

Citation:

Haga, H. & F.M.Jaeger, On the Symmetry of the Röntgen-patterns of Trigonal and Hexagonal Crystals, and on Normal and Abnormal Diffraction-Images of birefringent Crystals in general, in: KNAW, Proceedings, 18 I, 1915, Amsterdam, 1915, pp. 542-558

point $D_{1.1}$, therefore, corresponds with the point 1, the points b and $D_{1.2.2}$ with the points 3 and 4 of fig. 5]. When we take a diagonal succession of the phases, then we have, starting from point G : G , $D_{1.2.2}$, L_b , KCl and $D_{1.1}$. In the P, T -diagram the succession of the curves must be, therefore:

$$(G)_b, (D_{1.2.2})_b, (L)_b, (KCl)_b, (D_{1.1})_b$$

As is apparent from fig. 8, this succession has been found also experimentally. We find the metastable parts of these curves (not drawn in fig. 8) by a similar discussion, as has led us to fig. 6.

(To be continued).

Crystallography. — “*On the Symmetry of the RÖNTGEN-patterns of Trigonal and Hexagonal Crystals, and on Normal and Abnormal Diffraction-Images of birefringent Crystals in general.*”

By Prof. H. HAGA and Prof. F. M. JÄGGER.

§ 1. In connection with the peculiar phenomena observed some time ago with respect to a number of RÖNTGEN-patterns of birefringent, and more especially of rhombic crystals¹⁾, we thought it necessary to investigate in a rigorously systematical way, what kind of symmetry would be found in the diffraction-patterns of uniaxial crystals, if radiated through in directions perpendicular to the optical axis. For if the supposition were right, that the suppression of the symmetry-planes expected by theory in the RÖNTGEN-patterns of rhombic crystals were connected in any way with the double refraction, — as was thought at that time by one of us, — then we might expect something of the kind also in the case of the patterns obtained by means of planeparallel sections of uniaxial crystals, if cut parallel to the optical axis, and radiated through in a direction perpendicular to that axis.

To obtain the closest analogy in the orientation with that present in the case of the rhombic crystals, which were always cut parallel to the three pinacoidal faces $\{100\}$, $\{010\}$ and $\{001\}$, we investigated in the case of tetragonal crystals those sections, which were parallel to the first and the second prisms $\{100\}$ and $\{110\}$; in the case of trigonal and hexagonal crystals we used in the same way the sections parallel to the prism-faces $\{10\bar{1}0\}$ and $\{\bar{1}2\bar{1}0\}$. In the last mentioned crystals thus the sections parallel to $\{10\bar{1}0\}$ will be analogous to those parallel to $\{100\}$ in the case of rhombic crystals, the sections parallel to $\{\bar{1}2\bar{1}0\}$ corresponding in the same way to those parallel to $\{010\}$ in the mentioned biaxial crystals.

¹⁾ These Proceedings, 17, 1204, (1915),

To deduce the symmetry of the RÖNTGEN-patterns of these crystal-sections, as it may be expected after the theory of the phenomenon, it must be kept in mind, that this symmetry will be the same, as in the case of the corresponding sections of a fictive crystal, whose symmetry would be that of the investigated crystal after addition of the symmetry-centre there-to. Indeed, for the phenomenon of the RÖNTGEN-radiation the symmetry-centre would play the rôle of "additive" symmetry-element; and inversely this supposition may be judged satisfactorily proved, if the experiments will show on the other hand a complete concordance between the facts and the theoretical deduction.

In the accompanying table therefore the theoretically expected symmetry of the RÖNTGEN-patterns, as deduced from the now adopted theory, is summarized for all the optically uniaxial crystals from the classes 9 to 27. From this table the expected symmetry of the diffraction-image for all uniaxial crystals can immediately be seen.

§ 2. In the following pages we publish the results obtained in

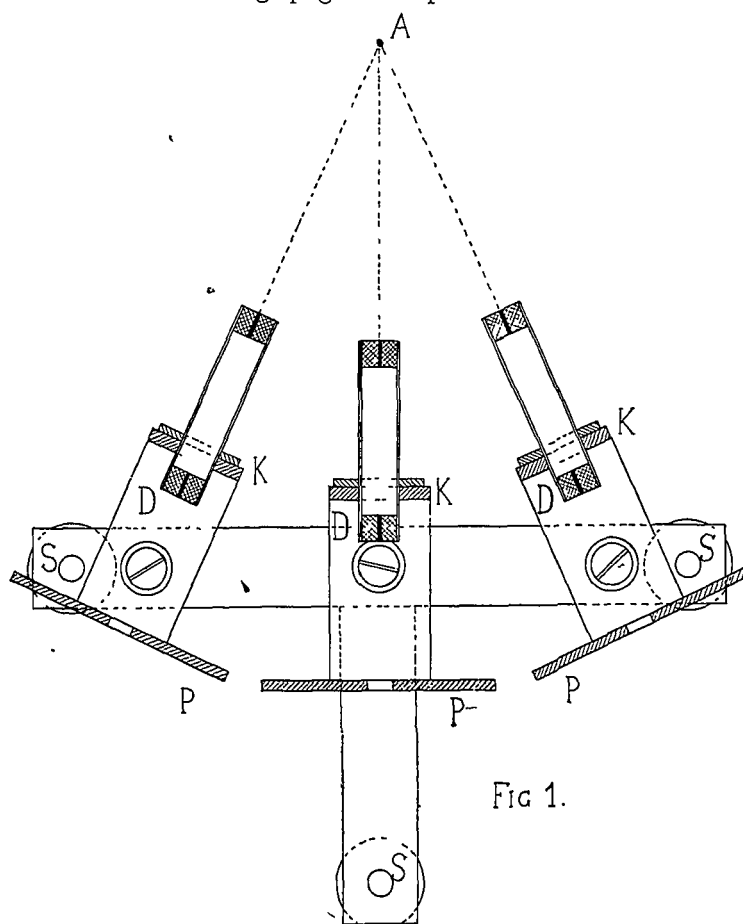


FIG 1.

the study of trigonal and hexagonal crystals; the data relating to the investigations made with tetragonal crystals will be published by us later-on in a separate communication.

Most of these researches were executed by means of RÖNTGEN-tubes with platinum-anticathode, some of them, however, by the aid of the COOLIDGE-tube with wolframium-anticathode and separate heating-coil. In most of these experiments we used an apparatus, which enabled us to make *three* RÖNTGENograms (in the case of rhombic crystals, by radiation along the three principal crystallographical axes, or perpendicular to the first and second prism) *at the same time.* This apparatus was arranged in the following way (vid. the horizontal projection in fig. 1 p. 543).

On a T-shaped brass support, provided with three levelling-screws *S*, (dimensions: 3 c.m. broad, 1 c.m. thick, longer beam: 28 c.m., shorter beam: 12,5 c.m.), three similar "crystal- and plate-holders" *D* (vid. also fig. 2) were fixed in the right position by means of strong screws. Every one of these bearers (fig. 2) consists of a brass

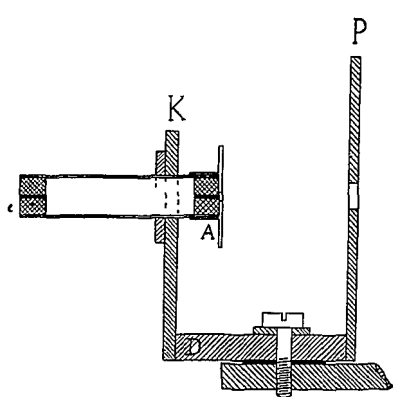


FIG 2.

bar *D* of 1 c.m. thickness, whose limiting faces are turned on the lathe perfectly rectangularly and parallel to each other. At the one end is fixed the likewise rectangularly turned-off plate-holder *P*, — whose dimensions are 9,5 c.m. broad, 12 c.m. high and 3 m.m. thick; at the other end, however, the special crystal-support *K* (high: 9 c.m., broad: 4 c.m. and thick: 5 m.m.) was immovably fixed by good screws. In a hole in *K* a brass tube of 8 c.m. length is fixed, which is closed at both ends by two lead-cylinders *e* of 1 c.m. length, these being pierced along their axes by a straight canal of 1 m.m. diameter. An accurately fitting cover *A* (fig. 2) can be pushed on that end of the brass tube, which is next to *P*; its front face consists of a small brass plate with a central hole of 2 m.m. diameter.

The crystals were smoothly pressed against this brass plate, and then held in position by means of sticking-wax.

As a result of the careful finishing of this apparatus, one could be sure, that the RÖNTGEN-rays, after having passed the small canals in the lead cylinders, progress in a thin pencil, which is perpendicular as well to the crystal-plate, as to the photographic plate.

The dimensions are chosen in such a way, that the distance from the front face of the cover to the sensitive film in P is precisely 50 m.m.; of course the thickness of the fluorescent screen and of the two black paper covers, with which the plate and screen are protected, are taken into account here.

The photographic plate, with the fluorescent "Eresco"-screen pressed against the sensitive film, was wrapped in two covers of black paper and then firmly pressed against P ; it had an opening measuring 8×8 cm., and the whole apparatus thus was held together much in the same way as in the case of a photographic copying-press.

The three plate-bearers D could be adjusted into the right position with sufficient accuracy by means of three straight, thin knitting-needles, which after being pushed through the canals in the lead cylinders, must meet in the same point A . For the purpose of making the anti-cathode coincide with this point A , the wooden bearer of the RÖNTGEN-tube was fixed on a heavy brass support, which had smoothly running sliding-motions in three perpendicular directions; thus it was made possible, to fix the RÖNTGEN-tube exactly in such position that the three pencils of RÖNTGEN-rays generated three equally strongly luminous little spots on a fluorescent screen, which was placed behind P . In the plates P three central holes of 1 cm. diameter were bored to enable us to see these luminous spots. To protect the photographic plate against undesired attack by direct or secondary RÖNTGEN-rays, some larger lead screens were interposed between the RÖNTGEN-tube and the plate-holders with a total thickness of 2 c.m.; in the same way the three crystal-, and plate-holders themselves were surrounded by a lead cover, which could be closely fitted to the large lead screens. In the backside face of the lead cover three holes were bored, big enough to let the undiffracted RÖNTGEN-rays freely pass.

An inconvenience, met with in our former experiments when using the fluorescent screen, was the abnormal size of the central spot on the photos, which spot would even seem still larger in the reproductions from the negatives ¹⁾. The extension of this spot must be caused by the action of the secondary RÖNTGEN-rays, which were produced by the passing of the undiffracted pencil through the glass and the sensitive film; these secondary rays will provoke a rather strong fluorescence of the vicinal parts of the screen and thus an intense

¹⁾ The diameter of the image of the undiffracted rays was about 2 m.m., as can also be calculated from the used dimensions of our apparatus: by photographic irradiation or by the mentioned secondary rays however, the central spot on the photos appeared to be about 8 m.m. in some cases.

action on that place of the photographic plate. We were able to eliminate this obstacle for the greater part, by cutting from the centre of the screen a small disc of about 1 c.m. diameter, and to cover the inner rim of the hole with a layer of black ink. On the photo however a very small halo was still visible in some cases; but this could be easily removed by covering the central part of the negative during the reproduction with a small disc of black paper. In this way the disturbance of the image by the above mentioned causes was finally completely prevented.

§ 3. From the representative of each crystal-class, necessary for our purpose, not all could be obtained in a sufficiently excellent quality, or they could not be used from some other cause in our experiments.

So for instance the *sodium-periodate*-crystals were unsuitable, because of their very rapidly occurring efflorescence and loss of their water of crystallisation; the crystals of *benzil* on the other hand appeared to show optical anomalies and peculiar phenomena to be described in a later communication. Notwithstanding much trouble it was impossible to obtain larger crystals of *cinnabar*, which were not at the same time twins or appeared to be too inhomogeneous. From *zincite* we could have only badly disturbed and lamellar crystals; in the case of *nephelite* the obtained crystals still appeared finally to be polysynthetic twins, notwithstanding the choice of very small, clear-looking individuals.

Completely reliable results we obtained finally in the case of the following minerals: *phenakite*, *dolomite*, *quartz*, *turmaline*, *calcite*, *apatite* and *beryl*, while also our experience with some *nephelite*-preparations, and with *cinnabar* cut perpendicularly to the *c*-axis, can be judged as to be in agreement with the theoretical deduction.

§ 4. Description of the examined substances.

a. Turmaline. For our observations we used a beautiful, dark green turmaline-crystal of Brazil. The image obtained by radiation through the direction of the optical axis, was already formerly reproduced¹⁾; it possesses the expected symmetry, namely: one ternary principal axis and three vertical symmetry-planes (vid. the stereographical projection in fig. 1, Plate VI).

The first crystal-plate parallel to {1010} had a thickness of 3,05 m.m.; a second one however only of 1,15 m.m. Both images (vid. Plate I, fig. 1 and 2, and Plate VI fig. 2.) show only one

¹⁾ Vid. these Proceedings, 17. 1204. (1915); Plate I, fig. 4; Plate IV, fig. 4.

single plane of symmetry, perpendicular to the prism-face. The spots in the image of the thick crystal-plate are very heavy and *not* oval-shaped, but *rectangular*. We have already drawn attention to this phenomenon on a previous occasion, in the cases of *sodium-chlorate*, of *sylvine*¹⁾, etc.

It now becomes clear that it is principally connected with the *thickness* of the crystal-plate: the formerly described patterns of sodiumchlorate and sylvine are indeed also obtained by means of *very thick* plates.

This peculiarity was also stated by us in many other cases, if thicker plates of not very strongly absorbing substances were used in the experiments; often the spots appear to be double ones in such cases, which by joining finally give the impression of a more or less rounded rectangular shape. We think that an explanation can be given in this way: that in the case of not powerfully absorbing substances so great a number of successive molecular layers contribute to the intensity of the spot on the photographic plate, that the images of the outer layers of the whole pile will appear in a discernible distance from each other on the film, because of the different distance of these outer layers from the sensitive plate. If the spots thus properly produced will coalesce with each other, the rounded rectangular shape of the resulting image is easily explained.

The fourth turmaline-plate was cut parallel to $\{\bar{1}2\bar{1}0\}$; the RÖNTGEN-pattern shows as a single symmetry-element, a binary axis coinciding with the plate-normal. (Plate I, fig. 3). The results of the experiments are therefore in this case in complete accordance with those of the theoretical deductions.

b. Phenakite. We had at our disposition very beautiful, colourless and lustrous *phenakite*-crystals from *San Miguel, Minas Gerais, in Brazil*.

The crystal plate cut perpendicularly to the *c*-axis, showed in convergent polarized light, a uniaxial interference-image of positive character; it manifested however a small abnormality in the form of a feebly biaxial image with extremely small axial angle. However this abnormality did not appear to have any influence on the diffraction-pattern. The plate had a thickness of 1,1 mm.; the photographic image was not very beautiful, and the most important spots appeared to be covered by the strong irradiation of the central spot. Later-on we obtained by means of our newer apparatus described previously, a feeble but completely symmetrical image, which was used in the

¹⁾ Ibid. 1207, note 1.

construction of the stereographical projection in Plate VI, fig. 3. Evidently there is only one ternary axis present, but no planes of symmetry in the pattern.

The plate parallel to $\{10\bar{1}0\}$ was 1,20 m.m., that parallel to $\{\bar{1}2\bar{1}0\}$ was 1,15 m.m. thick; we obtained with them two very beautiful photos, reproduced in Plate I, fig. 4 and Plate II, fig. 5; in these photograms the direction of the c -axis is vertical. The diffraction-patterns are wholly unsymmetrical; the results are therefore exactly what could be expected from the theory.

c. In the same symmetry-group also *Dolomite* must be placed. From a splendid, perfectly translucent crystal of *Binnenthal* in *Switzerland*, three faultless plates parallel to $\{0001\}$, $\{10\bar{1}0\}$ and $\{\bar{1}2\bar{1}0\}$ were carefully cut. The plate perpendicular to the c -axis had a thickness of 0,92 m.m.; the beautiful interference-image of negative character appeared to be exactly central. The plate parallel to $\{10\bar{1}0\}$ was 1,14 m.m. thick; that parallel to $\{\bar{1}2\bar{1}0\}$ was 1,11 mm.

The very beautiful diffraction-patterns obtained are reproduced in fig. 6, 7 and 8 on Plate II, and in stereographical projection on Plate VI, in fig. 4 to 6. The image perpendicular to the c -axis possesses only a ternary axis; both the other images are completely unsymmetrical, just as in the case of *phenakite*. Also in this case therefore experience and theoretical deduction are in full agreement with each other.

d. *Calcite*. From a lustrous calcite-crystal from *Iceland* two plates were cut: the plate parallel to $\{10\bar{1}0\}$, as well that parallel to $\{\bar{1}2\bar{1}0\}$ were 1,15 m.m. thick. Both images were too feeble to allow good reproduction; they are however reproduced as stereographical projections in fig. 7 and 8 on Plate VI. The RÖNTGEN-pattern for a plate perpendicular to the c -axis was published some time ago by BRAGG ¹⁾: the image exhibits a ternary axis and three vertical planes of symmetry. The symmetry of all these patterns is the same, as was found in the case of the *turmaline*, — just as could be expected from the theory. It must be remarked that the image parallel to $\{10\bar{1}0\}$, although possessing only a single (vertical) plane of symmetry, shows, however, a very strong approximation to a case, where two perpendicular symmetry-planes were present.

e. *Beryl*. We had very beautiful plates at our disposition, cut from a splendid, colourless, translucent crystal from the *Aduntschilom*-mountains in the *Transbaical*. The plate parallel to $\{0001\}$ had a

¹⁾ W. L. BRAGG Vid. Zeits. f. Anorg. Chem. **90**. 206. (1914); Proc. Royal Soc. A. **89** 248. (1913).

thickness of 1.10 mm., that parallel to $\{10\bar{1}0\}$ 1.17 mm., and that parallel to $\{\bar{1}2\bar{1}0\}$ 1.16 mm.

The diffraction-image parallel to $\{0001\}$ (vid. Plate III, fig. 9), shows a senary axis and six vertical planes of symmetry. Thus it is again proved, that the *beryl* is really *dihexagonal*, and that the arguments against this supposition, formerly brought to the fore by VIOLA¹), can hardly be considered as valuable any more.

The two remaining images (Vid. Plate III, fig. 10 and Plate IV, fig. 11) are, quite in concordance with the theory, symmetrical after two perpendicular planes of symmetry. They are reproduced as stereographical projections in fig. 9—11, on Plate VI. The image of the plate parallel to $\{10\bar{1}0\}$ appears to be somewhat sloping, evidently caused by not wholly perfect orientation of the crystal-section.

f. Apatite. From a beautiful crystal from *Zillerthal*, in *Tyrol*, two plates were cut. The image of the plate parallel to $\{0001\}$ was reproduced already previously²). The second plate was parallel to $\{10\bar{1}0\}$; its thickness was 1,30 mm. The beautiful diffraction-pattern is reproduced in fig. 12 on Plate IV, and both images as stereographical projections on Plate VI, fig. 12 and 13. The pattern parallel to $\{10\bar{1}0\}$ exhibits only one horizontal plane of symmetry, quite in agreement with the theoretical expectations.

g. Quartz. From a translucent crystal from the *St. Gothard* four plates were cut. The image of a plate perpendicular to the *c*-axis was too feeble to make reproduction by any means possible. A schematic drawing of the most important, — and always *double*, — spots, is given in fig. 14, Plate VII. The pattern shows a ternary axis and three vertical planes of symmetry.

Two different plates, each of which was parallel to $\{\bar{1}2\bar{1}0\}$, and having a thickness of 1,12, resp. 1,05 m.m., gave particularly remarkable patterns. For although both plates were very accurately orientated, and did not manifest, with the microscope, any differences, nor any inhomogeneity discernible by optical means, — the image obtained with the first mentioned plate appeared to be *symmetrical after two perpendicular planes*; the other image however, notwithstanding its being composed of precisely the same spots, showed quite another distribution of their intensities, in such a way, that the pattern was *only symmetrical after a single binary axis*. On repeating the experiment with the first-mentioned quartz-plate, which

¹) VIOLA. Z. f. Kryst. 34. 381. (1901).

²) loco cit. 17. Plate I.

now was radiated through in another place, its abnormal symmetry was found once more.

Here now we could, for the first time, observe in the case of a uniaxial crystal a very particular abnormality: indeed it appears, that properly a plane of symmetry perpendicular to the trigonal axis seems to be added to the crystal, which involves at the same time the addition of three new vertical-planes of symmetry passing through the *c*-axis, making this axis necessarily a *senary* one. In the original negatives this different symmetry in both cases is very evident, somewhat less, however, in the reproductions (Plate IV, fig. 13 and 14); but the differences between the normal and the abnormal pattern are clearly expressed in the stereographical projections, which here are given together in fig. 3 and 4.

The same abnormality, i.e. the addition of a horizontal plane of symmetry perpendicular to the ternary axis, seems to be also present in the RÖNTGEN-ogram, obtained with a crystal-plate parallel to $\{10\bar{1}0\}$; this plate had a thickness of 1,10 m.m. Although this plate was parallel to the *c*-axis, it appeared to be not completely parallel to the prismface; the pattern, which therefore very probably did not show a true *vertical* symmetry-plane, is here not reproduced. The stereographical projection of the normal patterns are given in fig. 14, 15 and 16, Plate VII.

A careful microscopical examination of both the plates parallel to $\{\bar{1}2\bar{1}0\}$, did however *not* reveal any optical differences.

One might be inclined to suppose, that the plate parallel to $(\bar{1}2\bar{1}0)$ which had given the abnormal pattern, were really a twin-formation after the brasilian rule; i.e. with a plane of $(\bar{1}2\bar{1}0)$ being the twinning-plane. Because perpendicularly to $(\bar{1}2\bar{1}0)$ there is a binary axis present, the RÖNTGEN-ogram therefore should indeed show a symmetry with respect to two planes, perpendicular to each other. But by this supposition it could never be made evident, that the diffraction-pattern obtained with a plate cut from *the same* crystal parallel to $(10\bar{1}0)$, shows very probably *also a horizontal plane of symmetry*. Thus the said explanation can hardly be considered a final one already for the peculiar RÖNTGEN-patterns which were obtained parallel to $(12\bar{1}0)$ and to $(\bar{1}0\bar{1}0)$. The observed abnormality therefore cannot be said to be explained fully, and we intend to make further experiments on this phenomenon in future.

h. Nephelite. From a small, clear crystal from the *Vesuvius*, three crystal-plates were investigated. The plate perpendicular to the optical axis showed a well-centred, uniaxial interference-image, possessing only a slightly abnormal character. The plate had a

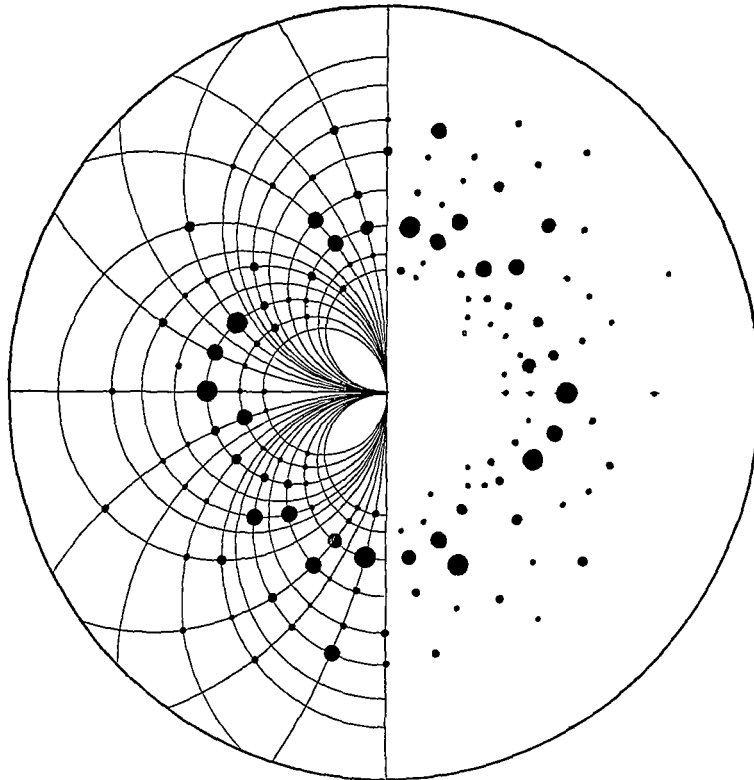


Plate 3. Stereographical Projection of the Röntgen-pattern of dextrogyratory Quartz. Plate parallel to $(\bar{1}2\bar{1}0)$. (Normal Image).

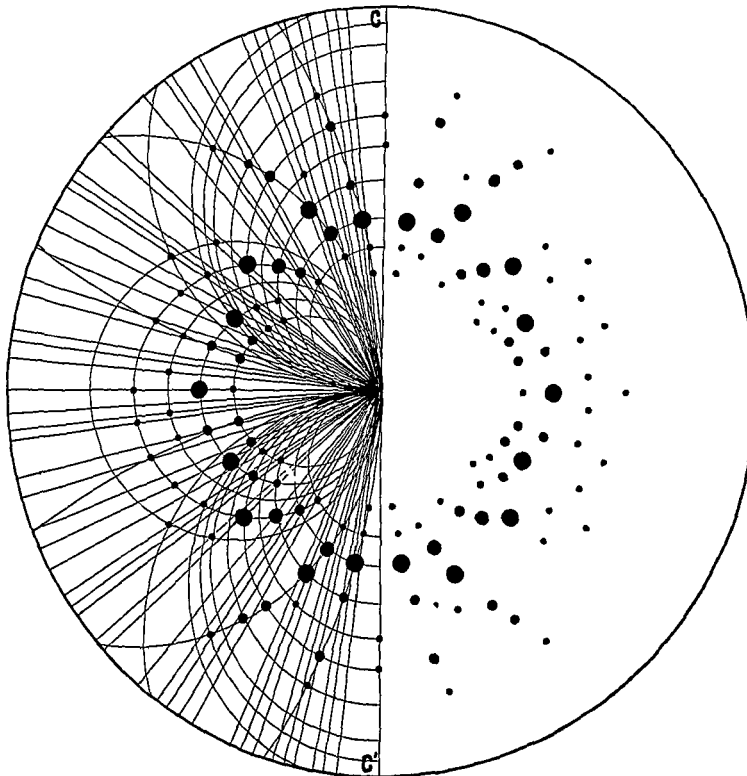


Fig. 4. Stereographical Projection of the Röntgen-pattern of dextrogyratory Quartz. Plate parallel to $(\bar{1}2\bar{1}0)$. (Abnormal Image).

thickness of 0.70 m.m. The obtained diffraction-image was extremely feeble: the spots, which, — as in the case of the quartz, — were all *double-ones*, — were situated very far from the centre and were so feeble, as to make any reproduction impossible. It was however possible to see, that the pattern possessed a senary axis (schematical projection in fig. 17, Plate VII); *no* vertical planes of symmetry were present.

The plate parallel to $\{10\bar{1}0\}$ was 0,78 m.m. thick, and gave a rather good image, which as a stereographical projection is reproduced on Plate VII, fig. 18. All spots here were also doubled, and the axes of the oval impressions were inclined to each other, giving to each pair of spots the shape of an arrow-point; this seems to indicate a twin-formation of the used mineral. The pattern was merely symmetrical after a horizontal plane. The third plate was too disturbed and inhomogeneous, to give any suitable image.

i. That *cinnabar*, if radiated through in the direction of the *c*-axis, will give a RONTGEN-pattern, whose symmetry is in full concordance with the theory, was already formerly recorded¹⁾. The stereographical projection of the RONTGEN-ogram is reproduced here once more in fig. 19, Plate VII. Finally in fig. 15, Plate V, the very beautiful photo of *pennine* is reproduced; although this mineral is only *mimetic* and clearly shows optical abnormalities, the structure of the lamellae is evidently here a so regular and perfect one, that the whole pile cannot be distinguished from a real trigonal crystal. Attention must be drawn to the remarkable fact, just as formerly stated in the case of *sylvine*, that the central spot seems to irradiate in about eighteen directions; it seems, that this irradiation is connected in some way with the presence of certain gliding-planes in the crystal. The thickness of the dark green, positively birefringent, and clearly optically anomalous crystal-plate, was 0,81 m.m.

§ 4. If we now review all the results hitherto obtained in these researches, it becomes clear, that, — with the exception of the phenomena observed in the case of the two quartz-plates, which phenomena undoubtedly are to be considered as true "abnormalities", — *the symmetry of the RONTGEN-patterns is always in agreement with that predicted by the now adopted theory of the diffraction-phenomenon.* On the other hand the correctness of the supposed *central symmetry* of the said phenomenon is thus sufficiently proved in this way. Our experience can be considered evidently as a strong argumenta-

¹⁾ These Proceed. 17. p. 1204: vid. Plate IV, fig. 5.

tion *against* the supposition, that the particular fact of the disappearance of certain symmetry-planes in the RÖNTGEN-patterns of birefringent crystals would have anything to do with their optical anisotropy. For if this were true, it would be hardly possible to understand, why not one of the numerous patterns of uniaxial crystals, which were radiated through in the direction of their optical axis, and thus likewise are birefringent plates, exhibited the formerly described phenomenon. On the other hand the case of the quartz-images makes prudence necessary: for evidently the symmetry of the patterns can by yet partially unknown *secondary* causes, appear *otherwise* than may be expected from the theory of the diffraction phenomena, — as well of *higher* symmetry (quartz) as of *lower* symmetry (rhombic crystals).

§ 5. In connection with these considerations, we have recommenced our studies with some optically biaxial (rhombic) crystals, and have begun with a renewed investigation of *the same*, translucent and very beautiful plate of *hambergite* parallel to {010}, which formerly¹⁾ had given a so strongly abnormal image. After having radiated through in another place, we now repeatedly obtained a perfectly normal diffraction-image, quite symmetrical after two perpendicular planes. The normal pattern is reproduced in fig. 16, Plate V, as a photo, and both images by the side of each other as stereographical projections, in fig. 5 and 6. Using the normal image as standard, it may be called very remarkable, that the abnormal image appears in comparison to it as a “distorted” normal pattern, as if the crystal-plate were rotated round the vertical principal direction at a certain angle. Very striking indeed is the completely different intensity-distribution of the spots, and also their altered position in both cases. Microscopically *no* differences could be found in the one place of the plate and the other: with a very strong enlargement the crystals showed certainly very small and long-shaped inclusions, but these were in precisely the same way and arrangement present also in both the other *hambergite*-plates, cut parallel to {100} and {001}, which plates however in striking contrast to the one mentioned, *always gave completely normal* patterns. From this it must follow, that these inclusions cannot be the cause of the phenomenon observed.

On repeating our experiments with *the same* plates of *sodium-ammoniumtartrate*, as we used formerly, we obtained with the, — now superficially somewhat rougher, — plate parallel to {100}, *the same abnormal* pattern as previously: only a few of the spots

¹⁾ These Proceed. 17, 1204, Plate II, fig. 8; Plate IV, fig. 11.

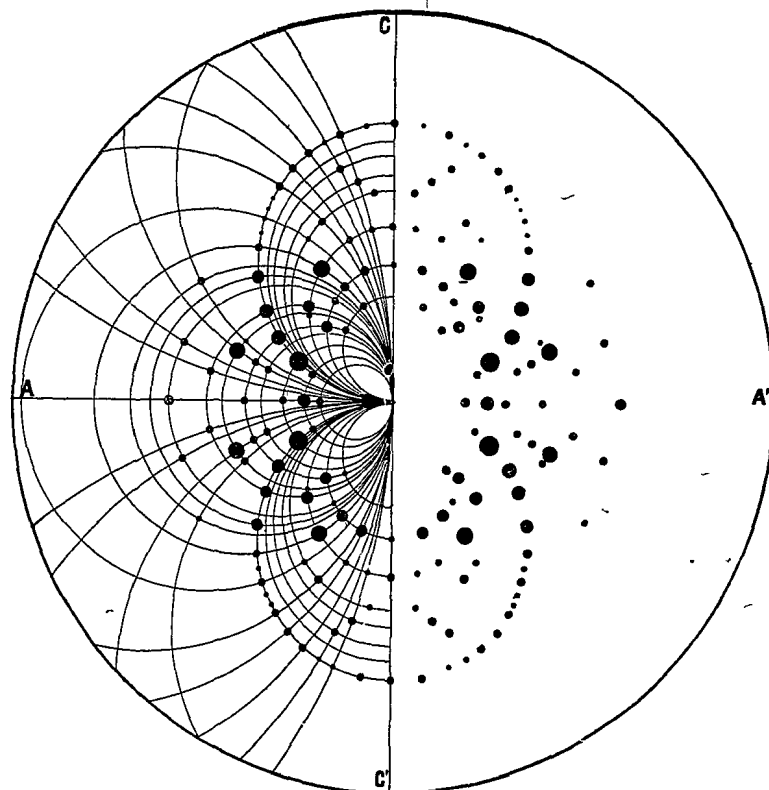


Fig. 5. Stereographical Projection of the Röntgen-pattern of Hambergite. Plate parallel to 010). (Normal Image).

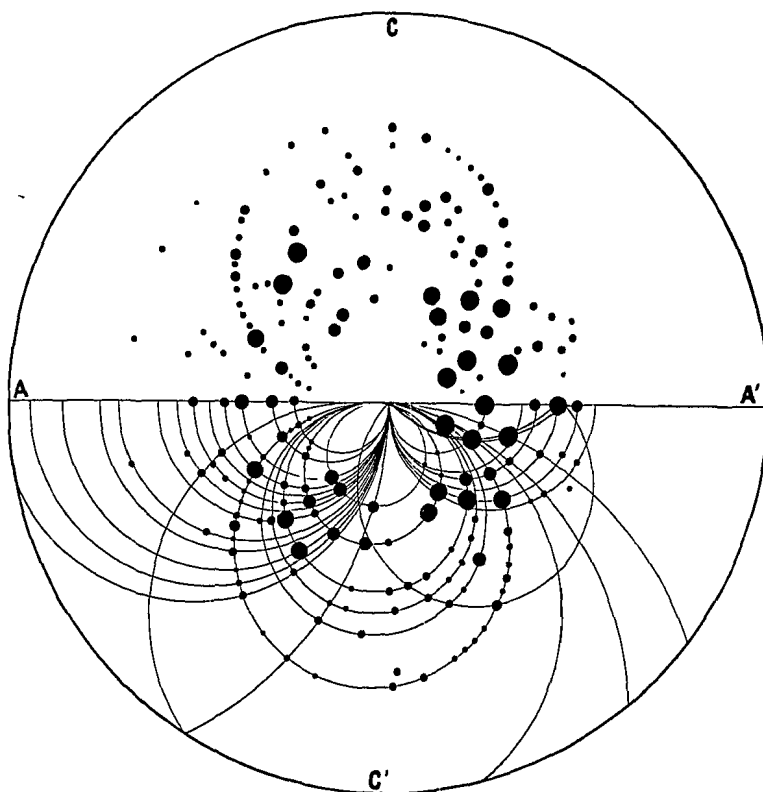


Fig. 6. Stereographical Projection of the Röntgen-pattern of Hambergite. Plate parallel to (010). (Abnormal Image).

appeared to be absent on comparison with the former image. The also superficially somewhat rougher plate parallel to {010} however, gave now undoubtedly also an *abnormal* image, but as a very remarkable fact: *just the other (second) plane of symmetry as before was now manifested in the pattern, notwithstanding the same position of the crystalplate!* Formerly this plate had given an abnormal image, which was symmetrical after the plane {001}; now it showed a symmetry-plane parallel to {100}. As both plates of the *tartrate* were superficially a little altered by a feeble efflorescence, we prepared from a fresh, translucent crystal of the salt three new plates, which were examined in the same way.

With these plates we were now able to obtain RONTGEN-patterns, which were symmetrical after *two* planes; in this way it was possible at the same time to compare the partial symmetry of both the abnormal images parallel to {010} with the normal pattern. The three normal and the abnormal images are reproduced as stereographical projections in fig. 20 to 25 on Plate VII.

§ 6. We have convinced ourselves by especially arranged experiments of the fact that a deviation from the true plane-parallel shape of the *hambergite*-plate could *not* be the cause of the partial symmetry of its pattern formerly obtained. Moreover, in our numerous experiments with *cordierite*, with material from all localities, we *never* obtained other images than *abnormal* ones, which were only symmetrical after one single plane of symmetry. With this mineral the phenomenon thus seems to manifest itself *constantly*. In our way of experimenting, with the use of fixed, on the lathe rectangularly turned-off crystal-supports, a somewhat appreciable deviation from the true orientation is highly improbable. Moreover the same crystals, adjusted by the same apparatus, appeared formerly often to give quite normal patterns, if radiated through in one or two of the other principal directions, so that a *systematical* error of the whole arrangement can hardly be considered to be the cause of the phenomenon. If this were true, or if deviations in the right orientation of the prepared crystal-sections were the cause of the phenomenon observed, it could furthermore not be understood, why *never* a distortion of the normal pattern after *another* direction than only after the two principal ones of the plate, were till now observed. The fact, that the planes of symmetry of the rhombic crystals just play the preponderant rôle in this, proves sufficiently, that *no accidental* causes are responsible here, but that these are of such

a nature, as to be connected intimately with the proper, internal molecular structure of the crystals.

But a further and persuasive illustration of this question is given also by the case of the rhombic *zinc-sulphate*. Here we used a splendid transparent crystal-plate, obtained by direct *cleavage* of the crystal along the plane of *perfect* cleavability {010}, whose perfectly right orientation could be controlled very rigorously by optical examination, the *b*-axis being at the same time the first bisectrix. Notwithstanding this, however, the corresponding diffraction-image appeared to be constantly *abnormal*, and to possess only one single plane of symmetry parallel to {001}, — i. e. parallel to the optical axial plane. (Vid. Plate V, fig. 17 and Plate VII, fig. 26).

The above mentioned observations undoubtedly must bring the conviction, that *the cause of the observed phenomenon must be ascribed to the crystal-plates themselves, — faultless as they may appear even on more detailed examination. Indeed further experiments* taught us, that also with other rhombic crystals than with *hambergite*, it is eventually possible to obtain *perfectly normal* patterns, with the aid of the same apparatus. In the following paper we will reproduce the photos and projections of the images, which we obtained with the plates of a number of biaxial minerals and artificial substances, cut parallel to the three pinacoidal faces. They will, besides some new cases of abnormal diffraction-patterns, also show many, which indeed must be judged to be quite “normal” ones; the fundamental exactness of the original theory thus being convincingly proved. As Prof. RINNE of *Leipzig*, who supposed already some time ago, that special secondary causes might be connected with the observed phenomena, wrote to one of us, — he obtained in the case of the *anhydrite* as well normal as abnormal diffraction-images, and with *calamine* parallel to {010} only abnormal ones. With respect to our own results with these minerals, we can refer here to the following paper.

§ 7. As a result of our completed experiences, we finally can make the statement, *that the now adopted theory of the diffraction-phenomena, really can describe sufficiently the general behaviour of crystals with respect to RÖNTGEN-rays; and that the peculiar partial symmetry of the RÖNTGEN-patterns, as observed till now in many cases and especially with rhombic crystals, must be caused by secondary circumstances, connected with a particular kind of disturbances of the internal molecular structure of the crystals, and which at the moment can be examined by no other physical means.*

Of course the question immediately arises; of what kind are these causes? On deviations in the right orientation of the crystal-plates, — (which are always present in a less or higher degree), — it is hardly necessary to expatiate: after a longer practice one learns to evaluate quite exactly the smaller and very typical distortions, arising from that source, and to pass over them as over the typographical errors in an ordinary text. But the anomalies here considered *are of a totally different order*; they must be caused by a breaking-up of the stratographic position of the molecular layers, by which certain parts of the parallel planes of the molecules will be locally rotated round one of the principal directions in the crystal, — in an analogous way, as on our earth the inversions and the folding of geological strata may be observed. But in every case these disturbances must be here *of molecular dimensions*; they can evidently not be studied or observed by other available means at the moment than by the RÖNTGEN-radiation, because the crystal-medium, disturbed in its molecular relations, behaves in respect to all other known physical actions like a continuum, with exception just in respect to the extremely small wave-lengths of the RÖNTGEN-rays.

If there are present in rhombic crystals some directions of higher or less perfect cleavability, which are parallel to the principal sections of the crystal, then it will be probable, that such “internal vicinal planes” of the molecular layers will appear to be turned exactly round these principal cleavage-directions as axes, — here round the one, and there round the second of them. It will then depend on the place, where the crystal will be radiated through, if the diffraction-image will show a symmetry after the one or after the perpendicular plane. It must be remarked here however, that exactly in the case of the *sodiumammoniumtartrate*, where the mentioned phenomenon was observed by us, *no* such directions of typical cleavability are present. It seems therefore, that the principal directions of the molecular structure can play this remarkable rôle also in the case, that they are *not* at the same time directions of distinct cleavability.

§ 8. We do not deny, that the explanation given here has some weak points, especially if it must be supposed, that *all* molecular layers, contained in the whole thickness of the crystal-plate, contribute their part to the final impression on the photographic film, while notwithstanding that, only for a certain number of these molecular layers the presence of such “internal vicinal planes” can be accepted, because otherwise they would manifest themselves at

the surface of the crystal-plates in some typical way, e.g. as irregularities of that surface. In this connection it may be of interest to mention the fact, that really in some few cases we found such abnormal phenomena with crystalplates, cut parallel to some of such "striated" faces of the crystal.

Moreover the question may arise: why is this abnormal behaviour observed relatively so *often* in the case of *biaxial* crystals, while it occurs evidently hardly ever in the case of uniaxial crystals?

Finally we may yet draw attention to the following case: If a *pseudo-symmetrical* (mimetic) crystal is built-up by lower-symmetrical lamellae, it cannot a priori be understood, why such a combination, radiated through in the direction of the (new) optical axis, would in any way manifest its polysynthetic twin-structure. Indeed this conclusion appears to be verified here by our experience with the *penmine*. But if that lamellar structure can cause in any way the presence of such "internal vicinal planes", so that the molecular layers can be turned a little round these two, three, four or six directions of intergrowth, the possibility can then be foreseen, that these irregularities will be brought *accidentally* in one of these directions more strongly to the fore, than in the remaining ones: *that one direction will then appear in the diffraction-pattern as a single plane of symmetry of it*, and in this way the appearance of this can be considered to be an indirect proof for the lamellar structure of the investigated crystal. This was evidently the case with *apophyllite*¹⁾, *benitoite*²⁾, and the *racemic triethylenediamine-cobalti-bromide*³⁾; moreover we found it a short time ago also in the case of *benzil*, if cut perpendicular to the optical axis of the pseudo-trigonal complex. We expect to elucidate in every case these questions by systematical experiments, and especially to determine finally the true nature of these internal disturbances, evidently intimately connected with the normal molecular structure.

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

Groningen, August 1915.

¹⁾ These Proceed. **4**. 438.

²⁾ Ibidem, **17**. 1204. (1915), Plate IV, fig. 14. (1915).

³⁾ Ibidem, **18**. 50. (1915).

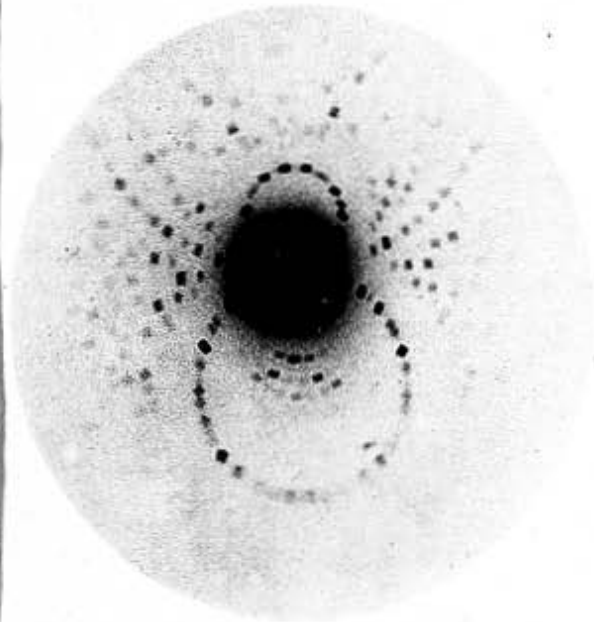


Fig. 1.
Turmaline. Plate parallel to $(01\bar{1}0)$.

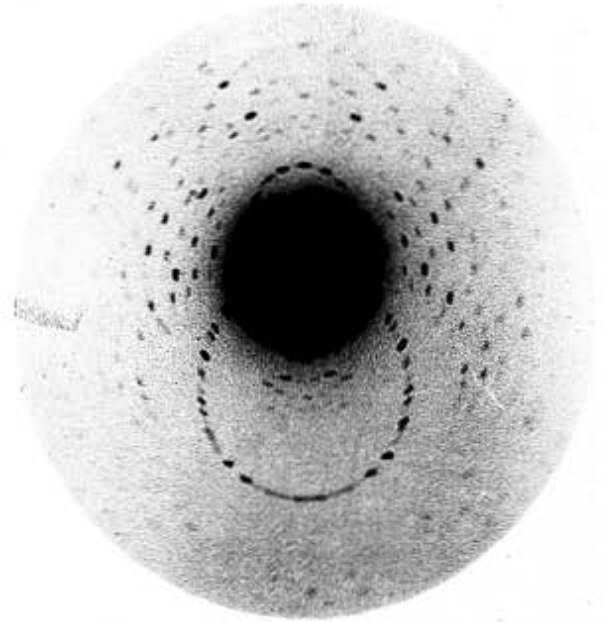


Fig. 2.
Turmaline. Plate parallel to $(01\bar{1}0)$.

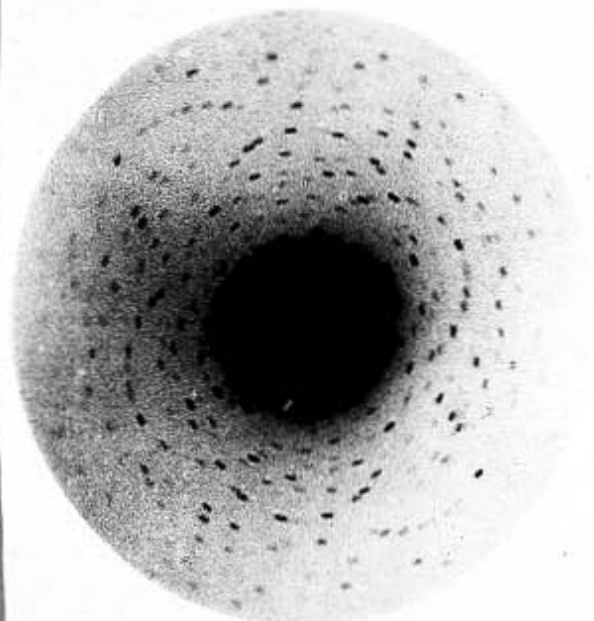


Fig. 3.
Turmaline. Plate parallel to $(\bar{1}2\bar{1}0)$.

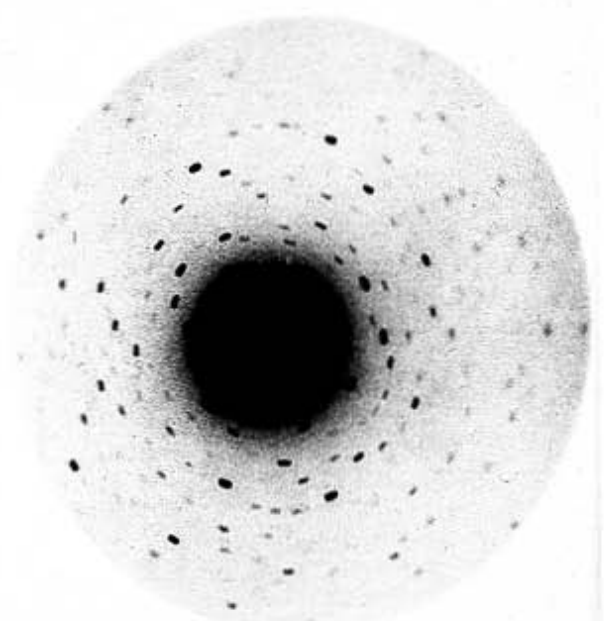


Fig. 4.
Phenakite. Plate parallel to $(01\bar{1}0)$.

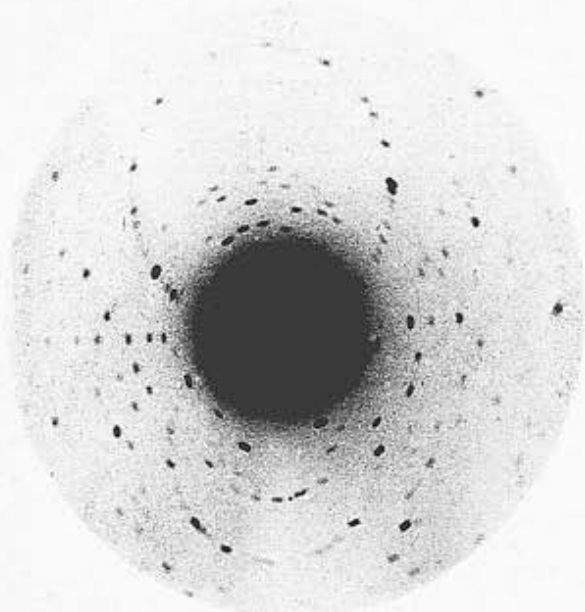


Fig. 5.
Phenakite. Plate parallel to $(\bar{1}2\bar{1}0)$.

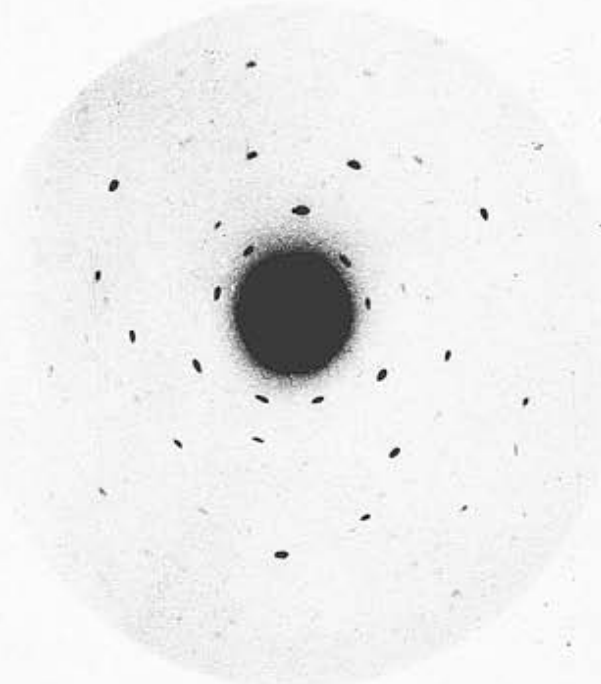


Fig. 6.
Dolomite. Plate parallel to (0001) .

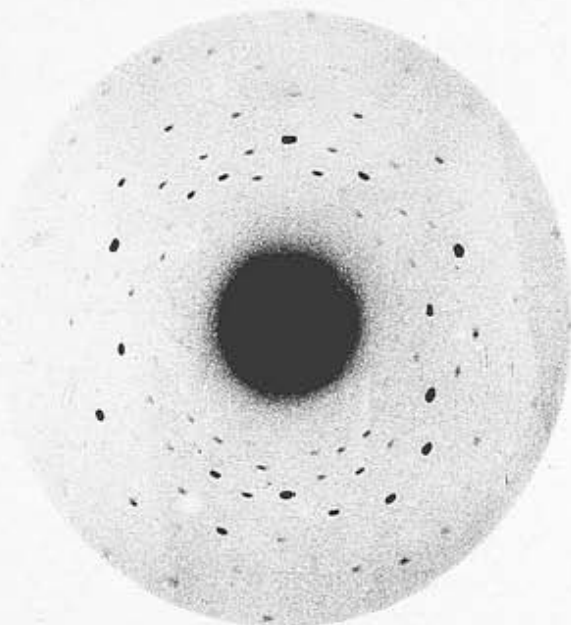


Fig. 7.
Dolomite. Plate parallel to $(01\bar{1}0)$.

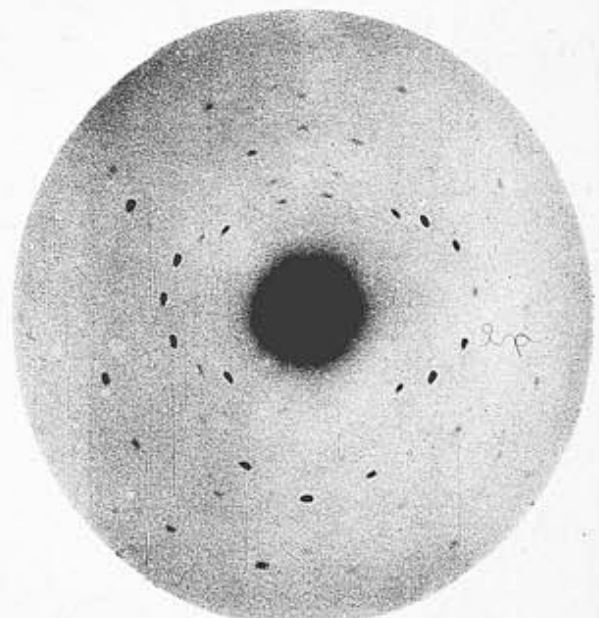


Fig. 8.
Dolomite. Plate parallel to $(\bar{1}2\bar{1}0)$.

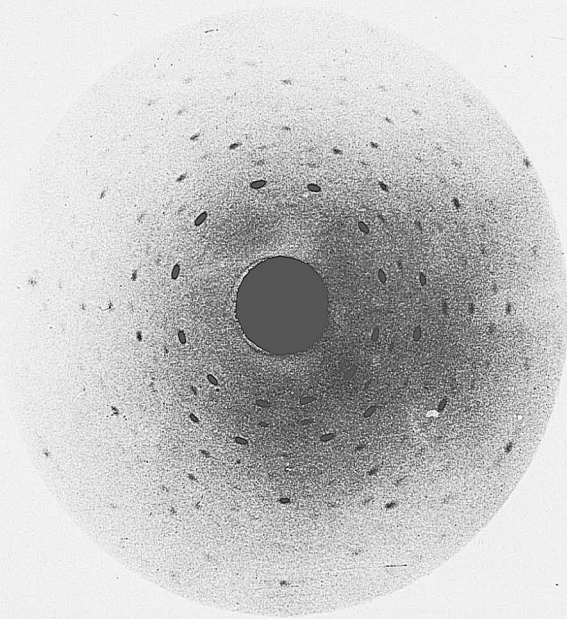


Fig. 9.
Beryl. Plate parallel to (0001).

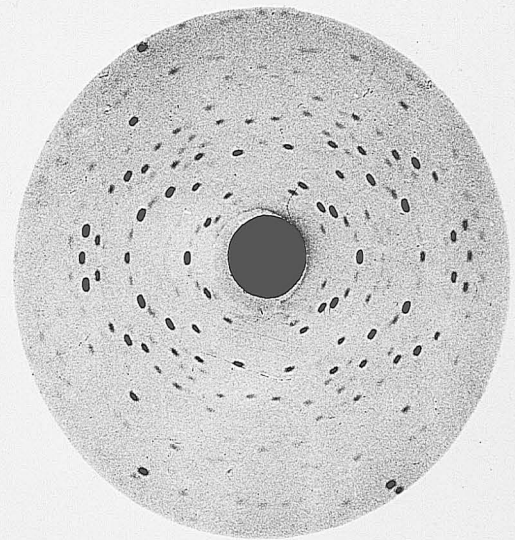


Fig. 10.
Beryl. Plate parallel to (10 $\bar{1}$ 0).

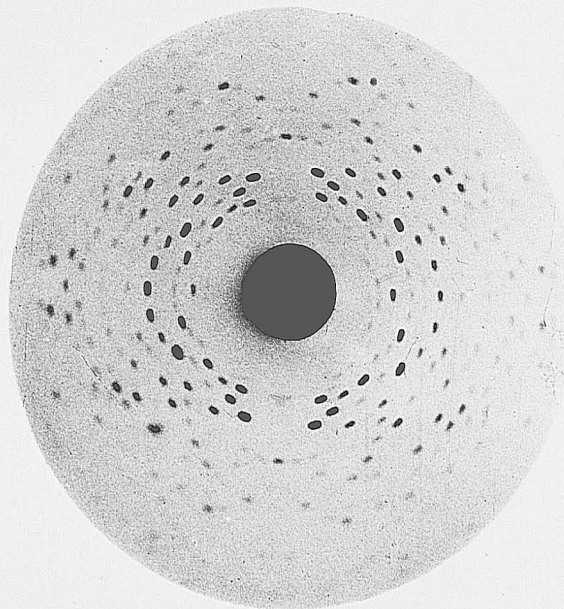


Fig. 11.
Beryl. Plate parallel to (1 $\bar{2}$ $\bar{1}$ 0).

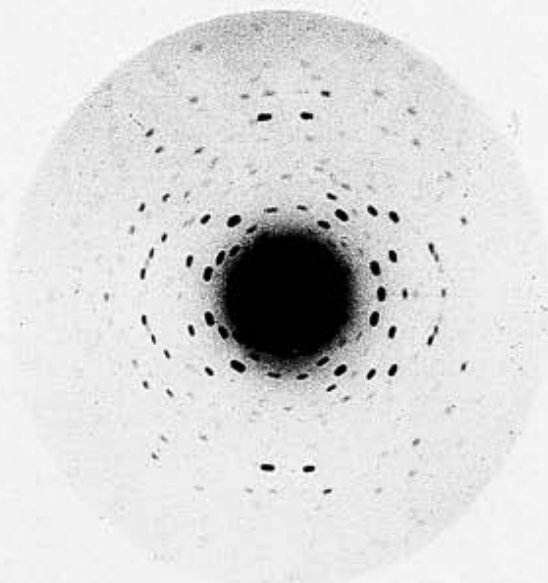


Fig. 12.
Apatite. Plate parallel to $(10\bar{1}0)$.

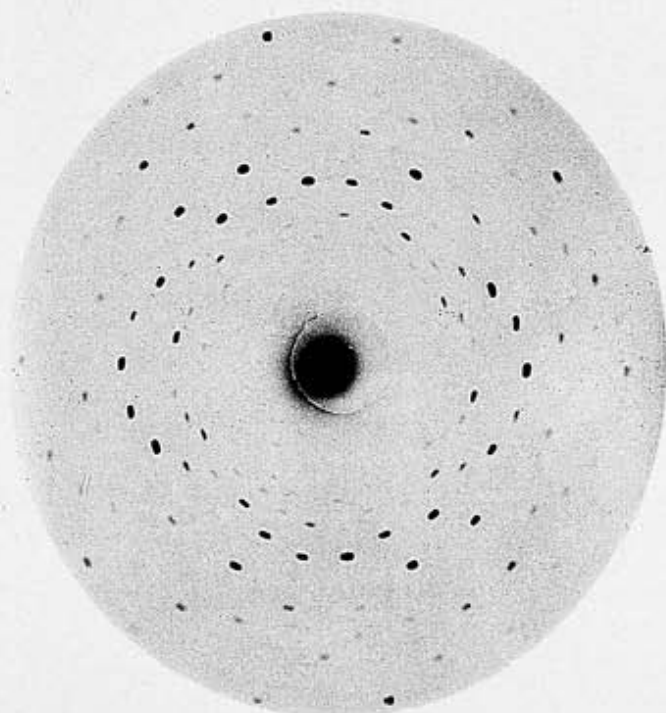


Fig. 13.
Quarz. Plate parallel to $(\bar{1}2\bar{1}0)$.
(Normal Pattern).

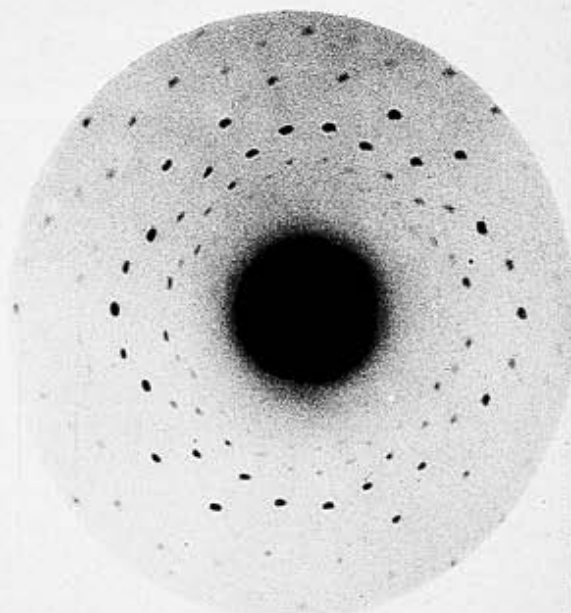


Fig. 14.
Quarz. Plate parallel to $(\bar{1}2\bar{1}0)$.
(Abnormal Pattern).

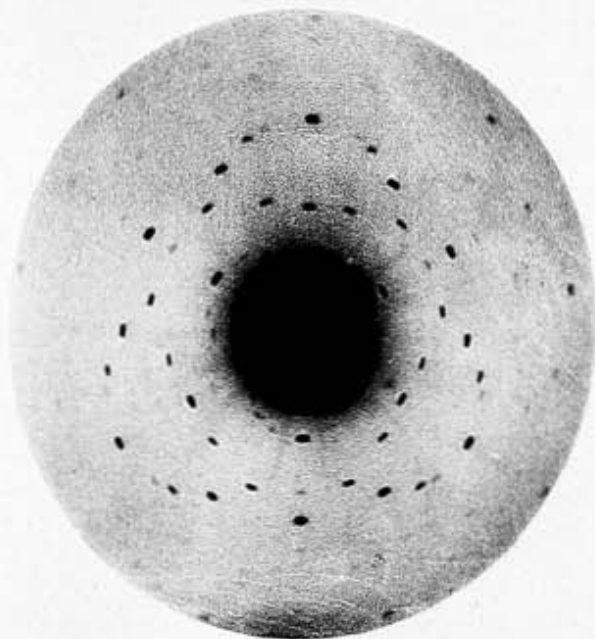


Fig. 15.
Pennine. Plate parallel to (0001).

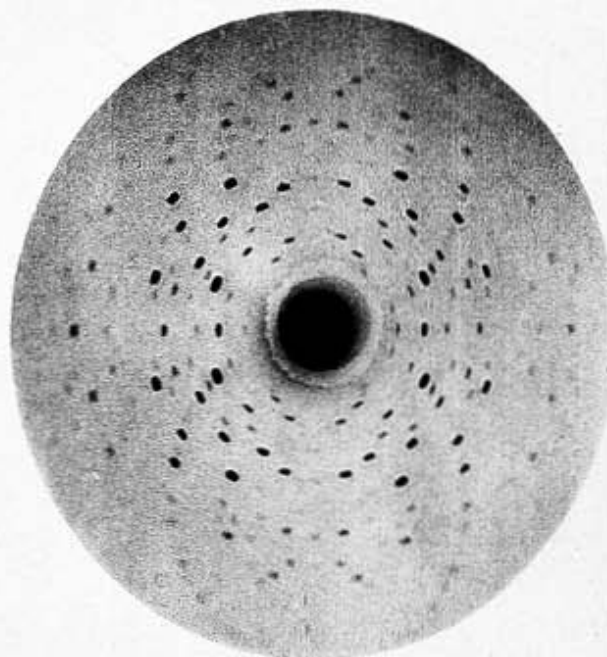


Fig. 16.
Hambergite. Plate parallel to (010).
(Normal Pattern).

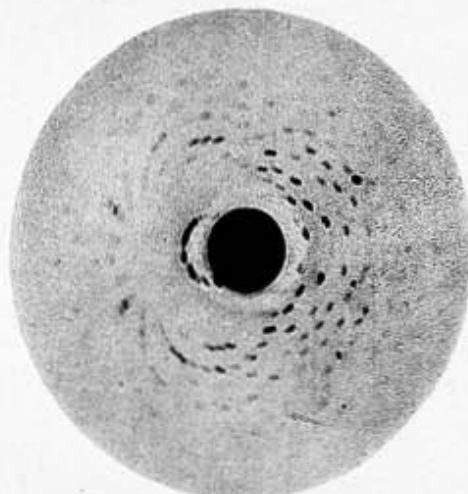


Fig. 17.
Zinc-Sulphate. Cleavage-lamella, exactly parallel to (010).
(The b -axis is 1st bissectrix).

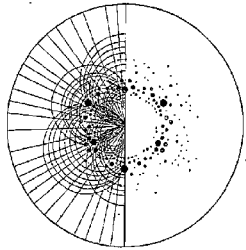


Fig. 1. Stereographical Projection of the Röntgenogram of Turmaline. Plate parallel to (0001) .

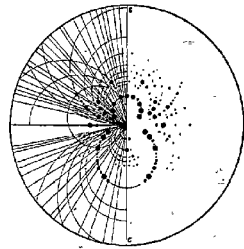


Fig. 2. Stereographical Projection of the Röntgenogram of Turmaline. Plate parallel to $(01T0)$.

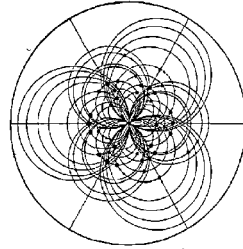


Fig. 3. Stereographical Projection of the Röntgenogram of Phenakite. Plate parallel to (0001) .

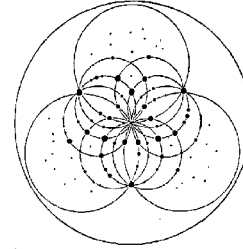


Fig. 4. Stereographical Projection of the Röntgenogram of Dolomite. Plate parallel to (0001) .

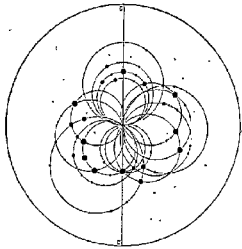


Fig. 5. Stereographical Projection of the Röntgenogram of Dolomite. Plate parallel to $(01T0)$.

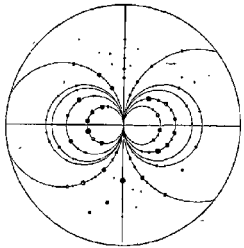


Fig. 6. Stereographical Projection of the Röntgenogram of Dolomite. Plate parallel to $(T2T0)$.

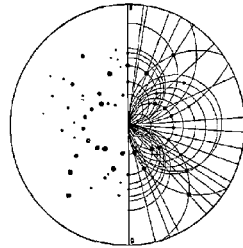


Fig. 7. Stereographical Projection of the Röntgenogram of Calcite. Plate parallel to $(10T0)$.

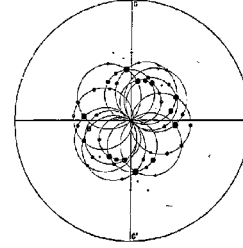


Fig. 8. Stereographical Projection of the Röntgenogram of Calcite. Plate parallel to $(T2T0)$.

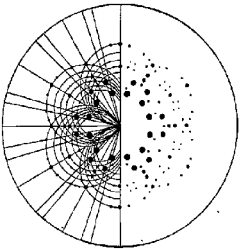


Fig. 9. Stereographical Projection of the Röntgenogram of Beryl. Plate parallel to (0001) .

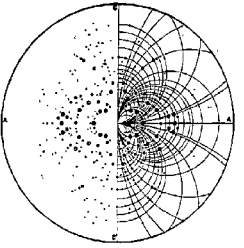


Fig. 10. Stereographical Projection of the Röntgenogram of Beryl. Plate parallel to $(10T0)$.

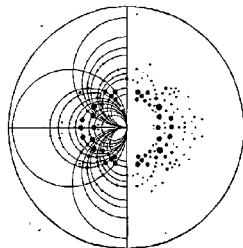


Fig. 11. Stereographical Projection of the Röntgenogram of Beryl. Plate parallel to $(T2T0)$.

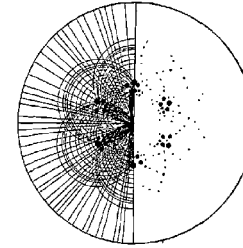


Fig. 12. Stereographical Projection of the Röntgenogram of Apatite. Plate parallel to (0001) .

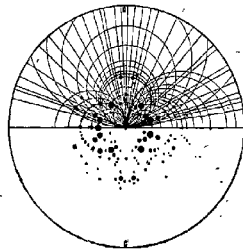


Fig. 13. Stereographical Projection of the Röntgenogram of Apatite. Plate parallel to $(10T0)$.

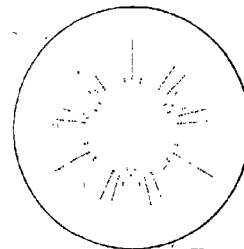


Fig. 14. Stereographical Projection of the Röntgenogram of dextrogyatory Quartz. Plate parallel to (0001) .

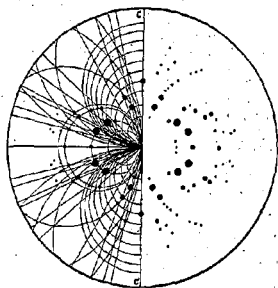


Fig. 15. Stereographical Projection of the Röntgenogram of dextrogyatory Quarz. Plate parallel to $(10T0)$.

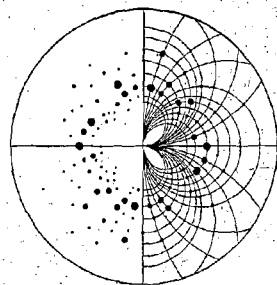


Fig. 16. Stereographical Projection of the Röntgenogram of dextrogyatory Quarz. Plate parallel to $(T2T0)$. (Normal Image).

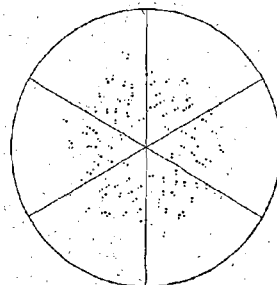


Fig. 17. Stereographical Projection of the Röntgenogram of Nepheline. Plate parallel to (0001) . (Schematic).

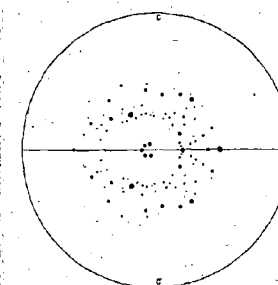


Fig. 18. Stereographical Projection of the Röntgenogram of Nepheline. Plate parallel to $(T2T0)$.

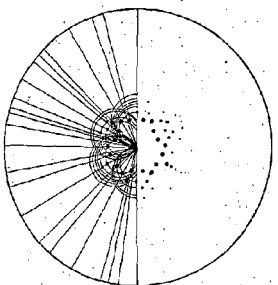


Fig. 19. Stereographical Projection of the Röntgenogram of dextrogyatory Cinnabar. Plate parallel to (0001) .

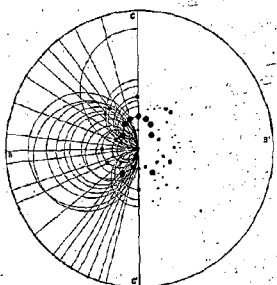


Fig. 21. Stereographical Projection of the Röntgenogram of dextrogyatory Sodium-Ammonium-Tartrate. Plate parallel to (100) . (Abnormal Image).

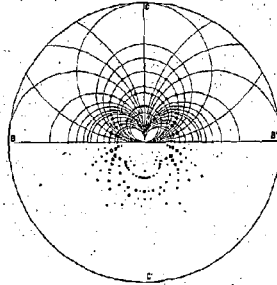


Fig. 21. Stereographical Projection of the Röntgenogram of dextrogyatory Sodium-Ammonium-Tartrate. Plate parallel to (100) . (Normal Image).

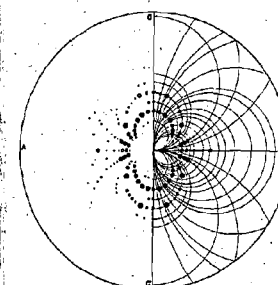


Fig. 22. Stereographical Projection of the Röntgenogram of dextrogyatory Sodium-Ammonium-Tartrate. Plate parallel to (010) . (Normal Image).

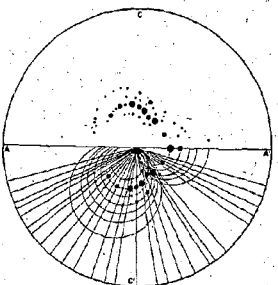


Fig. 23. Stereographical Projection of the Röntgenogram of dextrogyatory Sodium-Ammonium-Tartrate. Plate parallel to (010) . (Abnormal Image).

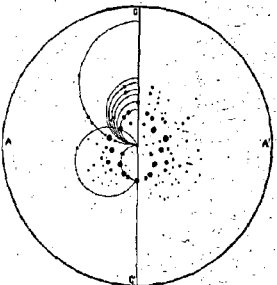


Fig. 24. Stereographical Projection of the Röntgenogram of dextrogyatory Sodium-Ammonium-Tartrate. Plate parallel to (010) . Abnormal Image, perpendicular to the first one, with the same position of the plate.

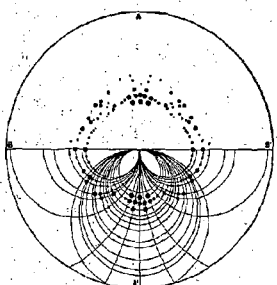


Fig. 25. Stereographical Projection of the Röntgenogram of dextrogyatory Sodium-Ammonium-Tartrate. Plate parallel to (001) .

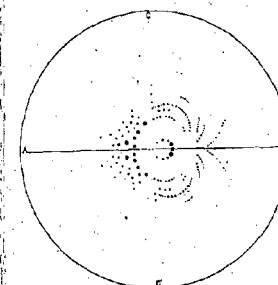


Fig. 26. Stereographical Projection (schematic) of the Röntgenogram of Zinc-sulphate. Plate parallel to (010) . Abnormal Image, obtained with a perfectly clear lamella, prepared by cleavage, and exactly perpendicular to the first bissectrix.