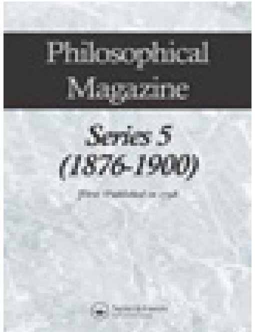

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XIX. On the change in the double refraction of quartz produced by electric forces

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XIX. *On the Change in the Double Refraction of Quartz produced by Electric Forces.* *By* Prof. W. C. Röntgen*.

It is well known that Sir W. Thomson has endeavoured to explain pyroelectric phenomena by assuming that the interior of pyroelectric crystals is constantly in a condition of electric polarization; the external action of this polarization is neutralized by a constant electric charge of the surface, so long as the polarization remains unaltered. Changes in the temperature of the crystals alter this condition; and the pyroelectric phenomena observed are the consequence of this change.

This view is supported by the phenomena recently observed by Messrs. J. and P. Curie†, confirmed by Hankel ‡, and distinguished as *piezoelectric*, as well as by the experiments made by J. and P. Curie § on the changes of form produced by electricity in pyroelectric crystals; at least these phenomena are naturally explained by the hypothesis in question.

I will not now dwell upon the difficulties which, in my opinion, militate against the acceptance of this hypothesis, but will simply explain how this view induced me to make the experiments here described, the results of which are certainly in themselves worthy of attention. The consideration from which I started was the following:—If an electric polarization were constantly present in a pyroelectric crystal in definite directions, and if it is allowable to conclude from the recently discovered effects of statical electricity on the optical properties of singly-refracting media that not only the polarization produced by external electric forces, but also any natural polarization already existing, would exert an influence on the vibrations of light transmitted through the crystal, then the optical properties of a pyroelectric crystal would be affected in different ways, according as the natural polarization was weakened or strengthened by the action of external electric forces.

Taking a quartz crystal as example, the result of piezoelectric experiments with it may be described as follows, at least for normal crystals of simple formation:—A section of the crystal at right angles to the principal axis may be divided by three straight lines intersecting each other in any point

* Translated from a separate impression from the *Berichte der Oberh. Ges. für. Natur- und Heilkünde*, communicated by the Author.

† *Compt. Rend.* xci. pp. 294, 383 (1880); xcii. pp. 186, 350; xciii. p. 204 (1881).

‡ *Abhandl. der kön. sächs. Gesellschaft.* vol. xii. p. 459 (1881).

§ *Compt. Rend.* xciii. p. 1137 (1881).

at angles of 60°, into six fields, which possess the following properties :—A pressure exerted upon the crystal in any direction through the point, or in any parallel direction, causes the crystal to become electrified at the two points of pressure, the one becoming positive, the other negative. If we change from one direction of pressure to another lying in an adjacent field, the sign of the electricity at the points of pressure changes whenever the direction of pressure crosses the boundary between adjacent fields.

Hence it follows that a pressure exerted in the direction of one of the three lines mentioned can produce no piezoelectricity ; on this account I propose to call these three directions the axes of no piezoelectricity. In the three directions bisecting the angles between these axes, there must be a maximum of piezoelectricity produced ; these directions may therefore be called the axes of maximum piezoelectricity. They coincide more or less exactly with the so-called secondary axes, the lines joining the two opposite edges of the quartz crystal. I am not yet able to decide whether or not they coincide exactly, since the experiments which I have made to test the point are not sufficiently numerous ; with some crystals, however, it seems to be really the case. If this were so, the axes of no piezoelectricity would have the same direction as the intermediate axes of the quartz.

Let us suppose that the three axes of maximum piezoelectricity give the three directions of the natural polarization : if we consider the ends of each axis as positive or negative, corresponding to the natural distribution of electricity in the interior, then these ends, if we follow them round in order, must be alternately positive and negative. The electricity produced by pressure is correspondingly positive or negative; and this holds good, as already remarked, for the whole field in which any axis lies.

If now a piece of quartz be so exposed to the inductive action of static electricity, that at any place the lines of force run at right angles to the principal axis, and at the same time do not run in the direction of an axis of no piezoelectricity, these forces will produce an increase or decrease of the natural polarization at this place, and with it, in accordance with the hypothesis stated at the outset, we shall have an increase or a decrease of the natural double refraction of rays which traverse the crystal at right angles to the principal axis and to the lines of force. The occurrence of the one case or of the other will depend wholly upon which of the three pairs of opposite fields the direction of the lines of force lies in, and in what direction they traverse it. There would be no change of the natural double refraction to be observed under the conditions assumed, if the lines of force run in the direction of one of the three axes of no piezoelectricity.

These conclusions, that the double refraction of the quartz may be increased or diminished at pleasure by the action of statical electricity, and that under certain defined conditions the double refraction is incapable of any such change, have been found to be confirmed by experiment.

The preliminary experiments were made with two rectangular parallelepipeds of pure Brazilian quartz, in which optical experiment showed no deformation. The pieces of quartz, which were obtained from Messrs. Steeg and Reuter, were 2·0 centim. long, and were made exactly of a uniform width and breadth of 1·2 centim. According to my instructions, the longest axis of the parallelepipeds should have coincided with a secondary axis; but, owing to a misunderstanding on the part of the workman, little weight was attached to this condition. Subsequent inquiries, as well as determinations made by means of Leydolt's* etched figures, showed that in both pieces this direction deviated but little from that of a secondary axis. It may further be remarked that it is sufficient for the investigations in hand that the direction of length should not coincide with an axis of no piezoelectricity: the piezoelectric experiments have shown that this was the case. Two of the lateral surfaces are exactly at right angles to the principal axis; and the other two lateral surfaces are therefore strictly parallel to the principal axis and nearly parallel to a secondary axis.

Each piece had a perforation in the direction of its length of about 0·2 centim. width, starting from the centre of the end faces; the coaxial perforations do not quite reach each other in the middle of the crystal, but leave a thickness of 0·2 centim., which forms the portion of the crystal whose electro-optical properties are to be investigated.

Both crystals were examined; but in each experiment only one crystal was placed in the electric field; the other was used to compensate the natural double refraction of the first: for this purpose the pieces were cemented together with a little isinglass, so as to have their principal axes at right angles to each other. Light polarized in a plane making an angle of 45° with the principal axes traversed the crystal at right

* *Berichte der Wiener Akademie*, vol. xv. p. 59 (1855).

angles to the plane containing the principal axis and a secondary axis of one of the crystals, and consequently at right angles to two lateral surfaces.

When placed between crossed Nicols, the centre of the field of view (that is, the place between the perforations) was nearly uniformly dark (employing sodium-light, the intensity of which was quite sufficient for these experiments), not regarding certain small irregularities, probably resulting from pressure during boring. Brass wires of about 0·15 centim. thickness, with well-rounded ends, were placed in the two perforations of the crystal under examination, which were so connected with the electrodes of a Holtz machine as to admit of rapid reversal of the connexions. The difference of potential between the two electrodes could be varied at pleasure and continuously, whilst the machine was rotated with uniform velocity by means of a secondary connexion containing a variable air-resistance. This method, which I have employed for some time, consists in connecting one electrode with an insulated sharp point, the other with an insulated metal plate. The point and plate are opposed to each other; and their distance apart can be altered at pleasure; the further they are apart the greater is the resistance of air for the dark discharge, and the greater therefore is the difference of potential.

In order to avoid the undesirable passage of sparks between the wires within the crystal, which is liable to occur when the difference of potential becomes too great, the crystal was placed in a small flask filled with sulphide of carbon, and in the later experiments with benzol; the polarized light entered one side at right angles where the flask was perforated, and the opening closed by a piece of plate-glass, and passed out at the opposite side, which was perforated in a similar manner.

For the purpose of control I have also examined the pieces of quartz in air, and have observed in all essential points the same behaviour as when they were immersed in sulphide of carbon or in benzol.

The direction of the secondary axis, the axis of length of the crystal examined for electro-optical effect, was placed vertical; the direction of the lines of force in the middle of the crystal was therefore vertical; and the principal plane of the Nicol consequently made angles of 45° with these lines of force (the arrangement previously designated "Position I." of the Nicol*).

* Compare *Ber. d. Ob. Ges.* vol. xix. p. 1 (1880); Wied. *Annal.* vol. x. p. 77 (1880).

To distinguish the pieces of quartz from each other they may be called "Crystal I." and "Crystal II."; one end face of each is marked, and in what follows is called "the marked end."

The action which the electricity exerts upon the light passing through the crystal was compared with the action which was caused by the compression, in a vertical or horizontal direction, of a piece of glass inserted between the analyzer and crystal. If we have noted, for example, as follows,

"Below +, above −; the same action as vertical compression,"

it is to be understood that charging the ends of the secondary axis below with positive and above with negative electricity produced the same optical change in the centre of the field of view as a compression of the plate of glass in a vertical direction.

Experiment I. Crystal I., marked end at the bottom.

Below +, above −; same action as *vertical* compression.
Below −, above +; „ *horizontal* „

Experiment II. Crystal I., marked end at the top.

Below +, above −; same action as *horizontal* compression.
Below −, above +; „ *vertical* „

Experiment III. Crystal I., marked end *above.*

(*a*) The centre of the field of view was made somewhat darker by vertical compression of the glass plate, then, while the glass plate remained compressed, "Below +, above −" caused an increase of brightness. This action could be compensated by stronger compression in a vertical direction.

(*b*) By vertical compression of the glass plate the centre of the field of view was again rendered darker; "Below −, above +" again rendered the field of view brighter; but this brightening was not now compensated, but rather increased by stronger compression in the vertical direction.

Experiment IV. (after the crystals had been cemented together in the opposite way). Crystal II., marked end at the top.

Below +, above −; same action as *horizontal* compression.
Below −, above +; „ *vertical* „

Experiment V.—In the above experiments the quartz was surrounded by sulphide of carbon; in the following ones it was surrounded by air.

Crystal I., marked end at the *top*.

Below +, above −; same effect as *horizontal* compression.
Below −, above +; „ *vertical* „

Experiment VI. Crystal II., marked end at the *top*.

Below +, above −; same effect as *horizontal* compression.
Below −, above +; „ *vertical* „

The experiments were repeated at very different times and under varied conditions: thus in the later experiments there was often only one crystal in the flask filled with benzol, the other, which served as compensator, being surrounded by air; sometimes plates of mica or other pieces of quartz were chosen as compensator; but the same results described above were always obtained, and never any thing different.

It is well known that a compressed glass plate behaves optically as a negative crystal the principal axis of which coincides with the direction of compression. Since quartz is a positive crystal, the results obtained above may be expressed by saying that the double refraction of the pieces of quartz examined increased when the marked end of the secondary axis was charged with positive electricity and the unmarked end with negative, and that, on the contrary, the double refraction diminishes when the marked end of the axis is negative and the unmarked end positive.

It was now further examined how these ends behave in piezoelectric relationship. The experiment was made by covering the end faces of the crystal with tinfoil and compressing it in a screw-press between plates of ebonite in the direction of its length: the one strip of tinfoil was connected with a delicate Fechner's goldleaf electroscope, capable of indicating the charge of the insulated pole of a Daniell's element by a marked deflection, the other was connected with earth.

The experiment frequently repeated gave uniformly the same result, that the marked end of the secondary axis of both pieces of quartz became negatively electrified upon increase of pressure, and positively electrified upon decrease of pressure; the unmarked end became respectively positively or negatively electrified.

We may therefore express the result of the electro-optical investigation thus: The double refraction of quartz increases if positive electricity is communicated to that end of a secondary axis which, upon increase of pressure acting in the direction of this secondary axis, becomes negatively electrified, and if at the same time negative electrity is communicated to the other end. The double refraction diminishes, on the other hand, when the distribution of the communicated electricities is the opposite.

If we adopt the view that a piezoelectric crystal is in a condition of electric polarization, whose direction, in the particular case of quartz, seems to coincide with the direction of a secondary axis, and that the observed piezoelectricity is a consequence of the change of polarization produced by pressure, then simple reasoning shows that the end which becomes negative upon increase of pressure is that towards which the negative side of the electrically polarized molecules is turned. But we found before that the double refraction increases when positive electricity is communicated to this end and negative electricity to the other; the polarization must be strengthened by this electrification; and we consequently obtain the result, that the double refraction of quartz increases or decreases according as the natural polarization is increased or weakened by external electric forces.

Having thus found the first of the consequences mentioned at the beginning confirmed by experiment, I proceeded further to put the second to the experimental test. The experiments described had shown me that, as far as intensity was concerned, there was at any rate no great difference between the increase and the decrease of the double refraction produced by equal electric forces; thence I concluded that it must be possible to find a direction in the quartz possessing the property that electric forces acting in this direction would produce no perceptible change in the double refraction. From what has been said above, this direction was to be sought for in an axis of no piezoelectricity—consequently in, or at any rate in the neighbourhood of, an intermediate axis of the crystal. I obtained therefore from Messrs. Steeg and Reuter a square plate of quartz of 1·5 centim. in the side and 0·25 centim. thickness, which was cut accurately parallel to a side face, and consequently at right angles to an intermediate axis. The principal axis is parallel to a side of the square; a secondary axis is consequently parallel to one of the sides at right angles to the first named. The four narrow side faces are polished.

First of all I examined whether the intermediate axis of this crystal was really an axis of no piezoelectricity. The result obtained was that even great changes of pressure in the direction of the intermediate axis gave rise to no perceptible quantities of electricity at the points of pressure, and that this direction is consequently an axis of no piezoelectricity. It may be remarked that a pressure parallel to the principal axis gave the same result, that, on the contrary, a pressure parallel to the secondary axis produced considerable quantities of electricity.

Next the plate was perforated in the centre of a square end face so that a nearly hemispherical depression was formed there (depth 0·1 centim). A further examination for piezo-electricity gave the same result as before.

The plate so prepared was placed upon the brass plate employed in my former electro-optic experiments between two thin strips of glass cemented to it, and introduced into the little flask filled with benzol. The end of a well-rounded brass wire projected into the depression in the plate, and formed the upper electrode, the plate being the lower electrode. The rays of light traversed the crystal parallel to the secondary axis, and consequently at right angles to the principal axis and to a secondary axis. The Nicols were placed in position I.

In order to compensate the natural double refraction, I employed the same means as in the experiments with the quartz parallelepiped; a second square plate of quartz, which was cut at right angles to the principal axis and had nearly the same dimensions as the first, was placed between the analyzer and glass flask, upon a stand movable about three axes at right angles to each other, and so placed that its principal axis was at right angles to that of the first plate. The double refraction could not be exactly compensated; but it was very easy to decide whether any such change of double refraction as took place with the first plate occurred here, by observing one of the vertical dark bands which crossed the field of view when the compensating plate was turned a little about a vertical axis. Any change in the double refraction would have been perceived by a displacement of the band to the right or to the left.

The experiment was made by adjusting a band in the middle of the field of view, consequently exactly under the bulb-shaped depression, which was to see if any displacement occurred when the difference of potential between the electrodes was subjected to rapid change. I have never been able to observe any such displacement, however varied the conditions under which the experiment has been made. Hence it follows that a change in the double refraction is not produced in any perceptible degree by electric forces acting in the direction of the axis of no piezoelectricity*.

* It is obvious that I cannot assert that no trace of electro-optic effect could have been observed in the direction of an axis of no piezoelectricity if much greater differences of potential than those in my experiments,

The bands were next adjusted first to the left and then to the right of the depression, but remaining close to it; in these positions also no effect of electrification upon the double refraction was observed; neither the lower nor the upper end of the band altered its position.

The observation that also the upper end of the band was not displaced is of importance; for since the lines of force which issue from the hemispherical depression run there horizontally, and coincide with the direction of the principal axis to the left and to the right of the upper electrode, it follows that also in the direction of the principal axis of the quartz no perceptible change can be produced in the double refraction by means of electric forces.

The remark made already in a footnote of course holds here also. The piezoelectric investigation, as remarked, had shown that change of pressure in the direction of the principal axis did not produce any electricity at the points of pressure. This result seemed worthy of direct proof. For this purpose the plate cut at right angles to the principal axis was provided with a central hemispherical depression just like the plate cut parallel to the axis, and introduced into the flask in the place of the latter; the plates were simply interchanged. Having by this arrangement obtained an interference-band under the depression (that is, at the point where the lines of force run parallel to the principal axis), I was unable to produce any displacement of the band by increasing or decreasing the difference of potential between the electrodes; consequently in this plate also the double refraction was not perceptibly altered by the action of electric forces in the direction of the principal axis. This plate also, upon pressure parallel to the principal axis, gave no piezoelectricity at the points of pressure. If the band were situated at one side of the depression, but very near to it, I observed a phenomenon upon electrification which furnished a very welcome confirmation of the results obtained with the quartz parallelepiped. The lower end of the vertical band remained fixed; but the upper end inclined towards the right or towards the left; and the direction of the motion changed with the sign of the electricity upon the electrodes. Further, I observed that, when the sign of the electricity remained the same, the upper end of a band moved in a different direction according

and a more intense source of light had been employed. If such a change were to be observed, it would certainly be much smaller than that which takes place in the direction of an axis of maximum piezoelectricity. The above experiments would therefore not lose their significance.

as the band was situated to the right or to the left of the centre. Vertical or horizontal compression of the ntercalated glass plate produced a displacement of the whole band parallel to itself to the left or to the right.

The explanation of these phenomena is easy to find if we consider that as the experiments described show the direction on the right, so on the left; that is, the horizontal direction at right angles to the rays of light in the crystal employed does not exactly coincide with an axis of no piezoelectricity. The phenomena are thus easily deduced from the first-described experiments:—The lines of force in the upper part of the plate run nearly horizontally in the neighbourhood of the depression; some of them therefore coincide with directions in which the double refraction is capable of being altered: in the lower part of the plate, on the other hand, the lines of force are vertical; they consequently run in the direction of the principal axis, and therefore produce no change of double refraction. Consequently it is only the upper end of the band which is displaced, and not the lower. The observation that the direction of displacement changes when the electrification or the position of the band changes is in complete agreement with the observed fact that the increase of the double refraction of quartz is changed into a decrease if the direction of the lines of force is reversed. We have seen above that from the distribution of piezoelectricity we can predict with certainty, for a given direction of the lines of force, whether an increase or a decrease of the double refraction will take place; and the question consequently arises whether with the new crystal the established rule will be found confirmed or not.

The plate was examined for piezoelectricity. A pressure upon the square surface in the direction of the principal axis produced no distinctly perceptible quantity of electricity at the points of pressure. The four narrow side faces, however, behaved differently; they may be designated *a*, *b*, *c*, *d* in order. An increase of pressure in the direction parallel to *b* and *d* produced positive electricity at *a*, and negative electricity at *c*; a decrease of pressure, electricities of opposite sign. An increase of pressure in the direction parallel to *a* and *c* gave negative electricity at *b*, positive electricity at *d*; a decrease of pressure, the opposite. In both cases I obtained vigorous deflection of the electroscope*. The plate was again

* Between the two directions, parallel to *b* and *d* and parallel to *a* and *c*, there must be a field which in piezoelectric relation would behave oppositely to the two fields to which these directions belong. In fact, an increase of pressure in the direction of the diagonal of the square which joins the corner *a d* with the corner *b c*, gave negative electricity at *a d* and positive electricity at *b c*; a decrease of pressure, the opposite.

placed in the flask full of benzol, and the electro-optical experiments repeated. The optical action of the electricity was also compared with the action of a glass plate compressed in a horizontal or in a vertical direction. In what follows such a note as

"Below +, above −; upper end of band left; horizontal compression"

is to be understood thus:—When the lower electrode was positive and the upper one negative, the upper end of the band lying on the left of the centre inclined towards the side towards which the whole band was displaced by a compression of the glass plate in a horizontal direction.

Experiment I. The rays of light traversed the plate parallel to *a* and *c*; *a* on the left, *c* on the right.

Below +,	above −;	upper end	of band	left;	vertical	compression.
" −,	" +;	"	"	"	horizontal	"
" +,	" −;	"	"	right;	horizontal	"
" −,	" +;	"	"	"	vertical	"

Experiment II. Rays of light parallel to *a* and *c*; *a* right, *c* left.

Below +,	above −;	upper end	of band	left;	horizontal	compression.
" −,	" +;	"	"	"	vertical	"
" +,	" −;	"	"	right;	vertical	"
" −,	" +;	"	"	"	horizontal	"

Experiment III. Rays of light parallel to *b* and *d*; *b* right, *d* left.

Below +,	above −;	upper end	of band	left;	vertical	compression.
" −,	" +;	"	"	"	horizontal	"
" +,	" −;	"	"	right;	horizontal	"
" −,	" +;	"	"	"	vertical	"

Experiment IV. Rays of light parallel to *b* and *d*; *b* left, *d* right.

Below +,	above −;	upper end	of band	left;	horizontal	compression.
" −,	" +;	"	"	"	vertical	"
" +,	" −;	"	"	right;	vertical	"
" −,	" +;	"	"	"	horizontal	"

It is easy to convince one's self that these data are in all respects in complete accordance with the results obtained from the experiments previously described.

It is necessary to mention that the phenomena described above may be explained by means of two known facts. One of these facts has been recently demonstrated by MM. J. and P. Curie*, as follows:—If electricities of opposite sign be communicated to the ends of a secondary axis of a quartz crystal, the crystal contracts or expands in the direction of this axis, according as the signs of the electricities communicated to the ends of the axis are opposed to, or the same as the signs of the piezoelectricities produced at these ends by a pressure exerted in the direction of the axis. It seems to me very probable that this result, found in the first instance for the direction of a secondary axis, would be found to hold good also for every direction at right angles to the principal axis, and that consequently the direction of the intermediate axis or the axis of no piezoelectricity possesses the property that electrical forces acting in this direction produce no perceptible change in the form of the quartz. So far I have had no opportunity to test the accuracy of Curie's experiment, and to extend it in the direction indicated; but inasmuch as those experiments interest me very much because of the close relationship in which they stand to my former experiments on the so-called electrical expansion†, I shall undertake this investigation at the earliest possible opportunity.

The second fact, admitting of easy confirmation, is that a mechanical compression of quartz at right angles to the principal axis exerts the same qualitative effect upon the rays of light which traverse the crystal at right angles to the principal axis and to the direction of compression as a compression exerted in the same direction upon an interposed plate of glass.

We shall find without difficulty that the phenomena de scribed are even in detail completely in agreement with the properties of quartz just now described.

I hope shortly to give an account of the remarkable phenomena which I have observed when the rays of light traverse the crystal parallel to the principal axis.

Giessen, November 25, 1882.

* *Compt. Rend.* vol. xciii. p. 1137 (1881).
† *Ber. d. Oberh. Ges.* vol. xx. p. 1 (1881).