Physics. — "Double refraction by regular crystals". By Prof. H. A. LORENTZ.

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1. It is well known that crystals of the regular system are anisotropic as to their elastic properties. Their three constants of elasticity are not connected by the same relations as those of isotropic substances. Therefore geometrically equal rods cut from the crystal in different directions are bent or twisted to different degrees.

Substances as rocksalt and fluor-spar on the contrary are single refracting to a first approximation. The Fresnel ellipsoid from which in crystal optics all phenomena are derived, is a sphere; this is also in accordance with the electromagnetic theory of light on the assumption that the optical properties are defined by the dielectric constant. Crystals with three equivalent mutually perpendicular principal directions can have but one dielectric constant.

More detailed considerations however teach that this optic isotropy can only exist as long as the distance  $\sigma$  of the molecules is very small compared with the wavelength  $\lambda$ . When  $\lambda$  becomes of the same order to this distance, we have for each direction of propagation two mutually perpendicular directions of vibration, the "principal directions" to which belong different velocities of propagation.

2. In 1877 I was led to the treatment of this problem 1) by the discussion of the explanation of the chromatic dispersion that was often excepted in those days. The unequal velocities of rays of different wavelength were explained by the assumption that the mutual distances of the molecules may not be neglected compared with the wavelength, which assumption may f.i. still be found in old papers of Kelvin. In the cited paper I explained how this assumption is in contradiction with the fact, that with a few exceptions, the regular crystals are single refracting; when namely the ratio  $d/\lambda$  was so great that it could give rise to the dispersion, this should necessarily be accompanied by a detectable double refraction.

<sup>&</sup>lt;sup>1)</sup> H. A. LORENTZ: Over het verband tusschen de voortplantingssnelheid van het licht en de dichtheid en samenstelling der middenstoffen. Verh. der Akad. van Wetenschappen te Amsterdam, 1878.

The true value of  $\sigma$  was then taken from the first estimations of van der Waals. Nowadays however we know the absolute dimensions of the molecules and also the distance  $\sigma$ . Moreover, thanks to the investigations on the interference of the Röntgen rays we can trace the structure of the crystals in detail. This made it desirable to take the problem at hand once more. While the wavelength of the Röntgen rays is comparable with  $\sigma$ , the question is, whether already for light rays the molecular discontinuity is of influence, whether there are any indications that  $\sigma/\lambda$  may not be quite neglected.

In my former calculations I made use of the theory of MAXWELL in the form given to it by Helmholtz. I followed this way because I had not yet penetrated deeply enough into the ideas of MAXWELL. In the first place, therefore, the calculations had to be repeated and to be based upon the theory of MAXWELL and the theory of electrons. The new calculations gave the same results as the first ones.

3. It will suffice to consider a cubical arrangement of the molecules. The equations for the light motion were derived on the supposition that equal particles are placed at the points of a cubic lattice. Further it has been assumed that in each molecule an electric force is excited and a corresponding electric moment in the direction of that force.

From considerations on the symmetry of the crystal we may easily deduce that for some definite directions of propagation comparable with the axis of monoaxial crystals we have only one velocity of propagation; these directions are those of the edges of the cubic lattice and of the diagonals of the elementary cube. By the diagonals of the side-faces of this cube however those directions of propagation are given for which we may expect (and this is confirmed by the experiments) the anisotropy in question to be felt most strongly. Further on we shall always assume the direction of propagation to coincide with such a diagonal of a side-face of the cube. Then the principal directions of propagation  $R_1$  and  $R_2$  may be indicated immediately.

The first one is that of the edge of the cube perpendicular to that side-face, the other one that of the second diagonal of that face.

The velocities of propagation belonging to these directions of vibration will be indicated by  $v_1$  and  $v_2$ , while the corresponding values of the refraction index  $\frac{c}{v_1}$  and  $\frac{c}{v_2}$  (c the velocity in vacuum) will be represented by  $\mu_1$  and  $\mu_2$ . For the difference between these last

quantities we find by making use of some simplifications that are allowed because of the smallness of this difference:

$$\mu_1 - \mu_2 = 0.11 \, \frac{n^2 \, \delta^2 \, \mu \, (\mu^2 - 1)^2}{\pi c^2} \, . \quad . \quad . \quad . \quad (1)$$

Here n is the number of vibrations in the time  $2\pi$ ,  $\sigma$  the distance of the molecules viz. the edge of the cubic lattice and  $\mu$  one of the refraction indices  $\mu_1$  and  $\mu_2$ , or rather the mean of both; we may say the observed refraction index.

From (1) we see that the sign of  $\mu_1 - \mu_2$  is the same as for a plate of calcite the optical axis of which coincides with the direction  $R_2$  and also the same as for a glass plate that is compressed in the direction  $R_2$ .

During the propagation in the crystal over a distance D a difference of phase arises between the vibrations in the directions  $R_1$  and  $R_2$ . Expressed in periods or wavelengths this difference in phase is determined by

$$\omega = (\mu_1 - \mu_2) \frac{D}{\lambda} = 0.44 \pi \mu (\mu^2 - 1)^3 \frac{D\sigma^2}{\lambda^3}, \quad . \quad . \quad . \quad (2)$$

where  $\lambda$  is the wavelength in vacuum.

For rock-salt  $d = 2.80 \cdot 10^{-8}$  cm.

With this  $\sigma$  we find for a thickness of 1 cm the following values of  $\omega$ .

$$\lambda = 5.9 \cdot 10^{-5}$$
 (sodium light); 5.1 \cdot 10^{-5}; 3.1 \cdot 10^{-5}; 1.8 \cdot 10^{-5}   
\omega = 0.016 ; 0.025 ; 0.14 ; 2.1.

Even in the visible spectrum these numbers are great enough to let us expect that under favourable circumstances the effect of the double refraction will be detectable between crossed Nicols.

4. I have sought for the phenomenon in several pieces of rock-salt, in which the faces of the cube were obvious and which were bounded by two side-faces perpendicular to the direction indicated above with L; the distance between these phases was about one cm. The side-faces could be easily polished, but we met with the difficulty, that they loose their polish even when the crystal is kept in dry air. To avoid this the crystal was put into a tube a little longer than the tickness of the rock-salt and shut on both sides by glass plates from Hilger with neglectable double refraction. The remaining space in the tube was filled with a mixture of carbonic disulphide and benzol, of which for the mean yellow light, the refraction index is equal to that of the rock-salt. Under these circumstances the crystal is beautifully transparent even when the side-faces have not

been polished particularly well. Neither is it now necessary, that these faces are perfectly plane and exactly parallel to each other; it suffices when the glass-plates are rather exactly parallel to each other. We had only to take care that these plates are not exposed to a too high pressure, so that the closeness (with caoutchouc rings between the glass plates and plane metal rims) was not perfectly tight. This however was of no importance.

5. When after having been fixed in this way the crystal was placed between crossed Nicols the light reappears again and when we fix the eye on the crystal we see irregular light spots over its extension (about 1 cm²). These are due to inner structure deviations and tensions and might retain us from further investigation, when not the differences in phase in this "accidental" double refraction proved to be rather small. When anywhere they reached the value of half a wavelength we should see dark spots between parallel Nicols. There is no question of this; a rotation of one of the Nicols over a few degrees from their crossed situation sufficed to obtain a uniform distribution of the light.

The irregular differences in phase being so small, it might be expected that the effect of a regular anisotropy, the same over the whole cross-section of the light beam could be observed when it was superposed on the irregular differences. In fact this was proved to be the case when a thin plate of mica was adjusted before the crystal and rotated in its own plane.

6. Now the experiment was made on the following way. A small round aperture into which the rays of a glow-lamp with opaque bulb are folling is placed in the focus of a collimator lens. After passing this lens the rays fall in a telescope that is focussed for parallel rays.

We then see a sharp image of the lighted aperture, which image is extincted by two Nicols placed between the collimator lens and the telescope.

When now the crystal is placed between the Nicols and when these are rotated, we see in positions differing 90° minima of the intensity of the light. These minima are not always equally pronounced, but they are always easy to recognize.

That in the mentioned positions the light is not perfectly extinguished, must of course be ascribed to the accidental double refraction, which now however causes a uniform illumination. As we have namely focussed not at the crystal but at the lighted aperture,

we have at every point of the image rays that have crossed the crystal at different points of the section. We may say, that of that double refraction we observe the mean over the whole section.

It may be remarked, that we can also speak of such a mean with respect to the succeeding layers of the crystal and that the great thickness which we use is in a certain respect an advantage. When namely the disturbing double refraction changes irregularly along the ray we may assume the intensity caused by it to be proportional with the thickness, while a regular double refraction gives an intensity proportional with the second power of the thickness as long as the difference in phase is small.

By these disturbing anomalies a sharp focusing at the minimum is impossible. In order to form an estimation of the accuracy of the experiment and therefore of the regularity of the phenomenon we read the positions of the crystal on a divided circle after having focussed at a minimum as accurately as possible. When one of the crystals was rotated continually we obtained the following readings in degrees:

17 14	107	<b>202</b>	294
	105	196	293
19	108	196	291

When the numbers of the second column are diminished by 90, those of the third one by 180 and those of the fourth one by 270 and when we then take the mean of the 12 numbers, we find 18, so that the principal positions would be

With the other crystals we obtained similar results, sometimes a little better, sometimes a little worse.

8. The positions of extinction were always those, for which the directions indicated with  $R_1$  and  $R_2$  coincide with the directions of vibration of the Nicols. As to this, the theoretical expectation is therefore confirmed.

In order to see whether also the sign of the double refraction agrees with (1) we used a glass plate compressed in one direction which was placed in the way of the rays and which was rotated in its plane. It was found then, that for all ten crystals that were investigated and which were cut from different pieces of rock-salt, the effect could be compensated by the glass plate, when the direction of compression coincided with  $R_*$ . Taking into consideration § 4 we come to the conclusion that the sign is opposite to that given by form (1).

Though this contradiction is not satisfactory, our astonishment must not be too great, to my opinion, as in the light of our present knowledge, the theory from which the equation has been derived is so very imperfect. There the assumption has been made that equal molecules were placed at the points of the crystal lattice and that in each of these an electric moment is excited as will be the case when the molecule contains a quasi-elastically and moreover isotropically bound electron. According to the present opinion however the sodium and chlorine nuclei are placed alternately along each edge of the lattice, while round these nuclei and perhaps also round the lines of connection electrons are circulating. When this circulation takes place in planes, the position of these planes may give rise to an anisotropy.

Perhaps the only thing that can make plausible the old theory is, that an anisotropy may be expected which like that determined by (1) is proportional with  $\frac{\sigma^2}{\lambda^2}$ . I have not tried a calculation based upon the new points of view. First we shall have to be further in the general treatment of light vibrations.

That the old theory is imperfect in several respect may be seen from the following. The difference in phase determined by (2) strongly increases with diminishing wavelength and when working with white light, we should therefore see the field distinctly coloured. This is however not at all the case.

As to the value of the difference in phase, it has hardly been possible to determine it because of the imperfectness of the extinctions.

For the crystals used it could not be measured with the compensator of Babinet. The only thing that could be done was to determine with this means the difference in phase of the compressed glass plate by which the double refraction of the rock-salt was compensated rather satisfactorily. In this way it was found, that the difference in phase was a small fraction, about  $\frac{1}{20}$  or  $\frac{1}{40}$  of a wavelength.

The idea suggests itself to work also with crystals, the side-faces of which are perpendicular to an edge of the lattice, in which case a double refrection as has been described above cannot exist.

To my astonishment even now we can often distinguish two mutually perpendicular positions in which the intensity is a minimum, so that we get the impression that also for the mean of the accidental double refraction taken over the cross-section of the crystal, we can speak of two principal directions. The phenomena however were doubtlessly less regular than for the crystals with which the

former experiments had been made. The minima were less pronounced and with the exception of a single case we could not focus on them as accurately as in the preceding case. Sometimes the focusing was much more dubious. Besides the positions of extinction were not always the same; sometimes the two edges of the lattice which were now lying in a plane perpendicular to the light beam, nearly coincided with the directions of vibration of the nicols, but sometimes they made with these angles of 30° or 40°. My final impression is however, that the double refraction in question really exists; but I hope that others will have the opportunity to repeat the experiments with better crystals than those that were at my disposal.