THE ELECTRO-OPTICAL KERR EFFECT IN GASES

BY E. C. STEVENSON AND J. W. BEAMS

UNIVERSITY OF VIRGINIA

ABSTRACT

The electric double refraction or Kerr electro-optical effect in CO_2 has been investigated as a function of the density and of the temperature. A method of study has been developed by means of which it is possible to work at several hundred atmospheres pressure and with high electric field strengths. The polarizing prisms and the Kerr cell are enclosed in the same pressure chamber thus eliminating errors due to strains in the windows. The double refraction of the emergent light is measured by means of a photoelectric cell arrangement. The Kerr constant B in CO_2 was found to vary with density over the range 0.08 to 0.18 gm/cm^2 by about the amount to be expected from theory. The temperature variation, also, was in approximate accord with theory. Preliminary results show an easily measurable effect in oxygen and nitrogen, the former being larger.

THE phenomenon of electric double refraction was discovered by J. Kerr[1] in 1875 and is usually known as the Kerr electro-optical effect. In his early experiments Kerr was able to establish the relationship connecting the magnitude of the double refraction with that of the electric field strength. Briefly, the law states that if n_1 and n_2 are the refractive indices for the components of the light vibrating parallel and perpendicular to the lines of force in the substance, respectively, their phase difference D in radians after passing through the electric field E is

$$D = \frac{2\pi l(n_1 - n_2)}{\lambda} = 2\pi B l E^2 \tag{1}$$

where λ is the wave-length of the light, l is the length of the light path through the electric field, and B is called the Kerr constant. The latter, however, has been found to vary with different substances, wave-lengths, and temperatures.

The theory of the Kerr effect as developed by Langevin,[2] Born,[3] Raman and Krishnan,[4] and others[5] connects the Kerr constant B with the index of refraction, the dielectric constant, and the light scattering coefficient (depolarization constant); the last of which has been extensively studied by Rayleigh, Raman, and many others.[6]

The fundamental assumptions of this theory are that the molecules

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[1] Kerr, Phil. Mag. **1**, 337 (1875); **7**, 85 (1879); **9**, 157 (1880).

[2] Langevin, Le Radium **7**, 249 (1910).

[3] Born, Ann. d. Physik **35**, 117 (1918).

[4] Raman and Krishnan, Phil. Mag. **3**, 713 (1927); Proc. Roy. Soc. **A117**, 1 (1927).

[5] R. de L. Kronig, Zeits. f. Physik **45**, 458, 508 (1927); **47**, 702 (1927); See Born and Jordon, Elementare Quantenmechanik 259, 1930.

[6] Gans, Wien-Harms, Handbuch der Experimental Physik **19**, 338–408 (1928).

possess an intrinsic optical and electrical anisotropy and hence, as a consequence of the orientative influence of the electric field upon the induced and permanent electrical doublets in the molecules, the substance as a whole becomes doubly refracting. It is shown[7,4] that

$$B \equiv B_m N = \frac{(n^2 - 1)(n^2 + 2)}{4n\lambda} \left(\frac{\epsilon + 2}{3}\right)^2 \frac{\theta_1 + \theta_2}{\alpha_0} \tag{2}$$

where N is the number of molecules per unit volume, λ is the wave-length of the light, n is the index of refraction with zero field strength, ϵ is the dielectric constant, α_0 is the polarizability. B_m as defined by the above relation will be called the Kerr constant per molecule. Further

$$\theta_1 = \frac{1}{45kT}[(a_{11} - a_{22})(b_{11} - b_{22}) + (a_{22} - a_{33})(b_{22} - b_{33})$$

$$+ (a_{33} - a_{11})(b_{33} - b_{11})]$$

$$\theta_2 = \frac{1}{45k^2T^2}[(\mu_1^2 - \mu_2^2)(b_{11} - b_{22}) + (\mu_2^2 - \mu_3^2)(b_{22} - b_{33})$$

$$+ (\mu_3^2 - \mu_1^2)(b_{33} - b_{11})],$$

where T is the absolute temperature, k is Boltzmann's constant, b_{11}, b_{22}, b_{33} are the moments induced in a molecule along its three principal axes of optical anisotropy by unit electrical force in the light wave acting along the three axes respectively. a_{11}, a_{22}, a_{33} are the moments induced in the same directions respectively by unit electrostatic force arising from the applied field, while μ_1, μ_2, μ_3, are the components of the permanent electric moment along these same directions.

It is obvious that the above theory, as well as any other that might at the present time be proposed, should be expected to hold strictly only in the case of gases or vapors, because our insufficient knowledge of the liquid and solid states does not permit us to determine definitely all the essential factors necessarily involved. Although the Kerr effect has been extensively studied in liquids, the data for gases is comparatively meager. However, where the data for gases are available, the agreement between theory and experiment is as good as the precision of the experiment will justify.[4,8] Nevertheless, there is need for better experimental data, especially on the way B varies with different factors, in order to subject the theory to a rigid test. With this accomplished, the Kerr effect can be utilized as a powerful tool in studying not only the structure of the molecules themselves, but the influences of the molecules on each other which occur in gases under high pressure, and in the liquid and solid states. We have, therefore, undertaken a study of the effect in gases as a function of the density and temperature.[9]

[7] Debye Marx Handbuch der Radiologie, 754, 1925.

[8] Szwessy, Handbuch der Physik **21**, 724–955 (1929).

[9] Beams and Stevenson, Phys. Rev. **35**, 1440 (1930); Stevenson and Beams, J.O.S.A. **21**, 147 (1931).

The scarcity of experimental data in the case of gases results from the fact that the effect in gases and vapors is very small compared to that in liquids and solids, and hard to measure with accuracy. Previously the most precise methods of measurement consisted in plane polarizing the incident light at 45° to the lines of force by means of a Nicol prism and analyzing the emergent elliptically polarized light by means of a Brace half-shade arrangement. The "Kerr cell" containing the gas under investigation was enclosed in a vessel with glass ends placed between the Nicol and the half-shade instrument. With this apparatus it is possible to work only at pressures in the neighborhood of one atmosphere because with greater pressure the resulting strains produce enough double refraction in the transparent windows to render the Brace half-shade method unreliable. This necessity of working in the region of atmospheric pressure makes the effect exceedingly small for most substances, because of the limitations set by the small densities and the low sparking potentials.

In the present work we have succeeded in developing a new experimental method by which it is possible to study the Kerr effect up to pressures of several hundred atmospheres and at field strengths many times the sparking
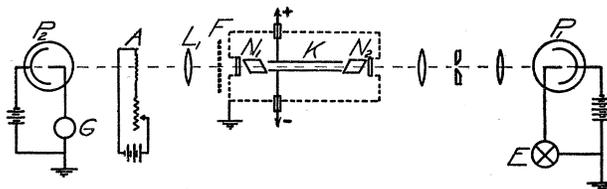

Fig. 1. Diagram of optical arrangement.

potential at atmospheric pressure. These conditions are accomplished by enclosing both polarizing and analyzing prisms together with the plates of the Kerr cell in a heavy pressure chamber with glass windows in the ends to permit the passage of light. The prisms under uniform pressure are not strained and no errors can be caused by strains in the windows. The increased pressure of the gas permits the use of very high field strengths because the sparking potential increases almost linearly with density according to Paschen's law.[10] This gives a very important advantage for, as can be seen from Eq. (1), the Kerr effect increases as the square of the field strength. The Brace half-shade method for measuring the double refraction was discarded because it not only required the measurement of small angles inside the tube but also depended upon the skill of the observer in matching light intensities. Instead, a photoelectric cell has been substituted which has given excellent results.

In Fig. 1 the arrangement of the optical parts is given. The light source A is a mercury arc or an incandescent lamp. A particular portion of the spectrum is selected by filters or a monochromator and the rays made parallel by the lens L_1. After being plane polarized by the polarizing prism N_1 they pass

[10] See J. J. Thompson, Conduction of Electricity through Gases, 2nd Edition p. 451, 1906.

between the oppositely charged parallel plates of the Kerr cell, through the analyzing prism N_2 and are finally brought to a focus on the surface of a vacuum photoelectric cell P_1. The latter is provided with a guard ring to prevent leakage and when used with a Swann electrometer E, was found to be linear for varying light intensities. The prism N_1 is set at a fixed angle with the plane of the plates such that the plane of vibration of the polarized light entering the cell is inclined at 45° to the direction of the lines of electric force between the plates. The prism N_2 may be oriented at any desired angle.

With N_1 and N_2 crossed and no field between the plates, no light enters the photoelectric cell and the rate of charge of the electrometer is zero except for an occasional small electrical or light leak for which a correction is made. When a potential is applied to the Kerr cell, the gas between the plates assumes the properties of a uniaxial crystal with its optic axis parallel to the lines of force. The light striking N_2 is therefore elliptically polarized and a small portion of the initial total intensity will pass on to the photoelectric cell. For particular settings of N_2 and the corresponding light intensities measured by the electrometer, the amount of double refraction in the Kerr cell can
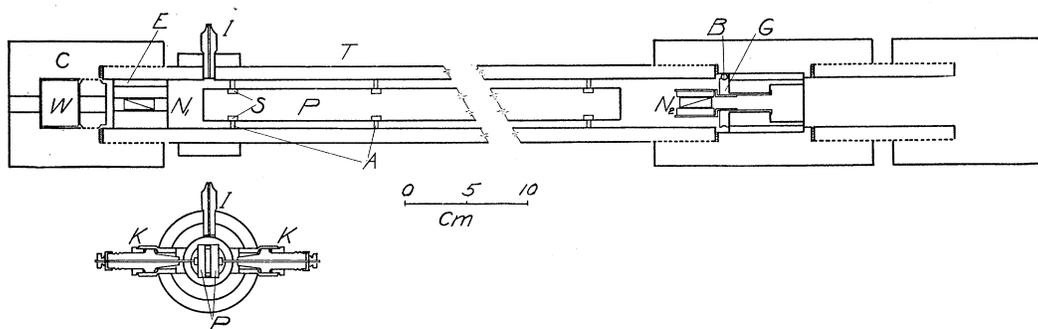

Fig. 2. Cross-sections of pressure chamber.

be calculated. Obviously, the intensity of the light source must remain constant, and to insure this, a second photoelectric cell P_2 and galvanometer are used to detect and correct for any fluctuations in A. The pressure chamber is indicated by the dotted enclosure.

In Fig. 2 are shown sections of the pressure tube. The barrel is a cold drawn steel tube of 3.8 cm inside diameter and walls 1.25 cm thick. The heavy caps on the ends contain plate glass windows 3 cm thick and 3 cm in diameter. The hole allowing passage of the light is 1.25 cm in diameter. The polarizing prism N_1 is mounted in the front end of the tube and the plates P are held in place by bakelite rings A. They are also insulated from each other by bakelite separators S. The plates used throughout this work were Starrett steel straight-edges 90 cm in length, 2.5 cm wide and 4 mm apart. The intermediate section F contains a worm and gear mounting BG for the analyzing prism N_2 by which it can be turned from the outside of the tube. Lead washers at all joints provide gas tight seals. Electrical connections to the plates are made by the use of ordinary spark plugs KK and the gas inlet is at I. Where the

gas inlet and spark plugs enter, the tube is reinforced by a steel ring shrunk on. Gases are admitted to the tube from commercial steel bottles through copper pressure tubing, and the pressures are measured on a special spring gauge with telescope mirror and scale attachment. This was calibrated by a dead-weight piston gauge. The densities were then calculated from the data of Amagat.[11]

Fig. 3 is a diagram of the method for obtaining and measuring the electric potential applied to the plates. One side of the secondary of a five-kilowatt x-ray transformer is grounded, while the other goes to two kenotrons in parallel. These charge the opposite sides of the condenser C through the resistances RR. The two sides of the condenser are connected to the plates of the
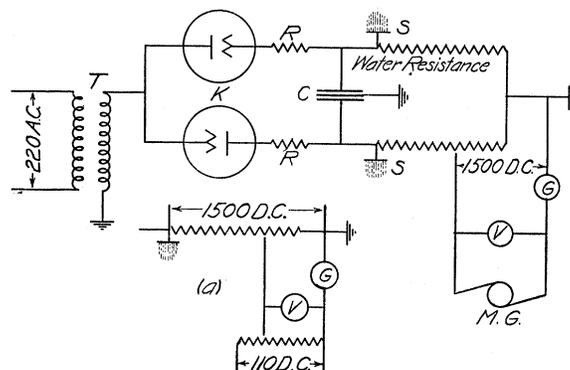

Fig. 3. Diagram of electrical arrangement.

Kerr cell and to the ends of a long continuous-flow water resistance. The water enters at ground potential, divides into two branches and is expelled through sprinklers at the end of each half. The use of sprinklers to break the stream obviates the need of a return path and thus reduces electrical leakage from the condenser. The total potentials are determined by a potentiometer method.[12] Voltage differences across segments of each half of the water resistances are balanced against a 1500 volt motor-generator set, the voltmeter V recording voltages directly and the galvanometer G being used only as a null instrument. Voltage readings across the segments are multiplied by the proper factors to give the total voltage across each half of the resistance. The multiplying factors are obtained by calibration in which the 1500 volt generator is connected across the half resistance and the voltage across the segments balanced against a 110 volt line source or batteries as shown in Fig. 3 (a). The potentials can be measured to 0.2 percent while the ripple was less than this. The temperature of the gas is controlled by immersing the whole tube in a water or oil bath regulated by a mercury in glass thermostat to less than 0.02 of a degree centigrade.

In order to determine D in Eq. (1) with the above apparatus it is only necessary to recall that the substance between the plates of the Kerr cell

[11] Amagat, Comptes Rendus, 663, 1885.
[12] Gunn, J.O.S.A. and R.S.I. **14**, 257 (1927).

takes the optical properties of a uniaxial crystal with the optic axis in the direction of the lines of electric force in the cell. Hence the intensity of light I passing the second or analyzing Nicol N_2 is[13]

$$I = [\cos^2 (\alpha - \beta) - \sin 2\alpha \sin 2\beta \sin^2 (D/2)] \tag{3}$$

where α and β are the angles that the planes of vibration of the transmited light of N_1 and N_2 make respectively with the lines of force. In the case where N_2 is crossed with respect to N_1 and the plane of transmission of N_1 makes an angle of $45°$ with the lines of force,

$$I_1 = I_0 \sin^2 \frac{D}{2} \cdot \tag{4}$$

When the Nicols are parallel, $\alpha = \beta = 45°$, and

$$I_2 = I_0 \left(1 - \sin^2 \frac{D}{2} \right). \tag{5}$$

Hence

$$\frac{I_1}{I_2} = \tan^2 \frac{D}{2} = \tan^2 \pi N B_m l E^2 \tag{6}$$

or

$$N B_m = \frac{1}{\pi l E^2} \tan^{-1} \left(\frac{I_1}{I_2} \right)^{1/2}. \tag{7}$$

Thus if the light intensities are measured by the above photoelectric cell method for conditions of crossed and uncrossed prisms I_0 is eliminated and $N B_m$ can be determined. The accuracy attainable in measuring D by this method is as good as is needed at the present time for it is better than that with which it is possible to measure the other factors entering into Kerr's law. The principal difficulty in this measurement arises from the fact that I_1 is usually very much smaller than I_2 making it necessary to measure photoelectric currents of widely different orders of magnitude.

In this work, it was especially desired to study the variations of the Kerr constant per molecule, B_m, with density and with temperature, so that greater precision was obtained by holding as many of the factors as possible in Eq. (1) constant rather than determining their absolute magnitudes. Rewriting Eq. (4)

$$\sin^{-1} \left(\frac{I_1}{I_0} \right)^{1/2} = \pi B_n N l E^2 = \left(\frac{I_1}{I_0} \right)^{1/2} \text{(approximately)}. \tag{8}$$

(The error due to the above approximation is less than one part per thousand because of the smallness of D). If now T, I_0, l, λ and E are held constant and $\sin^{-1}(I/I_0)^{1/2}$ is plotted as a function of the density a straight line should result, provided B_m is independent of the density. Fig. 4 shows an example of

[13] See Wood, Physical Optics 320, 1923; Robertson, Physical Optics 290, 1929.

such a graph for a density variation from 0.08 to 0.18 gm/cm³. It will be observed that over this range the curve within the limits of experimental error is a straight line. However, the line should pass through the origin if B_m is completely independent of the density, but the experimental values show clearly that this is not the case. B_m must, therefore, vary with density. If the curve were extended either to larger or smaller densities it would no doubt
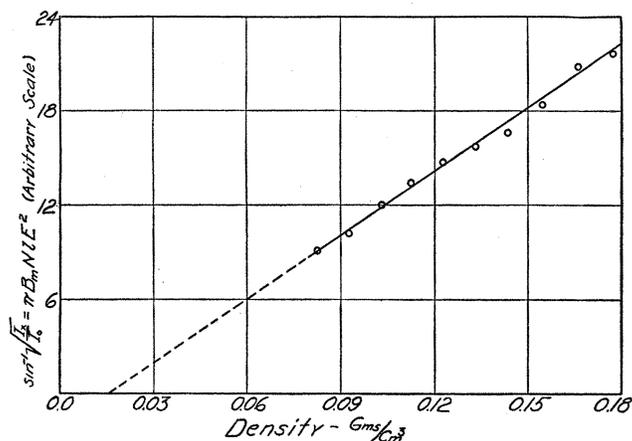
Fig. 4. Variation of the Kerr effect with density, temperature 34.3°C, λ4300–4700 A.

deviate from a straight line. It is interesting to compare this result with what should be expected from theory and the known behavior of the index of refraction and the dielectric constant.

Since CO_2 is non-polar θ_2 in Eq. (2) is zero and if we also substitute for α_0 the value $\alpha_0 = 3/4\pi N \cdot n^2 - 1/n^2 + 2$

$$B = B_m N \propto \frac{(n^2 + 2)^2(\epsilon + 2)^2}{n\lambda T}. \tag{9}$$

From the data of Phillips[14] for the index of refraction of CO_2 and that of Keyes and Kirkwood[15] for the dielectric constant of CO_2 the values for n and ϵ can be obtained and the variation of $B_m N$ with density calculated. Fig. 5 shows a graph of $(n^2+2)^2 \cdot (\epsilon+2)^2/n\lambda T$ plotted against the density over the same interval as our experimental values in Fig. 4. It will be observed that the calculated values also lie on approximately a straight line, which, however, does not pass through the origin, but cuts the density axis at approximately the same value as the experimental curve in Fig. 4. In fact, the agreement between theory and experiment is as good as should be expected considering that there is some choice in the way the lines should be drawn. The index of refraction and dielectric constant data indicate that if we were to extend our measurements to higher densities the variation of B_m with density would be found quite marked. The fact that the relation expressed in Eq. (1) is not ap-

[14] Phillips, Proc. Roy. Soc. A97, 226 (1920).
[15] Keyes and Kirkwood, Phys. Rev. 26, 754 (1930).

preciably changed by a variation in density indicates that the same causes which produce a variation in ϵ and in n with increasing ρ also produce the corresponding changes in B_m. However, as far as the writers know a satisfactory hypothesis concerning the changes in the structure of the molecule itself or its association with other molecules, resulting from increased density, has not been proposed that will quantatively explain all the experimental results. It will perhaps be necessary to extend the measurements to greater densities and to other gases before a clear understanding can be obtained.
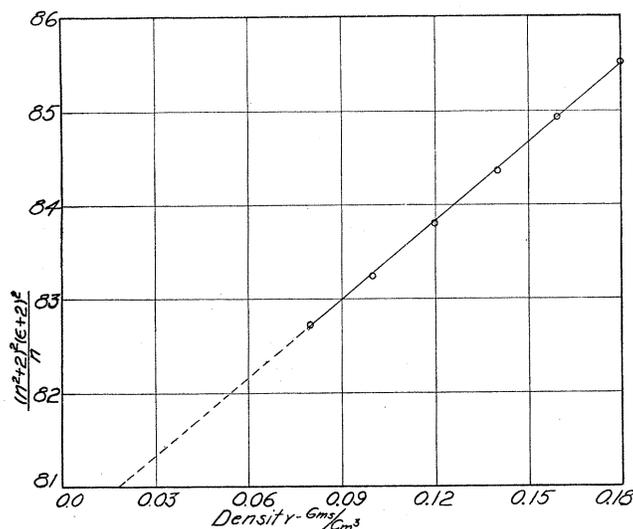

Fig. 5. Calculated variation of the Kerr effect with density from the data of
Phillips, for n, and Keyes and Kirkwood, for ϵ.

Referring again to Eq. (9) it will be noticed that B should vary inversely as the absolute temperature holding the remaining factors constant. We have made preliminary measurements of the changes of B with temperature from 20° to 45°C and find that the results are in approximate agreement with theory.

We have also looked for the Kerr effect in oxygen and nitrogen and found it large enough to be measurable by the method. The effect is larger in oxygen than in nitrogen but we did not obtain satisfactory absolute values because of a fire with oxygen that terminated the experiment before it was complete. In hydrogen our preliminary results indicate that the effect is much smaller than in nitrogen.