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## Citation:

Haga, H. \& F.M.Jaeger, On the Symmetry of the Röntgen-patterns of Rhombic Crystals, in: KNAW, Proceedings, 18 I, 1915, Amsterdam, 1915, pp. 559-572

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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 ${ }^{1}$ ), the Rontgrn-pātterns of plates from rhombic cry stals of bisphenoidical, pyramidal, or bipyramidal sy mmetry, 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 durection 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 $K C$ the distance of the crystal from that plate. This distance $(=A$ ) was in our experiments always $50 \mathrm{~mm} .{ }^{3}$ ); 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 viewpont of uc projection is 0 ; the planes $X C Y(=P), X C Z$, and $Y C Z$ 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 $C X, C Y$ and $C Z$ 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, $K z$ represents a "possible" zône-axis of the crystal, the value of $\frac{C z}{C K}$ must be: $n \cdot \frac{1}{c}$, where $\frac{1}{c}$ is the parameter-relation in

[^0]

Fig. 1.
the directions $C Y$ and $C Z$, while $n$ is some integer. If $K z^{\prime}$ is equally such a zône-axis, then just in the same way $\frac{C z^{\prime}}{C K}=m \cdot \frac{a}{c}$, where $m$ is also an integer. The projected spots $S$ and $S^{\prime}$, corresponding with the reticular planes $\{101\}$ and $\{011\}$ of the crystal, are therefore in the directions $C X$ and $C Y$ of the stereograptical projection situated in distances from the central spot $C$, of : $2 A \frac{a}{c}$ and $\frac{2 A}{c}$, or in our case of: $\frac{100 a}{c}$ and $\frac{100}{c} \mathrm{~mm}$. In the same way for spots corresponding with the reticular planes of the symbols $\{h o 1\}$ and $\{o k 1\}$, these distances from $C$, become : $\frac{1}{h} \cdot \frac{100 a}{c}$, resp $\cdot \frac{1}{l} \cdot \frac{100}{c}$ m.m.

The point of intersection $M$ of both zône-circles, corresponding with the zône-axes $K z$, resp. $K z^{\prime}$, possessing the symbols [ho1] and [ok 1], represents the stereographical projection of a spot, corresponding with a plane of the structure with the symbol $\left\{2 k l_{\}}\right.$; etc. In this way it is possible to determine the indices of every spot in the
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 spertral dissolvingpower of the different molecular planes is connected with their reticular density.

We have chosen the case of a radiation perpendicular through a plate parallél to $\{001\}$. In the same way however it can be found, that: On $\{100\}$ the spot corresponding with $\{101\}$, is situated in the durection 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 stituated in the direction
of $C$-axis, at a distance of 100 cm .m.
On $\{010\}$ the spot corresponding with $\{110\}$, is struated in the direction
of $A$-axis, at a distance of 100 am m .
all distances reckoned from the image-centre ( ${ }^{1}$ ).
§ 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 erystallography. However it is also possible to construct directly the stereographical projection of the reticular planes, with which the spots on the photo correspond, fiom the stereographical projection of the Rontgen-pattern itseif. The way to do this, is as follows:
The part to the right of fig. 2 relates to the original stereographical projection of the Ronigen-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

[^1]the point $z$ is the centre of a zône-circle CMS, and thus also the point of intersection of the zône-axis $K Q$ with the plane' of the photographic plate $P$, this being in fig. 2 the plane of projection of the stereographical figure to the left.


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

Let now $m$ 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 $K Q$ as their zone-axis. The azimuth of every spot in the plane $P$ and 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 onginal spot, and to pass this straight line
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 $50 \mathrm{~m} . \mathrm{m}$. is used; the contractions of the original distances to $C$, , which thus can be represented by $m z$, - 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 $2 A$ ( $100 \mathrm{~m} . \mathrm{m}$.) ; then it is possible to execute the different constructions by means of Wulerf's stereographical projection plat with a diameter of $20 \mathrm{c} . \mathrm{m}$. 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 detall the Röntarn-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 $100 \mathrm{~m} . \mathrm{m}$. 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 $112.04 \mathrm{~m} . \mathrm{m}$. from $C$
" ", ., ", $B$-axis ," 111.95 ," "
For a plate parallel to $\{010\}$ :
in the direction of the $C$-axis at $100.08 \mathrm{~m} . \mathrm{m}$. from $C$

$$
\text { " " " }, \quad, A \text {-axis , } 89.32 \text { " " ", }
$$

For a plate parallel to $\{001\}$ :
in the direction of the $A$-axis at $89.25 \mathrm{~m} . \mathrm{m}$. from $C$
", ", ", $B$-axis , 99.92 ," ",
If now we calculate from the measured distances of the spots on the photographic plate of anhydrite, using $A=50 \mathrm{~m} . \mathrm{m}$., the values of $\operatorname{tg} 2 r$, then $r$, and finally $100 \operatorname{tg} r$, - 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 $48,3 \mathrm{~mm}$.

Now the thickness $d$ of the crysialplate was in our case precisely $1,64 \mathrm{~mm}$.; and thus we must conclude, that in this case we must use in our calculations of the angles $\rho$, 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 $=50 \mathrm{~mm}$., 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 $O P$ is always kept $=100 \mathrm{~mm}$.; but that ordinarily a sufficient agreement between calculation and experiment would result, if $A$ daring 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 Röntgrin-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).
pholos 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 $\varphi$, and to diminish the distance of $50 \mathrm{~m} . \mathrm{m}$., if rather thick plates are used, with half the thickness of them ${ }^{2}$ ). The projections reproduced on Plate IV of this paper all relate to such stereographical projections, for which the distance $O P$ is $75 \mathrm{~m} . \mathrm{m}$.: in such cases the diameter of the figure is also kept equal to $75 \mathrm{~m} . \mathrm{m}$.

In most of the drawings of Plate IV we have calculated the symbols $\{h k 0\},\{0 k l\}$ and $\{h o l\}$ 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.
${ }^{1}$ ) This is connected with the specific absorbing power for Röntacn-rays of the crystallised material. As this abserption is stronger, the distance of $50 \mathrm{~m} . \mathrm{m}$. will have to be diminished with a smaller part of the thirkness $d$.


Fig. 4. Stereographical Projection of the Rön'rgenogram of Anhydrite. Plate parallel to (010).


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 ${ }^{2}$ ).


§ 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: anhydnite; 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 rhombc-bipyramidal class first ${ }^{2}$ ).
a. Anhydrite $\left(\mathrm{CaSO}_{4}\right)$. The used anhydite-crystal was from Stassfurt. It was lustrous and translucid, and evidently quite homogeneous. Parallel to the three directions of cleavage : $\{100\},\{010\}$ and $\{001\}$,

[^2]three rather thick crystal-plates were prepared. whose thickness w $\varepsilon$ resp. $1.64 \mathrm{~mm} ., 1,72 \mathrm{~mm}$. and $2,09 \mathrm{~mm}$. 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 cas of the anhydrite we used more particularly a Coondage-tube, with volframium-anticathode and separate heating-spiral.

The three photographs are reproduced in fig. 1-3 on Plate ] 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 perpendicilar planes of symmetry; the normal to the plat (direction of radiation) is thus at the same time a binary axis of th Rodetgen-patterns.
b. Arragonite $\left(\mathrm{CaCO}_{3}\right)$. Our clear, lustrous crystals were fror Horschenz in Bohemen. The crystal-plate parallel to $\{100\}$ had thickness of $0,96 \mathrm{~mm}$., that parallel to $\{010\} 1,06 \mathrm{~mm}$., and tha parallel to $\{001\} 1,10 \mathrm{~mm}$. The photos are reproduced in fig. 40 Plale I, and in fig. 5 and 6 on Plate II; their stereographical pro jections in fig. 1--3 on Plate IV. Also in this case the pattern appear to be symmetrical with respect to two planes of symmetr: 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. $\left(A l_{2}(F, O H)_{2} \mathrm{SiO}_{4}\right)$. The topaz-crystal used by us wa very homogeneons, vitreous and translucid; it possessed a yellowisl hue, and originated from Sitwony. The thickness of the three plate parallel to $\{100\},\{010\}$ and $\{001\}$, was from $1,20 \mathrm{~mm}$. to $1,27 \mathrm{~mm}$. ; the time of exposition again two and a half hours. The plate paralle 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 aftes two perpendicular planes, as might be expected from the theory.

To this same class belong furthermore the crystals of cordieritt and of hambergite, already previously ${ }^{1}$ ) discussed.
d. In the case of cordierite the patterns of crystal-plates paralle. 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 anong these were present splendid, lustrous crystals. Only the pattern obtained with a crystal-plate parallel tc $\{001\}$, appeared to exhibit the normal symmetry.

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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 Rontgrn-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 $\left.\cdot\left(\mathrm{NH}_{4}\right) \mathrm{MgPO}_{4}+6 \mathrm{H}_{2} \mathrm{O}\right)$ ), and the calamine: $\mathrm{Zn}_{2}(\mathrm{OH})_{3} \mathrm{SiO}_{3}$.
$f$. 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 $1,26 \mathrm{~mm}$. The time of exposition was two and a half hours.

The three very beautiful Rontgrn-patterns are reproduced in the fig. 9, 10 and 11 on Plate III, and as stereographical projections in fig. $10-12$ on Plate $I V$. Also in these images two planes of symmetry perpendicular to each other are evidently manfested; 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 calcmine we obtained for crystal-plates parallel to $\{100\}$ and $\{010\}$, cut from a very beautiful crystal ${ }^{1}$ ), 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 contaned moreover such a great number of vers small and feeble spots, that also in the projection of fig. 13 on Ylate IV, only the most important spots could be reproduced.

The Ronteenogram 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.

## $\$ 7$. Finally we used from the crystals of the rhombic-bisphenodical

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Anhydrite. Plate parallel to (100).


Fig. 2.
Anhydrite. Plate parallel to (010).


Fig. 3.
Anhydrite. Plate parallel to (001).


Fig. 4.
Arregonite. Plate parallel to (100).


Fig. 5.
Arragonite. Plate parallel to (0to).


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


Fig. 7.
Topaz. Plate parallel to (100).


Fig. 8.
Topaz. Plate parallel to (001).


Fig. 9.
Struvite. Plate parallel to (100).


Fig. 10.
Struvite. Plate parallel to (010).


Fig. $I$.
Struvite. Plate parallel to (001).


Fig. 12.
Calamine. Plate parallel to (001).

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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 asparruyine, crystal-plates parallel to $\{100\},\{010\}$ and $\{001\}$ were cut. The plate parallel to $\{100\}$ had a thickness of $1,21 \mathrm{~mm}$., that parallel to $\{010\}$, of $1,06 \mathrm{~mm}$., 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 Rontgex-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.

[^5]Proceedıngs Royal Acad. Amsterdam. Vol XVIII


[^0]:    $\left.{ }^{1}\right)$ These Proceed. 17, 1204: (1915).
    2) However attention must here be drawn to the remarks in $\S 4$

[^1]:    ${ }^{1}$ ) 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 $C S=2 A \cdot \operatorname{tg} \varphi$ and $C V=A \cdot \operatorname{tg} 2 \varphi$, if $V$ is the original spot, and $S$ its stereographical piojection on $P$. If $C T^{-}(=a)$ is measured on the photography, then $\operatorname{tg} 2 f=0,02 a$; from this $\rho$ and $\operatorname{tg} \rho$ are calculated, and thus $C S=100 \operatorname{tg} \varphi$ is found in m.m..

[^2]:    ${ }^{1}$ ) Note that these numbers relate to a projection distance $O P=100 \mathrm{~m} . \mathrm{m}$.; our figures then have also a diameter of $100 \mathrm{~m} . \mathrm{m}$. But for the drawings on Plate IV, , which are reduced to $3 / 4$ size, all these values need to be also multiplied with $3 / 4$.
    ${ }^{2}$ ) 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: $a: b: c=09804: 1: 0.5631$; topaz: $a: b: c=05985: 1: 0.9539$; struvite: $a: b: c=0.5667: 1: 0.9121$; calamine : $a: b: c=0.7835: 1: 0.4778 ; ~ l$-asparagine : $a: b: c=0.4737: 1: 08327$; sodium-ammonium-tartrate: $a: b: c=0.8233: 1: 0.4200$; hambergite $: a: b: c=0.7988$ : 10.7968.

[^3]:    $\left.{ }^{1}\right)$ These Proceed. 17, 430, 1204. (1915).

[^4]:    ${ }^{1}$ ) For this very beautiful crystal we are much indebted to our colleague, Prof. Molengraaft at Dellt, whom we render our best thanks here once more.

[^5]:    ${ }^{1}$ ) 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 durections of perfect cleavability.

