COMPARATIVE MEASUREMENTS OF THE TEMPERATURE DERIVATIVES OF VISCOSITY, DENSITY AND REFRACTIVE INDEX OF PURE LIQUIDS AND SOLUTIONS

Part II.—Dilatometric, Refractive Index and Flow Activation Energy Measurements on Benzene at Intervals of 1°C. to 2°C. in the Range of 10°C. to 50°C.

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In part I of this paper, some measurements on the coefficient of dilatation β of ethylene glycol in the range of 20°C. to 80°C. showed a roughly sinusoidal variation with peak-to-peak amplitude seven times the experimental error, and period 5° to 10°C. The majority of minima coincided within $\pm 1°$ C. with sharp jumps already observed in E/R, and a similar correlation between activation energy of viscous flow E/R, refractive index and coefficient of dilatation was earlier observed in the case of water. To elucidate these phenomena, measurements on benzene, a non-hydroxylic liquid are now reported.

The values of $\beta \times 10^4$ ow fairly regular cyclic behaviour, and maxima are observed at 15°C., 20°C., 23.5°C., 27°C., 31.5°C., 37°C., 40°C., 44°C., and 49°C., which coincide exactly in position with the previously found maxima in E/R. The peak-to-peak variation is 0.8 to 1 unit, which is about 15 times the standard deviation. A comparison is made with previous data on density reported by various workers, which shows evidence of 5 or 6 maxima. Measurements are also made for the temperature derivative of refractive index with Na'D' and Cd green lines, and show reasonably cyclic variation.

It is found that the peaks based on refractive index occur at slightly higher temperatures than those for E/R and β , the average temperature excess being 1.2°C., which is one quarter of a complete cycle (4°C.,). This indicates that $\frac{dn}{dT}$ is parallel either to the first derivative or to the integral of E/R and β . Further work on aqueous alcohol solutions is in hand.

Introduction

In part I of this series, some experiments were described¹ on the measurement of coefficient of dilatation β of ethylene glycol* at intervals of 1°C. to 2°C. in the range of 20°C. to 80°C., using a dilatometer with a long calibrated capillary tube. The mean of two sets of measurements of β had an accuracy of $+0.1 \times 10^{-5}$, which is considerably inferior (in absolute terms) to that obtained in the standard data on water below 20°C. Nevertheless, a plot of our mean data showed a number of roughly sinusoidal undulations, of peak-to-peak amplitude 7 times the experimental error, and periods of 5° C. to 10° C. The majority of the minima in this plot coincided within ± 1°C. with the sharp jumps already observed in the activation energy E_{η} for flow of ethylene glycol by Ahsan-ullah and Qurashi.² This lends further credence to a similar correlation pointed out in an earlier communication in the case of (i) the dilatation,

(ii) E_{η} , and (iii) the refractive index of purewater by Ahsanullah and Qurashi.³

While efforts are being made to improve the precision of the dilatometric measurements on ethylene glycol, it was considered desirable at this stage to make a parallel study of a non-hydroxylic liquid, and benzene was chosen for this purpose because of its relatively large coefficient of expansion ($120 \times 10-5$) and because preliminary measurements have already been made on the activation energy of flow, Eq in the range of 8°C. to 46°C., and had shown a sinusoidal behaviour,4 possibly going over into steps below 18°C.

In an attempt to explore fully the nature, the inter-relationship, and the physical basis of these sinusoidal variations and discontinuities, it was thought worthwhile to undertake coordinated measurements of co-efficient of dilatation, refractive index, and activation energy. This communication presents some measurements of thistype on benzene.

^{*}It may be mentioned that in part I the symbol α was used in place of β generally adopted for the coefficient of dilatation of liquids.

Experimental Technique

The density measurements on benzene were carried out with a standard density bottle of volume 10 ml., having a thermometer attached in its stopper, and provided with an auxillary narrow limb in order to enable the expelled liquid to come out when heated to a higher temperature. The limb is provided with a cap on top, to prevent the evaporation and contamination of the experimental liquid. Before starting the actual experiment, the empty weight of the clean, dry bottle is taken with a chemical balance, capable of reading to 0.2 mg. directly, and, by careful eye estimation,

to 0.02 mg. (i.e. $\frac{I}{10}$ th of the scale division). The

bottle is then filled with the experimental liquid and dipped upto the neck in a constant-temperature water bath, by hanging vertically with a rigid attachment. The desired temperature is maintained for 25 to 30 minutes to within $\pm 0.02^{\circ}$ C., inorder to ensure that the experimental liquid attains a uniform temperature throughout. The bottle is then taken out from the bath, the liquid wiped out completely from the outer surface with chamois leather, and the bottle weighed again to find out the mass 'm' of the contained liquid. The temperature of the bath is then raised by 2° C., and the whole process is repeated. The apparent density ρ of the liquid is calculated by the usual formula,

$$\rho = \frac{m}{V} \dots \dots \dots \dots \dots (1)$$

where V is the volume occupied by the experimental liquid, which was determined from the weight of water filling the bottle at 22° C. The coefficient of dilatation β as a function of temperature is then calculated from

$$\beta = + \frac{I}{V_{IO}} \; \frac{\Delta V}{\Delta T} = \; - \frac{I}{\rho_{IO}} \; \frac{\Delta \rho}{\Delta T}, \quad \dots \dots (2) \label{eq:beta}$$

where ρ_{10} indicates the density at 10°C., and $\Delta \rho$ is the change in density in a temperature ΔT . In this measurement we used ρ_{10} as the reference density, because of the experimental difficulty in measuring the density of benzene at 0°C.

For purification, the benzene (Analar grade) is treated with pure concentrated H_2SO_4 in a separating funnel, till the acid does not change its colour after vigorous shaking, and then washed with distilled water and treated with Na₂CO₃ solution to remove traces of acid. This is again washed with excess of water till clean benzene

free of sodium carbonate is obtained. The clear benzene is then kept in a bottle containing $CaCl_2$ to remove water. This is then distilled and the fraction between $80^{\circ}-82^{\circ}C$. is collected. This fraction is further dehydrated with metallic sodium wire. This dehydrated and distilled benzene is then re-distilled, and the fraction between $80-80.5^{\circ}C$. is collected and used. (In one case, the benzene then obtained was frozen at $5^{\circ}C$., and the crystals of benzene collected for measurement of M.P.)

Preliminary Experiment with Distilled and Dried Benzene

The first experiment was carried out in a cylindrical Gallenkamp thermostatic water-bath in the range from 20°C. to 55°C., with temperature interval $\Delta T=2.5$ °C., the temperature of the bath being controlled to ± 0.02 °C. The selection of temperature range from 20° to 55°C. was found to be most suitable for obtaining the best control in temperature, as well as to avoid undue error due to evaporation. The mass of the liquid filling the bottle is determined two to three times at each temperature, and the mean values are taken for the calculation of density. The full set of experiments is performed only in the rising temperature sequence, and in one working day, in order to minimise the experimental time and to avoid difficulties with the evaporation of the test liquid.

The average scatter of the different weighings of the liquid at each temperature is of the order of $\pm 0.000,04$ g. i.e. ± 0.04 mg. This gives density measurements accurate to the fifth decimal place, the sixth decimal place being in error by ± 4 units. This yields an accuracy of approximately 3 parts per 1000 in β as seen in Table 1, where the measured values of density ρ and calculated values of β are given.

The values of $\beta \times 104$ are plotted as crosses in Fig. 1, being labelled as Expt. I, and one can easily observe five maxima at 24°C., 31.5°C., 37°C., 44°C. and 49°C. The maximum-to-minimum variation is of the order of 0.8 unit, which is above 10 times the estimated standard deviation and provides quite satisfactory evidence for the existence of cyclic variations in β .

Experiments with $\Delta T = 2^{\circ}C$. in the Range from 10°-55°C. and Comparison with Standard Data

To confirm the nature and magnitude of these variations of β as a function of temperature, a second set of experiments was carried out with a

Temperature Derivatives of Viscosity, Density and Refractive Index of Liquids. Part II 13 Table 1.—Temperature, the Mass of 9.999,84 C.C. of Benzene, and the Calculated Values of $\beta = -\frac{I}{\rho_{IO}} \frac{\Delta \rho}{\Delta T}$ with Measuring Interval $\Delta T = 2.5^{\circ}$ C., in the Range from 20°-55°C. (Mass of Empty Density Bottle m_I=48.557,36 g.; Volume=9.999,84 C.C. at 20°C.).

Temperature Mean °C. + be	mass of bottle m_{2g} .	(m ₂ -m ₁) g.	Measured density 'p' g./C.C.	$\Delta ho imes 104$	Mean tempera- ture °C.	β×104
	2					
20.00 57.344	,86±0.06 mg. 8.78	7,50 <u>+</u> 0.06 mg.	$0.878,764 \pm 0.06$			
22.50 57.318	.85+0.05 8.76	1.49 ± 0.05	$0.876.163 \pm 0.05$	26.01 ± 0.08	21.25	11.69 ± 0.07
		1,17 0100	01010,100 _ 0100	27.10 ± 0.06	23.75	12.18 ± 0.06
25.00 57.291	$,75\pm0.03$ 8.73	4,39±0.03	0.873,452±0.03	26 00+0 06	26.25	11 69-10 06
27.50 57.265	$,75\pm0.05$ 8.70	8,39±0.05	0.870,852±0.05	20.00±0.00	20.25	11.07±0.00
30.00 57.230	7310.03 9.68	1 37 1 0 03	0 969 150 1 0 03	27.02 ± 0.06	28.75	12.14 ± 0.03
50.00 57.237	,75±0.05 8.08.	1,57±0.05	$0.606,150\pm0.05$	27.53 ± 0.04	31.25	12.38 ± 0.02
32.50 57.211	$,20\pm0.02$ 8.65	3,84±0.02	$0.865,397 \pm 0.02$	25 67 1 0 04	22 75	11 5410.00
35.00 57.185	,53+0.03 8.62	8,17+0.03	0.862,830+0.03	23.07±0.04	33.75	11.54 <u>±</u> 0.02
27 50 57 159	02 1 0 04 0 6 60	0 (7 1 0 04	0.000010.04	27.50 ± 0.04	36.25	12.36 ± 0.02
57.50 57.158	,03±0.04 8.60	0,07±0.04	$0.800,080\pm0.04$	26.30 ± 0.07	38.75	11.82+0.04
40.00 57.131	,73±0.05 8.57	4,37±0.05	$0.857,450 \pm 0.05$	26 10 1 0 07	41 05	11 76 10 04
42.50 57.105	,63±0.04 8.54	8,27±0.04	$0.854,840 \pm 0.04$	20.10±0.07	41.25	11.76±0.04
45.00 57.079	(0 L 0 05 0 52	1 24 1 0 05	0 952 127 1 0 05	27.03 ± 0.07	43.75	12.12 ± 0.04
45.00 57.078	,60±0.05 8.52	1,24±0.05	0.852,137±0.05	25.75 ± 0.07	46.25	11.58±0.04
47.50 57.052	$,85\pm0.05$ 8.49	5,49±0.05	$0.849,562 \pm 0.05$	07 20 1 0 05	40 75	10 29 1 0 02
50.00 57.025	.46+0.02 8.46	8,10+0.02	0.846,823+0.02	27.39±0.05	48.75	12.38±0.02
E2 E0 EC 000		1 10 1 0 05	0.0111(010.05	26.61 ± 0.05	51.25	11.97±0.02
52.50 56.998	$5,85\pm0.05$ 8.44	1,49±0.05	0.844,102±0.05	26.15+0.09	53.75	11.76+0.04
55.00 56.972	$2,70\pm0.08$ 8.41	$5,34\pm0.08$	$0.841,547 {\pm} 0.08$			

temperature interval $\Delta T = 2^{\circ}C$. The experiment was performed in a "Townson-Mercer" thermostatic water bath, where temperature control to $\pm 0.005^{\circ}C$. and better was maintained. The results thus obtained are given in Table 2(a). The best curve drawn through these points, (solid circles) labelled as Expt. II in Fig. 1, brings out clearly the five maxima observed in the preliminary experiment, together with others at intervals of nearly 4°C. The peak-to-peak amplitude is of the order of 0.9 unit, which is above 10 times the estimated experimental error in $\beta \times 104$.

In an attempt to obtain still more information about the character of this more-or-less cyclic variation in the co-efficient of dilatation, a third experiment was performed, but this time the temperatures were selected in such a way that the plotted points fall *in between* the solid circles of the previous experiment II. So that, we can finally get an experimental point at every degree centigrade. The new results are given in Table 2(b)and plotted (as hollow circles) and labelled Expt. III in Fig. 1. The graphs of experiments II and III show satisfactory agreement in the positions of maxima and minima, as shown by the vertical broken lines, and so a *combined plot* is shown in the top-most graph of Fig. 1 for experiments II (solid circles) and III (hollow circles). This combined plot is a considerable improvement on the individual graphs, and enables us to draw a nicely regular graph down to 10° C. The peak-to-peak amplitude varies from 0.8 to 1.6 units, i.e. about 15 times the estimated standard deviation of 0.08 unit obtained from the mean graph. The period is about 4° C., so that the change from one maximum to the minimum is quite sharp, cf. the previously reported E_{η}/R graph4 for benzene by Ahsanullah, Ali and Qurashi.

At this stage, it was thought desirable to make a comparison with previously reported density data on benzene. J. Timmermans⁵ (Physico-chemical constants of pure organic compounds 1958) gives the values of independent sets of determinations (going upto the fifth decimal place) by



Fig. 1.—Series of graphs showing the results of experiments on benzene to determine the values of $\beta = -\frac{1}{-\rho} \frac{\Delta \rho}{\Delta T}$ at various temperatures.

(i) Lowest curve (crosses) marked "Expt. I" is a plot of measurements made with ΔT =2.5°C., and shows five probable maxima.

(ii) Second curve (solid circles) marked "Expt. II" gives the results of the next experiment, which was carried out from 10°C. to 55°C. with a measuring interval of $\Delta T=2^{\circ}C$. All the maxima of Expt. I are reproduced and in addition some more.

(iii) Curve marked "Expt. III" (hollow circles) shows repetition of Expt. II., but at temperatures displaced by 1°C. There is a general agreement with Expt. II.

(iv) Uppermost curve (solid and hollow circles) is a combined plot of Expt. II and III, and good concordance is obtained, the mean graph having a nicely regular appearance.

Cohen and Burij (1937), B.E. (1926), Patterson (1902) and others. Cohen and Burij give the values of p at an interval of 4°C. in the range from 20° to 68°C. This data, believed to be accurate and consistent to $\pm 0.1 \times 10^{-4}$, is given in Table 3(a) together with the calculated values of β , which are plotted in the graph (solid circles) marked as curve I in Fig. 2(a). The individual density values obtained by the various observers (B.E. 1926), Patterson (1902) and others at an interval of 5°-6°C. in the range 10°-50°C. differ by 2-3 unit in the fifth decimal place, and values of β based on the mean of these are shown in the right hand column of Table 3(b), and plotted as hollow circles on curve marked II in Fig. 2(a). A combined plot of the two sets of values is given (solid and hollow circles) in the top of Fig. 2(a). The standard deviation of the individual points about the graph is of the order of 0.10 unit, as expected. The graph shows a series of maxima and minima with an average period of 6.5°C., the peak-to-peak amplitude being of the order of 0.8 units; five of the six peaks agree (Table 4(a)) with those found in our measurements, which are replotted as the lower curve in Fig. 2(b).

Comparison with E_{η}/R Curve

A comparison can now be made with the maxima in $E\eta/R$ as shown in the top curve of Fig. 2(b), where the vertical broken lines show a one-to-one correspondence between the two sets of phenomena. The measure of agreement between the sharp maxima in β and the maxima in $E\eta/R$

(actually
$$E_{\nu}/R = E_{\eta}/R - \frac{T^2}{1000} \Delta \ln \rho / \Delta T$$
) is further

brought out in Table 4(b), the last row of which gives the means of the pairs of corresponding temperatures in the two graphs, together with the deviations of the values for temperatures (at the various maxima and minima) in the two separate properties. The r.m.s. value of these deviations is of the order of $\pm 0.4^{\circ}$ C, whereas the aver-age interval between the successive maxima is nearly 4.0° C. Moreover, the larger peaks in one graph correspond to the larger ones in the other. It can therefore, be concluded that the two sets of maxima and minima correspond fully with each other, and that the sharp variations in β and E_{η}/R correspond in detail, thus providing evidence for the similar physical basis of both these phenomena. Of course, $\Delta \ln \rho / \Delta T$ is only one-tenth of $\Delta \ln \nu / \Delta T$, so that the oscillations in E_{γ} (and E_{ν}) are about ten times the effect of the density through $v = \eta/\rho$.

Here, it is instructive to also make a comparative analysis of the recent measurements of Hu and Parsons⁶ on the viscosity of benzene, which are given to 10⁻⁴ centipoise. After obtaining a straight-line fit through their data in the form $\log \eta = \alpha' + \frac{\beta'}{T}$, they plot the outstanding deviations

$$\delta \times 10^{3} = \left\{ \log \eta - (\alpha' + \frac{\beta'}{T}) \right\} \times 10^{3}$$

against 1/T, and this graph is reproduced photographically in Fig. 3(bottom), in which we have inserted the broken line passing through the points to represent the best experimental curve. This curve is replotted in Fig. 3(top) (solid circles) for comparison with the corresponding values of $\delta \times 103$ deduced from our (kinematic) viscosity measurements (hollow circles and triangles), which form the basis of the undulatory graph for E/R shown in Fig. 2(b). It is seen that (i) our two graphs (broken lines) as well as the mean (full line) run

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TEMPERATURE DERIVATIVES OF VISCOSITY, DENSITY AND REFRACTIVE INDEX OF LIQUIDS. PART II 15. TABLE 2 (a).-(FIRST SET) TEMPERATURE, THE MASS OF 9.999,84C. C. OF BENZENE, AND THE CALCUlated Values of $\beta\!=\!-\frac{I}{\rho_{IO}}~\frac{\Delta\rho}{\Delta T}$ with Measuring Interval $\Delta T\!=\!2\,^{\circ}C.,$ in the Range FROM 10° TO 54°C. (Mass of Empty Bottle $m_1 = 48.557,36$ g., Volume=9.999,84c.c. at 20°C.).

Temperature °C.	$\begin{array}{l} \text{Mean mass of bottle} \\ + \text{ benzene} = m_2 \\ \text{g.} \end{array}$	$(m_2 - m_1)$ g.	Measured density pg./c.c.	Δρ×104	Mean temperature °C.	β×10 4
				an bha an		
10.00 ± 0.01	57.451,95±0.03mg.	8.894,59±0.03mg.	0.889,473±0.03×10 -4	20.67±0.04	11.00	11.62+0.04
12.00 ± 0.00	57.431,28±0.02	$8.873,92 \pm 0.02$	$0.887,406 \pm 0.02$	21.00+0.04	13.00	11.80 ± 0.04
14.00 ± 0.00	57.410,28±0.04	$8.852,92 \pm 0.04$	$0.885,306 \pm 0.04$	21.50 ± 0.07	15.00	12 08+0.06
16.00 ± 0.01	57.388,78±0.06	8.831,42±0.06	$0.883,156 \pm 0.06$	20.72 ± 0.06	17.00	11 64 1 0 05
18.00 ± 0.02	57.368,06±0.02	8.810,70±0.02	$0.881,084 \pm 0.02$	20.72 ± 0.00	10.00	12.42 ± 0.03
20.00 ± 0.00	57.345,95±0.05	8.788,59±0.05	0.878,873±0.05	22.11 ± 0.05	19.00	12.42 ± 0.04
22.00 ± 0.01	57.324,62±0.03	8.767,26±0.03	$0.876,740 \pm 0.03$	21.33±0.05	21.00	11.98±0.05
24.00 ± 0.00	57.303,19±0.04	8.745,83±0.04	$0.874,\!596\pm0.04$	21.44 ± 0.05	23.00	12.05 ± 0.04
26.00 ± 0.00	57.282,61±0.05	8.725,25±0.05	0.872,5 <mark>3</mark> 8±0.05	20.58 ± 0.06	25.00	11.56 ± 0.05
28.00 ± 0.01	57.260,48±0.03	8.703,12±0.03	$0.870,325 \pm 0.03$	22.13 ± 0.06	27.00	12.43 ± 0.05
30.00 ± 0.00	$57.239,98 \pm 0.02$	$8.682,62\pm0.02$	$0.868,262 \pm 0.02$	20.63 ± 0.04	29.00	11.52 ± 0.04
32.00+0.01	57.216,91+0.04	8.659,55+0.04	$0.865,968 \pm 0.04$	22.94 ± 0.05	31.00	12.97 ± 0.04
34.00 ± 0.00	$57.195.98 \pm 0.03$	$8,638.62\pm0.03$	$0.863.875 \pm 0.03$	20.93 ± 0.05	33.00	11.76 ± 0.04
$36,00\pm0,01$	$57, 173, 56 \pm 0, 02$	$8,616,20\pm0,02$	$0.861.633 \pm 0.02$	22.42 ± 0.04	35.00	12.60 ± 0.04
38.00 ± 0.00	57.15210 ± 0.03	$8,594,74\pm0,03$	$0.859.487\pm0.03$	21.46±0.04	37.00	12.06 ± 0.04
40.00 + 0.00	57 131 38 1 0 04	8 574 02 + 0 04	0.857,415 + 0.04	20.72 ± 0.05	39.00	11.64 ± 0.04
40.00±0.00	57.110.11 + 0.03	$8.574,02\pm0.04$	$0.857,413\pm0.04$	21.27 ± 0.05	41.00	11.96 ± 0.04
42.00±0.00	57.110,11±0.03	$8.532,75\pm0.03$	$0.853,288 \pm 0.03$	20.90±0.04	43.00	11.74 ± 0.04
44.00±0.00	57.089,21±0.02	8.551,85±0.02	$0.853,198\pm0.02$	21.17±0.05	45.00	11.89 ± 0.04
46.00 ± 0.00	57.068,04±0.04	8.510,68±0.04	$0.851,081\pm0.04$	20.83±0.06	47.00	11.71 ± 0.05
48.00±0.01	$57.047,21\pm0.05$	8.489,85±0.05	$0.848,998 \pm 0.05$	22.13 ± 0.08	49.00	12.43 ± 0.06
50.00 ± 0.00	$57.025,08\pm0.06$	8.467,72±0.06	0.846,785,±0.06	20.92±0.09	51.00	11.76 ± 0.06
52.00 ± 0.01	57.004,16±0.07	8.446,80±0.07	0.844,693±0.07	20.80 ± 0.08	53.00	11.69+0.06
54.00 ± 0.00	56.983,36±0.05	8.426,00±0.05	$0.842,613\pm0.05$	and an		

closely parallel to that for Hu and Parsons' data, (ii) the steep regions shown by vertical arrows at A,B,C and D in Hu and Parsons' data are found more clearly in the mean graph through the points

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for our data, and (iii) distinct undulations or at least inflexions are observed in our data between successive pairs of steep regions. This suggests (a) that there is general reproducibility between the A. K. M. AHSANULLAH, TAYEB M. QURESHI AND M. M. QURASHI

TABLE 2 (b).—(SECOND SET) TEMPERATURE, THE MASS OF 9.999,84 C.C. OF BENZENE, AND THE CALCU-LATED VALUES OF $\beta = -\frac{I}{\rho_{IO}} \frac{\Delta \rho}{\Delta T}$ with Measuring Interval $\Delta T = 2.5^{\circ}$ C. In the Range FROM 11° to 53°C. (Mass of Empty Density Bottle m₁=48.557,36 g., Volume= 9.999,84 C.C. At 20°C.).

Temperature °C.	Mean mass of bottle + benzene=m ₂ g.	(m ₂ —m ₁) g.	Measured apparent density p g./c.c.	Δρ×104	Mean temperature °C.	β×10 ⁴
11.00±0.01	57.442,01±0.03 mg.	8.884,65±0.03 mg.	$0.888,479 \pm 0.03 \times 10^{-4}$			
12 00 1 0 01	57 420 72 1 0 04	8 8(2 2() 0 04	0.99(250 1.0.04	21.29 ± 0.05	12.00	11.96 ± 0.04
13.00±0.01	57.420,72 <u>±0.04</u>	8.863,36±0.04	0.880,350±0.04	21.31 ± 0.04	14.00	11.98±0.04
15.00 ± 0.00	$57.399,41\pm0.02$	$8.842,05\pm0.02$	$0.884,219\pm0.02$	21.40 ± 0.04	16.00	12.02 ± 0.04
17.00 ± 0.02	$57.378,01\pm0.03$	8.820,65±0.03	$0.882,079 \pm 0.03$	21.02 ± 0.05	18.00	11.82 ± 0.04
19.00 ± 0.01	$57.356,99 \pm 0.04$	8.799,63±0.04	$0.879,977 \pm 0.04$	22.71 ± 0.06	20.00	12.76 ± 0.05
21.00 ± 0.00	57.334,28±0.04	8.776,92±0.04	$0.877,706 \pm 0.04$	20.26 ± 0.04	22.00	$11 39 \pm 0.04$
23.00 ± 0.00	57.314,02±0.02	8.756,66±0.02	$0.875,\!680{\pm}0.02$	20.20 ± 0.04	24.00	12.04 + 0.04
25.00 ± 0.00	57.292,60±0.04	8.735,24±0.04	0.873,537±0.04	21.45±0.04	24.00	12.04±0.04
27.00 ± 0.00	57.268,61±0.03	8.714,85±0.03	0.871,498±0.03	20.39 ± 0.05	26.00	11.46 ± 0.05
29.00 ± 0.01	57.248,98±0.03	8.691,62±0.03	0.869,175±0.03	23.23 ± 0.04	28.00	13.06 ± 0.04
31.00 ± 0.00	57.228,31±0.05	8.670,95±0.05	0.867,108±0.05	20.67 ± 0.06	30.00	11.62 ± 0.05
33.00+0.01	57.206,46+0.05	8.649,10+0.05	0.864,923+0.05	21.85 ± 0.07	32.00	12.28 ± 0.06
35.00+0.00	$57.185.28 \pm 0.04$	$8.627.92 \pm 0.04$	$0.862.805 \pm 0.04$	21.18 ± 0.06	34.00	11.90 ± 0.05
37.00 ± 0.02	57 162 86 \pm 0.04	8 605 50 10 04	$0.860.563 \pm 0.04$	22.42 ± 0.06	36.00	12.60 ± 0.05
30.00±0.00	57.141.20 ± 0.05	8.593.04 LO.05	0.859,400 ± 0.05	21.54 ± 0.06	38.00	12.10 ± 0.05
39.00±0.00	$57.141,52\pm0.05$	8.383,90±0.03	$0.838,409 \pm 0.03$	22.12 ± 0.08	40.00	12.43±0.06
41.00±0.00	57.119,20 <u>±</u> 0.06	8.561,84±0.06	$0.856,197\pm0.06$	20.80 ± 0.08	42.00	11.69±0.06
43.00 ± 0.01	57.098,40±0.05	$8.541,04\pm0.05$	$0.854,117\pm0.05$	21.32 ± 0.07	44.00	12.59 ± 0.06
45.00 ± 0.01	57.077,08±0.05	$8.519,72 \pm 0.05$	$0.851,985 \pm 0.05$	20.80 ± 0.08	46.00	11.69 ± 0.06
47.00±0.01	57.056,28±0.06	8.498,92±0.06	$0.849,905 \pm 0.06$	20.80 ± 0.08	48.00	\pm 11.69 \pm 0.06
49.00±0.00	57.027,48±0.06	8.470,12±0.06	$0.847,025\pm0.06$	$22 02 \pm 0.07$	50.00	$12 \ 37 \pm 0.06$
$51.00{\pm}0.01$	57.005,46±0.04	$8.448,10\pm0.00$	0.844,823±0.04	20.65 0.09	52.00	11 61 10 06
53.00±0.02	56.984,81±0.07	8.427,45±0.00	0.842,758±0.07	20.05±0.08	52.00	11.01±0.00

two independent measurements on benzene and (b) that the extra detail in the data of Ahsanullah, Ali and Qurashi4 deserves further investigation. It is unlikely to be due to absorbed moisture (<0.2%), and could be a consequence of increased stability (upto 1 part in 20,000) consequent on the adoption of the differential technique of experimentation.

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TABLE 3(a).-DENSITY OF BENZENE REPORTED BY COHEN AND BURIJ (1937), AND THE CALculated Values of $\beta = - \; \frac{I}{\rho} \; \frac{\Delta \rho}{\Delta T} \; \text{in} \;$ the

RANGE FROM 20°C. to 60°C. at an Interval of 4°C.

Temperature °C.	Density _f g./c.c.	Mean temperature °C.	β×104
20.0	0.87909		
	0.0	22.0	11.50
24.0	0.87485	26.0	11.85
28.0	0.87058	20.0	11.05
	- 000	30.0	12.05
32.0	0.80624	34.0	11.60
36.0	0.86206	51.	
10.0	0 8= 760	38.0	12.13
40.0	0.05709	42.0	11.83
44.0	0.85343	C	
48 0	0.84012	4.6.0	11.94
40.0	0.04915	50.0	12.33
52.0	0.84469		
56.0	0.84032	54.0	12.13
9	1-J-	58.0	11.94
60.0	0.83602		

Measurements of Temperature Derivative of Refractive Index

The basic technique of carrying out this type of differential measurement of refractive index for a liquid, using a critical incidence Pulfrich refractometer (reliable to 0.000,01) has been described in an earlier communication by Ahsanullah and Ourashi.³ The basic idea is to keep all the settings of the instrument fixed, and to alter only the tangent-screw drum position as the temperature is varied. The drum readings. directly give the change in angle of emergence $\Delta\theta'$ in minutes of are for a change ΔT in temperature. This method is adopted for the present series of experiments on benzene, the temperature being limited to about 50°C. toprevent excessive evaporation of benzene.

TABLE 3(b).--INDIVIDUAL DENSITY VALUES OB-TAINED BY VARIOUS OBSERVERS, VIZ., (B.E. 1926), PATTERSON (1902) AND OTHERS, AND

0					
THE	CALCULATED	VALUES OF (3 ==	I	Δρ.
				9	ΔT

Temperature °C.	Density e g./c.c.	Mean temperature °C.	β×104
10.0	o.88946±0		
15.0	0.88420 ± 2	12.5	11.68
	- 0-006	17.5	11.85
20.0	0.67680 ± 2	22.5	11.60
25.0	0.87364 ± 2	27.5	11.62
30.0	0.86841 ± 3	01.4	10 50
32.9	0.86514 ± 2	31.45	12.52
42.65	0.85462 ± 0	37.78	11.98
45.05	0.85104 ± 1	44.3	12.04
43.95	0.05104 ± 1	46.5	11.60
47.05	0.84989 ± 2	48.5	12.00
50.0	0.84670±4		

Table 4 (a).-Comparison of Temperatures at Sharp Maxima in 3 Found from (i) Present DATA, AND (ii) PREVIOUS DATA.

No. of maximum		1	2	3	4	5	6	7	8	9
Temperature °C. from present data		15.0	20.0	23.6	27.6	31.1	35.8	40.2	44.0	49.6
From previous data	· · · ·	15.6	-	-	26.0	31.6	38.4	_	44.6	50.4
Mean		$15.3 \\ \pm 0.3$	-		$\begin{array}{c} 26.8 \\ \pm 0.8 \end{array}$	$31.4 \\ \pm 0.3$		-	$\begin{array}{c} 44.3 \\ \pm 0.2 \end{array}$	50.0 ± 0.4

N.B. Average deviation of the order of $\pm 0.5^{\circ}$ C.



Fig. 2(a).—(i) Curve I (solid circles) shows the plot of $\beta = -\frac{1}{\rho} \frac{\Delta \rho}{\Delta T}$, calculated from the density data of benzene given by Cohen and Burij (1937). Three peaks are visible in this

given by Cohen and Burij (1957). Three peaks are visible in this -curve.

(ii) Curve II (hollow circles) shows the plot of β calculated from the individual density values obtained by various observers: (B.E. 1926), Patterson (1902), and others. Three peaks are visible here.

(iii) Upper curve (solid and hollow circles) is a combined plot of curves I and II, and the mean graph shows a series of maxima and minima; five of the six peaks are in good agreement with those found in our measurements reproduced in Fig. 2(b) above.

Fig. 2 (b).— Shows a graphical comparison of β (present measurements) with values of E/R by Ahsanullah, Ali and Qurashi (1963) in the top curve, where the vertical broken lines indicate a one-to-one correspondence between the two sets of phenomena.

Measurement with Cadmium Green Line .- The first measurements of $\Delta \theta'$ for $\Delta T = 2^{\circ}C$. were carried out by filling the thermostatted liquid cell with benzene, and taking the repeated drum readings both for heating and cooling temperature sequences at intervals of $\Delta T = 2^{\circ}C$, in the range from 20°C. to 51°C. The cadmium green line $(\lambda = 5086^{\circ}A)$ was chosen because it has a relatively high intensity and therefore it enables precise settings of the refractometer telescope crosshair to be made. The temperature control to +0.03°C. was attained by circulating water from an ordinary controlled bath through (i) the thermostatted block immersed in the liquid and (ii) the chamber around the liquid cell. The value of $\Delta \theta'$ in minutes directly equals $1.1 \times \Delta n/T \times 10^4$. The r.m.s. deviation for $\Delta \theta'$ is seen from Table 5(a) to be of the order of 2% i.e. 3 seconds of arc on the average. A plot of the results is shown in Fig. 4, and labelled as Expt. I. where the hollow circles refer to the rising temperature sequence and the solid circles to the falling temperature sequence. The best graph drawn through these solid and hollow circles shows a series of maxima and minima with a peak-to-peak amplitude of more than I minute of arc, which is 20 to 25 times the r.m.s. experimental error.

In order to improve further the accuracy of the experimental values of $\Delta \theta'$ for 2°C., and to examine the reproducibility of the experiment, the measurements were repeated (with some points measured several times over), still using the cadmium green line. The results thus obtained are shown in Table 5(b) and labelled as Expt. II in Fig. 4. The r.m.s. scatter in $\Delta \theta'$ is here seen to be nearer I second of arc. The experiments I and II show a satisfactory measure of agreement in the positions of maxima and minima, and the doubtful maximum at 31.0°C. in Expt. I appears more clear in the case of Expt. II. The mean values of experiments I and II are therefore also plotted in the middle of Fig. 4, and the smooth graph drawn through the mean plotted points is seen to be reasonably regular.

Table 4 (b).—Comparision of Temperatures of Sharp Maxima in (i) E/R and (ii) β .

No. of maximum	1	2	3	4	5	6	7	8
Temperature from $E\eta/R$	 16.0	20.0	23.6	26.8	32.5	37.5	40.5	44.3
Temperature from β	 15.0	20.0	23.6	27.8	31.1	35.6	40.2	44.0
Mean	 15.5 ± 0.5	$\overset{20.0}{\pm 0.0}$	$\overset{23.6}{\pm^{0.0}}$	$\overset{27.3}{\pm 0.5}$	$\substack{31.8\\\pm0.7}$	$\begin{array}{c} 36.6 \\ \pm 1.0 \end{array}$	$\substack{40.4\\\pm0.2}$	$\substack{44.2\\\pm0.1}$

N.B. Average deviation $\pm 0.4^{\circ}$ C.

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Fig. 3.—(top) The values of $\delta \times 10^3$ from 26°C. to 11°C. in Hu and Parsons' (1958) data replotted (solid circles) for comparison with the corresponding values of $\delta \times 10^3$ deduced from our (kinematic) viscosity measurements (hollow circles and triangles). The steep regions shown by vertical arrows at A,B,C, and D in Hu and Parsons' are found more clearly in the mean graph through the points for our data. Our two sets of data give two graphs that run closely parallel, and their mean compares intriguingly with Hu and Parsons' data.

(bottom) Hu and Parsons' plot of the deviations $\delta \times 10^3 = \{\log \eta - (\alpha' + \beta'/T)\} \times 10^3$ (from the st. line fit in the form $\log \eta = (\alpha' + \beta'/T)$, plotted against I/T from their recent measurements on the viscosity of benzene. This graph is reproduced here photographically, and we have inserted the fine broken line passing through the points to represent the best experimental curve from 26° C. to 11° C.

Measurements with Sodium D Line and Discussion

The above experiments were finally repeated using the sodium D-line. The values of $\Delta \theta'$ for $\Delta T = 2^{\circ}C$. are shown in Table 5(c), and are plotted above the mean graph in Fig. 4, with which they agree in regard to peak positions. The broken vertical lines connecting the corresponding peaks of these two top curves (giving $\frac{1}{2}$ weight to the curve with sodium light) are continued above for comparison with another set of vertical lines giving



Fig. 4.—Curves showing the results of several experiments on benzene to determine the values of $104 \frac{\Delta n}{\Delta T}$ (actually $\Delta \theta'$ for 2°C.) at various temperatures.

(i) Lowest curve marked "Expt I" (solid circles indicate falling temperature sequence, hollow circles rising temperature sequence) is a plot of measurements made with the cadmium green line. The graph shows a series of sharp maxima and minima.

(ii) Second curve, marked "Expt. II" is a repetition of Expt. I.. There is a peak-by-peak agreement with Expt. I.

(iii) The third curve is a combined plot of Expt. I and II, and brings out the regular shapes of the peaks.

(iv) The topmost curve (above the mean graph of Expt. I and II) is a plot of another experiment, using the sodium D-line in the range from 15°C. to 20°C. It reproduces all the maxima and minima observed in the mean graph with the cadmium green line source.

(v) The broken lines connecting the corresponding peaks of the two curves (for sodium and cadmium lines) are continued above for comparison with another set of vertical lines giving the mean positions of corresponding peaks as deduced from the E/R and β curves. A relationship between the two sets can be deduced if we ignore the peaks labelled as small.

the mean positions of corresponding peaks from the E/R and β curves of Fig. 2. It is seen that if we ignore the two small peaks at 23.6°C. and 30.6°C., then a definite relationship emerges between the two sets of peaks, namely that the peaks based on the refractive index curve occur at slightly higher temperatures than those based on E/R and β ; this temperature excess varies from 0.4°C. upto 2.8°C., with a mean value of 1.2°C.

Since a complete cycle has a mean period of 4.4° C., this figure of 1.2° C. is very nearly equal to

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	Ris	sing sequence		Falling sequence					
Temperature °C.	Mean angle • 0'	Mean temperature °C.	$\Delta \theta' \text{ for } 2^{\circ}\text{C.}$ = $\Delta n / \Delta T \times 10^4$	Temperature °C.	Mean angle '0'	Mean temperature °C.	$\frac{\Delta\theta' \text{ for } 2^{\circ}\text{C}}{= \Delta n / \Delta T \times 10^4}$		
		- 1	a. And the second	19.0	4°-56'-38"±2"				
20.0	4°-56′-58″±1′	, 21.0	0°-8'-53"-8' 88	21.0	4°-49'-18"+2"	20.0	$0^{\circ}-7'-20''=7'.33$		
22.0	4°-48′-5″±5″	22.0		21.0		22.0	$0^{\circ}-8'-31''=8'.50$		
24.0	4°-40'-37"±3"	, 23.0	$0^{\circ} - 7' - 28'' = 7'.46$	23.0	4°-40′-4/″±1″	24.0	0°-7'-21"=7'.35		
26.0	4°_32'_57"2"	25.0	$0^{\circ}-7'-40''=7'.66$	25.0	4°-33'-26"±2*	26.0	0°-7'-45"-7' 65		
20.0	4 -52 -57 ±2	27.0	0°-8'-12"=8'.20	27.0	4°-25′-41″±3″	, 20.0	0 -7 -45 - 7 .05		
28.0	4°-24'-45"±3"	29.0	$0^{\circ}-8'-22''=8'.36$	29.0	4°-17'-8"+2"	28.0	0°-8'-33"=8'.55		
30.0	4°-16'-23"±3"	21.0	00 01 01 01 02	21.0	10 0/ 15// 15/	30.0	0°-7′-53″=7′.88		
32.0	4°-8'-21"±3"	31.0	$0^{\circ}-8^{\circ}-2^{\circ}=8^{\circ}.03$	31.0	4°-9°-15°±5°	32.0	0°-7'-33"=7'.55		
34.0	4°-1'-0"-+1"	33.0	$0^{\circ}-7'-21''=7'.35$	33.0	4°-1′-42″±2″	34.0	0°-8'-29"-8' 48		
.04.0	4-(-0 ±1	35.0	0°-7′-58″=7′.96	35.0	3°-53'-13"±3'	, 54.0	0 -0 -27 =0 .40		
36.0	3°-53'-2"±2"	37.0	$0^{\circ}-8'-33''=8'.55$	37.0	3°-46'-3"+3"	36.0	$0^{\circ}-7^{\prime}-10^{\prime\prime}=7^{\prime}.16$		
38.0	3°-44'-29"±1*	20.0	00 (1 = 2" (1 00	20.0	20 20/ 21// 1 2/	38.0	0°-7′-32″=7′.53		
40.0	3°-37'-36"±4"	, 39.0	0-0-33 =0.88	39.0	5 -38 -31 ±3	40.0	0°-7-'-33"=7'.55		
42.0	3°-29'-39"+1"	, 41.0	$0^{\circ}-7'-57''=7'.95$	41.0	3°-30'-58"±2"	42.0	$0^{\circ}-7'-28''=7'.46$		
11.0	28 22/ 40// 1 24	43.0	$0^{\circ}-6'-50''=6'.83$	43.0	3°-23′-30″±2′	· · · · · · · · · · · · · · · · · · ·	09 7/ 20/ 7/ 53		
44.0	3°-22°-49°±3°	45.0	0°-8'-25"=8'.42	45.0	3°-15'-58"±2'	, 44.0	$0^{\circ} - 1^{\circ} - 32^{\circ} = 1^{\circ} .53$		
46.0	3°-14'-24"±2"	47.0	$0^{\circ}-8'-30''-8'$ 50	47.0	3°-7'-16"-1-4"	46.0	0°-8'-42"=8'.70		
48.0	3°-5′-54″±2″	77.0	0-0-00-00	1.0	5 -7 -10 ±4	48.0	0°-7′-9″=7′.15		
50.0	2°-58'-39"+3"	, 49.0	$0^{\circ}-7'-15''=7'.25$	49.0	3°-0'-7"±3"	50.0	$0^{\circ}-7'-42''=7'.70$		
50.0		, 51.0	$0^{\circ}-8'-9'' = 8'.15$	51.0	2°-52'-25"±2'	//			
52.0	2 -30 -30 -4								

TABLE 5 (a).—TEMPERATURE, MEAN READING OF DRUM^{θ}, and the Calculated Values of $\Delta \theta'$ for 2°C. Change in Temperature of Pure Benzene in the Range 20°-52°C. Using Cadmium Green Line with $\Delta T = 2°C$.

Table 5 (b).—Temperature, Mean Reading of Drum θ , and the Calculated Values of $\Delta \theta'$ for 2°C. Change in Temperature for Benzene, Using Cadmium Green Line, in the Range from 15°C. to 50°C.

	Heating	g sequence		Cooling sequence					
Temperature °C.	Mean angle ' θ'	Mean temperature °C.	$\frac{\Delta \theta' \text{ for } 2^{\circ}\text{C.}}{= \Delta n / \Delta T \times 10^4}$	Temperature °C.	Mean angle 'θ'	Mean temperature °C	$\Delta \theta' \text{for } 2^{\circ}\text{C.}$ $= \Delta n / \Delta T \times 10^{4}$		
(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)		
15.0	4°-36′-50″.6±	0″	0°-8'-25",6=8',43	16.0	4°-32′-32″.4+	-2"	- and says and		
17.0	4°-28′-25″.0±	3″ 18.0	0°-8′-31″.0=8′.52	18.0	4°-24′-14″.6±	17.0 _1″	0°-8′-17″.8=8′.29		
19.0	4°-19′-54″.0±	0″ 20.0	0°-7′-54″.0=7′.90	20.0	4°-15′-31″.8±	19.0 (=0"	0°-8′-42″.8=8′.71		
23.0	$4^{\circ}-12 = 0^{\circ}.0\pm 2$ $4^{\circ}-3'-10''.8\pm 1$	22.0	0°-8′-49″.2=8′.82	22.0	4°-6′-50″.2±	1″ 23.0 ($0^{\circ}-8'-41''.0=8'.05$		
25.0	3°-55′-34″.7+3	24.0 2″	0°-7′-36″.1=7′.60	24.0	3°-58′-47″.2±	_1″	0°-7′-54″.1=7′.90		
27.0	3°-47′-10″.8±	26.0 0″	0°-8′-23″.7=8′.40	26.0	3°-50′-53″.1±	2″ 27.0	0°-8'-31".8=8'.53		
		28.0	0°-8′-33″.0=8′.55	28.0	3°-42′-21″.3-	2" (Continu	ed on next page)		

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(Table 5	(b)) continued: —						
(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
29.0	3°-38′-37″.8±2″	30.0	0°-8'-37" 38' 62	28.0	3°-42′-5″.2±2″	29.0	0°-7′-54″ 5-7′ 91
31.0	3°-30'-0".5±2"	32.0	$0^{\circ}-8'-20'', 9=8', 35$	30.0	3°-34'-10".7±1"	31.0	$0^{\circ}-8'-27''.4=8'.46$
33.0	3°-21′-39″.6±2″	34.0	$0^{\circ}-8'-32'', 9=8', 55$	32.0	3°-25′-43″.3±1″	33.0	$0^{\circ}-7'-47''.1=7'.78$
35.0	3°13′-6″.7±2″	36.0	0°-8′-2″.5=8′.04	34.0	3°-17′-56″.2±1″		
37.0	3°-5′-4″.2±0″	38.0	0°-7′-51″.5=7′.86	32.0	3°-25′-40″.0±2″	33.0	0°-7′-53″.3=7′.89
39.0	$2^{\circ}-57'-12'''.7\pm 2''$			34.0	$3^{\circ}-17'-46''.7\pm1''$	35.0	0°-7′-48″.1=7′.80
37.0	$3^{\circ}-4'-21'', 7+1''$	36.0	0°-8′-21″.1=8′.35	38.0	$3^{\circ}-1'-13'', 7+1''$	37.0	0°-8′-44″.9=8′.75
39.0	2°-56′-29″.4±0″	38.0	0°-7′-52″.3=7′.87	40.0	2°-53'-10".6±1"	39.0	0°-8′-3″.1=8′.05
41.0	2°-48'-12".3±1"	40.0	0°-8′-17″.1=8′.28	34.0	3°-17′-58″.1±0″		00 51 104 1 51 50
43.0	2°-40′-0″.4±0″	42.0	$0^{\circ}-8'-11''.9=8'.20$	36.0	3°-10′-16″.0±2″	35.0	$0^{\circ} - 7' - 42'' \cdot 1 = 7' \cdot 70$
45.0	2°-31′-45″.0±2″	46.0	$0^{\circ}-8^{\circ}-0^{''}, 0=8^{\prime}, 00$	38.0	3°-1′-37″.2±1″	39.0	$0^{\circ}-7'-39''.1=7'.65$
47.0	2°-23'-45".0±1"	48.0	0°-7′-50″.9=7′.85	40.0	2°-53′-58″.1±2″		
49.0	2°-15′-54″.1±3″			38.0	3°-1′-16″.5±1″	39.0	0°-7′-45″.1=7′.75
				40.0	$2^{\circ}-44'-40'', 2+3''$	41.0	0°-8′-51″.2=8′.85
				44.0	2°-37′-7″.3±1″	43.0	0°-7′-32″.9=7′.55
				46.0	2°-28'-19".7±1"	45.0	0°-8′-47″.6=8′.79
				48.0	2°-20'-14".2±1"	47.0	$0^{\circ}-8'-5''.5=8'.09$ $0^{\circ}-7'-42''.9=7'.72$
				50.0	2°-12′-31″.3±3″	49.0	0 -1 -42 .9=1.12

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Table 5 (c).—Temperature, Mean Reading of Drum θ , and the Calculated Values of $\Delta \theta'$ for 2°C. Change in Temperature for Benzene, Using Sodium-D-Line.

Temperature °C. (1)	Mean angle 'θ'	Mean temperature	°C. $\Delta \theta'$ for 2°C. $=\Delta n/\Delta T \times 10^4$	Temperature °C.	e Mean angle θ	Mean temperature ^o C	$\Delta \theta' \text{ for } 2^{\circ}\text{C.}$ $= \Delta n / \Delta T \times 104$
(1)	(2)	(3)	(1)				
		(-)	(+)	(5)	(6)	(7)	(8)
16.0 39.40)' 57" 2 L 2"			15.0 3	3°-45′-2″.6±1″	16.0.	0° 8' 28" 8-8' 48
18.0 3 ² -30	$2^{-38''} 5 \pm 2^{''}$	17.0	0°-8′-18″.7=8′.31	17.0 3	3°-36,-33".8±1"	18.0 ()°-8'-39" 6=8' 66
20.0 3°-23	3'-42".8+2"	19.0	0°-8′-55″.7=8′.93	19.0	3°-27′-54″.2±2″	20.0 (0°-8'-11".4=8'.19
22.0 3°-15	5′-19″.4±1″	21.0	0°-8'-23".6==8'.39	21.0	3°-19′-42″.8±2″	22.0 0)°-8'-37".8=8'.63
24.0 3°-8'	-11".6±2"	23.0	0°-8'-7".8=8'.13	23.0	3°-11′-5″.0±2″	24.0 0	0°-8′-0″.0=8′.00
26.0 2°-59	9′-44″.0±1″	25.0	0°-8'-27".6=8'.46	25.0	3°-3′-5″.0±2″	26.0 (0°-8'-28".8=8'.48

(Table continued on next page)

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Table 5(c) continued:-

(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
26.0	2°-59'-7".5±1"	27.0	0°-8'-51" 2-8' 85	23.0	3°-11′-30″.8±1″	24.0	0°-8′-0″ 7-8′ 01
28.0	2°-50'-16".3±2"	29.0	$0^{\circ}-8'-3''$, $1=8', 05$	25.0	3°-3'-30".1±1"	26.0	$0^{\circ}-8'-27'', 0=8', 45$
30.0	2°-42'-13".2±2"	31.0	$0^{\circ}-8'-10''.8=8'.18$	27.0	2°-55′-3″.1±1″	28.0	$0^{\circ}-8'-0'', 1=8', 00$
32.0	2°-34'-2".4±2"	33.0	0°-7′-58″.2=7′.97	29.0	2°-47′-3″.0±2″	30.0	0°-8'-28".9=8'.48
34.0	$2^{\circ}-26'-4''.2\pm 0''$			31.0	2°-38'-34".1±1"	32.0	0°-7′-53″.4=7′.89
24.0	3°-8'-1".0±2"	25.0	0°-8'-28".8=8'.48	33.0	2°-30'-40".7±2"	34.0	0°-8'-19".3=8'.32
26.0	2°-59'-32".2±2"	27.0	0°-8'-48".2=8'.80	35.0	2°-22'-21".4±2"	36.0	0°-8'-13".2=8'.22
28.0	2°-50'-44".0±2"	29.0	0°-8′-0″.0=8′.00	37.0	2°-14′-8″.2±2″	38.0	0°-8′-0″.6= 8′.01
30.0	$2^{\circ}-42'-44'' \cdot 0\pm 2''$	31.0	0°-8'-24".2=8'.40	39.0	$2^{\circ}-6^{\circ}-7^{\prime\prime}.6\pm1^{\prime\prime}$	40.0	0°-8′-11″.5=8′.19
32.0	$2^{\circ}-34^{\circ}-19^{\circ}.8\pm1^{\circ}$	33.0	0°-8′-12″.6=8′.21	41.0	$1^{-}-57^{-}-56^{-}.1\pm 2^{-}$		
36.0	$2^{\circ}-18'-8'' 5+1''$	35.0	0°-7′-58″.7=7′.98	43.0	$1^{\circ}-49'-24'' 0 \pm 0''$	42.0	0°-8′-21″.2=8′.35
38.0	$2^{\circ}-9'-40'', 9+0''$	37.0	0°-8′-27″.6=8′.46	45.0	$1^{\circ}-41'-23'', 7+0''$	44.0	0°-8′-0″.3=8′.01
40.0	$2^{\circ}-1'-57''.7+1''$	39.0	0°-7′-43″.2=7′.72	47.0	$1^{\circ}-33'-20''.0+1''$	46.0	0°-8′-3″.7=8′.06
40.0	2°-1′-58″.0±0″			49.0	1°-25′-14″.5±1″	48.0	0°-8'-5".5=8'.09
42.0	1°-53′-45″.5±1″	41.0	$0^{\circ}-8'-12''.5=8'.21$				
44.0	1°-45′-37″.2±0″	43.0	0°-8′-8″.3=8′.14				
46.0	1°-37′-18″.0±1″	45.0	$0^{\circ}-8^{\prime}-19^{\prime\prime}.2=8^{\prime}.32$				
48.0	1°-29′-15″.1+1″	47.0	$0^{-8} - 2^{-9} = 8^{-05}$				

one-quarter of the complete period, and indicates

that $\frac{dn}{dT}$ runs parallel to either the first derivative

or the integral of E/R (and of course β). This is a highly significant observation, particularly because, according to Gladstone and Dale's law,

$$\frac{dn}{dT}$$
 should be proportional to $\frac{d\rho}{dT}$ i.e. to β if the

polarization remains constant. We may therefore infer that the cyclic variation in dn/dT is due to corresponding changes of polarization. Before following up this line of thought, similar measurements are being made on n-Hexane aqueous alcohol and a few other simple systems in order to tie together the data relating to (i) step-formation and (ii) oscillatory behaviour of E_{η} curves.

References

- M.M. Qurashi and S.W. Ali, Pakistan J. Sci. Ind. Res., 7, 157 (1964).
- 2. A.K.M. Ahsanullah and M.M. Qurashi, Brit. J. Appl. Phys., 13, 334 (1962).
- 3. A.K.M. Ahsanullah and M.M. Qurashi, Pakistan, J. Sci. Ind. Res., **6**, 243 (1963).
- 4. A.K.M. Ahsanullah, S.R. Ali and M.M. Qurashi, Pakistan J. Sci. Ind. Res., **6**, 136 (1963).
- 5. J. Timmermans, *Physico-chemical Constants of Pure Organic Compounds* (Elsevier Publishing Co., Inc., New York, 1958), p. 145.
- 6. P.M. Hu and R.W. Parsons, Proc. Phys. Soc., **72**, 454 (1958).