## ENERGY SPECTRUM OF CASCADE ELECTRONS IN LIGHT MATERIALS 107

The results of the calculations of these functions are shown in Tables I and II.\* An investigation of the computed functions (cf. for example, Refs. 2 and 3, in which this type of analysis was first applied) shows that in the present case the error in the electron energy spectrum is approximately 10 percent. We may note that the method of moments is so far the only method which makes it possible to determine with reasonable accuracy the average behavior of an electron-photon shower in the energy region being considered. In calculating the moments, the photon absorption coefficient  $\sigma$  has been assumed constant and equal to its asymptotic value  $\sigma = \sigma_0$ . However, in computing the distribution functions the photon absorption coefficient has been assumed equal to  $\sigma_{\min} = 0.65$ . As follows from the results of Ref. 1, this method makes it possible to take account of the approximate dependence of  $\sigma(E)$  on energy, leading to better agreement of theory and experiment. The value 0.65 corresponds to the value of  $\sigma_{\min}$  in air; the computed curves, however, apply to the development of showers in materials with atomic number Z approximately up to 30 with good accuracy.<sup>4</sup> The numerical results which have been presented may be useful in the analysis of certain cosmic-ray experiments.

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<sup>3</sup>I. P. Ivanenko, J. Exptl. Theoret. Phys. (U.S.S.R.) 32, 491 (1957), Soviet Phys. JETP 5, 413 (1957). <sup>4</sup>P. S. Isaev, J. Exptl. Theoret. Phys. (U.S.S.R.) 28, 374 (1955), Soviet Phys. JETP 1, 379 (1955).

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# CONTRIBUTION TO THE THEORY OF SCATTERING OF LIGHT NEAR POINTS OF SECOND-ORDER PHASE TRANSITIONS

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The scattering of light by ferroelectric and ferromagnetic crystals in the vicinity of secondorder transition points is considered. Calculations are performed both for single component crystals and for solid solutions. By way of illustration light scattering by  $BaTiO_3$  type crystals as well as by Rochelle salt or  $KH_2PO_4$  type crystals is examined. The effect of an external field on the scattering is investigated.

I. Ginzburg<sup>1</sup> has shown that at temperatures close to the temperature of a second-order transition there occurs additional scattering of light which is particularly intense near the critical point, that is, near the point where the curve for second-order transitions merges into the curve for transitions of the first order. This effect has been experimentally detected by Iakovlev, Mikheeva, and Velichkina<sup>2</sup> who investigated the dispersion of light in quartz near the point of the  $\alpha \neq \beta$ -transformation. Herein we shall carry out a further theoretical investigation of the problem, primarily where ferroelectric materials are con-

<sup>\*</sup> In the first line of each column of the tables the first number denotes the energy of the initial electron or photon and the second the energy of the secondary electron.

cerned. We shall explicitly take into account the anisotropy of the fluctuation of the dielectric constant  $\epsilon^0$  (for light), which obtains even in cubic (or almost cubic) crystals, and the depolarization of light associated therewith. We shall investigate the influence of the electric field on the intensity of scattering. In addition to the case considered by Ginzburg, when there are only square terms in the expansion of  $\delta\epsilon^0$  in powers of the components of the polarization vector, we shall also examine the case which obtains in certain ferroelectrics (for example, in Rochelle salt), when this expansion also comprises linear terms. We shall also consider the scattering of light by solid solutions of ferroelectrics.

2. The additional scattering of light near the second-order transition points is associated with the inhomogeneities of the dielectric constant  $\epsilon^0$ , caused by the fluctuation of the internal parameters that characterize the long range order in the less symmetric phase (in the case of ferroelectrics, such internal parameters are the components of the polarization vector). Near second-order transition points inhomogeneities of this type can always be regarded as static. For anisotropic crystals the question of scattering of light by inhomogeneities of the dielectric constant, caused by thermal vibrations of the lattice, has been examined by Motulevich.<sup>3</sup> Carrying out exactly the same examination for light scattering by the static distribution of the inhomogeneities of the dielectric constant tensor, we find that the intensity of the scattered light is

$$\frac{J_1}{J_0} = K_1 K_2 \frac{\omega^4 V^2}{16\pi^2 c^4 R^2} \frac{n_2}{n_1} \cos^{-2} \delta_1 \cdot \cos^{-2} \delta_2 \left| \sum_{\alpha,\beta=1}^3 \varepsilon_{\alpha\beta\gamma} e_{\alpha\beta\gamma}^1 e_{\alpha\beta\gamma}^2 e_{\beta\gamma}^2 \right|^2.$$
(1)

Here  $J_1$  is the intensity of the scattered wave,  $J_0$  is the intensity of the wave incident on the crystal, V is the volume of the crystal, c is the speed of light, R is the distance from the point of observation to the scattering crystal (the size of which is assumed to be appreciably smaller than R),  $k_1$  and  $k_2$  are the wave vectors of the incident and scattered waves (in the crystal),  $e^1$  and  $e^2$  are the unit polarization vectors of these waves (having the components  $e^1_{\alpha}$  and  $e^2_{\beta}$ , where the indices  $\alpha$  and  $\beta$  designate the respective Cartesian coordinates),  $n_1$  and  $n_2$  are respectively the indices of refraction for the incident and scattered waves (in the crystal), respectively. The factors  $K_1$  and  $K_2$  take into account the decrease in the intensity of the incident and scattered rays in passing through the crystal-air interface and can be calculated by means of the familiar formulas of crystal optics.\* Lastly,  $\epsilon_{\alpha\beta q}$  denotes the q-th Fourier component of the deviation of the  $\delta\epsilon^0_{\alpha\beta}$  component of the dielectric-constant tensor from the mean value

$$\varepsilon_{\alpha\beta q} = \frac{1}{V} \int_{V} \delta \varepsilon_{\alpha\beta}^{0} e^{iqr} d\tau, \qquad (2)$$

where  $q = k_2 - k_1$ . In deducing expression (1) it was assumed that rotation of the plane of polarization in the crystal is absent or can be neglected. It was further assumed that light absorption in the specimen is negligible.

In the case of cubic crystals, in which the index of refraction is independent of the direction of the wave vector and of the polarization vector and the angles  $\delta_1$  and  $\delta_2$  are zero, the expression for the intensity of the scattered radiation is simplified to

$$\frac{J_1}{J_0} = K_1 K_2 \frac{\omega^4 V^2}{16\pi^2 c^4 R^2} \left| \sum_{\alpha, \beta=1}^3 \varepsilon_{\alpha\beta q} e_{\alpha}^1 e_{\beta}^2 \right|^2.$$
(3)

The change in the dielectric constant connected with the fluctuation of polarization in this case is also a tensor (not a scalar) quantity.

The further examination will be carried out in terms pertaining specifically to ferroelectric crystals. In the vicinity of the temperature  $T_0$  of the transition to the ferroelectric state we can retain only the first terms of the expansion of the components of the tensor  $\epsilon^0_{\alpha\beta}$  in powers of the components of the polarization vector:

$$\varepsilon^{\mathbf{0}}_{\alpha\beta} = \varepsilon^{\mathbf{0}}_{\alpha\beta0} + \lambda_{\alpha\beta,\gamma} P_{\gamma} + \lambda_{\alpha\beta,\gamma\delta} P_{\gamma} P_{\delta}, \tag{4}$$

<sup>\*</sup> Repeated reflections from the other faces of the crystal, which greatly complicate the picture, can be eliminated by blackening these faces.

where the coefficients do not depend on the polarization. Here and below summation from 1 to 3 over all twice-repeating indices (in a given term) is implied. The polarization vector **P** can be regarded as consisting of a mean value  $\overline{\mathbf{P}}$  and a fluctuating part  $\Delta \mathbf{P}$ . Replacing **P** in (4) by the sum  $\mathbf{P} + \Delta \mathbf{P}$ , we determine  $\delta \epsilon_{\alpha\beta}^0$ , retaining only the terms linear in  $\Delta \mathbf{P}_{\gamma}$ , and then, invoking (2), express the Fourier components of the dielectric constant tensor in terms of the Fourier components of the polarization vector  $\mathbf{P}_{\gamma \mathbf{q}}$ 

$$\varepsilon_{\alpha\beta\mathbf{q}} = \lambda_{\alpha\beta,\gamma} P_{\gamma\mathbf{q}} + 2\lambda_{\alpha\beta,\gamma\delta} \bar{P_{\delta}} P_{\gamma\mathbf{q}}.$$
(5)

Substituting (5) in (1), we obtain the following formula for the intensity of the scattered light, connected with the fluctuations of the polarization:

$$\frac{J_1}{J_0} = \frac{1}{2} F(1, 2) V \left[ \lambda_{\alpha\beta,\gamma} \lambda_{\alpha'\beta',\gamma'} + 4 \lambda_{\alpha\beta,\gamma} \lambda_{\alpha'\beta',\gamma'\delta'} \overline{P}_{\delta'} + 4 \lambda_{\alpha\beta,\gamma\delta} \lambda_{\alpha'\beta',\gamma'\delta'} \overline{P}_{\delta} \overline{P}_{\delta'} \right] e_{\alpha}^1 e_{\alpha'}^1 e_{\beta}^2 e_{\beta'}^2 \left( \overline{P_{\gamma q} P_{\gamma' q}^*} + \overline{P_{\gamma q}^* P_{\gamma' q}} \right), \tag{6}$$

where the function

$$F(1, 2) = K_1 K_2 \frac{\omega^4 V}{16\pi^2 c^4 R^2} \frac{n_2}{n_1} \cos^{-2} \delta_1 \cdot \cos^{-2} \delta_2$$
(7)

may be regarded as independent of the temperature.

The mean values of the squares and products of the Fourier terms constituting the fluctuation of the components of the polarization vector can be determined by means of the thermodynamic theory of fluctuations. The distribution of fluctuation probabilities, as is known, is given by the expression

$$w \sim \exp\left(-\Delta \Phi / kT\right),$$
 (8)

where  $\Delta \Phi$  is the change in the thermodynamic potential incident to the appearance of fluctuation. In the case when the polarization of the crystal undergoes fluctuational changes,  $\Delta \Phi$  can be written in the form

$$\Delta \Phi = \frac{1}{2} \int_{V} \left[ \varphi_{\alpha\beta} \Delta P_{\alpha} \Delta P_{\beta} + A_{\alpha\beta\gamma\delta} \frac{\partial P_{\alpha}}{\partial x_{\gamma}} \frac{\partial P_{\beta}}{\partial x_{\delta}} \right] d\tau.$$
(9)

Here  $\varphi$  is the thermodynamic potential per unit volume,\*  $A_{\alpha\beta\gamma\delta}$  is a fourth rank tensor and  $x_{\gamma}$  and  $x_{\delta}$  are Cartesian coordinates ( $\gamma$  and  $\delta = 1, 2, 3$ ); the last term takes into account the changes in the thermodynamic potential incident to the appearance of inhomogeneities in the fluctuation distribution; terms with higher order derivatives can be disregarded, provided (as in the case of scattering of light) the predominant part is played by sufficiently smooth fluctuations.

The thermodynamic potential per unit volume of a ferroelectric in the presence of an external electric field E can be written as  $\varphi = f(\mathbf{P}) - \mathbf{EP}$ , where  $f(\mathbf{P})$  is independent of E. Hence differentiating the equilibrium conditions  $\varphi_{Q} = 0$  with respect to  $\mathbf{P}_{\beta}$ , we obtain

$$\varphi_{\alpha\beta} \equiv \partial^2 f / \partial P_{\alpha} \partial P_{\beta} = \partial E_{\alpha} / \partial P_{\beta}.$$
(10)

Comparing Eq. (10) with the equation for the dielectric susceptibility tensor  $\kappa_{\alpha\beta}: \mathbf{P}_{\alpha} = \kappa_{\alpha\beta}\mathbf{E}_{\beta}$ , we find that the derivatives  $\varphi_{\alpha\beta}$  are components of a tensor that is the reciprocal of the dielectric susceptibility tensor:

$$\varphi_{\alpha\beta} = \varkappa_{\alpha\beta}^{-1}. \tag{11}$$

Replacing the fluctuations  $\Delta P_{\alpha}$  by their Fourier expansions

$$\Delta P_{\alpha} = \sum_{\mathbf{x}}' \left( P_{\alpha \mathbf{x}} e^{i \mathbf{x} \mathbf{r}} + P_{\alpha \mathbf{x}}^{\bullet} e^{-i \mathbf{x} \mathbf{r}} \right), \tag{12}$$

from Eqs. (8), (9), and (11), we obtain

$$\omega \sim \exp\left\{-\frac{V}{2kT}\sum_{\mathbf{x}}'\left(\mathbf{x}_{\alpha\beta}^{-1} + A_{\alpha\beta\gamma\delta}\,\mathbf{x}_{\gamma}\,\mathbf{x}_{\delta}\right)\left(P_{\alpha\mathbf{x}}\,P_{\beta\mathbf{x}}^{*} + P_{\alpha\mathbf{x}}^{\bullet}\,P_{\beta\mathbf{x}}\right)\right\}.$$
(13)

<sup>\*</sup> For simplicity of notation we shall designate the partial derivatives with respect to  $\varphi$  through indices:  $\varphi_{\alpha} \equiv \partial \varphi / \partial P_{\alpha}$ ,  $\varphi_{c} \equiv \partial \varphi / \partial c$ , etc.

By means of the distribution of probabilities (13) we can express the mean value of the products of the Fourier components corresponding to any value of  $\kappa$  in terms of the elements of the matrix that is the reciprocal of the  $\kappa_{\alpha\beta}^{-1} + A_{\alpha\beta\gamma\delta}\kappa_{\gamma}\kappa_{\delta}$ . Substituting the values so obtained for the case of  $\kappa = q$  in place of  $\overline{P_{\gamma q} P_{\gamma' q}^*} + \overline{P_{\gamma q}^* P_{\gamma' q}}$  in Eq. (6), we obtain

$$\frac{J_1}{J_0} = F(1,2)kT \left[\lambda_{\alpha\beta,\gamma}\lambda_{\alpha'\beta',\gamma'} + 4\lambda_{\alpha\beta,\gamma}\lambda_{\alpha'\beta',\gamma'\delta'}\overline{P_{\delta'}} + 4\lambda_{\alpha\beta,\gamma\delta}\lambda_{\alpha'\beta',\gamma'\delta'}\overline{P_{\delta}}\overline{P_{\delta'}}\right] e_{\alpha}^1 e_{\alpha'}^1 e_{\beta}^2 e_{\beta'}^2 (\mathbf{x}_{\gamma\gamma'}^{-1} + A_{\gamma\gamma'\nu\nu'}q_{\nu}q_{\nu'})^{-1}.$$
(14)

In the case of scattering of light, when the wavelength is much greater than the lattice constant, the second addend in the last factor in (14) – except for a narrow region near the temperature of the second-order phase transition, (where some components of the tensor  $\kappa_{\alpha\beta}^{-1}$  vanish) – is appreciably smaller than the first term. In this case the expression for the intensity of the scattered light assumes the more simple form

$$\frac{J_{1}}{J_{0}} = F(1,2)kT[\lambda_{\alpha\beta,\gamma}\lambda_{\alpha'\beta',\gamma'} + 4\lambda_{\alpha\beta,\gamma}\lambda_{\alpha'\beta',\gamma'\delta'}\overline{P}_{\delta'} + 4\lambda_{\alpha\beta,\gamma\delta}\lambda_{\alpha'\beta',\gamma'\delta'}\overline{P}_{\delta}\overline{P}_{\delta'}]e_{\alpha}^{1}e_{\alpha'}^{2}e_{\beta}^{2}e_{\beta'}^{2}\kappa_{\gamma\gamma'}.$$
(15)

All the quantities in formula (15) can be determined from independent measurements. Thus  $\lambda_{\alpha\beta,\gamma}$ and  $\lambda_{\alpha\beta,\gamma\delta}$ , according to (4), can be found, for example, by investigating the influence of an external electric field on the index of refraction, while the constants in expression (7) for F (1, 2) can be determined from ordinary optical measurements. Thus formula (15) enables us to calculate, in the general case of a ferroelectric with an arbitrary structure, the intensity and polarization of the part of the scattered light associated with the fluctuations of polarization. If the second addend in the last factor of Eq. (14) cannot be neglected, a more complicated formula must be used for evaluating  $J_1/J_0$ . Components of the tensor  $A_{\alpha\beta,\gamma\delta}$  entering into (14), which are also important in determining the intensities of scattering of x-rays, thermal neutrons, and other types of waves at temperatures near the second-order transition temperature, can be determined from comparisons of theory with experiment. They can also be calculated with the aid of the statistical theory of ferroelectrics based on a specific atomic model.

In many crystals all the components of the tensor  $\lambda_{\alpha\beta,\gamma}$  vanish identically, owing to the requirements imposed by symmetry of the crystal. In particular, this occurs in crystals with centers of symmetry in the nonferroelectric phase. (It was exactly this case that was examined in Ref. 1.) Temperature dependence of the intensity of the scattered light near the second-order phase transition temperature  $T_0$  differs markedly for crystals in which  $\lambda_{\alpha\beta,\gamma} = 0$  as compared with crystals in which  $\lambda_{\alpha\beta,\gamma} \neq 0$ . In the first case with  $T \ge T_0$ , when P = 0, according to (14) the intensity of the scattered radiation associated with fluctuations of polarization becomes zero.\* With  $T < T_0$  the ratio  $J_1/J_0$  increases very quickly, attains a maximum value and then falls off. When  $\lambda_{\alpha\beta,\gamma} \neq 0$ , the intensity of this scattering is not zero even with  $T > T_0$ . The ratio  $J_1/J_0$  attains a maximum at  $T = T_0$  and then decreases with further reduction of the temperature.

<sup>\*</sup>It must be emphasized that in the present contribution we consider only scattering by fluctuations of polarization and do not take into account scattering by thermal oscillations or by fluctuations of the internal parameters characterizing the short-range order the orientation of the dipole moments of different cells. The presence of inhomogeneities associated with fluctuations of these internal parameters must lead to additional scattering, which must occur both above and below the temperature  $T_0$ . There is reason to think, however, that in the most ineresting cases in the vicinity of the critical point for crystals with  $\lambda_{\alpha\beta,\gamma} = 0$  or near the usual transition point for crystals with  $\lambda_{\alpha\beta,\gamma} \neq 0$ , when the intensity of the scattering under examination here becomes anomalously great (see below), the scattering by fluctuations of polarization were taken into account) must be determined for constant values of the short-range order parameters plays a relatively insignificant values of the short-range order parameters, corresponding to thermodynamic equilibrium. Consequently, generally speaking, these quantities will differ somewhat from the corresponding quantities determined from the dependence of the equilibrium value of  $\epsilon^0$  on the intensity of the electric field, inasmuch as in the last case the short-range order parameters as well as P change.

For the purpose of a detailed investigation of the dependence of the intensity of scattering on the temperature and on the intensity of the electric field we shall examine below certain specific cases of ferroelectrics.

3. Let us start our examination with crystals of cubic symmetry (in the nonferroelectric state), having a center of symmetry, and in which the spontaneous polarization is directed along the cubic axis. In particular, barium titanate, one of the important ferroelectrics where practical engineering applications are concnerned, possesses such symmetry.<sup>4,5</sup> For simplicity let us assume that the electric field is directed along the ferroelectric axis which we shall choose as the OZ axis.

In crystals of this symmetry all the components of the tensor  $\lambda_{\alpha\beta,\gamma}$  vanish identically, while the tensor  $\lambda_{\alpha\beta,\gamma\delta}$  in the nonferroelectric state has only three nonzero components:  $\lambda_{11,11}$ ,  $\lambda_{12,12}$  and  $\lambda_{11,12}$ . In the ferroelectric state the crystal has tetragonal symmetry with which the nonzero components of the tensor  $\lambda_{\alpha\beta,\gamma\delta}$  are those identified by the indices 11, 11; 33, 33; 11, 22; 11, 33; 33, 11; 12, 12; 13, 13; 11, 13; 13, 11. (If the crystal has a center of symmetry,  $\lambda_{11,13} = \lambda_{13,11} = 0$ .) Below the transition temperature the dielectric susceptibility tensor has two nonzero components:  $\kappa_{33}$  and  $\kappa_{11} = \kappa_{22}$ .

It follows from Eq. (5) that in the given case in the region where one can neglect the terms square in q the intensity of the scattered light is

$$J_{1}/J_{0} = F(1, 2) \overline{P}^{2}kT \{4\lambda_{33,33}^{2} \times_{33} (e_{3}^{2})^{2} (e_{3}^{2})^{2} + 4\lambda_{11,33}^{2} \times_{33} (e_{1}^{1}e_{1}^{2} + e_{2}^{1}e_{2}^{2})^{2} + 8\lambda_{33,33}\lambda_{11,33} \times_{33} e_{3}^{1}e_{3}^{2} (e_{1}^{1}e_{1}^{2} + e_{2}^{1}e_{2}^{2}) + \lambda_{13,13}^{2} \times_{11} [((e_{1}^{1})^{2} + (e_{2}^{1})^{2})(e_{3}^{2})^{2} + (e_{3}^{1})^{2} ((e_{1}^{2})^{2} + (e_{2}^{2})^{2}) + 2(e_{1}^{1}e_{1}^{2} + e_{2}^{1}e_{2}^{2})e_{3}^{1}e_{3}^{2}]\}.$$
(16)

Neglecting the relatively minor tetragonality in calculating the factor F(1,2) we can set

$$F(1.2) = K_1 K_2 \omega^4 V / 16\pi^2 c^4 R^2.$$
(17)

Equation (16) enables us to determine the dependence of the intensity of scattering on the orientation of the polarization vectors of the incident and scattered radiation relative to the crystal axes and to investigate the depolarization of the light. Thus, for example, if the vectors  $e^1$  and  $e^2$  lie in a plane perpendicular to the Z axis and are mutually perpendicular, the intensity of scattering according to (16) goes to zero. On the other hand, if these vectors have components parallel to the Z axis, then even if  $e^1$  is perpendicular to  $e^2$  the intensity of scattering is not zero. In the narrow temperature interval near  $T_0$ , where one cannot neglect the terms square in q, the dependence of the scattering intensity on the direction of the vectors  $e^1$  and  $e^2$  becomes appreciably more complicated. Above the temperature  $T_0$ , as has already been noted, the scattering associated with the fluctuations of polarization in the crystals of this type vanishes.

In order to find the spontaneous polarization and the dielectric susceptibility tensor, which, according to (16), determine the temperature dependence of the scattering intensity, let us make use of the expansion of the thermodynamic potential per unit volume  $\varphi$  in powers of  $P_{\alpha}$ , introduced in other investigations<sup>6-12</sup> on the basis of the theory of phase transitions of the second order:<sup>13</sup>

$$\varphi = \varphi_0 + \frac{1}{2}a\left(T - T_0\right)P^2 + \frac{1}{4}B_1P^4 + \frac{1}{2}B_2\sum_{\alpha, \alpha'=1}^3 P_{\alpha'}^2 P_{\alpha'}^2 + \frac{1}{6}C_1P^6 + \frac{1}{2}C_2\sum_{\alpha, \alpha'=1}^3 P_{\alpha'}^4 P_{\alpha'}^2 + C_3P_1^2 P_2^2 P_3^2 - \mathbf{EP}.$$
 (18)

The expansion has been carried out to terms containing  $P_{\alpha}$  to the sixth power inclusive, in order to allow for a more detailed examination of the vicinity of the critical point where the intensity of scattering becomes particularly great.

The equilibrium value of the spontaneous polarization is determined from the condition  $\varphi_{\alpha} = 0$  ( $\alpha = 1$ , 2, 3). In the examined case (when  $\mathbf{P} \parallel OZ$ ), if  $\mathbf{E} = 0$ , the solution of this equation is of the form:

$$\overline{P}^2 = \overline{P}_3^2 = \left[-B_1 + \sqrt{B_1^2 + 4aC_1(T_0 - T)}\right] / 2C_1^2.$$
(19)

The dielectric susceptibility tensor can be found by substituting (18) in (11). Bearing in mind the equilibrium condition  $\varphi_3 = 0$ , we obtain that with  $\mathbf{E} = 0$ , the nonzero components of the tensor  $\kappa_{\alpha\beta}$  are described by

$$1/\varkappa_{33} = a \left(T - T_{0}\right) + 3B_{1}\overline{P}^{2} + 5C_{1}\overline{P}^{4} = 4a \left(T_{0} - T\right) + 2B_{1}\overline{P}^{2} = 2B_{1}\overline{P}^{2} + 4C_{1}\overline{P}^{4} - 4a \left(T_{0} - T\right) - B_{1}^{2}/C_{1} + \frac{B_{1}}{C_{1}}\sqrt{B_{1}^{2} + 4aC_{1}(T_{0} - T)}; \quad 1/\varkappa_{11} = 1/\varkappa_{22} = B_{2}\overline{P}^{2} + C_{2}\overline{P}^{4}.$$
(20)

Substituting (20) and (19) in Eq. (16), we obtain

$$\frac{J_{1}}{J_{0}} = F(1,2)kT \{ [2\lambda_{33,33}^{2}(e_{3}^{1})^{2}(e_{3}^{2})^{2} + 2\lambda_{11,33}^{2}(e_{1}^{1}e_{1}^{2} + e_{2}^{1}e_{2}^{2})^{2} + 4\lambda_{33,33}\lambda_{11,33}e_{3}^{1}e_{3}^{2}(e_{1}^{1}e_{1}^{2} + e_{2}^{1}e_{2}^{2})](B_{1}^{2} + 4aC_{1}(T_{0} - T))^{-1/2} + \lambda_{13,13}^{2}[((e_{1}^{1})^{2} + (e_{2}^{1})^{2})(e_{3}^{2})^{2} + (e_{3}^{1})^{2}((e_{1}^{2})^{2} + (e_{2}^{2})^{2}) + 2(e_{1}^{1}e_{1}^{2} + e_{2}^{1}e_{2}^{2})e_{3}^{1}e_{3}^{2}]/(B_{2} + C_{2}P^{2})\}.$$
(21)

At the critical point where the second-order phase transition curve becomes the first-order transition curve,  $B_1 = 0$ . Consequently near this point, where the phase transition curve  $B_1$  can be represented in the form of the expansion  $b(p - p_c)$  (here p is the pressure and  $p_c$  is the critical pressure), the scattering intensity becomes particular great, as may be seen from (21). The greatest contribution near the critical point is made by the first three terms in expression (21) (provided, of course, the constant  $B_2$  does not by chance also become small near the critical point). The temperature dependence of  $J_1/J_0$  is determined primarily by the factor  $[B_1^2 + 4aC_1(T_0 - T)]^{-1/2}$ . If  $B_1^2 \gg 4aC_1(T_0 - T)$ , then

$$(B_1^2 + 4aC_1(T_0 - T))^{-1/2} \approx 1/b (p - p_c),$$
(22)

that is, the intensity of scattering near the critical point must be strongly dependent on the pressure. In the vicinity of the critical point even a small decrease of temperature below the phase transition temperature results in the opposite condition:  $B_i^2 \ll 4aC_1(T_0 - T)$ . In this case, with some approximation,

$$J_1/J_0 \sim 1/\sqrt{(T_0 - T)}$$
. (23)

If the transition corresponds to the critical point  $(p = p_c)$ , the relationship (23) begins to hold on condition that the terms with  $A_{\gamma\gamma'\nu\nu'}q_{\nu}q_{\nu'}$  can be neglected.

In a first-order transition at the transition point  $aC_1(T - T_0) = 3B_1^2/16$ , that is,

$$[B_1^2 + 4aC_1(T_0 - T)]^{-1/2} = 2/B_1$$

and  $J_1/J_0$  is also approximately inversely proportional to the difference  $(p-p_c)$ .

In the presence of an electric field parallel to the Z axis the scattering intensity can be determined from Eq. (16), replacing  $\overline{P}$  by the solution of the equation

$$a(T - T_0)P + B_1P^3 + C_1P^5 = E$$
(24)

and determining  $\kappa_{33}$  by means of the first part of the equality (20) for  $1/\kappa_{33}$ . Specifically, in weak fields

$$\bar{P} = \bar{P}_0 + \varkappa_{033} E, \tag{25}$$

where  $\overline{P}_0$  and  $\kappa_{033}$  are, respectively, the values of spontaneous polarization and the dielectric susceptibility  $\kappa_{33}$  with no field. In this case application of the field will lead to replacement of the factor  $[B_1^2 + 4aC_1(T_0 - T)]^{-1/2}$  in Eq. (21) by

$$[B_1^2 + 4aC_1(T_0 - T)]^{-1/2} \left[ 1 - \frac{(B_1 + 6C_1\overline{P}_0^*) \varkappa_{033} E}{(B_1 + 2C_1\overline{P}_0^*) \overline{P}_0} \right].$$
(26)

Changes in the factor  $(B_2 + C_2 \overline{P}^2)^{-1}$  can be neglected at temperatures close to the phase transition temperature, when  $C_2 \overline{P}^2 \ll B_2$ . It will be seen from Eq. (26) that if the electric field is parallel to the spontaneous polarization, the scattering intensity decreases, while if the field is directed in the direction opposite to the polarization, the intensity of scattering increases. In view of the fact that in the vicinity of the phase transition point the dielectric susceptibility is very great while the spontaneous polarization is small, there should be observed in relatively weak fields a strong influence of the electric field on the scattering of light, i.e., a unique electro-optical effect. Thus, for example, assuming  $\overline{P}_0 \sim 10^4$  cgs electrostatic units ( $\sim \frac{1}{5}$  the saturation polarization for BaTiO<sub>3</sub>) and  $\kappa_{033} \sim 500$ , we obtain that the electric field substantially changes the intensity of light scattering when the field intensity is of the order of  $10^3 - 10^4$  v/cm.

Proceeding in an analogous fashion one can readily obtain the expressions for the scattering of light by ferroelectrics in which the spontaneous polarization vector is not directed along the cubic axis at  $T < T_0$ .

4. Crystals such as Rochelle salt and  $\text{KH}_2\text{PO}_4$  exhibit piezoelectric attributes not only in the ferroelectric but also in the nonferroelectric state.<sup>14,7</sup> Fluctuation of the polarization in such crystals even in the nonferroelectric region produces a proportional fluctuation of the deformation. Inasmuch as deformation

113

of the crystal in turn changes the dielectric constant (for light) by an amount proportional to this deformation, the components of the tensor  $\lambda_{\alpha\beta,\gamma}$  in Eq. (4) for crystals of this type are nonzero. In these crystals there is a preferred ferroelectric axis (OZ) at temperatures above the transition temperature, while in the vicinity of this temperature only the fluctuation of the component P<sub>3</sub> of the polarization vector becomes abnormally great. As follows from the general formulas (14) and (15), in the nonferroelectric region the terms containing the components of the tensor  $\lambda_{\alpha\beta,\gamma\delta}$  vanish. In the ferroelectric region these terms can be neglected near the transition temperature. As a result Eq. (14) in the given case assumes the form

$$J_1 / J_0 = g(1,2) / (\mathbf{x}_{33}^{-1} + B_{33\nu}q_\nu + A_{33\nu\nu'}q_\nu q_\nu),$$
<sup>(27)</sup>

where the quantity

$$g(1,2) = F(1,2) kT \left( \lambda_{\alpha\beta,3} e_{\alpha}^{1} e_{\beta}^{2} \right)^{2}$$
(28)

can be assumed to be temperature independent near the phase transition temperatre. This quantity depends on the direction of propagation of the incident and scattered waves and on their polarization relative to the crystal and can be determined through measurements of the index of refraction (for different values of  $\mathbf{k}$  and  $\mathbf{e}$ ) and its dependence on the electric field intensity. If the values of  $\lambda_{\alpha\beta,\gamma\delta}$  and  $P_{3H}$  (where  $P_{3H}$  is the saturation polarization) are much greater than  $\lambda_{\alpha\beta,\gamma}$ , then somewhat below the transition temperature it is necessary to take account of the terms containing  $\lambda_{\alpha\beta,\gamma\delta}$ . In this case g(1, 2) will strongly depend on the temperature:

$$g(1,2) = F(1,2) kT \left[ (\lambda_{\alpha\beta,3} + 2\lambda_{\alpha\beta,33} P_3) e_{\alpha}^1 e_{\beta}^2 \right]^2.$$
(29)

In order to determine the temperature dependence of the intensity of scattering let us make use of the expansion<sup>\*</sup> of the thermodynamic potential per unit volume in powers of  $P_3$ .<sup>7</sup>

$$\varphi = \varphi_0 + \frac{1}{2} a \left( T - T_0 \right) P_3^2 + \frac{B_1}{4} P_3^4 + \frac{C_1}{6} P_3^6 - E_3 P_3.$$
(30)

It follows from Eqs. (11), (30), and (28) that in the nonferroelectric region above the second-order phase transition temperature  $T_0\kappa_{33} = 1/a(T - T_0)$  and that the intensity of scattering is

$$J_1/J_0 = g(1,2)/[a(T-T_0) + B_{33\nu}q_{\nu} + A_{33\nu\nu'}q_{\nu}q_{\nu'}].$$
(31)

Thus, in contrast to the case of  $BaTiO_3$  crystals, scattering of light by fluctuations of the polarization occurs in the present case not only in the ferroelectric but also in the nonferroelectric region. Moreover the intensity of scattering becomes abnormally great not only in the vicinity of the critical point but also near the second-order phase transition point. (The sum  $A_{33\nu\nu'}q_{\nu}q_{\nu'}$  becomes comparable with a  $(T - T_0)$  only at very small values of  $T - T_0$ .)

Below the temperature  $T_0$  the spontaneous polarization is determined by expression (19) and the scattering intensity, according to (27), (11), (30), and (19), equals (cf. Ref. 15):

$$J_1/J_0 = g(1, 2)C_1/[4aC_1(T_0 - T) + B_1^2 - B_1\sqrt{B_1^2 + 4aC_1(T_0 - T)} + C_1B_{33\nu}q_\nu + C_1A_{33\nu\nu'}q_\nu q_{\nu'}].$$
(32)

If  $B_1^2 \gg 4aC_1 (T_0 - T)$ , then

$$J_1 / J_0 = g (1, 2) / [2a (T_0 - T) B_{33\nu} q_{\nu} + A_{33\nu\nu'} q_{\nu} q_{\nu'}]$$
(33)

The inequality  $B_1^2 \gg 4aC_1(T_0 - T)$  always holds in the case of ordinary phase transitions of the second order as well as in second-order phase transitions close to the critical point if  $T_0 - T$  is sufficiently small. In the case of transitions close to the critical point, however, the inequality  $B_1^2 \ll 4aC_1(T_0 - T)$  begins to obtain at relatively small values of  $T_0 - T$  and in this case

<sup>\*</sup> In the expansion (30) it is assumed that the component of the strain tensor  $u_{\alpha\beta}$  are already expressed through the polarization  $P_3$  (proportional to them) and eliminated from the expression for the thermodynamic potential. Analogously, in the expansion of  $\epsilon^0_{\alpha\beta}$  the tensor  $u_{\alpha\beta}$  is also eliminated, i.e., the expansion (4) is carried out not for a constant deformation but for a constant pressure.

## M. A. KRIVOGLAZ and S. A. RYBAK

$$J_1/J_0 = g(1,2)/[4a(T_0-T) + B_{33\nu}q_\nu + A_{33\nu\nu\nu'}q_\nu q_{\nu'}].$$
(34)

In the case of a first-order phase transition close to the second-order transition, utilizing the expressions<sup>15</sup> for the second derivatives of the thermodynamic potential, we find that above the transition temperature in the nonferroelectric phase

$$J_1/J_0 = g(1,2)C_1/[3B_1^2/16 + C_1B_{33\nu}q_{\nu} + C_1A_{33\nu\nu'}q_{\nu}q_{\nu'}],$$
(35)

while below the transition temperature in the ferroelectric phase

$$J_1/J_0 = g(1,2) C_1/[3B_1^2/4 + C_1B_{33\nu}q_{\nu} + C_1A_{33\nu\nu'}q_{\nu}q_{\nu'}].$$
(36)

Thus, if  $C_1 A_{33\nu\nu'} q_{\nu} q_{\nu'} \ll B_1^2$ , then in case of a phase transition the first order to the ferroelectric phase the intensity of scattering is reduced by a factor of four.

5. The above analyses pertained only to single-component crystals. In binary solid solutions, in addition to fluctuations of the polarization, there are also fluctuations of the concentration of one of the solution components. These fluctuations are not statistically independent (cf. Ref. 16), which can lead to the appearance of new effects.

When there are concentration fluctuations  $\Delta c$  (with constant polarization), the components of the tensor  $\epsilon^{0}_{\alpha\beta}$  change by amounts  $\lambda_{\alpha\beta,4}\Delta c$  proportional to  $\Delta c$ . Consequently in solid solutions, where there are fluctuations of both the polarization and composition, the Fourier component of the dielectric constant is defined by an expression which is a generalization of (5):

$$\varepsilon_{\alpha\beta,\,\mathbf{q}} = [\lambda_{\alpha\beta,\,\mathbf{q}} + 2\lambda_{\alpha\beta,\,\gamma\delta}\,\overline{P}_{\delta}]\,P_{\gamma\mathbf{q}} + \lambda_{\alpha\beta,\,4}\,c_{\mathbf{q}},\tag{37}$$

where  $c_{\mathbf{q}}$  is the Fourier component of the concentration while the quantities  $\lambda_{\alpha\beta,4}$  can be regarded as independent of **P** near the transition temperature. These constants correspond to changes in  $\epsilon_{\alpha\beta}^0$  with changes of c at constant values of  $P_{\alpha}$ . They are connected with the constants  $\lambda'_{\alpha\beta,4}$ , corresponding to changes in  $\epsilon_{\alpha\beta}^0$  with changes in c at equilibrium values of  $P_{\alpha}$  (which vary with changes in the composition) by the simple relationship

$$\lambda_{\alpha\beta,\,4} = \lambda'_{\alpha\beta,\,4} - \frac{\partial \varepsilon_{\alpha\beta}}{\partial P_{\gamma}} \, \frac{dP_{\gamma}}{dc}$$

Substituting Eq. (37) in (1) we find that the light scattering intensity is expressed through the mean values  $\overline{P_{\gamma q} P_{\gamma' q}^*}$ ,  $\overline{P_{\gamma q} c_q^*}$  and  $\overline{|c_q|^2}$ . These mean values, as before, can readily be found by considering the terms proportional to  $\Delta P_{\alpha} \Delta P_{\beta}$ ,  $\Delta P_{\alpha} \Delta c$  and  $(\Delta c)^2$  in the expansion of the thermodynamic potential. Carrying out the same calculations as for the single-component ferroelectric, we find that in ferroelectric solid solutions the scattering intensity associated with fluctuations of polarization and composition is given by the expression

$$J_1/J_0 = F(1,2) kT \left[\lambda_{\alpha\beta, i} \lambda_{\alpha'\beta', i'} K_{ii'}^{-1} + 4\lambda_{\alpha\beta, i} \lambda_{\alpha'\beta', \gamma'\delta'} \overline{P}_{\delta'} K_{i\gamma'}^{-1} + 4\lambda_{\alpha\beta, \gamma\delta} \overline{P}_{\delta} \overline{P}_{\delta'} K_{\gamma\gamma'}^{-1}\right] e_{\alpha}^1 e_{\alpha'}^2 e_{\beta}^2 e_{\beta'}^2.$$

$$(38)$$

[instead of Eq. (15)].

Here i and i' go through 1, 2, 3, and 4 (while  $\gamma$  and  $\gamma'$  run through 1, 2, and 3), the quantities  $K_{ii'}^{-1}$  are matrix elements of the matrix that is the reciprocal of the matrix  $K_{ii'}$ , while the matrix elements of the latter matrix are

$$K_{ii'} = \varphi_{ii'} = \varkappa_{ii'}^{-1} \quad \text{with } i = 1, 2, 3 \qquad K_{i4} = K_{4i} = \varphi_{ic} \quad \text{with } i = 1, 2, 3 \quad K_{44} = \frac{\partial^2 \varphi}{\partial c^2}.$$
(39)

Since the equilibrium condition  $\varphi_{\alpha} = 0$  is fulfilled at all concentrations, we have

$$d\varphi_{\alpha}/dc = \varphi_{\alpha\alpha'}dP_{\alpha'}/dc + \varphi_{\alpha c} = 0.$$

Therefore

$$\varphi_{\alpha c} = -\varphi_{\alpha \alpha'} dP_{\alpha'} / dc = -\kappa_{\alpha \alpha'}^{-1} dP_{\alpha'} / dc.$$
(40)

In the nonferroelectric region the derivatives  $dP_{\alpha}/dc$  equal zero and the fluctuations of the polarization and of the composition are statistically independent. In the ferroelectric region some of the derivatives

 $\varphi_{\alpha c}$  are nonzero; consequently these fluctuations become statistically dependent. Further the derivative  $\varphi_{cc}$  near the second-order phase transition temperature can be written in the form

$$\rho_{cc} = N \left( \partial \mu_A / \partial c - \partial \mu_B / \partial c \right), \tag{41}$$

where N is the number of molecules per unit volume and  $\mu_A$  and  $\mu_B$  are the chemical potentials of the first and second components of the solution in the nonferroelectric region.

Thus all the quantities entering into expression (38) for the intensity of scattering by solid solutions of ferroelectrics, just as in the case of single component ferroelectrics, can be determined by means of independent experiments. To find these quantities one must determine the index of refraction and its dependence on the electric field intensity and the concentration, the static dielectric susceptibility tensor, and the dependence of the spontaneous polarization and the chemical potentials of the components on the composition. In the immediate vicinity of the transition temperature the tensor  $K_{ii'}$  should be replaced by

$$K_{ii'} + B_{ii'\nu} q_{\nu} + A_{ii'\nu\nu'} q_{\nu} q_{\nu'} (i,i' = 1, 2, 3, 4; \nu, \nu' = 1, 2, 3),$$

where  $B_{ii'\nu}$  and  $A_{ii'\nu\nu'}$  near  $T_0$  can be regarded as temperature independent.

In the case of cubic ferroelectrics in which the spontaneous polarization is directed along the cubic axis and which have a center of symmetry,  $\lambda_{\alpha\beta,i} = 0$  with  $i = 1, 2, \text{ and } 3, \lambda_{\alpha\beta,4} = \lambda_4 \delta_{\alpha\beta}$  and the only one of the derivatives  $\varphi_{\alpha C}$  that is nonzero is  $\varphi_{3C}$ . In this case for  $T < T_0$ , we obtain the following expression for the intensity of scattering

$$\frac{J_{1}}{J_{0}} = F(1,2) kT \left\{ \frac{4\bar{P}^{2} \varkappa_{33}}{(-(\varphi_{3c})^{2} \varkappa_{33}/\varphi_{cc}} \left[ \lambda_{33,33}^{2} \left( e_{3}^{1} \right)^{2} \left( e_{3}^{2} \right)^{2} + \lambda_{11,33}^{2} \left( e_{1}^{1} e_{1}^{2} + e_{2}^{1} e_{2}^{2} \right)^{2} + 2\lambda_{33,33} \lambda_{11,33} e_{3}^{1} e_{3}^{2} \left( e_{1}^{1} e_{1}^{2} + e_{2}^{1} e_{2}^{2} \right) \right] \\
+ \lambda_{13,13}^{2} \bar{P}^{2} \varkappa_{11} \left[ \left( \left( e_{1}^{1} \right)^{2} + \left( e_{2}^{1} \right)^{2} \right) \left( e_{3}^{2} \right)^{2} + \left( e_{3}^{1} \right)^{2} \left( \left( e_{1}^{2} \right)^{2} + \left( e_{2}^{2} \right)^{2} \right) + 2 \left( e_{1}^{1} e_{1}^{2} + e_{2}^{1} e_{2}^{2} \right) e_{3}^{1} e_{3}^{2} \right] \\
+ \frac{4\lambda_{4} \bar{P} \varphi_{3c} \varkappa_{33} \left( e^{1} e^{2} \right)}{\varphi_{cc} \left[ 1 - \left( \varphi_{3c} \right)^{2} \varkappa_{33} \right] \varphi_{cc}^{1}} \left[ \lambda_{33,33} e_{3}^{1} e_{3}^{2} + \lambda_{11,33} \left( e_{1}^{1} e_{1}^{2} + e_{2}^{1} e_{2}^{2} \right) \right] + \frac{\lambda_{4}^{2}}{\varphi_{cc} - \left( \varphi_{3c} \right)^{2} \varkappa_{33}} \right\},$$
(42)

where F(1,2) is defined by Eq. (17). In the nonferroelectric region with  $T > T_0$ 

$$\frac{J_1}{J_0} = F(1,2) kT \lambda_4^2 / \varphi_{cc}.$$
 (43)

In solid solutions there is thus scattering of light by fluctuations of the concentration with  $T > T_0$  as well. The intensity of scattering increases in the ferroelectric region.

The intensity of scattering can also be expressed in terms of the constants entering into the expansion (18) of the thermodynamic potential in powers of  $P_{\alpha}$ . Invoking Eqs. (19) and (20) and the expression  $\varphi_{3C} = -a\overline{P}dT_0/dc$  which is valid near the transition temperature, we obtain

$$\frac{J_{1}}{J_{0}F_{1}(1,2)kT} = [B_{1}^{2} + 4aC_{1}(T_{0} - T)]^{-1/2} \left[ 1 - a^{2} \left( \frac{dT_{0}}{dc} \right)^{2} \left( 2\varphi_{cc} \sqrt{B_{1}^{2} + 4aC_{1}(T_{0} - T)} \right)^{-1} \right] \varphi_{cc}^{-1} \left\{ 2\varphi_{cc} \left[ \lambda_{33,33}^{2} \left( e_{3}^{1} \right)^{2} \left( e_{3}^{2} \right)^{2} + \lambda_{11,33}^{2} \left( e_{3}^{1} \right)^{2} \left( e_{3}^{2} \right)^{2} + \lambda_{11,33}^{2} \left( e_{3}^{1} \right)^{2} + 2\lambda_{33,33} \lambda_{11,33} e_{3}^{1} e_{3}^{2} \left( e_{3}^{1} + e_{2}^{1} e_{2}^{2} \right) \right] - 2\lambda_{4}a \frac{dT_{0}}{dc} \left( e^{1} e^{2} \right) \left[ \lambda_{33,33} e_{3}^{1} e_{3}^{2} + \lambda_{11,33} \left( e_{1}^{1} e_{1}^{2} + e_{2}^{1} e_{2}^{2} \right) \right] + \lambda_{4}^{2} \sqrt{B_{1}^{2} + 4aC_{1}(T_{0} - T)} + \frac{\lambda_{13,13}^{2}}{B_{2} + C_{2}\overline{P^{2}}} \left[ \left( (e_{1}^{1})^{2} + (e_{2}^{1})^{2} \right) \left( e_{3}^{2} \right)^{2} + (e_{3}^{1})^{2} \left( (e_{1}^{2})^{2} + (e_{2}^{2})^{2} \right) + 2 \left( e_{1}^{1} e_{1}^{2} + e_{2}^{1} e_{2}^{2} \right) e_{3}^{1} e_{3}^{2} \right] \right\}.$$

$$(44)$$

In solid solutions the critical point lies at the point where the second-order phase transition curve goes over into the decomposition curve.<sup>17,13</sup> According to Landau,<sup>17</sup> instead of the condition  $B_1 = 0$ , which is true for single component crystals, we have at this point the condition

$$\varphi_{cc} B_1 = \frac{1}{2} (a dT_0 / dc)^2.$$

It is evident from (44) that in solid solutions there should also be observed abnormally great scattering in the vicinity of the critical point. This scattering is associated both with fluctuations of the polarization and with fluctuations of the concentration. Even if the index of refraction is only weakly influenced by the polarization, i.e., if the values of  $\lambda_{\alpha\beta,\gamma\delta}$  are small, extremely intense scattering of light (provided  $\lambda_4$ is not small) due to fluctuation of the composition should be observed in solid solutions close to the critical point.

In solid solutions of ferroelectrics such as Rochelle salt or  $KH_2PO_4$ , where the components of the ten-

sor  $\lambda_{\alpha\beta,\gamma}$  ( $\gamma = 1, 2, 3$ ) are nonzero, in ordinary second-order phase transitions (far from the critical point) fluctuations of the composition near the temperature  $T_0$  play a relatively minor part in the scattering of light. Hence above the transition temperature, where  $\varphi_{3c} = 0$ , we can as before use expressions (27) or (31). Below the transition temperature in the ferroelectric region one should replace  $1/\kappa_{33}$  by  $(1/\kappa_{33})[1 - (\varphi_{3c})^2\kappa_{33}/\varphi_{cc}]$  in the denominator of (27). In terms of the thermodynamic theory of phase transitions of the second order this reduces to replacement of  $2a(T_0 - T)$  in the denominator of Eq. (33) by

$$2a(T_0 - T)[1 - a^2(dT_0/dc)^2/2B_1\varphi_{cc}].$$

6. The results deduced above, after appropriate modification of the designations, can obviously be fully applied to transparent ferromagnetic crystals. The results obtained by means of the theory of phase transitions of the second order can also be applied to antiferroelectric and antiferromagnetic materials in the absence of an external electric field where the former are concerned or a magnetic field where the latter are concerned. In the presence of an external electric field the state of an antiferroelectric is characterized by two vector parameters and the formulas for the intensity of light scattering, just as in the cases discussed above, can be obtained on the basis of the thermodynamic theory of phase transitions.<sup>18</sup>

All the expressions given above pertain to the scattering of light by a single crystal having no domains. A single crystal of this type can be realized by placing the ferroelectric in a capacitor.<sup>7</sup> Our results can also be applied to the scattering of light by individual domains provided their dimensions are appreciably greater than the wavelength of the light. If, however, the dimensions of the domains are of the same order of magnitude or smaller than the wavelength, then in addition to the scattering examined above there must also be evinced an additional extremely intense scattering by the domain walls.

In addition to scattering of light by the fluctuations of the polarization and the composition, in crystals there is always also some scattering connected with lattice vibrations. The intensity of this part of the scattering can be calculated<sup>3</sup> if the elastic and elastic-optical constants of the crystals are known. Both types of scattering can often be partially separated experimentally inasmuch as they depend in different ways on the temperature and external fields.

It would be of great interest to carry out an experimental investigation of light scattering near the points of second-order phase transitions in different ferroelectric materials with simultaneous measurements of the static dielectric constant, the index of refraction and its dependence on the electric field and the composition.

#### CONCLUSIONS

1. We have deduced equations for determining intensity and polarization of light scattered both by single-component ferroelectrics (or ferromagnets) of arbitrary structure and by solid solutions. All the quantities entering into the expressions for the scattering associated with fluctuations of the polarization (magnetization) and of the composition can be found (or evaluated) by means of independent measurements of the index of refraction and its dependence on the external field and the composition, static dielectric constant and the concentration dependence of the spontaneous polarization and the chemical potentials (for solid solutions).

2. If the expansion of the dielectric constant (for light) in powers of the components of the polarization vector contains linear terms (the case, for example, in crystals such as Rochelle salt or  $\rm KH_2PO_4$ ), then both below and above the second-order phase transition temperature there must be observed anomalously great scattering. If there are no linear terms in this expansion (for example, in crystals such as  $\rm BaTiO_3$ ), then scattering of the type in question will occur only in the ferroelectric (ferromagnetic) region and will be absent (in single component crystals) above the transition temperature.

3. The temperature dependence of the intensity of light scattering by crystals of the  $BaTiO_3$  and crystals of the type of Rochelle salt and  $KH_2PO_4$  was evaluated by means of the thermodynamic theory of phase transitions of the second order. In crystals of the  $BaTiO_3$  type (both in single component ferroelectrics and solid solutions) particularly intense scattering should occur near the critical point on the second-order phase transition curve.

4. There should be observed an appreciable change in the intensity of scattering by ferroelectrics in an external electric field; this should occur in relatively weak fields (for  $BaTiO_3$  type crystals, in fields

of the order of  $10^3 - 10^4$  v/cm). In the vicinity of the critical point the scattering should be strongly dependent on the pressure.

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# ON THE EFFECTIVE FIELD IN A PLASMA

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The Bogoliubov equations for the "partial distribution functions" are used to compute the effective field acting on charged particles in a plasma. It is shown that the effective field differs from the mean field by a small quantity of the order 1/N where N is the number of particles within a sphere whose radius is equal to the Debye radius. This result also holds in the presence of a magnetic field.

As is well known, the electric field acting on an individual particle of a medium is not equal to the average field in the medium. For example, in a gas of free dipoles the effective field  $\mathbf{E}_{eff}$  is given by the Lorentz formula  $\mathbf{E}_{eff} = \mathbf{E} + 4\pi \mathbf{P}/3$  where  $\mathbf{E}$  is the average field and  $\mathbf{P}$  is the polarization of the medium. This formula is obtained on the assumption that the molecular dipoles are mutually impenetrable so that each dipole behaves as if it were placed inside a cavity in a polarized medium.

In the case of ionized plasma there is, of course, no basis for such an assumption. However, the effective field in a plasma should also, generally speaking, differ from the average field because there exists