

THE THEORY OF LIQUID CRYSTALS.

BY C. W. OSEEN.

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In my monograph "*Die anisotropen Flüssigkeiten, Tatsachen und Theorien*" Fortschritte der Chemie, physikalischen Chemie und Physik. Bd. 20, 1929, I have among other things given a review of the results to which my theory had led by that time. The object of this article is to give a review of the results arrived at later on. In this connection I shall also enter upon some questions still unsolved.

1. On the Forces that give Rise to Liquid Crystals.

As far as I know, the physicists who have paid attention to liquid crystals have all taken it for granted that the forces which cause molecules to combine so as to form a liquid crystal, are not the chemical valence forces, but belong to the large group of molecular forces. This assumption has been verified by Vorländer,¹ whose statements are based upon the examination of a very large amount of chemical material. Now, as is well known, Debye and others have shown that the molecular forces may to a large extent be interpreted as electrostatic forces between the different nuclei and electrons that constitute molecules. Under these circumstances the question arises whether the forces between the molecules of a liquid crystal can also be supposed to be of an electrostatic nature. In my opinion the answer to this question must be negative.

Hitherto it has been possible in the theory of liquid crystals to ascribe to molecules rotational symmetry about an axis. For the determination of the positions and orientations of two molecules with regard to a definite co-ordinate system, then, ten quantities are needed. These may be, for either molecule, the three co-ordinates of the centre of gravity and the two angles that fix the direction of the axis of symmetry. Now there are ∞^6 equivalent co-ordinate systems. From this it follows that the positions of the two molecules in relation to each other must be determined by four quantities. These quantities may be chosen as the distance between the centres of gravity (r_{12}), the cosine of the angle that the axis of symmetry of the first molecule, when drawn in a given direction, forms with the line joining the centres of gravity, drawn from molecule 1 to molecule 2, the cosine of the angle that the axis of symmetry of the second molecule, when drawn in the same direction, forms with the line of connection just mentioned, drawn from molecule 2 to molecule 1, and the cosine of the angle that the two axes of symmetry form with each other. If by \mathbf{r}_{12} is meant the vector that is directed from the centre of gravity of molecule 1 to that of molecule 2, and if by $\mathbf{L}^{(1)}$ (or $\mathbf{L}^{(2)}$) is

¹ D. Vorländer, *Chemie der kristallinen Flüssigkeiten. Z. Kristallographie*, 79, 64.

meant a vector of unit length which has the same direction as the axis of symmetry of the molecule 1 (or 2), then these quantities will be :

$$r_{12}, \quad \mathbf{L}^{(1)} \cdot \frac{\mathbf{r}_{12}}{r_{12}}, \quad \mathbf{L}^{(2)} \cdot \frac{\mathbf{r}_{21}}{r_{21}}, \quad \mathbf{L}^{(1)}\mathbf{L}^{(2)} \quad . \quad . \quad . \quad (1)$$

$$(\mathbf{r}_{21} = -\mathbf{r}_{12}).$$

It is at once obvious, however, that these four quantities do not yield a single-valued determination of the positions of the two molecules relative to each other, but only a two-valued. The angle between the two planes which cut one another along the line joining the two centres of gravity and which contain each one of the two axes of symmetry, is determined as to its magnitude, but not with regard to its sign. For a single-valued determination of the positions of the two molecules relative to one another a further quantity is necessary, and this may be chosen to be :

$$\mathbf{L}^{(1)} \times \mathbf{L}^{(2)} \cdot \frac{\mathbf{r}_{12}}{r_{12}} \quad . \quad . \quad . \quad . \quad (2)$$

The square of this quantity, but not the quantity itself can be rationally expressed in the quantities (1). Accordingly it is only the sign of this quantity that introduces anything new.

From this it will be seen that there are two different kinds of liquid crystals. On the one hand there is a group characterised by the fact that the potential energy of two molecules may be expressed as a uniform function of the four quantities (1). On the other hand there is a group in the case of which such a description of the potential energy of two molecules is not possible, whereas this energy may be expressed as a uniform function of the four quantities (1) and of the quantity (2). It is one of the most important results of the theory, that the last-mentioned group of liquid crystals is identical with the group of cholesterine-nematic substances. From this must be inferred that the molecular forces that give rise to liquid crystals, are of such nature that the potential energy of two molecules may depend on the sign of the quantity (2); in other words, of such nature that it may occur that the potential energy of two molecules is not invariant for a reflection at a plane in space. This requirement excludes electrostatic forces. Electrostatic energy, which depends only on the distances between the charges, cannot change when the whole system is reflected at a plane, since at this reflection all charges and all distances remain unchanged.

If the force between two molecules in a liquid crystal of the cholesterine-nematic type cannot be regarded as purely electrostatic, then it will be an obvious conclusion that the non-electrostatic part of the potential energy of two molecules is of a magnetic nature. This theory is corroborated by the fact that the magnetic force actually has a directing effect upon anisotropic molecules. From the point of view of atomic theory, too, there are reasons to believe that two molecules can, among other things, also exert magnetic forces on each other. If a molecule has an axis of symmetry, the total moment of momentum of the molecule, which is composed of the mechanical moment of momentum and the spin-moment of the electrons, must have a component along the axis of symmetry, which for every state with a fixed energy value has also a fixed value. In the case of liquid crystals only those states are at present taken into consideration where the moment of momentum coincides with the axis of symmetry. Evidently two molecules con-

structed in this way will exert magnetic forces on each other. On closer examination, however, it becomes evident that even in this way we do not obtain a satisfactory interpretation of the molecular forces that appear in liquid crystals. A moment of momentum along the axis of symmetry may, so far as the physical effects are concerned, to a first approximation be substituted by a magnetic moment in the direction of the axis of symmetry. According to the interpretation sketched above the cholesterine-nematic substances must necessarily be paramagnetic. But, so far as we know, this is not the case. Besides, even in that case we should not be any nearer the solution of the problem. For magneto-static energy, like electrostatic, is invariant for reflection at a plane.

We have seen that, according to the theory, cholesterine-nematic substances are characterised by the fact that the potential energy of two molecules is not invariant for reflection. This agrees well with Vorlander's proposition that cholesterine-nematic substances are always so constructed that the molecule contains at least one asymmetric carbon atom.² Now, as is well known, the asymmetric carbon atom is characterised by optical rotary power. There naturally arises the question whether this effect does not suggest where the solution of the problem is to be found. Now the theories of optical rotary power that were started at the same time by M. Born and myself, both show that the essential fact here is the difference of phase that will exist between the different vibrators of the active molecule. It may be asked, then, whether the finite velocity with which the force is transmitted does not effect the interaction between two molecules. If we imagine two molecules which contain electrons rotating about axes in them, the interaction between them may, to a first approximation, be described by means of electric and magnetic dipoles, quadrupoles, and so on. But in an exact description the finite velocity of propagation of the force must also be taken into consideration. Here it is possible to understand how, in the case of cholesterine-nematic substances, the potential energy of two molecules can undergo a change on reflection at a plane. If this is correct, the solution of the problem of liquid crystals is to be found in a chapter of atomic physics that is still unwritten.

2. On Singularities of Structure in Nematic Substances.

If we suppose that the force between two molecules decreases rapidly enough as the distance increases, and if further we suppose that the density of the substance considered is constant, we shall find that the laws holding for the orientation of the axes of the molecules may be summed up in the requirement that an expression of the form

$$\int \rho^2 \{ K_1 \mathbf{L} \text{ rot } \mathbf{L} + K_{11} (\mathbf{L} \text{ rot } \mathbf{L})^2 + K_{22} (\text{div } \mathbf{L})^2 + K_{33} ((\mathbf{L} \nabla) \mathbf{L})^2 + 2K_{12} \text{div } \mathbf{L} \cdot \mathbf{L} \text{ rot } \mathbf{L} \} d\omega, \\ (\text{where } d\omega = dx_1 dx_2 dx_3),$$

will have an extreme value. In the case of nematic substances $K_{12} = 0$, $K_{22} = K_{33}$. In this case we obtain a solution:

$$\mathbf{L}_1 = \cos \phi, \quad L_2 = \sin \phi, \quad L_3 = 0, \\ \phi = \frac{K_1 x_3}{2(K_{11} - K_{33})} + f(x_1, x_2), \\ \frac{\partial^2 f}{\partial x_1^2} + \frac{\partial^2 f}{\partial x_2^2} = 0.^3$$

² *Loc. cit.*, p. 84.

³ Cf. my above-mentioned monograph, *Die anisotropen Flüssigkeiten*, p. 49.

If we put: $f = \pm \psi + \text{constant}$, $\pm \frac{1}{2}\psi + \text{constant}$, $\psi = \tan^{-1} \frac{x_2}{x_1}$, we obtain structures with a single line $x_1 = x_2 = 0$, which must optically show the phenomena described by Lehmann under the names of whole *Kernpunkte* ($+\psi$), whole *Konvergenzpunkte* ($-\psi$), half *Kernpunkte* ($+\frac{1}{2}\psi$), half *Konvergenzpunkte* ($-\frac{1}{2}\psi$). But as pointed out in the before-mentioned monograph, the theory also meets with difficulties. The energy of such a structure would be infinitely great, and, besides, it seems difficult to see why structures of the type:

$$f = k \log r$$

should not also be found.

We obtain a solution of these difficulties, if we abandon the assumption that the density is constant. Obviously the theory thus obtained will be far more complicated than before, and the formulæ are indeed so long that they cannot be reproduced here. For the new theory it is a matter of vital importance that the thermodynamic function which is to take a minimum value is the free energy. The entropy of an æolotropic substance is not known empirically. According to the theory every state of an element characterised by the distribution of the axes of the molecules in the different directions in space has its entropy. What is here to be taken into consideration, is, however, the most probable distribution and the entropy corresponding to this. Per unit of mass this entropy may be expressed as a function of the density and the absolute temperature $F(\rho, T)$. Hence we may write:

$$S = \int F(\rho, T) \rho d\omega.$$

If we develop this theory by the methods of mathematical physics, we shall obtain a system of three differential equations for the determination of the direction of the axis and of the density. If we specially want to examine plane structures, *i.e.*, structures in which, for instance, $L_3 = 0$, and L_1 and L_2 only depend on x_1, x_2 , the equations are reduced to two. In order that these equations may represent a nematic substance it is necessary to impose certain conditions on the coefficients. If we do so and put $L_1 = \cos \phi$, $L_2 = \sin \phi$, we obtain for the determination of ϕ and ρ the equations:

$$\Delta \phi - \frac{1}{\rho} \left(\frac{\partial \phi}{\partial x_1} \frac{\partial \rho}{\partial x_1} + \frac{\partial \phi}{\partial x_2} \frac{\partial \rho}{\partial x_2} \right) = 0,$$

$$C_2^{(0)} \Delta \rho + 2C^{(0)} \rho + 2K_{22} \rho \left[\left(\frac{\partial \phi}{\partial x_1} \right)^2 + \left(\frac{\partial \phi}{\partial x_2} \right)^2 \right] = \beta + 2m^2 T \left(F(\rho, T) + \rho \frac{\partial F}{\partial \rho} \right).$$

Obviously these equations may be satisfied by putting

$$\phi = k\psi, \psi = \tan^{-1} \frac{x_2}{x_1}, \rho = f(r) = f(\sqrt{x_1^2 + x_2^2}).$$

For the determination of the function f we obtain an ordinary differential equation of the second degree. For this differential equation $r = 0$ is a singular point. By means of successive approximations we can find a solution of this equation which fulfils, for $r = 0$, the conditions $f = 0$, $f' = 0$. Our system of equations has, however, no solutions of the form $\phi = k \log r$, $\rho = f(\psi)$. The two above-mentioned difficulties inherent in the theory of the singularities of structure of nematic substances, are thus removed.

A singular line in a nematic substance, the direction of which coincides with that of view, gives rise to a *Kernpunkt* or *Konvergenzpunkt* and so

forth. If its direction is perpendicular to the direction of view, it gives rise to other phenomena. It appears, then, that we have to distinguish two cases. It may occur that the image of the singular line is on both sides surrounded by a half-shade, owing to the fact that one of the two rays in the aeolotropic substance, to which the incident ray has given rise, undergoes a deviation in the neighbourhood of the singular line, which prevents it from penetrating into the microscope. It may, however, also occur that there appears no such half-shade. These facts give rise to the question whether the difference between singular lines with a half-shade and such lines without a half-shade has any connection with the distinction between whole *Kernpunkte*, whole *Konvergenzpunkte*, half *Kernpunkte*, half *Konvergenzpunkte*. In order to solve this problem the author has made a geometrical-optical examination of the progress of the rays in the vicinity of a singular line. The result of this examination is that in all these cases the extraordinary ray undergoes a deviation in the vicinity of the singular line, but that this deviation is generally small, if the singular line is of the type "whole *Kernpunkt*" ($\phi = \psi + \text{Konst.}$), whereas it is always great, if the singular line belongs to any of the other types. Though this result is to be received with a certain reserve, as the methods of geometrical optics are not applicable when a ray undergoes sharp deviations, yet it seems probable that a singular line without a half-shade is always of the type "whole *Kernpunkt*" ($\phi = \psi + \text{Konst.}$), and that a singular line with a half-shade is in general of another type than this.

3. On the Iridescent Structures of Cholesterine-nematic Substances.

A substance, for which $K_{12} = 0$, $K_1 \neq 0$ has a tendency to appear in a structure that may be characterised as helicoidally-twisted. Mathematically such a structure may be described by means of the simple formulæ:

$$L_1 = \cos \alpha x_3, L_2 = \sin \alpha x_3, L_3 = 0.$$

Here \mathbf{L} is as usual a vector of unit length which indicates the direction of the molecular axis. In my monograph and in the discussion arranged by Professor Ewald and published in *Z. Kristallographie*, I have given some information on the remarkable optical qualities of such structures. Here I shall give a more detailed account.

If the dielectric coefficient is regarded as a tensor with the components

$$\epsilon_{jk} = (\epsilon_3 - \epsilon_1)L_jL_k + \delta_{jk}\epsilon_1 (\delta_{jk} = 0, \text{ if } j \neq k, = 1, \text{ if } j = k)$$

Maxwell's equations takes the form:

$$\text{rot } \mathbf{H} = \frac{\epsilon_3 - \epsilon_1}{c} \left(\mathbf{L} \frac{\partial \mathbf{E}}{\partial t} \right) \mathbf{L} + \frac{\epsilon_1}{c} \frac{\partial \mathbf{E}}{\partial t}, \quad \text{rot } \mathbf{E} = - \frac{1}{c} \frac{\partial \mathbf{H}}{\partial t}.$$

We consider first the solutions of these equations that correspond to waves propagated in the direction of the x_3 axis. It appears that these solutions find their simplest expressions if, in addition to the fixed x_1 , x_2 axes, we introduce a moving (x'_1 , x'_2) system so chosen that the x'_1 axis has at every point the same direction as the vector \mathbf{L} and that, consequently, the x'_2 axis is perpendicular to \mathbf{L} and to the x_3 axis. We put:

$$\frac{1}{2} \left[\alpha^2 + \frac{\omega^2}{2c^2} (\epsilon_1 + \epsilon_3) \right] = a, \quad \frac{1}{4} \left[\alpha^2 - \frac{\omega^2}{2c^2} (\epsilon_1 + \epsilon_3) \right]^2 - \frac{\omega^4}{16c^4} (\epsilon_1 - \epsilon_3)^2 = b,$$

$$\Omega_1 = \sqrt{a + \sqrt{b}} + \sqrt{a - \sqrt{b}}, \quad \Omega_2 = \sqrt{a + \sqrt{b}} - \sqrt{a - \sqrt{b}},$$

and prescribe that, if $b > 0$, in which case Ω_1 and Ω_2 are real, the roots shall be taken with their positive values, and, if $b < 0$, in which case Ω_1 is real and Ω_2 purely imaginary, the signs shall be so chosen that Ω_2 takes the form $-ki$ ($k \geq 0$), whereas the real part of Ω_1 is still positive. If then we put:

$$-\Omega_1 = \Omega^{(1)}, \quad -\Omega_2 = \Omega^{(2)}, \quad \Omega_1 = \Omega^{(3)}, \quad \Omega^2 = \Omega^{(4)}$$

we obtain the following expressions for the components of the strengths of the electric and the magnetic fields in the moving system just mentioned.

$$E_1' = \frac{1}{2} \left[(\Omega^{(j)} + \alpha)^2 - \frac{\omega^2 \epsilon_1}{c^2} \right] e^{i(\omega t + \Omega^{(j)} x_3)},$$

$$H_1' = \frac{i}{2} \left[\frac{c}{\omega} (\Omega^{(j)} + \alpha)^2 (\Omega^{(j)} - \alpha) + \frac{\omega}{c} (\epsilon_1 \alpha - \epsilon_3 \Omega^{(j)}) \right] e^{i(\omega t + \Omega^{(j)} x_3)},$$

$$E_2' = \frac{i}{2} \left[(\Omega^{(j)} + \alpha)^2 - \frac{\omega^2 \epsilon_3}{c^2} \right] e^{i(\omega t + \Omega^{(j)} x_3)},$$

$$H_2' = -\frac{1}{2} \left[\frac{c}{\omega} (\Omega^{(j)} + \alpha)^2 (\Omega^{(j)} - \alpha) + \frac{\omega}{c} (\epsilon_3 \alpha - \epsilon_1 \Omega^{(j)}) \right] e^{i(\omega t + \Omega^{(j)} x_3)}.$$

A discussion of these expressions already gives valuable information on the strange optical qualities of helicoidally-twisted strata. If $b > 0$, which occurs, if ω lies outside the interval $c\alpha/\sqrt{\epsilon_1}$, $c\alpha/\sqrt{\epsilon_3}$, all $\Omega^{(j)}$, as observed are real. Our formulæ show that in this case there are two elliptically polarised waves in either direction. Their velocities of propagation are ω/Ω_1 and ω/Ω_2 . During the propagation the ellipse of vibration rotates thus that its principal axes always fall along the x_1' and x_2' axes. If, on the contrary, $b < 0$, that is, if ω lies within the interval $c\alpha/\sqrt{\epsilon_1}$, $c\alpha/\sqrt{\epsilon_3}$, Ω_2 and consequently $\Omega^{(2)}$ and $\Omega^{(4)}$ are purely imaginary. The corresponding waves are, referred to the moving co-ordinate system, to be taken as standing waves. These waves, too, are elliptically polarised so that the principal axes of the ellipse of vibration fall along the x_1' and x_2' axes. The amplitude increases, for the wave [4], exponentially with x_3 . For the wave [2] the amplitude decreases exponentially with x_3 . Within the interval $c\alpha/\sqrt{\epsilon_1}$, $c\alpha/\sqrt{\epsilon_3}$ there will generally be a region where $kd \gg 1$, d being the thickness of the æolotropic stratum. This region is called the interior part of the interval. In this part of the spectrum there is then a wave the upper amplitude of which is enormous compared to the lower, and another wave the lower amplitude of which is enormous compared to the upper. The above-mentioned propositions are of fundamental importance for the study of the reflection of a wave in a cholesterine-nematic stratum of iridescent structure, and of its passage through this stratum. Let us imagine that a wave that comes from the region $x_3 < 0$, meets the æolotropic substance in the plane $x_3 = 0$ and leaves it in the plane $x_3 = d$. The requirement that in the plane $x_3 = d$ there shall be no incident radiation, gives rise to two linear connections between the vector components $E_1(d)$, $E_2(d)$, $H_1(d)$, $H_2(d)$. Let us now specially consider a wave that may in the interior of the substance be represented by the formulæ:

$$E_1' = \sum_{j=1, 2, 4} A_j \frac{1}{2} \left[(\Omega^{(j)} + \alpha)^2 - \frac{\omega^2 \epsilon_1}{c^2} \right] e^{i(\omega t + \Omega^{(j)} x_3)},$$

$$E_2' = \sum_{j=1, 2, 4} \frac{1}{2} \left[(\Omega^{(j)} + \alpha)^2 - \frac{\omega^2 \epsilon_3}{c^2} \right] e^{i(\omega t + \Omega^{(j)} x_3)}.$$

The conditions just mentioned, applicable to $x_3 = d$, make it possible to express A_1 and A_4 in the forms $A_2 e^{-kd} F_1(\alpha, \omega, C, \epsilon_1, \epsilon_3)$ and $A_2 e^{-kd} F_2(\alpha, \omega, C, \epsilon_1, \epsilon_3)$, respectively. Hence it will be seen that A_1 and A_4 belong, roughly speaking, to the same order of magnitude as $A_2 e^{-kd}$. In the interior of the interval $c\alpha/\sqrt{\epsilon_1}$, $c\alpha/\sqrt{\epsilon_3}$ they are infinitesimal compared to A_2 . Hence in the vicinity of the plane $x_3 = 0$ we obtain with very great accuracy:

$$E_1' = A_2 \frac{1}{2} \left[(\alpha - \Omega_2)^2 - \frac{\omega^2 \epsilon_1}{c^2} \right] e^{i\omega t - kx_3}.$$

From these expressions for E_1' , E_2' , H_1' , H_2' we can now determine the corresponding expressions $E_1^{(i)}$, $E_2^{(i)}$, $H_1^{(i)}$, $H_2^{(i)}$ for the incident wave, as well as the components $E_1^{(r)}$, $E_2^{(r)}$, $H_1^{(r)}$, $H_2^{(r)}$ of the reflected wave. In both cases we obtain comparatively simple expressions showing that both waves are elliptically polarised. We can also determine the components, $E_1^{(g)}$. . . of the wave coming from the plane $x_3 = d$. It appears, as may also be concluded from what is said above, that the amplitude of this wave bears to that of the incident wave a proportion of the order of magnitude e^{-kd} : 1. The incident wave thus undergoes a total reflection, when it meets the æolotropic stratum.

It is interesting to examine more strictly the expressions for the incident and the reflected wave. We refer them, of course, to the fixed (x_1, x_2, x_3) system. In order to obtain the simplest possible formulæ we put:

$$E_1^{(i)} + iE_2^{(i)} = E_1^{*(i)}, \quad E_1^{(i)} - iE_2^{(i)} = E_2^{*(i)},$$

and in the same way:

$$H_1^{(i)} + iH_2^{(i)} = H_1^{*(i)}, \quad H_1^{(i)} - iH_2^{(i)} = H_2^{*(i)}.$$

We have in these denominations:

$$E_1^{*(i)} = -iH_1^{*(i)} = \frac{\omega}{4c}(\epsilon_3 - \epsilon_1) \left(\frac{\omega}{c} - \alpha + \Omega_2 \right) A_2 e^{i\omega(t - x_3/c)},$$

$$E_2^{*(i)} = iH_2^{*(i)} = \frac{c}{2\omega} \left[(\alpha - \Omega_2)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] \left(\frac{\omega}{c} + \alpha + \Omega_2 \right) A_2 e^{i\omega(t - x_3/c)},$$

$$E_1^{*(r)} = iH_1^{*(r)} = \frac{\omega}{4c}(\epsilon_3 - \epsilon_1) \left(\frac{\omega}{c} + \alpha - \Omega_2 \right) A_2 e^{i\omega(t - x_3/c)},$$

$$E_2^{*(r)} = -iH_2^{*(r)} = \frac{c}{2\omega} \left[(\alpha - \Omega_2)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] \left(\frac{\omega}{c} - \alpha - \Omega_2 \right) A_2 e^{i\omega(t - x_3/c)}.$$

The quantities $\Omega^{(j)}$ ($j = 1, 2, 3, 4$) are roots of the equation:

$$\Omega^4 - 4a\Omega^2 + 4b = 0.$$

This equation may also be written:

$$\left[(\alpha - \Omega)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] \left[(\alpha + \Omega)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] = \frac{\omega^4}{4c^4}(\epsilon_3 - \epsilon_1)^2.$$

If Ω_2 is purely imaginary, it follows that

$$\left| \frac{\frac{\omega^2}{2c^2}(\epsilon_3 - \epsilon_1)}{(\alpha + \Omega_2)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3)} \right| = 1.$$

There is then such a number t_0 that :

$$\begin{aligned}\frac{\omega^2}{2c^2}(\epsilon_3 - \epsilon_1) &= \left[(\alpha + \Omega_2)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] e^{-i t_0} \\ &= \left[(\alpha - \Omega_2)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] e^{+i t_0}.\end{aligned}$$

On account of this our expressions for $E_1^{*(r)}$ and $E_2^{*(r)}$ may be written :

$$E_1^{*(r)} = \frac{c}{2\omega} \left[(\alpha + \Omega_2)^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] \left(\frac{\omega}{c} + \alpha - \Omega_2 \right) A_2 e^{i\omega(t - t_0 + x_3/c)},$$

$$E_2^{*(r)} = \frac{\omega}{4c} (\epsilon_3 - \epsilon_1) \left(\frac{\omega}{c} - \alpha - \Omega_2 \right) A_2 e^{i\omega(t - t_0 + x_3/c)}.$$

If we write $E_1^{(i)}$ and $E_2^{(i)}$:

$$E_1^{(i)} = R \{ a e^{i\omega(t - x_3/c) + i\alpha} \} = a \cos \{ \omega(t - x_3/c) + \alpha \},$$

$$E_2^{(i)} = R \{ i b e^{i\omega(t - x_3/c) + i\beta} \} = -b \sin \{ \omega(t - x_3/c) + \beta \},$$

$E_1^{(r)}$ and $E_2^{(r)}$ will take the form :

$$E_1^{(r)} = R \{ a e^{i\omega(t - t_0 + x_3/c) - i\alpha} \} = a \cos \{ \omega(t - t_0 + x_3/c) - \alpha \},$$

$$E_2^{(r)} = R \{ -i b e^{i\omega(t - t_0 + x_3/c) - i\beta} \} = b \sin \{ \omega(t - t_0 + x_3/c) - \beta \}.$$

The formulæ for $E_1^{(r)}$ and $E_2^{(r)}$ will be transformed into the formulæ for $E_1^{(i)}$, $E_2^{(i)}$, if $t_0 - t$ is substituted for t . This proves that the ellipses of vibration of the incident and the reflected wave have exactly the same form and the same size. With regard to the different directions of propagation it also shows that in both cases the vibration, regarded by an observer towards whom the wave advances, takes place in the same direction. There is then no difference between the two waves but the opposite directions of propagation and a difference of phase.

We have found that there exists a wave of light, elliptically polarised in a given manner, which is subjected to a total reflection in the helicoidally twisted structure, at which reflection the reflected wave has, but for a difference of phase, exactly the same character as the incident wave. In order to master the problem of reflection we must also take another wave into consideration. We obtain it by examining a wave which in the æolotropic substance is determined by the formulæ :

$$E_1' = \sum_{j=1,3,4} B_j \frac{1}{2} \left[(\Omega^{(j)} + \alpha)^2 - \frac{\omega^2 \epsilon_1}{c^2} \right] e^{i(\omega t + \Omega^{(j)} x_3)},$$

$$E_2' = \sum_{j=1,3,4} B_j \frac{1}{2} \left[(\Omega^{(j)} + \alpha)^2 - \frac{\omega^2 \epsilon_3}{c^2} \right] e^{i(\omega t + \Omega^{(j)} x_3)}.$$

In this case I shall only mention the result of the computation. We put :

$$-1 + \frac{c^2}{\omega^2} \frac{\Omega_1^2 - \Omega_2^2}{\alpha(\epsilon_1 + \epsilon_3 - 2)} \left(\frac{\omega}{c} + \alpha + \Omega_2 \right) = M_1,$$

$$\frac{\Omega_2}{\Omega_1} + \frac{c^2}{\omega^2} \frac{\Omega_1^2 - \Omega_2^2}{\alpha \Omega_1 (\epsilon_1 + \epsilon_3 - 2)} \left(\frac{\omega}{c} + \alpha + \Omega_2 \right) \left(\frac{\omega}{c} + \alpha \right) = M_2,$$

and :

$$\begin{aligned}M_1 \cos(d\Omega_1) + i M_2 \sin(d\Omega_1) &= N_1, \\ M_2 \cos(d\Omega_1) + i M_1 \sin(d\Omega_1) &= N_2.\end{aligned}$$

With this notation we have :

$$\begin{aligned}
 E_1^{*(i)} &= \frac{\omega}{4c}(\epsilon_3 - \epsilon_1) \left\{ \left(\frac{\omega}{c} + \alpha \right) N_1 - \Omega_1 N_2 \right\} B_4 e^{i\omega(t - x_3/c)}, \\
 E_2^{*(i)} &= \frac{c}{2\omega} \left\{ \left[\left(\frac{\omega}{c} + \alpha \right) \left(\alpha^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right) + \left(\frac{\omega}{c} - \alpha \right) \Omega_1^2 \right] N_1 \right. \\
 &\quad \left. + \Omega_1 \left[\frac{2\alpha\omega}{c} - \alpha^2 + \Omega_1^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] N_2 \right\} B_4 e^{i\omega(t - x_3/c)}, \\
 E_1^{*(r)} &= \frac{\omega}{4c}(\epsilon_3 - \epsilon_1) \left\{ \left(\frac{\omega}{c} - \alpha \right) N_1 + \Omega_1 N_2 \right\} B_4 e^{i\omega(t + x_3/c)}, \\
 E_2^{*(r)} &= \frac{c}{2\omega} \left\{ \left[\left(\frac{\omega}{c} - \alpha \right) \left(\alpha^2 - \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right) + \left(\frac{\omega}{c} + \alpha \right) \Omega_1^2 \right] N_1 \right. \\
 &\quad \left. + \Omega_1 \left[\frac{2\alpha\omega}{c} + \alpha^2 - \Omega_1^2 + \frac{\omega^2}{2c^2}(\epsilon_1 + \epsilon_3) \right] N_2 \right\} B_4 e^{i\omega(t + x_3/c)}.
 \end{aligned}$$

For the wave that emerges from the plane $x_3 = d$ we finally obtain :

$$\begin{aligned}
 E_1^{*(g)} &= \frac{\epsilon_3 - \epsilon_1}{2\alpha(\epsilon_1 + \epsilon_3 - 2)} (\Omega_1^2 - \Omega_2^2) \left(\frac{\omega}{c} + \alpha + \Omega_2 \right) B_4 e^{i\omega(t - x_3/c) + i\alpha d}, \\
 E_2^{*(g)} &= -(\Omega_1^2 - \Omega_2^2) \left\{ 1 + \frac{1}{2\alpha} \left(\frac{\omega}{c} + \alpha + \Omega_2 \right) \right. \\
 &\quad \left. - \frac{c^2}{\omega^2 \alpha (\epsilon_1 + \epsilon_3 - 2)} \left(\frac{\omega}{c} + \alpha + \Omega_2 \right) \left[\left(\frac{\omega}{c} + \alpha \right)^2 - \Omega_1^2 \right] \right\} B_4 e^{i\omega(t - x_3/c) - i\alpha d}.
 \end{aligned}$$

From these formulæ it will be seen that if a plane wave, advancing in the direction of the normal, meets a helicoidally-twisted structure, the intensity of the reflected wave as well as that of the passing one must in general be a periodic function of $\Omega_1 \cdot d$. Now, if the two bounding surfaces of the ælotropic substance are not exactly parallel, but form a small angle, then both in the reflected light and in that which has passed there will appear dark stripes at constant distances from one another, all parallel to the line of intersection of the bounding surfaces. If, however by means of an elliptical polariser we cause the incident ray to be of the first type considered above, these dark stripes will not appear. The conclusions here drawn from the theory agree perfectly with the observations that have been made with relation to the so-called stripes of Grandjean. It cannot very well be assumed that this agreement is due to a mere chance. There will then be no reasons why the so-called planes of Grandjean should really exist.

We have mentioned above some consequences of the theory which agree with known facts. I shall now point out some consequences, the correctness of which is questionable. According to the statements of our experimentalists, the iridescent structures of cholesterine-nematic substances have an optical rotary power which in certain cases is very great. Now the above formulæ show that in the interior of the region $c\alpha/\sqrt{\epsilon_1}$, $c\alpha/\sqrt{\epsilon_3}$, there is no optical rotary power in the usual sense of the word. Every wave which, having passed through the ælotropic stratum, emerges from the plane $x_3 = d$, must be obtainable through a linear combination of those emergent waves that correspond to the two types of elliptically polarised waves in the structure. We know, however, that the former type, which gives rise to a total reflection, does not give any perceptible wave emergent from the plane $x_3 = d$. The ellipse of

vibration of the wave that emerges from this plane has then exactly the same form, and the principal axes of this ellipse have exactly the same directions, whatever the incident wave may be. Hence a rotary power in the usual sense of the word is out of the question. As we see, the argument stated here is based on the existence of an elliptically polarised wave that on incidence in the substance is subjected to a total reflection. Now this consequence of the theory agrees with facts. Then it seems probable that the conclusion that has here been drawn from this consequence, also agrees with facts. According to Friedel the optical rotary power actually changes its sign for a certain wave-length, taking then an infinitely large value. But in the place of the spectrum where this infinite rotary power was supposed to be found, no rotation at all can be shown, because, according to Friedel, the emergent wave is circularly polarised. If we leave Friedel's theory of the infinitely great rotary power out of the question and keep to what can be directly observed, the theory seems to agree with facts on this point too.

We shall now leave the region $\alpha\lambda/\sqrt{\epsilon_1}$, $\alpha\lambda/\sqrt{\epsilon_3}$ and consider the parts of the spectrum for which ω falls outside this interval. The result of the theory is that in these regions of the spectrum there is no rotary power in the usual sense of the word. It is true that an incident, plane-polarised wave that passes through the stratum, is changed into an elliptically-polarised wave whose greater axis of vibration forms an angle with that of the incident wave, but this angle does not depend linearly on the thickness of the stratum, but in a far more complicated way. If, as the experimentalists state, the iridescent structure of a cholesterine-nematic substance really has an optical rotary power of the usual kind, then, on this point, the theory in its present state disagrees with facts. It must be added, however, that our experimentalists seem to have taken it for granted that, when a rotation of the plane of polarisation is perceptible, there will also be a rotary power in the usual sense of the word. A careful examination of phenomena of this kind is at present one of the most pressing needs with reference to liquid crystals.

An account has been given of the laws which hold for the perpendicular incidence of a wave in the iridescent structure of a cholesterine-nematic substance. But it is also very interesting to study the reflection and refraction of a non-perpendicular wave. It is found empirically that, in the case of a non-perpendicular wave also, there is a region in the spectrum in which a component of the wave undergoes a total reflection. As the incidence of the wave becomes oblique instead of perpendicular, the region of total reflection is displaced towards the violet part of the spectrum. It is this fact, above all, which the theory has to explain. The investigation shows that an incident wave, for which the components of the electromagnetic field are proportional to :

$$e^{i\omega\left(t - \frac{\beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3}{c}\right)} \quad (\text{where } \beta_1^2 + \beta_2^2 + \beta_3^2 = 1)$$

gives rise to a wave of the type :

$$E_j = e^{i\omega\left(t - \frac{\beta_1 x_1 + \beta_2 x_2}{c}\right)} e^{\frac{i h \omega x_3}{c}} F_j(x_3),$$

(where $j = 1, 2, 3$).

The functions $F_j(x_3)$ are periodic with the period $2\pi/\alpha$. They satisfy the conditions :

$$F_j\left(x_3 + \frac{\pi}{\alpha}\right) = -F_j(x_3).$$

h is a transcendent function of c , ω , α , E_1 , E_3 and $\beta^2 = \beta_1^2 + \beta_2^2$. In order that a total reflection may occur, it is necessary that h should have a complex value. This is, as we know, the case for $\beta = 0$ if ω falls in the interval $c\alpha/\sqrt{\epsilon_1}$, $c\alpha/\sqrt{\epsilon_3}$, i.e., if the wave-length lies within the limits :

$$\frac{2\pi}{\alpha}\sqrt{\epsilon_1} \quad \text{and} \quad \frac{2\pi}{\alpha}\sqrt{\epsilon_3}.$$

Now, if $\beta^2 > 0$ but so small that β^4 may be neglected compared with unity these limits will be displaced to :

$$\frac{2\pi}{\alpha}\sqrt{\epsilon_1} - \beta^2 \quad \text{and} \quad \frac{2\pi}{\alpha}\sqrt{\epsilon_3 - \beta^2 \frac{\epsilon_1 + \epsilon_3}{2\epsilon_1}}.$$

We see that the theory explains why the displacement takes place towards the side of the short waves. Besides the region of total reflection, which remains for the case of $\beta = 0$, there is, however, an infinite number of other such regions. They all lie on the violet side of the region first found. Now, if we pass from perpendicular incidence to oblique, it will not only occur that the region of reflection already present is displaced towards the violet part of the spectrum, but new colours will appear on the violet side of this region.

One of the most remarkable properties known as to the iridescent structures of cholesterine-nematic substances, is the fact that their colour depends both on the angle that the incident light forms with the normal and on that which the reflected light forms with this normal. By this it seems that one of the oldest optical laws which exists, namely that according to which the angle of reflection is determined by the angle of incidence and equal to this, does not hold for these structures. The theory developed does not explain this fact. A theory of it may, however, be given, which is based upon the properties of helicoidally-twisted strata. We only have to remember that no body has an absolutely plane surface and that, if two plane surfaces existed, it would never be possible to adjust them so as to be perfectly parallel to each other. From this it follows that the theory concerning the iridescent strata of cholesterine-nematic substances must also take into account the case where a bounding surface divides the structure obliquely, so that the preparation takes the form of a wedge. In this case other phenomena will appear. If, for the sake of simplicity, we assume the stratum to be infinitely thick, the most important result of the theory can be expressed as follows : A plane polarised wave incident on the plane bounding surface of a helicoidally twisted stratum, the normal of which makes an angle with the axis of the stratum, gives rise to a reflected wave, which is composed of an infinite number of plane waves with different directions of propagation. The relation between the angle of incidence and that of reflection is therefore not single-valued. An infinite number of angles of reflection correspond to a given angle of incidence.

4. On the Motion of Æolotropic Liquids.

It has been known for some long time that many liquids do not move in accordance with the Navier-Stokes equations of motion. In 1925 Kruyt summed up the investigations then made and arrived at the result that disperse-systems with globular particles follow the law given by Poiseuille which holds for a liquid passing through a tube, whereas

deviations from this law appear in the case of systems with non-spherical particles. Further material since 1925 has confirmed the correctness of Krüy's conclusion. Under these circumstances Dr. A. Anzelius⁴ set himself the task of formulating laws of motion valid for a liquid with oblong molecules. We shall give below a survey of the theory of Anzelius and the results to which it has led hitherto.

Anzelius assumes that in a liquid consisting of oblong molecules with symmetry of rotation, the direction of the axis of a molecule will vary continuously from point to point, even if the liquid moves, so that at a certain moment there will be a certain direction of axes for every element of volume. Under these circumstances we can no longer suppose that Navier's and Stoke's assumptions concerning the tensor of viscosity

$$\tau_{jk} = -p\delta_{jk} + \mu\left(\frac{\partial u_j}{\partial x_k} + \frac{\partial u_k}{\partial x_j}\right)$$

holds good. Instead, we must assume that this tensor will depend also on the direction of the axis of the molecule, *i.e.*, on \mathbf{L} . On the presumption that a dissipation-function exists, Anzelius determines the form of this function. His results may be summed up in the statement that the dissipation function, F , must have the form:

$$A_1 \sum_{\substack{j,k=1,2,3}} D_{jk}^2 + A_2 \sum_k \left(\sum_j L_j D_{jk} \right)^2 + A_3 \left(\sum_{j,k} L_j L_k D_{jk} \right)^2 \\ + 2A_4 \sum_{j,k} L_j L_k D_{jk} \sum_m D_{mm} + A_5 \left(\sum_m D_{mm} \right)^2,$$

where

$$D_{jk} = \frac{1}{2} \left(\frac{\partial u_j}{\partial x_k} + \frac{\partial u_k}{\partial x_j} \right),$$

and $A_1 \dots A_5$ are constants. From F we obtain the tensor of viscosity by the formulæ:

$$\tau_{jk} = -p\delta_{jk} + \frac{1}{2} \frac{\partial F}{\partial D_{jk}}.$$

Now the question arises, how the directions of the axes of the molecules are to be determined. Anzelius assumes that between the molecules there are forces in action that have rotary moments about the centres of gravity. The analytical expression for these forces he takes from the present author's theory of liquid crystals. But he assumes further that the motion of the liquid has a directive effect on the molecules. This, he supposes, is due to the fact that on account of the motion the impacts of the molecules give rise to a rotary moment. Taking the conditions of symmetry for his basis he finds for this moment, \mathbf{M} , an expression, which may be written in the form:

$$\mathbf{M} = \{(C_1 - C_2)(\mathbf{LD}) + \frac{1}{2}(C_1 + C_2)(\mathbf{L} \times \text{rot } \mathbf{u}) \\ + (C_{11} + C_{12})(\mathbf{L} \times (\mathbf{LD})) - \frac{1}{2}(C_{11} - C_{12})\text{rot } \mathbf{u}\} \times \mathbf{L}.$$

C_1, C_2, C_{11}, C_{12} are constants. (\mathbf{LD}) is the vector whose components are $L_j D_{j1}, L_j D_{j2}, L_j D_{j3}$. Here, a term in which the same index appears twice, will, with regard to this index, be summed up over the values 1, 2, 3.

Finally I should mention the boundary condition which Anzelius introduces into his theory. He assumes that, in a flowing liquid, asymmetrically constructed molecules arrange themselves perpendicularly to the wall.

⁴ A. Anzelius, *Über die Bewegung der anisotropen Flüssigkeiten*. The Annual of the University of Uppsala, 1931.

Basing our computations on these assumptions we may now proceed to calculate the motion of a liquid in given cases. As appears from what is said above, one of the most interesting cases is that of a liquid flowing through a tube, because in this case a liquid with oblong molecules does not give the same result as a liquid with spherical molecules. Anzelius summarises the results of his calculations on this case as follows :

1. The theoretical relation between the pressure gradient and the amount of liquid passing per unit of time agrees with the experimental relation, if $A_3 < 0$.

2. When A_3 is negative, the distribution of velocity over the transverse section of the tube is different from that found in the case of isotropic substances, in that the velocity in the middle of the tube is comparatively greater. Ostwald supposes, though he cannot refer to any theory or observations, that in flowing æolotropic liquids an equalisation of the velocities will take place in the transverse section, analogous to that which occurs in turbulent isotropic liquids. The result arrived at does not agree with Ostwald's assumption.

3. The orientation of the molecules depends on the sign of the pressure gradient, that is, on the direction of the flow. This agrees with an observation by Professor R. Fåhræus, Uppsala, according to which blood that is absorbed by a capillary, seems to change its colour, when the direction of the flow is reversed.

4. As the velocity of the flow increases, the molecules tend to take a direction which is parallel to the axis of the tube. This phenomenon is analogous to the rotation of the molecules in electric and magnetic fields, which may, if the field strength is sufficiently high, cause the destruction of the edge stratum.

Anzelius has further investigated the behaviour of an æolotropic liquid in the case of Couette's arrangement, that is, when the liquid is enclosed between two coaxial cylinders, the inner of which is stationary, whereas the outer one rotates with constant velocity round the common axis. If the inner radius is r_1 and the outer one r_2 if $(r_2 - r_1)/r_1 = E$, and if ω is the angular velocity of the outer cylinder, and M_d is the rotary moment acting per unit of length on the inner cylinder, he finds :

$$M_d = \alpha\omega + \beta\omega^3$$

where :

$$\alpha = \frac{2\pi A_1 r_1 r_2}{2E - E^2 + E^3}, \quad \beta = \frac{\pi A_3 C_2^2 r_1^3 r_2^3}{60K_{22}^2} E \text{ for asymmetrical molecules,}$$

$$\text{or} = \frac{\pi A_3 C_1^2 r_1^3 r_2^3}{60K_{22}^2} E \text{ for symmetrical molecules.}$$

To this must be added an expression for the largest angle that the axis of a molecule forms with the radius vector from the axis of the cylinder :

$$\lambda_{\max.} = \frac{C_2}{8K_{22}} r_1 r_2 \omega E.$$

We see that by determining ω , M_d , $\lambda_{\max.}$ we shall also be able to determine three constants A_1 , A_3 and C_2/K_{22} . K_{22} is a constant taken from the author's theory of liquid crystals.

Anzelius has further investigated the manner in which, in Couette's case, the stability of the motion is affected by the æolotropy of the molecules. In order to make the complicate calculations possible he had to assume that the axis of a molecule adjusts itself radially everywhere.

The investigation is of interest, because in this case, too, one must assume $A_3 < 0$.

I hope that the above review of the theory of Anzelius has shown the great interest it deserves. Attention must, however, be called to a difficulty inherent in this theory. During the motion of a liquid the moment \mathbf{M} has to do work. It is not clear how this work stands with regard to the principle of energy.

On account of these doubts as to the theory of Anzelius the author has developed another theory of the motion of an ælotropic liquid. According to this theory the state of an element is characterised by a vector \mathbf{L} , whose components are the mean values of the unit vectors that indicate the directions of the axes of the molecules belonging to the element. Accordingly the direction of the vector \mathbf{L} indicates the mean directions of the axes of these molecules. Its length indicates the ælotropy of the element. For the flow of the liquid and for the changes of the vector \mathbf{L} the following laws can be formulated:

$$\rho \frac{du_j}{dt} = X_j - \frac{\partial p}{\partial x_j} - \rho \frac{\partial}{\partial x_j} \int F(x, x', \mathbf{L}, \mathbf{L}') dm' + \sum_k \frac{\partial \tau_{jk}}{\partial x_k},$$

$$\rho a^2 \frac{d^2 L_j}{dt^2} = \Lambda_j + \rho A \frac{L_j}{a} - \rho \frac{\partial}{\partial L_j} \int F(x, x', \mathbf{L}, \mathbf{L}') dm' - \sigma_j.$$

Here X_j ($j = 1, 2, 3$) are the components of the external force that acts on the liquid, Λ_j are the components of a generalised external force, for instance the magnetic. A is a quantity characteristic of the theory and called the pressure of orientation. $\int F dm$ is the potential energy of the element dm in the neighbourhood of the point x and of the element dm' in the neighbourhood of the point x' . In order to determine the tensor of viscosity τ_{jk} and the corresponding vector σ we assume that they depend on a dissipation function $G(D_{jk}, dL_j/dt; \mathbf{L}, \text{rot } \mathbf{u})$ so that

$$\tau_{jk} = \frac{\partial G\left(D_{jk}, \frac{dL_j}{dt}; \mathbf{L}, \text{rot } \mathbf{u}\right)}{\partial D_{jk}},$$

$$\sigma_j = \frac{\partial G}{\partial \frac{dL_j}{dt}}.$$

The heat produced per unit of volume and unit of time is then $2G$.

The two equations given above do not suffice to determine the motion. To them must be added the condition of continuity:

$$\rho \frac{dv}{dt} = \sum_j \frac{\partial u_j}{\partial x_j},$$

and the equation of the conduction of heat:

$$\rho T \frac{d}{dt} \frac{\partial \zeta}{\partial T} + 2G = \text{div } \mathbf{j}.$$

Here $v = 1/\rho$ is the specific volume, ζ the free energy, and \mathbf{j} the current of heat. Further the equations of the state of the liquid must be added:

$$p = - \frac{\partial \zeta}{\partial v}, \quad A = - \frac{\partial \zeta}{\partial A}.$$

The theory expressed in these formulæ agrees with the principle of energy.

According to the opinions generally accepted it may be assumed that the dissipation function G is a homogeneous, quadratic function of the nine quantities D_{jk} , dL_j/dt the coefficients may be functions of L_j and $\text{rot } \mathbf{u}$. A natural hypothesis enables us to determine G more closely. It is a plausible assumption that all forces of friction vanish, when the liquid moves like a solid body. This assumption is equivalent to the assumption that G is a homogeneous, quadratic function of the eight quantities D_{jk} , $\mathbf{L} \frac{d\mathbf{L}}{dt}$, $\frac{d\mathbf{L}}{dt} \text{rot } \mathbf{u}$. The coefficients may as before depend on \mathbf{L} and $\text{rot } \mathbf{u}$.

The theory sketched above has not yet been used for the calculation of given problems of motion. I have mentioned it here, because it seems to me to throw light upon one of Lehmann's most remarkable observations on liquid crystals. He found that in certain cases a substance, spread out between two glass surfaces, would be put into motion, when influenced by a flow of heat coming from below, during which motion the different drops of liquid seemed to be in violent rotation. Further investigations convinced Lehmann that in this case it was not the drop itself, but the structure, that moved. According to my opinion the observed motion was due to the fact, that the molecules rotated with the same speed round vertical axes drawn through their centres of gravity. At this rotation $\mathbf{u} = 0$, $\mathbf{L} \frac{d\mathbf{L}}{dt} = 0$. Consequently all forces of viscosity vanish. The "violent speed" is hereby explained.

5. On the Forms of Liquid Crystals.

When Lehmann introduced the term "liquid crystals," he did it not only on account of the double refraction found in certain liquids, but also on account of the form that drops of certain substances, namely those which are now usually called the smectic ones, will take under favourable conditions. That this form is not spherical is the first important fact in this connection that a theory of liquid crystals has to explain. Lehmann's further investigations showed that smectic drops can assume a multitude of different forms, some of which are characterised by considerable grace and symmetry. To find these forms theoretically is the final object of the theory in this connection.

In my contribution to the discussion on liquid crystals arranged by Professor Ewald and published in *Z. Kristallographie*, I formulated a mathematical problem, the solution of which was calculated to throw light upon the above questions. Roughly speaking, it stands to the exact thermodynamic treatment of them as Laplace's theory of capillarity stands to Van der Waals' theory. The problem was formulated like this: It is required to determine a closed surface, S , a region outside it, U , and a vector function \mathbf{L} which fulfils the condition $L^2 = 1$, in the interior of S so that the expression:

$$\frac{\rho^{(a)2}}{2m^{(a)2}} \int_K \{K_{11}(\mathbf{L} \text{rot } \mathbf{L})^2 \times K_{33}(\mathbf{L}\Delta)\mathbf{L})^2\} d\omega$$

$$+ \frac{1}{2} \int_S \left\{ \frac{\rho^{(a)2}}{m^{(a)2}} f_1(L_n) + \frac{2\rho^{(a)}\rho^{(i)}}{m^{(a)}m^{(i)}} f_2(L_n) + \frac{\rho^{(i)2}}{m^{(i)2}} G_2 \right\} dS,$$

where

$$\rho^{(a)}, \rho^{(i)}, m^{(a)}, m^{(i)}, K_{11}, K_{33}, G_2$$

are constants and K is the region enclosed by S , takes the smallest value that is compatible with the conditions :

$$\rho^{(a)} \int_K d\omega = Nm^{(a)}, \quad \rho^{(i)} \int_U d\omega = Mm;$$

N and M being here constant, positive numbers.

The above problem has been subjected to an investigation by Mr. N. E. Larsson. The following is for the most part based upon this investigation.

If we want to solve a problem in the calculus of variations, we may begin by formulating the necessary conditions to be fulfilled by the solution, which conditions follow from the requirement that a small variation of the wanted functions must leave unchanged the expression that is to take the minimum value. In the problem under consideration we can firstly vary the directions of the axes of the molecules, or in other words, the vector \mathbf{L} , and secondly the form of the crystal, in other words that of the closed surface S . Through the former variation we obtain the laws holding for the orientation of the molecules in a liquid crystal. These laws, which must of course agree with the laws earlier formulated by the author, have the form :

$$[(K_{11} - K_{22})\{2 \mathbf{L} \text{ rot } \mathbf{L} \cdot \text{rot } \mathbf{L} - \mathbf{L} \times \text{grad } (\mathbf{L} \text{ rot } \mathbf{L})\} + K_{33} \text{rot rot } \mathbf{L}] \times \mathbf{L} = 0 \quad (1)$$

Further we obtain a condition that must be fulfilled at the bounding surface S . If, for the sake of simplicity, we put :

$$\frac{m^{(a)2}}{\rho^{(a)2}} \left\{ \frac{\rho^{(a)2}}{m^{(a)2}} f_1(L_n) + \frac{2\rho^{(a)}\rho^{(i)}}{m^{(a)}m^{(i)}} f_2(L_n) + \frac{\rho^{(i)2}}{m^{(i)2}} G_2 \right\} = f(L_n),$$

this condition may be written :

$$(K_{11} - K_{33})L_n \cdot \mathbf{L} \text{ rot } \mathbf{L} \cdot \mathbf{L} - K_{11} \mathbf{L} \text{ rot } \mathbf{L} \cdot \mathbf{n} + K_{33} L_n \cdot \text{rot } \mathbf{L} - \frac{1}{2} f'(L_n) \cdot \mathbf{n} \times \mathbf{L} = 0 \quad (2)$$

The variation of the form of the crystal gives a new condition which the surface S must fulfil :

$$K_{11}(\mathbf{L} \text{ rot } \mathbf{L})^2 + K_{33}((\mathbf{L}\Delta)\mathbf{L})^2 + f'(L_n) \text{div } \mathbf{L} + \sin \psi \frac{df'(L_n)}{dt} + \{f(L_n) - f'(L_n) \cdot L_n\} \left(\frac{1}{R_1} + \frac{1}{R_2} \right) = \text{constant} = K \quad (3)$$

Here ψ is the angle between the vector \mathbf{L} and the normal \mathbf{n} drawn outwards. df'/dt is the derivative of the function f' in the direction given by the projection of the vector \mathbf{L} on the tangent plane of the surface. R_1 and R_2 are the radii of curvature of the surface, taken as positive, if the corresponding centre of curvature lies within the surface S .

As the problem is difficult, it will be allowable to begin by investigating simple cases.

It appears from Lehmann's photographs that one structure, which is at least approximately realised in many smectic drops, is so shaped that the axes of the molecules are parallel, and the form of the drop is determined by the fact that its bounding surfaces are a right circular cylinder and two planes perpendicular to this. The direction of the axes of the molecules coincides with the direction of the generatrices of the cylinder. Now the question arises whether this structure agrees with the theory developed.

It is at once obvious that the law holding for the structure of the interior of a liquid crystal, is fulfilled. The first boundary condition, too, is fulfilled, if we make the plausible assumption that $f'(0) = 0$. The second boundary condition is fulfilled by the cylindrical boundary surface. But it is not fulfilled with regard to the plane end surfaces, because the left-hand member of the equation (3) has the value 0, whereas the right-hand member must have the value K . From this it follows that our assumptions concerning this structure cannot all be correct. We see also where we shall have to make a correction. The assumption that the non-cylindrical bounding surfaces are plane, cannot be absolutely correct. From this follows that in the vicinity of these surfaces \mathbf{L} cannot be a constant. If the end surfaces have a different form, for instance spherical, it will not be difficult to satisfy the equation (1) and the first boundary condition. This may be done thus, that the axes of the molecules are assumed approximately to fulfil the condition $\text{rot } \mathbf{L} = 0$ and to arrange themselves perpendicularly to the end surfaces. But the question still remains to be answered why these end surfaces are but slightly curved. We see, however, from our formula (3) that an explanation is afforded at once. If we apply this formula to the cylindrical bounding surface, we shall obtain $f(0)$ as coefficient of the medium curvature. If, on the contrary, we apply it to the end surfaces, we shall obtain $f(1)$. We only have to assume $f(1) \gg f(0)$ and the approximately plane form of the end surfaces will be explained. It should be noticed that the assumption $f(1) \gg f(0)$ is also necessary for the explanation of the prolate form that is characteristic of the smectic drops considered. The fact that the molecules at the greatest part of the bounding surface adjust themselves so as to be parallel to this surface, proves that $f(1) \gg f(0)$. The theory thus traces the prolate form and the approximately plane end surfaces to a common origin.

Much work will yet be required before the strange and beautiful forms that smectic drops will often take are explained. But the framing of a theory of these forms has begun.