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Physics. — "The effect of temperature and transverse magnetisation on the resistance of graphite." By David E. Roberts. (Communicated by Prof. H. E. J. G. Du Bois).

The investigations of Grunmach and Weidert 1), Patterson 2) and others on the effect of transverse magnetisation on electrical resistance show that paramagnetic and diamagnetic metals exhibit an increase of resistance when magnetised, while the three ferromagnetic metals, at least in sufficiently strong transverse fields, show a decrease. Although as yet no simple relation may be given between the order of magnitude of this effect and the corresponding magnetic susceptibility, it may be noticed that the effect increases in the ratio of one to a hundred as we pass from paramagnetic tantalium to diamagnetic cadmium and suddenly again a thousandfold as we pass on to bismuth. This element, as is well known, possesses rather a high diamagnetic specific susceptibility (-1,40.10<sup>-6</sup>). Soon after Morris OWEN 3) found Ceylon graphite to show the highest value yet observed, Dr. W. J. DE HAAS was led, by analogy, to anticipate that graphite might exhibit a variation of resistance of an even higher order when magnetised and suggested to me to search for the effect. The preliminary experiments 1) performed with powdered graphite pressed into a thin plate, with irregularly shaped pieces and with ordinary pencils amply satisfied expectation and justified an extended investigation of the phenomenon.

Well defined crystals of graphite are exceedingly rare and could not be procured; the ordinary material occurs in lamellar agglomerations, cleavable with great ease along surfaces parallel to the base of the hexagonal system. From a chemical point of view the structure is possibly very complicated; graphite is generally considered, above 372°, the most stable of the three allotropic carbon modifications.

The conductivity for heat of this substance has lately been studied by Koenigsberger and Weiss<sup>5</sup>). The resistivity as formerly determined by several observers<sup>6</sup>) is as follows:

<sup>1)</sup> L. GRUNMACH and F. WEIDERT, Ann. der Phys. 22 p. 141, 1907.

<sup>2)</sup> J. Patterson, Phil. Mag. (6) 3 p. 643, 1902.

<sup>&</sup>lt;sup>3</sup>) Morris Owen, Versl. Afd. Natuurk. **20** p. 673, 1911. Ann. der Physik. **37** p. 657, 1912.

<sup>4)</sup> When magnetised transversely in a field of 20 kilogauss, the compressed powdered Ceylon graphite gave an increase in resistance of  $52\,^{0}/_{0}$ ; an irregularly shaped piece gave  $219\,^{0}/_{0}$ ; HB and 5B pencils by A. W. Faber gave only  $3^{0}/_{0}$  increase.

<sup>&</sup>lt;sup>5</sup>) J. Koeningsberger and J. Weiss, Ann. der Physik. 35 p. 27, 1911. Verh. d. Deutsch. physik. Ges. 14 p. 9, 1912.

<sup>6)</sup> See Handb. der Anorg. Chemie 8 (2 Abtl.) p. 54, 1909.

The best of my samples gave a resistivity as small as 0,5.10<sup>-4</sup>, i.e. roughly about half that of mercury (0,96.10<sup>-4</sup> at 18°): this increased with rise of temperature by about 0,001 per degree. The resistivity of amorphous carbon has always been found to be much larger and is well-known to decrease with rise of temperature; the coefficient diminishes, however, as the transformation into the graphitic modification proceeds¹), although it has never been observed to change its sign.

With regard to the effect of magnetisation Patterson 1. c. found the resistance of a glow-lamp filament to increase by 0,027 percent in a transverse field of 25 kilogauss. According to Clay?) the resistivity of such a filament decreases by 24 °/0 on heating from — 255° to 0°. Laws 3) has investigated the effect for transverse magnetisation of glow-lamp filaments, pencils and graphite without finding it to be of a high order. He found, at ordinary temperatures, the increase of resistance of the graphite in a field of 11 kilogauss to be about 1 °/0 of the resistance when outside the field, while at the temperature of liquid air the effect was increased threefold. Within this small range the increase of resistance was found proportional to the square of the field and between the temperatures 18° and —186° inversely proportional to the absolute temperature. As will be seen these results are not in agreement with those found in the present research.

## EXPERIMENTAL ARRANGEMENT.

The specimens most used in this investigation were prepared from the same Ceylon graphite as that used by Owen in his researches on its thermo-magnetic properties; a chemical analysis has not yet been made. Short rectangular pieces (7—10 mm. long, 1—2 mm. wide and 0,1—0,5 mm. thick) were obtained by careful cleavage and those selected for investigation which appeared of most pronounced and uniform crystalline structure. For the determination of the effect of transverse magnetisation they were, in general, supported in the magnetic field so that the cleavage planes were perpendicular to the field i. e. the crystallic axis was parallel to the lines of force. On supporting the pieces freely in a magnetic field it was observed

<sup>1)</sup> See Handb der Physik 4 p. 380, 1905. G. Wiedemann, Elektrizität, 1 p. 539, 1882.

<sup>2)</sup> J. CLAY. Dissert., Leiden 1908.

<sup>3)</sup> S. C. Laws, Phil. Mag. (6) 19, p. 694, 1910; his graphite was obtained from the Morgan Crucible Co., London.

that they moved so that the crystallic axis set itself perpendicular to the field, this axis thus coinciding with the direction of maximum diamagnetic specific susceptibility, which according to Owen may reach -- 15 millionths.

The magnetic field of the latest large type model of the DU Bois half-ring electromagnet was used. To obtain the higher fields at ordinary temperatures special prism-shaped pole end-pieces were used — 13 mm. long and 1,2 mm. wide. — With these end-pieces (0,7 mm. apart) and a pair of extra polar coils a field of 50 kilogauss could be easily attained. For observations at low and high temperatures the same arrangement was used as that adopted by DU Bois and Wills in conjunction with the large type electromagnet 1). The magnetic fields were measured by means of an exploring coil and a ballistic galvanometer 2) in the usual way. It was assumed provisionally that the fields were appreciably the same at all the temperatures used for a given current through the electromagnet.

The resistance of the graphite specimens, both in and out of the field, was determined by a potentiometer method ), being compared directly with known resistances (0,1-1,0 ohm). The current through the graphite during a series of measurements was varied between 2 and 0.5 milliamperes according to its resistance. In order to eliminate thermo-electric junction effects the current in the main circuit as well as the potentiometer connections were successively reversed. The changes of resistance involved being considerable it was found necessary to adjust the sensitiveness of the potentiometer arrangement during a single series of readings; this was initially sufficient to detect differences of 1/10000 ohm. Small irregular variations in the resistance of a particular specimen were observed after it was subjected to the action of magnetic fields or to widely different temperatures. This change, however, amounted in general to less than 1°/<sub>a</sub>. Through the kindness of Dr. Hoffmann the resistance of specimen G. 15 — that used in the experiments at different temperatures — was re-determined at 18° in the Phys. Techn. Reichsanstalt by means of Diesselhorst's "compensation apparatus" ) and a differential galvanometer; good agreement was found. Some of the preliminary measurements had been made with Wheatstone's bridge method and, when repeated potentiometrically, practically the same results were obtained.

<sup>1)</sup> H. pv Bois. Ztschr., für Instr.kunde 31, p. 362, 1911.

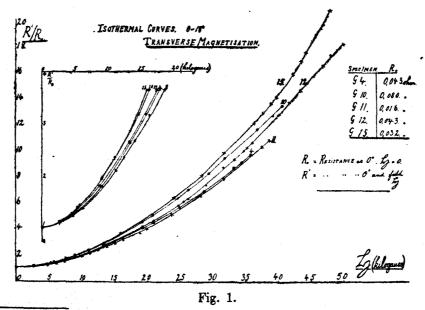
<sup>2)</sup> H. DU Bois. The magnetic circuit in theory and practice, p. 300, London 1896.

<sup>3)</sup> F. Kohlrausch, Prakt. Physik. 11 Auflage p. 422, 1910.

H. Diesselhorst, Zeitschr. für Instr.kunde. 26 pp. 173, 297, 1906; 28 pp. 1, 38, 1908.

## Experiments at ordinary temperature [18°].

About twenty specimens of Ceylon graphite were investigated, which all gave variations of resistance of a high order, the increase of resistance in a field of 20 kilogauss varying however between 300 and 500% of the resistance in zero field. Considering the difficulty of obtaining specimens of graphite of definite crystalline structure and having regard to the impurities occurring in the natural substance these variations in the magnitude of the effect are not surprizing. About five specimens, which gave a variation of resistance of greatest order were investigated more particularly; by analogy with the well-known behaviour of more or less pure bismuth 1) the assumption appeared justifiable that these were more likely to be pure and perhaps of more uniform crystalline structure. Some of the specimens were supported free between thin mica or glass plates; when imbedded in sodium silicate, collodion or Canada balsam allowed afterwards to solidify they did not experience any change in the magnitude of their increase of resistance in the magnetic field, thus eliminating any doubts that the effects were due to bodily strains in the graphite. In the final experiments at different temperatures the graphite pieces were supported by fine flexible wires between thin mica plates so as to avoid any strain due to possible expansion or contraction. The specimens could be mounted with their connections so that the total thickness amounted to less than 0.7 mm, thus enabling them to be examined in fields up to 50 kilogauss. Some of



the isothermal curves obtained for different specimens, at 18°, with the cleavage plane normal to the field, are shown in Fig. 1.

Attempts to identify the curves with such equations as

$$\frac{R'}{R_o} = \frac{R}{R_o} + b \mathfrak{I} + c \mathfrak{I}^2 + \dots$$

failed; it was found however that all the curves obtained at ordinary temperatures could, well within experimental errors, be represented by the formula

where  $R_{\rm o}$ , resistance at 0° for  $\mathfrak{H}=0$ ,

R, ,  $\theta^{\circ}$  for  $\hat{\mathfrak{D}}=0$ ,

R', ,,  $\theta^{\circ}$  in transverse field  $\phi$ .

A,n Constants.

Owing to the difficulty of determining the dimensions of the specimens it is unfortunately impossible to give their absolute resistivity with any exactitude.

From equation (1) we have, taking logarithms

$$\log \frac{R'-R}{R_0} = \log A + n \log \mathfrak{H}$$

which can be represented by a straight line, the coordinates being  $log(R'-R) R_0$  and  $log \mathfrak{P}$ .

The values of  $log(R'-R)R_o$  and log f, corresponding to the curves shown in fig. 1, when plotted were found to lie on straight lines practically parallel to one another, indicating that n is the same constant for each of these specimens. In the case of specimen G. 15 — the one which gave an increase of the greatest order — equation (1) did not hold as well as for the other specimens although the mean value of n was the same for this as for the others.

TABLE 1.

Isothermals at 18°		
Specimen	R <sub>0</sub>	$R'/R_0 = R/R_0 + A_{n}n$
G. 4	0.0430 ohm.	1.01 + 0.0171 pj1.745
G. 10	0.0792 "	1.01 + 0.0205 p1.745
G. 11	0.0162 "	1.004 + 0.0162 101.746
G. 12	0.0430 "	1.014 + 0.0188 p 1.746
G. 15	0.0316 "	1.02 + 0.0214 m 1.748

The values of A and n obtained for the different specimens are given in Table 1. For each specimen  $n = 1.74^{5}$ .

A specimen was also prepared for investigation from another piece of Ceylon graphite out of the laboratory collection. This graphite very easily split up along its cleavage surfaces but pieces of uniform structure of suitable form were difficult to obtain. The best piece I could prepare gave an increase of resistance of only  $182^{\circ}/_{\circ}$  in a field of 20 kilogauss, the resistance out of the field being 0.0427 Ohm.

A piece of graphite from Himbuluwa (Ceylon), which was investigated, on the other hand, gave quite different results. The upper side of this graphite possessed a quite smooth and polished surface underneath which however it appeared to be of a fine granular structure. A thin piece of this upper layer was removed and the variation of its resistance found when transversely magnetised. An increase of resistance of  $220^{\circ}/_{\circ}$  was observed in a field of 20 Kgs, the resistance out of the field being 0.0786 ohm. A thin piece removed from the under side of the same material, and having a high natural polish on both of its cleavage surfaces gave the anomalous results. Its resistance outside the field was several hundred ohms and diminished very rapidly with increase of temperature. In a magnetic field however no change in its resistance could be observed, while rough experiments indicated that it was apparently paramagnetic; no test for the presence of ferroginous impurities was made.

Specimen G 12 was also tested with its cleavage plane parallel to a transverse field, the crystallic axis being therefore at right angles to the lines of force. In a field of 26 kgs the value  $R'/R_{\bullet}$  was found to be only 1,15 while for the usual position this ratio is rather more than 6. This evidently proves the necessity for very accurate adjustment of the angle between the crystallic and field axes  $^{2}$ ); an analogous question is known to arise in the behaviour of nickel and other ferromagnetic wires.

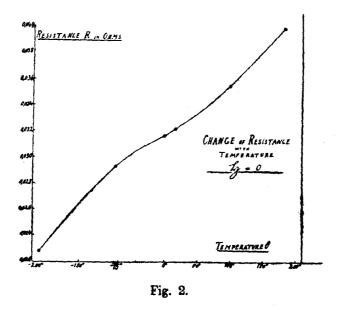
## EXPERIMENTS AT LOW AND HIGH TEMPERATURES.

Observations were taken at temperatures of  $-179^{\circ}$ ,  $0^{\circ}$ ,  $+18^{\circ}$ ,  $+95^{\circ}$  and  $+179^{\circ}$ , the field being varied from 0 to 40 kilogauss.

<sup>1)</sup> Within the experimental errors the exponent may also be  $n = \sqrt{3} = 1{,}732$  or n = 7/4.

<sup>2)</sup> The effect of longitudinal magnetisation was also observed. The increase of resistance involved was found to be independent of the direction of the current and of the same order as that observed in this last described position. Experiments are in progress to study the effect in both these cases at different temperatures.

The method of measurement was the same as at  $18^{\circ}$ ; the determinations afforded no difficulty, the resistances being quite steady. At the lower and higher temperatures thermo-electric effects were sometimes evident but by successive reversals these were eliminated. It was incidentally observed that these thermo-electric effects — when occurring at the connections of the graphite and therefore within the magnetic field — were also influenced by the field  $^{\circ}$ ). Thus in one case the thermo-electric effect was increased fourfold by a field of 38 kgs. For all the specimens examined (with the exception of the piece from Himbuluwa) the resistance of the graphite out of the field was found to increase with the temperature, the coefficient of increase of resistance being of the order 0.001 per degree. The ordinary temperature curve R = funct. ( $\theta$ ) for  $\mathfrak{H} = 0$  is given in Fig. 2 for G 15. Very

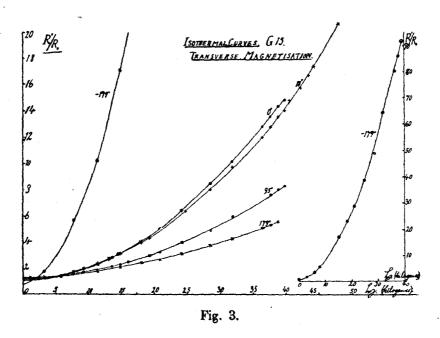


nearly the same type of curve was obtained in the case of specimen G 11. It is interesting to compare this with the curves obtained by KAMERLINGH ONNES and NERNST<sup>2</sup>). The temperature during a series of readings, the graphite being in the field, was determined as follows. Before commencing, the current required to be sent in the reverse direction through the magnet to reduce the residual field to zero, was determined. Then, to measure the temperature, the graphite

<sup>1)</sup> These effects are being subjected to further detailed investigation.

<sup>2)</sup> H. Kamerlingh Onnes, Versl. Afd. Nat. 19. p. 1187, 1911. W. Nernst, Sitz. Ber. Berl. Akad. p. 306, 1911.

being in position, this reverse current was set up and the resistance of the graphite found. The temperature of the graphite was then deduced from the temperature curve  $(\mathfrak{H}=0)$  fig. 2. Owing to the difficulty of exactly getting rid of the residual field without setting np a field in the opposite direction, and on account of the small change of resistance with temperature, this method of determining the temperature does not seem to be susceptible of great accuracy. The isothermals at low and high temperatures were determined for G 11 and G 15. Except for the difference in the magnitude of the changes of resistance concerned similar results were found. The results obtained with specimen G 15 are shown as isothermal curves (fig. 3) from which the so-called isopedic curves  $(\mathfrak{H}=\text{constant})$  may



easily be deduced. As will be seen, the increase of resistance is much greater at low temperatures. At the temperature of liquid air the increase is 9300 % for a field of 38,8 kgs, the increase at 18° being 1250 %.

The isothermal curves for the lower temperatures cannot be represented by an equation of the form (1); at higher temperatures this seems to be the case, although more accurate measurements appear desirable.

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