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COMMUNICATIONS

FROM THE

PHYSICAL LABORATORY

AT THE

UNIVERSITY OF LEIDEN

BY

PROF. DR. H. KAMERLINGH ONNES.

Director of the Laboratory.

N^o. 37-48.

APRIL 1897-APRIL 1899.

EDUARD IJDO — PRINTER — LEIDEN.

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Dr. E. VAN EVERDINGEN Jr. *On the increase of the resistance of bismuth in the magnetic field, in connection with the dissymmetry of the HALL-effect.*

In a communication to the Academy, presented in the Session of 18 April 1895, Dr. A. LEBRET <sup>1)</sup> has pointed out, that the dissymmetry of the HALL-effect in bismuth may be described mathematically by supposing the resistance of bismuth to increase unequally in the magnetic field for different directions. At the same time the possibility was pointed out of a connection between these directions and the principal crystallographic directions.

Also in my communications on this subject, presented in the Session of 30 May 1896 <sup>2)</sup>, this description is continually used, and the value of the difference in increase of resistance was calculated for some plates. Besides, the connection with the crystallographic directions was established almost beyond doubt. It was a matter of importance to decide whether the existence of this difference in resistance might be proved directly.

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<sup>1)</sup> Communications N<sup>o</sup>. 19, p. 23.

<sup>2)</sup> Communications N<sup>o</sup>. 26.

For this purpose the following experiments were made

1. After determining the axes of symmetry in a round plate, a square was cut out of it with sides parallel to those axes. From the remaining borders two oblong pieces of bismuth were obtained; to each of them by means of WOOD'S metal two „resistance-electrodes” were soldered on the same side-plane, which were connected through a resistance-box and one of the coils of a differential-galvanometer. Then the pieces were placed between the poles of the magnet so as wholly to cover each other, though they remained apart, and a current was sent through them traversing one after the other. In one of the resistance-boxes the resistance was left constant, say 100 Ohms, and in the other was determined for different magnetic fields the resistance necessary to annul the deflection observed at the differential-galvanometer on closing the primary current. In order to eliminate the error, caused by HALL-effect, the mean was taken of the values, obtained thus for both directions of the magnetic field.

Indicating by  $a$  the ratio of the resistances when not in the field, that same ratio was found to be

1,005  $a$  in a magnetic field of 5500 c. g. s.

1,022  $a$  „ „ „ „ „ 7800 c. g. s.

2. In order to decide, whether the difference thus found between the resistances in different directions might be discovered also in the plates themselves, which served for the determination of the dissymmetry, „resistance-electrodes” were pressed against them along the directions]of the axes, and the resistance was measured by means of the compensative-current. (See communi-

cation of 18 April 1895<sup>1)</sup>). Hence the primary current flows through the rheotan-wires (see that same communication) and the plate of bismuth in succession. The result was not satisfactory. The current in these plates flows by no means exclusively in the direction of the line joining the primary electrodes. Accordingly I had not expected to obtain in this manner the true ratio of the resistances. The unfavourable result however induced me to calculate the difference of potential between the primary electrodes, taking into account the difference in the increase of resistance in different directions, which calculation brought the result, that the difference of potential contains no term with the first power of the difference in resistance. So this method could afford no results and was abandoned accordingly.

3. One of the round plates used for other experiments was made out of a piece of bismuth, the crystalline structure of which looked homogeneous. From the remaining piece two little bars of bismuth were cut, with their greatest dimension parallel to the plane of the plate (which itself was parallel to a cleavage-plane of the crystalline piece of bismuth), in two perpendicular directions. These little bars were fastened in a frame of ebonite between two brass screws, which entirely covered the limiting planes whilst two thin resistance-electrodes were screwed on to one of the sides. The little bar was traversed by the primary current, and the resistance between the resistance-

<sup>1)</sup> Communications N<sup>o</sup>. 19, p. 5.

electrodes was compared as in § 2 with that of the rheotan-wires of the compensative-current. It was found that in a magnetic field of  $\pm 5700$  c. g. s., the resistance of N<sup>o</sup>. 1 had increased 5,4 perct., that of N<sup>o</sup>. 2 7,4 perct. The specific resistance of these bars was  $\pm 154000$  in c. g. s. units<sup>1)</sup>; 2 perct. of this is  $\pm 3100$  c. g. s. The dissymmetry of the HALL-effect ( $K_{11}-K_{22}$  of LEBRET) observed in one of the positions of the plate was  $\pm 2700$  c. g. s. The directions of the edges of the bars with respect to the piece out of which they had been cut did not coincide wholly with those in which the axes of symmetry of the plate had pointed with respect to the same piece; moreover in round plates  $K_{11}-K_{22}$  is proportional indeed but not wholly equal to the dissymmetry; the agreement between the above mentioned numbers is satisfactory, if the inevitable errors of observation are taken into account.

4. Also with this method of observation sometimes different values for the resistance before and after the reversing of the field were obtained. The inquiry into the cause of this phenomenon is related in the next communication. From the experiments mentioned in § 1 and § 3 we may however conclude, that a different increase of resistance in the magnetic field really exists for different directions in crystalline bismuth.

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<sup>1)</sup> In this determination a rather large error may occur, as the distance between the electrodes was only 6 m.M.

Dr. E. VAN EVERDINGEN Jr. *On the relation between the 'crystallographic directions and the resistance, the magnetic increase of resistance and the HALL-effect in bismuth.*

1. The researches published in the preceding communication induced me to put to myself the question, in what manner the coefficient of magnetic increase of resistance in a fixed plane is related to the position of this plane and the direction of the magnetic force with respect to the crystallographic axis of bismuth. During closer inquiry also the question rose, in what manner the HALL-coefficient is related to the same direction. The answer to these questions is given in § 3. Let me describe the course of experiments in close connection with the former communication.

2. The increase of resistance in the magnetic field being determined for the little bars, mentioned in the latter part of the former communication, the same experiments were repeated with a bar, cut from the same crystalline piece in a direction  $\perp$  the former two and  $\perp$  the principal cleavage-plane. It appeared, not only that this bar had a greater specific resistance, but also that it showed a much greater magnetic increase of resistance. Whereas for instance with N<sup>o</sup>. 2 a resistance was found of 154000 c. g. s. when not in the field, and a magnetic increase of resistance of 7,4

perct., here the corresponding numbers were 176000 and 12,2 perct. A difference in the resistance when not in the field was observed already in 1855 by MATTEUCCI<sup>1)</sup>, who mentions that the conductivity of bismuth in the direction  $\perp$  the (principal) cleavage-plane is related to the conductivity // that plane as 1:1,16.

Hence it seemed a matter of importance to try to obtain plates in which this third direction, the direction of greatest resistance in a zero magnetic field, should be parallel to the plane sides; for such plates a large dissymmetry of the HALL-effect was expected. The examination of these plates ought then to be completed by that of three little bars, cut in the principal directions. As none of the crystalline pieces at hand was large enough to obtain a suitable plate from,  $\pm 300$  G. of bismuth were melted, cast in a porcelain shell and cooled slowly in the manner, described in the communication of 30 May 1896, p. 54<sup>2)</sup> From this piece of bismuth were cut:

1<sup>o</sup>. A round plate with its sides vertical, i. e.  $\perp$  the horizontal surface of the congealed mass of bismuth (R 7).

2<sup>o</sup>. A round plate with its sides horizontal. (R 8).

3<sup>o</sup>. From the bismuth close to where the first plate had been cut two vertical and two horizontal bars. (1,2,I,II).

4<sup>o</sup>. A horizontal bar,  $\perp$  the last mentioned ones, taken from the bismuth close to where the second plate had been cut (3).

The examination of the plates gave results not answering the expectations. R 7 had its axes just in the

<sup>1)</sup> C R. T. XL p. 541, 914, 1855.

<sup>2)</sup> Communications N<sup>o</sup>. 26, p. 14.

directions of the bars, not so however R 8. The dissymmetry ( $K_{11}-K_{22}$  of LEBRET) was in neither particularly large, in R 7 for instance  $\pm 5200$  c. g. s. in a field of  $\pm 7700$  c. g. s. (For comparison we refer to the results of plate R 2 on p. 55 of the above mentioned communication<sup>1)</sup>, where we may calculate for a field of 8600 c. g. s. a dissymmetry ( $K_{11}-K_{22}$ ) of  $\pm 14000$  c. g. s.) It will appear hereafter (§ 8) that the anisotropy in resistance when not in the field has little to do with the magnetic increase of resistance. Moreover, the plates appeared to show a different HALL-effect; to this we shall revert in § 5.

3. Also the examination of the bars gave results that demanded closer inquiry. (See § 4 of the preceding communication). Remarkable was the very large difference occurring with some among them between the values, obtained before and after reversing the magnetic field (Magn. A., Magn. B.), perpendicular to one of the oblong side-planes, for instance with the little bar N<sup>o</sup>. II in a field of 7700:

|               | Magn. A | Magn. B | Mean  |
|---------------|---------|---------|-------|
| Increase in % | 41,2    | 23,6    | 32,4. |

It looked unacceptable, that such differences might be caused by HALL-currents, received in degrees differing for the two electrodes, though these touched the same side-plane, as these differences represented a considerable part of the total HALL-effect to be expected if the electrodes were placed on *different* sides of the bar.

4. In order to detect the character of this phenome-

<sup>1)</sup> Communications N<sup>o</sup>. 26, p. 16.

non and to ascertain that no errors were occasioned by the method of observation, it appeared desirable to take experiments:

*a.* With primary currents of various strength.

*b.* In four positions (1, 2, 3, 4), obtained by turning the bars about the line joining the primary electrodes, each time through an angle of 90°.

*c.* In four corresponding positions, but with front and back interchanged (viewed from the magnet-poles).

*d.* In the same positions, after reducing the dimensions of the cross-section to about one half of their original value.

*e.* With a plate deposited by electrolysis.

These experiments gave the following results:

*a.* The results are wholly independent of the strength and direction of the current.

*b.* With the little bar N<sup>o</sup>. II.

| Position. | Magn. A | Magn. B | Mean   | Difference |
|-----------|---------|---------|--------|------------|
| 1         | 41,2 %  | 23,6 %  | 32,4 % | + 17,6     |
| 2         | 12,2    | 32,1    | 22,2   | - 19,9     |
| 3         | 37,1    | 25,0    | 31,0   | + 12,1     |
| 4         | 11,5    | 39,8    | 25,7   | - 28,3     |

*c.* With the same little bar

|   |      |      |      |        |
|---|------|------|------|--------|
| 1 | 20,7 | 34,0 | 27,4 | - 13,3 |
| 2 | 31,8 | 16,6 | 24,2 | + 15,2 |
| 3 | 18,6 | 34,8 | 26,7 | - 16,2 |
| 4 | 41,7 | 10,8 | 26,2 | + 30,9 |

*d.* <sup>1)</sup> The same little bar.

|   |      |      |      |       |
|---|------|------|------|-------|
| 1 | 28,6 | 35,8 | 32,2 | - 7,2 |
|---|------|------|------|-------|

<sup>1)</sup> In the original communication by mistake a slightly different series was published.

|   |      |      |      |        |
|---|------|------|------|--------|
| 2 | 28,7 | 20,8 | 24,7 | + 7,9  |
| 3 | 27,6 | 31,0 | 29,3 | - 3,4  |
| 4 | 32,0 | 19,7 | 25,8 | + 12,3 |

*e.* For both directions of magnetic force (A and B) quite the same increase of resistance.

Here we observe:

1<sup>o</sup>. Agreement between the positions 1 and 3 or 2 and 4; difference between 1 and 2 or 3 and 4. In the positions 2 and 4 the mean is always smaller.

2<sup>o</sup>. Interchanging front and back reverses the sign of the difference. Remarkably high or low values occur now with opposite magnetisation.

3<sup>o</sup>. In *d* the differences have been reduced on an average to less than half their value.

5. A tolerably probable explanation of these particulars we may derive from the observation of the HALL-effect in the plates R 7 and R 8. For whereas R 7 showed in a field of 7700 c. g. s. a HALL-constant of 3,36 c. g. s., R 8 gave 6,39 c. g. s., that is to say almost twice as much. The differences between the various bars show clearly enough, that the bismuth had not crystallised perfectly regularly. If we suppose the crystals at one end of the bar to be placed otherwise than those at the other end, then also at one end a HALL-effect may occur of double strength, and the full HALL effect of plate R 7 may appear as an error in the determination of the resistance. After the bar turning 90° about its longest axis, the stronger HALL-effect appears there, where first the weaker effect was found, at least if the crystals in one cross-section are nearly

parallel: thence the reserve of sign in the difference. Front and back having been interchanged, at the electrode where first the strong HALL-effect was found, now the weak effect appears (the resistance electrodes touch the lower side); once more the sign of the difference is reversed. If the dimensions of the cross section are reduced to one half of their former value, the resistance is multiplied by *four*, the HALL-effect only by *two*, the relative error is divided by *two*.

This explanation was confirmed by the same regularities being generally observed also with the other bars.

In order to test it more closely, as for the moment indeed no certainty had yet been obtained that the HALL-effect depended solely on the position of a plane in the crystal with regard to the magnetic force, the HALL-effect of these little bars was determined directly in a frame of ebonite, composed expressly for this purpose, in the positions 1, 2, 3 and 4. (See § 3). Quite in accordance with the expectation, the result was that in many cases very different HALL-coefficients were obtained.

So we may put the differences to the account of the HALL-effect and henceforth take the mean of the values, obtained before and after reversing the magnetic field, and also of the positions 2 and 4 or 1 and 3, in order to obtain the value of the resistance. In the same manner we will proceed with regard to the experiments, made after the above mentioned ones with a set of three little bars, cut at right angles to each other out of one of the crystalline pieces, presented by the

„Königliches Blaufarbenwerk: Oberschlema” <sup>1)</sup>, which showed on experiment the same phenomena, but for the greater part much more regularly; and with the three little bars from the crystalline piece, mentioned in the preceding communication (obtained from MERCK), which were now subjected to a renewed and more extensive investigation, and showed still greater regularity.

6. In order to get a survey of the experiments we should direct our attention to what follows:

We know that bismuth crystallises in the hexagonal system, in rhomboids differing but little from cubes. The principal axis ends in the most acute solid angles. This principal axis coincides with FARADAY'S magne-crystal-axis. According to MATTEUCCI it is moreover the axis of greatest resistance; also my experiments lead to this result. The most important result of these experiments is, that the increase of resistance for the directions of a plane perpendicular to the direction of the magnetic field is smallest in bismuth, when the principal axis coincides with the lines of force, and that also the HALL-effect is much weaker, when the axis is placed in this position.

For the cast bismuth of course we know nothing about the position of the principal axis. With regard to this bismuth we will compare the direction of greatest resistance with the directions perpendicular to it. For the sake of simplicity we will enter only a mean value for the two other directions under „second direction”, and mention here only, that the differences between

<sup>1)</sup> See Communications N<sup>o</sup>. 26, p. 14.

these two are generally very small and never so large as those between one of them and the principal axis.

*Cast bismuth (§ 2).*

| Direction:                        | Principal axis<br>(⊥ plate R 7) | Second direction | N <sup>o</sup> . | Remarks                                                                                                                                                                                 |
|-----------------------------------|---------------------------------|------------------|------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Specific resistance               | 147000                          | 130000           | 3<br>1,2,I,II    |                                                                                                                                                                                         |
| Increase of resistance in percts. | 21,5                            | 27,9             | 1                | The figures in the first column represent not the increase of the resistance 147000, but the increase of the resistance 130000, when the princ. axis coincides with the lines of force. |
|                                   | 25,4                            | 28,5             | 2                |                                                                                                                                                                                         |
|                                   | 25,0                            | 29,6             | II               |                                                                                                                                                                                         |
|                                   | 32,6                            | 28,6             | I                |                                                                                                                                                                                         |
|                                   |                                 | 33,5             | 3                |                                                                                                                                                                                         |
| HALL-constant.                    | 1,77                            | 7,77             | 1                | The figures behind the accolades are mean values.                                                                                                                                       |
|                                   | 3,36                            | 8,73             | 2                |                                                                                                                                                                                         |
|                                   | 5,12                            | 4,78             | II               |                                                                                                                                                                                         |
|                                   | 5,22                            | 5,65             | I                |                                                                                                                                                                                         |
|                                   |                                 | 9,17             | 3                |                                                                                                                                                                                         |
|                                   | 3,36                            | 6,39             | R 7<br>R 8       |                                                                                                                                                                                         |

Though large differences remain, yet we clearly see that the direction adopted as „principal axis” possesses the enumerated qualities. The mean value of the HALL-constants for the four little bars (1, 2, I, II) in the

position, corresponding, with that of plate R 7 between the poles, 3,87, differs not much from the value obtained with R 7, 3,36; also the mean value 6,53 agrees with the value 6,39 of R 8.

The differences in the first column for increase of resistance should explain the dissymmetry observed in R 7. Combining them to two mean values, we find 23,5 (1,2) and 28,3 (I, II), hence difference = 4,8%.

The original resistance is for both directions 130000, so we calculate for the dissymmetry 6240, whilst we observed  $\pm 5200$ . Hence the difference of resistances found is more than sufficient for explaining the dissymmetry. Moreover it appeared on examination, that also the sign of the dissymmetry agreed with the observed differences.

*Crystalline piece from OBERSCHLEMA.*

| Direction:                       | Principal axis | Second direction | N <sup>o</sup> . | Remarks        |
|----------------------------------|----------------|------------------|------------------|----------------|
| Specific resistance              | 146000         |                  | 4                | 6 had a burst. |
|                                  |                | 122000           | 5                |                |
|                                  |                | 156000?          | 6                |                |
| Increase of resistance in percts | 17,5           |                  | 5                |                |
|                                  | 26,8           |                  | 6                |                |
|                                  |                | 30,5             | 4, 5, 6          |                |
| HALL-constant.                   | 0,96           |                  | 5                |                |
|                                  | 4,43           |                  | 6                |                |
|                                  |                | 7,59             | 4, 5, 6          |                |

Very regular were especially the results for increase of resistance obtained with 5, partly because this was the longest of the three bars. Both the values, obtained before and after reversing the field, and the mean values for positions differing by  $180^\circ$  were always equal within a fraction of a percent. The HALL-coefficients observed were 0,96 and 7,37 <sup>1)</sup>.

*Crystalline piece from MERCK.*

| Direction:                        | Principal axis<br>( $\perp$ plate R6) | Second direction | N <sup>o</sup> . | Remarks                                                               |
|-----------------------------------|---------------------------------------|------------------|------------------|-----------------------------------------------------------------------|
| Specific resistance               | 172000                                | 151500           | 3                |                                                                       |
|                                   |                                       |                  | 1,2              |                                                                       |
| Increase of resistance in percts. | 6,5<br>7,7                            | 16,4             | 1                |                                                                       |
|                                   |                                       |                  | 2                |                                                                       |
|                                   |                                       |                  | 1, 2, 3          |                                                                       |
| HALL-constant.                    | 1,28<br>1,47<br>$\pm 2,00$            | 7,30             | 1                |                                                                       |
|                                   |                                       |                  | 2                |                                                                       |
|                                   |                                       |                  | 1, 2, 3<br>R 6   | This value was deduced from an experiment in a weaker magnetic field. |

With these bars almost always the same resistance is found before and after reversing the field. The

<sup>1)</sup> With this bar the experiments have been repeated at very low temperatures; the results will be published afterwards.

greater resistance and smaller increase of resistance we may ascribe perhaps to a very slight impurity and rapid cooling.

7. Attempts have been made also to measure in these bars the dissymmetry of the HALL-effect. These however have not met with success, probably because, with these bars, it is not allowed to neglect the dimension in the direction of the magnetic force. Now in this direction the resistance is not or but little increased, and dissymmetry appears as soon as the HALL-electrodes are not placed one just over the other. In this manner we may perhaps explain why with the bars at hand dissymmetry was observed, though the above-given explanation supposes that no dissymmetry appears when one of the crystal-directions coincides with the magnetic force (in this case the increase of resistance depends indeed for all the directions of the plane of our plate on *one* vector).

In concluding we wish to point out, how the above-described phenomena may serve to explain a great many irregularities, appearing, in dissymmetry and mean HALL-effect, at the examination of the same plate in different positions <sup>1)</sup>. As indeed the plates are generally not homogeneously crystalline, both observed quantities will to some extent depend on the position of the crystals happening to be placed at the electrodes, and we ought to expect a priori a different HALL-effect as soon as the plate is clamped in a different position. Only the regular differences between positions of sym-

<sup>1)</sup> See Communications N<sup>o</sup>. 26, p. 7.

metry and dissymmetry will not yet be cleared up in this way.

8. In order to explain the above mentioned phenomena we should connect them with the magnetisation. We might suppose the state of matters to be as follows:

In a zero magnetic field the resistances in different directions in a crystal of bismuth may be represented by the radii-vectores in an ellipsoid of revolution the greater axis of which coincides with the principal axis of the crystal.

A magnetic force, directed along the principal axis, causes a stronger magnetisation than the same force acting in a direction perpendicular to this axis. If we construe (the directions of the axes being the same as before) two other ellipsoids of revolution, one with axes in the ratio of the square roots of the values for the magnetisation in the two cases mentioned, the other with axes in the ratio of those values themselves, then, for a given direction of magnetic force the direction of magnetisation is indicated by the radius vector towards the point where the tangent plane  $\perp$  the magnetic force touches the first ellipsoid; the relative value of the magnetisation is measured by the length of this radius vector to where it meets the second ellipsoid<sup>1</sup>).

The strength of the HALL-effect in a plane plate, made of a crystal of bismuth, depends on the component of magnetisation  $\perp$  the plane of the plate. When the

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<sup>1</sup>) In the original communication was erroneously stated, that both direction and magnitude of magnetisation might be found with the aid of *one* ellipsoid.

direction of magnetisation coincides with the principal axis the HALL-coefficient for the plane perpendicular to it will have a smaller value, than when the magnetisation is in a direction perpendicular to that axis. Very likely we shall, for an arbitrary position of the plane for which we wish to know the HALL-coefficient, find that coefficient with the aid of the ellipsoid of revolution construed with the extreme values.

The magnetic increase of resistance we know to be much smaller in the direction of the magnetic force than in the directions perpendicular to it. Hence we put forward the following *hypothesis*: in the magnetic field the resistance is increased only in all directions of a plane perpendicular to the *magnetisation*; in this plane all resistances are increased in equal proportion. The magnitude of the increase of resistance depends on the direction of magnetisation. We find the smallest increase when this direction coincides with the principal axis, and the greatest when it is perpendicular to the principal axis. Very likely for an arbitrary position the coefficient of increase of resistance will be found with the aid of an ellipsoid of revolution, construed with the extreme values.

After this increase of resistance the ellipsoid of resistances in general will possess three unequal axes. The resistances in directions at right angles in a plane section will generally be increased in different proportions; hence the possibility exists of dissymmetry of the HALL-effect.

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COMMUNICATIONS  
FROM THE  
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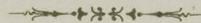
AT THE  
UNIVERSITY OF LEIDEN

BY  
PROF. DR. H. KAMERLINGH ONNES,

*Director of the Laboratory.*

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No. 38.

(REPRINT).



Dr. L. H. SIERTSEMA. On the effect of pressure on the natural rotation of the plane of polarisation in solutions of cane-sugar. (continued).

(Translated from: Verslagen van de Afdeeling Natuurkunde der Kon. Akad. van Wetenschappen te Amsterdam, 29 Mei 1897, p. 24—28)

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EDUARD IJDO — PRINTER — LEIDEN.

Dr. L. H. SIERTSEMA. *On the effect of pressure on the natural rotation of the plane of polarisation in solutions of cane-sugar (Continued).*

1. To the measurements, described in the former communication ¹⁾ new ones, with a greater concentration have been added. In making them the two quartz-plates, used on a former occasion have been employed together for the compensation.

The following results have been obtained:

III. Thickness of the quartz-plates 6.88 + 13.835 mM. = 20.715 mM.

<i>c</i>	$\Delta\lambda$	α	$\Delta\alpha_k$	$\Delta\alpha_s$	α_s	$\Delta\alpha_s/\alpha_s$	<i>n</i>
27.84	601 +	1°.007 +	0°.086	1°.093	439° +	0.00249	10
»	539	1°.455	0°.108	1°.563	555°	282	15
»	»	1°.399	»	1°.507	»	272	15
»	»	1°.440	»	1°.557	»	281	15

Conc. 27.84, mean value of $\frac{\Delta\alpha_s}{\alpha_s} = + 0.00273 \pm 0.00007$

In the same manner as before we find for the relative variation per unit of length:

<i>c</i>	$\Delta\beta/\beta$
27.84	0.00270

¹⁾ Communication No. 35.

From this result, compared with the preceding ones, it appears that a relation of this quantity to the concentration cannot be deduced from these measurements.

2. It is interesting to test the results here obtained by a theory proposed by TAMMANN ¹⁾. According to this theory the coefficient of variation of the specific rotatory power by external pressure is equal to the coefficient of variation by internal pressure, which latter can be varied by adding sugar, or any inactive salt.

We therefore will proceed to the calculation of the specific rotatory power γ . This quantity is related to the rotation per unit of length by the equation $\beta = c\gamma$. If now the external pressure varies, all three quantities, appearing in this equation, will vary, and we find

$$\frac{\Delta\beta}{\beta} = \frac{\Delta c}{c} + \frac{\Delta\gamma}{\gamma}$$

No measurements are known of the quantity $\frac{\Delta c}{c}$, which is equal to the coefficient of compressibility $-\frac{\Delta v}{v}$.

Accepting the hypothesis of TAMMANN, we can, in the manner pointed out by him, deduce this coefficient from the researches of AMAGAT on the compressibility of water at high pressures ²⁾.

For doing this let us take a volume v_0 of the solution at an external pressure = 0, in which the internal pressure is greater by ΔK atm. than in water and further an equal volume $w_{\Delta K}$ of water at an external pressure

¹⁾ See e. g. TAMMANN, Zeitschr. f. phys. Ch. XIV, p. 433 (1894).

²⁾ TAMMANN, Zeitschr. f. phys. Ch. XVII, p. 620 (1895).

ΔK , so that $v_0 = w_{\Delta K}$. Supposing we increase for both fluids the external pressure with p , according to the hypothesis of TAMMANN both volumina will still be equal, hence $v_p = w_{\Delta K + p}$. Defining the coefficient of compressibility μ for water by the relation

$$\mu = -\frac{1}{w_0} \cdot \frac{dw_p}{dp} \quad 1)$$

in which μ is a function of p , we find

$$w_p = w_0 \left(1 - \int_0^p \mu dp \right)$$

and further

$$v_0 = w_{\Delta K} = w_0 \left(1 - \int_0^{\Delta K} \mu dp \right)$$

$$v_p = w_{\Delta K + p} = w_0 \left(1 - \int_0^{\Delta K + p} \mu dp \right)$$

and for the wanted quantity $\frac{\Delta c}{c}$ we find

$$\frac{\Delta c}{c} = -\frac{\Delta v_0}{v_0} = \frac{v_0 - v_p}{v_0} = \frac{\int_0^{\Delta K + p} \mu dp}{\Delta K} \cdot \frac{1}{1 - \int_0^{\Delta K} \mu dp}$$

in which we must take $p = 100$.

Assuming moreover with TAMMANN the empiric relation

¹⁾ Our μ is identical with $\frac{\delta v}{\delta p}$ of TAMMANN, see l. c. p. 622.

$\mu = \frac{A}{p + B}$, in which A and B are constants, we find

$$\frac{\Delta c}{c} = \frac{Al \frac{B + \Delta K + p}{B + K}}{1 - Al \frac{B + \Delta K}{B}}$$

The values of ΔK have been deduced by TAMMANN from observations of MARIGNAC on thermal expansion¹⁾; the constants A and B have likewise been calculated by TAMMANN from the researches of AMAGAT.

With these values (mean temperature 10°) we find:

c	ΔK	$\frac{\Delta c}{c}$	$\frac{\Delta \beta}{\beta}$	$\frac{\Delta \gamma}{\gamma} = \frac{\Delta \beta}{\beta} - \frac{\Delta c}{c}$
9.48	238	+ 0.00449	+ 0.00268	- 0.00181
18.70	465	418	252	166
27.84	669	398	270	128

3. We now have to test the theory of TAMMANN by comparing the quantity $\frac{\Delta \gamma}{\gamma}$ with the coefficients of variation by increasing concentration. This comparison will be made in the following manner. We begin with deducing from experimental data the coefficient of variation of the specific rotatory power. Assuming with TAMMANN that the coefficient of variation found by us agrees with a variation of the internal pressure $\Delta K = 100$, we shall find from this how much the internal pressure changes for $\Delta c = 1$, and this value we shall compare with the one which follows from the values deduced by TAMMANN in different other ways.

¹⁾ TAMMANN, Zeitschr. f. phys. Ch. XIII, p. 179 (1894); XXI, p. 532 (1896).

From the formulae of TOLLENS on the relation between specific rotatory power and concentration we deduce

c	$\frac{\Delta \gamma}{\gamma}$ (for $\Delta c = 1$)	$\frac{\Delta \gamma}{\gamma}$ (observed)	Δk
9.48	- 0.000238	- 0.00181	13.1
18.70	243	166	14.7
27.84	068	128	5.6

This last quantity represents the value of ΔK for $\Delta c = 1$, which according to TAMMANN follows from the formulae of TOLLENS. The values of ΔK , deduced by TAMMANN from other phenomena, give for this the much greater value 23.4.

4. Further we can test the theory of TAMMANN by researches of FARNSTEINER¹⁾ on the variation of the salt. Knowing the variation of $\frac{\Delta \gamma}{\gamma}$ which agrees with $\Delta K = 100$ from our measurements, connected with the hypothesis of TAMMANN, we can calculate ΔK from some of the measurements of FARNSTEINER, and then compare these values with those deduced by TAMMANN from other phenomena.

The results of this comparison are found in the following table. In this we represent by

- A the values of $\frac{\Delta \gamma}{\gamma}$ for $\Delta K = 100$, as found above,
 B the values of $\Delta \gamma$ from measurements of FARN-

¹⁾ FARNSTEINER, Ueber die Einwirkung einiger anorganischen Salze auf das optische Drehungsvermögen des Rohrzuckers. Diss. Jena, 1890.

STEINER, for an addition of a weight 1 of salt to n of water,

C the values of $\frac{\Delta\gamma}{\gamma}$, following from B ,

ΔK the increase of the internal pressure, calculated from our measurements for the addition mentioned above,

ΔK_T the values of this quantity according to those found by TAMMANN in other ways.

	c	A	B	n	C	ΔK	ΔK_T
NaCl	9.48	-0.00181	1.42	9.93	-0.0213	1120	1090
	18.70	166	3.01	4.73	452	2720	2030
KCl	9.48	-0.00181	1.04	9.93	-0.0156	862	636
	18.70	166	2.06	4.73	309	1860	1150
BaCl ₂	6.48	-0.00181	0.39	9.93	-0.00584	323	526
	18.70	166	0.57	4.73	855	515	990
CaCl ₂	9.48	-0.00181	1.13	9.93	-0.0169	934	1140
	18.70	166	2.38	4.73	357	2150	2400

The agreement is satisfactory for some salts, in other cases on the contrary we find important differences.

5. Finally we come to the conclusion that the variation of the specific rotatory power by pressure, by variation of concentration and by the addition of an inactive salt is a more complicated phenomenon than we are led to suppose by the hypothesis of TAMMANN.

COMMUNICATIONS

FROM THE

PHYSICAL LABORATORY

AT THE

UNIVERSITY OF LEIDEN

BY

PROF. DR. H. KAMERLINGH ONNES,

Director of the Laboratory.

No. 39.

(REPRINT).

A. VAN ELDIK. Measurements of the capillary ascension of the liquid phase of a mixture of two substances in equilibrium with the gaseous phase (*With two plates*).

(Translated from: *Verlagen van de Afdeling Natuurkunde der Kon. Akad. van Wetenschappen te Amsterdam*, 29 Mei 1897, p. 18-24, en 21 Juni 1897, p. 74-78.)

A. VAN ELDIK. *Measurements of the capillary ascension of the liquid phase of a mixture of two substances in equilibrium with the gaseous phase.*

1. If, with a view to v. D. WAALS's theory of capillarity and in connection with his theory of the ψ -surface, we wish to measure the changes of the capillary ascension of the liquid phase of a mixture of two substances, during successive variations in composition along a connode-curve presenting a plaitpoint, as far as next to this plaitpoint, several experimental difficulties occur.

In the first place care should be taken that the mixtures to be experimented with, are not mixed, even in the smallest degree, with other substances. Indeed, the experiments concerning the ψ -surface, the plaitpoints and capillarity, made by KUENEN, DE VRIES and VERSCHAFFELT at Leiden, have sufficiently shown that it is of much importance to secure a high degree of purity, if one wishes to obtain trustworthy results from this kind of investigations.

Secondly in these measurements some conditions should be considered, the satisfying of which requires particular precautions. Indeed, in performing those measurements we must be sure, that both spaces separated by the meniscus are filled with one single phase

each; i. e. that in each of those spaces apart, the composition is everywhere the same as near the meniscus. And further it is generally necessary, in measuring the ascensions, to bring the meniscus at different heights in the capillary tube, and to renew repeatedly the liquid phase in this tube.

Now, again according to the experiments of KUENEN, it is very difficult to satisfy the first condition, and to exclude the phenomena of retardation, if the phases are not thoroughly stirred. In order to satisfy the second condition, we might support the liquid phase by a movable column of mercury. But in moving this, we should disturb the equilibrium, supposed to have been reached by stirring, if by doing so the space containing the total quantity of the substance were diminished. Hence this space should be kept constant, notwithstanding the moving of the mercury-meniscus.

Of course it is necessary to keep continually the temperature rigorously constant, in order to maintain the equilibrium.

By the arrangement of the apparatus to be described hereafter, the difficulties opposing the satisfying of the enumerated conditions were overcome, and constant values could be obtained for the ascensions, among others with mixtures of Methylchloride and Ethylene. In giving this description we suppose that we are experimenting with the said substances at a temperature a little above the temperature of the room, so that f. i. the point of 23° for a mixture of Methylchloride and Ethylene is investigated.

2. *Experimenting-tube.* This tube is represented in

fig. II. The part, serving for measuring the ascensions is the wide glass tube *B*, which contains the capillary tube, centred by two narrowings. At the top *B* carries a piece of narrow glass tube, which ends as a capillary tube within the wider piece *A*, in order to prevent the particles of dust, perhaps carried along by the gas streaming in, from reaching the tube *B*, serving for the measurements, *A* serves at the same time to receive the Methylchloride which may have condensed already in the copper supplying tubes, and so may have grown impure by the contact with the copper cocks, etc.

In order to allow us to move the meniscus at liberty — which is necessary also if we wish to make a series of experiments, beginning with the same quantity of Methylchloride, because at high pressure the gas dissolves to a high degree in the liquid, causing the latter to increase considerably — *B* is not closed at the nether end, but provided with a reservoir *C*, connected along the glass tube *b*, the steel capillary tube *c* and the glass tube *d*₁, with the pressure-cylinder *D*₁, filled with mercury. The pressure of an hydraulic pump is transmitted along copper tubes filled with glycerine to the mercury in this cylinder, and so in *C* the mercury-meniscus may be moved at liberty. The steel capillary tube, connected to the glass tubes *b* and *d*₁ by two brass pieces *e*, fastened to the glass by means of sealing wax, consists of two pieces, joined by a steel cock *I*, which closes the experimenting-tube every time when the surface of the liquid has been raised to a suitable level; in this way any leakage of pressure-cylinder or pump

was rendered harmless. With the pressure cylinder D_1 is also connected the air-manometer M , placed in another cylinder D_3 , and so the pressure, occurring in the experimenting tube, (the parameter giving the places of the phases on the connode-curve) is read. By means of the cock III the manometer may be disconnected from the remaining part of the apparatus.

The reservoir C is useful, as explained above, to secure sufficient room for the moving of the meniscus, and moreover to hasten the mixing by means of a stirring-apparatus.

For this reason in this reservoir, rather large in order to allow us always to mix the liquid phase with a sufficient quantity of the gas, a little soft-iron bar is placed, of about 6 cM. length, wholly covered with glass, in order to allow the cleaning of the tube with acids, and which is provided with two glass knobs, preventing it from adhering to the side of the glass tube, and facilitating the mixing by increasing the surface of it.

This stirring-bar is moved as in KUENEN'S experiments by means of electro-magnetism.

Instead of placing the windings of the coil on the iron cylinder itself, surrounding the experimenting tube, I used a common coil K (containing about 500 windings, 7 cM. in diameter and 14 cM. long), which is moved outside the water-bath surrounding the experimenting tube, whilst within it, close round the latter, a soft-iron cylinder k is suspended to the same support as the coil K , and so may be moved simultaneously with the latter.

By this means a rather strong magnetic field is obtained along the axis of the coil, so as to enable us to apply a sufficient force on the stirring bar with a moderate current (± 2 Amp., obtained from 2 Bunsen cells), generating but little heat in the coil. In order to prevent the iron from touching the glass tube, the little iron cylinder was wholly covered with sealing wax.

The current through the coil was closed only during the stirring, which consists in moving the stirring-bar up and down through the surface of the liquid. As soon as by stirring the liquid in C together with the gas above it, the thermodynamical equilibrium of the two phases, corresponding to the given temperature and pressure, has been reached, in forcing up the mercury in C , we bring the surface of the liquid into B , and there read the height of ascension.

The condition, that the whole volume, occupied by the substance, should not be altered by moving the mercury in C is now satisfied, by moving simultaneously with the mercury meniscus in C another mercury column as much in opposite direction in the reservoir E , which is connected with the space in the experimenting tube. The reservoir E is connected with the upper end A of the observation tube B by a copper tube h and the T-piece T . Along the steel tubes f it is connected with the glass tube d_2 of the mercury cylinder D_2 , from which it may be disconnected by the steel cock II.

Both parts: the experimenting tube $A.B.C.$ and the reservoir E I placed one above the other in the same water bath in order to secure as much as possible the

same temperature for the whole space filled with the mixture; to this end the T-piece *T* received the shape represented in fig II. In the brass piece *g* is soldered with silver the copper tube *h*, which at the nether end carries a screw-nut, to be applied on the brass screw, which is fastened with sealing wax on the tube *A*. In order to prevent the aperture of the leather ring from being stopped up in applying the nut on the screw, a piece of steel capillary tube (± 1 cM. long) is put into this aperture.

A similar arrangement is made at the second branch *j* of the T-piece, which leads to the reservoir *E*.

The third branch leads to the supplying tube *l*, along a capillary tube and the capillary cock IV.

The inferior end of *E* is connected by an arrangement *m* of the same kind, but which, on account of the presence of mercury, has been made in steel, with the steel tubes *f*₁ and *f*₂, and the steel cock II, leading to the pressure cylinder *D*₂.

3. *The Moving of the Phases.* In order to enable us to give to both mercury-niveaus equal and opposite displacements, — which will always be of use in experiments on equilibrium of phases — a compound hydraulic pump was constructed, as represented by a scheme (*V*) in fig. I. It consists in a strong brass cylinder, divided into two parts by a piston. These two parts are connected through the cocks V and VI, each to one of the cylinders *D*₁ and *D*₂, and so the displacement of the piston is transmitted on the mercury in *C* and *E*.

The two other taps of the pump are connected with

each other by the cocks VII and VIII, and further through the cock IX with a common hydraulic pump *P* (pump for testing manometers, from SCHÄFFER und BUDENBERG, who also constructed the above described apparatus). This arrangement enables us to displace both menisci either separately or simultaneously.

In order to move only the meniscus in *C* the cocks I, V, VII, IX are opened, VIII is shut; in order to move only the meniscus in *E*, II, VI, VIII, IX should be open, VII shut. The motion is then obtained by means of the hydraulic pump *P*. For the simultaneous motion of the two niveaus VII and VIII are shut, and I, II, V, VI are opened.

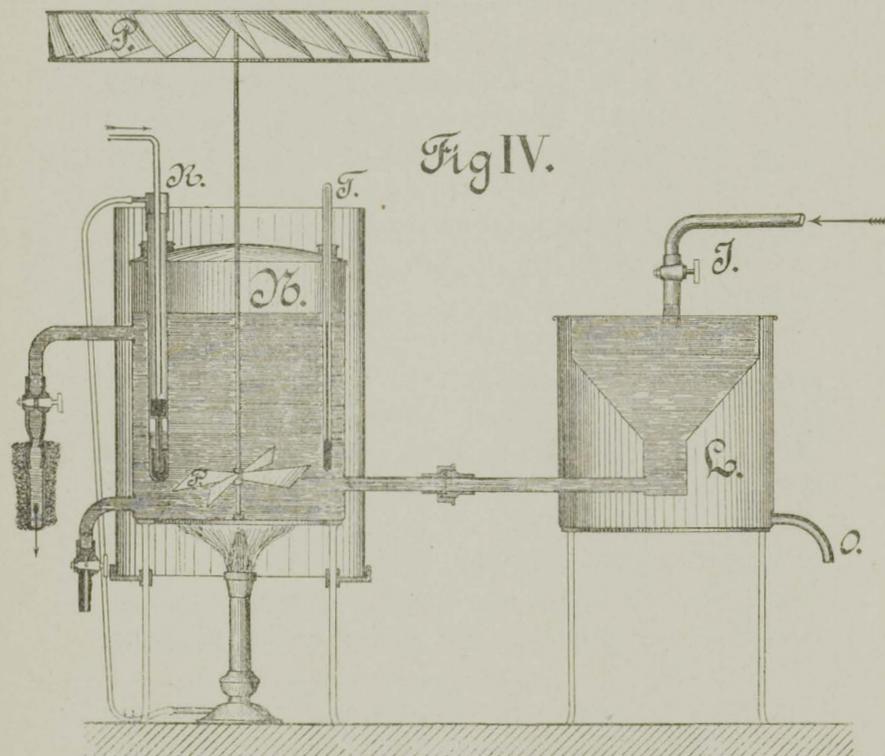
Further particulars are shown by fig III.

In order to make really equal the quantities of glycerine, displaced on both sides of the piston, the piston-rod is prolonged on both sides outside the cylinder; it passes through leather packing-rings. The piston-rod ends free at one end *S*₁; the other end *S*₂ is fastened to the frame *F* and follows its motion, which is obtained by turning the wheel *H* riveted to the screw *G*. By means of this wheel we may give a very slow motion to the piston.

4. In order to get a suitable and sufficiently constant temperature, a warm water bath is used, as represented in fig. I.

The water, streaming through the water bath, is heated in a vessel, placed at a height of about $4\frac{1}{2}$ M., and provided with a thermoregulator. In order to cause the water to stream out of this vessel under a constant difference of level, and so with a constant velocity, the

arrangement represented in fig. IV, was made. The cock *I* supplies the funnel *L*, which communicates with the heating vessel *N*, with a little more water than streams down out of *N* after being heated. The super-



fluous water flows into the vessel placed round *L*, and streams off through *O*.

Besides the thermoregulator *R* and the thermometer *f* a stirring-apparatus *P* (after the example of OSTWALD) is placed in the vessel *N*, and is made to revolve by the

heated air ascending from a Bunsen-burner. The flame placed under *N* however is not sufficient for this purpose; a second flame is wanted, placed outside the asbestos screen standing round *N*. The water heated thus streams through wool-covered tubes, and enters from above the cylindrical space between the two glass tubes, the inner of which contains the experimenting tube, and is filled with water in rest. This water is slowly heated by the warm water streaming along, and after the state of equilibrium of temperature has been reached, it renders quite imperceptible the still possible small variations of temperature. Accordingly the temperature was kept constant for hours within 0.1 ; moreover it was possible to adjust it on various days exactly at the same value. As on the bursting of the experimenting tube, which occurred repeatedly, the two tubes of the water-bath were usually crushed likewise, the whole apparatus was placed in the oaken case, already described by DE VRIES. ¹⁾

5. *Apparatus for obtaining the Mixtures.* In fig. V a scheme is given, representing the purificating apparatus, by means of which the substances, used by me, were subjected to the process of purification, indicated by KUENEN ²⁾. The cross-shaped copper tube *n*, connected through the cock *X* with the copper tube *l* leading to the experimenting tube, joins the two parts, each of which serves for one of the two components of the mixture. The piece *n* carries also the

¹⁾ Diss. Leid. 1893.

²⁾ Arch. Néerl. 26. 1893. p. 354.

two cocks XV and XVI, one of which, leading to the mercury-air-pump, enables us to evacuate every part of the apparatus at liberty, the other serving to let escape the gas. Through the cock XI the piece n is connected with the purifying apparatus for the Methylchloride, p is the copper cylinder containing the impure liquefied gas, such as is supplied by trade; q is a strong iron tube, filled with P_2O_5 , and r the strong copper cylinder, in which by cooling in a mixture of solid carbonic acid and alcohol the Methylchloride is liquefied. During the distilling the cocks XII and XIV are opened, XI shut. With short intervals now XIII is opened for a moment to admit the gas, dried in q , which then condenses in r , as is evident from the manometer m falling back every time. Not before this falling back ceases, thus indicating r to be wholly filled with the liquid, XIII is shut definitively; then through XI and XV we let boil away some Methylchloride to drive out the more volatile impurities, the less volatile impurities remaining in p .

For the Ethylene, (obtained at the Leiden Laboratory itself from alcohol and sulfuric acid, and kept in common iron cylinders under a pressure of 30–45 atm) the process is wholly the same.

As storing cylinders for the Ethylene I used the two strong copper cylinders r' and r'' , each provided with a cock. Having two cylinders is useful, first because in this way the quantity of Ethylene stored up is, as is necessary, greater than that of Methylchloride; secondly this arrangement enables us to reach with the Ethylene higher pressures in the experimenting tube, than would be the case in using one greater reservoir; for we first

admit as much as possible of the gas into the experimenting tube, and open r'' not before r' has been shut.

Easily and quickly so much Ethylene may be distilled from the large storing-cylinders into these reservoirs by cooling in alcohol and solid carbonic acid, that pressures of over 100 atm. are reached in returning to the temperature of the room.

6. *Experiments on mixtures of Methylchloride and Ethylene.* By means of the apparatus described above the changes were measured of the capillary ascension with variation of the pressure (the parameter, giving the places of the phases on the connode curve), the temperature remaining constant. The pressure was read from an air-manometer (*M.* fig. 1). In order to obtain the capillary ascension I measured the vertical distance h between the lowest point of the meniscus in the capillary tube, and the lowest point of the meniscus in the annular space, left between the capillary tube and the observation tube.

The real ascension, — i. e. the ascension which the liquid would show, if the capillary tube were placed amidst an infinite horizontal niveau — is obtained, as shown by VERSCHAFFELT, by adding to the measured height the correction

$$h' = h \frac{2d}{\frac{(r_3 - r_2)^2}{r_1} - 2d} + \frac{r_1}{3}$$

in which

h = the ascension as measured
 d = the height of the annular meniscus
 r_1 = the inner —
 r_2 = the outer radius of the capillary tube
 r_3 = the inner radius of the observation tube.

The thus obtained real ascensions $H = h + h'$ were then multiplied by $10r_1$, and so I obtained the real ascensions H_1 which would have been measured in a capillary tube with a radius of 0.1 mM. In order to obtain r_1 I used the value of the surface-energy of Methylchloride, which has been measured very exactly by VERSCHAFFELT¹⁾, who found the real ascension in a capillary tube with a radius = 0.1 mM. to be:

$$H_1 = 42.09 - 0.265 t.$$

As an example may serve the calibration of the tube IV. In order to estimate the correction h' I used the results of a microscopic measurement of r_1 at both ends of the tube

$r_1 = 0.104$ and 0.107 mM.: mean value = 0.106 mM.

r_2 was measured by means of a micrometer

$r_2 = 0.525$ and 0.555 mM.: mean value = 0.540 mM.

By measuring with a cathetometer and by weighing a mercury-column I found

$$r_3 = 3.04 \text{ mM.}$$

Measuring the ascensions h and the height of the annular meniscus d at distances a mM. from the top of the tube, I obtained the following values:

¹⁾ Not yet published.

t	a	h	d	H	r_1
12°	65	34.84	1.31	36.53	0.1065
»	75	35.00	.30	.69	.1061
»	85	.16	.33	.85	.1056
»	95	.38	.34	37.07	.1050
»	105	.63	.31	.29	.1044
»	115	.78	.32	.47	.1039

I repeated the measurements with a renewed quantity of the liquid, and obtained agreeing results.

In order to obtain the mixtures, the ascension of which has to be measured, we may proceed in two different ways:

1°. Taking a quantity of Methylchloride, we may add repeatedly a small quantity of Ethylene, and so obtain successively several mixtures, each of which contains more Ethylene than the former. Thus at last the phenomena of the plaitpoint will be observed.

As at the temperature of my experiments (10° and 23°) the plait occupies nearly the whole breadth of the ψ -surface, and accordingly the plaitpoint is reached only when a very great quantity of Ethylene is dissolved, it is necessary to begin every time with different quantities of Methylchloride, in order to study different parts of the *Ascension-Pressure-Curve*.

So f. i. the volume of the liquid phase increased in the neighbourhood of the plaitpoint at both temperatures to about 30 or 40 times the original volume, which compelled me, on account of the dimensions of my apparatus, to begin with so small a quantity of Methylchloride, that I was not able to bring the meniscus to the suitable place in the capillary tube, (the quantity

of the liquid being too small), before the pressure had reached ± 30 atmosph. Besides on these occasions however I repeatedly renewed the mixtures, in beginning with a new quantity of Methylchloride, in order to rid myself of possible accidental impurities.

2°. The second way to obtain mixtures of different composition, is to begin with a mixture containing more Ethylene. In repeatedly letting boil off some Ethylene, I successively obtained different mixtures, each of which contained less Ethylene than the former. But also when proceeding in this way, it is necessary to begin with different quantities of Methylchloride, in order to study different parts of the curve.

Both methods I applied in my experiments, and in doing so I obtained well agreeing values.

This was especially the case in my second experimenting series (23°), where particularly the second method of preparing the mixtures, the method of decreasing pressures, led comparatively quickly to a constant result, every time however with the aid of the stirring apparatus. Usually I obtained already constant values of the ascension, after having twice brought the liquid into C with the aid of the compound-pump V (fig. I), and having stirred it there together with the gas.

With increasing pressures on the contrary, often a 10 times repeated stirring was necessary, before the ascension became constant. Very likely the chief reason of this is, that the small quantity of the liquid that remains in the capillary tube is renewed very slowly and so reaches the thermodynamical equilibrium, ap-

pertaining to the given pressure, only by repeatedly moving the meniscus up and down; with the method of decreasing pressures on the contrary, this small thread of liquid is immediately broken and driven out of the capillary tube by the Ethylene boiling away.

7. *Changes of the capillary ascension with variations of pressure as far as the plaitpoint.* A first series of experiments was made at the temperature of the water from the supply, about 10°.4 C, only a little over the critical temperature of Ethylene (9°). At slightly differing temperatures, I repeatedly observed the decrease of the ascension with increasing pressures as far as the plaitpoint. In the immediate neighbourhood of the plaitpoint however it appeared nearly impossible to obtain constant values, on account of the great disturbances caused there by gravitation and by changes of temperature, even when nearly imperceptible. So f. i. I sometimes noticed a sudden change of the ascension, without any perceptible cause, and sometimes the ascension even turned negative. Yet even then the meniscus remained sensibly concave, thus incontestably proving this phenomenon to be caused by the liquid not being homogeneous within and without the capillary tube. The plaitpoint-pressure at this temperature I derived from the plaitpoint-pressure at 11°.6 — which I found to be 56.07 ± 0.10 atm. — by means of a correction, obtained graphically by using the plaitpoint-pressure (50.15 ± 0.10 atm.) of 23° C., observed in my second series of experiments.

In Table I are given the results of the measurements made at 10°.4 C.

A second series of experiments, the results of which are given in Table II, was made at the temperature of 23°, kept constant by the apparatus described above.

In consequence of the greater distance from the critical temperature of Ethylene and of the temperature being much more constant, I succeeded here much better in obtaining constant and well agreeing values, as is evident from the graphical representation in fig. VI. There the curve *A* gives the the ascensions and the pressures of pure Methylchloride at different temperatures. The two series of observations concerning the mixtures of Methylchloride and Ethylene at 10°.4 and at 23° are represented by the curves *B* and *C*.

Table I.

Ascension-Pressure-curve at 10°.4 C.

<i>t.</i> ° C	<i>P.</i> atm.	<i>H</i> ₁ mM.
10°.4	3.60	39.33
2	15.30	30.13
4	19.54	26.96
6	23.61	23.50
3	29.50	19.64
2	32.17	17.74
3	37.93	14.24
2	39.65	13.49
4	44.05	10.55
3	46.28	8.67
4	49.10	6.55
2	52.04	3.63
4	55.20	Plaitpoint (derived from that at 11°.6.).

Table II.

Ascension-Pressure-curve at 23°.0 C.

<i>t.</i> ° C.	<i>P.</i> atm	<i>H</i> ₁ mM.
23.00	5.25	36.00
14	20.62	25.16
08	26.45	23.20
15	30.64	18.50
05	35.20	15.53
15	40.71	12.40
07	41.56	11.79
00	43.26	10.92
02	45.85	9.71
07	50.74	7.39
07	52.48	6.20
00	54.11	4.92
00	59.15 ± 0.09	Plaitpoint.

Fig. I.

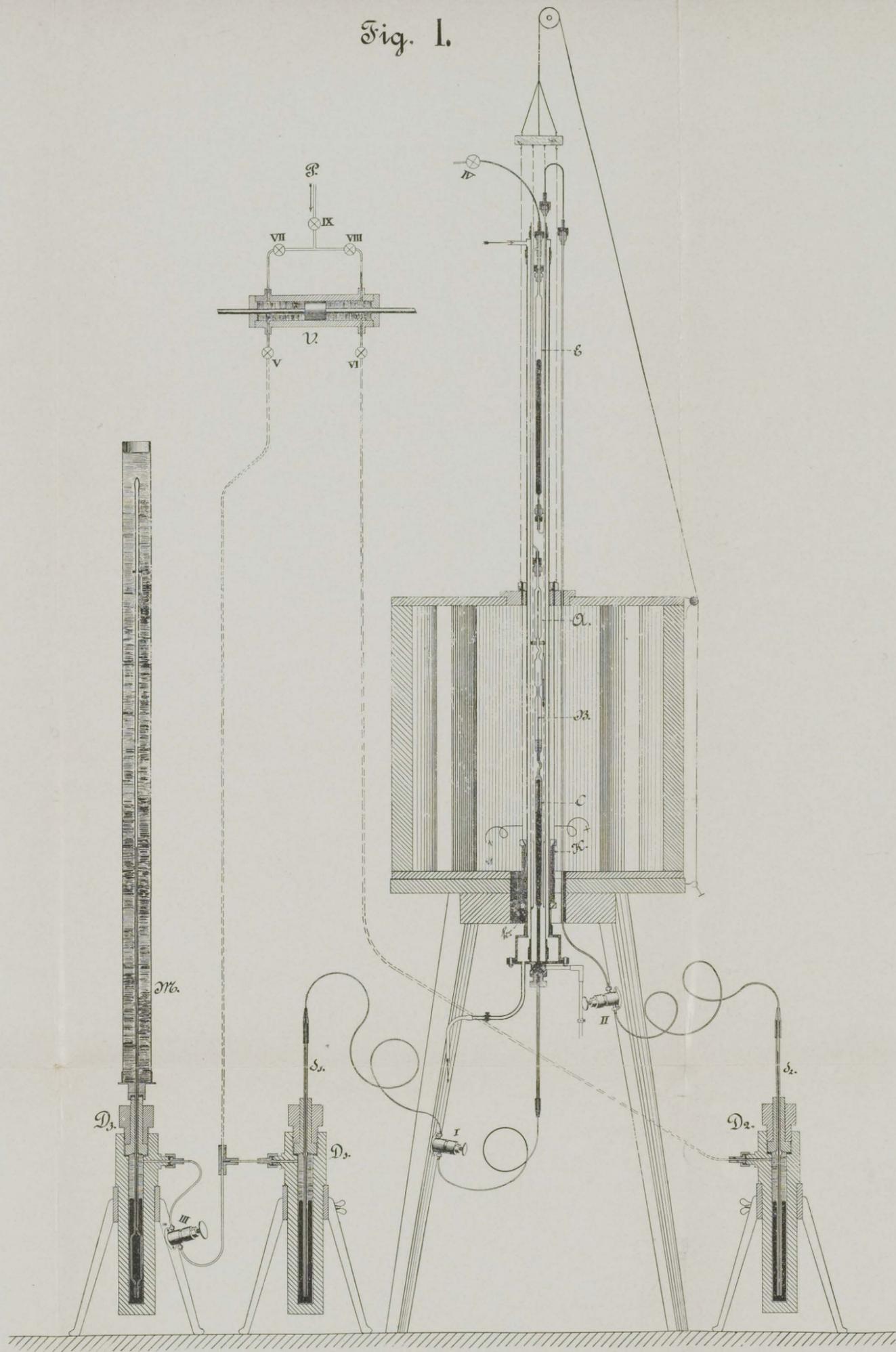
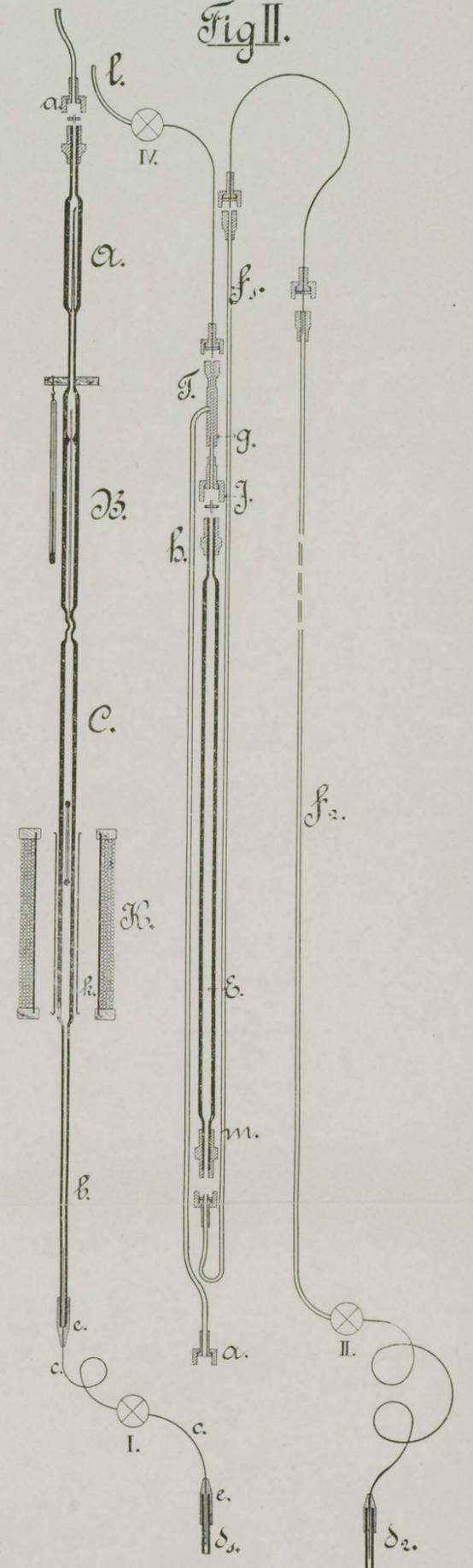


Fig II.



56.

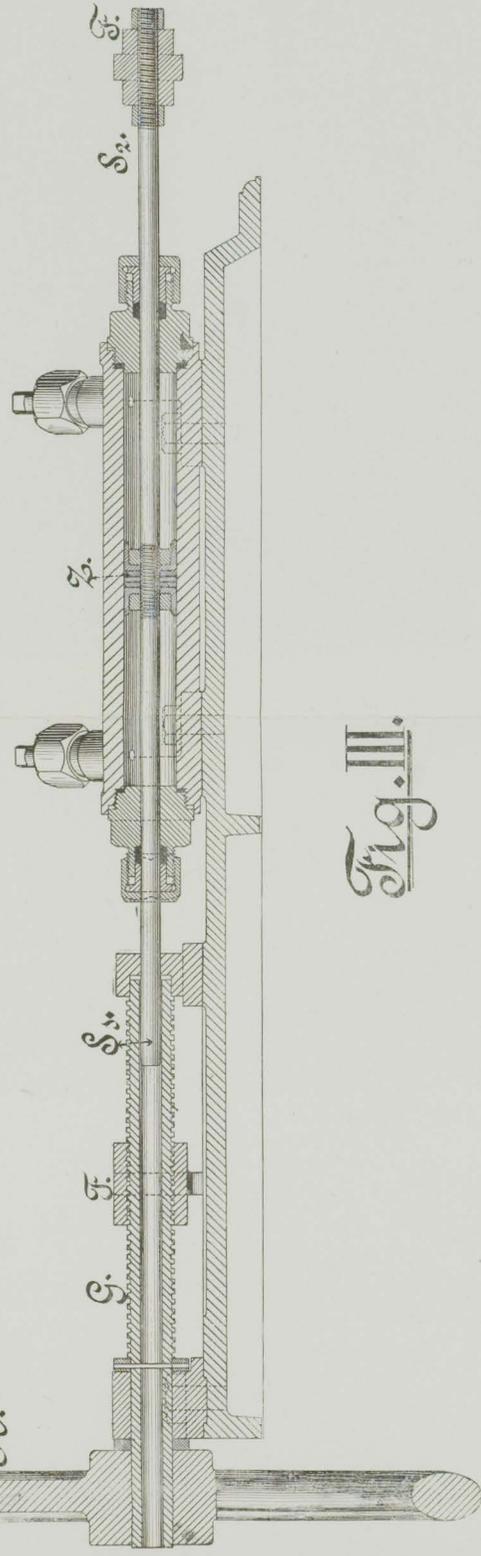


Fig. III.

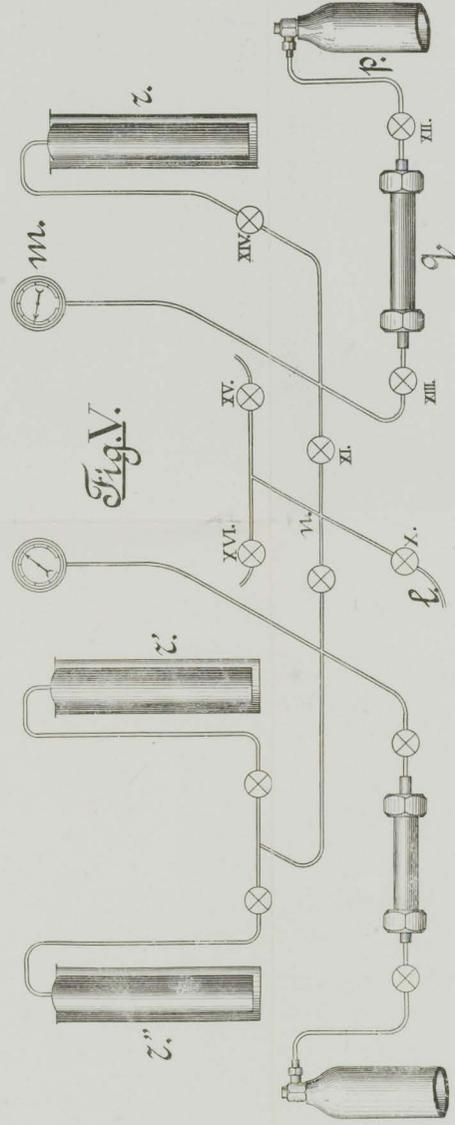
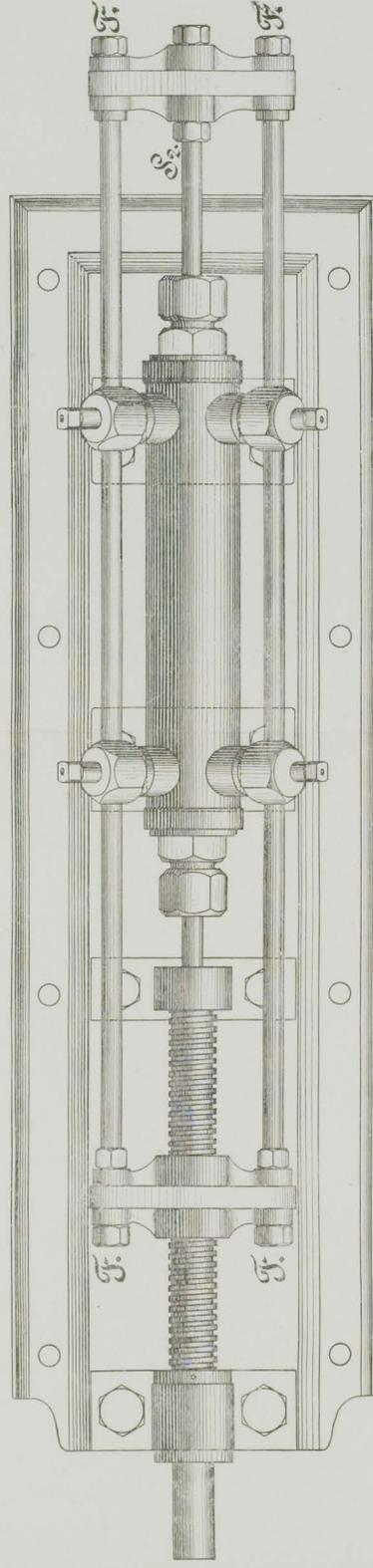


Fig. V.

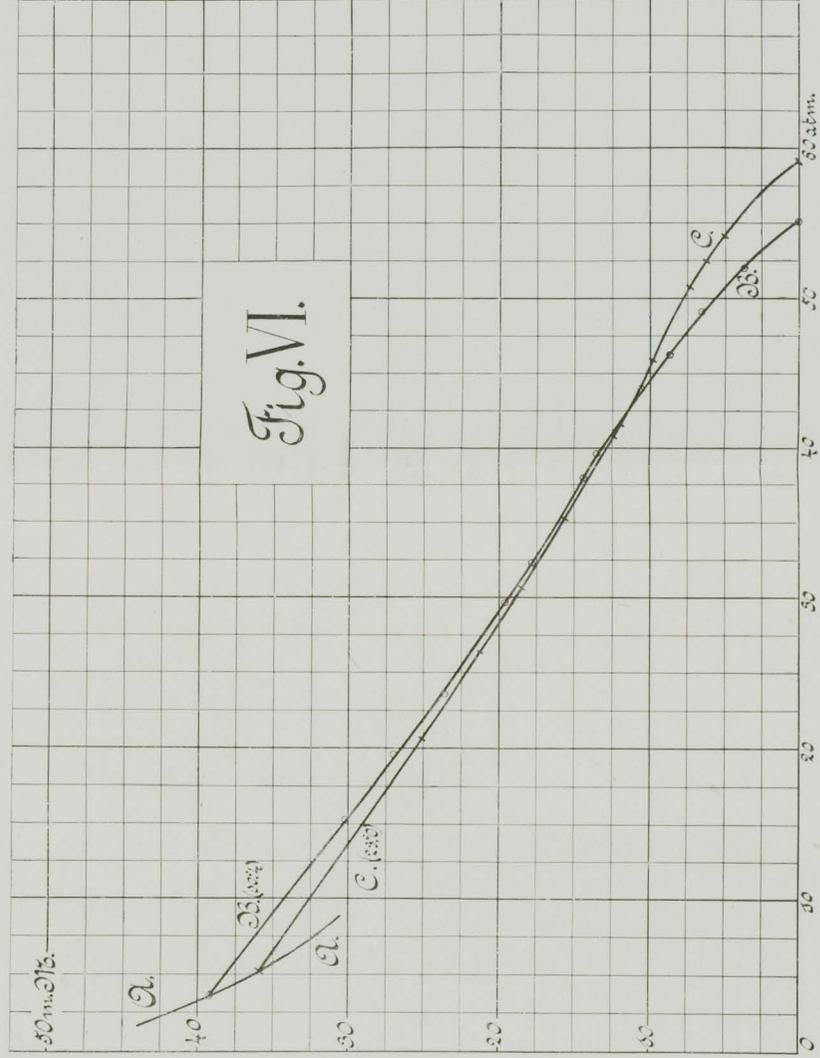


Fig. VI.

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COMMUNICATIONS
FROM THE
PHYSICAL LABORATORY
AT THE
UNIVERSITY OF LEIDEN

BY
PROF. DR. H. KAMERLINGH ONNES,
Director of the Laboratory.

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**No. 40.**

(REPRINT).

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Dr. E. VAN EVERDINGEN Jr. On the HALL-effect and the
magnetic increase of resistance in bismuth (*with a plate*).
(*Translated from: Verslagen van de Afdeling Natuurkunde der
Kon. Akad. van Wetenschappen te Amsterdam, 26 Juni 1897,*
p. 68—74. Communications No. 26, 37 and 40 contain the most
important part of the inaugural dissertation of 7 July 1897.

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Dr. E. VAN EVERDINGEN Jr. *On the HALL-effect and the magnetic increase of resistance in bismuth.*

1. At the end of my last communication <sup>1)</sup> is mentioned, „we shall, for an arbitrary position of the plane for which we wish to know the HALL-coefficient, find that coefficient with the aid of the ellipsoid of revolution, construed with the extreme values”. In the accompanying figure 1 is further indicated how this should be understood. The plane of the plate is indicated by *Pl*, the principal crystallographic axis by *OA*. The intersection of the magnetisation-ellipsoid with the plane through *OA* and *OH*, the perpendicular to *Pl*, at the same time direction of magnetic force, is represented by the ellipse *AB*. Moreover the intersection *BD* is indicated of a second ellipsoid of revolution with this plane, whilst

$$\frac{OD}{OB} = \sqrt{\frac{OA}{OB}}$$

The direction of the magnetisation is given by the point where the tangent plane perpendicular to *OH* touches the second ellipsoid; the magnitude of the magnetisation by the length *OM* of the radius-vector through this point to where it meets the first ellipsoid. Now the HALL-coefficient is given

<sup>1)</sup> Communications N<sup>o</sup>. 37, p. 19.

by  $OR$ , the length of that same radius-vector to where it meets the ellipsoid of revolution, the section of which is indicated by  $EF$ .

By a single line is indicated already in this figure the intersection with the plane  $AOH$  of the plane perpendicular to  $OM$ , in which the greatest increase of resistance is found. In the § mentioned we remarked, that the resistance-ellipsoid will generally get three different axes. This is illustrated by fig. 2.  $Pl$ ,  $OH$  and  $OM$  have the same signification as before;  $W$  is the plane perpendicular to  $OM$ . Round  $OS$ , the principal crystallographic axis as axis we imagine the resistance-ellipsoid, of which are drawn the intersection  $ACD$  with plane  $SOH$  and the intersection  $BD$  with plane  $W$ . The length  $OP$  of the radius-vector  $OM$  in the ellipsoid of revolution, the intersection of which with  $SOH$  is indicated by  $GK$ , determines the value of the increase of resistance, which is equal for all directions in the plane  $W$ . Thence the ellipse  $BD$  is enlarged proportionally to  $B'D'$ . The new ellipsoid of resistances passes through this ellipse and touches in  $M$  the old one. The principal directions (axes of symmetry) of the plate are still  $OA$  and  $OB$ ; but the proportion between  $OA$  and  $OA'$  is another than that between  $OB$  and  $OB'$ .  $OC$  is not sensibly increased.

2. In my preceding communications on the above mentioned subjects <sup>1)</sup> some questions were proposed, which we will now try to answer, as far we are able to do so by this time.

<sup>1)</sup> Communications N<sup>o</sup>. 26 and 37.

In Communications N<sup>o</sup>. 26 we find on p. 6 a discussion of the question, how far we are allowed to write for the difference of potential at the secondary electrodes

$$e = \left\{ H + \frac{1}{2} \sin 2\alpha (K_{11} - K_{22}) \right\} \frac{I}{a}.$$

The correctness of this formula was then called in question first because of the small mean HALL-effect, found in all the round plates used. This argument no longer exists; later round plates gave sometimes very high values for the HALL-coefficient, and it is almost certain, that the small effect in the first plates was caused by a particular position of the principal crystallographic axis. (See Comm. N<sup>o</sup>. 37 § 8)

A second reason for doubt was given by the observation of differences in mean HALL-effect after turning the plate about the lines of magnetic force through angles of 45°, 90° etc. In connection with this it was thought desirable to construct two differently situated quadratic plates out of *one* round plate and to investigate them.

This investigation has been performed indeed, but appeared not to possess sufficient demonstrative power, since the two halves may show large differences even in *corresponding* positions. Hence it was necessary to proceed as follows:

In a round plate of bismuth the axes of symmetry are determined and the amount of dissymmetry is measured.

Then the plate is sawed into two thinner halves ( $a$  and  $b$ ); each of the halves is tested in the 8 principal positions, after a repeated determination of the axes.

Thereupon out of one a quadratic plate is cut with

axes of symmetry as diagonals, out of the other one a quadratic plate with sides parallel to the axes of symmetry. These quadratic plates are tested each in 4 positions.

The following proportions of the HALL-effect in plates *a* to that in plates *b* resulted from such an investigation:

mean of the 4 positions of symmetry  
and the 4 positions of dissymmetry in round plates 1,016  
mean of the 4 pos. of diss. in *b*  
to mean of the 4 pos. of symm. in *a* „ „ „ 1,035  
mean of the 4 pos. of diss. in *b*  
to mean of the 4 pos. of symm. in *a* „ quadratic „ 1,136.

So we find the difference between positions of symmetry and dissymmetry also in quadratic plates, even in a higher degree than in the round ones.

3. These experiments rendered it rather probable, that not the round form of the plates caused the observed differences. Certainty with regard to this matter was obtained by calculating the difference of potential between the secondary electrodes of a round plate of bismuth, placed in the magnetic field, to within terms of the third order. This calculation, in which  $H$  and  $(K_{11} - K_{22})$  were regarded as very small compared with  $(K_{11} + K_{22})$ , and which consisted in an approximation <sup>1)</sup> successively of terms without  $H$  or  $(K_{11} - K_{22})$ , terms with the first power of those quantities, etc., is too long to be given here; therefore I will communicate merely the results.

<sup>1)</sup> For this idea I am indebted to Mr. J. WEEDER, phil. doct. at Leiden.

1°. For round plates, in whatever position they stand with regard to the primary electrodes, the HALL-effect is always equal to

$$H \frac{I}{d},$$

and therefore always the full HALL-effect is found.

2°. Whether we adjust the secondary electrodes so as to annul the difference of potential in the zero magnetic field, or measure this difference of potential and allow for the increase of resistance <sup>1)</sup>, always the observed dissymmetry is equal to

$$\frac{4}{\pi} \sin 2\alpha \frac{I}{d} \{ (K_{11} + K_{22}) \Delta \left( \frac{K_{11} - K_{22}}{K_{11} + K_{22}} \right) \}$$

where  $\Delta$  means the increase in the magnetic field of the quantity in brackets.

The result from this calculation as to the total resistance of the plate between the primary electrodes was given in the communication of 20 April 1897 (§ 2) <sup>2)</sup>.

4. Also after the investigation mentioned in § 2 the difference between the values of the mean HALL-effect in positions of symmetry and of dissymmetry remains unexplained. As no theoretical meaning could be given as yet to this difference, it seemed that it had to be attributed to some new phenomenon; but doubt arose again because with some round plates tested afterwards the sign of the difference was reversed, viz. the greatest HALL-effect was found in the positions of dissymmetry. For the present we might still assume, in spite of the

<sup>1)</sup> See Communications N°. 26, p. 5.

<sup>2)</sup> Communications N°. 37, p. 5.

apparent regularity of the phenomenon <sup>1)</sup>, that the difference is due to the influence of irregular crystallisation (see communication 2 of 21 April 1897, § 7<sup>2)</sup>) especially as the electrodes in the positions of dissymmetry stand in places quite different from those of the positions of symmetry.

5. Many observations have been made also of the differences between the mean HALL-effect in the 4 possible positions of symmetry or the 4 possible positions of dissymmetry, to which as little theoretical meaning could be given as yet.

Here were taken into account the possible influences of:

Instability of the magnetising current (escaping observation by inaccuracy of the ampère-meter).

Non-homogeneity of the field.

Differences of temperature.

Bad contacts at the secondary electrodes.

The influence of these sources of errors was annulled or diminished by the use of more accurate instruments, by measuring the temperature and the contact-resistance, etc. but in spite of all this in the observed differences generally no traces of regularity could be discovered, and even in one position at different periods rather different values for the mean HALL-current were found. *These* differences therefore may very likely be ascribed to irregularity in crystallisation.

<sup>1)</sup> This came out strongly especially in plate R 3 where in a later series for all positions of symmetry  $\pm 9,10$ , for all positions of dissymmetry 7,80 was found.

<sup>2)</sup> Communications N<sup>o</sup>. 37, p. 17.

6. With a view to the results of the calculation communicated in § 3, the calculation of the resistances in the magnetic field in two directions at right angles, contained in § 1 and 2 of the second communication of 30 May 1896 <sup>1)</sup>, should be revised. We will confine ourselves here to the calculation for plate R 1. As nothing is known about the resistances in the zero magnetic field in the directions of the axes of symmetry, we must assume those resistances to have equal values. The formula for the dissymmetry becomes then simply

$$\frac{4}{\pi} (K_{11} - K_{22}) \frac{I}{d}$$

This therefore represents  $e_A - e_B$ .

If we mean by:

$w_A$  and  $w_B$  the compensative resistances for the two directions of magnetisation;

$w_r$  the resistance of the rheotan-wires of the compensative current;

$w_s$  " " in the secondary circuit;

then the difference of the secondary currents is:

$$I \left( \frac{w_r}{w_A} - \frac{w_r}{w_B} \right)$$

So  $e_A - e_B = I \cdot w_r \cdot w_s \cdot \left( \frac{1}{w_A} - \frac{1}{w_B} \right)$ .

The quantity  $D$ , always used as measure for the dissymmetry, is <sup>2)</sup>

$$1000 \left( \frac{1}{w_A} - \frac{1}{w_B} \right)$$

<sup>1)</sup> Communications N<sup>o</sup>. 26, p. 13 and 16.

<sup>2)</sup> " " N<sup>o</sup>. 26, p. 11.

whence we find

$$K_{11} - K_{22} = \frac{\pi}{4} d \frac{w_r \cdot w_s \cdot D}{1000} \cdot 10^9 \text{ c. g. s.}$$

By means of this formula we found for plate R 1 in a field of 8600 c. g. s.

$$K_{11} - K_{22} = \pm 4200.$$

7. The general formula for the dissymmetry and the conception of the influence of a magnetic field with the resistance of bismuth, worked out in § 1, enable us to inquire, what we may expect the variation of the dissymmetry and the increase of resistance to be in varying magnetic field and temperature. A very simple calculation shows for instance, that if we call  $p$  the increase of resistance in % for the direction  $OB$  (fig. 2) and neglect higher powers of  $p$ , the increase of resistance in the direction  $OA$  is given by  $p \cos^2 \alpha$ , where  $\alpha$  represents the value of the angle  $MOH$ . So long as  $\alpha$  remains constant, also the ratio between the two increments of resistance will remain the same, and therefore the dissymmetry will be proportional to the mean increase of resistance. If however  $\alpha$  varies, this proportionality cannot exist any longer. Now the value of  $\alpha$  is determined wholly by the ratio of the permeability in the directions  $OA$  and  $OB$  (fig. 1). Hence from the observations with plate R 2<sup>1)</sup> would follow, that in this plate the ratio spoken of varies considerably, when we alter the magnetising force.

8. In Communications N<sup>o</sup>. 26, § 4, p. 20 attention was directed to a similarity in the variation with tem-

<sup>1)</sup> Communications No. 26, p. 15.

perature of conductivity and HALL-effect in bismuth. Inter alia we deduced from the observations of HENDERSON, that probably in a field of 6000 c. g. s. units a maximum conductivity would be found for bismuth at a temperature below 0°; in connection with the observation of such a maximum at low temperature for the HALL-effect with bismuth II by LEBRET and with bismuth I by myself this was regarded as a confirmation of the similarity mentioned. Since the publication of that communication the maximum conductivity for electrolytic bismuth has really been observed by FLEMING and DEWAR<sup>1)</sup> below 0°; but my further experiments at low temperatures (from 0° to -70°) with bismuth from MERCK and bismuth from OBERSCHLEMA show for neither of these specimens a maximum HALL-effect, not even an approach towards a maximum. Hence the similarity between conductivity and HALL-effect is not confirmed by these later experiments.

<sup>1)</sup> Proc. Roy. Soc. 60, p. 425.

Fig. I.

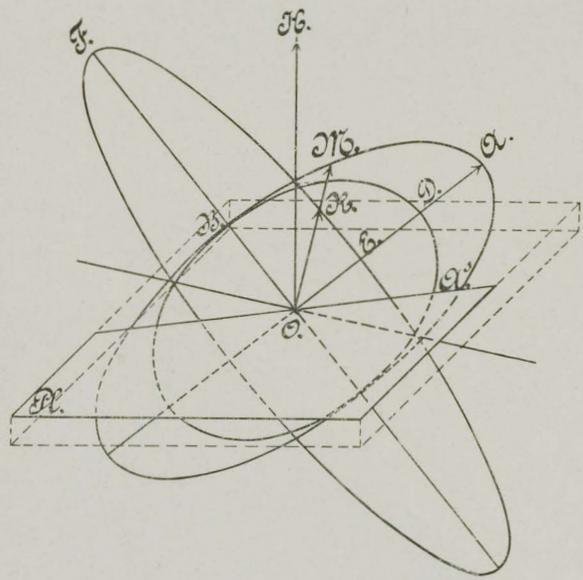


Fig. II.

