CHAPTER IV

MICROWAVE CONDUCTIVITY AND DIPOLE MOMENT OF SOME SUBSTITUTED BENZENE

Introduction:

In recent years, much attention has been drawn to study the concentration variation of high frequency conductivity (Ghosh S K et al, 1980 and Datta S K et al 1981) to yield the dielectric relaxation parameters such as: relaxation time (\mathcal{T}), dipole moment (\mathcal{M}_j) of polar dielectrics, heat enthalpy ($\Delta H_{\mathcal{T}}$), entropy of dielectric relaxation ($\Delta S_{\mathcal{T}}$) and the activation energy ($E_{\mathcal{T}}$) etc. In the previous chapter (III) it has been shown that under the microwave electric field the dipole moment is not a fixed quantity, but varies with temperature though it is supposed to be constant under the low frequency and the d.c. electric fields.

The present chapter will report the permanent dipole moment and bond moment of the following seven dielectric liquids. The dipole moment were calculated from the concentration variation of microwave conductivity method. The theory and method of determination of dipole moment (\mathcal{M}_j) from variation of h.f. conductivity with weight fraction W_j are given in chapter (III).

The seven dielectric liquids (i) m-aminobenzotrifluoride (ii) O-nitrobenzotrifluoride (iii) m-nitrobenzotrifluoride (iv) O-chlorobenzotrifluoride at 3.0166 cm wave length, (v) O-chloronitrobenzene (vi) 4-chloro-3-nitrobenzotrifluoride and (vii) 4-chloro-3-nitrotoluene at 3.0 cm wavelength electric field, the systems are very interesting regarding the sizes and shapes of the molecule whose dipole moments and bond moment have been reported.

Result and Discussion:

The microwave conductivity K_{ij} of polar nonpolar liquid mixtures were calculated with the help of the fundamental eq. (3.4), i.e.

$$K_{ij} = \frac{\omega}{4\pi} \left(\epsilon_{ij}^{"2} + \epsilon_{ij}^{'2} \right)^{1/2} \dots 4.1$$

Further the total conductivity K_{ij} for the h.f. region is represented by eq. (3.6) i.e.

$$K_{ij} = K_{\infty} + K'_{ij} / \omega \tau_s$$
 ... 4.2

where K_{∞} and T_{S} are constants (d.c. conductivity and relaxation time of the solute molecules in the dilute solution respectively). Therefore the total conductivity can be written with the help of eq. (3.8) and eq. (4.2) as

$$K_{ij} = K_{\infty} + \frac{\mu_j^2 N \rho_{ij} F_{ij}}{3 M_j k T} \left(\frac{\omega}{1 + \omega^2 \tau_s^2} \right) W_j \quad \dots \quad 4.3$$

The eq. (4.3) clearly shows that the K_{ij} is a function of weight fraction. Hence the variation of K_{ij} with W_j could be expressed according to mathematical relation

$$K_{ij} = \alpha + \beta W_{ij}$$
 ... 4.4

where \swarrow and β are constants. The constant β is the most important factor for determining the τ_s and \mathcal{M}_j . The slope β could be obtained from the graphical plot of K_{ij} against W_j at $W_j \rightarrow 0$ or from the eqn. (4.4). The K_{ij} of the seven respective solutions computed by using the following least square fit relations are

i)	$K_{ij} = 10^{-10} = 1.1106 + 2.3536 W_{j}$	
ii)	$K_{ij} = 10^{-10} = 1.1169 + 4.1557 W_j$	
iii)	$K_{ij} = 10^{-10} = 1.1262 + 1.7747 W_{j}$	
iv)	$K_{ij} = 10^{-10} = 1.1199 + 2.0159 W_{j}$	
V)	$K_{ij} = 10^{-10} = 1.1265 + 4.0539 W_{j}$	
vi)	$K_{ij} = 10^{-10} = 1.1286 + 1.1894 W_{j}$	and
vii)	$K_{ij} = 10^{-10} = 1.1250 + 2.8594 W_{j}$	

To calculate the variation of K_{ij} at various weight fraction W_j , the experimental data of ϵ'_{ij} and ϵ''_{ij} given the paper of (Arrawatia et al.1977),

and Gupta P C et al, (1978) have been considered.

The K_{ij} values thus obtained are tabulated in table (4.1a, 1b). The nature of variation of K_{ij} with weight fraction W_j and also the nature of their fitted curves are shown in fig. (4.1a -4.1 g).

The dipole moments of seven systems of interest have been computed with the help of eq. (3.13) i.e.

$$\mathcal{M}_{j} = \left(\frac{3M_{j}kT}{N\rho_{j}F_{i}} \cdot \frac{\beta}{\omega b}\right)^{/2}$$

and it is observed that the dipole moment values of many molecules measured in solution were consistently different from one solvent to another and this effect has been considered as due to solvent effect. So the correct values of induce dipole moment calculated from the most convenient Müllers empirical equation

$$\mathcal{M}_{j}^{(S)} = \mathcal{M}_{j} \left[1 - C \left(\epsilon_{j} - 1 \right)^{2} \right]$$

are also tabulated in table (4.2) along with the available literature data for comparison. The \mathcal{M}_j data have also been computed from the bond dipole moment and bond angles, assuming that the benzene molecule is a planer one. The molecular parameters needed for this computation of theoretical values of \mathcal{M}_j are shown in fig. 4.2 and placed in table (4.3).















The microwave conductivity data in the low concentration region are of much interest as they reveal a completely different situation in comparison to those of high concentration which is shown in the previous chapter (III). This is the reason which led us to study the systems reported here and to calculate their dipole moments. High concentration conductivity data of a polar nonpolar liquid mixture is liable to yield the dipole moment due to dimer formation while those in low concentration region behave linearly with the weight fraction and give the dipole moment due to monomer formation i.e. solute-solvent association. The computed dipole moments agree with those reported . experimentally as well as computed theoretical ones, as illustrated in table (4.2). This fact suggests the basic soundness of the method adopted here.

The variation in the \mathcal{M}_j values for these substituted molecules in benzene might be due to (i) the difference in values of group moments and (ii) inductive effect. The dipole moment of 0-nitrobenzotrifluoride is always greater than that of m-nitrobenzotrifluoride (Table 4.2) both shown by the computed and theoretical \mathcal{M}_j 's due to the two different positions of the NO₂ (shown in fig. 4.2) group in the compound.



Fig4.2-Conformation of the different molecules studied showing the , orientation of the bond axes and the dipole moments and also bond moments: [1, m-Aminobenzotrifluoride: 2, o-nitrobenzotrifluoride: 3, *m*-nitrobenzotrifluoride: 4. o-chlorobenzotrifluoride. 5, o-chloronitrobenzene: 6, 4-chloro-3-nitrobenzotrifluoride & 7, 4-ct 4-chloro-3-nitrotoluene]

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The other two compounds, viz. m-aminobenzotrifluoride and O-chlorobenzotrifluoride have smaller values of dipole moments due to the same reasons as mentioned earlier.

The group moment of -CF3 groups acts in the reverse direction to that of - Cl group and hence its net effect is to reduce the total moment along the common axis. 4-chloro-3-nitrobenzotrifluoride thus shows the lower value of \mathcal{M}_{i} in benzene than in 0-chloronitrobenzene. The group moment of - CH3 group. acts in the direction of -Cl group and hence the resultant dipole moment increases along the common axis. That is why the last compound in table (4.2) and in fig. (4.2), i.e. 4-chloro-3-nitrotoluence has higher dipole moment in benzene than in O-chloronitrobenzene. The close agreement between the computed \mathcal{M}_{i} values with the theoretical ones shows that the dipole moments of these compounds have a preferred conformation, as depicted in fig. (4.2). Regarding the group moments assumed by us, they compensate each other to a certain extent.

Table 4.1a

Values of dielectric constant (ϵ'_{ij}), loss factor (ϵ''_{ij}) conductivity (K_{ij}) and weight fraction (W_j) at wave length $\lambda = 3.0166$ cms.

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	Systems	W ;	€' _{ij}	ε" ε" ε	$\begin{array}{c} K_{ij} \mathbf{x} \\ 10 \\ 10 \\ 10 \\ 10 \\ 10 \\ 10 \\ 10 \\ 1$
1.	m-Aminobenzo- trifluoride	0.0142 0.0265 0.0379 0.0561 0.0682	2.306 2.351 2.412 2.480 2.556	0.053 0.088 0.131 0.178 0.233	1.147 1.170 1.201 1.236 1.276
2.	O-Nitrobenzo- trifluoride	0.0085 0.0167 0.0244 0.0335 0.0402	2.315 2.386 2.445 2.515 2.571	0.058 0.108 0.159 0.205 0.255	1.151 1.188 1.218 1.255 1.285
3.	m-Nitrobenzo- trifluoride	0.0096 0.0173 0.0245 0.0326 0.0380	2.302 2.322 2.347 2.375 2.404	0.032 0.060 0.082 0.106 0.128	1.145 1.155 1.168 1.182 1.197
4.	0-chlorobenzo- trifluoride	0.0081 0.0142 0.0225 0.0312 0.0374	2.278 2.315 2.350 2.368 2.404	0.027 0.048 0.061 0.087 0.106	1.133 1.151 1.169 1.178 1.197

Table 4.1b

Values of dielectric constant (ϵ'_{ij}), loss factors (ϵ''_{ij}) conductivity (K_{ij}) weight fractions (W_j) at wavelength $\lambda = 3.0$ cms.

	System	* 1 1 1 1 1	W _j	€́ij	ϵ''_{ij}	K_{ij} $x 10^{-10}$ in esu
5.	0-chloronitro- benzene		0.0109 0.0173 0.0217 0.0280 0.0330	2.340 2.393 2.422 2.479 2.511	0.066 0.100 0.126 0.165 0.192	1.170 1.198 1.213 1.242 1.259
6.	4-chloro-3-nitro- benzotrifluoride		0.0096 0.0154 0.0275 0.0410 0.0519	2.266 2.289 2.316 2.369 2.386	0.021 0.035 0.058 0.093 0.112	1.133 1.145 1.158 1.185 1.194
7.	4-chloro-3-nitro- toluene	- - .	0.0072 0.0144 0.0224 0.0323 0.0453	2.298 2.322 2.375 2.426 2.499	0.046 0.088 0.133 0.179 0.252	1.149 1.162 1.189 1.216 1.256

Table 4.2

The Computed Values of b, $\mathcal{M}_{j}^{(S)}$, \mathcal{M}_{j} (Compt.), \mathcal{M}_{j} (Rept) at 35°C									
System	ь (12 T _S ×10 Rept)	(Compt)			Mjin D			
	$\overline{\lambda}$	= 3.01	<u>66 cm.</u>						
m-Aminobenzo- trifluoride	0•7234	9.9	3.1971	3.40	3.16	2.48			
0-Nitrobenzo- trifluoride	0.6913	10.7	4.7332	5.0 3	4.54	6.18			
m-Nitrobenzo- trifluoride	0.6403	12.0	3.2138	3.42	3.33	3.74			
O-Chlorobenzo- trifluoride	0 .7356	9.6	3.1062	3.30	3.15	3.98			
$\Lambda = 3.0 \text{ cm}$.									
0-Chloronitro- benzene	0.5816	13.5	4.6137	4.91	4.71	5.28			
4-Chloro-3- nitrobenzo- trifluoride	0.7088	10.2	2.7084	2.88	3.17	3.78			
4-Chloro-3- nitro-toluene	0.3670	20 .9	5.0899	5.41	4.88	5.58			

Table 4.3

Electric moment of groups and bonds.

Group or bond	Electric moments in Debye units.
- CF3	2.86
- NH ₂	1.166
- NO ₂	4.23
- Cl	1.69
- CH3	-0.4

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Microwave Conductivity & Dipole Moment of Substituted Benzenes

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The slope of the curve of the variation of the microwave conductivity with concentration at infinite dilution, is used to obtain the dipole moments of some substituted benzotrifluorides and substituted benzenes at 35°C, subjected to the 3.0166 and 3.0 cm wavelength electric fields respectively. The data thus estimated are in agreement with the reported μ_j values, suggesting the uniqueness of the method adopted.

In recent years, much attention has been drawn to study the concentration variation of high frequency conductivity^{1,2} to yield the dielectric relaxation parameters: relaxation time (τ) , dipole moment (μ_j) of polar dielectrics, heat of enthalpy (ΔH_{τ}) , entropy of dielectric relaxation (ΔS_{τ}) and the activation energy (E_{τ}) , etc. Recently, it has been shown by Acharyya *et al.*³ that under the microwave electric field the dipole moment is not a fixed quantity, but varies with temperature though it is supposed to be constant under the low-frequency and the dc electric fields.

The microwave conductivities (K_{ij}) of (i) maminobenzotrifluoride, (ii) o-nitrobenzotrifluoride, (iii) m-nitrobenzotrifluoride, (iv) o-chlorobenzotrifluoride at 3.0166 cm wavelength electric field are found to vary linearly with the concentration expressed in weight fractions (W_i) . Similarly do those of (v) ochloronitrobenzene, (vi) 4-chloro-3-nitrobenzotrifluoride and (vii) 4-chloro-3-nitrotoluene at 3.0 cm wavelength electric field, thereby yielding a constant slope β at any stage of dilution and thus makes the procedure more reliable than those done earlier³. Moreover, the systems mentioned above are very interesting regarding the sizes and shapes of these molecules. Of the seven systems, five are benzotrifluorides and two are comparatively less simple. The variation of K_{ij} with W_j could be expressed by $K_{ij} = \alpha$ + βW_i of which β is the most important to yield both τ_s as well as μ_j of the polar liquids. The K_{ij} of the respective solutions are computed by using the following relations:

$$K_{ii} \times 10^{-10} = 1.1106 + 2.3536 W_{ii}$$

 $K_{ij} \times 10^{-10} = 1.1169 + 4.1557 W_j$ $K_{ij} \times 10^{-10} = 1.1262 + 1.7747 W_j$ $K_{ij} \times 10^{-10} = 1.1199 + 2.0159 W_j$ $K_{ij} \times 10^{-10} = 1.1265 + 4.0539 W_j$ $K_{ij} \times 10^{-10} = 1.1286 + 1.1894 W_j$ $K_{ij} \times 10^{-10} = 1.1250 + 2.8594 W_j$

with the help of the available data^{4.5}.

Theoretical Formulations—The high frequency conductivity⁶ of a polar-nonpolar liquid mixture is given by $K_{ij} = K'_{ij} + j K''_{ij}$ where K'_{ij} is the real part of the conductivity $= \omega \varepsilon''_{ij}/4\pi$, and K''_{ij} is the imaginary part of the conductivity $= \omega \varepsilon'_{ij}/4\pi$. Now the magnitude of the total high frequency conductivity is usually computed from the relation

$$K_{ij} = \frac{\omega}{4\pi} \left[\varepsilon_{ij}^{\prime\prime 2} + \varepsilon_{ij}^{\prime 2} \right]^{1/2} \qquad \dots (1)$$

The ε'_{ij} of the solution in the microwave region is very small and equals the optical dielectric constant, but still $\varepsilon'_{ij} \ge \varepsilon''_{ij}$ and ε''_{ij} is responsible for the absorption of electrical energy to yield resistance to polarization; hence the real part K'_{ij} of hf conductivity of solution of weight fraction W_j of polar solute at temperature T K is given by

$$K'_{ij} = \frac{\mu_j^2 N \rho_{ij} F_{ij}}{3 M_j k T} \left(\frac{\omega^2 \tau}{1 + \omega^2 \tau^2} \right) W_j \qquad \dots (2)$$

where μ_j is the dipole moment of polar solute of molecular weight M_j , N is the Avogadro number, k is the Boltzmann constant and $F_{ij} = \left(\frac{\varepsilon_{ij}+2}{3}\right)^2$ is the local field. Hence, it requires that the total conductivity should be

$$K_{ij} = \frac{\omega}{4\pi} \varepsilon'_{ij} \qquad \dots (3)$$

But for hf region it can be shown that

$$\varepsilon_{ij}' = \varepsilon_{ij,\,\infty} + \varepsilon_{ij}''/\omega\tau \qquad \dots (4)$$

where $\varepsilon_{ij,\infty}$ is the optical dielectric constant of the solution. From Eqs (3) and (4) the total hf conductivity of the solution takes the form

$$K_{ij} = K_{\infty} + K'_{ij} / \omega \tau_s \qquad \dots (5)$$

where K_{∞} is a constant and τ_s is the relaxation time of the solute molecule in solution. As K_{ij} is a function of W_j , we have from Eq. (5)

$$\left(\frac{\mathrm{d}K_{ij}}{\mathrm{d}W_j}\right)_{W_j\to 0} = \omega \tau_s \left(\frac{\mathrm{d}K_{ij}}{\mathrm{d}W_j}\right)_{W_j\to 0} = \omega \tau_s \beta \qquad \dots (6)$$

where β is the slope of $K_{ij} - W_j$ curve at $W_j \rightarrow 0$ the density ρ_{ij} of the solution becomes ρ_i , the density of the

solvent, and the local field F_{ij} of the solution is F_i = the local field of the solvent $=\left(\frac{\varepsilon_i+2}{3}\right)^2$. Under the circumstances, Eq. (2) on being differentiated with respect to W_j and as $W_j \rightarrow 0$ takes the form

$$\left(\frac{\mathrm{d}K'_{ij}}{\mathrm{d}W_j}\right)_{W_j \to 0} = \frac{\mu_j^2 N \rho_i F_i}{3M_j kT} \left(\frac{\omega^2 \tau_s}{1 + \omega^2 \tau_s^2}\right) \qquad \dots (7)$$

Taking

$$b = \beta \left/ \left(\frac{\mu_j^2 N \rho_i F_i}{3M_j kT} \cdot \omega \right) \right. \dots (8)$$

We have from Eqs (6) and (7)

$$b = \frac{1}{1 + \omega^2 \tau_s^2}$$

and whence

$$\tau_{s} = \frac{\lambda}{2\pi C} \sqrt{\frac{1}{b} - 1} = \frac{\lambda \times 10^{-10}}{18.84} \sqrt{\frac{1}{b} - 1} \qquad \dots (9)$$

Now from Eq. (8) the dipole moment of the solute molecule can be written in the form

$$\mu_{j} = \left(\frac{3M_{j}k\Gamma}{N\rho_{i}F_{i}} \cdot \frac{\beta}{\omega b}\right)^{1/2} \qquad \dots (10)$$

The Eq. (9) is used to compute b from the reported τ_s data and thence the dipole moment of polar solute $\mu_j^{(S)}$ when it is dissolved in a solvent. From Eq. (10), Müller⁷ gave a relation between $\mu_j^{(S)}$ and μ_j

$$\mu_{j}^{(S)} = \mu_{j} \left[1 - C(\varepsilon_{i} - 1)^{2} \right] \qquad \dots (11)$$

where $\mu_j^{(S)}$ is the dipole moment of the solute in a solvent and μ_j the dipole moment of the polar solute, C = 0.038 which is claimed to be a constant for a large

number of liquids, and ε_i the dielectric constant of the solvent.

Results and discussion—The dipole moments of seven systems of interest have been computed and listed in Table 1 with the reported μ_j data available from the literature for comparison. The μ_j data have also been computed from the bond dipole moment and bond angles, assuming that the benzene molecule is a planar one. The molecular parameters needed for this computation of theoretical values of μ_j are shown in Fig. 1.

The microwave conductivity data in the lowconcentration region are of much interest as they reveal a completely different situation in comparison to those of high-concentration region, as recently shown by Acharyya et al.⁸ This is the reason which led us to study the systems reported here and to calculate their dipole moments. High-concentration conductivity data of a polar-nonpolar liquid mixture is liable to yield the dipole moment due to dimer formation while those in low-concentration region behave linearly with the weight fraction and give the dipole moment due to monomer formation, i.e. solute-solvent association. The computed dipole moments agree with those reported experimentally as well as computed theoretical ones, as illustrated in Table 1. This fact suggests the basic soundness of the method adopted here.

The variation in the μ_j values for these substituted molecules in benzene might be due to (i) the difference in the values of group moments and (ii) inductive effect. The dipole moment of *o*-nitrobenzofluoride is always greater than that of *m*-nitrobenzotrifluoride (Table 1), both shown by the computed and theoretical μ_j 's due to the two different positions of the $-NO_2$ group in the

	Table 1	—The Compu	ited Values of	$(b, \mu_j^{\langle s \rangle}, \mu_j)$	(Compt), μ_j (F	Rept) at 35°	2
	System	b	$\tau_s \times 10^{12}$ (Rept)	µ́s ^{s)} (Compt)	μ _j (Compt)	μ_j (Rept)	ر (Theo)
			$\lambda = 3$.0166 cm			
	<i>m</i> -Aminobenzo- trifluoride	0.7234	9.9	3.1971	3.40	3.16	2.48
•	o-Nitrobenzo- trifluoride	0.6913	10.7	4.7332	5.03	4.54	6.18
	<i>m</i> -Nitrobenzo- trifluoride	0.6403	12.0	3.2138	3.42	3.33	3.74
	o-Chlorobenzo- trifluoride	0.7356	9.6	3.1062	3.30	3.15	3.98
			· λ =	3.0 cm			
	o-Chloronitro- benzene	0.5816	13.5	4.6137	4.91	⁵ 4.71	5.28
	4-Chloro- 3-nitrobenzo- trifluoride	0.7088	10.2	2.7084	2.88	3.17	3.78
	4-Chloro-3-nitro- toluene	0.3670	20.9	5.0899	5.41	4.88	5.58



Fig. 1—Conformation of the different molecules studied showing the orientation of the bond axes and the dipole moments and also bond moments: [1, *m*-Aminobenzotrifluoride; 2, *o*-nitrobenzotrifluoride; 3, *m*-nitrobenzotrifluoride; 4, *o*-chlorobenzotrifluoride; 5, *o*-chloronitrobenzene; 6, 4-chloro-3-nitrobenzotrifluoride & 7, 4-chloro-3-nitrotoluene]

compound. The other two compounds, viz. *m*-aminobenzotrifluoride and *o*-chlorobenzotrifluoride have smaller values of dipole moments due to the same reasons as mentioned earlier.

The group moment of $-CF_3$ group acts in the reverse direction to that of -Cl group and hence its net

effect is to reduce the total moment along the common axis. 4-Chloro-3-nitrobenzotrifluoride thus shows the lower value of μ_j in benzene than in ochloronitrobenzene. The group moment of $-CH_3$ group acts in the direction of -Cl group and hence the resultant dipole moment increases along the common axis. That is why the last compound in Table 1 and in Fig. 1, i.e. 4-chloro-3-nitrotoluene has higher dipole moment in benzene than in o-chloronitrobenzene. The close agreement between the computed μ_j values with the theoretical ones shows that the dipole moments of these compounds have a preferred conformation, as depicted in Fig. 1. Regarding the group moments assumed by us, they compensate each other to a certain extent.

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