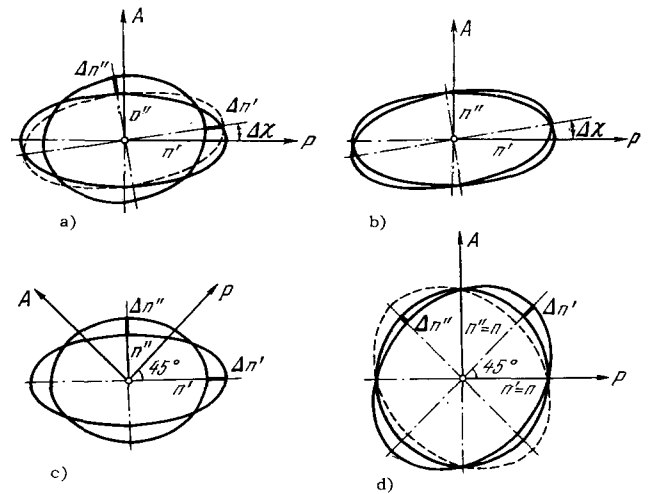

FIG. 1. Various types of changes in the optical indicatrix of a crystal under the action of an electric field and the orientation of the cross section of the indicatrix in crossed polarizers, used in various types of change of the indicatrix. A — analyzer; P — polarizer; n' and n" — refractive indices; $\Delta n'$ and $\Delta n"$ — increments in the refractive indices; $\Delta\chi$ — angle of rotation of the elliptical cross section of the optical indicatrix.

may change into a circle of different radius or may transform into an ellipse oriented along a certain direction (in particular, at an angle of ± 45° to the crystallographic axes), or it may remain unchanged (if the effect is absent).

Depending on the nature of the change in the optical indicatrix induced by a field, the electro-optical effect and its applications are nowadays investigated using mainly the following four types of optical systems (Fig. 2):

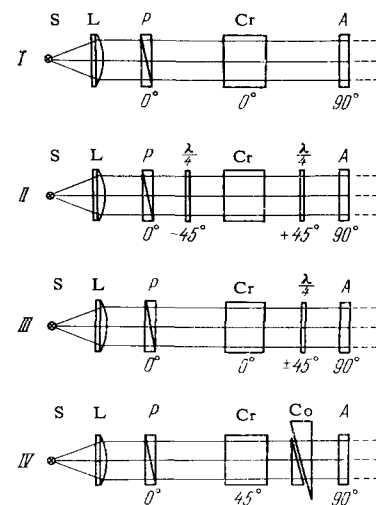I. A crystal is placed between crossed flat

FIG. 2. Types of optical systems used to observe the electro-optical effect, and to modulate and interrupt light beams. S — source of light; L — lens; P — polarizer; A — analyzer; $\lambda/4$ — quarter-wave plate; Cr — crystal; Co — compensator. The orientations of the elements are given in degrees.

polarizers; a parallel or convergent monochromatic beam, recorded by a photomultiplier, is passed through the system.

II. A crystal is placed between left-handed and right-handed circular polarizers, consisting of polaroids and $\lambda/4$ mica plates, whose crystallographic axes make, respectively, $\pm 45°$ with the plane of polarization of the crossed polaroids.

III. Only one $\lambda/4$ plate, oriented at $\pm 45°$ as in system II, is placed in front of a polarizer in system I.

IV. System I is supplemented by some type of birefringent compensator, which is placed behind the polarizer or in front of the analyzer. This system is used mainly for making measurements, while the first three systems are applied to the modulation of light.

The orientation of the crystal plate in an optical system depends on the nature of the change in the elliptical cross section of the optical indicatrix and on the type of problem to be solved (measurement of constants, modulation of light, etc.).

If the cross section of the indicatrix is elliptical, the plate in system I is placed normal to the rays or in the extinction position. This position is used when the electric field mainly rotates the elliptical cross section of the indicatrix and only slightly deforms it. The intensity of the transmitted light will increase as the intensity of the field applied to the crystal is increased.

If the effect of a field on a crystal is simply to deform the elliptical cross section of the indicatrix, the plate is placed in the position of maximum transmission in the absence of a field, i.e., so that the axes of the ellipse make an angle of 45° with the planes of polarization of the crossed polarizers. The intensity of the transmitted light will change when the electric field is applied and increased. The light transmitted by a system in the absence of a field may be extinguished by means of a birefringent compensator introduced into the system (for example, an exactly identical plate but rotated by 90° about the axis of the system, which corresponds to case IV).

If the cross section of the indicatrix is circular, we may use systems I, II, and III. Then, if the circle becomes elongated into an ellipse under the influence of a field, in the absence of a field, the plate is placed in such a way that the principal axes of this ellipse are rotated by 45° with respect to the direction of vibrations in the crossed polaroids. As the field is increased, the intensity of light will rise from zero (in the case of systems I and II), reaching its maximum when the path difference is equal to half the wavelength.

It should be mentioned that a crystal plate whose indicatrix has a circular cross section does not require special orientation in system II with the right-handed and left-handed circular polarizers and analyzers. When system III is used, in the presence of a plate not subjected to a field the intensity of the transmitted light is not equal to zero but decreases or increases

depending on the direction of the applied field vector. It decreases to zero or increases to a maximum value in a field which makes the additional path difference equal to $\lambda/4$. Therefore, 100% modulation of light is obtained in system III using an effective field intensity half that required in systems I and II.

In the absence of a field, light is not transmitted through systems I, II, and IV and the frequency of modulation is twice as high as the frequency of the alternating electric field applied to a crystal. On the other hand, system III transmits half the maximum intensity of light in the absence of a field, and when an alternating field is applied to a crystal the light is modulated at the frequency of the applied field. Figure 3 shows the characteristics of the light modulation for systems I, II and IV, and separately for system III.
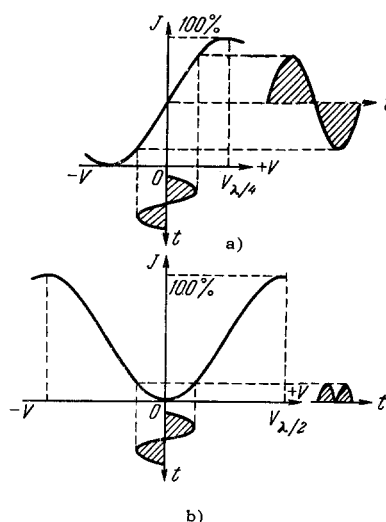
FIG. 3. Nature of the transmission and modulation of light in electro-optical systems: a) I, II, IV; b) III. J — transmission; V — voltage; t — time.

Fields are applied to crystals using a variety of electrodes. Opaque electrodes are used in those cases when light must pass through a crystal at right angles to the field. Transparent or grid electrodes are used when light should pass along the direction of the field. Transparent electrodes are deposited either directly on the crystal surface (zinc oxide, semitransparent gold electrode), or on glass plates (stannous oxide), which are in contact with the crystal. Sometimes, conducting liquid electrodes are used. Grid electrodes are deposited on the crystal surface in the form of sufficiently thick low-resistance films. The most rational form of electrodes is the ring or grid type.

Recently, microwave modulation of light has been achieved by placing a crystal at an antinode of a standing electric field wave in a waveguide. The light passes through the crystal along the direction of the vector of the applied electric field or at right angles

to it. The crystal is sometimes placed in a traveling wave of microwave frequency.

Because of their high resistance, transparent electrodes are used in low-frequency modulators[8,9] (up to 100 kc) and in constant fields. Grid electrodes allow us to increase the modulation frequency; when a ring electrode of indium is used, the modulation frequency may be raised to 50 Mc,[10] and the waveguide method of applying a field to a crystal imposes no frequency limit at all. In practice, it has been possible to measure the permittivity, employing the microwave technique, at 36 Gc.[11]

3. SOME REMARKS ON THE DIRECT DETERMINATION OF THE ELECTRO-OPTICAL COEFFICIENTS

The circular z cross section of the optical indicatrix of tetragonal ADP ($NH_4H_2PO_4$) and KDP (KH_2PO_4) crystals* is elongated, by the action of a field E_z, (along the optical axis), into an ellipse oriented at an angle of $\pm 45°$ to the crystallographic axes. When system I (Fig. 2) is used, the intensity of light transmitted along the axis of the system, for a suitable orientation of a crystal with respect to the field E_z, is given by the following formula:[12]

$$J = \frac{I}{I_0} = \sin^2 \frac{\pi V n_0^3 r_{63}}{\lambda} , \qquad (17)$$

where λ is the wavelength, n_0 is the ordinary refractive index, $V = E_z d$ is the voltage across the crystal. From this formula, it follows that the intensity will reach its maximum value ($J = 1$) at a voltage V' ensuring a path difference $\lambda/2$, i.e., when

$$r_{63} = \frac{\lambda}{2V'n_0^3} . \qquad (18)$$

Using this formula, we can determine the electro-optical coefficient r_{63} and its dispersion by passing monochromatic light of various wavelengths through a crystal and by measuring the corresponding voltages V' necessary to obtain the path difference $\lambda/2$.

When system II (Fig. 2) is used, the electro-optical coefficient r_{63} of ADP-type crystals may be determined from the angle of compensation, i.e., the angle through which the analyzer has to be turned in order to compensate (or to reduce to a minimum) the intensity of light obtained for a given voltage across a crystal. The angle of compensation Φ depends linearly on the value of the voltage across a crystal

$$\Phi = \frac{r_{63}n_0^3\pi}{\lambda} V,$$

which makes it possible to determine r_{63} for any voltage across the crystal.

---

*ADP and KDP crystals belong to the V_d class (a fourfold reflection axis is the optical axis of these crystals and they have also two twofold axes, which are perpendicular to the optical axis, and two symmetry planes passing through the fourfold reflection axis and making angles of 45° with the twofold axes).
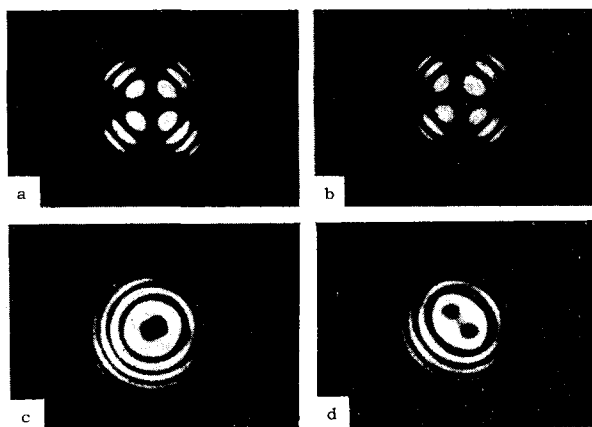

FIG. 4. Interference conoscopic patterns, obtained when light is transmitted through a z-cut plate of ADP crystal. Linearly polarized light, crossed polarizers: a) without any field; b) in a field $V_z = 5$ kV. Circularly polarized light, dark spots represent the emergence of the optical axes: c) without any field; d) in a field $V = 5$ kV.

The interference-conoscopic pattern, obtained when linearly polarized and circularly polarized light is transmitted through a z-cut plate made of a crystal with the ADP or KDP symmetry, is shown in Fig. 4.

We have pointed out that system I is used in those cases when the angle of rotation of the optical indicatrix depends on the field E. Knowing the dependence of the angle of rotation of the indicatrix on the voltage across a crystal, we can find the electro-optical coefficients from the experimentally determined angles of rotation of the optical indicatrix corresponding to a given voltage across the crystal. Thus, for example, in pentaerythritol crystals subjected to a field at right-angles to their cleavage planes, i.e., along the symmetry axis denoted by the index 2, the cross section of the optical indicatrix, which is elliptic in the absence of a field,[13] is rotated by an angle ξ_2 when light is passed along the field direction, the angle ξ_2 being given by[14]

$$\tan 2\xi_2 = \frac{2r_{52}E_y}{(a_0^2 - c_0^2) + (r_{12} - r_{32})E_y} , \qquad (19)$$

where a_0 and c_0 are the reciprocals of the principal values of the refractive indices for a given cross section in the absence of a field. Having determined experimentally the values of the angle ξ_2 for two different values of the voltage across a crystal and substituting these values in a system of two equations of the Eq. (19) type, we can determine r_{52} and $(r_{12} - r_{32})$.

In the case of crystals belonging to the D_3 class— in particular quartz crystals—the measurement of the electro-optical coefficients is carried out very simply by the use of two y-cut plates, placed between crossed nicols and oriented in such a way as to compensate their double refraction (x axis of the first plate coincides with the z axis of the second plate, and conversely). Here, an electric field is applied to one of the plates

along the x axis and light is propagated along the y axis.

When the principal axes of the optical indicatrices of these plates make angles of $\pm 45°$ with the planes of polarization of the nicols, then, in accordance with the formula for the intensity of light, the total (or maximum) transparency of the system is obtained when the field establishes a path difference of $\lambda/2$

$$\frac{1}{2}\frac{\pi d_y r_{11} n_0^3 E_x'}{\lambda} = \frac{\pi}{2}. \tag{20}$$

Here, d_y is the thickness of a plate along the y axis direction, n_0 is the ordinary refractive index of the crystal, λ is the wavelength of the light employed, and E_x' is the field intensity ensuring the path difference $\lambda/2$. The electro-optical coefficient r_{11} follows from Eq. (20):

$$r_{11} = \frac{\lambda}{E_x' d_y n_0^3}. \tag{21}$$

In the case of cubic crystals belonging to the T_d class (ZnS, CuCl, urotropin), the electro-optical coefficients can be determined relatively easily by using different directions of the light and the field. The transverse effect can be found very simply: the field is applied along the [110] direction and the light is propagated along the same direction. The double refraction induced by the field for this direction of propagation of light is represented by $\frac{1}{2}n_0^3 r_{41} E$. When the plate thickness in the direction of propagation is d, the path difference becomes

$$\Gamma = \frac{\pi}{\lambda} d n_0^3 r_{41} E, \tag{22}$$

where n_0 is the ordinary refractive index, E is the

field intensity, and λ is the wavelength. If the double refraction or the path difference is known, the value of the coefficient r_{41} can be determined.

4. LINEAR ELECTRO-OPTICAL EFFECT IN LINEAR DIELECTRICS

As mentioned earlier, all piezoelectric crystals should, in principle, exhibit the linear electro-optical effect. This follows, for example, from the fact that the electro-optical effect is described by a third-rank polar tensor, which for all crystals is identical with the piezoelectric tensor. However, one must point out, that because of the smallness of the effect, the difficulty of detecting and measuring it, and—until very recently—the few practical applications of the electro-optical properties of crystals, the electro-optical properties have been investigated only for a very small number of crystals (less than twenty). We must add that the electro-optical properties of even these twenty crystals were in most cases not fully investigated.

Quartz and tourmaline were the first crystals in which the electro-optical effect was discovered. [1,15,16] Only one electro-optical coefficient, r_{22}, has been determined for tourmaline (Table III).

For quartz, two electro-optical coefficients, r_{11} and r_{41}, have been determined. The electro-optical properties of crystals may be interpreted by the characteristic surfaces of third-rank polar tensors. In the case of quartz, the characteristic surface of the linear electro-optical effect is completely analogous to the characteristic surface of the piezoelectric effect.

Table III. Electro-optical coefficients of some linear dielectrics

Crystal	Symmetry	$r_{ij} \times 10^8$ cgs esu	Voltages necessary to produce path difference $\lambda/2$ ($\lambda = 5461$ Å), kV	Reference
1. Quartz	D_3	$r_{11}=0.59$ $r_{41}=1.4$	~ 33	1,15–20
2. Tourmaline	C_{3v}	$r_{22}=0.9$		1
3. NaClO$_3$	T	$r_{41}=1.19$	200	1
4. ZnS	T_d	$r_{41}=5.0$	~ 10	21, 22
5. CuCl	T_d	$r_{41}=18.4$	6.2	23
6. Pentaerythritol	$S_4(C_2)$	$r_{52}=4.38$ $r_{32}-r_{12}=2.09$		14
7. CdS	C_{6v}	$r_{51}=11.1$		24
8. Urotropin	T_d	$r_{41}=5.1$	~ 15	25, 26
9. GaAs	T_d	$r_{41}=12.5$		27
10. K$_2$S$_2$O$_6$	D_3	$r_{11}=0.79$		25
11. Cs$_2$C$_4$H$_4$O$_6$	D_3	$r_{11}=3.00$		25
12. (C$_6$H$_{12}$O$_6$)$_2$·NaBr·H$_2$O	D_3	$r_{11}=0.30$		25
13. LiKSO$_4$	C_6	$r_{11}-r_{33}=4.80$		25
14. (NH$_4$)$_2$Cd$_2$(SO$_4$)$_3$	T_d	$n_0^3 r_{41}=8.70$		28
15. (NH$_4$)$_2$Mn$_2$(SO$_4$)$_3$	T_d	$n_0^3 r_{41}=6.30$		28
16. Na$_3$SbS$_4$·9H$_2$O	T	$n_0^3 r_{41}=16.32$		29
17. ZnSe	T_d	$r_{41}=4.82$		30
18. Na$_8$(AlSiO$_4$)(OHCl)$_2$	T_d	$r_{41}=5.00$		31

The electro-optical properties of AT- and BT-cut plates of quartz were investigated[41-45] because of the use of quartz in the modulation of light and in optical range-finders. For BT-cuts it was found that, in a constant field $E_y = E \sin 49°$, the change in the ordinary refractive index due to a change in the field by 1 cgs esu is equal to $\Delta n_0/\Delta E_y = 3.3 \times 10^{-8}$, and in an alternating field of resonance frequency this change is 3.3×10^{-5}. Thus, because of the high value of the Q-factor of quartz, a field alternating at the resonance frequency alters the refractive index by an amount which is 1000 times greater than the change induced by an equal constant field. However, it must be mentioned that 100% modulation of light is not obtained in quartz plates at their resonance frequency because the fields required for such modulation are stronger than those which fracture quartz by strong mechanical deformation.

The values of the electro-optical coefficients of quartz listed in Table III represent the total effect, both true and false. Using Eq. (12), we can easily estimate the contribution of the false effect. Calculations show that $\approx 80\%$ of the coefficient r_{11} represents the true effect and $\approx 20\%$ the false effect. Evidently, in general, the contribution of the false electro-optical effect in linear piezoelectric materials is greater than the contribution of the spontaneous false electro-optical effect in ferroelectric materials (see below), at least in the phase transition region.

The electro-optical properties have been investigated recently for several other trigonal crystals [$K_2S_2O_6$, $Cs_2C_4H_4O_6$, $(C_6H_{12}O_6)_2 \cdot NaBr \cdot H_2O$] and one hexagonal crystal, $LiKSO_4$. The information about their electro-optical properties is also given in Table III.

The electro-optical properties have been investigated for several cubic linear dielectrics: sodium chlorate $NaClO_3$, sphalerite ZnS, copper chloride CuCl, gallium arsenide GaAs, urotropin $C_6H_{12}N_4$, langbeinite [$(NH_4)_2Cd_2(SO_4)_3$, $(NH_4)_2Mn_2(SO_4)_3$], sodalite type crystals [$Na_8(AlSiO_4)(OHCl)_2$, ZnSe] and several others. The electro-optical properties of cubic crystals are attracting much attention because of the possibility of using these crystals in light sources with wide-angle apertures. The values of the electro-optical coefficients of these crystals are listed in Table III. The electro-optical coefficient r_{41} of CuCl is large compared with other linear dielectrics. Unfortunately, good-quality CuCl crystals are difficult to synthesize.

The electro-optical properties of crystals of pentaerythritol $C(CH_2OH)_4$ are very characteristic. It has been found[13] that some growth pyramids of this crystal have the monoclinic symmetry (class C_2), due to mechanical stresses during growth, while a crystal formed of four growth pyramids has the S_4 symmetry. The latter symmetry is confirmed by the x-ray diffraction investigation of this crystal. Investigations of the electro-optical properties of pentaerythritol have been carried out only on monoclinically distorted samples. Because of the distorted symmetry of such crystals (class C_2), they have eight independent electro-optical coefficients (axis 2 of these crystals is taken as the y axis). The coefficients r_{52} and the difference $(r_{32} - r_{12})$ have been determined experimentally (cf. Table III). The field in these determinations was applied along the y axis, and the light was propagated along the same direction, in a plate placed between polaroids. The value of the coefficient r_{52} was calculated from the angle of rotation of the optical indicatrix under the action of a given field [cf. Eq. (19)].

The electro-optical properties were recently discovered in the hexagonal crystal CdS (class C_{6v}). The value of the electro-optical coefficient r_{52} of this crystal is also given in Table III. The electro-optical properties of CdS crystals are of considerable interest in view of the fact that the ultrasound may be amplified by the conduction electrons (excited by an internal photoeffect) in these crystals.

5. LINEAR ELECTRO-OPTICAL EFFECT IN FERROELECTRICS

The electro-optical properties of ferroelectrics depend considerably on whether the sample is electrically short-circuited (E = const) or open-circuited (D = const) during measurements. It follows from Eq. (8) that the coefficient r_{ijk} is obtained when the electric field intensity is constant and the coefficient m_{ijk} is obtained when the electric induction is constant. It is known that some coefficients, representing the properties of ferroelectrics in the phase-transition region (elastic constants, piezoelectric coefficients), vary slightly in that region if they are measured under the condition of constant electric induction (open-circuited state) but alter considerably if they are measured in a constant field (short-circuited state). It is found that the same applies to the electro-optical coefficients. When D = const, the electro-optical effect is independent of the change in the permittivity ϵ of a crystal. The electro-optical effect does, however, depend on a change in ϵ if the field intensity is constant. Thus, in the latter case, the electro-optical effect is governed not only by the spontaneous polarization, but also by an additional charge obtained from outside. In other words, the electro-optical properties of ferroelectrics are better described by Eq. (9) and represented by the coefficient m_{ijk}. Investigations of a number of ferroelectrics have shown that this coefficient has no anomalies in the phase-transition region.

The spontaneous electro-optical effect in barium titanate ($BaTiO_3$) can be deduced from the temperature dependences of the spontaneous polarization (Fig. 5) and refractive indices (Fig. 6). From these two figures, it follows that, in the tetragonal modification
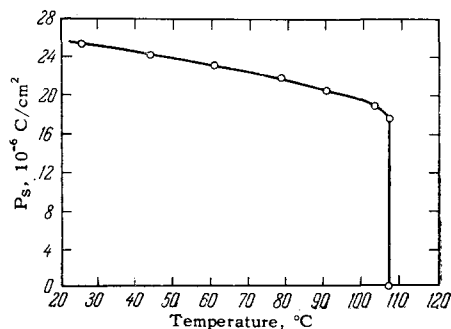
FIG. 5. Temperature dependence of the spontaneous polarization of $BaTiO_3$ (according to Merz [56]).
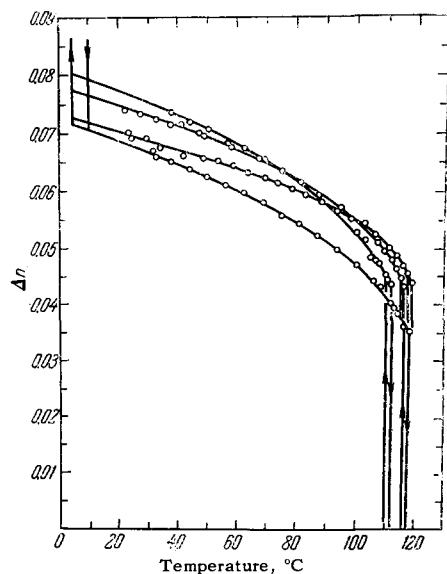

FIG. 6. Temperature dependence of the double refraction of four different crystals of tetragonal $BaTiO_3$ (according to Meyerhofer [57]).

(which exhibits the linear electro-optical effect), a polarization increment of 8×10^{-6} C/cm^2 corresponds to a change in the refractive indices by $\Delta n = -0.03$. This allows us to estimate the electro-optical coefficient m_{33} of this crystal:

$$m_{33} = 1.87 \cdot 10^{-7} \text{ cgs esu.}$$

An estimate of the product pg [cf. second expression in Eq. (12)] shows that only 5% of this quantity may be ascribed to the false electro-optical effect. No direct measurements of the electro-optical properties of the tetragonal modification of $BaTiO_3$ have yet been published.

The linear electro-optical effect in ferroelectrics has been investigated in greatest detail for KDP crystals. It should be mentioned that the generally pronounced electro-optical properties of this crystal (and other isomorphous compounds) have stimulated and greatly helped in the investigation and application of the electro-optical properties in general.

At room temperature, potassium dihydrogen phos-

phate KH_2PO_4 is an optically negative uniaxial crystal. Its refractive indices for the D-line of sodium at 15°C are: $n_1 = n_2 = n_a = 1.5095$ and $n_3 = n_c = 1.4684$, and the double refraction is represented by $\Delta n = n_3 - n_1 = -0.0411$. The appearance of the spontaneous polarization at $-150°$ C is accompanied by the linear electro-optical effect because before the transition, in the para-electric modification, the crystal is piezoelectric. Consequently, the optical indicatrix of the crystal transforms from an ellipsoid of revolution to a triaxial ellipsoid by compression and expansion of the ellipsoid of revolution along two mutually perpendicular directions, [110] and [1$\bar{1}$0]. This deformation of the optical indicatrix takes place simultaneously with a shear deformation of the crystal in the (001) plane and a dilation (compression) along the directions [110] and [1$\bar{1}$0]. The temperature dependence of the increment in the double refraction, due to the spontaneous polarization, is plotted in Fig. 7. This increment is superimposed on the double refraction mentioned above.
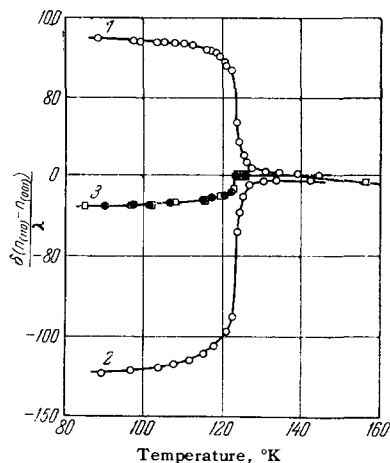

FIG. 7. Spontaneous change in the double refraction of a KH_2PO_4 crystal. 1) A crystal polarized "upward" by a field of $+3000$ V/cm; 2) crystal polarized "downward" by a field of -3000 V/cm; 3) curve, obtained by averaging the values for curves 1 and 2, identical with the curve for an open-circuited crystal (according to Zwicker and Scherer [32]).

In accordance with Eq. (12) and Table I, the spontaneous increment in the polarization constant a_6 of KH_2PO_4 due to the linear electro-optical effect can be found from the expression

$$a_6 = (m_{63}^* + p_{66}g_{36}) P_s \qquad (23)$$

or from the expression in terms of the refractive indices:

$$\frac{1}{n_1'^2} - \frac{1}{n_1^2} = (m_{63}^* + p_{66}g_{36}) P_s, \qquad (24)$$

where the primed refractive index represents the new value of the index after the phase transition to the ferroelectric region. The value of the coefficient m_{63} can be found from the above equation provided the

value of the refractive index n_1' is known (we may assume that $p_{66} = -2 \times 10^{-2}$ cgs esu, $g_{36} = 5 \times 10^{-7}$ cgs esu). Figure 7 allows us to calculate the value of this coefficient. In fact, curves 1 and 2 in Fig. 7 represent: 1) an increase in the double refraction, which is the difference between the highest refractive index n_1' = ($n_1 + \alpha$) and the index n_3'; 2) a reduction in the double refraction, which in this case is the difference between the reduced refractive index $n_2' = (n_1 - \alpha)$ and the index n_3' (α represents the change in the refractive indices $n_1 = n_2$ due to the spontaneous polarization; under experimental conditions, a change in the direction of the field orienting the domains leads to changes in these differences, because a change in the direction of polarization of the domains transforms the long axes of the optical indicatrix into short axes, and conversely). Thus, we find from Fig. 7 that at 100°K

$$n_1 + \alpha - n_3' - \Delta n = 85.5893 \cdot 10^{-8},$$

$$n_1 - \alpha - n_3' - \Delta n = -120.5893 \cdot 10^{-8}. \qquad (25)$$

Assuming that $\Delta n = -0.0411$, we obtain from Eq. (25) $n_1' = n_1 + \alpha = 1.5155$, $n_2' = n_1 - \alpha = 1.5035$, $n_3' = 1.4694$. Substituting now in Eq. (24) the values $n_1' = 1.5155$, * $P_S = 4 \times 10^3$ cgs esu (from Fig. 8), we find that $m_{63} = -2.43 \times 10^{-7}$, which gives $m_{63}^* = -2.33 \times 10^{-7}$ cgs esu. The relationship between the value of m_{63}^* and the product $p_{66}g_{36}$ shows that 96% of the electro-optical coefficient of KH_2PO_4 at the phase transition represents the true linear electro-optical effect and 4% represents the false electro-optical effect caused by the piezo-optical effect resulting from the converse piezoelectric effect.
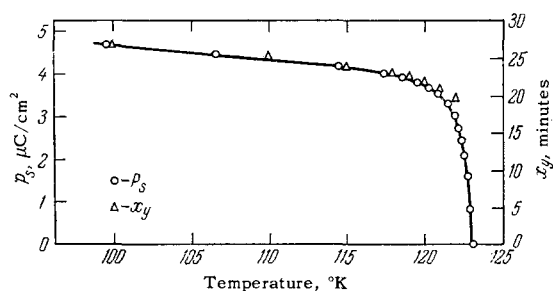

FIG. 8. Temperature dependence of the spontaneous polarization and of the spontaneous shift for KH_2PO_4 (according to Arx and Bantle [58]).

If we assume that the coefficient m_{63} is approximately the same for the ferroelectric and paraelectric modifications, then, using its value given above, we can estimate the electro-optical coefficient r_{63} of KDP.

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*It should be remembered that when the temperature of a crystal is altered, only some of the changes are due to changes in the spontaneous polarization (i.e., not all these changes represent the spontaneous electro-optical effect). When the temperature is varied, the optical properties change also due to the thermal expansion of the crystal (which is known as the "thermo-optical effect" [67,68]).

Assuming that the polarizability of KDP at room temperature is $\alpha = 1.67$ ($\epsilon \approx 22$), we obtain

$$r_{63} = -40.6 \cdot 10^{-8} \text{ cgs esu.}$$

A number of investigations have been published on the measurements of the electro-optical effect "induced" by an external electric field in KDP crystals. The value of the coefficient r_{63} can be determined experimentally using the method described in Sec. 3 or by other methods.

In particular, the value of this coefficient was found by investigating the "longitudinal" (light and field along the z axis) and the "transverse" (field along z axis and light along [110] or $[1\bar{1}0]$) electro-optical effects in KDP. [32] The use of the equation for the optical indicatrix with the field along the z axis (which is denoted by the index 3) of this class of crystal

$$\frac{x'^2}{n_1^2}(1 + r_{63}n_1^2E_3) + \frac{y'^2}{n_1^2}(1 - r_{63}n_1^2E_3) + \frac{z^2}{n_3^2} = 1 \qquad (26)$$

leads (in the "longitudinal" effect case) to the expression

$$n_{x'} - n_{y'} \cong n_1^3 r_{63}E_3, \qquad (27)$$

from which we can calculate the value of r_{63} if we know the value of the double refraction produced by the field and the value of the refractive index n_1 (Fig. 9a). Similar relationships are obtained also for the "transverse" effect (Fig. 9b):

$$n_z - n_{x'} \cong (n_3 - n_1) + \frac{1}{2}n_1^3 r_{63}E_3, \left.\begin{array}{c} \\ \\ \end{array}\right\}$$
$$n_z - n_{y'} \cong (n_3 - n_1) - \frac{1}{2}n_1^3 r_{63}E_3. \left.\begin{array}{c} \\ \\ \end{array}\right\} \qquad (28)$$

The considerable departure from the linear relationship between the relative increment in the double refraction and the field, observed in the transverse effect near the Curie temperature, is due to the quadratic electro-optical effect and is caused by a change in the coefficient n_z. In fact, the coefficient n_z may vary, in the field E_3, only as the square of this field [cf. Eq. (35) below]. At temperatures far from T_C, the variation of n_z is weak and the linear relationship between $\delta(n_{x'} - n_z)/\lambda$ and E_3 is governed mainly by the linear effect, which is the variation of $n_{x'}$ in direct proportion to the field.

The calculated values of r_{63}, obtained from the measurements of the "longitudinal" and "transverse" effects, coincide when plotted as a function of temperature (Fig. 10). It follows from Fig. 10 that the coefficient r_{63} obeys the Curie—Weiss law above T_C and has an anomalously large value at the transition point. The anomalies in r_{63} follow the anomalies in ϵ_C and are the direct consequence of the anomalies in ϵ_C. It then follows directly that the electronic polarizability of KDP crystals varies with temperature in the same way as the total polarizability, which represents the sum effect of all the mechanisms.
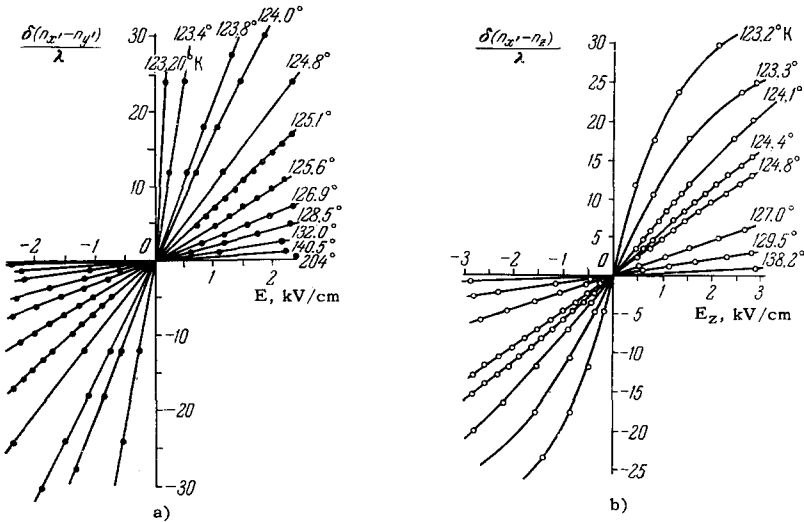
FIG. 9. Dependence of the relative change in the double refraction of a KDP crystal on the field intensity E_z at various temperatures above the Curie point (according to Zwicker and Scherer [32]).

a) $\dfrac{\delta(n_{x'}-n_{y'})}{\lambda} = \dfrac{\delta(n_{110}-n_{\bar{1}10})}{\lambda} = f(E_z)$,

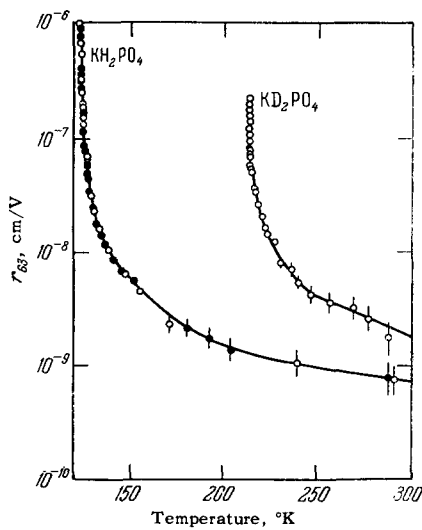b) $\dfrac{\delta(n_{x'}-n_z)}{\lambda} = \dfrac{\delta(n_{110}-n_{001})}{\lambda} = f(E_z)$.

FIG. 10. Temperature dependence of the electro-optical coefficient r_{63} of KH_2PO_4 and KD_2PO_4 crystals above the Curie point (according to Zwicker and Scherer [32]).

The numerous measurements of the coefficient r_{63} of KDP, including the values given in Fig. 10, produce the following room-temperature value of r_{63}:

$$r_{63} \cong -30 \cdot 10^{-8} \text{ cgs esu,}$$

which is in satisfactory agreement with the value of r_{63}, given above, estimated from the coefficient m_{63} at the phase-transition point.

When low-frequency alternating electric fields are applied to a crystal, the "free-crystal" conditions are obtained (the coefficient r is determined). At frequencies higher than the resonance frequency of the sample, the deformation is unable to follow the field and the crystal ceases to be mechanically free. In this case, the crystal exhibits the true electro-optical effect, described by the coefficient r*. Thus, by measuring the electro-optical effect at a low frequency and at a frequency higher than the piezoelectric resonance fre-

quency, we can determine the difference between the total and the true effects. This difference can also be found by calculation, using the following equations [cf. Eqs. (11) and (12)]: *

$$\left.\begin{array}{l} r_{ijk} - r^*_{ijk} = p_{ijlm}\, d_{klm}, \\ r_{ijk} - r^*_{ijk} = \pi_{ijlm}\, e_{klm} \end{array}\right\} \tag{29}$$

(where d and e are the piezoelectric coefficients).

The relationship between the total and true electro-optical effects corresponds to the relationship between the polarizabilities of "free" and "clamped" crystals. In turn, the latter relationship represents the electro-mechanical coupling in a crystal: other conditions being equal, the false electro-optical effect is greater in a crystal which has a larger electromechanical coupling coefficient k.

The permittivity ϵ of KDP (and ADP) crystals is independent of the frequency up to $\approx 10^{10}$ cps. We may expect, therefore, that the electro-optical coefficients of these crystals are also independent of the frequency up to wavelengths in the centimeter range.

The electro-optical coefficients, in accordance with their definition, should depend on the wavelength of light to the same extent as the refractive indices (polarization constants). Thus, in general, the electro-optical coefficients should exhibit dispersion. Figure 11 shows the dispersion of the coefficients r_{63} of KDP and ADP crystals.

All that has just been said about the electro-optical effect in ferroelectrics applies to single-domain crystals. In the case of KDP (and other) crystals, the presence of domains complicates the electro-optical phenomena. Thus, in KDP, the appearance of the spontaneous polarization along the z axis produces domains of opposite orientation; the elliptical cross sections of the optical indicatrices of oppositely oriented domains

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*Similarly, we can find the difference between the total and true electro-optical effects using the coefficients m_{ijk}.
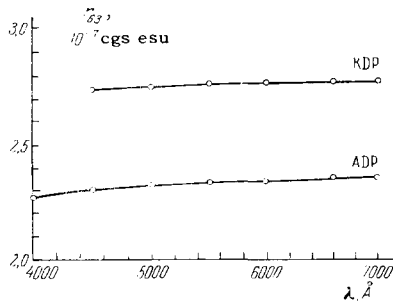
FIG. 11. Dispersion of the electro-optical coefficient r_{63} of ADP and KDP crystals (according to Blokh [59]).

in a (001) plane of a crystal are rotated by 90° with respect to one another. The application of an electric field along the z axis orients all the domains along the same direction; the re-orientation of the domains alters the sign of the electro-optical coefficients in the sense that the formerly long axis of the optical indicatrix becomes its short axis: assuming one of these axes to be x' and an axis perpendicular to it to be y', we shall then have a change in the sign of the difference $(n_{x'} - n_{y'})$. The dependence of the double refraction on the field in the ferroelectric region has the form of hysteresis loops, similar to the dielectric hysteresis loops.

The values of the electro-optical coefficient r_{41} of KDP and the values of the coefficients r_{ij} of several other isomorphous ferroelectrics are listed in Table IV. [12,32-36]

Rochelle salt (like KDP) is an interesting crystal from the point of view of the electro-optical effect.

Above its upper Curie point (+24° C), Rochelle salt is orthorhombic and optically biaxial; its refractive indices are: $n_1 = n_a = 1.4954$, $n_2 = n_b = 1.4920$, $n_3 = n_c = 1.4900$. The optical axes are in the (010) plane and the bisectrix of the acute angle between the optical axes (which is 69° 40') is the a axis. The appearance of the spontaneous polarization at the Curie point makes Rochelle salt monoclinic (dihedral axial class

C_2). In accordance with the symmetry of the paraelectric modification (class V in Table I), the optical indicatrix of Rochelle salt (or, more exactly, the indicatrix of a domain) is rotated by an angle α about the a axis due to the spontaneous linear electro-optical effect. The relationship between the angle α and the value of the spontaneous polarization, expressed in terms of the electro-optical coefficient, can be found from the indicatrix equation

$$\frac{x^2}{n_1^2} + \frac{y^2}{n_2^2} + \frac{z^2}{n_3^2} + 2m_{41}P_s r_{23} = 1, \qquad (30)$$

from which it follows that

$$\tan 2\alpha = \frac{2m_{41}P_s}{\frac{1}{n_2^2} - \frac{1}{n_3^2}}, \qquad (31)$$

or, bearing in mind the smallness of the angle, we can use the relationship

$$\alpha = \frac{m_{41}P_s}{\frac{1}{n_2^2} - \frac{1}{n_3^2}}. \qquad (32)$$

Assuming that, near 0° C, $\alpha \approx 0.8$ (Fig. 12), $P_s = 7.5 \times 10^2$ cgs esu (Fig. 13), and that the refractive indices n_2 and n_3 vary slightly in the investigated range of temperatures (see below), we obtain $m_{41} = 0.224 \times 10^{-7}$ cgs esu, which is almost one order of magnitude smaller than the analogous coefficient m_{63} of KH_2PO_4.

The value of m_{41} is determined here using the experimental value of the angle of rotation of the indicatrix. This angle is due to the true linear effect and to the piezo-optical effect, caused by the converse piezoelectric effect [shear of a crystal in a (100) plane]. The relationship between the true and false effects may be found by comparing the value of $m_{41} = 0.224 \times 10^{-7}$ cgs esu with the value of the product $p_{44}g_{14}$ [cf. Eq. (12)]. Assuming that $p_{44} = 0.009$ and $g_{14} = 6.7 \times 10^{-7}$ cgs esu, we obtain $p_{44}g_{14} = 0.6 \times 10^{-8}$ cgs esu, from which it follows that the false effect in Rochelle salt represents about 27% of the true effect.

Assuming that the permittivity of Rochelle salt at

Table IV. Electro-optical coefficients of some ferroelectrics

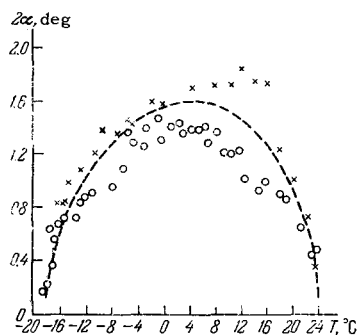| Crystal	Symmetry	r_{ij} $\times 10^8$ cgs esu	m_{ij} $\times 10^8$ cgs esu	Voltages necessary to produce path difference $_0\lambda/2$ (λ h 5461 Å), kV
1. KH_2PO_4 (KDP)	V_d	$r_{63} = -30$ $r_{41} = 26$	$m_{63} = -24.3$	~8
2. KD_2PO_4	V_d	$r_{63} = -70$		3.4
3. KH_2AsO_4	V_d	$r_{63} = -39$		6.2
4. RbH_2PO_4	V_d	$r_{63} = -33$		7.3
5. $KNaC_4H_4O_6 \cdot 4H_2O$ (Rochelle salt)	V	$r_{41} = -6$ $r_{52} = -5.1$ $r_{63} = 0.95$	$m_{41} = 2.24$	
6. $BaTiO_3$ (tetragonal modification)	C_{4v}		$m_{33} = 18.7$	
7. $NH_4H_2PO_4$ (ADP)	V_d	$r_{63} = -25$ $r_{41} = 62$		~10
8. $NH_4H_2AsO_4$	V_d	$r_{63} = -19$		13

FIG. 12. Temperature dependence of the angle between the extinction positions of neighboring domains in an a-cut plate of Rochelle salt crystal (according to Indenbom and Chernysheva [60]).
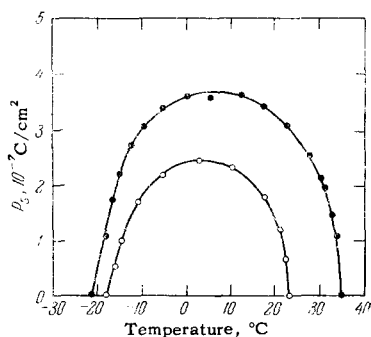

FIG. 13. Temperature dependence of the spontaneous polarization of Rochelle salt (according to Hablutzel [61]). 1) Ordinary Rochelle salt; 2) deuterated salt.

$t = 0°$ C is $\epsilon_a \approx 130$, we can estimate the value of the coefficient r_{41} from the coefficient m_{41}:*

$$r_{41} = 22.4 \cdot 10^{-8} \text{ cgs esu.}$$

This value is over three times as large as the experimental value [1 37-40] (cf. Table IV), determined at room temperature. In this connection, we may mention that the estimates of the coefficients m_{41} and r_{41} given above are only rough and that the experimental values of the electro-optical coefficients differ widely from sample to sample.

In the paraelectric modification (above $-124°$ C), $NH_4H_2PO_4$ (ADP) exhibits electrical properties similar to those of KH_2PO_4 (KDP). A comparison of the properties of these two crystals is given in Table IV.

As the temperature of the antiferroelectric phase transition is approached, the electro-optical coefficient r_{63} of a free ADP crystal increases in accordance with the Curie-Weiss law. In spite of the fact that this temperature is relatively low, r_{63} has a very high value. According to some investigators, the false ("piezoelectric") electro-optical effect in ADP is quite large and represents more than one third of the total effect (at $20°$ C).[10] The remarkably high value of the coeffi-

$$*r_{41} = \frac{\epsilon_a}{4\pi} m_{41}.$$

cient r_{41} (at room temperature) indicates a strong electronic polarizability of these crystals.

The appearance of the spontaneous antipolarization in $NH_4H_2PO_4$ makes this crystal (or its domains) optically biaxial. In accordance with the linear electro-optical tensor for crystals of the V_d class (Table I), the spontaneous antipolarization is accompanied by the spontaneous linear electro-optical effect.

6. QUADRATIC ELECTRO-OPTICAL EFFECT

The quadratic electro-optical effect of centrosymmetric crystals is of great interest since the linear electro-optical effect is absent in such crystals and the quadratic effect exists in its "pure" form unmasked by the linear effect (which is stronger than the quadratic).

The electro-optical effect is relatively strong in some ferroelectric crystals in the paraelectric centrosymmetric region near the Curie temperature. This applies primarily to ferroelectrics with the oxygen-octahedral structure. [46]

The refractive index $n_C = n_3$ of barium titanate changes suddenly near the Curie point ($120°$ C). This index, which is approximately 2.42 in the cubic modification, suddenly assumes a value of about 2.38 after the transition. At room temperature, $n_C \approx 2.35$ and, therefore, the tetragonal modification of $BaTiO_3$ is optically negative ($\Delta n = n_C - n_a < 0$). The refractive indices $n_1 = n_a = n_2 = n_b$ of the cubic and tetragonal modifications are practically independent of temperature. This is illustrated clearly by the temperature dependence of the double refraction of $BaTiO_3$ crystals given in Fig. 6.

The change in the optical properties when the spontaneous polarization appears may be called the spontaneous electro-optical effect. Since $BaTiO_3$ is not piezoelectric in the paraelectric modification, the electro-optical effect at the Curie point is not quadratic. In accordance with Eq. (15), the increment arising in the polarization coefficient Δa_3 when the polarization appears along the c axis (which is denoted by the index 3), will be written in the form

$$\Delta a_3 = (M^*_{33} + p_{3m}Q_{m3}) P_s^2. \tag{33}$$

Using the relationship

$$\Delta a_3 = \frac{1}{n_3'^2} - \frac{1}{n_3^2}$$

and assuming that at the Curie point $n_3' = 2.38$, $n_3 = 2.42$ ($\Delta n = -0.04$), $P_3 = 54 \times 10^3$ cgs esu, we find that the quantity in parentheses in Eq. (33) is equal to 1.88×10^{-12} cgs esu. The first term in the parentheses, i.e., the quantity M^*, describes the true quadratic electro-optical effect, and the second term gives the correction to this effect due to the electrostriction of the crystal. Using the values [47] $Q_{33}(= Q_{11} = Q_{22}) = 1.17 \times 10^{-12}$ cgs esu, $Q_{12} = Q_{13} = -0.53 \times 10^{-12}$ cgs esu and assuming that the piezo-optical coefficients

P_{3m} are (as in the majority of ionic crystals) equal to $\approx 10^{-1}$, we can estimate the second term in the parentheses to be $\approx 10^{-14}$ cgs esu. This allows us to deduce that $M_{33}^* = 1.87 \times 10^{-12}$ cgs esu. This value may be used also to describe the quadratic electro-optical effect of $BaTiO_3$ in the paraelectric modification above the Curie temperature ($M_{33} \approx M_{33}^*$).

Thus, at the Curie point, barium titanate exhibits the quadratic electro-optical effect, 99% of which is the true effect and 1% is the false effect. The coefficient M_{33}, which is related to the polarization, is practically independent of temperature and approximately the same in the paraelectric and ferroelectric regions. However, the coefficient L, which is related to the field intensity E, increases in accordance with the Curie—Weiss law when ϵ increases as the Curie point is approached. The published experimental value of the difference $M_{33} - M_{12}$ ($M_{33} = M_{11} = M_{22}$) for $BaTiO_2$ is listed in Table V. [48]

High values of the quadratic electro-optical coefficients M_{pq} have been found for the paraelectric modification of $KTaO_3$ ($T_C \approx 1°$ K) in the temperature range $2-77°$ K, for $SrTiO_3$ in the temperature range $4-300°$ K, and for $KTa_{0.65}Nb_{0.35}O_3$ (KTN) in the temperature range $285-310°$ K ($T_C \approx 283°$K). The information on these coefficients is also given in Table V. The electro-optical properties have also been found in $LiNbO_3$ crystals. [49]

The deformation of the optical indicatrix along the direction of the spontaneous polarization in KDP is solely due to the quadratic electro-optical effect (the electro-optical coefficient m_{33} of the V_d class crystals is equal to zero; cf. Table I). We shall estimate the value of the spontaneous quadratic electro-optical coefficient for the same direction using Eq. (15)

$$\Delta a_3 = (M_{33}^* + P_{3m}Q_{m3})P_s^2, \qquad (34)$$

which is written in terms of the refractive indices as follows

$$\frac{1}{n_3'^2} - \frac{1}{n_3^2} = (M_{33}^* + P_{3m}Q_{m3})P_s^2. \qquad (35)$$

Table V. Coefficients of the quadratic electro-optical effect for some ferroelectrics

Crystal	Symmetry	Refractive index	$M \times 10^{12}$ cgs esu
1. $BaTiO_3$	O_h	2,42	$M_{33} = 1.88$
			$M_{11} - M_{12} = 1.44$
2. $SrTiO_3$	O_h	2.38	$M_{11} - M_{12} = 1.55$
3. $KTaO_3$	O_h	2.24	$M_{11} - M_{12} = 1,77$
			$M_{44} = 1.33$
4. $KTa_{0,65}Nb_{0,35}O_3$	O_h	2.29	$M_{11} - M_{12} = 1.93$
			$M_{11} = 1.5$
			$M_{12} = 0,43$
			$M_{44} = 1.83$
5. KH_2PO_4	V_d		$M_{33} = -2.86$
6. $KNaC_4H_4O_6 \cdot 4H_2O$ (Rochelle salt)	V		$M_{11} = -1500$

Assuming, in accordance with the data given above, that $P_S = 14 \times 10^3$ cgs esu (T = 100° K), $n_3' = 1.4694$, $n_3 = 1.4684$, p = -2.0×10^{-2}, Q = 4.3×10^{-12} cgs esu, we can estimate the coefficient M_{33} for KDP:

$$M_{33} = -2.86 \cdot 10^{-12} \text{ cgs esu.}$$

97% of this value represents the true effect and 3% represents the false effect.

A comparison of the coefficients M_{33} for KDP and $BaTiO_3$ shows that in the case of KDP the value of this coefficient is about one and a half times greater than that for $BaTiO_3$. We should also mention the fact that $BaTiO_3$ has an exceptionally high refractive index. The spontaneous polarization of this crystal obviously brings its electronic polarizability to a state close to saturation. This explains precisely why the optical dielectric polarizability of the tetragonal $BaTiO_3$ is lower along the spontaneous polarization direction than at right angles to it. The refractive indices of KH_2PO_4 are small. The spontaneous polarization in that crystal does not saturate the electronic polarizability (at least along the c axis direction) which is responsible for the higher (compared with $BaTiO_3$) value of the coefficient of M_{33}^* of KH_2PO_4.

As in the case of KH_2PO_4, the electro-optical effect along the spontaneous polarization direction in Rochelle salt is quadratic. The value of the coefficient M_{11} for this effect is found from Eq. (15)

$$\Delta a_1 = (M_{11}^* + P_{1m}Q_{m1})P_s^2 \qquad (m = 1, 2 \dots 6) \qquad (36)$$

or, which is equivalent, from the following equation

$$\frac{1}{n_1'^2} - \frac{1}{n_1^2} = (M_{11}^* + P_{1m}Q_{m1})P_s^2.$$

It is known that, in the range of temperatures considered (0—30° C), the coefficient n_1 of Rochelle salt varies linearly as the temperature rises and that the change in this coefficient in the stated range of temperatures amounts to about 0.0014. This allows us to estimate the value of the expression in parentheses in Eq. (36), which is -1.5×10^{-9} cgs esu. Since the product $P_{1m}Q_{m1}$ is of the order of $\approx 10^{-13}$, it follows that the former value can be assumed to be equal to the true quadratic electro-optical coefficient M_{11}^* = 1.5×10^{-9} cgs esu. It is very important that this coefficient is three orders of magnitude greater than the corresponding coefficients of $BaTiO_3$ and KH_2PO_4. It indicates the exceptionally "unsaturated" nature of the electronic polarizability of Rochelle salt crystals along the ferroelectric axis.

The "induced" (by an external field) electro-optical effect in Rochelle salt has been investigated in detail. The results of the investigation of this effect, represented by an increment in the relative double refraction $\delta(n_x - n_y)/\lambda$, are given in Fig. 14a for Rochelle salt above its upper Curie point (symmetry V, here the linear effect, representing this increment, is absent because $r_{11} = r_{21} = 0$).
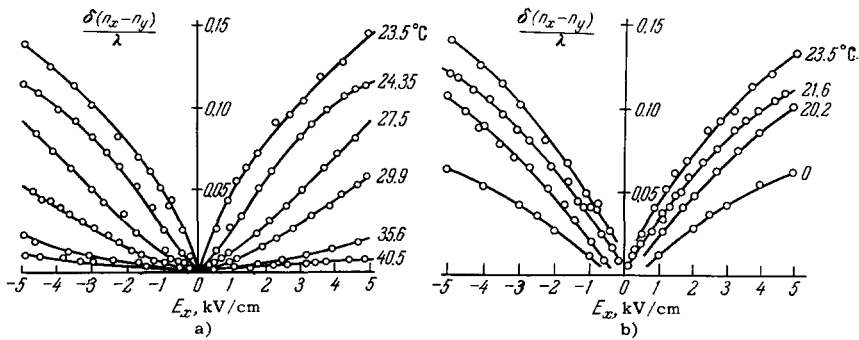
FIG. 14. Relative change in the double refraction of Rochelle salt as a function of the field intensity E_x (according to Müller [37]). a) $\delta(n_x - n_y)/\lambda = f(E_x)$ at various temperatures above the upper Curie point; b) $\delta(n_x - n_y)/\lambda = f(E_x)$ at various temperatures in the ferroelectric region.

Figure 14b shows an increment in the relative difference of the refractive indices n_x and n_y of Rochelle salt under the action of a field E_x in the ferroelectric region, below the upper Curie point. It is evident from Fig. 14b that this difference between the indices is a quadratic function of the field. This quadratic dependence can be understood as follows. Below its Curie point, Rochelle salt exhibits the linear effect (domains of the C_2 symmetry have non-zero electro-optical coefficients r_{11} and r_{21}), as well as the quadratic effect. Due to the presence of the quadratic effect, the relationship between the quantity $\delta(n_x - n_y)/\lambda$ and the field departs from linearity. The contribution of the quadratic effect, in accordance with its definition, should not change sign when the field is reversed. The contribution of the linear effect behaves as if it were the quadratic-effect contribution because, when the external field direction is altered, the domains are reoriented but the nature of the distortion in the optical indicatrix by the field remains the same. This feature of the linear effect, associated with the domain re-orientation, is similar to the behavior of the piezoelectric effect in a polydomain crystal which makes the piezoelectric effect appear as if it were the electrostriction. [2]

A comparison of the changes in the refractive indices with the value of the spontaneous polarization of ammonium sulfate $(NH_4)_2SO_4$ [50] gives a large value (as in the case of Rochelle salt) of the coefficient M'_{33} = 1.9×10^{-9} cgs esu.

7. PRACTICAL APPLICATIONS OF THE ELECTRO-OPTICAL PROPERTIES OF CRYSTALS

Electro-optical devices are finding an increasing range of applications. Thus, for example, light modulators are used in optical range-finders, optical communication, laser radiation modulation, sound recording on cine film, image reproduction (color television, phototelegraphy, electron microscopy), and automatic polarimeters.

Electro-optical shutters are used in photography and in high-speed cinematography, as well as in phototelegraphy. Electro-optical transducers are employed in tunable narrow-band interference-polarization filters, in devices used to measure high voltages,

and in optical elements of computers. We shall consider briefly some of these applications.

a) High-Frequency Modulation of Light

With the recent development of quantum electronics, there has been an increased interest in devices capable of the microwave modulation of light. KDP, ADP, quartz and $BaTiO_3$ (cubic modification) electro-optical crystals are used at present in the high-frequency modulation of light. Measurements show that ϵ of ADP crystals does not change up to 3.6×10^{10} cps. [51] We may expect, therefore, that ADP crystals (and probably KDP) can be used to modulate light up to these frequencies and above.

So far, modulation has been achieved at frequencies of the order of 10^9 cps. [11,52] At these frequencies, the electro-optical elements work under the piezoelectric "clamped" conditions (above the resonance frequency). In this case, the modulation is solely due to the true effect and, therefore, the 100% modulation requires voltages higher than those needed at frequencies below resonance. Thus, at λ = 550 mμ, a voltage of 14.8 kV must be applied to an ADP crystal and 8.5 kV to a KDP crystal, while at low frequencies 9.3 and 7.7 kV, respectively, are needed.

Quartz modulators work effectively under resonance conditions. Some quartz devices give 90% modulation in a field of 100 V using 1.8 W. Quartz modulators are used mostly in optical range-finders. [41-45]

Cubic crystals (CuCl, ZnS and others), which show promise as light modulators, allow the use of wide-angle light beams. $BaTiO_3$ single crystals can be used as effective electro-optical shutters, using the quadratic electro-optical effect above the Curie temperature (+120°C). At 125°C, a path difference of $\lambda/2$ (maximum transparency) may be obtained by applying only 150 V to a $BaTiO_3$ crystal. [46]

b) Recording of Sound [53,54]

One of the currently used methods for the recording of sound on motion-picture film employs a Kerr cell filled with nitrobenzene. However, the use of Kerr cells presents a number of difficulties, which can be avoided if crystalline modulators are employed.
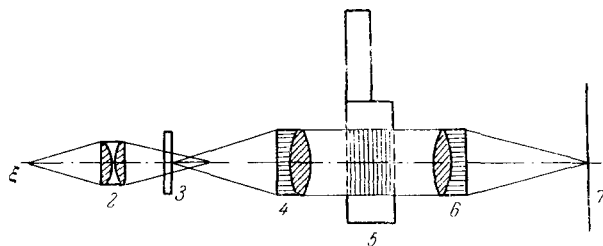
Figure 15 shows the optical system of a device

FIG. 15. Schematic representation of an electro-optical modulator for light. 1) Lamp filament; 2) condenser lens; 3) slit; 4) collimator; 5) cell consisting of a $\lambda/4$ mica plate and an ADP crystal, placed between crossed film polaroids; 6) focusing lens; 7) cine film.
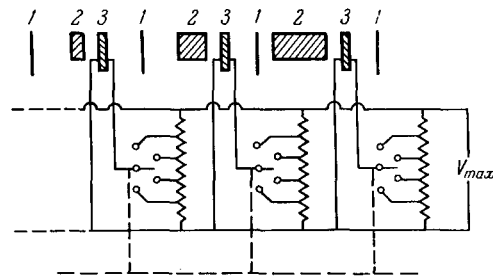

FIG. 16. Schematic representation of an electrically tuned interference-polarization filter used to shift the pass band to one of four discrete wavelengths. 1) Polaroids; 2) quartz plates; 3) ADP plates.

used to record sound on film. A monochromatic parallel beam passes through an ADP cell and is focused on the film. When an electric field is applied to the crystal, the slit shielding the central part of the conoscopic pattern becomes transparent. By applying an audio-frequency voltage to the crystal, we can record sound on film.

The same principle is used to measure high voltages. In this case, the value of the voltage applied to a crystal is determined from the value of the angle of compensation, i.e., the angle of rotation of an analyzer needed to compensate the transparency of the crystal induced by the field.

c) Narrow-Band Interference-Polarization Light Filters [55]

The action of interference-polarization light filters is based on the interference of polarized light when it passes through an anisotropic crystal. The light, linearly polarized at an angle of 45° to the principal axes of the optical indicatrix, is split into two beams which emerge from the crystal having a certain path difference. If the light is now passed through an analyzer, which is oriented in parallel with a polarizer, the two beams will interfere. If white light is passed through such a system, the spectrum of the emergent light will consist of correctly alternating bright and dark bands.

The interval between the nearest bright bands is inversely proportional to the thickness of the crystal plate. If a system consists of a number of plates whose thicknesses are in the ratio 1:2:4, etc., the spectra produced by each plate will be superimposed and the result will be a spectrum consisting of a series of widely separated bands, each of which can be selected by an ordinary filter.

The separation between the bands will be equal to the distance between the transmission maxima of the the thinnest crystal; the width of the bands will be equal to half the width of the transmission maximum of the thinnest element.

A tunable filter is also based on the variation of the plate thickness in the ratio 1:2:4, i.e., path differences

are varied in accordance with this ratio. In tunable filters, this is achieved by regulating the voltage across electro-optical crystals.

Figure 16 shows a tunable filter, consisting of quartz plates whose thicknesses are in the ratio 1:2:4, giving fixed transmission bands. The introduction of ADP plates into the system allows us to shift and change the pass band of the filter by applying an electric field to these plates. The pass band width may be varied from a fraction of an angstrom to several hundreds of angstroms. Moreover, the pass band may be shifted into any desired region. Tunable light filters can obviously be used for the frequency modulation of light.

Additional literature on the applications of electro-optical devices is given in the reviews [4,7].

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