

Theory of Elliptic Polarization of Light Reflected from Isotropic Media

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With reflection of light viewed as interference of waves radiated by molecules and atoms of a medium, equations that take into account the effect of the transition layers are derived for the amplitudes of the reflected light. No special assumptions are made concerning the molecular structure of the medium or of the transition layer. The equations are valid for layers of any thinness, including monomolecular layers. At the end of the work, the equations are supplemented by second order terms in $f\gamma_m$. Complex mathematical calculations are avoided by converting radiation from volume sources to radiation from surface sources.

1. THE latest investigations by Kizel¹ make timely the development of a theory of elliptic polarization of light reflected by an isotropic medium. This theory is free of the limitations imposed by the phenomenological treatment and does not introduce simplified concepts with respect to the molecular structures of the reflecting medium and of the transition layer. Results following from this theory were given in part in Ref. 2. However, the proof given there is easier stated than derived. Here we introduce the single hypothesis that the elliptic polarization is the result of the existence of transition layer on the reflecting surface of the medium, with no assumptions made at all concerning the molecular structure of the medium or of the transition layer. To free ourselves of the restrictions of the phenomenological theory, we shall dispense also with phenomenological description of the transition layer in terms of the dielectric constant or the index of refraction. At the same time no limitations whatever are imposed on the lower thickness limit of the transition layer. To exhibit more clearly the physical nature of the theory, we propose to employ elementary calculations almost exclusively or to avoid calculations where possible.

2. We know that the reflected wave is the result of interference of secondary waves, radiated by the molecules or atoms of the medium. Under the influence of the incident wave and of the radiation of the remaining molecules and atoms these molecules or atoms acquire dipole moments and therefore produce radiation. Since we are interested in the radiation field at distances from the medium that are very large compared with the molecular dimensions and the intermolecular distances, we can replace the discrete radiation centers in the

medium by continuously distributed sources. Let us introduce two types of such sources--volume sources and surface sources. If a plane monochromatic wave is incident on the medium the polarization vector of the medium consists of a homogeneous-wave term:

$$\mathbf{P} = \mathbf{P}_0 e^{i(\omega t - kr)} \quad (1)$$

and of a supplementary polarization of the transition layer. By virtue of the polarization term (1), each volume element of the medium can be considered as a radiating Hertz dipole of moment $\mathbf{P} dV$. In addition to these volume sources, it is also necessary to account for the radiation from the transition layer. This can be done numerically by assuming that each elementary area dS on the surface of the body is a radiating Hertz dipole of moment $\vec{\tau} ds$. The vector is the supplementary

$$\vec{\tau} = \vec{\tau}_0 \exp \{i(\omega t - k_x x)\} \quad (2)$$

mentary dipole moment per unit surface of the body (the surface of the medium is taken to be the XY coordinate plane; the Z axis is directed downward inside the medium; X and Z lie in the plane of incidence). This vector and the proposed theory make possible a description of the properties of transition layer, inasmuch as we deal with a first-order approximation. The question of the origin of this layer is not touched upon in this theory. The layer may be either the result of soiling or processing the reflecting surface, or the result of the molecular structure of the medium near the surface, or finally the result of a difference between the effective field near the surface and the effective field inside the medium.

3. The radiation from the volume sources can be reduced formally to a radiation from surface sources. Let us subdivide the entire medium into plane-parallel layers of equal thickness. These layers we shall call zones, in analogy with the Fresnel zones. The radiation field in the upper half space

¹V. A. Kizel', J. Exptl. Theoret. Phys. (U.S.S.R.) 26, 228 (1954); 29, 658 (1955).

²D. V. Sivukhin, J. Exptl. Theoret. Phys. (U.S.S.R.) 18, 976, (1948); 21, 367 (1951).

due to each of these zones is a plane wave, propagating in the direction of the wave vector \vec{f}' of the reflected wave. The direction and length of this vector as well as of the wave vector \vec{f} of the incident wave are determined by the following conditions:

$$f^2 = f'^2 = \omega^2 / c^2, \quad f_x = f'_x = k_x; \quad (3)$$

$$f_z = -f'_z = \sqrt{f^2 - f_x^2}.$$

Let the thickness of each zone be $L = \pi / (k_z + f_z)$. The plane waves radiated by the neighboring zones are then of opposite phase, and the radiation field in the upper half space is represented by an alternating series:

$$\mathbf{E} = \mathbf{E}_1 - \mathbf{E}_2 + \mathbf{E}_3 - \dots, \quad (4)$$

in which every term represents the radiation field due to the corresponding zone. Were the polarization wave (1) strictly homogeneous, and were the spherical waves radiated by the volume elements of the medium to experience no absorption at all, the absolute values of all terms of series (4) would be equal and the series itself would diverge (oscillate). Actually, however, absorption should cause the terms of series (4) to decrease monotonically, thereby insuring its convergence. If the absorption is slight, the decrease should be in geometric progression, since the attenuation of a plane wave of light passing through an infinitesimally thin layer of a medium is proportional to the thickness dx of this layer, owing to the linearity of the field equations. Denoting the denominator of the progression by q , we can therefore write $\mathbf{E} = \mathbf{E}_1 / (1 - q)$.

In the limiting case of infinitely small absorption $q = -1$, and we obtain $\mathbf{E} = \mathbf{E}_1 / 2$. Thus the field intensity produced by radiation from the entire medium in the upper half space is equal to half the field intensity produced by radiation from the first zone.

Let us subdivide the first zone into a large number of subzones of equal thickness and employ the vector-diagram method (Fig. 1). The complex amplitudes of the waves produced in the upper half space by the individual subzones are represented on the vector diagram by small arrows, forming half a regular polygon, and becoming a semicircle in the limit. The diameter $AB = D$ of this semicircle, is the complex amplitude of the wave radiated by the entire first zone. Were all phases of the waves radiated by the individual subzones identical and equal to the phase of the wave radiated by the first subzone, we would obtain (instead of

a semicircle) a straight-line segment of length $\pi D/2$. This would increase the wave $\pi/2$ times, and the phase of the resultant wave would lead the phase of the wave radiated by the entire first zone by $\pi/2$. Considering that the amplitude of the wave radiated by the entire medium is equal to half the amplitude of the wave radiated by the first zone, we obtain the following theorem: If a half space bounded by a plane is filled with a medium the polarization vector of which is given by the plane wave (1), the radiation produced outside the medium is equivalent to the radiation due to an infinitely thin layer, placed on the surface of the medium, provided the dipole moment per unit area of this surface is:

$$\begin{aligned} \mathbf{A} &= \frac{L}{\pi} \mathbf{P}_0 \exp \{i(\omega t - k_x x - \pi/2)\} \\ &= -\frac{i}{f_z + k_z} \mathbf{P}_0 \exp \{i(\omega t - k_x x)\}. \end{aligned} \quad (5)$$

The proven theorem is valid for the radiation field only at distances from the surface of the radiating medium that are large compared with the molecular dimensions and intermolecular distances. Incidentally, this limitation is not as rigid as appears at first glance. Exact calculations by Ewald³ have shown that the radiation field of a crystal lattice is practically equal to the radiation of a solid medium even at distances on the order of the lattice constant.

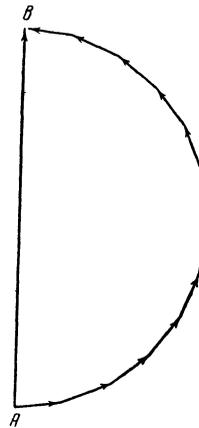


FIG. 1.

4. To obtain the field of the reflected wave, it is necessary also to take into account the added radiation produced by the transition layer. This can be done by adding the vector $\vec{\tau}$ to expression (5). This leads to the following theorem. The

³ P. P. Ewald, Ann. Physik 49, 117 (1916).

field of the reflected wave is equivalent to the field due to an infinitesimally thin layer, placed on the surface of the medium and having the following dipole moment per unit area:

$$A^{(r)} = (\vec{\tau}_0 - \frac{i}{f_z + k_z} \mathbf{P}_0) \exp \{i(\omega t - k_x x)\}. \quad (6)$$

This theorem is valid not only for infinitesimally thick transition layers, but also for layers of any thickness. In fact, in a manner similar to that used for the radiation due to the medium, the radiation produced by the transition layer in the upper half space can be replaced by the radiation due to an infinitesimally thick layer on the surface of the medium, provided only that $\vec{\tau}_0$ is suitably defined.

5. Let us now replace the field of the incident wave by the radiation from an infinitely thin layer, also placed at the boundary of the medium. This can be done in the following manner: The field at point Q , located within the medium where the polarization wave is practically homogeneous, i.e., far from the boundary, it can be represented in the form $E_Q = E^{(e)} + E_{l_0} + E_{l_a}$, where $E^{(e)}$ is the field intensity produced at Q by the incident wave; E_{l_0} -- the field intensity due to radiation from the medium filling the lower half space, calculated under the assumption that the polarization wave of the medium is homogeneous everywhere; E_{l_a} -- intensity of the additional radiation field due to the transition layer.

If we now imagine for an instant that the medium fills all the infinite space both above and below the coordinate plane XY , and that a homogeneous polarization wave (1) is propagated in this medium, then it is obvious that the field at point Q remains unchanged, since, by assumption, the polarization of the medium does not vary at this point. But in this case we can write $E_Q = E_{l_0} + E_u$, where E_u is the radiation field due to the upper half of the medium. Comparison with the preceding equation gives: $E^{(e)} = E_u - E_{l_a}$. The effect of the upper half of the medium can be reduced to the radiation field due to an infinitely thin layer, placed in the coordinate plane XY , similar to the manner used for the lower half of the medium. The effect of the transition layer in the lower half space can be reduced to the radiation due to an infinitely thin layer with a dipole-moment surface density:

$$\vec{\tau}' = \vec{\tau}_0 \exp \{i(\omega t - k_x x)\}. \quad (7)$$

This leads to the following theorem: The field due to an incident wave in the lower half space is the same as the field radiated by an infinitely thin layer, placed at the boundary of the medium, provided the dipole moment per unit surface area is:

$$A^{(e)} = (-\vec{\tau}'_0 + \frac{i}{k_z - f_z} \mathbf{P}_0) \exp \{i(\omega t - k_x x)\}. \quad (8)$$

Where we write $\vec{\tau}'_0$ instead of $\vec{\tau}_0$, since we deal with radiation produced by the transition layer in the lower half space. If the transition layer is thick the decrease in the phases of the sources in the Z direction causes the radiation in the upper and lower half space to be different. Consequently $\vec{\tau}'_0 \neq \vec{\tau}_0$ in general. Only in the case of very thin transition layers, when the thickness is quite small compared with the wave length, can we neglect the changes in phase over the thickness of the layer; in this approximation we can assume $\vec{\tau}'_0 = \vec{\tau}_0$.

6. The problem of computing the amplitudes of the reflected light has thus been reduced to a comparison of the radiation produced by two infinitesimally thin layers with dipole-moment surface densities (6) and (8). It is necessary to recall here that the radiation from a dipole depends on the relative orientation of the dipole and of the radiation: the only effective dipole-moment component is the one perpendicular to the direction of the radiation; the parallel component produces no radiation. Taking this into account, let us introduce the following symbols:

$$a_m = \tau_{0m} / P_{0m}; \quad a'_m = \tau'_{0m} / P_{0m} \\ (m = x, y, z)$$

(by virtue of the symmetry $a_x = a_y$; $a'_x = a'_y$) and let us examine two cases.

First case. Electric vector perpendicular to the plane of incidence. Evidently,

$$R_s / \mathcal{E}_s = A_y^{(r)} / A_y^{(e)},$$

where \mathcal{E}_s and R_s are the complex amplitudes of the incident and reflected waves. Inserting (6) and (8) we obtain by simple transformations

$$\frac{R_s}{\mathcal{E}_s} = - \frac{\sin(\varphi - \psi) [1 + ia_y f (\cos \varphi + n \cos \psi)]}{\sin(\varphi + \psi) [1 - ia'_y f (\cos \varphi - n \cos \psi)]}, \quad (10)$$

where φ is the angle of incidence, ψ the angle of refraction, and n the index of refraction of the medium.

Second case. Electric vector lies in the plane of incidence. Let \mathbf{e} and \mathbf{e}' be unit vectors in the plane of incidence, perpendicular to the incident and reflected waves respectively, and directed toward the normal to the surface of the medium. Then

$$R_p/\mathcal{E}_p = (\mathbf{A}^{(r)}\mathbf{e}')/(\mathbf{A}^{(e)}\mathbf{e}),$$

where \mathcal{E}_p is the complex amplitude of the incident wave, and R_p that of the reflected wave. Inserting

$$\frac{R_p}{\mathcal{E}_p} = \frac{\operatorname{tg}(\varphi - \psi)}{\operatorname{tg}(\varphi + \psi)} \frac{1 - if(a_z \sin \varphi - a_x \cos \varphi \operatorname{ctg} \psi) \operatorname{tg}(\varphi + \psi)}{1 + if(a'_z \sin \varphi + a'_x \cos \varphi \operatorname{ctg} \psi) \operatorname{tg}(\varphi - \psi)}. \quad (11)$$

7. For all their generality Eqs. (10) and (11) have the substantial shortcoming that the quantities a_m and a'_m remain undetermined. However, in the case of thin transition layers it is possible to make definite conclusions concerning these quantities. They have the dimensionality of length, and we shall call the transition layer thin if a_m and a'_m are quite small compared with the wave length. Let us expand a_m and a'_m in power series in f and let us keep only the zero-order terms of these expansions. In this approximation we neglect the variation of the phase of the source with the thickness of the layer. Therefore $a_m = a'_m = \gamma_m$, where γ_m is the common zero-order term in the expansions for a_m and a'_m . Substituting this value into (10) and (11) and disregarding the terms in $(f\gamma_m)^2$ we obtain:

$$\frac{R_s}{\mathcal{E}_s} = -\frac{\sin(\varphi - \psi)}{\sin(\varphi + \psi)} [1 + 2if\gamma_y \cos \varphi]; \quad (12)$$

$$\frac{R_p}{\mathcal{E}_p} = \frac{\operatorname{tg}(\varphi - \psi)}{\operatorname{tg}(\varphi + \psi)} \times \left[1 + 2if \cos \varphi \frac{\gamma_x \cos^2 \psi - \gamma_z \sin^2 \varphi}{\cos^2 \psi - \sin^2 \varphi} \right]. \quad (13)$$

The parameters $\gamma_x = \gamma_y$ and γ_z are independent of the field intensity and of the frequency of the incident wave (neglecting dispersion). These parameters indeed characterize the properties of the transition layer in the approximation employed here.

8. Assume that the incident light is linearly polarized at a 45° angle with the plane of incidence, i.e., $\mathcal{E}_s = \mathcal{E}_p$. Let us introduce the designation

$$R_p/R_s = \eta e^{i\delta}. \quad (14)$$

The polarization of the reflected light will in general be elliptic. We shall call the ratio of the minor to the major axes of the corresponding el-

(6) and (8) and allowing for the transverse nature of the reflected wave ($\mathbf{k} \cdot \mathbf{P} = 0$) we obtain by a simple transformation:

lipse the ellipticity coefficient of the reflected light. If the reflected light is passed through a Nicol-prism analyzer, it becomes again linearly polarized, whereby η (which can always be made positive by proper choice of δ) will equal to the ratio of the p and s components of the electric vector of the reflected waves, i.e., to the tangent of the azimuth of the restored polarization of the reflected light. In the experiments one usually measures η and δ . The ellipticity coefficient ρ is calculated from the following equation:

$$\rho^2 = \frac{1 + \eta^2 - [(1 + \eta^2)^2 - 4\eta^2 \sin^2 \delta]^{1/2}}{1 + \eta^2 + [(1 + \eta^2)^2 - 4\eta^2 \sin^2 \delta]^{1/2}}. \quad (15)$$

We stopped to discuss these known definitions and relationships because in the literature (see, for example, Ref. 1), ρ is sometimes confused with η . Actually, η characterizes only the position of the planar polarization of the reflected light after its linear polarization has been restored by a Nicol-prism analyzer. The quantity η alone cannot establish the form and orientation of the ellipse of oscillations, and δ must also be known for this purpose. In particular, if $\delta = \pi/2$, we have $\rho = \eta$.

9. If we determine R_p/R_s from (12) and (13) and compare the results with (14), we readily obtain

$$\eta \cos \delta = -\frac{\cos(\varphi + \psi)}{\cos(\varphi - \psi)}; \quad (16)$$

$$\eta \sin \delta = 2f(\gamma_z - \gamma_x) \frac{\cos \varphi \sin^2 \varphi}{\cos^2(\varphi - \psi)}.$$

These relationships are accurate as long as the $(f\gamma_m)$ terms do not exceed the first power; they yield:

$$\operatorname{tg} \delta = 2f(\gamma_z - \gamma_x) \cos \varphi \sin^2 \varphi / (\sin^2 \varphi - \cos^2 \psi). \quad (17)$$

As far as η goes, equations (16) cannot be used to compute this quantity with an accuracy greater than that given by the simple Fresnel equations. It would be necessary for this purpose to carry out all calculations with an accuracy up to $(f\gamma_m)^2$.

The only exception occurs when the light is incident at the Brewster angle or very nearly so. In this case the zero term in the expansion for $\eta \cos \delta$ vanishes, so that the expansion itself starts with a second-degree term which affects only the third-order quantities in the expression for ρ and can be discarded. If the incidence in this approximation is at the Brewster angle, the second equation of (16) yields

$$\eta = \rho = (\pi/\lambda) \sqrt{n^2 + 1} (\gamma_z - \gamma_x), \quad (18)$$

since in this case $\delta = \pi/2$. Here n is the index of refraction of the medium, and λ is the wavelength in vacuum.

Thus, independently of the structure of the medium and of the transition layer, relationships (17) and (18) should hold, provided only that the causes of ellipticity are the transition layers at the reflecting surface of the medium. There are not enough experimental data available to decide with full assurance whether these relationships are true or not. In particular, the corollary of Eq. (18), that the ellipticity of the reflected light is inversely proportional to the wavelength, has never been checked at all. According to Eq. (17), $\tan \delta$ is inversely proportional to λ . This consequence has also not been checked. The dependence of δ on the angle of incidence φ has not been studied sufficiently. Before we can study the structure of the reflecting surfaces of media by reflected light it is necessary to investigate all these problems and thus corroborate or reject the basic initial premises of the theory, namely that the ellipticity of the reflected light results from the existence of transition layers. We can hope that the continuing investigations by Kizel' will throw light on this problem.

The dependence of the ellipticity coefficient ρ or of the quantity η on the angle of incidence φ in the vicinity of the Brewster angle cannot serve as a criterion for the correctness of the theory proposed, for the theory is true only with an accuracy to within first-order terms in $f\gamma_m$. To determine ρ or η in the vicinity of the Brewster angle it is necessary to carry out all the calculations with an accuracy at least including second-order terms. We shall therefore dwell on the form of the equations in the "second approximation."

Since we are interested in the radiation far away from the transition layer, we can replace the discrete radiation centers by sources that are continuously distributed through the volume of the layer. This can be done if we assume that within the transition layer we add to the polarization (1) a supplementary polarization

$$\mathbf{P}' = \mathbf{P}'_0(z) e^{i(\omega t - \mathbf{k}\mathbf{r})} \quad (19)$$

It is easy to see that the radiation in the upper half space, due to such a polarization, is equivalent to the radiation, due to an infinitesimally thin layer, placed in the XY coordinate plane, provided the dipole moment (2) per unit area has the amplitude

$$\tilde{\tau}_0 = \int_0^l \mathbf{P}'_0(z) \exp \{-i(k_z + f_z)z\} dz, \quad (20)$$

where l is the thickness of the transition layer. In fact, the phase of the wave radiated from point 0 (Fig. 2) to point A is equal to $\omega t - f'r$. Let us isolate an infinitesimally thin layer of thickness dz and let us extend the radius vector r in the backward direction until it intersects this layer. The phase of the wave radiated from $B(R)$ to $A(r)$ is $\Phi = \omega t - f'(r+R) - \mathbf{k}\cdot\mathbf{R} = \omega t - f'r + (f'\cdot\mathbf{k})\cdot\mathbf{R}$, or since $f'_x = f_x = k_x$ and $f'_z = -f_z$, we have

$$\Phi = \omega t - f'r - (k_z - f_z)z.$$

Integrating with respect to z and taking the phases of the radiated waves into account, we obtain Eq. (20).

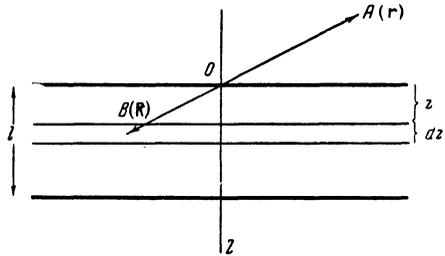


FIG. 2.

Analogously, the wave due to the supplementary polarization (19) radiated from the transition layer into the lower half space is equivalent to the radiation from an infinitely thin layer with a surface density of dipole moment (7), whereby

$$\tilde{\tau}'_0 = \int_0^l \mathbf{P}'_0(z) \exp \{-i(k_z - f_z)z\} dz. \quad (21)$$

The vector $\mathbf{P}'_0(z)$ depends on the frequency of the incident wave and also on the angle of incidence. However, rather than introduce these two arguments it is more convenient to use the arguments k_z and f_z . Let us expand the vector $\mathbf{P}'_0(z)$ in powers of k_z and f_z and let us terminate the expansion with the first-degree terms:

$$\mathbf{P}'_0(z) = \mathbf{P}'_{00}(z) - ik_z \mathbf{P}'_{01}(z) - if_z \mathbf{P}'_{02}(z),$$

where $\mathbf{P}'_{00}, \mathbf{P}'_{01}, \mathbf{P}'_{02}$ no longer depend on the frequency (neglecting dispersion) or on the angle of incidence. In this approximation we can replace (20) and (21) by

$$\vec{\tau}_0 = \mathbf{d} - ik_z \mathbf{q}_1 - if_z \mathbf{q}_2, \quad (22)$$

$$\vec{\tau}'_0 = \mathbf{d} - ik_z \mathbf{q}_1 + if_z \mathbf{q}_2,$$

$$\mathbf{d} = \int_0^l \mathbf{P}'_{00}(z) dz; \quad \mathbf{q}_1 = \int_0^l (\mathbf{P}'_{01} + z\mathbf{P}'_{00}) dz; \quad (23)$$

$$\mathbf{q}_2 = \int_0^l (\mathbf{P}'_{02} + z\mathbf{P}'_{00}) dz.$$

It is important to note that in the case of non-absorbing media the vectors \mathbf{d} , \mathbf{q}_1 , and \mathbf{q}_2 are all real. It is enough to prove that $\mathbf{P}'_{00}, \mathbf{P}'_{01},$ and \mathbf{P}'_{02} are real. This follows from the reversibility principle for non-absorbing media. Let us reverse at the instant $t=0$ the magnetic field in all points of space. Then, according to the reversibility principle, the electric field, and consequently also the polarization vector of the medium, will have at the instant t exactly the same values that they had at the instant $-t$. In particular, the supplementary polarization vector is obtained at the instant t by replacing t in (19) by $-t$. This gives

$$\mathbf{P}'(t, \mathbf{r}) = \mathbf{P}'_0(z, \mathbf{f}, \mathbf{k}) e^{-i(\omega t + \mathbf{k}\mathbf{r})}.$$

On the other hand, the above reversal of vector H also reverses the vectors \mathbf{f} , \mathbf{f}' , and \mathbf{k} . Therefore the value of $\mathbf{P}'(t, \mathbf{r})$ can be also obtained by reversing the signs of \mathbf{f} and \mathbf{k} in (19):

$$\mathbf{P}'(t, \mathbf{r}) = \mathbf{P}'_0(z, -\mathbf{f}, -\mathbf{k}) e^{i(\omega t + \mathbf{k}\mathbf{r})}.$$

Comparison with the preceding expression gives:

$$\mathbf{P}'_0(z, \mathbf{f}, \mathbf{k}) = \mathbf{P}'_0^*(z, -\mathbf{f}, -\mathbf{k}), \quad (24)$$

from which the above statement follows directly.

The vector \mathbf{d} can be considered as the dipole moment per unit area of the transition layer. On the other hand, the vectors \mathbf{q}_1 and \mathbf{q}_2 have the sense of quadrupole moments per unit area of the same layer. Thus the second-order theory differs from the first-order theory by further inclusion of the quadrupole radiation from the transition layer.

If we introduce along with parameters γ_x and γ_z the four new parameters:

$$\gamma'_m = q_{1m}/P_{0m}; \quad \gamma''_m = q_{2m}/P_{0m} \quad (m = x, z), \quad (25)$$

we obtain

$$a_m = \gamma_m - ik_z \gamma'_m - if_z \gamma''_m; \quad (26)$$

$$a'_m = \gamma_m - ik_z \gamma'_m + if_z \gamma''_m.$$

In the second approximation the properties of the transition layer are thus characterized by six parameters: $\gamma_x, \gamma_z, \gamma'_x, \gamma'_z, \gamma''_x$ and γ''_z . As to second order effects which may occur in experiments, this problem is exhaustively discussed in Ref. 4, and I will not dwell on it any further. These effects reduce to slight differences in the values of the polarization angle, of the principal angle of incidence, and of the Brewster angle, which according to the first-order theory should all equal each other.

11. If the thickness of the transition layer is large compared with the molecular dimensions and intermolecular distances (but naturally small compared with the wavelength), the transition layer can be phenomenologically characterized by the index of refraction. Equations (13) and (12) then become the Drude equations²; in second-order theory we obtain the Maclaurin equation.⁴ On the other hand, if this condition is not satisfied, such a method is not suitable, and it becomes necessary to know the molecular structure of the medium and of the transition layer before the parameters γ_m can be calculated. These data cannot be obtained by optical investigations alone. An investigation of the reflected field can hardly serve as a basis for single-value conclusions concerning the molecular structure of the reflecting medium and of the transition layer. An attempt to calculate γ_x and γ_z making very rough assumptions concerning this structure is found in reference 2 (let us note that parameters γ_m employed there differ in sign from the parameters used here). In this respect the theory requires further development and refinement.

⁴D. V. Sivukhin, J. Exptl. Theoret. Phys. (U.S.S.R.) 13, 361 (1943).