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Physics. — "*On a New Phenomenon Accompanying the Diffraction of Röntgenrays in Birefringent Crystals.*" By Prof. Dr. F. M. JAEGER. (Communicated by Prof. H. HAGA.)

(Communicated in the meeting of March 27, 1915).

§ **1.** A short time ago HAGA and JAEGER [1]) made some observations on the diffraction of RONTGEN-rays in crystals of *cordierite*, from very beautiful, perfectly transparent and homogeneous examples of which suitable plates were cut parallel to the three pinacoïdal faces {100}, {010} and {001}. On this occasion the RÖNTGENogram of the plate parallel to {001} of this mineral hitherto considered rhombic-bipyramidal, appeared in fact to possess two symmetry-planes perpendicular to each other, as well as a binary axis; the patterns however, obtained by the transmission of RONTGEN-rays through the plates parallel to {100} and {010}, appeared to possess only one single symmetry-plane. This combination of symmetry-elements is just the essential of rhombic-hemimorphic crystals.

It must be remarked however, that this fact is contrary to the consequences which follow from the theory of these phenomena, as far as it regards the expected symmetry of the RÖNTGEN-patterns.

The question, what will eventually be the symmetry of the RONTGENograms of crystals of a certain symmetry-class, can be answered comparatively easily. Deductions of this kind were made for the first time in 1913 by G. FRIEDEL [2]), who concluded, that under *no* circumstances such symmetry of crystals, as were characterized by the absence of a centre of symmetry, could be revealed in their RÖNTGEN-patterns.

The reasoning of FRIEDEL is principally as follows. He deduces the complex of symmetry-properties which is characteristic of hemihedrical and tetartohedrical crystals, from those belonging to the holohedrical forms, by the suppression of certain symmetry-elements in the latter groups, thereby making use of the wellknown fact, that in the holohedrical crystals every plane of symmetry corresponds to a binary axis perpendicular to it. This results from the fact, that all holohedrical crystals possess a centre of symmetry, and that such a centre, if combined with either a plane of symmetry or with an axis of pair period, necessarily will cause the presence of the other

[1]) H. HAGA and F. M. JAEGER, Proc. of the R. Acad. Amsterdam, **17**. 430. (1914).

[2]) G. FRIEDEL, Compt. rend. de l'Acad. des Sciences, Paris **157**, 1533, (1913).

of any of those three symmetry-elements; thus the combination of a symmetry-centre with a symmetry-plane having always the presence of a binary axis perpendicular to that plane as a consequence; and a centre combined with an axis of pair period always involving the existence of a symmetry-plane perpendicular to that axis. If now all hemihedrical and tetartohedrical crystals are considered as polyhedra, whose symmetry-groups correspond to complete secondary groups of the symmetry-complex of the holohedrical forms of the same system, then those secondary groups can be mathematically deduced from the primary groups, by suppression of definite symmetry-properties from the primary groups; from a mathematical standpoint nothing can be objected to such a way of reasoning; only it is necessary to keep always in mind, that from a *cristallogenetical* standpoint the hemi- and tetartohedrical crystalforms have of course nothing to do with the holohedrical ones.

Just because the centre, the plane of symmetry and the binary axis perpendicular to it, are always connected two and two in the way described before, it follows, that the deduction of the hemihedrical and tetartohedrical secondary groups from the holohedrical ones, can occur only by *simultaneous* suppression of *two* of them, in the symmetrygroups of the holohedrical forms. This suppression can be made in three ways:

a) So that one or more symmetry-planes + symmetry-centre are eliminated.

b) So that one symmetry-plane + a binary axis perpendicular to it are eliminated. –

c) So that a binary axis + the symmetry-centre disappear.

If now in a holohedrical crystal of any system, S_1 and S_2 are two secondary Röntgen-rays, which will be equivalent by symmetry to a certain plane V, they will also be symmetrically situated with respect to the binary axis perpendicular to the plane V; S_1 and S_2 will moreover always be centrically symmetrical to themselves, because every particle of the space-lattice, if reached by the ether-motion, will start as a centre of a secondary radiation in all directions.

If now in the holohedrical form of the system we imagine the centre of symmetry suppressed, then:

in the case of *a*) S_1 and S_2 will still remain symmetrically arranged to the binary axis, perpendicular to the simultaneously disappearing plane; and:

in the case of *c*), they will remain symmetrical with respect to

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the plane, perpendicular to the binary axis, which disappears at the same time as the symmetry-centre.

Thus in both these cases the symmetry of the RÖNTGEN-patterns will evidently remain *unchanged*; they will show the same symmetry as the RÖNTGEN-patterns of the holohedrical forms of the system would possess. From this results that all merohedrical crystals whose symmetry can be derived in the ways described sub *a*) and *c*), will give RÖNTGENograms of the same symmetry as in the case of the corresponding holohedrical crystals.

Only for the merohedrical crystals of the type mentioned sub *b*), the RÖNTGENogram will possibly manifest a different symmetry, than may be expected in the case of the holohedrical forms.

The sub *a*) mentioned symmetry-groups are characteristic of all crystals, which only possess *axial* symmetry; that is to say: for all crystals of those eleven classes, whose forms are different from their mirror-images, and which can exist therefore as *enantiomorphous* polyhedra. *Thus all dextro- and laevogyratory antipodes will necessarily manifest identical* RÖNTGEN-*patterns*.

Furthermore to the groups derived sub *a*) and *c*) will belong all those crystals whose symmetry is that of *hemimorphic* crystals; in the latter therefore the absence of the symmetry-centre will *not* be shown by the RÖNTGENograms in any other way than in the case of crystals of other symmetry-classes.

On more detailed consideration it appears that the cause, *why* the absence of a symmetry-centre in the crystals can never be revealed in the RÖNTGEN-patterns, is to be ascribed to the fact, that the generated secondary radiation is in itself of a centrically symmetrical nature, just as in the case of ordinary light-waves. If this were *not* the case, then the symmetry of the RÖNTGENograms could be discerned in the same 32 symmetry-classes, just as with the polyhedrical crystalforms themselves, which are generated under the influence of the one-sided forces of crystallisation. However it will appear that even such a supposition would not be sufficient to give an explanation of the new phenomena to be recorded here.

§ **2.** The problem we have had before us for a long time, and which evidently could only be answered by means of numerous experiments, was just this: what symmetry will eventually be revealed in the RÖNTGEN-patterns of all kinds of merohedrical crystals.

Originally it seemed as if the experience hitherto obtained fully supported the correctness of the above mentioned theoretical deductions.

In the RÖNTGENogram [1]) of the *sphalerite*: *ZnS*, which crystallizes in the hexacistetrahedrical class, *no* other symmetry could be stated than that which corresponds to the hexacis-octahedrical crystals.

On the other hand there is certainly another symmetry present in the case of *pyrite* [2]): FeS_2, which belongs to the dyacisdodecahedrical class, and which possesses thus a centre of symmetry.

The right-handed and left-handed rotating crystals of *sodiumchlorate*: $NaClO_3$ gave, on transmission of a pencil of RÖNTGENrays, *identical* patterns (Table I, fig. 1 and 2), which, if the plates were parallel to the faces of the cube, were of the same apparent symmetry, as the images of the pyrite, — just as EWALD and FRIEDRICH [3]) have also stated. It can be easily shown, that the same crystal (dextrogyratory), if radiated through perpendicularly to each of the cubefaces, always gave the same image, absolutely corresponding with that of a laevogyratory crystal, under the same conditions of experiment. Thus the absence of a symmetry-centre in this case could *not* be stated; both the symmetry-planes, perpendicular to each other, which in the case of the pyrite can be attributed to the crystals themselves, appear here in the RÖNTGENogram, because the symmetry-centrum of the radiation is superimposed on the symmetry of the chlorate-crystals, which symmetry is characterised by the presence of only three perpendicular binary axes and the four ternary ones [2]).

According to FRIEDEL, plates of dextro- and laevogyratory quartz, if cut parallel to {0001}, will give *identical* RÖNTGEN-patterns. Just in the same way, the crystals of dextro- and laevogyratory *luteo-triaethylenediamine-kobaltibromide* [4]): $\{Co(Aein)_3\}Br_3+2H_2O$, which crystallizes in the tetragonal system, gave identical RÖNTGEN-patterns, showing the presence of four vertical planes of symmetry.

1) LAUE, FRIEDRICH und KNIPPING, Bayr. Ak. der Wiss. München, 303, (1912).

2) It is remarkable, that the spots have not an *oval*, but a *rectangular* shape here; this fact cannot be explained in the way suggested by BRAGG, by the incomplete parallellism of the incident rays of the pencil. Such phenomena were observed likewise with some other crystals, so e g., with *sylvine* (*KCl*). perpendicular to the ternary axis. With *sylvine* also perpendicular to a quaternary axis the rectangular spots were observed; moreover the central spot here shows a radiation in eight directions, parallel to the faces of the rhombusdodecahedron, a phenomenon quite unexplicable at this moment. As to the rectangular shape of the spots, we are persuaded now that it is principally connected with the thickness of the crystalplates: the phenomenon manifests itself only in the case of thick plates, being more prominent, if the plate is thicker.

3) EWALD und FRIEDRICH, Ann. der Phys (4), **44**, 1183 (1914); vide also: FRIEDRICH, Deutsche Naturforscher und Ärztetag, Wien, (1913); BRAGG, Proc. R. Soc. London, **89** A, 477, (1914).

4) F. M. JAEGER, Verslagen Kon. Ak. Amst. April, (1915). (Still to be translated in these Proceedings).

On the other hand, in the case of the neutral *ethylsulfates of the rare earth-metals* [1]), which possess hexagonal-bipyramidal symmetry, the hemihedrical structure was found to be expressed quite clearly in the RÖNTGENogram, which is also in full accordance with the consequences of the theory, as this represents the case above mentioned sub *b*).

Further control of the exactness of these conclusions by experiment was finally only possible to me with the aid of the beautiful RÖNTGENograms obtained at the Physical Laboratory of this University by my friend and colleague H. HAGA with the crystallographical material especially selected by me for this purpose. Without his aid and kindness this investigation would have been quite impossible, and I therefore wish to express to him here my sincere thanks once more.

In fig. 1 the corresponding photo of the *apatite*: $Ca_5Cl(PO_4)_3$ is reproduced in stereographical projection [2]).

The plate used here was cut perpendicularly to the *c*-axis; it gave a very beautiful photographic image (Table I, fig. 3). The presence of a senary axis, but the absence of all vertical symmetry-planes is immediately recognisable here.

§ **3.** For the purpose of obtaining further data of this kind, plane-parallel plates were cut from crystals of *ferric-ammonia-alum*: $Fe_2(SO_4)_3 \cdot (NH_4)_2SO_4 \cdot 24H_2O$, and of *potassium-chromic-alum*: $K_2SO_4 \cdot Cr_2(SO_4)_3 \cdot 24H_2O$, — in both cases perpendicularly to a ternary axis.

In fig. 2 and 3 the patterns obtained are reproduced in stereographic projection. It is immediately evident that these images only show a ternary axis, but *no* planes of symmetry whatever. This is in full agreement with the theory: for the *alums*, just like *pyrite*, are dyacisdodecahedrical, and from the theoretical considerations mentioned above it follows, that they may eventually manifest their hemihedrical character in their RÖNTGENograms. But because these crystals do not possess any other planes of symmetry than those which are parallel to the faces of the cube, the image perpendicular to the ternary axis will in fact manifest no other symmetry-properties than those which follow from the mere presence of the ternary axis itself.

[1]) F. M. JAEGER, these Proceed. **16**. 1095. (1914); Receuil des Trav. des Chim. des Pays Bas et de la Belgique, T. **33**, 343. (1914).

[2]) In all these projections, *d* signifies the thickness of the crystalplate, *A* the distance between the frontal face of the crystal plate and the sensitive film of the photographic plate.

Thus as far as experience goes, the phenomena observed in crystals of the *regular* system seem to be in all cases in full agreement with the postulations of the theory. In this connection it may here be definitely stated, that the conclusions made by HAGA and JAEGER[1]) some time ago, from their experiments with *boracite* above and below 400° C., are now seen to be completely justified. For boracite at room-temperature, — *if* it were really regular, — would be of hexacis-tetrahedrical symmetry; and thus its RÖNTGENogram would possess just the same symmetry as that of the sphalerite; i. e. that the image would be identical with the pattern of boracite, which was obtained *above* 400° C., — because this corresponds to the holohedrical symmetry of the regular system. But the image obtained by the authors at room-temperature, now only shows the presence of two perpendicular planes of symmetry and a binary axis: therefore it can only correspond either to a rhombic structure, or to a dyacisdodecahedrical, or to a tetrahedrical-pentagonedodecahedrical crystal. The last mentioned two symmetry-groups however must be excluded definitely because of the characteristic development of the boundary forms of the boracite; thus the symmetry of the RÖNTGEN-pattern at room-temperature can only correspond to a *rhombic* arrangement of the molecules, the optical behaviour (biaxial) of the composing lamellae being in full agreement with this supposition. The internal change of symmetry of the boracite, if heated above 400° C., seems therefore to be incontestably proved by the authors in this experiment.

§ **4.** If now we leave for the present out of consideration the cases of the composite pseudo-symmetrical (mimetic) crystals hitherto studied, it seems really, as if in *all* cases, where regular or uniaxial crystals were studied, the results of the experiments were in full agreement with the conclusions which necessarily follow from the now adopted theory of the said phenomena.

However, the case of the rhombic *cordierite* is in flagrant contradiction with it: for from the theory it follows immediately, that *crystals of all three classes of the rhombic system must give Rontgenograms whose symmetry corresponds to that of the holohedrical forms.*

Thus plates parallel to the three pinacoidal faces: {100}, {010} and {001}, must always give patterns which are symmetrical with respect to two perpendicular planes of symmetry; their intersection, i. e. the line perpendicular to the photographic plate, must therefore in all cases be a binary axis.

[1]) H. HAGA and F. M. JAEGER, These Proceed. **16.** 792. (1914).

If the *cordierite* therefore were really hemimorphic, even then its hemimorphy could under *no* circumstances be revealed in its RONTGENpatterns in the way formerly observed by us! Notwithstanding that however, nobody can doubt the fact, that the RÖNTGENograms of plates parallel to {100} and {010}, only manifest one single vertical symmetry-plane. There must be some unknown cause therefore, why the other planes of symmetry in the images have vanished.

As long as this case was the only one known, it was allowable to regard it as quite accidental.

The following experiments however carry the conviction, that the theoretical views demonstrated in the beginning of this paper, are quite insufficient to explain the phenomena, as soon as they are studied in biaxial crystals, instead of in optically isotropous or uniaxial crystals, radiated through parallel to their optical axis.

The nature of these deviations may be seen from the facts described further-on; it will however be adviceable first to say something more in connection with the RÖNTGENograms of some uniaxial crystals, from which plates cut perpendicularly to the optical axis were studied.

Turmaline is ditrigonal-pyramidal; according to the above mentioned theory the RONTGEN-pattern must show the same symmetry as *calcite*, which is of ditrigonal-scalenohedrical symmetry. Just in the same way the pattern of the strongly dextrogyratory *cinnabar*: *HgS*, which crystallizes in the trigonal-trapezohedrical class, should manifest the same symmetry.

Indeed, it can be seen from fig. 4 and 5, — which represent in stereographical projection the RONTGEN-patterns, obtained with plates perpendicular to the optical axis, — that these images do not only possess a ternary principal axis, but moreover three vertical planes of symmetry; and they thus really show the same symmetry in their RONTGENograms, as the calcite.

From this it follows immediately, that e.g. the images for dextro- and laevogyratory cinnabar, if radiated through perpendicularly to the basal face {0001}, must be quite *identical.*

In Plate I fig. 4 the original photograph for *turmaline* is reproduced. Whether crystalplates of these minerals, when parallel to $\{\bar{1}2\bar{1}0\}$ will now really show the presence of a binary axis in their RONTGENograms as follows from the theory, we have yet to find out by more numerous experiments[1]). But in any case we can say,

[1]) Really we have found now this conclusion confirmed by experiment, as will be published in a short time.

that if these crystals are radiated through in a direction, in which they are *optically-isotropous*, hitherto nothing could be found which indicated a divergence between the theory and he experiments.

§ **5**. Quite different however are the phenomena observed in the cases of biaxial crystals, i.e. of such crystals, which are birefringent in *all* directions, and in which therefore the anisotropy of the ether will manifest itself in *all* directions.

In order to study the simpler cases first, we started with crystals of *rhombic* symmetry. Plates were cut from them going exactly parallel to the three pinacoidal faces· {100}, {010} and {001}. It might be expected therefore, that every image would appear symmetrical with respect to a pair of perpendicular symmetry-planes, while the normal on the photographic plate would be a binary axis.

The experiments were first of all made with plates of a beautiful, perfectly transparent crystal of dextrogyratory *sodium-ammonium-tartrate*: $Na(NH_4)C_4H_4O_6 + 4H_2O$. The crystallographic measurements were in perfect agreement with those of RAMMELSBERG: the salt is rhombic-bisphenoidical, and thus possesses as symmetry-elements only three perpendicular binary axes, but neither a plane nor a centre of symmetry.

In fig. 6, 7, and 8 the stereographical projections of the obtained RÖNTGENograms are given; on Plate II, in fig. 5 and 6, the original photographs of plates parallel to {010} and {100} are further reproduced; they were, as in all following cases, obtained with the use of a screen "Eresco" behind the photographic plate.

Although the time of exposure was fully three hours, the impressions on the photographic plate in the case of a plate parallel to {001} were extremely feeble; this fact could perhaps be partially caused by the rather great distance of the spots from the centre of the plate. The characteristic symmetry towards both perpendicular planes (vid. projection fig. 8) is however immediately recognisable.

The same fact, that the transmission of the RONTGEN-rays is so much less effective in one direction of the crystal than that in the others, will be found in other cases also, e.g. in that of the *hambergite* which will be described afterwards.

It is immediately evident, that these results are in total disharmony with the postulations of the theory.

The stereographical projection (fig. 7) of the image obtained by transmission of the RONTGEN-rays in the direction of the *b*-axis (plate parallel to {010}), only possesses one single plane of symmetry: there is neither a binary axis nor a symmetry-centre present. The plane of symmetry i.e. is parallel to {001}. The plane of the optical axes of the crystal being parallel to {100}, while the *c*-axis coincides with the first bisectrix of negative character, it is evident, that the homologous spots are missing in the photograph which lie in the directions parallel to the *a*-axis, i.e. parallel to the direction of the *smaller* optical elasticity of the crystal plate.

The figure corresponding to the image of a plate parallel to {100} (vid. fig. 6), also possesses only one single plane of symmetry; but it is now just the plane {001}, which has disappeared as such, while {010} remains. Here we therefore miss the spots which would correspond to directions parallel to the *c*-axis; thus on the photograph the spots have vanished, completely or partially, which would lie in the direction of the *greater* optical-elasticity of the crystal-plate.

On the other hand the image of a plate perpendicular to the *c*-axis (fig. 8) shows two perpendicular planes of symmetry, as well as a binary axis; the intensity of the spots is however very feeble indeed.

It must here be remarked, that the combination of symmetry-properties observed in these three Röntgen-patterns is geometrically quite impossible for the crystals themselves. The case considered is therefore again more convincing than that of the cordierite. Hence the cause of the newly discovered phenomenon can *not* be sought in the special symmetry-character of the crystals; there must be still some unknown factor, which determines the phenomenon of the unexpected disappearance of the planes of symmetry.

§ **6.** As a second case of this kind we have reproduced here the Röntgenograms which were obtained in the same way with plates of *hambergite.*

The choice of this very rare mineral, got at *Helgaråen, Langesundford, Norway,* was made with a view to its chemical composition: $Be_2(OH)BO_3$; the compound being composed of the lighter elements in the periodical system, whose atomic weights are all smaller than 20. Hambergite is rhombic; its parameter-ratio is: $a:b:c = 0.7988:1:0.7267$. The crystals were glassy and splendidly homogeneous; they showed the forms {110}, {100}, {010} and {011}, and had a prismatic aspect. A perfect cleavage is present parallel to {010}, a good one parallel to {100}. The crystals are very strongly birefringent; the birefringency is about: 0.072. The plane of the optical axes is parallel to {010}, the first bisectrix, which has positive character, coincides with the vertical axis. The dispersion round the first bisectrix is only feeble, with $\rho < v$.

In fig. 9, 10 and 11 the stereographical projections of the Röntgen-patterns obtained are reproduced. When perpendicular to the c-axis, the photograph, remained very feeble even after $3\frac{1}{4}$ hours' exposure; in both the other principal directions however, even after much shorter exposure, the photographs were very sharp.

The plate perpendicular to the c-axis, in fact shows two normal planes of symmetry, as well as the binary axis: in the direction of the a-axis (parallel to the plane (010)), the density of the spots seems to be greatest, but this is only slight in comparison with that of the very numerous and intense spots on both the other patterns.

When the RONTGENogram is perpendicular to the a-axis (fig. 9), the expected symmetry is also unmistakably present; the circles containing most spots here lie in the direction of the c-axis (parallel to the plane (010)).

The photograph however, obtained with a plate perpendicular to the b-axis manifests only one single plane of symmetry, namely that parallel to {001}. It is evident, that this combination of symmetry-properties would be also crystallographically quite impossible, and the only suitable explanation in this case is, that the plane of symmetry (parallel to {100}) has *disappeared* for some reason. Fig. 10 proves, that the above mentioned symmetry-plane is really expressed in that photo; but if the RONTGEN-rays are transmitted parallel to the direction of the optical normal (i.e. perpendicularly to the directions of maximum and minimum optical elasticity) evidently the spots which would be expected in the direction of *maximum* elasticity either come out in the photo not at all or only partially.

In table II moreover two of the original photographs in figs. 7 and 8 are also reproduced; they show the said phenomenon very clearly.

In the case of cordierite only the two vertical planes of symmetry were found, for plates cut parallel to {100} and {010}. In cordierite {100} is the optical axial plane, and the c-axis is the first bisectrix, and of negative character. However cordierite is a but feebly birefringent mineral (about: 0.010) and the optical elasticity in the directions of a- and b-axis is only slightly different in it.

Both pinacoides {100} and {010} have thus in an optical sense about the same relation to the direction of the c-axis, and therefore we observe that the plane of symmetry parallel to {001} disappears on the corresponding photos. In both plates the spots which lie in directions parallel to the *greater* elasticity have thus disappeared completely, just as in the case of hambergite.

§ **7.** Finally we point out the peculiarities found in the study of *benitoite*. This very beautiful mineral, which is used as a gem, and whose chemical composition is: $BaTiSi_3O_9$, has some importance from the mineralogical standpoint, because it has been considered by mineralogists as the only representative of the trigonal-bipyramidal, or, — with greater probability, — of the ditrigonal-bipyramidal class.

The first opinion was expressed by ROGERS[1]), the last by PALACHE[2]), who as a proof of the correctness of his view emphasized the presence of a form $\{22\bar{4}1\}$ in many crystals. Later on however JEŽEK[3]) made it probable that the ternary axis was of a polar nature, and that *benitoite*-crystals were twins with respect to the basal face $\{0001\}$; by this author *benitoite* is considered to be a ditrigonal-pyramidal mineral.

Evidently the question about the real symmetry of this remarkable, enormously dichroitic mineral, is not yet settled; for that reason this object was chosen for the study of the diffraction phenomena of RÖNTGEN-rays.

We had at our disposition very beautiful pink crystals from *San Benito County* in *California*, where *benitoite* is accompanied by natrolithe and neptunite. They were flattened parallel to $\{0001\}$, and showed a combination of the forms: $\{0001\}$, $\{10\bar{1}1\}$, $\{01\bar{1}1\}$, $\{10\bar{1}0\}$ and $\{0\bar{1}10\}$. From a beautiful, homogeneous crystal three plates were prepared exactly parallel to $\{0001\}$, to $\{10\bar{1}0\}$ and to $\{\bar{1}2\bar{1}0\}$. The optical investigations of the plate perpendicular to the *c*-axis, very soon proved that the crystals show only *pseudo*-trigonal symmetry, and that they are in reality not uniaxial, but biaxial, with a very small axial angle, and with positive character of their first bisectrix which coincides with the direction of the *c*-axis. In no position was the plate completely dark when between crossed nicols. On rotating the microscope-table the interference-image often showed a deformation of the central part and distinct lemniscate-shaped inner rings, as well as the transformation of the dark cross into two branches of a black hyperbola; the plane of the optical axes is evidently perpendicular to $\{10\bar{1}0\}$, with the *c*-axis as the direction of the first bisectrix, which has a positive character. The birefringence of the mineral is strong; in basal sections local disturbances of the image are also observed, suggesting at once the *mimetic* character of the

[1]) ROGERS, Science, **28**, 676, (1908).

[2]) PALACHE, Amer. Journ of Science, **27**, 398, (1909).

[3]) JEŽEK, Bull. intern. de l'Acad. des Sciences de Bohême, Prague (1909). Vide also on benitoite: LOUDERBACK. Publ. of Calif. Univ. **5**, 9, 149, (1907); KRAUS, Science **27**, 696, 710, (1908).

benitoite; it appears to be composed of lamellae, which with respect to each other are turned through an angle of 120°, and seem to possess rhombic-hemimorphic or monoclinic symmetry.

The plates parallel to $\{10\bar{1}0\}$ and $\{\bar{1}2\bar{1}0\}$ also betrayed this lamellar structure in a more or less convincing way: the plate parallel to $\{\bar{1}2\bar{1}0\}$ showed this lamellar character very clearly, and was composed of two sets of nearly perpendicular crossing lamellae, which made about 53° with the *c*-axis, while an irregular partition in fields of different colour and dichroism could be observed in some cases besides.

The crystals are very strongly dichroitic: for vibrations parallel to the *c*-axis the crystals have a deep blue colour, for such perpendicular to the *c*-axis, they are almost colourless, with a very faint lilac hue.

The cleavage is very imperfect, and parallel to $\{10\bar{1}1\}$; from the goniometrical measurements it follows, that the pseudo-trigonal complex has an axial ratio of: $a : c = 1 : 0.7319$.

In figures 12, 13, 14, are reproduced the stereographical projections of the very fine RÖNTGENograms which were obtained in our experiments. Figures 9, 10, and 11 on plate III are reproductions of the original photographs.

The plate perpendicular to the *c*-axis (fig. 14) gave a RÖNTGEN-pattern, which possessed no more than one single plane of symmetry, parallel to $\{10\bar{1}0\}$ notwithstanding its undeniable trigonal design.

In agreement with this, the image in fig. 13, obtained with a plate perpendicular to $\{\bar{1}2\bar{1}0\}$, shows a vertical symmetry-plane. It may appear doubtful whether this image also possesses a horizontal plane of symmetry: a very slight but noticeable difference in the intensity of the spots at the ends of the vertical axis seems to be present.

The question is however, whether this would indicate a real, and in that case very feeble polarity of the *c*-axis, or if it should be considered as a photographic effect, caused perhaps by a slight deviation of the plate from its normal position. In fig. 12, obtained by transmission of the pencil of RÖNTGENrays in a direction perpendicular to $\{10\bar{1}0\}$, the polarity of the *c*-axis is however very much more easily recognisable, — not only in the differences in intensity, but also in the different arrangement of the spots.

However, whether one considers the *c*-axis a polar one or not, the combination of the symmetry-properties observed is here geometrically quite impossible also; for if the *c*-axis is of a polar nature, then fig. 14 must be symmetrical with respect to both perpendicular planes; and if the *c*-axis is not of that kind, fig. 14 should necessarily possess the same symmetry.

In every case therefore, one plane of symmetry must have disappeared in fig. 14; here also no other supposition is possible than that there must be some reason why the expected spots in directions parallel to the intersection (0001) : $(10\bar{1}0)$ are completely or partially suppressed. The real symmetry of the pseudotrigonal complex of lamellae can thus be regarded after this as a matter of secondary importance; for it is very well possible, that in fig. 12 also a second symmetry-plane, parallel to {001} has disappeared, and in that case the resulting combination of symmetry-properties would be geometrically impossible too, just as in all preceding cases.

§ **8.** *We here thus meet the extremely remarkable phenomenon, that in biaxial crystals, in striking contradiction to the experience hitherto gathered from optical isotropous or uniaxial crystals if studied perpendicular to their optical axis, certain symmetry-elements of the* RÖNTGEN*ograms which were to be expected according to the* LAUE-BRAGG-*theory absolutely vanish.* Thereby a complex of symmetry-properties is revealed in the complete set of RÖNTGENpatterns of the same crystal, which is geometrically impossible, and which therefore cannot be a representation of the special symmetry of the crystal itself.

As far as experience now goes, and provided that the more complicated case of the mimetic *benitoite* is for the present left out of consideration, the suppression of the spots occurred in two of the cases studied, in those images which are obtained by the transmission of the RÖNTGEN-rays parallel to the optical normal; i.e. the spots disappeared there in the plane in which the differences of the optical elasticity of the crystal are *as great as possible.* In the case of the *sodiumammoniumtartrate* the suppression occurred for crystal-plates either parallel to the optical axial plane, or perpendicular to the second bisectrix; i.e. in the directions of the *greatest* and *smallest* elasticity, not however in the direction of the optical normal.

One would be inclined to explain these phenomena, —just because they are observable exclusively in those crystals whose optical anisotropy is manifested in *all* directions, — by supposing some condition of polarisation of the generated secondary waves, which polarisation would finally find its expression, — somewhat in the same way as in the case of ordinary light-waves, — in an unequivalence of perpendicular directions. Or one again would be inclined to suppose an anisotropy in the mode of motion of the particles

A NEW PHENOMENON, ACCOMPANYING THE DIFFRACTION OF RÖNTGEN-RAYS IN BIREFRINGENT CRYSTALS.

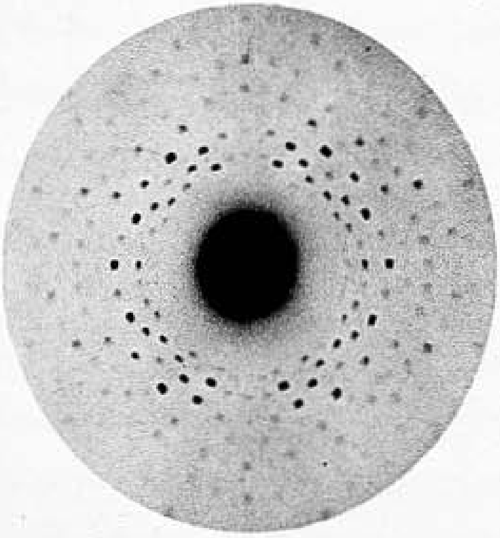

Fig. *1*.
Dextrogyratory Sodiumchlorate. Plate parallel to (100).
d = 2.95 m.M.; A = 45.8 m.M.

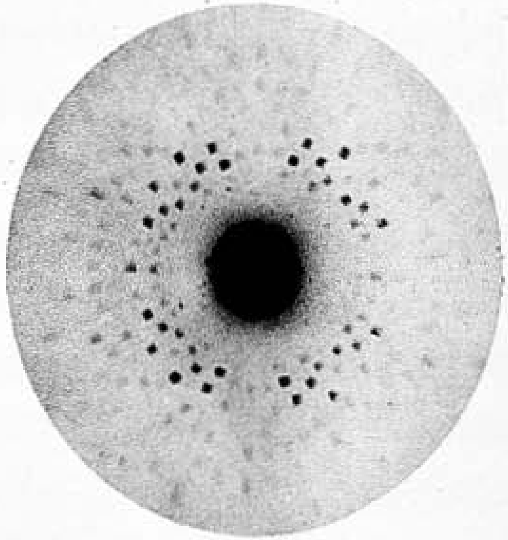

Fig. 2.
Laevogyratory Sodiumchlorate. Plate parellel to (100).
d = 2.25 mM.; A = 45.7 m.M.

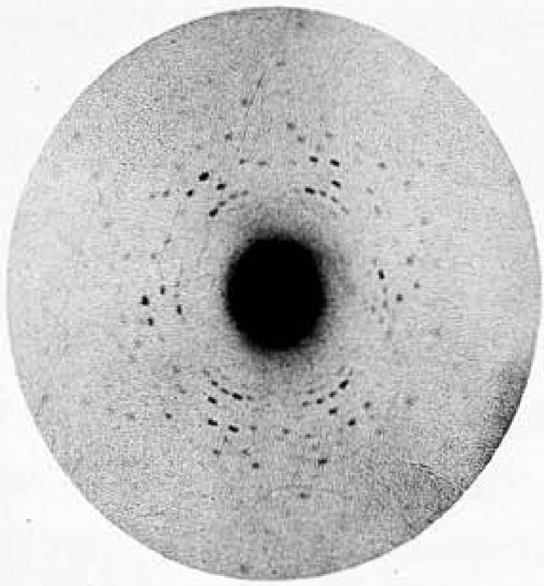

Fig. *3*.
Apatite; plate perpendicular to the c-axis.
d = 1.5 m.M.; A = 43 m.M.

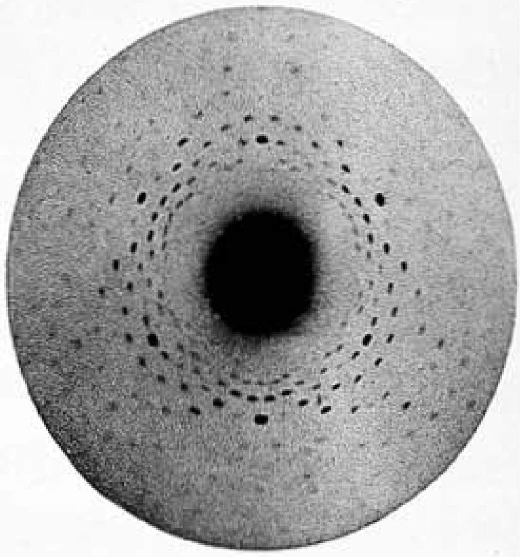

Fig. *4*.
Turmaline; plate perpendicular to the c-axis.
d = 1.1 m.M.; A = 44 m.M.

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A NEW PHENOMENON, ACCOMPANYING THE DIFFRACTION OF RÖNTGEN-RAYS IN BIREFRINGENT CRYSTALS.

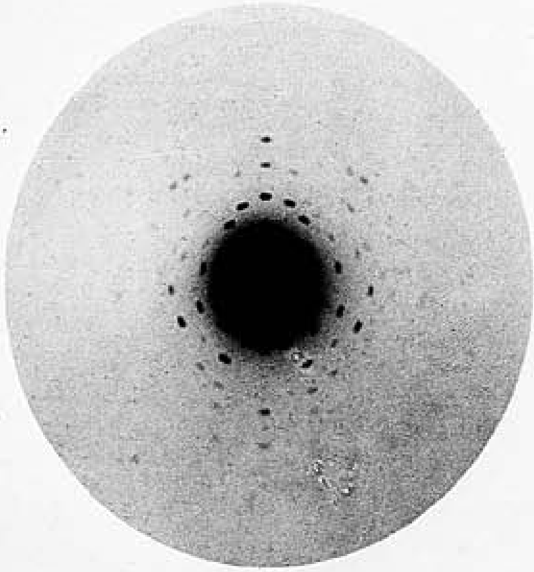

Fig. 5.
Dextrogyratory Sodium-Ammonium Tartrate ($4\,H_2O$).
Plate perpendicular to the *a*-axis.

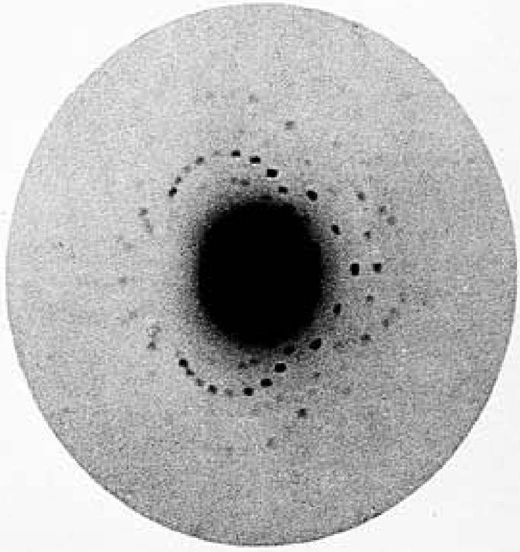

Fig. 6.
Dextrogyratory Sodium-Ammonium Tartrate ($4\,H_2O$).
Plate perpendicular to the *b*-axis.

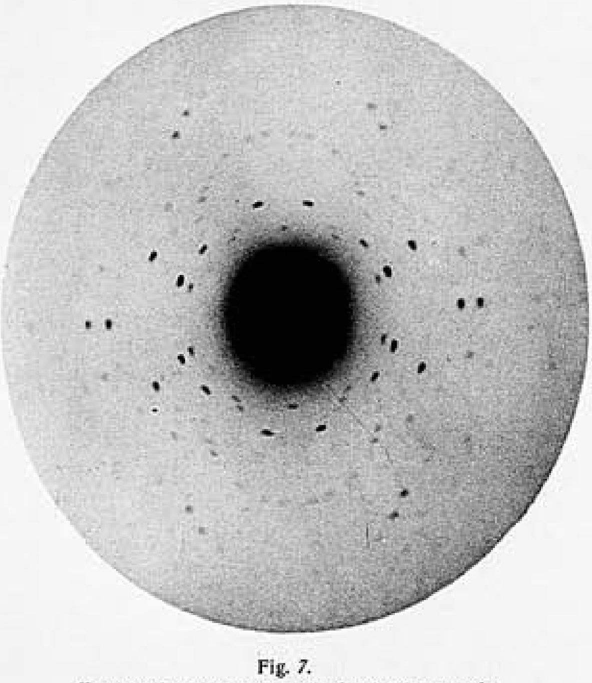

Fig. 7.
Hambergite; plate perpendicular to the *a*-axis.
$d = 1.56$ m.M.; $A = 50$ m.M.

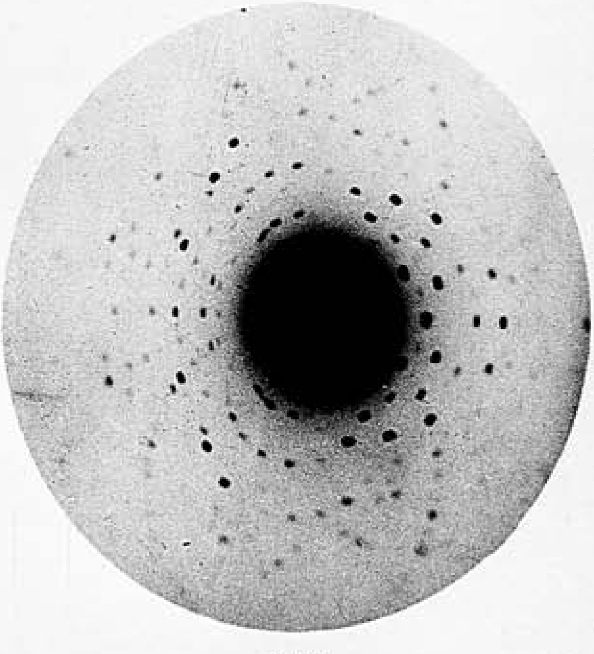

Fig. 8.
Hambergite; plate perpendicular to the *b*-axis.
$d = 1.54$ m.M.; $A = 50$ m.M.

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A NEW PHENOMENON, ACCOMPANYING THE DIFFRACTION OF RÖNTGEN-RAYS IN BIREFRINGENT CRYSTALS.

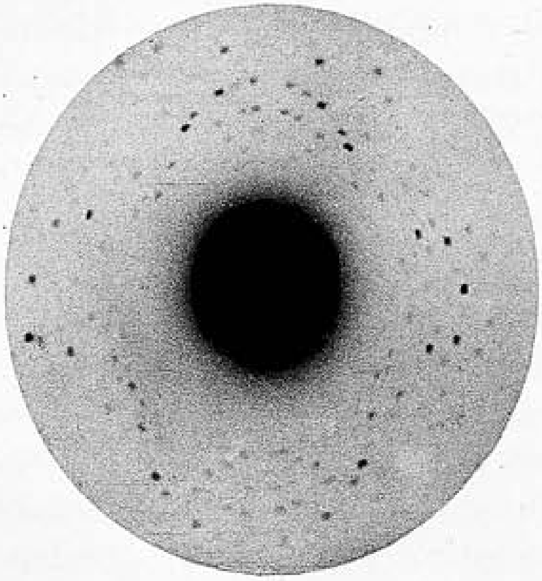

Fig. 9.
Hambergite; plate perpendicular to the *c*-axis.
$d = 1.52$ m.M.; $A = 50$ m.M.

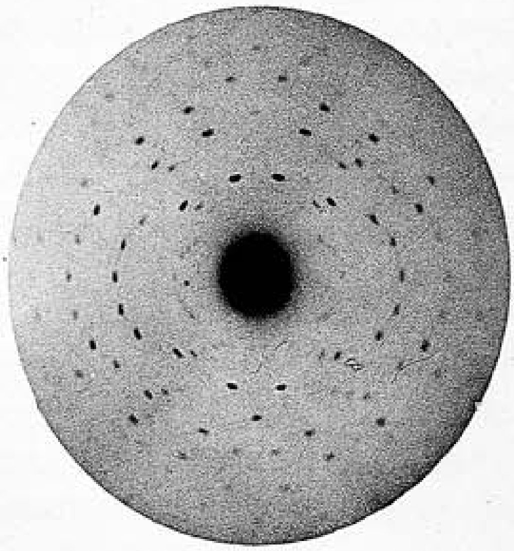

Fig. 10.
Benitoite; plate parallel to $\{10\bar{1}0\}$
$d = 1.52$ m.M.; $A = 50$ m.M.

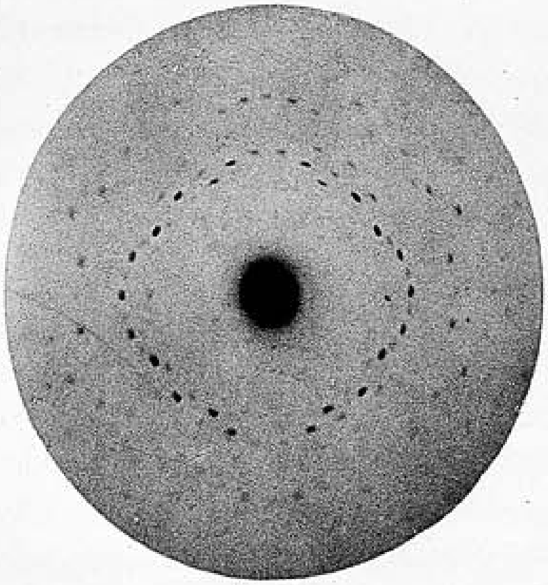

Fig. 11.
Benitoite; plate parallel to $\{\bar{1}2\bar{1}0\}$
$d = 1.50$ m.M.; $A = 50$ m.M.

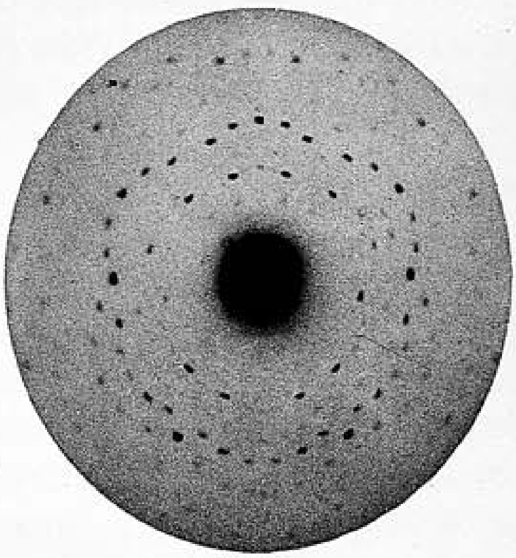

Fig. 12.
Benitoite; plate parelle to $\{0001\}$
$d = 1.50$ m.M.; $A = 50$ m.M.

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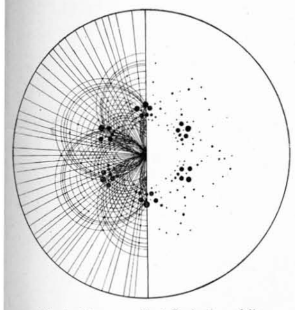

Fig. 1. Stereographical Projection of the Röntgen-pattern of *Apatite*. Crystalplate perpendicular to the *c*-axis.

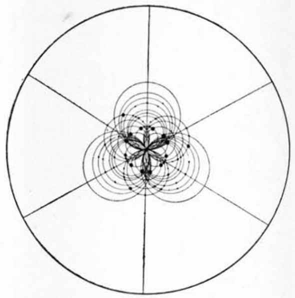

Fig. 2. Stereographical Projection of the Röntgen-pattern of *Ferric-Ammonium-Alum*. Crystalplate parallel to (111).

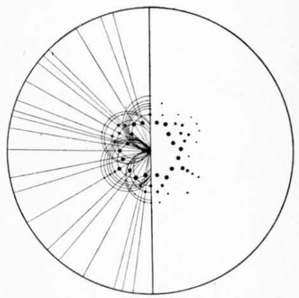

Fig. 5. Stereographical Projection of the Röntgen-pattern of dextrogyratory *Cinnabar*. Plate parallel to (0001).

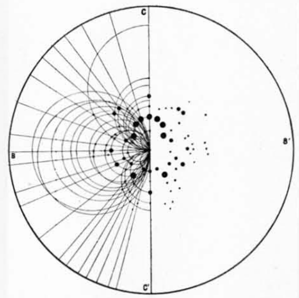

Fig. 6. Stereographical Projection of th Röntgen-pattern of dextrogyratory *Sodium-Ammonium-Tartrate*. Plate parallel to (100).

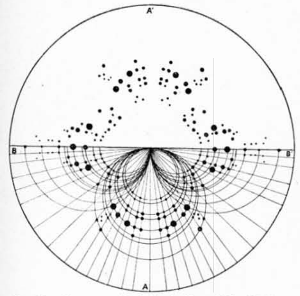

Fig. 10. Stereographical Projection of the Röntgen-pattern of *Hambergite*. Plate parallel to (~~010~~). 001

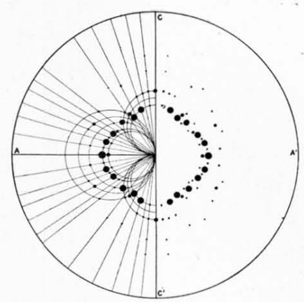

Fig. 13. Stereographical Projection of the Röntgen-pattern of *Benitoite*. Plate parallel to $(\bar{1}2\bar{1}0)$.

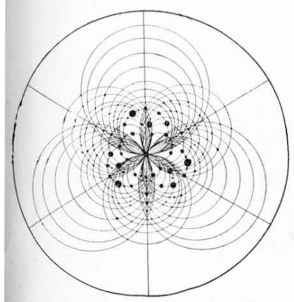

Fig. 3. Stereographical Projection of the Röntgen-pattern of *Potassium-Chromic-Alum*. Plate parallel to (111).

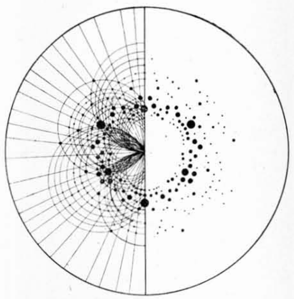

Fig. 4. Stereographical Projection of the Röntgen-pattern of *Turmaline*. Plate perpendicular to the *c*-axis.

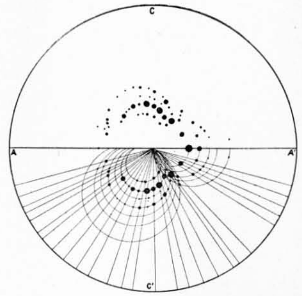

Fig. 7. Stereographical Projection of the Röntgen-pattern of dextrogyratory *Sodium-Ammonium-Tartrate*. Plate parallel to (010).

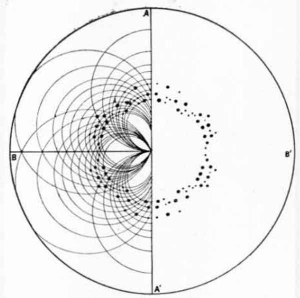

Fig. 8. Stereographical Projection of the Röntgen-pattern of dextrogyratory *Sodium-Ammonium-Tartrate*. Plate parallel to (001).

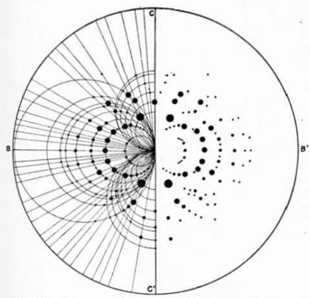

Fig. 12. Stereographical Projection of the Röntgen-pattern of *Benitoite*. Plate parallel to $(10\bar{1}0)$.

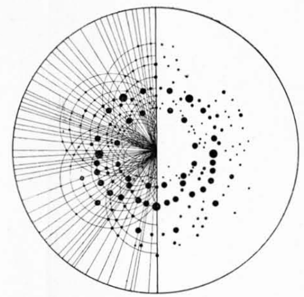

Fig. 14. Stereographical Projection of the Röntgen-pattern of *Benitoite*. Plate perpendicular to the *c*-axis.

affected by the impulse of the incident rays, in three perpendicular directions, and to investigate the consequences of such a supposition for the process of the generating of the spots on the photographic plate [1]).

In connection with this last supposition, the question could then be considered once more, whether the unequal deviations of the law of FRANTZ and WIEDEMANN in the principal directions of crystallised bismuth and hematite formerly observed, [2]) were not perhaps to be explained in some analogous way?

But let be it as it may, a final explanation of the phenomona observed here cannot be given at this moment. In any case it has become quite evident, that in the temporarily adopted theory for the diffraction-phenomena of RONTGEN-rays in crystals, a certain factor is yet missing, which has the result that the consequences of the theory are in agreement with the experimental results only if it is applied to isotropous crystals or to those in which the transmission of the RÖNTGEN-rays takes place in a direction, in which the crystal behaves like an optically isotropous one.

Only in the last-mentioned cases do the facts appear as full illustrations of the theoretical deductions.

But as long as that theory is unable to explain *why* the facts observed with biaxial crystals do *not* coincide with those expected by it, the theory can hardly be said to give a final explanation of the diffraction-phenomena in crystals at all.

Systematical experiments with the purpose to elucidate these phenomena as well as possible, are momentaneously going on.

Groningen, *Laboratories for Physics and for Inorganic*
March 15, 1915. *Chemistry of the University.*

Chemistry. — *"Researches on* PASTEUR'S *Principle of the Connection between Molecular and Physical Dissymmetry."* I. By Prof. Dr. F. M. JAEGER. (Communicated by Prof. VAN ROMBURGH).

(Communicated in the meeting of March 27, 1915)

§ **1.** It is now matter of common knowledge among scientists how the classic investigations [3]) of L. PASTEUR, regarding the connection between the so-called "molecular dissymmetry" of organic compounds and their optical behaviour, and especially those investi-

[1]) The case of the *tartrate* has in so far some analogy with that of *cordierite*, that in this case also the optical elasticities in the directions of both the *a*- and *b*-axes, do not differ very appreciably, in comparison with that in the *c*-axis.

[2]) F. M. JAEGER, These Proc. **15**. 27, 89. (1907).

[3]) L PASTEUR, Récherches sur la Dissymmetrie Moléculaire; Leçons professées devant la Société Chimique de Paris, (1860).