

## DIPOLE MOMENTS AND THE STRUCTURE OF THE MOLECULES OF SOME OXADIAZOLE DERIVATIVES

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The molecular diagrams of 2-phenyl- and 2,5-diphenyl-1,3,4-oxadiazoles, their derivatives with chlorine, nitro, and dimethylamino substituents in the para positions, 3,5-diphenyl-1,2,4-oxadiazole, and 3,4-diphenyl-1,2,5-oxadiazole have been calculated by the MO LCAO method. In the heterocycle of a substituted oxadiazole there is not, as in aromatic rings, a single system of electrons present in conjugation (the  $\pi$ -electrons of the C=N bond and the pairs of nongeneralized electrons of the oxygen atom). The moment of the C=N bond in the heterocyclic compounds is 2.47 D and is directed from the carbon to the nitrogen. Considerable conjugation occurs between the  $\pi$ - or the p-electrons of the substituents in the phenyl rings of the diphenyloxazoles and the  $\pi$ -electrons of the C=N bonds of the heterocycle.

The question of the chemical structure (aromaticity of the 1,2,5-, 1,3,4-, and 1,2,4-oxadiazoles still remains undecided [1-3]. Since the dipole moments ( $\mu$ ) of molecules enables some information to be obtained on the distribution of the electrons in the heterocycle and on the interaction of the latter with  $\pi$ -electronic systems attached to it, it appeared of interest to consider the values of  $\mu$  of various oxadiazole derivatives. The importance of a systematic study of the dipole moments of heterocyclic compounds has recently been pointed out [4]. Nevertheless, up to the present time values of  $\mu$  have been known only for some dialkyl- and diaryloxadiazoles [5, 6]. In view of this, we have determined the values of  $\mu$  for 23 phenyl derivatives of 1,3,4-oxadiazole (mainly the 2,5-diphenyl derivatives) substituted in the phenyl rings. The experimental values  $\mu_e$  in D have been compared with those calculated both by the semi-empirical MO method and by a simple vectorial scheme.

## EXPERIMENTAL

The synthesis of the compounds studied and their purification (chromatographically on alumina or activated carbon) has been described previously [7]; the solvent—benzene—was prepared as described in the literature [8], and the method of measuring the dielectric constants and the densities of the solutions has been given previously [9]. The molar polarization at infinite dilution ( $P_{2\infty}$ ) was calculated by Hedestrand's method [10] and  $\mu_e$  from the formula  $\mu_e = 0.22(P_{2\infty} - P_{E+A})^{1/2}$ , where  $P_{E+A}$  was taken as  $1.05 R_D$  (where  $R_D$  is the molar refraction calculated by an additive scheme). The values of the coefficients  $\alpha$  and  $\beta$ ,  $P_{E+A}$ ,  $P_{OR}$ , and  $\mu_e$  are given in Table 1.

## Calculation of the Dipole Moments by the Molecular Orbitals Method

The distribution of the electron density and the mobile sequence of the bonds for seven of the oxadiazoles studied (Figs. 1 and 2) were calculated by the semi empirical MO method on a EVM-20 machine using Hückel's program. The coefficients  $b_X$  and  $K_{C-X}$  for the parameters  $\alpha_X = \alpha_C + b_X \beta_{C-C}$  and  $\beta_{C-X} = K_{C-X} \beta_{C-C}$  were taken from Orgel [11], Streitwieser [12], and Pullman [13], and for the nitro groups those proposed by Stone and Maki [14] were used, namely:  $b(-N=) = 1$ ;  $b(Cl) = 2$ ;  $K_{C-N} = 1.2$ ;  $K_{C-O} = 1.4$ ;  $b(-O-) = 3.2$ ;

$$b(O=) = 1.4; K_{C-Cl} = 0.4; K_{C-N} \leftarrow = 1.25; b(-N-) = 1.5; \\ b(-N \begin{array}{l} \nearrow O \\ \searrow O \end{array}) = 2.2; K_{N-O} = 0.6; K_{C-NO_2} = 1.2; K_{N \rightarrow O(NO_2)} = 1.67.$$

The influence of the heteroatoms on the carbon atoms attached to them was taken into consideration by means of an auxiliary induction parameter  $\delta_X = 0.1 \Sigma(\alpha_X - \alpha_C)$ . Table 2 gives the total electronic charges in the heterocycle and the phenyl groups and also the values  $\mu_\pi = e \Sigma_i r_i$  (where  $r_i$  is the radius vector of the i-th atom) calculated from the effective charges ( $q_i$ ). It was assumed that the heterocycle is a plane regular hexagon with all the bonds of the same length (1.40 Å); the length of the N-O bond in the nitro group was taken as 1.20 Å and that of the C-Cl bond as 1.80 Å. In the calculation of  $\mu_O$ , the following values and directions of the moments of the individual bonds were taken:  $\mu_{C-H} \leftarrow = 0.4$  D;  $\mu_{C-N} \rightarrow = 0.45$  D;  $\mu_{C-O} \rightarrow = 0.8$  D;  $\mu_{N-O} \rightarrow = 0.5$  D. The over-all moments was determined as the vectorial sum of  $\mu_O$  and  $\mu_\pi/1.6$  [11].

The calculated values of  $\mu_{tot}$  prove to be fairly close to  $\mu_e$ . Only in the case of 3,4-diphenyl-1,2,5-oxadiazole is there an appreciable deviation which is apparently connected with the nonconsideration of the steric effects between the two adjacent phenyl rings and the consequent impossibility of their arrangement in the same plane as the heterocycle. In the compounds studied with para substituents in the phenyl ring,  $\mu_e$  is always somewhat higher than  $\mu_{tot}$ , which may be connected with the nonconsideration of the electronic interaction of the p- and  $\pi$ -electrons of the substituents through the phenyl ring with the  $\pi$ -electrons of the heterocycle. The closeness of the calculated value of  $\mu_{tot}$  to  $\mu_e$  shows that the selection of Orgel's parameters (including the ratio  $\mu_\pi/1.6$ ) used for calculating the electron density distribution is evidently the most successful for substituted heterocycles as well, and the distribution itself is close to the true one [11]. As can be seen (Fig. 1 and Table 2), the introduction of phenyl groups into the oxadiazoles causes an appreciable change in the electron density distribution in the heterocyclic ring. Thus, in comparison with the electron distribution in the unsubstituted 1,3,4-oxadiazole, calculated by means of the same parameters [11], and its 2-phenyl derivative there is an increase in the negative charge on  $N_3$ , a decrease in the positive charge on the oxygen atoms and  $C_2$  with almost no change in the densities on  $N_4$  and  $C_5$ . At the same time, the total excess charge in the heterocycle of the 2-phenyl derivative has a negative value. The introduction of a second phenyl radical in position 5 causes an increase in the negative charge on  $N_4$ , a decrease in the positive charge on  $C_5$ , and a further small decrease in this charge on the oxygen atom, the total excess negative charge on the heterocycle increasing almost twofold in comparison with that calculated for the monophenyl derivative (Table 2). This effect of phenyl groups appears still more sharply in 3,4-diphenyloxadiazole as compared with the unsubstituted heterocycles [11]. In all the compounds, the total excess charge on the phenyl groups has a positive sign. Evidently, in the interaction of the  $\pi$ -electron systems of the phenyl radicals and the heterocycle the electron-accepting effect of the latter prevails, increasing with an increase in the number of nitrogen atoms directly attached to oxygen in the heterocycle. This was to be expected in view of the greater electronegativity of nitrogen than carbon in the C=N bond. At the same time, judging from the results for the 2-phenyl and 2,4-diphenyl derivatives, only one of the C=N bonds and not the whole  $\pi$ -electron system of the heterocycle enters into conjugation with each of the phenyl radicals; the p-electrons of the oxygen

Table 1  
Dipole Moments of Oxadiazole Derivatives

No.	Compound	$\alpha$	$\beta$	$P_{2\infty}$	$P_{E+A}$	$P_{Or}$	$\mu_E$	$\mu_{lit}$	$\mu_c$	$\Delta\mu$
1	2-Phenyl-1,3,4-oxadiazole	18.54	0.506	306.75	42.35	264.40	3.58	—	—	—
2	2,5-Diphenyl-1,3,4-oxadiazole	18.58	0.815	323.95	67.13	256.82	3.52	3.86 <sup>[5]</sup>	—	—
3	3,5-Diphenyl-1,2,4-oxadiazole	5.16	0.609	133.02	67.13	65.89	1.78	3.45 <sup>[6]</sup>	—	—
4	3,4-Diphenyl-1,2,5-oxadiazole	26.30	0.644	442.50	67.13	375.47	4.26	1.56 <sup>[5]</sup>	—	—
5	2-Bromomethyl-5-phenyl-1,3,4-oxadiazole	18.19	1.044	316.94	54.73	262.21	3.56	4.74 <sup>[5]</sup>	—	—
6	2-(4-Chlorophenyl)-5-phenyl-1,3,4-oxadiazole	16.39	0.978	298.51	72.78	225.73	3.30	4.20 <sup>[11]</sup>	—	—
7	5-Phenyl-2-(4-tolyl)-1,3,4-oxadiazole	21.19	0.607	373.43	72.52	300.91	3.81	—	3.39	-0.09
8	2-(4-Methoxyphenyl)-5-phenyl-1,3,4-oxadiazole	21.88	0.970	377.85	74.24	303.61	3.83	—	3.66	+0.15
9	2-(4-Nitrophenyl)-5-phenyl-1,3,4-oxadiazole	28.00	1.325	462.05	79.57	382.48	4.30	—	2.70	+1.13
10	2-(4-N-Dimethylaminophenyl)-5-phenyl-1,3,4-oxadiazole	38.85	1.210	624.29	80.96	543.33	5.13	—	4.45	-0.15
11	2-(4-Bromophenyl)-5-phenyl-1,3,4-oxadiazole	16.24	1.501	297.22	75.72	221.50	3.27	—	3.46	+1.67
12	2-(p-Biphenyl)-5-phenyl-1,3,4-oxadiazole	20.13	0.921	369.35	92.98	276.37	3.66	—	3.38	-0.11
13	2,5-Di(p-bromophenyl)-1,3,4-oxadiazole	8.93	2.426	186.84	83.76	103.08	2.23	—	—	—
14	2,5-Di(p-methoxyphenyl)-1,3,4-oxadiazole	24.42	1.176	419.15	86.24	332.91	3.99	—	—	—
15	2-( $\alpha$ -Naphthyl)-5-styryl-1,3,4-oxadiazole	24.42	1.109	436.64	95.89	338.75	4.04	—	—	—
16	2-(p-Biphenyl)-5-(p-dimethylaminophenyl)-1,3,4-oxadiazole	37.75	1.134	636.39	99.78	536.61	5.29	—	3.58	+1.71
17	5-(p-Dimethylaminophenyl)-2-( $\alpha$ -naphthyl)-1,3,4-oxadiazole	42.87	1.142	710.47	95.85	614.62	5.45	—	—	—
18	2-(p-Methoxyphenyl)-5-styryl-1,3,4-oxadiazole	25.32	0.485	452.00	83.45	368.55	4.22	—	—	—
19	2,5-Di(p-aminophenyl)-1,3,4-oxadiazole	30.79	0.861	493.84	76.72	417.12	4.58	—	—	—
20	2-(o-Chlorophenyl)-5-phenyl-1,3,4-oxadiazole	21.60	1.538	357.95	72.78	285.17	3.71	—	—	—
21	2-(o-Methoxyphenyl)-5-phenyl-1,3,4-oxadiazole	27.95	0.909	468.89	74.24	394.65	4.37	—	—	—
22	2-(m-Methoxyphenyl)-5-phenyl-1,3,4-oxadiazole	19.95	0.819	354.12	74.24	279.28	3.67	—	—	—
23	2-(o-Nitrophenyl)-5-phenyl-1,3,4-oxadiazole	22.32	1.066	386.45	79.57	306.88	4.85	—	—	—
24	2,5-Di(o-aminophenyl)-1,3,4-oxadiazole (benzene)	7.50	1.090	162.95	76.72	86.23	2.05	—	—	—
	2,5-Di(o-aminophenyl)-1,3,4-oxadiazole (dioxane)	9.32	1.154	177.56	76.72	100.89	2.22	—	—	—
25	2,5-Di(o-hydroxyphenyl)-1,3,4-oxadiazole (dioxane)	25.72	0.549	428.96	70.86	358.10	4.16	—	—	—

Table 2  
Total Charges and Moments of the Diphenyloxadiazoles

No.	Compound	Total electronic charge			$\mu_{\pi} \cdot D$ ( $\varphi^\circ$ )	$\mu_{\sigma} \cdot D$ ( $\varphi^\circ$ )	$\mu_{tot} \cdot D$ ( $\varphi^\circ$ )	$\mu_e$
		hetero-cycle	phenyl (1)	phenyl (2)				
1	2-Phenyl-1,3,4-oxadiazole	-0.0638	—	+0.0652	5.35 (10)	0.16 (10)	3.48 (10)	3.52
2	2,5-Diphenyl-1,3,4-oxadiazole	-0.1265	+0.0635	+0.0635	5.77	0.16	3.53	3.52
3	3,4-Diphenyl-1,2,5-oxadiazole	-0.2131	+0.1069	+0.1069	5.70	2.10	4.95	4.26
4	3,5-Diphenyl-1,2,4-oxadiazole	-0.2060	+0.0814	+0.1254	3.39 (71)	0.79 (71)	2.08 (172)	1.78
5	2-(p-Chlorophenyl)-5-phenyl-1,3,4-oxadiazole	-0.1220	+0.0587	+0.0640	5.25 (350)	0.48 (90)	3.01 (0)	3.30
6	2-(p-Dimethylaminophenyl)-5-phenyl-1,3,4-oxadiazole	-0.1387	+0.0764	+0.0624	6.68 (349)	1.20 (268)	4.70 (315)	5.13
7	2-(p-Nitrophenyl)-5-phenyl-1,3,4-oxadiazole	-0.1022	+0.0358	+0.0667	5.54 (60)	0.35 (17)	4.35 (36)	4.30

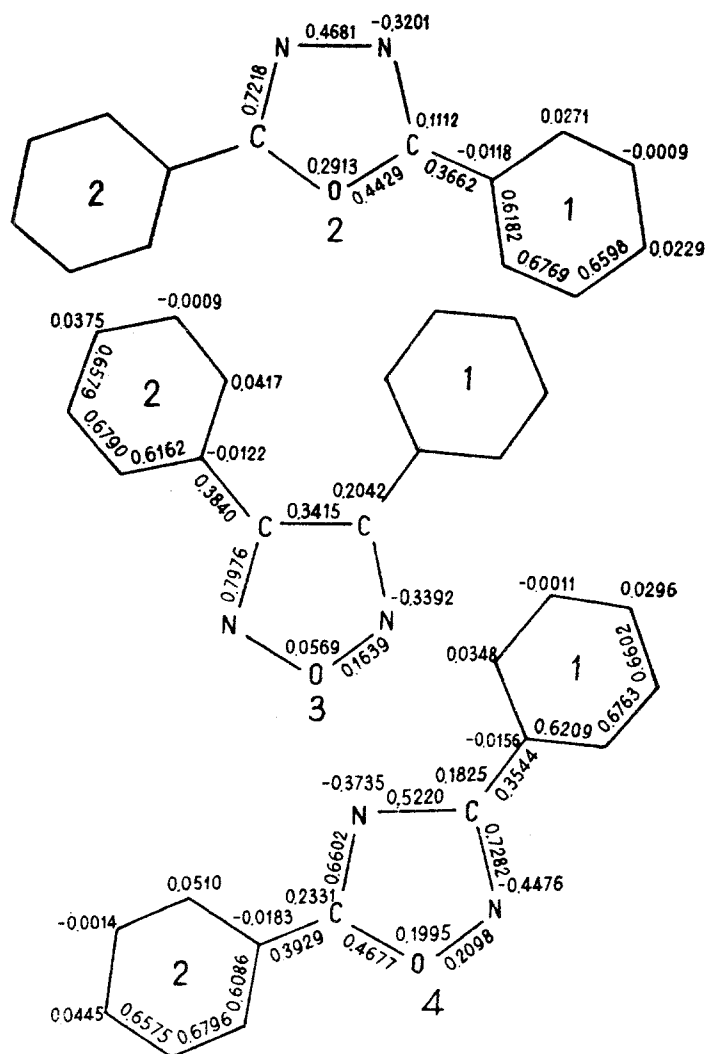


Fig. 1. Molecular diagrams of compounds 2, 3, and 4 (numbering given in Table 2).

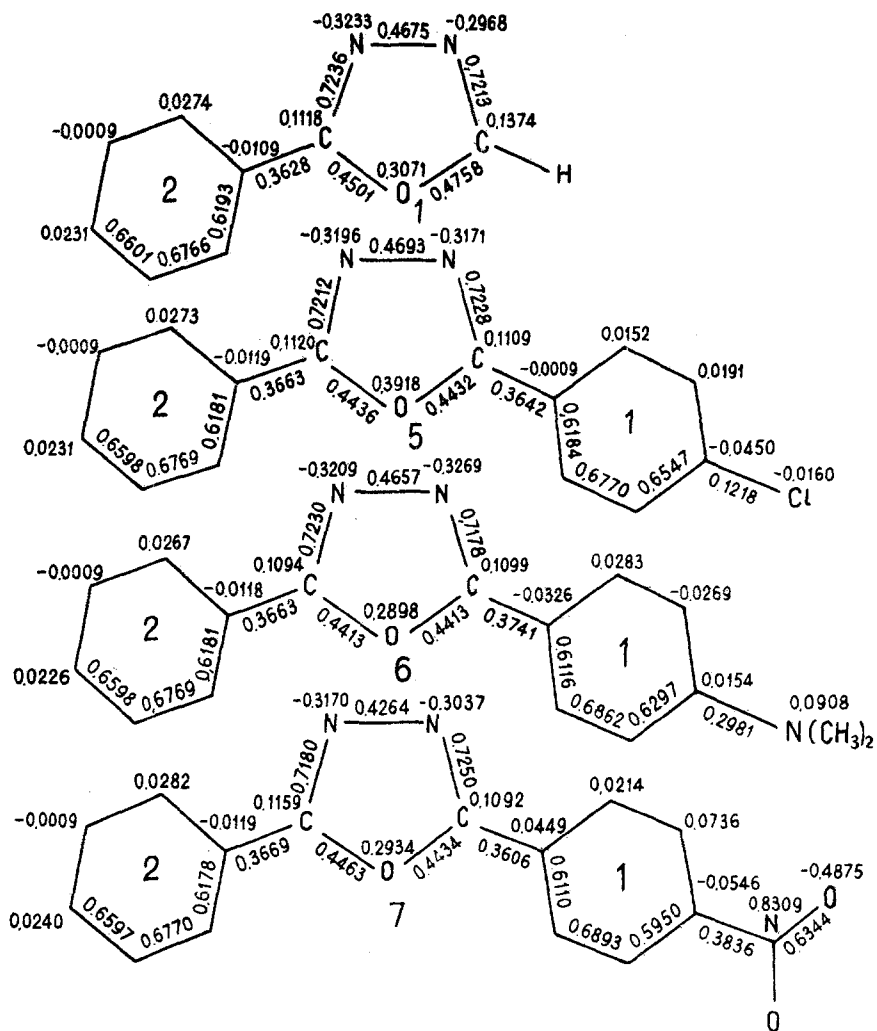


Fig. 2. Molecular diagrams of compounds 1, 5, 6, and 7 (numbering given in Table 2).

take only a very slight part in the conjugation considered between the  $\pi$ -electrons of the C=N bonds and the  $\pi$ -electronic systems of the phenyl groups.

The introduction into the para position of one of the phenyl groups of 2,5-diphenyl-1,3,4-oxadiazole of electron-accepting substituents (Cl, NO<sub>2</sub>) causes a decrease in the total negative charge on the heterocycle and the total positive charge in that phenyl radical that contains the substituents (Fig. 2); in the unsubstituted phenyl group, on the oxygen atom of the heterocycle, and on the atoms of the C=N bond adjacent to this phenyl radical (N<sub>4</sub> where there is a p-nitrophenyl group in position 2), the charge either does not change at all or undergoes an insignificant change. However, when an electron-donating substituent is introduced into the para position of one of the phenyl radicals, the opposite effect is observed; the excess negative charge on the heterocycle and positive charge on the phenyl group bearing the substituent (a dimethylamino group) rise, and the distribution of the charges in the unsubstituted phenyl group remains unchanged, while on the atoms of the heterocycle connected with this C<sub>6</sub>H<sub>5</sub> group there is only an insignificant change in charge. As can be seen, between the different phenyl rings of the compounds considered there is no appreciable  $\pi$ -electronic interaction whatever through the heterocycle. The nature of the influence of the phenyl groups and the substituents on the distribution of the electron density mentioned above shows the absence from the heterocycles of the oxadiazoles of an equalization of the bonds which is characteristic for aromatic rings. The same thing has been established in the case of 1,2,4-oxadiazole on the basis of the NMR and UV spectra [2].

#### Vector Analysis of the Dipole Moments

The values of  $\mu$  for the compounds studied show, first of all, the fairly high resultant moment of the heterocycle itself in the case of 1,3,4- and 1,2,5-oxadiazoles and the considerable decrease in this moment in 1,2,4-oxadiazole [1,5]. This is also confirmed by the data for  $\mu$  of unsubstituted furazan (1,2,5-oxadiazole) [3] and likewise by the considerable increase in the dipole moments of the derivatives substituted in the phenyl rings above the moment of the corresponding monosubstituted benzenes. Apparently, the dipole moments of the compounds considered are determined mainly by the moment of the C=N bond, which is directed from the carbon to the nitrogen. In the case of the 1,3,4- and 1,2,5-oxadiazoles, the vectors of the moments of the two C=N bonds are added, and in 1,2,4-oxadiazole they almost

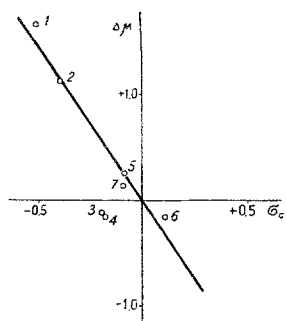


Fig. 3. Correlation of  $\Delta\mu$  and  $\sigma_c$  for compounds of the series C1=CN2C(=O)N=C2C1-c1ccc(X)cc1: X =: 1) N(CH<sub>3</sub>)<sub>2</sub>; 2) OCH<sub>3</sub>; 3) Br; 4) Cl; 5) CH<sub>3</sub>; 6) NO<sub>2</sub>; 7) C<sub>6</sub>H<sub>5</sub>.

compensate one another and the value of  $\mu$  for the heterocycle is close to that of the compound with the ether grouping C—O—C. From this point of view it also becomes clear why  $\mu$  for 1,2,5-oxadiazole is higher than for 1,3,4-oxadiazole in which the direction of the vector of the moment of the C=N bond is opposite to that of the moment of

the C—O—C grouping. The resultant moment in 1,3,4-oxadiazole must be directed towards the nitrogen atoms and in 1,2,5-oxadiazole towards the oxygen atom; in view of the symmetry of these molecules (C<sub>2v</sub>), the vector of the resultant moment lies on the axis of symmetry passing through the center of the oxygen atom and the middle of the N—N or C—C bond.

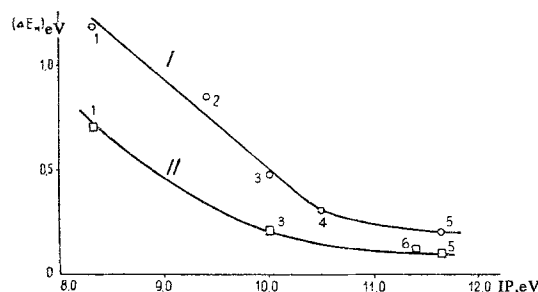


Fig. 4.  $\Delta E_m$  of compounds of the series C1=CN2C(=O)N=C2C1-c1ccc(X)cc1 on the ionization potential of CH<sub>3</sub>X: X =: 1) N(CH<sub>3</sub>)<sub>2</sub>; 2) NH<sub>2</sub>; 3) OCH<sub>3</sub>; 4) Br; 5) CH<sub>3</sub>; 6) Cl.

The value of the moment of the C=N bond was calculated from the values of  $\mu$  for 2,5-diphenyl-1,3,4-oxadiazole on the basis of the assumption that the heterocycle is a regular plane pentagon with an internal angle between the bonds of 108° (for furazan, this has been shown on the basis of its microwave spectrum [3]); the moments of the individual bonds were taken as:  $\mu_{\text{C}_{\text{heter}}-\text{C}_{\text{phen}}} = 0.4$  D;  $\mu_{\text{C}-\text{H}} = 0.4$  D;  $\mu_{\text{C}-\text{O}} = 0.8$  D; for  $\mu_{\text{C}_{\text{heter}}-\text{C}_{\text{phen}}}$ , in agreement with the calculated electron density distribution the vector is directed from C<sub>heter</sub> to C<sub>phen</sub>. The value of  $\mu_{\text{C}=\text{N}}$  proved to be 2.47 D. The values of  $\mu$  for the molecules of 3,4-diphenyl-1,2,5-oxadiazole and 3,5-diphenyl-1,2,4-oxadiazole calculated with this value of  $\mu_{\text{C}=\text{N}}$  (with the assumption that  $\mu_{\text{C}-\text{N}} = 0.45$  D and  $\mu_{\text{N}-\text{O}} = 0.5$  D) and also the values for the molecules of such heterocycles as pyrazole, imidazole or 4,5-diphenylimidazole, etc. ( $\mu_{\text{N}-\text{H}} = 1.65$  D) agree satisfactorily with the observed values [15]. The proposed value of  $\mu_{\text{C}=\text{N}}$  almost coincides with or is close to those given in some other papers [16–18], but is considerably higher than the calculated moment of this bond in aliphatic aldimines [19].

The values of  $\mu$  for 2,5-diphenyl-1,3,4-oxadiazoles substituted in one of the phenyl groups show, in contrast to that given in [2], a considerable interaction between the  $\pi$ -electrons of the C<sub>6</sub>H<sub>5</sub> group and the heterocycle. This can be seen particularly clearly from the fact that the values of  $\mu$  for the dimethylamino and nitro derivatives are considerably higher than the vectorial sum of the moments of the initial compound and dimethylaniline or nitrobenzene, as the case may be. This interaction can be evaluated quantitatively from the values  $\Delta\mu = \mu_c - \mu_e$ , where  $\mu_c$  is the moment of the molecule calculated from the figures for the components along the X and Y axes of the moments of 2,5-diphenyloxadiazole and the corresponding monosubstituted benzene. The calculation has been carried out only for the para isomers with values of  $\mu_{\text{C}_6\text{H}_5\text{X}}$  for X = Cl, Br, CH<sub>3</sub>, NO<sub>2</sub>, OCH<sub>3</sub>, and N(CH<sub>3</sub>)<sub>2</sub> of, respectively, 1.58, 1.56, 0.4, 4.00, 1.30, and 1.50 D, and for the two latter substituents X the angles of inclination to the plane of the benzene ring were taken from the literature [20, 21] as 67° and 33° C, respectively. The calculated values  $\mu_c$  and  $\Delta\mu$  are given in Table 1. As can be seen, in the case of electron-donating groups  $\Delta\mu$  falls in the sequence N(CH<sub>3</sub>)<sub>2</sub> > OCH<sub>3</sub> > CH<sub>3</sub>, and for electron-accepting groups in the sequence NO<sub>2</sub> > Hal; this agrees with the capacity of the electrons of the groups mentioned for participating in conjugation with the  $\pi$ -electronic system of the phenyl ring. Another confirmation of this is the fairly good correlation of the values of  $\Delta\mu$  with Taft's constants for the functional groups mentioned (Fig. 3) [22]. The existence of conjugation between the phenyl groups and the heterocycle follows from the nature of the change in the other prop-

erties of the compounds studied when the substituents in the phenyl groups are changed; for example, the change in the value of  $\Delta E_M$  (in eV) of the absorption band with the maximum in the case of 2-phenyloxadiazole at 245 nm [23] with a change in the nature of the electron-donating substituent (X) is subject to a general law, namely it falls with a rise in the ionization potential of  $CH_3X$  (Fig. 4) [24].

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