THE FARADAY EFFECT IN DIAMOND

BY S. RAMASESHAN

(From the Department of Physics, Indian Institute of Science, Bangalore)

1. INTRODUCTION

SINCE diamond is optically isotropic and has a large dispersive power, it may be expected to show an easily observable Faraday effect. A search of the literature however reveals only a single observation on the Faraday effect of diamond recorded by H. Becquerel (1877) in the following words, " In a remarkable crystal (octahedral) belonging to M. Fizeau, we obtained a feeble rotation of near about 27 minutes in yellow light." The thickness of the specimen was 2·875 mm. and the effective field was 7330 gauss. The Verdet's constant for diamond calculated from these data comes out as 0·0128 minutes per centimeter per gauss. This is actually less than that constant for common crown glass and, as we shall see in the course of the paper, it is only one-half of the correct value. The lack of any further observations with diamond in the century which has elapsed since Faraday's original discovery is evidently due to the difficulty of obtaining suitable specimens for study. As is wellknown, diamond commonly exhibits birefringence in greater or less measure, and the restoration of light due to this effect when the crystal is observed between crossed nicols completely overpowers the feeble restoration due to the action of the magnetic field. That Becquerel could at least observe the Faraday effect shows that the particular specimen was better than most samples. The low value obtained by him may be ascribed to the difficulty of measuring the Faraday effect in the presence of residual birefringence. Indeed, the Faraday rotation in isotropic bodies is known to be markedly diminished in the presence of birefringence induced by strain (Schutz, *Magneto Optik*, 1936).

However, contrary to the belief that has been frequently expressed in recent literature, birefringence is *not* always to be observed in diamond. The observations of Raman and Rendall (1944) on this point indicate that it is present only when there is an admixture of physically different allotropic modifications of diamond and is absent when there is no such admixture. In Sir C. V. Raman's collection of crystals and of cleavage plates, there are several specimens of diamond of which the birefringence is negligibly small.

Amongst these, the cleavage plates N.C. 157, N.C. 177 and N.C. 73 having approximately parallel faces were found on examination to be suitable for Faraday effect studies. The thickness of these plates were however rather small (0·58, 0·85 and 0·68 mm. respectively). Observations were also possible with a large diamond crystal in its natural form, N.C. 4, which gave an easily observable Faraday rotation on account of its considerable thickness (4·8 mm.). Owing, however, to the curvature of the faces of this crystal and the consequent deviation of the rays in passing through it, the magnitude of the Verdet's constant deduced from it is not as trustworthy as that deduced from the observations on the other three plates in spite of their comparatively smaller thickness.

Darwin and Watson (1927) have shown that the Faraday effect in most transparent bodies is accurately expressed by the modified Becquerel formula

$$V = \gamma \, \frac{e}{2mc^2} \, \lambda \frac{dn}{d\lambda}$$

where V is the Verdet's constant (rotation per cm. per gauss), e and m are the electronic charge and mass respectively, c is the velocity of light, λ the wavelength of the light used and n the refractive index of the substance. γ is a multiplying constant which may be called the *magneto-optic anamoly*. (γ is usually expressed as a percentage). As has been shown by Darwin and Watson, γ remains constant over the whole of the visible and near ultra-violet spectrum, provided the part of the dispersion associated with the infra-red absorption is eliminated from the dispersive power appearing in the formula. The value of γ for the great majority of carbon compounds lies between 40% and 60%. Accordingly in the present investigation, the Faraday rotation has been determined for different regions in the visible spectrum to determine the magneto-optic anamoly of diamond and to ascertain whether or not it is independent of the wavelength of the light employed.

2. The Experimental Procedure

The magnetic field.—A large electromagnet of the Rutherford type capable of giving fields upto 23,000 gauss was used for these investigations. Each of the two pole pieces supplied by the manufacturers had a longitudinal hole about 2 cms. in diameter. When the pole pieces are brought quite close, the large size of the holes would result in the field between them being low and also sensibly non-uniform. To eliminate this difficulty, specially designed soft iron plugs were introduced which reduced the diameter of the holes to 0·6 cm. The pole pieces after this modification, had a flat area of about 2 sq. cms. They were kept at a distance of 0·95 cm. apart. A

measurement of the field with a small search coil and fluxmeter over various regions in the space between the pole pieces showed that the variations in the field along the lines of force were not greater than 2 to 3 %. The magnitude of the field with 6 amps. current through the exciting coils was about 16,000 gauss.

The source of light.—An image of the tungsten bead of a powerful point-o-lite lamp was focussed on the slit of a constant deviation spectrograph, which was used as a monochromator. A slit of width less than one mm. selected out various regions of the spectrum. The spectral band coming through the slit was not more than 50 Å. in the green and blue regions and was about 100 Å. in the red and yellow regions. The mean wavelength of the band was determined with a calibrated direct vision spectroscope. For accurate monochromatisation a mercury point-o-lite or a sodium lamp was substituted for the tungsten lamp. Since a monochromator with a single prism was used, the scattered light gave a continuous background. This was not considerable when the brighter regions of the spectrum, *i.e.,* from 6000 Å. to 4800 Å. are used, but became a serious source of error for wavelengths below 4800 Å. The magnetic rotation of diamond was therefore studied only over a range of wavelengths between 6000 Å. and 4800 Å. where the background intensity did not affect the observations. On certain occasions the monochromator was dispensed with, and sodium or mercury lamps with adequate filters were used.

Two wide-angled nicols giving perfect extinction were used as polariser and analyser respectively. The rotation of the analysing nicol was measured with a lamp and scale, the scale being at 80 cms. from the mirror. The diamond plate was introduced between the pole pieces in a wooden holder and care was taken to see that the crystal was not strained thereby introducing birefringence. The surface of the diamond was kept normal to the incident light, as otherwise, there would be a rotation of the plane of polarisation due to refraction at oblique incidence. Although this scarcely affected the Faraday effect, it was more convenient to avoid such rotation.

The general procedure of the experiment was as follows. The light of the necessary wavelength was focussed on the slit and the diamond introduced in position and the nicols then accurately crossed. A large number of readings (usually 25) were taken for this crossed position. It was found that settings could be made within 1·5 mm. when the scale was at 80 cms. distance. The mean of these readings was taken as the position of extinction. The magnetic field was then put on by passing a current of 6 amps. through the coils and the nicols crossed again. The mean of 25 readings gave the

position of extinction. Similarly, the extinction position when the field was reversed was found. The rotation of the plane of polarisation was calculated by taking half the difference between the two extinction positions, the zero-reading acting as a check on the accuracy of measurement.

Half-shades and other appliances for finding out the exact extinction position could not be successfully used in the case of diamond. One seldom obtained complete extinction in all the regions of the diamond when it was observed between crossed nicols. To get the correct extinction position it was, therefore, necessary to view only those regions which gave total extinction. The half shade gave the mean extinction position for the whole plate of diamond and hence gave less accurate results than when ordinary nicols were used. In fact when a half-shade was used, rotations of the same order as those observed with nicols were obtained, but the differences between individual readings were larger and hence the method was untrustworthy. The numerous concordant values obtained when nicols were used bore testimony to the accuracy of this method.

The absolute value of the Verdet's constant of diamond was determined by comparing the rotation produced by it with that produced by a thin crystal of rock salt placed exactly in the same position as the diamond. This procedure was adopted so that if there was a minute rotation due to the effect of the magnetic field on the nicols, the cover slips protecting the nicols and other optical parts, the same could be eliminated. Separate measurements of these spurious effects were made and they were found to be less than 5% of the rotation produced by diamond. From the knowledge of the magnetic rotation due to a crystal of rock salt of known thickness and of the accurate Verdet's constant for it (from the International Critical Tables), it is possible to calculate the effective field. Table I gives the effective fields calculated from the rotation by substances of different thicknesses. The standard values of the Verdet's constant for these substances for the sodium 5890 line which were used for the calculation of the field are also given. The value obtained with a search coil (0·7 cm. long) is also given. These measurements were carried out for two different field strengths which were used in the determination of the magneto-optic rotation. The value of the field appears lower when the thickness of the substance used is small. This is, as already mentioned, due to the rotation produced (in the opposite direction) by the effect of the magnetic field on the optical system. At the foot of the table has been entered the ratio of the Verdet's constant for water for the wavelengths 5890 Å. and 5460 Å. observed experimentally by the present writer and that given in the International Critical Tables.

The two values agree quite closely, indicating the accuracy with which the settings could be made.

TABLE I

Verdet's constant at 25°C. for 5890A.

NaCl   ..   0.03585 mins. per cm. per gauss.

Water   ..   0.01306   ,,   ,,

A. Distance between pole pieces, 0.95 cms.

    Distance of scale from mirror, 79 cms.

Substance		Thickness in cm.	Wavelength in Å. U.	Rotation in cms.	Rotation in mins.	Field in gauss
NaCl	..	0·046	5890	1·24	27·54	16700
		0·098	,,	2·66	59·1	16820
		0·417	,,	11·4	254·3	17010
Water	..	0·105	,,	1·03	23·0	16720
		0·372	,,	3·70	82·6	17010

    Field determined with search coil 17100 gauss.

B. Distance between pole pieces, 1·05 cms.

    Distance of scale from mirror, 81·7 cms.

Substance		Thickness in cm.	Wavelength in Å. U.	Rotation in cms.	Rotation in mins.	Field in gauss
NaCl	..	0·043	5890	1·16	24·4	15810
		0·093	,,	2·57	54·0	15880
		0·401	,,	10·85	227·8	15890
Water	..	0·372	5890	3·37	70·8	15860
		0·372	5460	3·96	83·6	15880

$\dfrac{V_{5460}}{V_{5896}}$ for water at 25° 1·184 (I. C. T.)

     ,,   ,,   ,,   1·175 (Observed)

3. OBSERVATIONAL DATA

The cleavage plate N.C. 157 was used for the study of the variations of the Verdet's constant with wavelength. Table II gives the Verdet's constant obtained for different wavelengths between 6000 Å. and 4800 Å. The last column in the table gives the magneto-optic anamoly, the method of calculation of which is indicated later. Two monochromatic wavelengths 5890 Å. (sodium) and 5460 Å. (mercury) were used in the measurement of the Verdet's constant in N.C. 177. The values are given in Table III, each of them being a mean of 100 readings. The ratio $\dfrac{V_{5460}}{V_{5890}} = 1\cdot191$ and the ratio calculated from the Becquerel formula is 1·179. Table IV gives the results

obtained with N.C. 73 and N.C. 4 and also those obtained for a combination of N.C. 157 and N.C. 73.

TABLE II

N. C. 157. Thickness of the plate 0·58 mm.
Distance between pole pieces 0·95 cm. Effective field 16700 gauss.
Distance of scale from mirror, 79 cms.

Wavelength in Å. U.	Rotation in cms.	Rotation in mins.	Verdet's constant in mins per cm. per gauss	γ
5890	0·97	22·3	0·0230	27·4
5800	1·00	23·0	0·0237	28·1
5600	1·07	24·6	0·0254	27·9
5400	1·20	27·6	0·0285	28·9
5200	1·27	29·3	0·0302	28·2
5025	1·35	31·1	0·0321	27·8
4860	1·41	32·4	0·0335	26·9

TABLE III

N. C. 177. Thickness of the plate 0·85 mm. Effective field 15800 gauss.
Distance of the scale from the mirror, 81·7 cms.

Wavelength in Å. U.	Rotation in cms.	Rotation in mins.	Verdet's constant mins. per cm per gauss	γ
5890 (Na) ..	1·36	31·3	0·0233	27·6
5461 (Hg) ..	1·62	37·3	0·0278	27·9

$$\frac{V_{5460}}{V_{5890}} = 1\cdot191 \text{ (experimental)}$$
$$= 1\cdot179 \text{ (from Becquerel's formula)}$$

TABLE IV

Effective field 16700 *gauss.   Distance of scale from mirror,* 79 *cms.*

Diamond	Thickness in mms.	Wavelength in Å. U.	Rotation in cms.	Verdet's constant	γ
N. C. 73 ..	0·68	5800	1·20	0·0243	28·9
		5400	1·35	0·0273	27·5
		5025	1·54	0·0312	26·9
N. C. 73 and N. C. 157	1·26	5800	2·22	0·0243	28·9
		5400	2·67	0·0292	29·6
N. C. 4 (Crystal) ..	4·8	5800	7·44	0·0209	25·4
		5400	9·08	0·0256	25·6

As already remarked, the result obtained with the crystal N.C. 4 is not so trustworthy as that obtained with the cleavage plates.

4. The Magneto-Optic Constants of Diamond

The value of γ was determined in the following way, Peter (1923) determined the refractive index of diamond from 6790 Å. to 2265 Å. and found the refractive index in this region to be well expressed by the formula

$$n^2 - 1 = \frac{\epsilon_1 \lambda^2}{\lambda^2 - \lambda_1^2} + \frac{\epsilon_2 \lambda^2}{\lambda^2 - \lambda_2^2}$$

where

$$\epsilon_1 = 0 \cdot 3306 \qquad \lambda_1 = 1750 \text{ Å.}$$

$$\epsilon_2 = 4 \cdot 3356 \qquad \lambda_2 = 1060 \text{ Å.}$$

From this formula $\lambda \frac{dn}{d\lambda}$ was calculated for those wavelengths for which the magneto-optic rotation has been determined. Using the relation

$$V = \gamma \frac{e}{2mc^2} \lambda \frac{dn}{d\lambda}$$

γ was found and is expressed as a percentage. γ is thus the percentage ratio of the observed magnetic rotation to that calculated from the unmodified Becquerel formula. The last columns of each of the Tables II, III and IV give the value of γ for the wavelengths indicated. It may be noticed that for a range of wavelengths from 6000 Å. to 4800 Å., the value of γ is, within the limits of experimental error, constant.

From the examination of the above results, the following can be taken as the standard values of the various magneto-optic constants for diamond.

Verdet's constant 5890 Å .. $0 \cdot 0233$ mins. per cm. per gauss

5460 Å .. $0 \cdot 0278$,,   ,,

$$\frac{V_{5460}}{V_{5890}} = 1 \cdot 191 \text{ (observed)}$$
$$= 1 \cdot 179 \text{ (calculated from the Becquerel formula)}$$

The mean value of γ for the visible spectrum $= 27 \cdot 8$.

Fig. 1 gives the experimental values of the magnetic rotation for various wavelengths, as also the theoretical values, calculated from the Becquerel formula with $\gamma = 28\%$. It is estimated that in the measurement of the magnetic rotation, an error of not more than 4% could have crept in.

5. The Magneto-Optic Constants of Zinc Blende

Since sphalerite (ZnS) has a crystal structure similar to that of diamond it was considered worthwhile to determine its Verdet's constant and the magneto-optic anamoly. Using the same arrangement as that described for
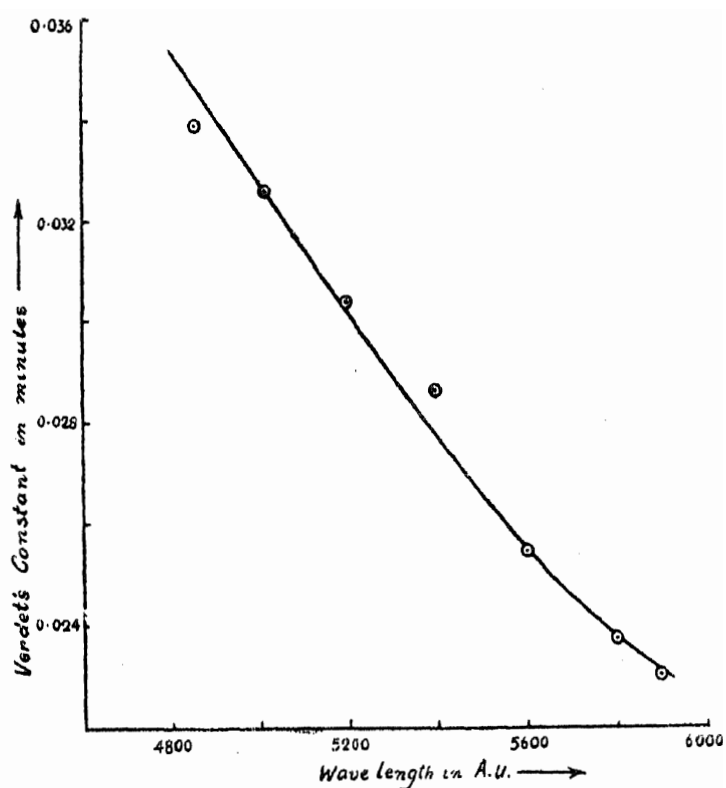
———— Theoretical curve from Becquerel's formula ($\gamma = 28\%$)
⊙ Experimental points (N.C. 157)

FIG. 1. Observed and Calculated Faraday Rotations of Diamond.

diamond, the magnetic rotation for a beautiful transparent plate of sphalerite in the possession of Sir C. V. Raman was determined for different wavelengths. Using the refractive index data of Maria Mell (1923) the magneto-optic anamoly for these wavelengths was determined. The results are tabulated in Table V. Table VI compares the results obtained by the present writer with those obtained by different authors. The agreement between these results is quite satisfactory.

TABLE V

Sphalerite (ZnS). Effective field, 16700 gauss. Thickness of the specimen, 5·55 mm.

Wavelength in Å. U.		Rotation in degrees	Verdet's Constant	γ %
6100	..	32·7	0·211	91
5890 (Na)	..	35·0	0·226	91
5780 (Hg)	..	38·1	0·246	90
5600	..	40·4	0·261	90
5460 (Hg)	..	44·5	0·287	92
5400	..	45·6	0·294	92
5200	..	52·4	0·333	91·5
4360 (Hg)	..	90·25	0·582	91

S. Ramaseshan

TABLE VI

Sphalerite (ZnS)

Constant	Author	Other observers
Verdet's constant		
5890 A	0·226	0·225 (Becquerel) 1877
5460 A	0·287	0·286 (Cotton) 1932
$\dfrac{V_{4360}}{V_{5460}}$	2·03	2·057 (,,)
$\dfrac{V_{5780}}{V_{5460}}$	0·852	0·863 (,,)

6. DISCUSSION OF RESULTS

Table VII gives the optical and the magneto-optic constants of some cubic crystals of relatively simple structure. The magneto-optic anamoly γ for all these crystals is constant over the range of the visible spectrum.

TABLE VII

Substance	Refractive Index 5890 Å	Dispersion 5890 Å	Verdet's constant 5890 Å	γ %
ZnS	2·3683	4611	0·226	91
NaCl	1·5443	669	0·03585	85
KCl	1·4904	586	0·02858	75
CaF$_2$	1·4338	242	0·00883	66
Diamond	2·4172	1298	0·0233	28

This indicates that the electrons which give rise to dispersion are also those responsible for the magneto-optic rotation. The value of γ for atoms and ions having the inert gas configuration has been shown (Van Vleck, 1932) to be 100%, *i.e.*, they obey the unmodified Becquerel formula. Hence we may legitimately infer that a deviation of γ from this value is an indication of the extent to which the electron configuration departs from the inert-gas state. The low value of γ in the case of diamond finds a natural explanation on this basis. The four electrons in the carbon atom are shared between the neighbouring atom and the electronic configuration is considerably different from that of the inert gas. Rock salt and sylvine are usually considered to be typical examples of crystals with ionic binding. In that case except for the slight perturbation produced by the crystal field, the sodium and chlorine ions in rock salt have the inert-gas structure and hence should have a value for γ approximating to 100%. Actually, as we see in Table VII the value of γ for these crystals is considerably less. This can only mean

that the alkali and halogen structures in these alkali halides deviate from the inert-gas configuration by a considerable extent. It seems possible that the binding between them may be at least partly covalent in nature. This is even clearer in the case of CaF_2. On this view, the fact that the value of γ for ZnS approaches that for an inert-gas configuration is significant. It indicates that the binding between Zn and S is much nearer being an electro-valent than of a covalent nature, in spite of the geometrical structure of the crystal being similar to that of diamond. Incidentally it may be remarked that in agreement with this view, ZnS is a strongly piezo-electric crystal.

In conclusion the author wishes to record his deep sense of gratitude to Professor Sir C. V. Raman, for his helpful interest and encouragement in this work.

7. SUMMARY

With three thin cleavage plates and a natural crystal of diamond, all of which had an almost negligible birefringence, the magnitude of the Faraday rotation was measured for a series of different wavelengths in the visible spectrum. The magnetic rotation was proportional to the dispersion and the Verdet's constant for the wavelengths 5890 Å. and 5460 Å. were 0·0233 and 0·0278 minutes per cm. per gauss respectively. The magnetic rotation followed the modified Becquerel formula with the multiplying factor γ, which remains sensibly constant in the visible region, equal to 28%. The value of this constant is much less than that for most carbon compounds available as liquids for which it is known. The magneto-optic constants for sphalerite (ZnS) were also determined and it was found that γ, in this case, was 91%. The significance of these results is discussed.

REFERENCES

Becquerel, H.	.. *Annales de Phys. et. Chem.*, 1877, **12**, 32.
Cotton, A., Dupuy, G., and Scherer, M.	*Comptes Rendus*, 1932, **194**, 405.
Darwin, C. G., and Watson	.. *Proc. Roy. Soc.*, London, (A), 1927, **114**, 474.
Maria Mell	.. *Zeit. für Phys.*, 1923, **16**, 244.
Peter, F.	.. *Ibid.*, 1923, **15**, 358.
Raman, C. V., and Rendall, G. R.	.. *Proc. Ind. Acad. Sci.*, 1944, **19A**, 265.
Schutz	.. *Handbuch der Experimental Physik*, 1936, **Band 16**, *Teil.*, 1, 49.
Van Vleck	.. *The Theory of Electric and Magnetic Susceptibility*, Oxford, 1932, 367.