The University of Chicago
FOUNDED BY JOHN D. ROCKEFELLER

ON THE RELATION BETWEEN DENSITY AND INDEX OF REFRACTION OF AIR

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BY
HENRY G. GALE

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IF a body is compressed or its temperature varied, its density, d, will change and so also will its index of refraction, n. The problem of finding the relation between the quantities d and n is an old one and one attended by some difficulties. In the case of solids and liquids the density can be made to vary only slightly and experiments cannot be conducted over wide limits. In the case of gases the density can be given a wide range, but the index of refraction differs but little from unity, and its variation with changes of density is small.

Gladstone and Dale[1] first proposed the very simple relation,

$$\frac{n-1}{d} = \text{const.} \tag{1}$$

This is the relation which would be expected if transparent substances owe their refractive power to their molecules alone, and if the molecular index of refraction does not change with the density.

Jamin[2] proposed the formula,

$$\frac{n^2-1}{d} = \text{const.} \tag{2}$$

a relation which follows necessarily on the basis of the emission

[1] Gladstone and Dale, Phil. Trans., p. 887, 1858; Phil. Trans., p. 317, 1863.

[2] Jamin, Ann. de Chim. et de Phys., 3e série, T. LII., p. 163, 1858; T. LXI., p. 358, 1861.

theory. For gases this relation is almost the same as that of Gladstone and Dale, since

$$\frac{n^2-1}{d}=\frac{(n-1)(n+1)}{d}\doteq\frac{2(n-1)}{d}$$

$n+1$ being very little different from 2.

Lorentz,[1] on the basis of the electro-magnetic theory of light, has proposed the following formula,

$$\text{(3)} \qquad \frac{n^2-1}{(n^2+2)d}=\text{const.}$$

which comes at once on substituting n for D in Clausius' equation $\frac{D+2}{D-1}d=\text{const.}$, where D is the dielectric constant. It is seen that the relation (3) amounts to the same thing as relation (1) in the case of gases since

$$\frac{n^2-1}{(n^2+2)d}=\frac{(n-1)(n+1)}{(n^2+2)d}\doteq\frac{2}{3}\frac{(n-1)}{d}$$

because $n+1$ is very nearly equal to 2, and n^2 differs but very little from unity.

Jamin[2] was among the first to conduct experiments designed to test these relations. He measured the index of refraction of water at different pressures and found $\frac{n^2-1}{d}$ very nearly constant, but Mascart[2] showed that Jamin's results satisfied still better the equation of Gladstone and Dale.

Quincke[3] verified the results of Mascart for water and found that for glycerine, Rüböl, carbon disulphide, alcohol and ether, equation (2) always gave too large computed values and equation (3) too small values, while the values computed from equation (1) were nearly correct, being sometines a little too large and sometimes too small.

[1] Lorentz, Wied. Ann., Bd. IX., S. 641.

[2] Mascart, Comptes Rendus, LXXVIII., p. 801; Pogg. Ann., Bd. CLIII., S. 154.

[3] Quincke, Wied. Ann., Bd. XIX., S. 401; Bd. XLIV., S. 774.

Röntgen and Zehnder[1] obtained results in agreement with Quincke's for water, but they found that for the other substances equation (1) gave results consistently a little too large, but nearer than equations (2) and (3).

Quincke attempted to measure the change in the index of refraction of solids with a change of pressure, but the variations he was able to produce were too small to be measured accurately.

Rudberg,[2] Fizeau,[3] Stefan,[4] F. Vogel,[5] Pulfrich,[6] Offret[7] and others have investigated the change in the index of refraction of many crystals and transparent bodies as the temperature was varied over wide limits.

Jamin,[8] Damien,[9] Pulfrich,[10] Rühlmann,[11] Wüllner,[12] B. Walter,[13] Ketteler[14] and others have found that the maximum index of refraction of water occurs at a temperature somewhat below that of maximum density, and have investigated the relation of n and d for water from temperatures below 0° C. to 100° C. They found that the quotient $\frac{n-1}{d}$ diminished slightly as the temperature was increased. The observations of Ketteler and Pulfrich for water give, for sodium light, the following values:

	$\frac{n-1}{d}$	$\frac{n^2-1}{d}$	$\frac{n^2-1}{(n^2+2)} \cdot \frac{1}{d}$
At — 10°	.33447	.78061	.20656
At 10°	.33363	.77922	.20618
At 56.8°	.33274	.77464	.20585
At 95.2°	.33189	.76998	.20583

[1] Zehnder, Wied. Ann., Bd. XXXIV., S. 91; Röntgen und Zehnder, Wied. Ann., Bd. XLIV., S. 1 und S. 24.

[2] Rudberg, Pogg. Ann., Bd., XXVI., S. 291.

[3] Fizeau, Ann. de Chim. et de Phys., 3e série, t. LXVI., p. 429; 4e série, t. II., p. 143; Pogg. Ann., Bd. CXIX., S. 87 u. 297.

[4] Stefan, Berichter der Wiener Akademie (2), LXIII., S. 223.

[5] F. Vogel, Wied. Ann., Bd. XXV., S. 87.

[6] Pulfrich, Wied. Ann., Bd. XLV., S. 609.

[7] Offret, Bulletin de la soc. franç. de min., t. XIII., p. 405, 1890.

[8] Jamin, Comptes Rendus, t. XLIII., p. 1191; Pogg. Ann., Bd. C., S. 478, 1857.

[9] Damien, Ann. de l'école norm., sup., 2e série, t. X., p. 257, 1881.

[10] Pulfrich, Wied. Ann., Bd. XXXIV., S. 326, 1888.

[11] Rühlmann, Pogg. Ann., Bd. CXXXII., S. 1 u. 177.

[12] Wüllner, Pogg. Ann., Bd. CXXXIII., S. 1.

[13] B. Walter, Wied. Ann., Bd. XLVI., S. 423.

[14] Ketteler, Wied. Ann., Bd. XXXIII., S. 353 u. 506.

When instead of n the constant A of Cauchy's formula, $n = A + \frac{B}{\lambda^2}$ was used, the following relations were obtained:

	$\frac{A-1}{d}$	$\frac{A^2-1}{d}$	$\frac{A^2-1}{A^2+2} \cdot \frac{1}{d}$
At 10°	.32516	.75867	.20185
At 20°	.32463	.75717	.20159
At 30°	.32445	.75644	.20154

Wüllner found that the best agreement was given by the formula $\frac{A-1}{d} = a + bt$, the constant b being negative for glycerine, alcohol and carbon disulphide, but positive for a saturated solution of zinc chloride in water. Knops,[1] Weegmann[2] and Dufet[3] obtained similar results.

Ketteler[4] proposed the formula $\frac{n^2-1}{n^2+x} \cdot \frac{1}{d} =$ const., and computed x from the observations of Knops and Weegmann who had extended their investigations to a great number of organic substances. Ketteler found values of x ranging from 2 to 8.4.

On varying the temperature of air, Mascart[5] found that the index of refraction decreased more rapidly than the density with increasing temperature, but von Lang[6] obtained an opposite result by measuring the index of refraction directly from air at a high temperature to air at ordinary temperatures.

Benoit[7] found that for air the diminution of the refraction was exactly proportional to the diminution of density when the temperature was raised, and Chappuis and Rivière[8] obtained the same result for cyanogen.

[1] Knops, Liebigs Ann., CCXLVIII., S. 175.

[2] Weegmann, Zeitschr. für physikal. Chemie, Bd. II., S. 218.

[3] Duet, Journal de Phys., 2e série, t. IV., pp. 389 et 477.

[4] Ketteler, Wied. Ann., Bd. XXX., S. 285; Bd. XXXIII., S. 353 u. 506; Bd. XXXV., S. 662.

[5] Mascart, Ann. Sc. de l'Ecole Norm. sup. (2), t. VI., p. 9, 1877; Comptes Rendus, t. LXXVIII., pp. 617, 679, 801, 1874; t. LXXXVI., pp. 321, 1182, 1878.

[6] v. Lang, Pogg. Ann., Bd. CLIII., S. 448, 1874.

[7] Benoit, Jour. de Phys. (2), t. VIII., p. 451, 1889.

[8] Chappuis et Rivière, Ann. de Chim. et de Phys. (6), t. XIV., p. 5, 1888.

Biot and Arago,[1] working between $-1.5°$ C. and $25°$ C., found that the "refractive power" $n^2 - 1$ was proportional to the density when d was computed by the formula, $d = \frac{d_0}{1 + at}$, a being the temperature coefficient of expansion. Their results, however, satisfy equally well the relation $\frac{n-1}{d} = \text{const.}$

Lorenz[2] and Prytz[3] found that the relation $\frac{n^2 - 1}{n^2 + 2} \cdot \frac{1}{d}$ held fairly well for many substances though the liquid into the gaseous state —markedly better, in fact, than the relation $\frac{n-1}{d}$ const. Some of their results are shown in the following tables:

	Liquid.		Gas.
	10°	20°	100°
Ether.	0.30264	0.30287	0.3068
Alcohol.	0.28042	0.28066	0.2825
Water.	0.20615	0.20608	0.2068
Chloroform.	0.17902	0.17909	0.1796

Mascart[4] conducted an extensive research on the effect of pressure on the index of refraction of air, using interference methods. Two tubes were filled with air under pressure, the one always a fixed amount higher than the other, and the relation between density and index of refraction was deduced from the number of fringes which passed as the air in the two tubes was brought to the same pressure. He found that the refraction $n - 1$ increased more rapidly than the pressure; the ratio increased about 1 per cent. when the pressure was increased to about 8 atmospheres. On taking into account variations of density in accordance with Regnault's formula, $d = AH(1 + BH)$, Mascart found that there were still discrepancies.

[1] Biot et Arago, Mem. de la prem. classe de l' Institut, t. VIII., p. 301, 1806; Mem. de l'Acad. des. Sciences, t. VII., p. 301, 1806; Gilberts Ann., Bd. XXV., S. 345, 1807; B. XXVI., S. 79, 1807.

[2] Lorenz, Wied. Ann., Bd. XI., S. 70, 1880.

[3] Prytz, Wied. Ann., Bd. XI., S. 104, 1880.

[4] Mascart, Ann. Sci. de l'Ecole Norm. sup. (2), t. VI., p. 9, 1877; Comptes Rendus, t. LXXVIII., pp. 617, 679, 801, 1874; t. LXXXVI., pp. 321, 1182, 1878.

which, however, he considered were within the limits of observational error.

Chappuis and Rivière[1] kept the pressure in one tube constan and measured the change dn in the index of refraction of the air in the other tube as its pressure was changed by an amount dH, Working with air and carbon dioxide up to twenty atmospheres they found that in every case the refractive power, $n - 1$, was proportional to the density.

Carnazzi,[2] however, working with a prism and a long beam of light, found that the ratio $\frac{n-1}{d}$ showed a decided increase in the case of air and hydrogen as the pressure was increased, but that the ratio decreased with increasing pressures in the case of carbon dioxide.

Since the work which had been done on the relation between density and index of refraction of air with a changing pressure did not appear to be entirely conclusive, an attempt has been made in the work described in this article to get some data on the subject. Interference methods naturally suggested themselves as the best for attacking the problem, but it was found advisable to modify the form of interferometer in common use. The pressure was measured in a simple way by applying the inverse principle of the McLeod gauge. The consideration of the investigation divides itself naturally into four parts: I. The pressure gauge, II. The optical arrangement, III. Observations, IV. Conclusions.

I. The Pressure Gauge.

The pressure gauge used is outlined in Fig. 1. AB is a steel rod about an inch in diameter, along the axis of which has been drilled a hole, C, about $\frac{1}{32}$ inch in diameter. D and E are steel stopcocks which were carefully ground with fine carborundum to fit as exactly as possible into tapering holes drilled through the rod AB. A piece of heavy copper pressure tubing of fine bore, screwed into one end of the rod and soldered in place, connected the pressure gauge to a large tank which contained air at the pressure to

[1] Chappuis et Rivière, Ann. de Chim. et de Phys. (6), t. XIV., p. 5, 1888.

[2] Carnazzi, Il Nuovo Cimento, 6, p. 385, 1897.

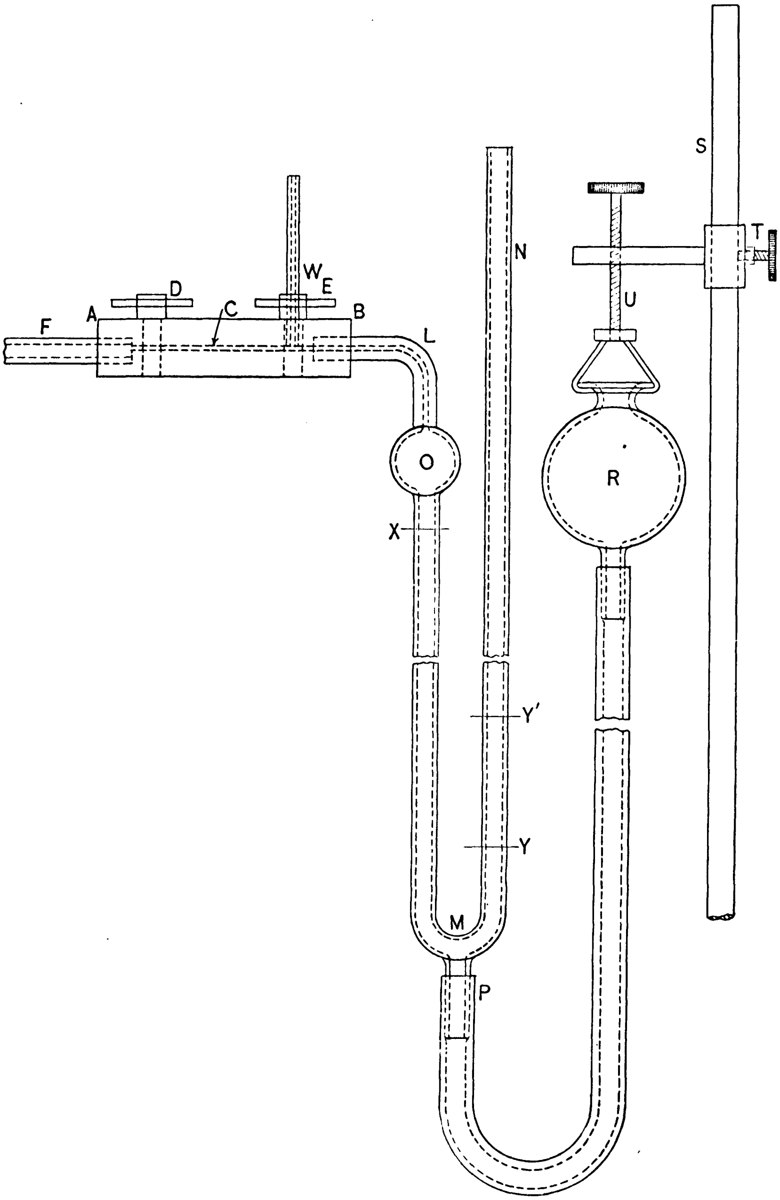

Fig. 1.

be measured. Into the other end of the rod *AB* was fastened one end of a glass tube *LMN* of the form shown in the figure. The bulb *O* had a capacity of about 10 c.c., and the tubes *OM* and *MN* had an internal diameter of about .5 cm. A bulb *R*, of about 500 c.c. capacity, for holding mercury, was connected with *P* by a piece of rubber tubing. The height of the mercury in the tubes *OM* and *MN* was regulated by sliding the clamp *T*, which held the bulb, up and down the rod *S*. A threaded rod *U*, which held the bulb, gave an opportunity for fine adjustment.

The stopcock *D* had a hole through it of small diameter in line with the hole *C*, through *AB*. The stopcock *E* was a three-way cock, having one hole through it in line with *C* and another half way through at right angles to the first. This stopcock was first set across so that the air could pass from *L* to the center of the stopcock and then out through the back of the rod *AB*, through a hole drilled to meet the hole *C*. A glass tube *W* was waxed into this hole to carry off the overflowing mercury.

As the bulb *R* was raised, mercury rose in the tube *NOL*, driving the air before it through the three-way cock *E*, and out into the tube at *W*. When the mercury began to rise in the tube at *W* the stopcock *E* was turned through 45°, thus closing all of its openings, and the bulb *R* was lowered until the mercury stood at some fixed point, as *X*, below the bulb *O*. The mercury then stood in the other arm of the tube at some point *Y*, almost a barometric column below *X*. A very high vacuum could not be expected in *O*, but by measuring with a cathetometer the difference of level between *X* and *Y* and subtracting this difference from the barometric height, the pressure of the gas left in *O* was found. The stopcock *D* was then opened and the small volume *C*, between the stopcocks, was allowed to fill with air at the pressure of the tank. Some little time was allowed for the pressure to adjust itself evenly, the stopcock *D* was then closed, and *E* was turned another 45°. This allowed the small volume of air in *C* at a high pressure to expand into the larger volume *O*, the mercury, of course, being forced down from *X*. The bulb *R* was then raised until the mercury came back exactly to its original position *X*, and a new reading was taken for the difference of level of the mercury in the two arms, the level in the second arm having been raised to some point *Y'*.

From these readings and the barometric height the pressure in the tank may be deduced in the following way: let V be the volume of the bulb and tube down to the point X, and call the pressure of the residual gas P. Let v' represent the volume of the space between the stopcocks, and p' the pressure of the gas in the tank. If the final pressure in the bulb, after the three-way cock has been opened and the mercury brought back to X, is represented by $\bar{P}$, the following equations will hold:

$$VP + v'p' = (V + v')\bar{P},$$

$$p' = \frac{V + v'}{v'}\bar{P} - \frac{V}{v'}P,$$

$$p' = k(\bar{P} - P) + P,$$

when

$$k = \frac{V + v'}{v'};$$

$$\therefore \frac{p'}{k} = \bar{P} - P + \frac{P}{k}.$$

Since $\bar{P} - P$ is equal to $Y' - Y$, this equation shows that the pressure p' in the tank is proportional to the change from Y to Y' if to this change be added the small quantity $\frac{P}{k}$ in which P stands for the residual pressure, which need never be more than a few mm., and k for the ratio of the total volume to the volume before expansion. This ratio need be found only roughly if it is desired to measure simply relative pressures. The value of the ratio in this experiment was about 30.

A further advantage was gained by the use of this form of pressure gauge. Since the density of a gas is not directly proportional to its pressure, if the pressure had been measured directly, the values of the density, computed from them by the simple law of Boyle, would have been too small. But if the gas is allowed to expand first by a definite amount and its pressure is measured at some smaller value, the departures from Boyle's law will be negligible, and the densities at the high pressures will be directly proportional to these lower pressures. This may be shown as follows:

$$M = V_0 D_0 = VD,$$

$$\frac{D}{D_0} = \frac{V_0}{V}.$$

But

$$V_0 P_0(1 + aP_0 + bP_0^2) = VP(1 + aP + bP^2);$$

$$\therefore \frac{D}{D_0} = \frac{V_0}{V} = \frac{P(1 + aP + bP^2)}{P_0(1 + aP_0 + bP_0^2)},$$

where D_0, P_0, and V_0 represent the density, pressure, and volume under some standard condition, D, P, and V, the same quantities at some high pressure, and $(1 + aP + bP^2)$ is a factor which gives with a high degree of accuracy the departure from Boyle's law. But if the high pressure P is allowed to fall by expansion to some small pressure P', the following relations will hold:

$$VP(1 + aP + bP^2) = V'P'(1 + aP' + bP'^2),$$

and

$$P = \frac{V'P'}{V} \frac{(1 + aP' + bP'^2)}{(1 + aP + bP^2)};$$

$$\therefore D = \frac{D_0 V'P'(1 + aP' + bP'^2)}{VP_0(1 + aP_0 + bP_0^2)}.$$

Since the ratio $\frac{V'}{V}$ is constant, and since P_0, D_0, and the factor $(1 + aP_0 + bP_0^2)$ are constants, the result found above may be written

$$D = CP'(1 + aP' + bP'^2)$$

where C is a constant. If the pressures are less than one atmosphere, the factor $(1 + aP' + bP'^2)$ will not differ sensibly from unity, and the density D at the high pressure will be expressed as proportional to the small pressure P'. The readings on the pressure gauge will, then, be proportional to the densities at the high pressures and not to the high pressures themselves.

II. Optical Arrangement.

The air of which the index of refraction was to be found was confined in one of two similar stout tubes (Fig. 2), the ends of which were closed by pieces of plate glass. Two plates of steel, held together by bolts running from one end of the tubes to the other, pressed the pieces of plate glass tight against the ends of the tubes. Rubber washers were placed on each side of the glass plates. One of the tubes was connected by copper pressure tubing to the tank which contained the air under investigation, and to the pressure gauge.

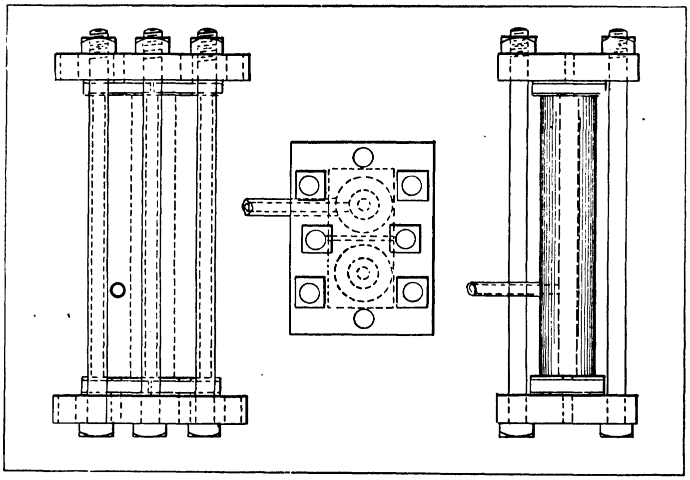

Fig. 2.

It was found convenient to arrange the interferometer mirrors in a way different from that in ordinary use, in a manner which is essentially a modification of Jamin's form. The objection to the latter is that the beams of light cannot be separated to any considerable extent except by the use of very thick plates. On the suggestion of Professor Michelson the mirrors were arranged as indicated in Fig. 3. *A* and *C* are "plane parallel" plates, each of which is coated with a thin film of silver, the plate *A* on the side nearer the source *S* and the plate *C* on the side nearer the telescope *T*. *B* and *D* are plane mirrors. Light coming from the source *S* strikes the thin film on *A*; half of the light is reflected to *B* and out through *C* to *T*. The rest of the light passes through *A* to *D* and is reflected to *C*. At *C* it is reflected, in part, by the thin film *C* to *T*. When the mirrors are properly adjusted, the conditions are right for interference, since the two parts of the original beam, when they arrive at *T*, have traveled nearly equal paths.

The tubes described above were placed between these mirrors, so that the light going from A to B passed down one tube, and that going from C to D passed down the other. When the apparatus was in adjustment and the fringes in view in the telescope, if the density of the air in one of the paths was changed, the fringes would shift.

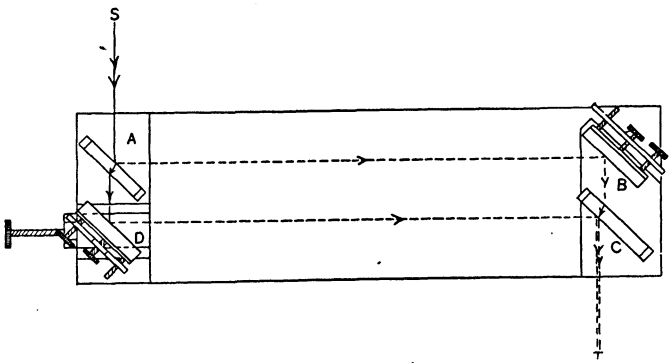

Fig. 3.

Let l equal the length of the tube in the path AB, λ the wave-length in a volume of the green light given out by incandescent mercury vapor in a vacuum tube, let n' and λ' be the index of refraction and wave-length, at a temperature t' and a pressure p'. Let N be the number of wave-lengths in the distance l in a vacuum, and N' the number at a temperature t' and a pressure p'. The following relations hold:

$$N = \frac{l}{\lambda},$$

$$N' = \frac{l}{\lambda'},$$

$$n' = \frac{\lambda}{\lambda'} = \frac{N'}{N},$$

$$n' - 1 = \frac{N'}{N} - 1 = \frac{N' - N}{N}.$$

Since N is constant this shows that the quantity $n' - 1$ under any conditions is proportional to $N' - N$, the number of fringes which would pass on removing from the tube the air of index n', *i. e.*, the air at a temperature t' and pressure p'.

In practice it was, of course, impossible to remove all the air from the tube, but since the ratio $\frac{n-1}{d}$ was known to be very nearly constant, especially at low pressures, the pressure in the tube was simply allowed to fall to the pressure of the room, and the number of fringes was counted. The small additional number which would have passed if all of the air had been removed was computed in the following way, and added to the observed number. Let n'' and d'' be the index of refraction and density respectively of air at the temperature and pressure of the room, and let N'' be the number of wave-lengths in the tube under the same conditions. Let C represent the value of the constant ratio $\frac{n_0-1}{d_0}=\frac{n''-1}{d''}=C$. Then

$$n''=Cd''+1,$$

$$n''=\frac{N''}{N},$$

$$n''-1=\frac{N''-N}{N},$$

$$N=\frac{l}{\lambda_0 n_0};$$

$$\therefore N''-N=(n''-1)N,$$

$$=C\frac{d''l}{\lambda_0 n_0},$$

$$=C'd'',$$

where C' is a constant equal to $\left(\frac{n_0-1}{d_0}\right)\frac{l}{\lambda_0 n_0}$.

The quantity $N''-N$ is proportional to the density d'' and is equal to the number of fringes which would pass on removing the air at the pressure and temperature of the room, and is therefore the number of fringes to be added to the number counted while the pressure was falling from p' to p'', in order to get the number which would have passed if the pressure had fallen from p' to o.

III. OBSERVATIONS.

In the taking of observations the pressure was measured several times, and since only the small volume between the stopcocks was drawn off at each measurement, the pressure in the tank was not sensibly diminished. In the process of counting the number of fringes, the valve at the tank was opened and the tube between the interferometer mirrors and the connecting tubes were allowed to fill. The valve at the tank was then closed, and the union which joined the pressure tubing to the tank was gradually opened and the air in the apparatus allowed to escape. It was not at all difficult to regulate the escape so that the fringes would pass the cross hairs of the observing telescope at any desired rate. On filling the apparatus again and repeating the count, it was found that the number of fringes was slightly less than before, which meant that the pressure in the tank had been slightly reduced by drawing off enough air to fill the tubes. This amount of diminution was constant and it was not difficult to deduce the number of fringes which would have passed at the measured pressure, since this number corresponded to the number of the first count. The succeeding counts simply served as a check on the first one.

After a set of readings at one pressure had been taken, the air was allowed to escape from the tank into the room until the pressure had fallen to some desired point, when another set of readings was taken.

One set of readings, taken at the lowest pressure, is given below.

Temp. 23.8°. Bar. Ht. 74.785.

x	y	y'	$\bar{P}-P$	P	$\bar{P}-P+\frac{P}{K}$
103.5	29.250	40.345	11.095	.535	11.113
"	29.170	40.085	11.085	.455	11.100
"	29.100	40.205	11.105	.385	11.118
"	29.175	40.245	11.070	.460	11.085
"	29.160	40.240	11.080	.445	11.094
"	29.080	40.145	11.065	.365	11.077
"	29.085	40.155	11.070	.370	11.084

Mean—11.096.
Reduced to 0° C.—10.163.

$l = 15.024$ cm. $\lambda_0 = .00005461.$ $n_0 = 1.0002928.$ $N'' - N = 72.6.$

Fringes 249.0
248.8
248.6
248.4

$N' - N = 321.6.$

$$\frac{N' - N}{\bar{P} - P + \frac{P}{K}} = 31.64.$$

A similar set of readings was taken for each of the other pressures, and the results are collected in the table below. In the first column is the approximate pressure in atmospheres; in the second column the pressure as read on the gauge, reduced to 0° C. The third gives the number of fringes, $N' - N$, and the fourth column gives the ratio

$$\frac{N' - N}{\bar{P} - P + \frac{P}{K}}$$

which is proportional to $\frac{n - 1}{d}$.

Atmos.	Press.	$N'-N$	$\frac{N-N'}{\bar{P}-P+\frac{P}{K}}$	
4.	9.989	316.7	31.70	
	10.146	321.2	31.66	31.67
	10.163	321.6	31.64	
7.2	18.281	579.2	31.68	31.70
	18.365	582.7	31.72	
10.6	26.932	852.6	31.66	
14.2	35.990	1142.1	31.69	
19.2	48.780	1545.1	31.68	

IV. Conclusions.

There is probably not an error of more than .1 in any of the numbers of fringes. The errors in the pressures however may run as high as one or two tenths of 1 per cent. Since the variations in the values of the ratio at different pressures do not amount to more than .2 per cent., any apparent variations in the value of the ratio may be due to errors of observation. It seems, therefore, that if there is any departure from the law of Gladstone and Dale up to twenty atmospheres in the case of air, this departure does not amount to more than about .1 per cent. The ratio between Lorentz's equation and that of Gladstone and Dale is so nearly equal to a constant, $2/3$ that an attempt to compare the two would be useless.

It is desired to express thanks to Professor Michelson, both for suggesting the experiment and for valuable advice and encouragement throughout the work. Thanks are due to Mr. F. B. Jewett for the drawings which accompany this article, and to Dr. Mann and Dr. Millikan for revision of the manuscript and checking of results.