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Crystallography. — "On the Symmetry of the Röntgen-patterns of Rhombic Crystals". **I.** By Prof. H. Haga and Prof. F. M. JAEGER.

§ 1. With the purpose to test yet further by experiments the conclusions with respect to the special symmetry of the diffractionpatterns of Rontgen-rays in crystals, as may be derived from the theory adopted till now, we have moreover made a series of experiments with optically biaxial crystals. In the following pages are recorded the results obtained in the investigations with *rhombic* crystals, and especially with representatives from each of the possible symmetryclasses of the rhombic system. As already formerly mentioned '), ; the RONTGEN-patterns of plates from rhombic crystals of *bisphenoidical*, *pyramidal*, or *bipyramidal* symmetry, must always show the symmetry which would be observed in the analogous crystal-sections of the rhombic-bipyramidal class. From this is deduced, that crystal-plates cut parallel to the three pinacoides {100}, {010} and {001}, in the case of crystals of each of the three possible classes of this system will give RONTGEN-patterns, which will always appear symmetrical with respect to twoperpendicular planes of symmetry, if they are radiated through in the direction of the plate-normal.

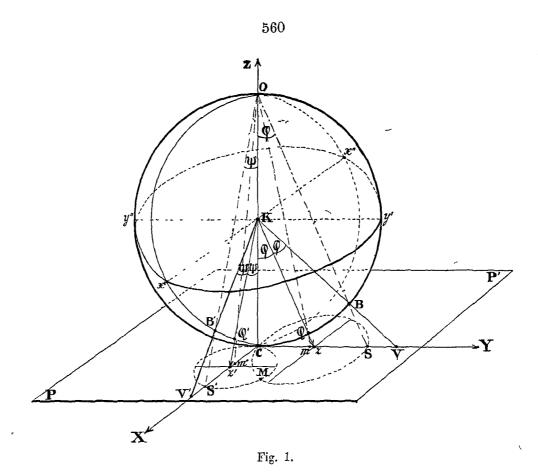
§ 2. The way, in which the true situation of the spots in the diffraction-pattern, or more correctly: in its stereographical projection, is related to the parameters a:b:c of such rhombic crystals, can be elucidated more particularly as follows.

Let P in Fig. 1 be the plane of the photographic plate, and KC the distance of the crystal from that plate. This distance (= A) was in our experiments always 50 mm.²); the diameter of the sphere by means of which the stereographical projection on the plane P will take place, is therefore always 100 mm. The viewpoint of me projection is 0; the planes XCY (= P), XCZ, and YCZ are supposed to coincide with (or to be parallel to) the three planes of symmetry of rhombic-bipyramidal crystals. Let the parameter-quotient in the directions CX, CY and CZ be: a:b:c, of which a and c are numbers, known by the measurements of the angular values, and b is taken arbitrarily = 1.

If, now, Kz represents a "possible" zône-axis of the crystal, the value of $\frac{Cz}{CK}$ must be: $n \cdot \frac{1}{c}$, where $\frac{1}{c}$ is the parameter-relation in

¹) These Proceed. 17, 1204. (1915).

2) However attention must here be drawn to the remarks in § 4



the directions CY and CZ, while *n* is some integer. If Kz' is equally such a zône-axis, then just in the same way $\frac{Cz'}{CK} = m \cdot \frac{a}{c}$, where *m* is also an integer. The projected spots *S* and *S'*, corresponding with the reticular planes {101} and {011} of the crystal, are therefore in the directions CX and CY of the stereographical projection situated in distances from the central spot *C*, of: $2A\frac{a}{c}$ and $\frac{2A}{c}$, or in our case of: $\frac{100a}{c}$ and $\frac{100}{c}$ mm. In the same way for spots corresponding with the reticular planes of the symbols {*ho1*} and {*ok1*}, these distances from *C*, become: $\frac{1}{\cdot h} \cdot \frac{100a}{c}$, resp. $\frac{1}{k} \cdot \frac{100}{c}$ m.m. The point of intersection *M* of both zône-circles, corresponding with the zône-axes *Kz*, resp. *Kz'*, possessing the symbols [*ho1*] and [*ok1*], represents the stereographical projection of a spot, corresponding with a plane of the structure with the symbol {*hkl*}; etc. In this

way it is possible to determine the indices of every spot in the

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stereographical projection P by means of the existent zône-relations, and to investigate, which reticular planes will give the most intense impressions on the photographic film; or, what is the same: it will be possible to find out, in what way the spectral dissolvingpower of the different molecular planes is connected with their reticular density.

We have chosen the case of a radiation perpendicular through a plate parallel to {001}. In the same way however it can be found, that: On {100} the spot corresponding with {101}, is situated in the direction

of C-axis, at a distance of $\frac{100 c}{a}$ m.m

On $\{100\}$ the spot corresponding with $\{110\}$, is situated in the direction

of the *B*-axis, at a distance of $\frac{100}{a}$ m.m.

On $\{010\}$ the spot corresponding with $\{011\}$, is situated in the direction of C-axis, at a distance of 100 c m.m.

On $\{010\}$ the spot corresponding with $\{110\}$, is situated in the direction of A-axis, at a distance of 100 a mm.

all distances reckoned from the image-centre C^{1}).

§ 3. If in this way the indices of the molecular planes are determined, it is possible to construct a stereographical projection of them, and by means of this projection to execute the ordinary calculations, as usual in crystallography. However it is also possible to construct *directly* the stereographical projection of the reticular planes, with which the spots on the photo correspond, from the stereographical projection of the Rönrgen-pattern itself. The way to do this, is as follows:

The part to the right of fig. 2 relates to the original stereographical projection of the RÖNTGEN-pattern of *potassiumchloride*, obtained by BRAGG, if the plate, cut parallel to a face of the cube, is radiated through in a direction perpendicular to that face. The part to the left of fig. 2 represents the stereographical projection of the molecular planes of the crystalline structure, corresponding with the spots in the part of fig. 2, situated to the right; the plane of projection is parallel to the same face of the cube. If now we again pay attention for a moment to fig. 1, we shall find that in this figure

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¹⁾ The relation between the situation of the spots on the photographical film, and that of the corresponding points in the stereographical projection on P, can also easily be seen from the fig 1. For $CS = 2A \cdot tg \varphi$ and $CV = A \cdot tg 2\varphi$, if V is the original spot, and S its stereographical projection on P. If CV (= a) is measured on the photography, then $tg 2\varphi = 0.02 a$; from this φ and $tg \varphi$ are calculated, and thus $CS = 100 tg \varphi$ is found in m.m.

the point z is the centre of a zône-circle *CMS*, and thus also the point of intersection of the zône-axis KQ with the plane of the photographic plate P, this being in fig. 2 the plane of projection of the stereographical figure to the left.

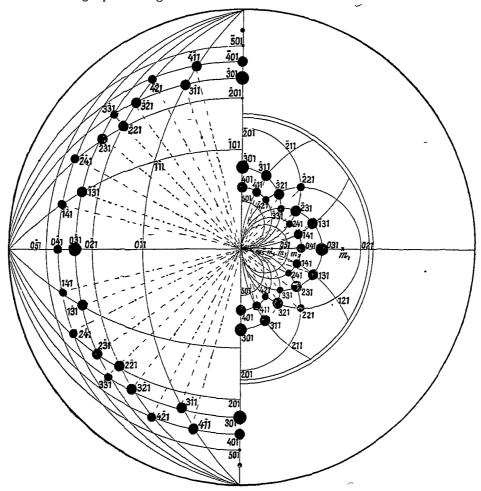


Fig. 2 Construction for the change of the Stereographical Projection of a RÖNTGENpattern (to the right: potassiumchloride after BRAGG), into the Stereographical Projection of the corresponding reticular planes of the Crystal-structure.

Let now $\neg m$ be the stereographical projection of the point Q, then, if the polar circle to m is drawn, this circle will contain all points, that are the projections of all tautozonal reticular planes, having KQ as their zone-axis. The azimuth of every spot in the plane Pand the azimuth of the stereographical pole of each corresponding reticular plane in P always being equal, the place of every one of these stereographical poles on the polar circle just obtained, may be localised by joining the original spot, and to pass this straight line

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ا ا _ سولاست through, until it intersects the constructed polar circle in the left part of the figure.

This point of intersection is the stereographical projection of the molecular plane, which corresponds with the spot in the right part of fig. 2. The points m can be easily found from the points z by an additional construction, in which a circle with a radius of 50 m.m. is used; the contractions of the original distances to C_{i} — which thus can be represented by mz, — are moreover for spots in the neighbourhood of C only so slight, that without considerable error, instead of m, the point z itself may be used; but at greater distances from C this of course is no longer allowable. The system of polar circles and poles of reticular planes obtained in this way, will give a direct review of the total zône-relation and of the indices of the molecular planes; while the calculation of the angles between the poles of those reticular planes and the plotting of graphical constructions etc., can be made in the way usual in crystallonomy. It is adviceable to keep the radius of the projection-circle in the left part of fig. 2 equal to 2A (100 m.m.); then it is possible to execute the different constructions by means of WULFF's stereographical projection plat with a diameter of 20 c.m. The indices of the poles of the reticular planes are the same as the original ones of the spots in the projection to the right of fig. 2.

Finally we can express the different intensities of the spots in the original figure, into the projection-figure of the corresponding molecular planes to the left of fig. 2; in this way a further discussion of the relations between the indices of the reticular planes and their spectral-resolving power can be made, in the way indicated for the first time by BRAGG.

§ 4. As an application of the discussions given above, we will consider here more in detail the Röntgen-patterns of the anhydrite. The parameters of this mineral are: a:b:c = 0.8932:1:1.0008; from this it follows, that for a distance of 100 m.m. between the plane of projection P and the viewpoint O of the projection, the projected spots for the faces $\{101\}$, $\{011\}$ and $\{110\}$ will be situated at the distances:

For a plate parallel to {100}:

in the direction of the *C*-axis at 112.04 m.m. from *C* ,, ',, ., ,, *B*-axis ,, 111.95 ,, ,, , For a plate parallel to {010}: in the direction of the *C*-axis at 100.08 m.m. from *C*

,, ,, ,, ,, ,, ,, A-axis ,, 89.32 ,, ,, ,,

For a plate parallel to {001}:

,,

in the direction of the A-axis at 89.25 m.m. from C

,, ,, ,, *B*-axis ,, 99.92 ,, ,, ,

If now we calculate from the measured distances of the spots on the photographic plate of anhydrite, using A = 50 m.m., the values of $tg 2\varphi$, then φ , and finally 100 $tg \varphi$, — we shall find that all these calculated values are a little smaller than the corresponding rational parts of the above mentioned fundamental distances in the directions of the resp. axes: moreover, these calculated values all appear to be diminished in about the same ratio.

In the case of the photo parallel to $\{100\}$ e.g. we measured for some spots:

in the direction of the C-axis: 31.2 mm.; 24.1 mm. and 19.9 mm. from the centre;

in the direction of the B-axis: 27.4 mm.; and 21.8 mm.

For the same points the *calculated* distances are:

in the direction of the C-axis: 32 mm.; 24.8 mm.; and 22.4 mm.in the direction of the B-axis: 28 mm.; and 22.4 mm.

But calculations and measurements now appear in much better agreement with each other, if we suppose A during the experiment to have had a *smaller* value, and to have been about 48,3 mm.

Now the *thickness* d of the crystalplate was in our case precisely 1,64 mm.; and thus we must conclude, that in this case we must use in our calculations of the angles φ , for A not the value of the distance from the *front* face of the crystal-plate to the photographic film, but that from the *backward*-face of it to the photographic plate.

In other analogous cases we indeed now learned, that if A was supposed to be = 50 mm., during the experiment, the distances from the projected spots to the centre C appeared to be always too small, if in the final projection the distance OP is always kept = 100 mm.; but that ordinarily a sufficient agreement between calculation and experiment would result, if A during the experiment is supposed to be $(50 - \frac{1}{2}d)$, where d is the thickness of the crystalplate used.

This influence of the plate-thickness becomes yet more evident, if of the same crystal Röntgen-patterns are obtained with plates of very different thickness; for in that case the photos, must manifest different distances from C for the same spots. We were able to observe something of the kind in the measurement of two analogous

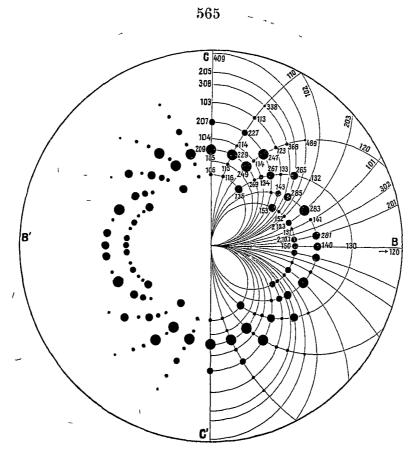


Fig 3. Stereographical Projection of the RÖNTGENOGRAM of Anhydrite. Plate parallel to (100).

photos of arragonite, obtained with crystalplates of different thickness.

Thus it seems undoubtedly necessary; to take into consideration the thickness of the crystal-plates in the calculations of the angles φ , and to diminish the distance of 50 m.m., if rather thick plates are used, with half the thickness of them ¹). The projections reproduced on Plate IV of this paper all relate to such stereographical projections, for which the distance *OP* is 75 m.m.: in such cases the diameter of the figure is also kept equal to 75 m.m.

In most of the drawings of Plate IV we have calculated the symbols $\{hko\}$, $\{okl\}$ and $\{hol\}$ of the zône-circles, (whose centres lie in the direction of the axes), in the way formerly described; the symbols of the most important spots in the figure can then be immediately seen from the indices of the zone-circles, after the method mentioned above.

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¹) This is connected with the specific absorbing power for Röntgen-rays of the crystallised material. As this absorption is stronger, the distance of 50 m.m. will have to be diminished with a *smaller* part of the thickness d.

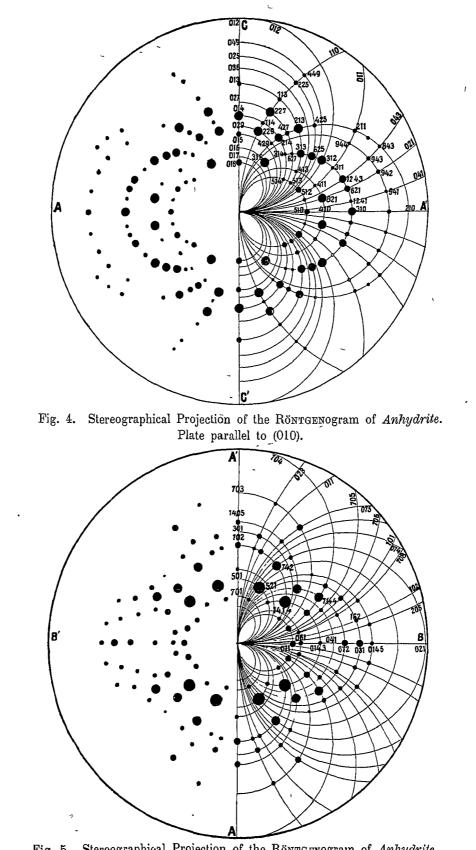


Fig. 5. Stereographical Projection of the Röntgenogram of Anhydrite. Plate parallel to (001). In connection with the crystals investigated up to this moment, the corresponding principal distances in the direction of the axes, as in the case of *anhydrite*, may be recorded here ¹).

	Struvite	Topaz	Anhydrite	Arrago- nite	Hamber- gite	Zinc- sulphate	Aspara- gine	Sodium- ammonium- tartrate	Calamine	Cordie- rite
On {100}: $\begin{cases} C-axis \frac{100c}{a} \\ B-axis \frac{100}{a} \end{cases}$	160.9					57.4	1	51.0	61.0	95.2
B -axis $\frac{100}{a}$	176.5	189.2	111.9	160.7	125.2	102.0	211.1	121.4	127.6	170.3
On $\{010\}$: $\begin{cases} C-axis 100c \\ A-axis 100a \end{cases}$	91.2 56.7	95.4	100.1	72.1	72.7	56.3	83.3		47.8	55.8
(A-axis 100a)	56.7	52.8	89.3	62.2	79.9	98.0	47.4	82.3	78.3	58.7
$(A-axis\frac{100a}{c})$	62.1	55.4	89.2	86.4	109.9	174.1	56.9	196.0	163.9	105.1
On {001}: $\begin{cases} A \text{-axis} \frac{100a}{c} \\ B \text{-axis} \frac{100}{c} \end{cases}$	109.6	104.8	99.9	138.8	137.6	177.6	120.1	238.1	209.3	179.1

§ 5. In previous papers we already discussed some crystals of rhombic symmetry, which will be reviewed again in connection with what is mentioned above. However we will principally discuss in this paper the results, to which our experiments till now have led us, with respect to the following crystals: anhydrite; arragonite; zinc-sulphate; topaz; struvite; l-asparagine and calamine. A following communication will then contain the results with other rhombic crystals, and at the same time we shall have then an opportunity to draw the attention to some problems, which are connected with the special choice of these crystals.

We will begin here with the crystals of the *rhombic-bipyramidal* class first ²).

a. Anhydrite $(CaSO_4)$. The used anhydrite-crystal was from STASS-FURT. It was lustrous and translucid, and evidently quite homogeneous. Parallel to the three directions of cleavage : {100}, {010} and {001},

¹⁾ Note that these numbers relate to a projection distance OP = 100 m.m.; our figures then have also a diameter of 100 m.m. But for the drawings on Plate IV, which are reduced to $\frac{3}{4}$ size, all these values need to be also multiplied with $\frac{3}{4}$.

²) The crystals discussed in this paper are supposed to have such a position, that their parameters become: anhydrite: a:b:c = 0.8932:1:1.0008; arragonite: a:b:c = 0.6224:1:0.7206; zinc sulphate: a:b:c = 0.9804:1:0.5631; topaz: a:b:c = 0.5285:1:0.9539; struvite: a:b:c = 0.5667:1:0.9121; calamine: a:b:c = 0.7835:1:0.4778; l-asparagine: a:b:c = 0.4737:1:0.8327; sodium-ammonium-tartrate: a:b:c = 0.8233:1:0.4200; hambergite: a:b:c = 0.7988:1:0.7268.

three rather thick crystal-plates were prepared, whose thickness we resp. 1.64 mm., 1,72 mm. and 2,09 mm. In this case and all other here we experimented again with the fluorescent screen "Eresco" the time of exposition was ordinarily about 2,5 hours. In this case of the anhydrite we used more particularly a Coolidge-tube, with wolframium-anticathode and separate heating-spiral.

The three photographs are reproduced in fig. 1—3 on Plate I their stereographical projections already in fig. 3, 4 and 5 of th text. All three images appear to be quite normal, and every on has two perpendicular planes of symmetry; the normal to the plat (direction of radiation) is thus at the same time a binary axis of th RONTGEN-patterns.

b. Arragonite $(CaCO_a)$. Our clear, lustrous crystals were from HORSCHENZ in Bohemen. The crystal-plate parallel to $\{100\}$ had thickness of 0,96 mm., that parallel to $\{010\}$ 1,06 mm., and tha parallel to $\{001\}$ 1,10 mm. The photos are reproduced in fig. 4 of Plate I, and in fig. 5 and 6 on Plate II; their stereographical projections in fig. 1--3 on Plate IV. Also in this case the pattern appear to be symmetrical with respect to two planes of symmetry perpendicular to each other, just as might be predicted from theory In the image parallel to $\{001\}$ moreover the well-known pseudo ditrigonal symmetry of the mineral is clearly recognisable.

c. Topaz. $(Al_2 (F, OH)_2 SiO_4)$. The topaz-crystal used by us wa very homogeneous, vitreous and translucid; it possessed a yellowisl hue, and originated from Saxony. The thickness of the three plate parallel to {100}, {010} and {001}, was from 1,20 mm. to 1,27 mm.; the time of exposition again two and a half hours. The plate parallel to {010} afterwards appeared to be a little inclined; therefore the corresponding photo was not reproduced here, but solely those of the other sections in fig. 7 and 8 on Plate II; their stereographica projections are to be found in fig. 4-6 on Plate IV.

Also in this case all three patterns appear to be symmetrical after two perpendicular planes, as might be expected from the theory.

To this same class belong furthermore the crystals of *cordierite* and of *hambergite*, already previously ¹) discussed.

d. In the case of *cordierite* the patterns of crystal-plates parallel to $\{100\}$ and $\{010\}$ appeared to be, till this moment, *always* abnormal, notwithstanding the fact that crystals of several localities were used in the experiments, and among these were present splendid, lustrous crystals. Only the pattern obtained with a crystal-plate parallel tc $\{001\}$, appeared to exhibit the normal symmetry.

¹) These Proceed. 17, 430, 1204. (1915).

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e. About the *hambergite* and its normal and abnormal images we have said already something in the foregoing paper. In fig. 7-9 on Plate IV we reproduce here again the more exact stereographical projections of the normal RONTGEN-patterns of this mineral, with indication of the corresponding indices of the reticular planes.

§ 6. Of the *rhombic-pyramidal* class, to which thus belong the hemimorphic crystals of the system, — we investigated here the *struvite* (= magnesium-ammonium-ortho-phosphate (NH_4)MgPO₄+6H₂O)), and the *calamine*: $Zn_2(OH)_2 SiO_3$.

f. From a big, brownish yellow and only little translucid crystal of *struvite* from HOMBURG, three plates were cut parallel to the three pinacoides $\{100\}$, $\{010\}$ and $\{001\}$, whose thickness was from 1,20 to 1,26 mm. The time of exposition was two and a half hours.

The three very beautiful RONTGEN-patterns are reproduced in the fig. 9, 10 and 11 on Plate III, and as stereographical projections in fig. 10—12 on Plate IV. Also in these images two planes of symmetry perpendicular to each other are evidently manifested; notwithstanding the polarity of the *c*-axis is very strongly revealed in the crystals themselves, the result is also in this case in full agreement with the theoretical prediction.

g. However in the case of calamine we obtained for crystal-plates parallel to $\{100\}$ and $\{010\}$, cut from a very beautiful crystal¹), always abnormal patterns, from which one parallel to $\{010\}$ is reproduced partially as a stereographical projection in fig. 13 of Plate IV; the image parallel to $\{100\}$ was quite analogous to that parallel to $\{010\}$, but it was too bad to allow in any way a reproduction of it. Both patterns contained moreover such a great number of very small and feeble spots, that also in the projection of fig. 13 on Plate IV, only the most important spots could be reproduced.

The RONTGENOGRAM of the *calamine* parallel to $\{001\}$ however was very beautifully regular (fig. 14 on Plate IV) and (fig. 12 on Plate III); quite in concordance with the theory, it is symmetrical with respect to two planes perpendicular to each other. Why it is the images parallel to $\{100\}$ and $\{010\}$, — (corresponding with those sections, that in the crystal itself do not possess the horizontal plane of symmetry), — where the plane of symmetry parallel to the *c*-axis is suppressed, can hardly be understood at this moment.

57. Finally we used from the crystals of the *rhombic-bisphenodical*

¹⁾ For this very beautiful crystal we are much indebted to our colleague, Prof. MOLENGRAAFF at Delft, whom we render our best thanks here once more.

class, besides the already formerly discussed crystals of *d*-sodiumaminonium-tartrate, moreover: those of *l*-asparagine: $(C_4H_8O_3N_2+H_2O)$, and of zinc-sulphate: $ZnSO_4 + 7H_2O$; of these compounds both the first named are optically active in solutions, while the zinc-sulphate does not change the plane of polarisation of the light, when passing through its solution.

h. Zinc-sulphate. From a beautiful crystal three rather thick plates were cut: that parallel to $\{100\}$ had a thickness of 2.11 mm., that parallel to $\{010\}$ of 3.30 mm., and that parallel to $\{001\}$ of 3.10 mm. Even with a time of exposition of two and a half hours, the patterns parallel to: $\{100\}$ and $\{001\}$ were too feeble, to allow of any reproduction; but in fig. 15 and 16 their stereographical projections are drawn.

These images are again symmetrical with respect to two perpendicular. planes. However the pattern obtained with a plate parallel to {010} appeared to be *always abnormal*; the respective photo is already reproduced in the foregoing paper on Plate V, while here in fig. 17 on Plate IV its stereographical projection is represented.

This last fact is indeed of high importance for our problem: for it may be supposed with good reason, that in cases, where such crystal-plates are prepared by cleavage along planes of very perfect cleavability, all chance to get a faulty orientation of the plate is altogether eliminated. Now in our case the very perfect orientation of this plate obtained by such cleavage, could moreover be very rigorously tested, because of the fact that the b-axis, being the direction through which radiation here takes place, is at the same time the first bisectrix of the crystal. Indeed the interference-image in convergent polarized light appeared after measuring with the microscope, to be accurately centred, so that no deviation between the b-axis and the normal on the plate could be found by any means. And while now the orientation of the perfectly clear and lustrous plate could hardly show any error exceeding a few minutes; the image was in two repeated experiments, absolutely abnormal in the way indicated here: evidently only the plane of symmetry parallel to the plane of the optical axes has remained.

This fact must convince us in a striking way, that the abnormalities occurring in the case of such crystals cannot have their origin in a faulty orientation of the crystal-sections. Indeed, they must be caused by *internal* disturbances of the molecular structure, which evidently, as here with the *zinc-sulphate*, cannot even be discerned by the usual optical means. At the same time it appears furthermore by this fact, that the probability of such "internal vicinal."

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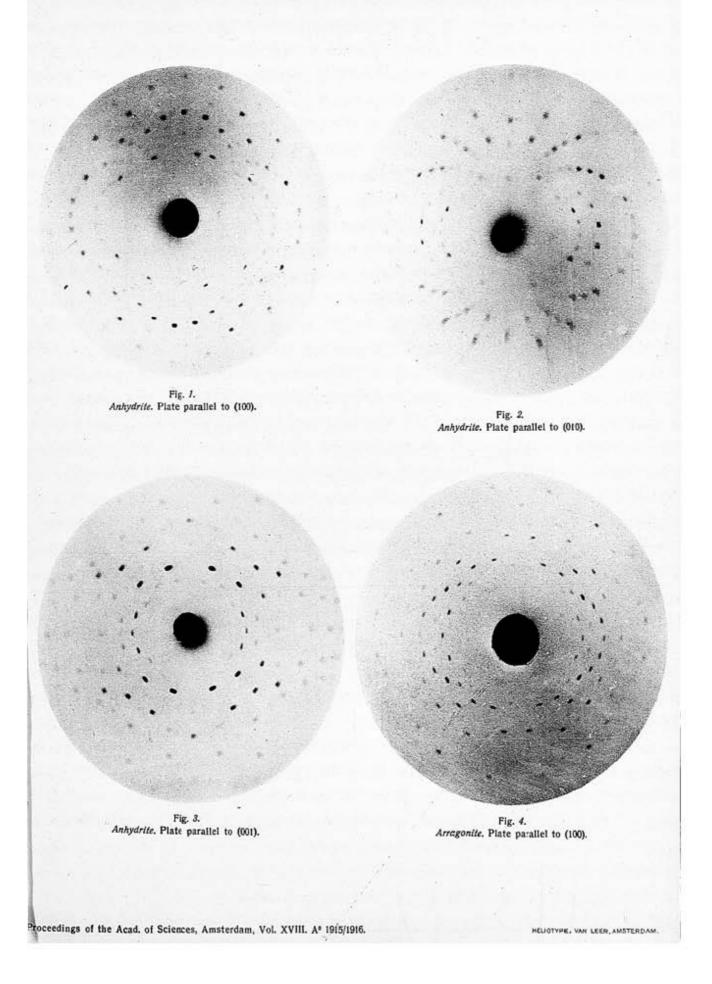
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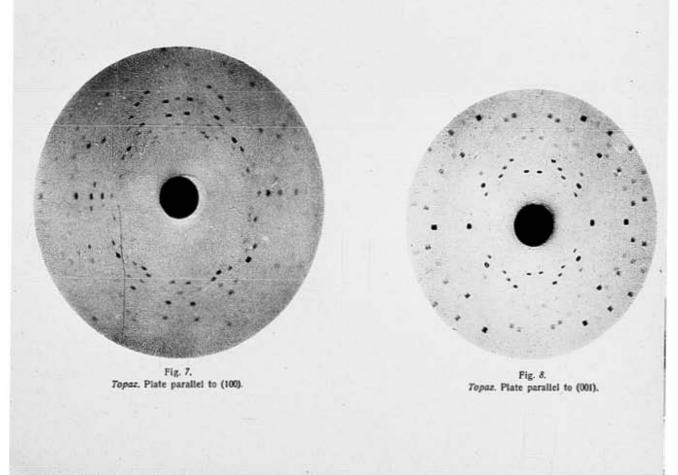
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F. M. JAEGER AND H. HAGA. ON THE SYMMETRY OF THE RÖNTGEN-PATTERNS OF RHOMBIC CRYSTALS. I.

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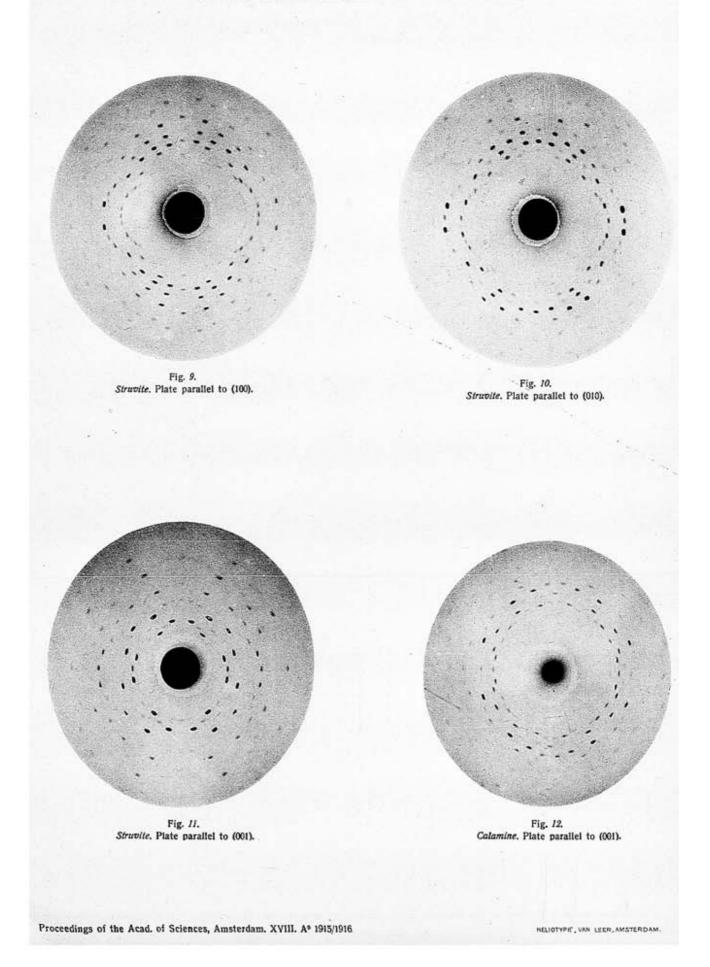
Proceedings of the Acad. of Sciences, Amsterdam. XVIII. At 1915/1916.

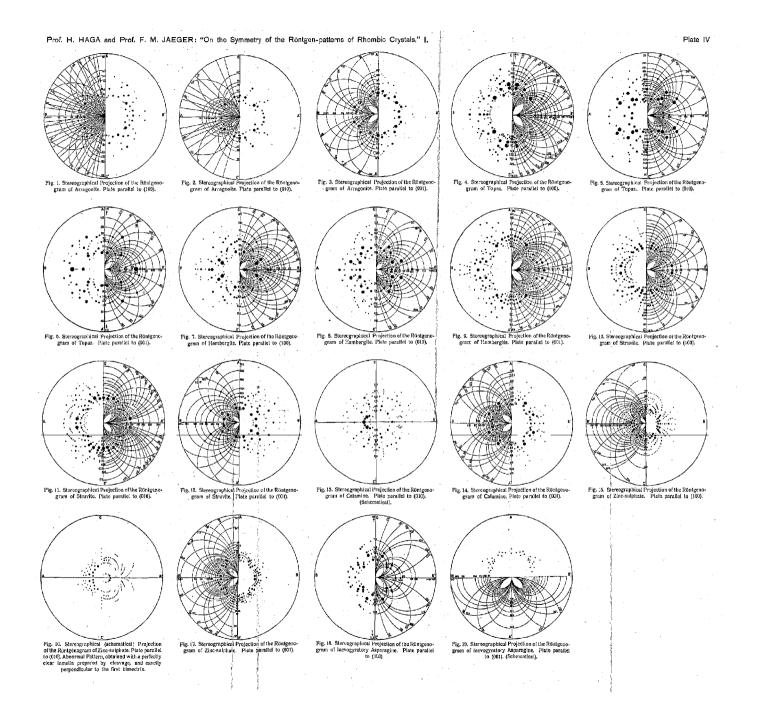
HEUOTYPE, VAN LEER, AMSTERDAM

Fig. 6. Arragonite. Plate parallel to (001).

PLATE II.

PLATE III.





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planes" is by no means diminished by the particular circumstance, that the considered molecular layers are just those, which play the role of directions of perfect cleavability in the crystals. (Thus being perpendicular to the direction of *minimal* cohesion ¹).

i. The RÖNTGEN-patterns of *d*-sodium-ammonium-tartrate, as well the normal as the abnormal ones, and all particulars observed in that case, have been discussed already in detail in our last paper. We can here therefore refer to the resp. figures; only it may be remembered here once more, that the patterns parallel to all three pinacoidal faces, in the normal case appeared to be symmetrical with respect to two perpendicular planes.

k. From big, colourless and perfectly transparent crystals of laevogyratory *asparagine*, crystal-plates parallel to $\{100\}$, $\{010\}$ and $\{001\}$ were cut. The plate parallel to $\{100\}$ had a thickness of 1,21 mm., that parallel to $\{010\}$, of 1,06 mm., and that parallel to $\{001\}$, of 1.22 mm.

The obtained Rontgen-patterns were all too feeble to make a direct reproduction possible. But in fig. 18 and 19 on Plate IV two of their stereographical projections are drawn. Also these images evidently are symmetrical with respect to two perpendicular planes. The third pattern was too disturbed to allow any valuable judgment about this question.

§ 8. From these researches, which will be still completed, it becomes clear even now, that in ordinary cases also with optically biaxial crystals, the theoretical predictions are in full concordance with experience.

The repeatedly observed suppression of one of the two expected planes of symmetry in the RONTGEN-patterns, must be considered also in these cases as a peculiar "abnormality", which undoubtedly is caused by `internal disturbances of molecular dimensions, whose true nature however at this moment cannot yet be more sharply defined.

> University-Laboratories for Physics and for Inorganic and Physical Chemistry.

Groningen, August 1915.

¹) In this connection a remark made a short time ago by P. EHRENFEST (these Proceed. 18. 180 (1915) is of interest, considering the possibility of cleavage along planes, which are "vicinal" with respect to such directions of perfect cleavability.

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