

photo cell was used from 240 to 400 $m\mu$, and the visible cell above 400 $m\mu$. For the most part spectra in the visible region were measured in 10^{-3} molar solution and ultraviolet spectra in 10^{-4} molar solution. In certain cases, notably samples 14, 18, 19 and 21, it was necessary to use 10^{-4} molar solutions for measurements in the visible. Run 15 was 10^{-3} molar in dye and 10^{-2} molar in hydrochloric acid for visible measurements, and this solution was diluted 1 to 10 for ultraviolet measurements. All solutions were made with Commercial Solvents Gold Shield ethanol.

Summary

Absorption spectra for a series of acylated and

unacylated *p*-phenylazophenyl β -D-glycosides have been measured. No significant differences depending on either gross or stereochemical structure were found. Spectra of two substituted *p*-phenylazophenyl tetraacetyl- β -D-glucosides have been measured and compared with those of the unsubstituted analogs. Spectra of all of the above glycosides have been compared with those of the parent dyestuffs and several other related compounds.

CHICAGO, ILLINOIS
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[CONTRIBUTION FROM THE ANALYTICAL AND PHYSICAL CHEMICAL DIVISION, NORTHERN REGIONAL RESEARCH LABORATORY¹]

Liquid-Vapor Equilibrium of Ethanol-Toluene Solutions

BY CARL B. KRETSCHMER AND RICHARD WIEBE

In continuation of this Laboratory's program of determining the physical properties of ethanol-hydrocarbon mixtures,² measurements have been made of the total and partial vapor pressures of ethanol-toluene solutions. The general purpose of the program is to select a representative member of each of the principal classes of hydrocarbons present in gasoline and to study their behavior in mixtures with ethanol.

The apparatus and methods described previously² were used unchanged except for a modification of the condensate trap and vaporizer as shown in Fig. 1. Any tendency toward stratification in the trap was prevented by means of the magnetic stirrer. The flash boiler of the type suggested and fully discussed by Jones, Schoenborn and Colburn³ simplified the operation of the still.

Density Measurements.—Commercially purified toluene was treated with concd. sulfuric acid until the isatin test⁴ for thiophene and methylthiophenes was negative, washed free of acid, and fractionated in the column previously described.² A melting point determination on the sample of toluene selected gave the value -95.048° , indicating an impurity content of 0.14 mole per cent.^{4a} The physical constants, *b. p.* $110.64 \pm 0.02^\circ$, d^{25}_4 0.86219, n^{25}_D 1.4940 are in good agreement with those reported by the National Bureau of Standards⁵ for toluene containing 0.10% impurity. Moisture was excluded by keeping the toluene un-

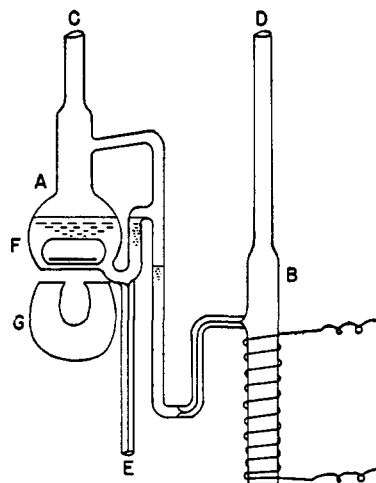


Fig. 1.—Condensate trap and vaporizer: A, trap; B, vaporizer; C, from condenser; D, to saturator; E, sampling line; F, nail in glass capsule; G, rotating magnet, 60 r.p.m.

der its own vapor pressure in a flask sealed to a vacuum manifold through a silicone-lubricated stopcock, and samples were taken by condensing vapor in bulbs attached to the manifold. The ethanol was purified as described previously.² Experimentally determined densities of ethanol-toluene solutions at 25° are given in Table I. As shown in column 4, the change in volume on mixing is positive at low ethanol concentrations but negative over the remainder of the range. The data are in qualitative agreement with those of Washburn and Lightbody,⁶ and the discrepancies may be accounted for by the fact that their solutions were made up by volume which is subject to greater errors than when made up by weight as was done in this work.

Vapor Pressure of Ethanol.—Vapor pressures of ethanol were measured at 10° intervals be-

(6) E. R. Washburn and A. Lightbody, *J. Phys. Chem.*, **34**, 2701, (1930).

(1) One of the laboratories of the Bureau of Agricultural and Industrial Chemistry, Agricultural Research Administration, U. S. Department of Agriculture. Article not copyrighted.

(2) Carl B. Kretschmer, Janina Nowakowska and Richard Wiebe, *THIS JOURNAL*, **70**, 1785 (1948).

(3) C. A. Jones, E. M. Schoenborn and A. P. Colburn, *Ind. Eng. Chem.*, **35**, 666 (1943).

(4) H. N. Holmes and N. Beeman, *ibid.*, **26**, 172 (1934).

(4a) Calculated using freezing point for zero per cent. impurity and cryoscopic constants listed by API Research Project 44 at the National Bureau of Standards, Selected Values of Properties of Hydrocarbons, Table No. 5z, dated Aug. 31, 1945.

(5) A. F. Forziati, A. R. Glasgow, C. B. Willingham and F. D. Rossini, *J. Research Nat. Bur. Standards*, **36**, 129 (1946).

TABLE I

DENSITIES OF ETHANOL-TOLUENE SOLUTIONS AT 25°

Wt. fract. Ethanol	Mole fract. ^a	d_{25}^4	$10^4 \Delta v$, ml./g.
0.0000	0.0000	0.86219	0
.0144	.0284	.86073	32
.0528	.1002	.85735	54
.1344	.2370	.85073	31
.1904	.3198	.84638	- 2
.2826	.4407	.83933	- 61
.2892	.4486	.83883	- 64
.3658	.5356	.83305	-110
.4647	.6345	.82567	-163
.5630	.7204	.81830	-192
.7249	.8405	.80611	-188
.8474	.9174	.79681	-135
.9129	.9544	.79180	- 88
1.0000	1.0000	.78508	0

^a Molecular weights: toluene 92.134, ethanol 46.068.

tween 35° and the normal boiling point. The following equation was fitted to represent these data as well as the earlier ones at 0, 25 and 50°²

$$\log p = 8.11576 - [1595.76/(t + 226.5)] \quad (1)$$

where p is in mm. and t in degrees C. Table II gives the observed temperatures and pressures,

TABLE II

VAPOR PRESSURE OF PURE ETHANOL

Temp., °C.	$p_{\text{obs.}}$, mm.	$p_{\text{obs.}} - p_{\text{calcd.}}$, ^a mm.
0	11.95	0.19
25.000	59.02	0.03
34.988	103.03	- .05
44.994	172.95	- .06
50.000	220.94	- .10
54.988	279.79	.05
65.000	438.04	.06
78.553	766.71	- .07

^a Calculated values are obtained from equation (1).

and deviations of the observed pressures from equation (1). The vapor pressures calculated by means of the above equation are in reasonable accord (within 0.3 mm.) with values obtainable from an equation developed by Scatchard and Raymond.⁷ Equation (1) gives a normal boiling point of 78.328° compared with 78.306° given by the equation of these two authors.

Liquid-Vapor Equilibrium.—Table III gives the liquid-vapor equilibrium data of the ethanol-toluene system at 35 and 55° in terms of the mole fraction of ethanol in the liquid and vapor and the total pressure. The vapor pressure given for pure ethanol was calculated from equation (1) while that for pure toluene was taken from the data of Willingham, *et al.*⁸ For illustration, Fig. 2 shows a plot of total pressures *vs.* liquid and vapor compositions at 35°.

(7) G. Scatchard and C. L. Raymond, *THIS JOURNAL*, **60**, 1278, 3099 (1938).(8) C. B. Willingham, W. J. Taylor, J. M. Pignocco and F. D. Rossini, *J. Research Nat. Bur. Standards*, **35**, 219 (1945).

TABLE III

LIQUID-VAPOR EQUILIBRIUM OF ETHANOL-TOLUENE SOLUTIONS AT 35° AND 55°

Mole fract. C ₂ H ₅ OH		p , mm.	Δy^a
Liquid, x	Vapor, y		
35°			
0.0000	0.0000	46.78
.0330	.4216	79.38	-0.0045
.0468	.4749	86.34	.0018
.1214	.5662	102.09	.0004
.2079	.6014	108.93	.0007
.3620	.6346	114.26	.0024
.4160	.6384	115.34	- .0031
.5930	.6730	117.90	- .0034
.7263	.7164	118.57	- .0002
.8519	.7848	116.56	.0005
.9701	.9318	107.64	.0026
1.0000	1.0000	103.14
55°			
0.0000	0.0000	113.56
.0439	.4369	196.64	-0.0059
.1157	.5679	247.70	.0005
.2497	.6319	279.24	.0008
.4034	.6649	294.17	- .0007
.4142	.6673	294.75	- .0004
.6282	.7150	305.48	.0002
.7186	.7431	307.81	- .0002
.8423	.8049	306.23	.0000
.9163	.8685	299.53	.0004
.9635	.9307	290.47	.0005
1.0000	1.0000	279.89

^a Observed minus calculated, using equation (4) and values of constants in Table IV.

If it is assumed that the pure vapors obey an equation of state of the type

$$p(V - \beta) = RT \quad (2)$$

and that the volume change on mixing the vapors at constant temperature and pressure is zero, the Gibbs-Duhem equation may be written as follows:

$$\ln(p/p_2) = \int_0^x \ln \alpha dx - x \ln \alpha - \ln(1 - y) + \ln(1 - x) - (1/RT)[(\beta_2 - v_2)(p - p_2) - (\beta_2 - \beta_1 - v_2 + v_1) \int_{p_2}^p x dp] \quad (3)$$

in which the relative volatility α is defined as $y(1 - x)/[x(1 - y)]$, p is the total pressure, and p_1 , p_2 , v_1 , v_2 , β_1 , β_2 are the vapor pressures, liquid molal volumes and second virial coefficients of ethanol and toluene, respectively, and x and y are the mole fractions of ethanol in the liquid and vapor respectively. Even though $\int_0^x \ln \alpha dx$ is quite suitable for numerical integration since the plot of $\ln \alpha$ *vs.* x does not differ markedly from a straight line, the evaluation was performed analytically with the aid of the empirical equation

$$\alpha = (A - Bx)/[(x + C)(1 - 2C + Cx)] \quad (4)$$

which permits a simple evaluation of the integral by means of $\int \ln z dz = z(\ln z - 1) + \text{constant}$. Values of the second virial coefficients were ob-

tained from the equation of state of Keyes, *et al.*,⁹ in the form given by Scatchard and Raymond.⁷ The values of $-\beta$ in cc. at 35 and 55° are as follows: for ethanol, 1090 and 865; for toluene, 3383 and 2577. Liquid molal volumes were calculated from equations given in "International Critical Tables."

At 35 and 55° the maximum differences between the observed vapor pressures and these calculated by means of equation (3) are 0.6 and 0.3%, respectively. Somewhat better agreement may be obtained by taking the second virial coefficient for toluene to be 0.9 of that calculated from the equation of state used, as Scatchard and co-workers¹⁰ have done in order to fit the equation to available data of state for non-polar vapors. However, the improvement is scarcely significant. In any case, the close agreement shows that our experimental data are quite consistent thermodynamically.

The equations of Van Laar and Margules, as well as various modifications proposed recently^{11,12a} will fit the present data only when an inconveniently large number of terms is included. Redlich and Kister^{12b} have derived an equation on the basis of continued association, which they have shown to represent alcohol-hydrocarbon systems quite well. The application of their equation to our data will be discussed in a future publication.

A. M. Clark¹⁸ has shown that the composition of the vapor in equilibrium with a given liquid in many binary systems may be represented by one or the other of the following equations

$$y/(1-y) = ax/(1-x) + b \quad (5a)$$

$$(1-y)/y = a'(1-x)/x + b' \quad (5b)$$

The two hyperbolas represented by these equations are tangent or nearly so at a certain value of x (conjugate point) and equations (5a) and (5b) apply to values of x above and below this conjugate point, respectively. Gordon and co-workers¹⁴ have successfully used equation (5b) to fit their data for the systems ethanol-acetone and ethanol-diethyl ether. We have found equations (5) useful for ethanol-hydrocarbon systems near mole fractions of 0 and 1, but the representation breaks down for the intermediate range. The somewhat similar equation (4) was found to represent our data for ethanol-toluene solutions very well, as indicated by the values of Δy in Table III, column 4.

(9) F. G. Keyes, L. B. Smith and H. T. Gerry, *Proc. Am. Acad. Arts Sci.*, **70**, 319 (1936).

(10) G. Scatchard, S. E. Wood and J. M. Mochel, *THIS JOURNAL*, **68**, 1957, 1960 (1946).

(11) K. Wohl, *Trans. Am. Inst. Chem. Engrs.*, **42**, 215 (1946).

(12a) O. Redlich and A. T. Kister, *Ind. Eng. Chem.*, **40**, 345 (1948).

(12b) O. Redlich and A. T. Kister, *J. Chem. Phys.*, **15**, 849 (1947).

(13) A. M. Clark, *Trans. Faraday Soc.*, **41**, 718 (1945).

(14) A. R. Gordon and W. G. Hines, *Can. J. Research*, **24B**, 254 (1946); A. R. Gordon and W. J. Hornibrook, *ibid.*, **24B**, 263 (1946).

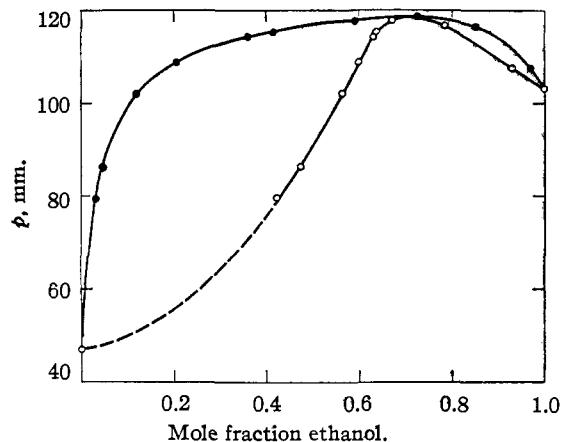


Fig. 2.—Vapor pressure of ethanol-toluene solutions at 35°.

Equation (4) was tested on a variety of systems, but was found applicable only to solutions of alcohols in hydrocarbons and compounds possessing a non-polar and highly symmetrical molecular structure like carbon tetrachloride and tetraethylsilane. We have fitted equation (4) to much of the reliable data available for such systems; the resulting constants are listed in Table IV. Data at

TABLE IV

CONSTANTS IN THE EQUATION $\alpha = (A - Bx)/(x + C)$
($1 - 2C + Cx$)

System	Temp., °C.	A	B	C	Ref.
Ethanol-toluene	35	1.589	1.234	0.045	a
Ethanol-toluene	55	1.782	1.347	.072	a
Ethanol-2,2,4-trimethylpentane	25	1.033	0.955	.021	b
	50	1.282	1.151	.029	b
Ethanol-n-heptane	30	1.207	1.147	.020	c
Ethanol-benzene	50	0.582	0.390	.060	d
Ethanol-cyclohexane	25	0.498	0.472	.015	e
Ethanol-tetraethylsilane	50	10.85	10.25	.035	f
Methanol-benzene	35	1.096	0.906	.040	g
Methanol-benzene	55	1.207	0.981	.048	g
Methanol-carbon tetrachloride	35	0.900	0.768	.015	g
	55	1.070	0.935	.030	g
Methanol-n-hexane	45	0.639	0.628	.004	h
Methanol-n-heptane	760 mm.	2.867	2.807	.020	i
Methanol-toluene	760 mm.	3.805	3.305	.070	i
n-Propanol-benzene	40°C.	0.1863	0.1345	.050	j

^a This work. ^b C. B. Kretschmer, J. Nowakowska and R. Wiebe. ^c J. B. Ferguson, M. Freed and A. C. Morris, *J. Phys. Chem.*, **37**, 87 (1933). ^d R. A. Lehfeldt. ^e E. R. Washburn and B. H. Handorf, *THIS JOURNAL*, **57**, 441 (1935). ^f N. Bjerrum and E. Josefowicz, *Z. physik. Chem.*, **A159**, 194 (1932). ^g G. Scatchard, S. E. Wood and J. M. Mochel. ^h J. B. Ferguson, *J. Phys. Chem.*, **36**, 1123 (1932). ⁱ M. Benedict, C. A. Johnson, E. Solomon and L. C. Rubin, *Trans. Am. Inst. Chem. Engrs.*, **41**, 371 (1945). ^j S. C. Lee, *J. Phys. Chem.*, **35**, 3558 (1931).

constant pressure appear to give about as close a fit as those at constant temperature. The agreement is in general within a few tenths mole per cent. with some greater scattering near the ends of the concentration range. Only two of the three constants in equation (4) are really independent, since equation (3) with $x = 1$ provides one relation between them. This relation was disregarded

in fitting the equation, but it was found to be closely satisfied in all cases for which the test was made. Using equations (3) and (4), it would be possible to calculate the complete vapor pressure and composition curves from experimental values at just one value of x (conveniently, the azeotrope), if the vapor pressures of the pure components were known.

Inspection of Table IV shows that C and $(A - B)/A$ become smaller with increasing tendency of the solution to separate into two phases; e. g., they are smaller at lower temperatures, smaller for methanol than for ethanol with the same hydrocarbon, and smaller for paraffins and naphthenes than for aromatic hydrocarbons with the same alcohol. A and B are roughly proportional to p_1/p_2 , since $\alpha = (p_1/p_2)(\gamma_1/\gamma_2)$, and the ratio of activity coefficients γ_1/γ_2 at a given value of x does not vary markedly among these systems.

The system ethanol-toluene has been studied at 50° by Lehfeldt¹⁵ and at 60 to 85° by Wright.¹⁶ The total pressure at 20 to 90° has been measured by Deveux and Schouteden.¹⁷ Since none of these data appeared to be sufficiently accurate for our purposes, the present work was undertaken.

Thermodynamic Properties.—The assumptions leading to equation (3) yield an equation for the excess free energy of mixing

$$G^E = RT\{x \ln (p_1/p_1^0) + (1-x) \ln [p_2(1-y)/p_2^0(1-x)]\} + x(\beta_1 - v_1)(p - p_1) + (1-x)(\beta_2 - v_2)(p - p_2) \quad (6)$$

Values of G^E at 35 and 55°, calculated by means of equation 6 using values of β previously given, are listed in Table V. Smoothed values of the heat of

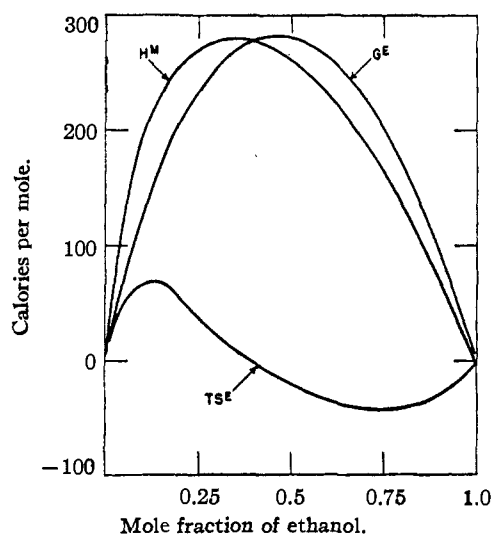


Fig. 3.—Excess thermodynamic functions of ethanol-toluene solutions at 35°.

(15) R. A. Lehfeldt, *Phil. Mag.*, [5] **46**, 46 (1898).

(16) W. A. Wright, *J. Phys. Chem.*, **37**, 233 (1933).

(17) J. Deveux and F. Schouteden, "Dampdruk en Verdampingswarmte van het Mengsel Ethanol-Toluën," Brussels, Palais des Academies, 1937.

TABLE V
EXCESS THERMODYNAMIC FUNCTIONS FOR ETHANOL-TOLUENE SOLUTIONS, CAL./MOLE

Mole fract. ethanol	GE, 35°	GE, 55°	TSE, 35°	HM
0.0439	63	61	47	110
.1157	143	137	68	211
.2497	236	232	36	272
.4034	280	281	- 2	278
.4142	281	282	- 4	277
.6282	256	258	-35	221
.7186	219	221	-41	178
.8423	143	147	-35	108
.9163	83	84	-25	58
.9635	37	38	-12	25

mixing H^M and the excess entropy S^E multiplied by absolute temperature at 35° are also given. These were calculated from the temperature dependence of G^E , assuming additive specific heats. The various functions are plotted in Fig. 3. The entropy curve is quite similar to that found by Scatchard¹⁰ for methanol in benzene, and the explanation for the negative excess entropy of mixing proposed in our previous article² applies here also.

The heats of mixing in Table V are approximately 1.5 times as large as Washburn and Lightbody's⁶ calorimetric values at 23°, though the two curves have the same unsymmetrical shape. More than half of this discrepancy can be eliminated by abandoning the assumption of additive specific heats and assuming a value of 2 cal. mole⁻¹ deg.⁻¹ for ΔC_p of mixing at 0.5 mole fraction, and the remainder can be attributed to entirely reasonable errors in our values of G^E and Washburn and Lightbody's values of H^M . For solutions of ethanol in benzene, Perrakis¹⁸ has found that ΔC_p is 1.55 cal. mole⁻¹ deg.⁻¹ at 0.5 mole fraction, and the data of Williams and co-workers¹⁹ for methanol-benzene mixtures yield a somewhat greater value of ΔC_p . Therefore 2 cal. mole⁻¹ deg.⁻¹ is not an unreasonable value to assume for the ethanol-toluene system. The result of these considerations is to make TS^E more negative than shown in Fig. 3 by as much as 30 or 40 cal., while still leaving a region of positive excess entropies at low alcohol concentrations.

Summary

The vapor pressure of ethanol has been determined from 25° to the boiling point. Densities at 25° and vapor compositions and pressures at 35 and 55° have been measured for solutions of ethanol in toluene. The results are quite consistent with the Gibbs-Duhem relation.

The excess free energy and entropy and the heat of mixing have been calculated from these data. The excess entropy has negative values similar to those found in other systems containing alcohols.

A simple empirical equation has been developed

(18) Perrakis, *J. chim. phys.*, **22**, 297 (1925).

(19) G. C. Williams, S. Rosenberg and H. A. Rothenberg, *Ind. Eng. Chem.*, **40**, 1273 (1948).

which represents the liquid-vapor equilibrium of alcohol-hydrocarbon systems quite accurately. Constants in this equation are presented for etha-

nol-toluene solutions and a number of systems previously investigated.

PEORIA 5, ILLINOIS

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[CONTRIBUTION FROM THE UNIVERSITY OF COLORADO]

Diamagnetic Susceptibilities of Some Chloro and Fluoro Derivatives of Methane and Ethylene¹

BY J. R. LACHER, RALPH E. SCRUBY AND J. D. PARK

In this Laboratory direct measurements of reaction heats of simple organic fluorine compounds are being made.² The results obtained to date show that the addition of chlorine to fluorinated olefins gives heats which deviate strongly from an additivity rule. For example, the vapor phase heat of chlorination of tetrafluoroethylene is -57.3 kcal./mole. When one fluorine atom is replaced by chlorine in this olefin to give trifluoro-chloroethylene, the heat of chlorination drops by 8.5 kcal./mole. On the other hand, if a fluorine atom is replaced by a trifluoromethyl group to give hexafluoropropene, the reaction heat falls off 10.1 kcal./mole. Hexafluorocyclobutene in the same reaction liberates only 37.3 kcal./mole.

These large variations in reaction heats are probably to be correlated with the fact that a C-Cl bond distance will undergo a shrinkage depending on whether or not one or two fluorine atoms are attached to the same carbon atom as is the chlorine.³ Further, the C=C bond distance is shorter in tetrafluoroethylene than it is in ethylene. If the bond distances change, so will the bonding energies, as will also the densities of the electron cloud associated with these bonds.

It is well known⁴ that molar diamagnetism is, to a first approximation, a measure of the sum of the mean cross sectional areas of the electron orbits present in a molecule. One might expect, therefore, to be able to correlate the strongly exothermic character of the reaction heat which results when chlorine adds to tetrafluoroethylene with a general shrinkage in the electron orbits as measured by the change in susceptibility associated with the reaction. On the other hand, since the reaction of chlorine with hexafluorocyclobutene is considerably less exothermic, one might expect in this case either no change or perhaps an expansion of the electron clouds. In order to test these ideas it is necessary to measure first the susceptibilities of the organic fluorine compounds involved. The present paper reports measurements

on some chloro and fluoro derivatives of methane and ethylene.

Very little experimental information is available concerning the molar susceptibilities of the simpler organic halogen compounds, particularly those containing fluorine. Recently, French and Trew⁵ have summarized the molar susceptibilities of some polyhalogen derivatives of methane. All of these compounds show strong deviations from Pascal's additivity rule. Pascal⁶ interprets the anomalous diamagnetism of organic polyhalogen compounds as being due to the mutual deformation of the electronic orbitals of the halogen atoms in contact. This effect is a maximum for iodine and is small for fluorine. The observed molar susceptibilities of the chlorine, bromine and iodine derivatives of methane can be interpreted empirically by assuming that they are the sum of atomic susceptibilities and six interaction terms directed along the edges of a tetrahedron.⁷

Experimental

The Quincke method,⁴ which is well suited for measurements on liquids, was used. The measurements were relative ones and the displacement of the meniscus of a given liquid was compared with that produced by a standard liquid in the same field strength and in an identical tube. For liquids boiling above room temperature, the tubes were made from 50-ml. distilling flasks using 3.0-mm. precision bore tubing. Since several of the compounds studied are gases at ordinary conditions, special tubes were constructed which would hold liquids under moderate pressures. One is shown diagrammatically in Fig. 1. The reservoir was constructed of brass and had an internal diameter of 4.3 cm. The movement of the meniscus was observed in a thick-walled glass precision bore capillary. "Teflon" gaskets were used to ensure a tight seal. With this arrangement, the susceptibility of the vapor above the meniscus is not negligible. However, if one assumes that the mass susceptibility of the vapor is identical to that of the liquid (which is undoubtedly true for the non-associated compounds used here), one can readily show that the change in height of the meniscus is independent of the vapor density. The reason is that the net magnetic force operating will then be directly proportional to the difference in density of the vapor and liquid. This is also true of the opposing force due to the hydrostatic head developed.

The electromagnet⁸ used operated on 21 to 25 amperes at 120 volts and gave fields around 18,000 gauss. Changes in liquid level were measured by means of a traveling microscope to an accuracy of ± 0.004 mm. Displacements be-

(1) Presented before the Division of Inorganic and Physical Chemistry, 114th Meeting of the American Chemical Society, Portland, Oregon, September 13-17, 1948. This work was supported by Contract N6onr-231, Task Order 6, with the Office of Naval Research, United States Navy.

(2) J. R. Lacher, J. D. Park, *et al.*, submitted to THIS JOURNAL.

(3) S. H. Bauer, MDDC-1494, U. S. A. E. C.

(4) P. W. Selwood, "Magnetochemistry," Interscience Publishers, Inc., New York, N. Y., 1943.

(5) French and Trew, *Trans. Faraday Soc.*, **41**, 439 (1945).

(6) P. Pascal, *Compt. rend.*, **217**, 657 (1943).

(7) J. R. Lacher, THIS JOURNAL, **69**, 2067 (1947).

(8) H. C. Staehle, Ph.D. Thesis, University of Colorado, 1933.