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UDC 547.77:541.67

Data on the dipole moments of azoles are correlated and systematized. The possibilities of the method of dipole moments in the solution of a number of problems of the chemistry of azole-containing systems – the electronic structures of azoles, intermolecular association, tautomeric and conformational equilibria, and reactivities – are demonstrated. The experimental values of the dipole moments of azoles are tabulated.

Azoles are five-membered heteroaromatic systems that contain two or several heteroatoms, one of which is an unsaturated nitrogen of the pyridine type. Azoles command the unflagging interest of chemists, biologists, and medical men in connection with the exceptional role of a number of their derivatives in the most important biochemical processes and in connection with their extensive use as physiologically active preparations and medicinals.

The results of investigations of the physicochemical properties, structures, and reactivities of azole molecules have been correlated in a number of reviews [1-3], which, however, because of the breadth of coverage of the material, are frequently sketchy. In this connection, a more detailed illumination of the individual problems associated with modern aspects of the physical chemistry of azole molecules seems extremely useful.

This paper is a correlation of primarily our investigations on the use of the method of dipole moments in the chemistry of azole-containing systems. Without dwelling upon the method itself, which has been rather fully illuminated in a monograph [4], we have strived, in the case of azoles, to show the possibilities of the method of dipole moments in the solution of a number of problems common to the chemistry of nitrogen-containing heterocyclic compounds.

Calculations of the Dipole Moments for Azole Molecules

The interpretation of the data obtained as a result of the determination of dipole moments is usually based on a comparison of the experiment (μ_{exp}) and calculated (μ_{cal}) values.

The calculation of the dipole moments of azole molecules, which involves the use of the bond and group moments adopted in the literature [4], is extremely crude and at best gives only approximate results. This is explained by the significant change in the bond moments as a result of mutual polarization and by the necessity, in a number of cases, to separately take into account the atomic dipole of the unshared pair of electrons of nitrogen. In this connection, it is better to use the moments of the individual fragments of the molecule for the vector calculation of the dipole moments of azoles.

In [5-10] and, subsequently in [11], the dipole moments of pyrazole and triazole molecules and their derivatives were calculated as the vector sums of components of two types, one of which includes a pyridine nitrogen atom, the other of which includes a pyrrole nitrogen atom. The data set forth in [6-11] sufficiently rigorously explain the facts of the satisfactory convergence of the μ_{exp} and μ_{cal} values for a number

Rostov-on-Don State University. Institute of Organic Synthesis, Academy of Sciences of the Latvian SSR, Riga. K. A. Timiryazev Moscow Agricultural Academy. Translated from *Khimiya Geterotsiklicheskikh Soedinenii*, No. 7, pp. 867-892, July, 1971. Original article submitted October 5, 1970.

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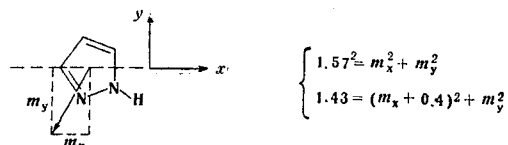
TABLE 1. Dipole Moments of Pyrazole Derivatives

Compound	μ_{exp}, D	$\mu_{\text{cal}}, D,$ from [11]	$\mu_{\text{cal}}, D,$ from [5-8]
1-(4-Pyridyl)-3,5-dimethylpyrazole	2,84	3,14	2,59
1-Phenyl-3-methyl-5-chloropyrazole	1,07	1,08	0,68
1-(4-Pyridyl)-3-ethyl-4-methyl-5-aminopyrazole	3,90	3,92	3,57
1-Phenyl-3-aminopyrazole	1,85	1,93	1,44

TABLE 2. Dipole Moments of Imidazole Derivatives

Compound	$\vec{\mu}_{\pi}$	$\vec{\mu}_{\sigma}$	$\vec{\mu} = \vec{\mu}_{\pi} + \vec{\mu}_{\sigma}$	μ_{cal}, D	μ_{exp}, D
N-Methylimidazole	$-0,80i + 3,35j$	$0,54i + 0,43j$	$-0,35i + 3,78j$	3,80	3,80
N-Phenylimidazole	$-0,59i + 2,71j$	$0,31i + 0,80j$	$-0,28i + 3,51j$	3,52	3,14
N-Methylbenzimidazole	$-2,22i + 3,09j$	$0,45i + 0,43j$	$-1,77i + 3,52j$	3,94	4,04
4,5,6,7-Tetrahydro-N-methylbenzimidazole	$+3,55j$	$0,45i + 0,43j$	$0,45i + 3,98j$	3,99	4,00
3-Ethyl-naphthimidazole	$-2,20i + 3,31j$	$0,45i + 0,43j$	$-1,75i + 3,74j$	4,13	4,04
1-Methylnaphthimidazole	$-2,20i + 2,98j$	$0,45i + 0,43j$	$-1,75i + 3,41j$	3,83	3,86
1-Ethylphenanthrenimidazole	$-1,67i + 3,12j$	$0,45i + 0,43j$	$-1,22i + 3,55j$	3,76	4,11
1-Ethyl-4,5-diphenylimidazole	$-1,49i + 3,27j$	$0,45i + 0,43j$	$-1,04i + 3,71j$	3,86	4,11

of unsubstituted azoles and azole-containing compounds with substituents that do not have significant polarizing action [12]. The presence of highly polar substituents, however, causes an appreciable redistribution of the electron density in the heterocycle molecule. The difference that arises between μ_{exp} and μ_{cal} , which is considered to be the interaction moment (μ_{int}), can give valuable information regarding the electronic interaction in molecules of substituted azoles but simultaneously limits the possibilities of the rigorous application of the method for the solution of problems of the fine structure of substituted azoles and bisheterocyclic systems. It should, however, be noted that this imperfection is organically inseparable from any vector method of calculation that uses the moments of bonds, groups, or entire fragments as constants. In this respect, the method of vector calculation of dipole moments proposed by Eda and Ito [13] has the greatest advantages. This method is based on finding the direction of the resulting moment of a parental compound by means of a comparison of its experimental dipole with the moment of an analogous derivative that differs by one regular group (CH_3 , F, NO_2). A considerable advantage of this method is the possibility of finding the resulting vector of a fragment that already includes one or more substituents. The use of this value for the calculation of molecules with additionally introduced polar groupings will naturally lead to considerably smaller errors in the calculated μ values. An illustration of this method is provided by the calculation of the direction of the vector of the pyrazole molecule carried out with the use of the dipole moments of pyrazole (1.57 D) and its 3-methyl derivative (1.43 D) [11]. The solution of a system of equations relative to the projections of the moments on the axes of coordinates (m_x and m_y), accompanied by the selection of the rational sign of the projection of m_y , gives the direction of the total vector of the pyrazole molecule in the selected system of coordinates: $m_x = -0.73$, $m_y = -1.39$.



The acceptability of this method for the calculation of the dipole moments of azoles is confirmed by the data in Table 1.

A method that differs fundamentally from the simple methods of calculation is the method based on the computation of the dipole moment from the electronic distribution calculated by means of quantum-mechanical methods. Among the most promising methods here is the Pople-Segal CNDO (complete neglect of differential overlap) method [14], in which the dipole moment is calculated with allowance for the σ and

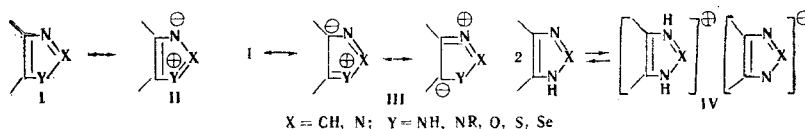
π components and the atomic dipoles. The extended Hückel method, which was used for the calculation of the dipole moments of azoles by Adam and Grimison [15] has, in principle, similar possibilities but, in the absence of self-consistency with respect to charge, it leads to markedly elevated μ values. Difficulties caused by the necessity of individual allowance for the σ moment arise in the use of other schemes that involve the σ, π approximation.

The σ moment is calculated on the basis of the moments of analogs that do not have double bond or by vector addition of tabulated σ moments. However, more accurate σ moments can be obtained by the Del Re method [16] proceeding from the magnitudes of the σ charges. A third possible approach to the calculation of the σ moment was realized by Brown and Coller [17], who proposed that only the contributions from the atomic dipoles of unshared electron pairs of the heteroatoms be taken into account.

A graphical illustration of the successful application of the MO method for the calculation of the dipole moments of azoles is provided by the data in Table 2 [18]. As another example, one can adduce the results of calculations of the dipole moments of 1-phenylimidazole, 1-phenylpyrazole, and 1-phenyltriazole [19]. The calculations made it possible to determine the magnitudes and directions of μ_{π} , μ_{σ} , and the total moment, as well as the magnitude of the additional π moment (μ_{int}^{π}) arising as a result of conjugation of the N-phenyl and heterocyclic rings. The resulting μ_{int}^{π} value (0.5-0.7 D) is in good agreement with the experimental estimates.

Dipole Moment and Electronic Structure of the Azole Ring

In early papers [22-24] the deviations of the experimentally found dipole moments of azoles from the values calculated for classical formulas (I) were explained by intramolecular structures II-III:



Finally, an ionogenic structure of the salt type (IV) was considered possible for imidazoles with unsubstituted NH groups on the basis of a study of the IR spectra [25]. Thus, considerable charge separation, which, in the case of IV went all the way to the development of ion pairs, was proposed in all of the studies pointed out above.

On the basis of a determination of the dipole moments of systems containing imidazole [26-28], pyrazole [5, 29], and 1,2,4-triazole rings [6], we expressed the assumption that the deviations in the experimentally found μ values from the additive values are actually associated with polarization of the azole ring in the direction of the C=N bond; however, the degree of polarization is far from that of complete charge separation. The latter is in agreement not only with the magnitudes of the dipole moments (2-4 D) but also with the results of quantum-mechanical calculations [18, 19]. The concepts regarding the development of ion pairs, however, do not stand up under criticism in view of the practical coincidence of the moments of N-unsubstituted and N-alkylated azoles (salt formation of the IV type is not realizable for the latter) [26,27]: 3.99 D for imidazole and 3.76 D for N-methylimidazole; 2.06 D for pyrazole and 2.28 D for N-methylpyrazole; 3.29 D for 1,2,4-triazole and 3.50 for its N-methyl derivative; 4.07 D for benzotriazole and 4.16 D for its N-methyl derivative, etc. [30].

In connection with what was pointed out above, one should bear in mind that even in the case of meso ionic compounds (sydnones [24, 34], nitrones [24,30], and antipyridines [29]), dipole moments of 7-9 D are not in agreement with the concepts regarding complete localization of the negative charge on the exocyclic atoms. The results of quantum-mechanical calculations that we performed for antipyridines [29] and their thio [32] and imino [29] analogs and that Bochvar [33] made for sydnones lead to the same conclusion.

The dipole moments of alkyl and aryl derivatives of azoles are determined principally by the polarities of the azole rings [6, 26, 27], although they also depend on the position and nature of the substituents in the heterocyclic ring. The dipole moments of most C- and N-methyl-substituted azoles differ by 0.3-0.4 D from the moments of the corresponding unsubstituted heterocycles. However, in the case of benzimidazole, 2-methylbenzimidazole, and 5(6)-nitrobenzimidazole and its 2-methyl derivative, this difference amounts to 1 D [28], which is probably caused by σ, π conjugation of the CH_3 group with the benzimidazole ring.

TABLE 3. Interaction Moments in Pyrazole Derivatives

No.	Compound	$\mu_{\text{exp}}^{\text{D}}$	$\mu_{\text{cal}}^{\text{D}}$	$\mu_{\text{int}}^{\text{D}}$
1	1-Phenyl-3-aminopyrazole	1,85	1,44	+0,41
2	1-Phenyl-3-chloropyrazole	3,76	3,49	+0,27
3	1-Phenyl-5-aminopyrazole	3,34	3,40	0
4	1-Phenyl-5-chloropyrazole	1,25	0,51	+0,74
5	1-Phenyl-5-bromopyrazole	1,33	0,49	+0,84
6	1-Phenyl-4-aminopyrazole	2,36	2,68	-0,32
7	1-Phenyl-4-chloropyrazole	2,28	2,38	-0,1
8	1-Phenyl-4-bromopyrazole	2,28	2,37	-0,09
9	3,5-Dimethyl-4-nitropyrazole	3,88	3,54	+0,34
10	3,5-Dimethyl-4-nitrosopyrazole	4,66	3,06	+1,60
11	1-(p-Nitrophenyl)pyrazole	4,40	2,36	+2,04
12	1-(p-Aminophenyl)pyrazole	2,97	3,00	0
13	1-(p-Hydroxyphenyl)pyrazole	2,71	2,72	0

Phenyl groups joined to the C atoms or condensed at the 4- and 5-positions of the heterocyclic rings also have only a slight effect on the dipole moments of azole-containing systems. The dipole moments of azoles and benzazoles [26, 30] and their C-alkyl and C-aryl derivatives [10, 21, 26] practically coincide. In the case of N-phenyl-substituted azoles, however, the dipole moments are lower than those for the corresponding unsubstituted or N-methylated heterocycles [19-21]. This result is associated with the development of an additional π moment, which is directed along the N-Ph axis and is subtracted from the total moment of the heterocyclic portion of the molecule [21].

Shifting of the extremely labile π -electron system of the azole rings under the influence of a polarizing substituent evokes deviations of the experimental dipole moments of azoles from the vectorially calculated values can be considered to be the "interaction dipole moment" (μ_{int}). The data in Table 3 indicate that substituents that have a +C effect (the amino group and halogens) in the 3- and 5-positions of the pyrazole ring give rise to positive μ_{int} values (compounds 1, 2, 4, and 5). In the 4-position of the pyrazole ring, substituents that have a +C effect, in contrast to the same substituents in the 3- and 5-positions, produce negative μ_{int} values (Table 3, compounds 6-8), while substituents that have -C effects (nitro and nitroso groups) give rise to positive μ_{int} values (Table 3, compounds 9 and 10). A high electron density on the C₄ carbon atom, as compared with the C₃ and C₅ atoms, apparently appreciably hinders migration of the electrons from an electron-donor substituent to the ring and promotes their migration from the azole ring to electron-acceptor substituents.

A conclusion regarding the high polarizability of azole rings (which leads to a change in the direction of the moment toward the nitro group) was drawn on the basis of the dipole moments of C- and N-nitrophenyl-substituted azoles [2, 6, 21, 28]. The experimental dipole moment for a molecule of compound 11 (Table 3) is 2 D higher than the calculated value, while the polarization is so strong that the center of negative charges is shifted toward the nitrophenyl substituent. Similar polarization of the azole ring is also observed for C- and N-nitrophenyl derivatives of imidazole [21], benzimidazole [26, 27], and 1,2,4-triazole [10].

Polar Properties and Intermolecular Interaction

Disruption of the linear dependence of the dielectric permeability of azole solutions on concentration ($\epsilon = \epsilon_1 + \alpha c$) attests [4, 37] to association, which is usually accomplished through intermolecular hydrogen bonding (IHB). The graphical function $\epsilon = f(c)$ in this case is curvilinear and is accompanied by either a symbatic dependence of the α coefficient on c (for a more polar polymer) or by an antibatic dependence (if the polymer is less polar than the monomer) [4]. When association is absent, the α coefficient is independent of c .

Significant deviations of the concentration dependence of the dielectric permeability from linearity are observed for the majority of azoles that contain unsubstituted NH groups: imidazole [27] and its derivatives [26, 28], pyrazole [5,6], 1,2,4-triazole [10], etc. [30]. Positive deviations in $\epsilon = f(c)$ are observed for non-alkylated imidazoles, while negative deviations are observed for pyrazoles. These differences in the behavior of the indicated heterocycles are explained by the fact that imidazole forms highly polar, linear associates, while pyrazole forms nonpolar, cyclic dimers and trimers [34, 35]. The dipole moments that Hückel found for imidazole (5.58 D) [22] and pyrazole (1.54 D) [30] are therefore considered to be erroneous; according to later results, they are 3.99 D [26, 27] and 2.06 D [5], respectively. In contrast to these hetero-

cycles, 4,5-diphenylimidazole [26] and naphth[1,2-d]imidazole [28] are not associated in dioxane solution: this is indicated by the linear character of the $\epsilon = f(c)$ dependence.

The determination of the dipole moments made it possible to express ideas regarding the reasons for the association of azole-containing systems with unsubstituted NH groups [26, 36].

While rather convincing evidence for the association of azoles through IHB of the NH . . . N type is presented in [26, 27, 36, 37], Otting [25], on the basis of the results of an investigation of IR spectra, considered that the development of ionogenic pairs (IV) and electrostatic intermolecular interaction were possible. As pointed out in the previous section, the latter point of view is completely refuted, not only by the comparatively low dipole moments of azole-containing systems, but also by the low polarities of unsubstituted and N-alkylated azoles [26-28]. The results of our investigation of the slope of the dielectric losses of benzene solutions of imidazole, benzimidazole, and their N-alkyl substituted derivatives are directly related to what has been stated above. The high dielectric losses (losses in conductivity) of unsubstituted azoles can be explained by the chain character of the association and by proton migration along a chain of H bonds.

Dipole Moments and Tautomerism of Azole-Containing Systems

The determination of the dipole moments gives satisfactory results in the solution of the problems of tautomerism only in the case of a considerable difference in the calculated moments of the individual tautomers. In connection with the inaccuracies in the computed values, it is expedient that they be replaced by the experimental values of dipole moments found for fixed tautomeric forms.

Meso hydrogen tautomerism (protomerism), associated with the fact that the proton is not fixed on a definite N atom of the azole ring but migrates between them, is postulated for azoles with unsubstituted NH groups. In addition, it is known that isomers of compounds that contain imidazole [1] and pyrazole [3,38] rings in a number of cases react only in one of the tautomeric forms. Moreover, protomers [39] can be separated for the hard-to-dissolve imidazole derivatives. These facts indicate the possibility of localization of the proton on one of the N atoms of the azole ring.

To verify the latter conclusion, we determined the dipole moments of a number of 3,4-disubstituted and 3,4,5-trisubstituted pyrazoles [7] and bis(3,5-dimethyl-4-pyrazolyl) [8]. The experimentally found moment of 3,4-dibromopyrazole attests to the fixation of the proton to the N₁ atom of the pyrazole ring. A similar result was also obtained for 3,4,5-trisubstituted pyrazoles and for bis(3,5-dimethyl-4-pyrazolyl) [8].

Thus the dipole moments of fixed tautomeric forms of C-substituted derivatives of pyrazole attest to the existence of a structure with a proton localized on only one of the N atoms of the pyrazole ring. Investigations of the polar properties of 1,2,4-triazole [10], whose protomers have different dipole moments, lead to the same conclusion. Here, however, we note that the predominant realization of the 1H-isomer of 1,2,4-triazole is characteristic only for nonpolar media. In polar solvents, the tautomeric equilibrium of the 1H \rightleftharpoons 4H forms is shifted to the right [40].

The method of dipole moments proved to be useful in the discussion of the keto-enol [29, 41], thione-thiol [41], and amine-imine [10] tautomerism that is extremely characteristic in a number of azole-containing compounds. The vectorially calculated μ values of the tautomers of benzothiazole derivatives differ substantially from one another: the hydroxy and oxo forms have μ_{cal} values of 2.4 and 3.8 D, respectively, while those for the thiol and thione forms are 2.5 and 4.2 D [41]. The experimental values (4.2 and 4.6 D) obtained for the O- and S-derivatives attest to a shift of the equilibrium to favor the keto forms. However, an increase in temperature in this case leads to an appreciable decrease in the μ values and indicates a shift of the equilibrium to favor the hydroxyl and thiol forms [41].

A study of the polar properties of pyrimidines makes it possible to suppose that the highly polar antipyrine structure (VII) [29] participates, in addition to the enol (V) and keto (VI) structures, in the keto-enol equilibrium:

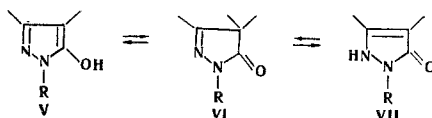


TABLE 4. Experimental Values of Dipole Moments of Azole-Containing Molecules

I		2		1		2	
Empirical formula and compound name*		Dipole moment and cond. under which it was deter.†		Empirical formula and compound name*		Dipole moment and cond. under which it was deter.†	
$\text{C}_2\text{H}_2\text{N}_4$ 1,2,3,4-Tetrazole		5.14, D, 25° 23, 5.11, 62 MW 62		$\text{C}_2\text{H}_2\text{N}_4$ 1-Methyl-1,2,3,4-tetrazole		5.38, B, 25° 23	
$\text{C}_2\text{H}_2\text{N}_4\text{S}$ 5-Amino-1,2,3,4-thiaziazole		5.77, D 63		5-Methyl-1,2,3,4-tetrazole		5.0, B, 25° 71	
$\text{C}_2\text{H}_2\text{N}_5$ 5-Amino-1,2,3,4-tetrazole		5.71, D 25° 72, 23		3-Amino-1,2,4-Triazole		3.38, D, 25° 10	
$\text{C}_3\text{H}_2\text{O}_2$ 1,2,4-Oxadiazole		1.2±0.3, MW 64		$\text{C}_2\text{H}_4\text{N}_4\text{S}$ 5-(N-Methylamino)-1,2,3,4-thiaziazole		5.72, D 83	
1,2,5-Oxadiazole		3.38±0.04, S 65		$\text{C}_3\text{HN}_2\text{Br}_3$ 3,4,5-Tribromopyrazole		2.48, B 25° 7	
1,3,4-Oxadiazole		3.04±0.04, MW 66		$\text{C}_3\text{H}_2\text{ONBr}$ 4-Bromoisoxazole		1.88, B, 25°, 1.99, D, 25° 75	
$\text{C}_3\text{H}_2\text{N}_6\text{S}$ 1,2,5-Thiadiazole		1.656±0.015, S 67		$\text{C}_3\text{H}_2\text{N}_2\text{Br}_2$ 3,4-Dibromopyrazole		4.66, B, 25° 8	
1,3,4-Thiadiazole		3.28±0.03, MW 68		$\text{C}_3\text{H}_3\text{ON}$ Isoxazole		1.5, MW 73	
$\text{C}_3\text{H}_2\text{N}_2\text{Se}$ 1,2,5-Selenadiazole		1.11±0.03, MW 69		Oxazole		2.81, B, 25° 23.	
$\text{C}_3\text{H}_2\text{N}_3\text{Cl}$ 3-Chloro-1,2,4-triazole		4.25, D 25° 10		Thiaziazole		2.76, B, 25°, 3.01, D, 25° 76;	
$\text{C}_2\text{H}_3\text{ON}_3$ 1,2,4-Triazol-5-one		3.30, D 25° 23		Isothiazole		2.90, MW 79	
$\text{C}_3\text{H}_3\text{N}_3$ 1,2,3-Triazole		1.77, B 25° 23		$\text{C}_3\text{H}_3\text{NS}$ Thiazole		1.64, B, 25° 23; 1.41, MW 2.4±0.2, MW 78	
1,2,4-Triazole		3.23, D, 20°; 3.16, D, 50°;		Imidazole		6.21, B, 50°; 6.17, B, 60°;	
		3.47, D, 96° 70; 3.24, D, 25° 22; 3.17, D 25° 23;				6.16, B, 70°; 4.84, D, 20°;	
		3.29, D, 25° 10				4.78, D, 50°; 4.58, D, 90° 70; 6.2, B, 25°; 4.49,	

* The compounds are arranged with respect to empirical formulas of increasing complexity; the elements in each formula are given in the following order: C, H, O, and N (C1, Br, I, S, and metals are in alphabetical order).

† B is benzene, D is dioxane, C is carbon tetrachloride, CH is chloroform, CY is cyclohexane, MW is the microwave method, and S is the Stark method.

TABLE 4 (continued)

	1	2	1	2
Pyrazole		D, 25° 23; 3,99, D, 25° 26, 27; 3,8 ± ±0,4, MW 64, 3,4, B 33	C ₄ H ₆ N ₂ 1,3-Dimethyl-5-aminotetrazole	4,02, B, 25° 31
C ₃ H ₄ N ₂ S 2-Aminothiazole		1,46, B, 20°; 1,64, B, 50°; 1,79, B, 70°; 2,19, D, 20°; 1,32, CY 70° 70; 1,47, B, 25°; 2,2, D, 25° 22; 1,57, B, 25° 23; 2,64, D, 25° 79;	C ₄ H ₆ N ₂ 3(5)-Methyl-4-nitro(5(3)-chloropyrazole	5,92, B, 25° 6, 12
C ₃ H ₅ O ₂ N Oxazolidone		2,06, B, 25°; 2,33, D, 25° 5, 12; 2,214 ± 0,015; MW 80	C ₄ H ₆ O ₂ N 3-Methylisoxazolone	2,90, B, 25° 84; 2,86, B, 25°; 2,89, D, 25° 74, 76, 18 3,12, B, 25° 75 3,04, B, 25°; 3,13, D, 25° 74, 76; 3,17, B, 25° 84
C ₃ H ₅ N ₃ 1-Methyl-1,2,4-triazole		1,71, B, 25° 23	C ₄ H ₆ O ₂ N 3-Methylisoxazolone	1,13, B, 25°; 2,02, D, 25° 75
C ₃ H ₆ ON ₄ 1,4-Dimethyltetrazolone		5,07, D, 30° 81	C ₄ H ₆ O ₂ N ₂ 3-Methyl-5-pyrazolone Dimethylloxadizole	2,54, D, 25° 23 3,31, D, 25° 87
C ₃ H ₆ O ₂ N ₄ 1,2,4-Trimethyltetrazolin-5-one		3,50, D, 25° 10	C ₄ H ₆ N ₂ 1-Methylimidazole	3,63, B, 20°; 3,8, D, 20° 70; 3,76, B, 20° 36; 3,81, B, 25° 91
C ₃ H ₆ N ₄ 1,5-Dimethyltetrazole		1,14, B, 25° 82	4(5)-Methylimidazole	6,4, B, 20°; 6,3, B, 50°; 3,7, B 84; 6,2, B, 70°; 5,1, D, 20°; 5,8, C, 18° 70; 6,3, B, 25°; 5,1, D, 25°; 5,8, C, 25° 22
1-Ethyltetrazole		1,14, B, 25° 81	1-Methylpyrazole	2,28, B, 25° 6, 12
C ₃ H ₆ N ₄ S 5-(N,N-Dimethylamino)-1,2,3,4-thiaziazole		5,30, B, 20° 31 4,4, D, 25° 10	3-Methylpyrazole	1,43, B, 25° 23
1-Methyl-5-aminotetrazole		2,65, B, 25° 31	C ₄ H ₆ N ₂ S 2-Amino-4-methylthiazole	1,8, B 88 3,3, B 88
1,4-Dimethyl-5-iminotetrazole		5,46, B, 25° 31	2-Imino-4-methylthiazoline	3,32, D, 25° 89 3,06, D, 25° 89

TABLE 4 (continued)

1		2		1		2	
$C_6H_6ON_2$	1-Methyl-2-formylimidazole			$C_8H_8N_2S$	2-Methylamino-4-methylthiazole		1,60 ⁸⁸
C_6H_7ON	3,4-Dimethylisoxazole		3,77, B, 25° ⁴⁶	$C_8H_{11}N_3Si$	N-Trimethylsilyl-1,2,3-triazole		1,54, B 129
	3,5-Dimethylisoxazole		3,18, B, 25° ⁷⁴ ; 3,15, B, 25°;	$C_8H_{11}N_6$	1-Ethyl-5-ethylaminotetrazole		7,36, B, 25° ⁸¹
	4,5-Dimethylisoxazole		3,18, B ⁸⁴ ; 3,08, B 25°; 3,10, D ^{25° 74, 78}	$C_8H_9N_2ClS$	6-Chlorobenzo-2,1,3-thiadiazole		0,84, B, 25° ⁸¹
$C_6H_7ON_3$	3,5-Dimethyl-4-nitropyrzazole		4,06, B, 25° ¹²	$C_8H_3N_2ClSe$	6-Chlorobenzo-2,1,3-selenadiazole		0,50, B, 25° ⁹³
$C_8H_9O_2N$	1,3-Dimethylisoxazolin-5-one		5,89, B, 25° ⁹⁰	$C_8H_4O_2N_2Se$	7-Nitroso-3,7-dihydrobenz-2,1,3-oxaselenazole		2,90±0,03 ⁹³
	1,4-Dimethylisoxazolin-5-one		5,15, B, 25° ⁹⁰	$C_8H_4N_6S$	Benzo-1,2,3-thiadiazole		3,57, B, 20° ⁹²
	3,4-Dimethylisoxazolin-5-one		4,73, B, 25°; 5,3, D, 25° ⁹⁰	Benzo-2,1,3-thiadiazole		1,73, B, 25° ⁹²	
$C_8H_9O_2N_6$	3,5-Dimethyl-4-nitropyrzazole		3,88, B, 25° ⁷	$C_8H_4N_2Se$	Benzo-2,1,3-selenadiazole		0,94, B, 25° ⁹²
$C_8H_9O_2N$	N-Acetyloxazolidone		2,81, D, 30° ⁸¹	$C_6H_5ON_2$	Benzo-1,2,3-triazole		4,03, B, 25° ⁹⁴
	5,5-Dimethyloxazolidine-2,4-dione		1,74±0,02, D, 30° ⁸¹	Benzo-1,2,3-triazole		4,07, D, 25° ²³	
$C_5H_7N_2Cl$	3,5-Dimethyl-4-chloropyrazole		2,43, B, 25° ^{7, 12}	$C_6H_8N_2S$	2,3-Dihydroimidazolino-4-methylthiazole		4,4, B ⁹⁸
$C_5H_7N_2Br$	3,5-Dimethyl-4-bromopyrazole		2,62, B, 25° ^{7, 12}	$C_6H_7O_3N$	3,5-Dimethyl-4-isoxazolecarboxylic acid		3,42, B ⁹⁵ ; 3,39, B, 25° ⁷⁴ ;
	1,2-Dimethyl-5-bromoimidazole		3,37, B, 25° ⁹¹	$C_6H_8N_2S$	2,3-Dihydroimidazolino-4-methylthiazole		3,39±0,02, B, 25°; 3,40±
$C_6H_7N_2J$	3,5-Dimethyl-4-iodopyrazole		2,19, B, 25° ^{7, 12}	C_6H_9ON	3,4,5-Trimethylisoxazole		±0,05, B ⁷⁶
$C_6H_8N_6$	1,2-Dimethylimidazole		3,74, B, 25° ⁹¹				
	3,5-Dimethylpyrazole		2,31, B, 25° ^{6, 12}				

TABLE 4 (continued)

1	2	1	2
<p>C_6H_9ON 3,4-Dimethyl-5-methoxyisoxazole</p> <p>1,3,4-Trimethylisoxazol-5-one</p> <p>3,4,4-Trimethylisoxazol-5-one</p> <p>C_6H_9NS 5,6-Dihydro-3-methyl-6-aminoimidazo-[2,1-b]thiazole</p> <p>$C_6H_{10}ON_2$ 1,2,3-Trimethylpyrazol-5-one</p> <p>$C_6H_{10}N_2$ 1-Propylimidazole</p> <p>$C_6H_{10}N_4$ 6,7,8,9-Tetrahydro-5H-tetrazoloazepine</p> <p>C_7H_9NClS 2-Chlorobenzothiazole</p> <p>C_7H_5ON 4,5-Benzisoxazole</p> <p>Benzoxazole</p> <p>C_7H_5ONS 2-Hydroxybenzothiazole</p> <p>$C_7H_5O_2N_3$ 5(6)-Nitrobenzimidazole</p> <p>C_7H_5NS Benzothiazole</p>	<p>2,16, D, 25° 75; 2,47—2,52, B, 25° 80</p> <p>3,78, B, 25°; 4,24, D, 25° 75; 5,68, B, 25°; 5,84, D, 25° 80</p> <p>5,00, B, 25°; 5,11, D, 25° 75</p> <p>1,8±0,2, B 96</p> <p>5,62, D, 25° 29</p> <p>4,12, B, 25° 20</p> <p>6,14, B, 25° 97</p> <p>2,28, B, 25°; 3,33, B, 40° 41</p> <p>3,03, B, 25° 23 1,47, B, 25° 23</p> <p>3,36, B, 25°; 3,33, B, 40° 41</p> <p>5,95, D, 25° 26, 28</p> <p>1,45, B, 25° 98</p>	<p>$C_7H_6NS_2$ 2-Mercaptobenzothiazole</p> <p>$C_7H_5N_2Br$ 4-Bromopyrazolo[2,3-a]pyridine</p> <p>$C_7H_6N_2$ Benzimidazole</p> <p>Indazole</p> <p>Pyrazolo[2,3-a]pyridine</p> <p>$C_7H_6N_4S$ 1,1'-Thiocarbonyldipyrazole</p> <p>$C_7H_{10}ON_2$ 3,5-Dimethyl-4-acetylpyrazole</p> <p>$C_7H_{10}N_6$ 4,5,6,7-Tetrahydrobenzimidazole</p> <p>$C_7H_{10}N_4S$ 3,5-Dimethyl-6-imino-5,6-dihydroimidazo-[2,1-b]thiazole</p> <p>$C_7H_{11}N_3S$ 3-Methyl-6-methylamino-5,6-dihydroimidazo[2,1-b]thiazole</p> <p>$C_7H_{13}N_2Al$ N-Dimethylaluminio-3,5-dimethylpyrazole</p> <p>$C_7H_{13}N_2Ga$ N-Dimethylgallio-3,5-dimethylpyrazole</p> <p>$C_7H_{13}N_2In$ N-Dimethylindio-3,5-dimethylpyrazole</p> <p>$C_7H_{15}N_3Si$ N-Trimethylsilyl-4,5-dimethyl-1,2,3-triazole</p>	<p>4,00, B, 25° 98; 4,67, B, 25°; 4,02, D, 25° 41</p> <p>2,44, B, 25° 8</p> <p>3,93, D, 25° 23; 4,08, D, 25° 100; 4,03, D, 25° 86; 27</p> <p>1,83, B, 25° 23 2,15, B, 25° 8</p> <p>3,19, B 108</p> <p>2,26, B, 25° 45, 53</p> <p>3,98, D, 25° 26, 27</p> <p>3,3±0,2, B 96</p> <p>1,6±0,2, B 96</p> <p>0, B, 25° 130, 131</p> <p>1,57, B, 25° 130, 131</p> <p>1,39, B, 25° 130, 131</p> <p>0,66, B 29</p>

TABLE 4 (continued)

1		2		1		2	
C₈H₅ONS				C₆H₈N₂			
2-Formylbenzothiazole				1-Methylbenzimidazole			4.04, D, 25° 26, 27
C₈H₆ON₂		2.84, B, 25° 45, 53		2-Methylbenzimidazole			4.65, D, 25° 28
2-Phenyl-1,3,4-oxadiazole		3.58, B ¹⁰¹		C₈H₈N₄			
C₈H₆O₂N₄				1-Methyl-5-phenyltetrazole			5.70, B, 25° 31
1-(p-Nitrophenyl)-1,2,4-triazole		2.33, D, 25° 10		1-Phenyl-5-methyltetrazole			5.64, B, 25° 31
3-(p-Nitrophenyl)-1,2,4-triazole		6.60, D, 25° 10		3-Phenyl-5-amino-1,2,4-triazole			3.57, D, 25° 10
C₈H₇ONS₂				C₈H₁₀N₃Ga			
N-Hydroxymethylbenzothiazolethione				N-Dimethylgalliobenzotriazole			1.83, B, 25° 180, 131
C₈H₇O₂N₃		4.58, B, 25° 102		C₈H₁₁O₂N			
2-Methyl-5(6)-nitrobenzimidazole		6.57, D, 25° 28		Ethyl 3,5-dimethylisoxazole-4-carboxylate			3.13, D, 25° 103
C₈H₇O₂N₅				C₈H₁₂ON₂			
1-Methyl-5-(p-nitrophenyl)tetrazole		3.87, B, 25° 71		1,3,5-Trimethyl-4-acetylpyrazole			2.79, B, 25° 45, 53
C₈H₇NS				C₈H₁₄O₂N₄			4.05 ¹⁰⁴
2-Methylbenzothiazole		1.33, B, 25° 88		1,3,4,6-Tetramethyltetrahydroimidazo-[4,5-a]imidazole-2,5-dione			
C₈H₇NS₂				C₉H₁₇N₃Si			
2-Methylmercaptobenzenothiazole		1.42, B, 25° 24, 1.33, B, 25° 102		N-Trimethylsilyl-4-propyl-1,2,3-triazole			1.05, B ¹²⁸
4-Methyl-2-mercaptobenzenothiazole		4.00, B, 25° 24		C₉H₆O₄N₄			
6-Methyl-2-mercaptobenzenothiazole		4.30, B, 25° 24		1-(o,p-Dinitrophenyl)imidazole			3.36, D, 25° 20
N-Methylbenzenothiazolethione		4.84, B, 25° 24		C₉H₇ON			
C₈H₇N₃				3-Phenylisoxazole			2.98, B, 25° 84, 2.80, B, 25° 75
1-Phenyl-1,2,3-triazole		4.08, B, 25° 23		4-Phenylisoxazole			2.95, B, 25° 75
2-Phenyl-1,2,3-triazole		0.97, B, 25° 23		5-Phenylisoxazole			3.28, B, 25° 64, 3.19, B, 25° 75; 3.34, B ⁹⁵
4-Phenyl-1,2,4-triazole		2.88, B, 25° 23		C₉H₇ON₂Br			
3-Phenyl-1,2,4-triazole		5.63, B, 25° 23		5-Phenyl-2-bromomethyl-1,3,4-oxadiazole			3.56, B, 25° 101
C₈H₇N₃Cl		3.10, D, 25° 10		C₉H₇O₂N			
1-(p-Chlorophenyl)-5-methyltetrazole		4.38, B, 25° 71		3-Phenylisoxazol-5-one			4.9, B, 30° 105
C₈H₇N₃Br				4-Phenylisoxazol-2-one			4.69, D, 25° 80
1-(p-Bromophenyl)methyltetrazole		4.45, B, 25° 71					

TABLE 4 (continued)

I	2	I	2
$C_9H_8O_2N_3$	5,90, B, 25°; 5,89, D, 25° ⁸²	$C_9H_8N_4$	3,94, B, 25° ^{48, 51}
1-(o-Nitrophenyl)imidazole	1,55, D, 25° ²¹	2-Phenylazobenzimidazole	3,96, B ⁸⁸
1-(p-Nitrophenyl)imidazole	3,63, D, 25° ²¹	$C_9H_8ON_3$	2,33, B, 25° ¹⁰²
1-(m-Nitrophenyl)imidazole	4,40, B, 25° ^{6, 12}	2-Amino-5-benzyl-1,3,4-oxadiazole	7,0±0,2, D, 25° ¹⁰⁸
1-(p-Nitrophenyl)pyrazole	4,44, B, 25° ¹⁰²	$C_9H_8ONS_2$	2,97, B, 25° ⁷
$C_9H_7O_2NS_2$		2-Hydroxyethylthiobenzothiazole	1,85, B, 25° ⁷
2-Carboxymethylthiobenzothiazole	3,76, B, 25° ⁷	$C_9H_8O_2N_2S_2$	2,36, B, 25° ⁷
$C_9H_7N_2Cl$	2,28, B, 25° ⁷	2-Sulfanilamidothiazole	3,34, B, 25° ⁷
1-Phenyl-3-chloropyrazole	1,25, B, 25° ⁷	$C_9H_8N_3$	4,61, D, 25° ²⁸
1-Phenyl-5-chloropyrazole	2,28, B, 25° ⁷	1-(p-Aminophenyl)pyrazole	6,8±0,2, D, 25° ¹⁰⁸
$C_9H_7N_2Br$	1,33, B, 25° ⁷	1-Phenyl-3-aminopyrazole	4,01, D, 25° ¹⁰
1-Phenyl-4-bromopyrazole	2,01, B, 25° ²¹	1-Phenyl-4-aminopyrazole	1,72, B, 25° ¹¹
1-Phenyl-5-bromopyrazole	3,434, B ⁸⁷	$C_9H_8ON_2$	2,80, B, 25° ^{130, 131}
1-(p-Bromophenyl)imidazole	1,93, B, 25° ^{45, 53}	2-Methyl-5(6)-methoxybenzimidazole	1,87, B, 25° ^{130, 131}
$C_9H_8ON_2$	2,71, D, 25° ⁷	$C_9H_{10}O_2N_4S_2$	1,22, B, 25° ^{130, 131}
2-Methyl-5-phenyl-1,3,4-oxadiazole	3,37, B, 25° ^{45, 53}	$C_9H_{10}N_4$	4,27, B ¹²⁹
2-Acetylbenzimidazole	2,18, D, 25° ⁷	1-Methyl-3-phenyl-5-amino-1,2,4-triazole	3,61, B ¹²⁹
1-(p-Hydroxyphenyl)pyrazole	2,43, B, 25° ⁷	1-(α -Pyriddy)-4-methyl-5-aminopyrazole	
1-Methyl-2-formylbenzimidazole	3,41, D, 25° ⁷	$C_8H_{11}N_2Al$	
1-Phenyl-3-hydroxypyrazole	5,48, D, 25° ²¹	N-Dimethylaluminobenzimidazole	
1-Phenyl-4-hydroxypyrazole	1,58, D, 25° ¹⁰	$C_8H_{11}N_2B$	
1-Phenyl-5-hydroxypyrazole	3,14, B, 25° ^{20, 3,50, B}	N-Dimethylborobenzimidazole	
1-(p-Hydroxyphenyl)imidazole	2,26, B, 25° ^{6, 12}	$C_8H_{11}N_2Ga$	
$C_9H_8ON_4$	2,00, B, 25° ^{6, 12}	N-Dimethylgalliobenzimidazole	
1-Benzoyl-5-amino-1,2,4-triazole	2,1, B, 25° ⁸⁸	$C_8H_{10}N_6Si$	
$C_9H_8N_6$		N-Trimethylsilylbenzotriazole	
1-Phenylimidazole		$C_8H_{10}N_2Al$	
3(5)-Phenylpyrazole		N-Dimethylaluminio-4,5,6,7-tetrahydrobenzimidazole	
1-Phenylpyrazole			
$C_9H_8N_2S$			
2-Amino-4-phenylthiazole			

TABLE 4 (continued)

1	2	1	2
$C_9H_{15}N_2Ga$ N-Dimethylgallio-4,5,6,7-tetrahydro- benzimidazole	1,29, B, 25° 130, 131	1-Phenyl-3-methylthio-5-pyrazolone $C_{10}H_{10}ON_2$	2,51, D, 25° 29
$C_9H_{10}N_4Si$ N-Trimethylsilyl-4-butyl-1,2,3-triazole	1,03, B, 25° 129	1-Methyl-2-acetylbenzimidazole 1-Phenyl-2-methyl-3-pyrazolone 1-Phenyl-2-methyl-5-pyrazolone 1-Phenyl-3-methyl-5-pyrazolone	2,07, B, 25° 45, 53 4,99, D, 25° 29 5,03, B, 25° 29 3,33, D, 25°; 2,68, CH 25° 29; 2,5°; 1,87, CH 2,11, D, 25°; 1,87, CH 25° 29 3,80, B, 25° 75
$C_{10}H_7O_3N$ 3-Phenylisoxazole-5-carboxylic acid 5-Phenylisoxazole-3-carboxylic acid 5-Phenylisoxazole-4-carboxylic acid	3,25, D, 25° 89 2,85, D, 25° 89 3,33, D ⁸⁶	1-Phenyl-5-methyl-3-pyrazolone 1-Phenyl-5-methoxy-pyrazole	
$C_{10}H_7O_4N_4Cl$ 1-(p-Nitrophenyl)-3-methyl-4-nitro- 5-chloropyrazole	2,97, B, 25° 6, 12	$C_{10}H_{10}ON_4$ 1-Acetyl-3-phenyl-5-amino-1,2,4- triazole	1,13, D, 25° 10
$C_{10}H_8ONBr$ 2-Bromomethyl-5-phenylloxazole	2,68, B 138	1-(p-Methoxyphenylazo)imidazole	3,73, B, 25° 48, 51
$C_{10}H_8ON_2$ 1-Phenyl-2-formylimidazole	3,53, B, 25° 46	$C_{10}H_{10}N_6$ 1-Methyl-3-phenylpyrazole 1-Methyl-5-phenylpyrazole 1-(o-Tolyl)imidazole 1-(m-Tolyl)imidazole 1-(p-Tolyl)imidazole	2,30, B, 25° 6, 12 2,45, B, 25° 6, 12 3,79, B, 25° 21 3,73, B, 25° 21 3,90, B, 25° 21 4,19, B, 25° 21
$C_{10}H_9ON$ 3-Methyl-5-phenylisoxazole 5-Methyl-3-phenylisoxazole	3,04, B, 25° 84 3,05, B, 25° 84	$C_{10}H_{11}N_3$ 1-(α -Pyridyl)-3,5-dimethylpyrazole 1-(β -Pyridyl)-3,5-dimethylpyrazole 1-(γ -Pyridyl)-3,5-dimethylpyrazole	1,87, B, 25° 11, 54 2,87, B, 25° 11, 54 2,84, B, 25° 11, 54
$C_{10}H_9ON_3$ 1-Acetyl-3-phenyl-1,2,4-triazole	1,98, D, 25° 10	$C_{10}H_{10}N_4$ 2-(p-Tolylazo)imidazole	3,76, B, 25° 48, 51
$C_{10}H_9ON_4$ 1-Benzoyl-3-methyl-5-amino-1,2,4-triazole	1,23, D, 25° 10	$C_{10}H_{10}N_6$ 1-Propylbenzimidazole	3,72, B, 25°; 3,95, D, 25° 20
$C_{10}H_9O_2N$ 3-Phenyl-N-methylisoxazol-5-one 4-Phenyl-N-methylisoxazol-5-one 3-Phenyl-4-methylisoxazol-5-one	5,97, B, 25° 90 5,40, B, 25° 90 4,9, B, 25° 75	$C_{10}H_{12}N_4S$ 1-Phenyl-3-methyl-5-methylthiopyrazole	2,80, D, 25° 29
$C_{10}H_9N_2Cl$ 1-Phenyl-3-methyl-5-chloropyrazole	1,07, B, 25° 11	$C_{10}H_{12}N_4S_2$ N-Dimethylaminomethylbenzothiazolone	4,39, B, 25° 102
$C_{10}H_{10}N_2S$ 2-Methylamino-4-phenylthiazole	2,3, B 86		

TABLE 4 (continued)

1	2	1	2
C ₁₀ H ₁₂ N ₄ Bis(3,5-dimethyl-4-pyrazolyl)	3,54, D, 25° 8	3-Methyl-4-phenyl-5-methoxyisoxazole 2,3-Dimethyl-4-phenylisoxazol-5-one	2,32, D, 25° 75 5,95, D, 25° 75; 5,65, B, 25° 90
C ₁₀ H ₁₆ O ₃ N ₂ N-Cyclohexyl-5-methyl-5-nitrotetrahydroxazole	4,45 ± 0.1, B, 20° 110	3-Phenyl-2,4-dimethylisoxazol-5-one 3-Phenyl-4,4-dimethylisoxazol-5-one	5,70, B, 25° 90 4,9, B, 25° 5,10, D, 25° 75
C ₁₀ H ₁₈ N ₄ 8-Isobutyl-6,7,8,9-tetrahydro-5H-tetrazaolepine	6,18, B, 25° 97	2,3-Dimethyl-5-phenylisoxazol-5-one	2,83, D, 25° 75 5,70, B, 25° 90
8-tert-Butyl-6,7,8,9-tetrahydro-5H-tetrazaolepine	6,20, B, 25° 97	C ₁₁ H ₁₁ N ₃ S 6-Amino-5,6-dihydro-3-phenylimidazo[2,1-d]thiazole	2,1 ± 0.2, B ⁸⁶
C ₁₁ H ₈ N ₂ Naphth[1,2-d]imidazole	4,12, D, 25° 86, 27	C ₁₁ H ₁₂ ON ₂ 1-Benzyl-3-methylpyrazol-5-one	3,10, D, 25° 25° 29 2,78, CH ₁
C ₁₁ H ₉ O ₃ N 3-Methyl-5-phenyl-4-carboxyisoxazole	3,32, B, 25° 75	1-Phenyl-2,3-dimethylpyrazol-5-one (5-anti-pyridine)	5,48, B, 25° 23, 5,50, B, 30° 105; 5,47, B, 25° 26
3-Phenyl-5-methyl-4-carboxyisoxazole	3,23, B, 25° 75	1-Phenyl-2,5-dimethylpyrazol-3-one (3-anti-pyridine)	5,69, D, 25° 108
C ₁₁ H ₁₀ ON ₂ 1-Benzyl-2-formylimidazole	3,37, B, 25° 48	C ₁₁ H ₁₂ O ₃ S ₂ 2-Benzothiazoliosulfene morpholide	1,73, B, 25° 102
1-(p-Acetylphenyl)imidazole	2,89, B, 25° 21	C ₁₁ H ₁₂ N ₂ S 1-Phenyl-2,3-dimethylpyrazole-5-thione (5-thiopyridine)	7,33, B, 25° 23, 7,60, D, 25° 29
1-Phenyl-4-acetylpyrazole	2,61, B, 25° 45, 53	1-Phenyl-3-methyl-5-methylpyrazole-5-pyridone	2,80, D, 25° 29
C ₁₁ H ₁₀ O ₅ N ₄ 1,5-Dimethyl-2-phenyl-4,4'-dinitro-pyrazol-3-one(4,4'-dinitroantipyridine)	4,6, B, 30° 105	C ₁₁ H ₁₂ N ₂ Se 1-Phenyl-2,3-dimethylselenopyrazol-5-one (5-selenopyridine)	7,91, D, 25° 29
C ₁₁ H ₁₀ N ₂ S 2,3-Dihydroimidazolino-4-phenylthiazole	4,2, B ⁸⁸	1-Phenyl-2,5-dimethylselenopyrazol-3-one (3-selenopyridine)	8,17, D, 25° 23
C ₁₁ H ₁₀ N ₄ N-(1-Methyl-2-benzimidazolyl)imidazole	4,45, D, 25° 58	2-(o,p-Xylylazo)imidazole	3,95, B, 25° 38, 53
N-(1-Methyl-2-benzimidazolyl)pyrazole	2,86, D, 25° 88	C ₁₁ H ₁₂ N ₄ S ₂ 1,1'-Thiocarbonylbis(3,5-dimethyl-4-chloropyrazole)	1,98, B ¹⁰⁸
C ₁₁ H ₁₁ ON ₂ Br 1,5-Dimethyl-2-phenyl-4-bromo-pyrazol-3-one(4-bromoantipyridine)	5,9, B, 25° 105		
C ₁₁ H ₁₁ O ₂ N 3,4-Dimethyl-4-phenylisoxazol-5-one	4,86, D, 25° 75		

TABLE 4 (continued)

1		2		1		2	
$C_{11}H_{10}N_3$	1-Phenyl-2,5-dimethyliminopyrazol-3-one (3-iminopyrine)	3,92, D, 25° 29		$C_{12}H_{14}N_2S$	1-Phenyl-3,4,4-trimethylthiopyrazol-5-one	3,16, D, 25° 29	
$C_{11}H_{10}N_3$	1-Phenyl-2,3-dimethyliminopyrazol-5-one (5-iminopyrine)	4,37, D, 25° 29		$C_{12}H_{16}N_2S_2$	N-Diethylaminomethylbenzothiazolone	4,38, B, 25° 102	
$C_{11}H_{13}ON_3$	1-(β -Pyridyl)ethyl-3-methyl-5-hydroxypyrazole	3,70, B, 25° 54, 55		$C_{12}H_{18}O_2N_4$	Bis(1,3,4-trimethyl-5-oxo-4-pyrazolyl)	meso, 1,95, B, 25° 126, racemate, 3,94, B, 25° 138	
$C_{11}H_{14}N_3$	1-(α -Pyridyl)-3-ethyl-4-methyl-5-aminopyrazole	1,71, B, 25° 11, 54		$C_{13}H_{18}N_2S$	Bis(1,3,5-trimethyl-4-pyrazolyl) sulfide	3,52, D, 25° 8	
$C_{11}H_{14}N_3$	1-(γ -Pyridyl)-3-ethyl-4-methyl-5-aminopyrazole	3,90, B, 25° 11, 54		$C_{13}H_{16}O_3N_2$	2-Phenyl-6-nitrobenzoxazole	4,23, D, 25° 61	
$C_{11}H_{14}N_4S_2$	1,1'-Thiocarbonylbis(3,5-dimethylpyrazole)	3,37, B, 25° 108		$C_{13}H_8O_2N_2$	2-(α -Hydroxyphenyl)-6-nitrobenzoxazole	5,50, D, 25° 61	
$C_{11}H_{15}N_3Si$	N-Trimethylsilyl-4-phenyl-1,2,4-triazole	1,09, B, 25° 129		$C_{13}H_8O_4N_4$	3-Nitro-2-(p-nitrophenyl)imidazol[1,2-a]pyridine	7,27, D, 25° 43	
$C_{12}H_6N_3$	1-Phenylpyrazolo[3,4-b]pyridine	4,29, B, 25° 8, 12		$C_{13}H_8O_2N$	2-(α -Hydroxyphenyl)benzoxazole	2,10, B, 25°; 2,15, D, 25° 61	
$C_{12}H_6N_3$	1-Phenylpyrazolo[4,5-b]pyridine	2,03, B, 25° 8, 12		$C_{13}H_9O_2N_3$	2-(α -Nitrophenyl)benzimidazole	5,32, D, 25° 52	
$C_{12}H_{10}N_3$	3-Methylnaphth[1,2-d]imidazole	3,86, D, 25° 26, 23		$C_{13}H_{11}O_2N_3$	2-(m-Nitrophenyl)benzimidazole	3,24, D, 25° 52	
$C_{12}H_{11}O_3N$	Ethyl 5-phenylisoxazole-4-carboxylate	3,28, D, 25° 107		$C_{13}H_{11}O_2N_3$	1-(α -Nitrophenyl)benzimidazole	5,91, D, 25° 52	
$C_{12}H_{14}ON_5$	1-Phenyl-3-methyl-4-ethylpyrazol-5-one	3,39, D, 25° 29		$C_{13}H_9NS$	1-(m-Nitrophenyl)benzimidazole	3,45, D, 25° 52	
$C_{12}H_{14}ON_5$	1-Phenyl-3,4,4-trimethylpyrazol-5-one	2,83, D, 25° 29		$C_{13}H_9NS$	2-Phenylbenzothiazole	0,94, B 112	
$C_{12}H_{14}ON_5$	1-Phenyl-3-methyl-5-ethoxypyrazole	2,66, D, 25° 29		$C_{13}H_{10}ON_2$	2-(α -Hydroxyphenyl)benzimidazole	4,49, D, 25° 61	
$C_{12}H_{14}ON_5S_2$	1-Isopropyl-2-acetylbenzimidazole	2,44, B, 25° 45, 53		$C_{13}H_{10}N_2$	3(5)-Phenyl-5(3)-(2-furyl)pyrazole	2,60, D, 25° 8	
$C_{12}H_{14}ON_5S_2$	N-Morpholinomethylbenzothiazolone	4,72, B, 25° 102		$C_{13}H_{10}N_2$	2-Phenylbenzimidazole	3,63, D, 25° 61	

TABLE 4 (continued)

1	2	1	2
C ₁₃ H ₁₀ N ₂ S ₂ 2-Benzothiazolesulfene anilide	2,38, B, 25° 72	H ₁₀ ON ₂ Cl 5-Phenyl-2-(o-chlorophenyl)-1,3,4-oxadiazole	3,71, B, 25° 101 3,30, B, 25° 101
C ₁₃ H ₁₀ N ₄ 1,5-Diphenyltetrazole	5,95, B, 25° 113	C ₁₄ H ₁₀ ON ₂ Br 5-Phenyl-2-(p-bromophenyl)-1,3,4-oxadiazole	3,27, B, 25° 101
C ₁₃ H ₁₂ O ₂ N ₂ 3-Ethyl-5-methyl-5H-(a)-(3,1)-benzoxazolin-5-one	α-lactone, 1,33, B 114; β-lactone, 2,53, B 114	C ₁₄ H ₁₀ O ₃ N ₃ 5-Phenyl-2-(o-nitrophenyl)-1,3,4-oxadiazole	4,85, B, 25° 101 4,30, B, 25° 101
C ₁₃ H ₁₀ N ₂ 1-Ethynaphtho[1,2-d]imidazole	4,04, D, 25° 26, 28	C ₁₄ H ₁₀ ON ₂ 2,5-Diphenyl-1,3,4-oxadiazole	3,56 ⁸⁷ , 3,45, B, 25° 115, 3,52, B 101
C ₁₃ H ₁₂ O ₂ N Ethyl 3-methyl-5-phenylisoxazole-4-carboxylate	3,11, D, 25° 103	3,5-Diphenyl-1,2,4-oxadiazole	1,78, B 116
Ethyl 3-phenyl-5-methylisoxazole-4-carboxylate	3,21, D, 25° 103	3,4-Diphenyl-1,2,5-oxadiazole	4,26, B 116
C ₁₃ H ₁₄ ON ₂ 1,3,5-Trimethyl-4-benzoylpyrazole	2,80, B, 25° 45, 53	2-Benzoylbenzimidazole	1,45, B, 25° 45, 53
C ₁₃ H ₁₆ N ₄ 1-(o-Pyridyl)-3-n-propyl-4-ethyl-5-aminopyrazole	1,76, B, 25° 11	C ₁₄ H ₁₀ ON ₂ S 2-(o-Hydroxyphenyliminomethyl)benzothiazole	1,71, B, 25° 48
1-(β-Pyridyl)-3-n-propyl-4-ethyl-5-aminopyrazole	3,75, B, 25° 11	C ₁₄ H ₁₀ O ₂ N ₂ 2,5-Di(o-hydroxyphenyl)-1,3,4-oxadiazole	4,16, D, 25° 101
1-[(β-Pyridyl)ethyl]-3-ethyl-4-methyl-5-aminopyrazole	3,32, B, 25°; 4,05, D, 25° 55	C ₁₄ H ₁₀ N ₄ N-(2-Benzimidazolyl)indazole	1,41, D, 25° 58
C ₁₄ H ₈ ON ₂ Benzylenebenzimidazole	1,97, B, 25° 100	C ₁₅ H ₁₁ O ₂ 2-(o-Methoxyphenyl)benzoxazole	2,21, B, 25° 61
C ₁₄ H ₁₀ ON ₂ Br 2,5-Di-(p-bromophenyl)-1,3,4-oxadiazole	2,23, B, 25° 101	C ₁₅ H ₁₂ ON ₂ 2,5-Di(o-aminophenyl)-1,3,4-oxadiazole	2,05, B, 25°; 2,22, D, 25° 101
C ₁₄ H ₈ N ₂ S ₃ 2,2-Bis(benzothiazolyl) sulfide	3,07, B, 25° 41	2,5-Di(p-aminophenyl)-1,3,4-oxadiazole	4,58, B, 25° 101
C ₁₄ H ₈ N ₂ S ₄ 2,2-Bis(benzothiazolyl) disulfide	3,52, B, 25° 41	C ₁₄ H ₁₂ N ₂ S ₂ 2-Benzothiazolesulfene methylamide	1,73, B, 25° 102
		3-Phenylamino-1-phenyl-1,2,4-triazole	3,54±0,02, B, 25° 117

TABLE 4 (continued)

1	2	1	2
$C_{15}H_{10}ONBr$ 2-(p-bromophenyl)-5-phenyloxazole 2,5-Diphenyl-4-bromoxazole	1,87, B, 25° 133 2,15, B, 25° 133	$C_{15}H_{12}N_4$ N-(1-Methyl-2-benzimidazolyl)benzimidazole N-(1-Methyl-2-benzimidazolyl)indazole	4,38, D, 25° 58 2,77, D, 25° 58
$C_{15}H_{10}ON$ 2,5-Diphenyl-4-iodooxazole 2,4-Diphenyl-5-iodooxazole	1,42, B, 25° 133 1,42, B, 25° 133	$C_{15}H_{13}ON_3$ 1-Methyl-3-(o-hydroxyphenylimino-methyl)indazole 1-Methyl-2-(o-hydroxyphenylimino-methyl)benzimidazole	3,68, B, 25° 118, 3,68, D, 25° 50 1,87, B, 25° 48
$C_{15}H_{10}ON_6$ 2-(p-Nitrophenyl)-5-phenyloxazole 2-Phenyl-5-(p-nitrophenyl)oxazole	5,11, B, 25° 133 3,56, B, 25° 133	$C_{15}H_{13}O_2N_4$ 1-Ethyl-2-(m-nitrophenyl)benzimidazole 1-(α -Pyridyl)-3-(p-methoxyphenyl)-5-hydroxypyrazole 1-(β -Pyridyl)-3-(p-methoxyphenyl)-5-hydroxypyrazole 1-(γ -Pyridyl)-3-(p-methoxyphenyl)-5-hydroxypyrazole	4,59, D, 25° 52 2,80, B, 25° 54 5,16, B, 25°, 4,99, D, 25° 54 5,79, D, 25° 54
$C_{15}H_{11}N_2Br$ 3,5-Diphenyl-4-bromopyrazole	2,62, B, 25° 7, 12	$C_{15}H_{13}N_5$ 1-Methyl-3-(phenyliminomethyl)indazole	2,48, B, 25° 118
$C_{15}H_{12}ON_2$ 5-Phenyl-2-(p-tolyl)-1,3,4-oxadiazole	3,81, B, 25° 101	$C_{15}H_{14}N_4$ 1-(β -Pyridyl)-3-benzyl-5-aminopyrazole 1-(β -Pyridyl)-3-(p-tolyl)-5-aminopyrazole	3,36, B, 25° 11, 54 3,85, B, 25° 11, 54
$C_{15}H_{12}ON_4$ 1-Benzoyl-3-phenyl-5-amino-1,2,4-triazole	0,84, D, 25° 10	$C_{15}H_{12}N_4$ 1-[(β -Pyridyl)ethyl]-3-n-propyl-4-ethyl-5-aminopyrazole	2,92, B, 25° 54, 55, 2,97, D, 25° 55
$C_{15}H_{12}O_2N_6$ 5-Phenyl-2-(o-methoxyphenyl)-1,3,4-oxadiazole 5-Phenyl-2-(m-methoxyphenyl)-1,3,4-oxadiazole 5-Phenyl-2-(p-methoxyphenyl)-1,3,4-oxadiazole	4,37, B, 25° 101 3,67, B, 25° 101 3,83, B, 25° 101	$C_{16}H_{15}O_2N_2$ 2-Phenyl-5H-pyrazole of 3,1- α -benzoxazin-5-one (o-lactone)	1,53, B, 4,114
$C_{15}H_{12}N_2$ 4,5-Diphenylimidazole 3,5-Diphenylpyrazole	4,39, D, 40° 26, 27, 4,40, D, 25° 52 2,30, B, 25° 6, 12	$C_{16}H_{15}O_3N_2$ 3,5-Diphenylisoxazole-4-carboxylic acid 2-(p-methoxyphenyl)-5-(p-nitrophenyl)-oxazole	5,94, D, 25° 107 4,96, B, 25° 53

TABLE 4 (continued)

1	2	1	2
$C_{16}H_{13}ON$ 2-Methyl-4,5-diphenyloxazole	1,7, B, 25° 24	$C_{16}H_{15}N_2S$ 1-(2-Benzothiazolyl)-3-methyl-2-(p-tolyl)formazan	2,96, D, 25° 120
$C_{16}H_{13}ON_3$ 1-Phenyl-4-(o-hydroxyphenylimino methyl)-pyrazole	2,94, B, 25° 50	$C_{16}H_{16}ON_4$ 1-[(α -Pyridyl)ethyl]-3-(p-aminophenyl)-5-hydroxypyrazole	5,41, D, 25° 55
1-Phenyl-2-(o-hydroxyphenylimino methyl)-imidazole	2,01, B, 25° 48	1-[(β -Pyridyl)ethyl]-3-(p-aminophenyl)-5-hydroxypyrazole	6,51, D, 25° 55
$C_{16}H_{13}O_2N$ 2-(p-Methoxyphenyl)-5-phenyloxazole	1,92, B, 25° 133	$C_{16}H_{17}N_5$ 1-(α -Pyridyl)-3-(p-aminophenyl)-4-ethyl-5-aminopyrazole	2,40, B, 25° 84
3,4-Diphenyl-5-methoxyisoxazole	2,28, D, 25° 75	1-[(α -Pyridyl)ethyl]-3-(p-aminophenyl)-5-aminopyrazole	3,01, D, 25° 55
3,4-Diphenyl-N-methylisoxazol-5-one	5,39, D, 25° 75	$C_{17}H_{13}O_3N$ Methyl 3,5-diphenylisoxazole-4-carboxylate	3,43, D, 25° 107
$C_{16}H_{13}N_3$ 1-Phenyl-4-(phenylimino methyl) pyrazole	2,24, B, 25° 50	$C_{17}H_{14}O_2N_2$ 5-(ω -Styryl)-2-(p-methoxyphenyl)-1,3,4-oxadiazole	4,22, D, 25° 101
$C_{16}H_{14}O_3N_2$ 2,5-Di-(p,p'-methoxyphenyl)-1,3,4-oxadiazole	3,99, B, 25° 101	$C_{17}H_{14}N_2$ N-Ethylphenanthrene[9,10-d]imidazole	4,11, D, 25° 23, 28
$C_{16}H_{14}O_3N_3$ 1-[(α -Pyridyl)ethyl]-3-(p-nitrophenyl)-5-hydroxypyrazole	6,80, D, 25° 55	$C_{17}H_{15}N_3$ 1-Phenyl-4-(p-tolylimino methyl)pyrazole	2,02, B, 25° 50
$C_{16}H_{14}N_2$ 1,5-Diphenyl-3-methylpyrazole	2,48, B, 25° 6, 12	$C_{17}H_{17}ON_3$ 1-[(α -Pyridyl)ethyl]-3-phenyl-4-methyl-5-hydroxypyrazole	4,17, B, 25° 55, 25° 84, 56
2,2'-Bis(1-methylbenzimidazolyl)	1,41, B, 25° 119	$C_{17}H_{17}ON_5$ 1-[(α -Pyridyl)ethyl]-3-phenyl-4-methyl-5-hydroxypyrazole	4,07, B, 25° 55, 25° 84, 56
$C_{16}H_{15}ON_3$ 5-Phenyl-2-(p-dimethylaminophenyl)-1,3,4-oxadiazole	5,13, B, 25° 101	$C_{17}H_{17}ON_5$ 1-[(β -Pyridyl)ethyl]-3-phenyl-4-methyl-5-hydroxypyrazole	4,64, B, 25° 5, 577, D, 25° 55
1-[(β -Pyridyl)ethyl]-3-phenyl-5-hydroxypyrazole	3,16, B, 25°; 3,45, D, 25° 55	$C_{17}H_{17}O_2N_3$ 1-[(α -Pyridyl)ethyl]-3-phenyl-4-methyl-5-hydroxypyrazole	4,07, B, 25° 55, 25° 84, 56
$C_{16}H_{15}ON_5$ 1-(2-Benzoxazolyl)-3-methyl-2-(o-tolyl)-formazan	3,32, D, 25° 120	$C_{17}H_{17}ON_5$ 1-[(β -Pyridyl)ethyl]-3-phenyl-4-methyl-5-hydroxypyrazole	4,64, B, 25° 5, 577, D, 25° 55
$C_{16}H_{15}N_5$ 1-Methyl-3-(p-tolylimino methyl)indazole	2,20, B, 25° 118		

TABLE 4 (continued)

1		2		1		2	
$C_{17}H_{15}O_2N_5$ Diethyl 1-phenyl-5,6-diazo-3a,4,5,6,7,7a-hexahydrobenzotriazole-5,6-dicarboxylate			3,26, D, 25° 122	$C_{19}H_{17}ON_2S$ 2-(α -methoxynaphthyliminomethyl)benzothiazole			0,43, B, 25° 48
$C_{18}H_{12}ON_2S$ 2-(α -Hydroxynaphthyliminomethyl)benzothiazole			2,20, B, 25° 48	$C_{19}H_{14}N_2S_2$ 2-Methyl-(α -thionaphthyliminomethyl)-benzothiazole			1,16, B, 25° 48
$C_{18}H_{14}O_2N_4S$ Bis(1-phenyl-4-pyrazolyl) sulfide			3,15, D, 25° 8	$C_{18}H_{15}ON_3$ 1-Benzyl-3,5-dimethyl-4-(α -hydroxyphenyl)iminomethylpyrazole			3,76, B, 25° 50
$C_{18}H_{14}O_2N_4S$ Bis(1-phenyl-4-pyrazolyl) sulfone			5,23, D, 25° 8	$C_{19}H_{20}N_4$ 2-Benzothiazolylsulfene dicyclohexylamide			3,01, B, 25° 102
$C_{18}H_{15}O_2N$ Ethyl 3,5-diphenyl-4-isoxazole-4-carboxylate			3,22 \pm 0,03, D, 25° 105	$C_{19}H_{30}N_4$ 1-[α -Pyridyl]ethyl-3-aryl-4-butyl-5-aminopyrazole			3,12, B, 25° 54, 55, 25° 55, 4,03, D
$C_{18}H_{15}O_2N_2S$ 1-(m, p-Methylenedioxyphephenethyl)-5-(2-methyl-5-thiazolyl)-1H-pyrid-2-one			4,24, B, 25° 123	$C_{20}H_{12}ON_2S_2$ p-Di-(benzothiazolyl) benzene			0,50, B, 20° 99
$C_{18}H_{17}ON_6$ 1-Phenyl-3,5-dimethyl-4-(α -hydroxyphenyliminomethyl)pyrazole			3,64, B, 25° 50	$C_{20}H_{17}ON_2$ 5-(ω -Styryl)-2-(α -naphthyl)-1,3,4-oxadiazole			4,04, B, 25° 101
$C_{18}H_{18}N_2$ 1-Propyl-4,5-diphenylimidazole			4,17, D, 25° 27	$C_{20}H_{16}ON_2$ 2-(α -Furyl)-4-phenyl-5-(p-tolyl)imidazole			3,76, D, 25° 52
$C_{18}H_{18}N_4$ Bis(1-ethyl-2-benzimidazole)			1,94, B, 25° 119	$C_{20}H_{16}N_4$ 4,5-Dihydro-1,4-diphenyl-3,5-phenylimino-1,2,4-triazole (nitron)			7,20, B, 30° 124
$C_{18}H_{20}ON_2S_3$ (3-Buryl-2-benzothiazolylidene)-3-ethyl-5-ethylideneethodanine			7,68, B, 25° 37	$C_{20}H_{17}ON_3$ 2-(α -Naphthyl)-5-(4-dimethylamino-phenyl)-1,3,4-oxadiazole			5,45, B, 25° 101
$C_{18}H_{20}O_2N_2S_2$ (3-Buryl-2-benzoxazolimidene)-3-ethyl-5-ethylideneethodanine			8,22, B, 25° 37	$C_{20}H_{22}N_4$ Bis(1-propyl-2-benzimidazole)			2,34, B, 25° 125
$C_{19}H_{14}ON_2$ 2-(α -Furyl)-4,5-diphenylimidazole			3,45, D, 25° 52	$C_{21}H_{15}ON$ 2-Biphenyl-5-phenyloxazole			1,65, B, 25° 133
				2-Phenyl-5-biphenylyloxazole			1,95, B, 25° 133

TABLE 4 (continued)

1	2	1	2
$C_{21}H_{15}O_2N_3$ 2-(m-Nitrophenyl)-4,5-diphenylimidazole 2-(p-Nitrophenyl)-4,5-diphenylimidazole	3,48, D, 25° 52 5,78, D, 25° 28, 6,10, D, 25° 52	$C_{22}H_{24}O_2N_4$ Bis(1'-phenyl-3,4-dimethyl-5-oxo-4-pyrazoliné)	meso, 0,98, B, 25° 128, <i>racemate</i> , 2,83, B, 25° 128
$C_{23}H_{15}O_2N_5$ 2-(p-Nitrophenylazo)-4,5-diphenylimidazole	6,04, B, 25° 46, 51	$C_{23}H_{15}ON_4$ 1-(2-Benzimidazolyl)-3-benzyl-4-phenyl-5-hydroxypyrazole	3,38, B, 25° 59
$C_{21}H_{15}N_2Br$ 2-(p-Bromophenyl)-4,5-diphenylimidazole	4,10, D, 25° 52	$C_{23}H_{18}N_4S$ 1-(2-Benzothiazolyl)-3-benzyl-4-phenyl-5-aminopyrazole	1,59, B, 25° 59
$C_{21}H_{13}N_4Br$ 2-(p-Bromophenylazo)-4,5-diphenylimidazole	4,67, B, 25° 46, 51	$C_{23}H_{20}N_4$ 2-(o,p-Xylylazo)-4,5-diphenylimidazole	4,48, B, 25° 48, 51
$C_{21}H_{16}N_2$ 1,3,5-Triphenylpyrazole 2,4,5-Triphenylimidazole	2,69, B 126, 2,47, B, 25° 6, 126 3,92, D, 25° 52	$C_{23}H_{21}N_3$ 2-(p'-Dimethylaminophenyl)-4,5-diphenylimidazole	4,38, D, 25° 28, 4,76, D, 25° 52
$C_{21}H_{16}N_4$ 2-Phenylazo-4,5-diphenylimidazole	4,69, B, 25° 46, 51	$C_{23}H_{22}N_4$ 1-[(α -Pyridyl)ethyl]-3-benzyl-4-phenyl-5-aminopyrazole	3,13, B, 25° 54, 55; 3,66, D, 25° 55
$C_9H_7O_2N_2$ 2-(α -Furyl)-4-phenyl-5-(p-ethoxyphenyl)-imidazole	3,94, D, 25° 52	$C_{23}H_{23}N_6$ 1-(1'-Benzyl-2'-benzimidazolyl)-2-(o-tolyl)-3-methylformazan	3,18, D, 25° 120
$C_{21}H_{18}N_4$ 1-(α -Pyridyl)-3-benzyl-4-phenyl-5-amino-pyrazole	1,32, B, 25° 11, 54	$C_{23}H_{22}N_4$ 2-(o,o',p'-Trimethylphenylazo)-4,5-diphenylimidazole	4,43, B, 25° 46, 51
$C_{22}H_{15}ON_2$ 2-(p-Methoxyphenyl)-4,5-diphenylimidazole	4,21, D, 25° 52	$C_{23}H_{16}ON_2S_2$ p,p'-Dibenzothiazoloyldiphenyl ether	1,13, B, 20° 99
$C_{22}H_{18}N_2$ 2-(p-Tolyl)-4,5-diphenylimidazole	3,87, D, 25° 52	$C_{33}H_{24}ON_4$ 1-(1-Benzyl-2-benzimidazolyl)-3-benzyl-4-phenyl-5-hydroxypyrazole	4,37, B, 25° 59
$C_{22}H_{18}N_4$ 2-(p-Tolylazo)-4,5-diphenylimidazole	4,51, D, 25° 46, 51	$C_{33}H_{28}N_6$ 1,2-Bis(1'-benzyl-2'-benzimidazolyl)-3-methylformazan	4,90, D, 25° 120

The dipole moment of 3-pyrazolone in dioxane (2.11 D) is close, and that of the same compound in chloroform (1.87 D) practically coincides with the moment of 1-phenyl-3-methoxy-5-methylpyrazole (1.80 D) (hydroxy model) [29], which attests to its existence in solution in the enol form, which has also been detected by spectroscopy [42].

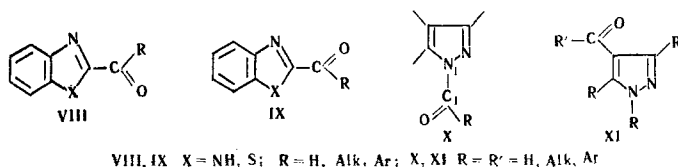
Dipole Moments and Three-Dimensional Structures of Azole Derivatives

The use of the method of dipole moments makes it possible to comparatively simply establish the position of a substituent in azole-containing compounds when there is an appreciable difference in the values of the moments calculated for different isomers. This possibility was used to determine the direction of acylation of 3-amino-1,2,4-triazole [10] and of nitration of 2-phenylimidazo[1,2-a]pyridine [43] and benzo-triazole [44].

The method of dipole moments has broad possibilities for the solution of the problems of the conformations of azole systems.

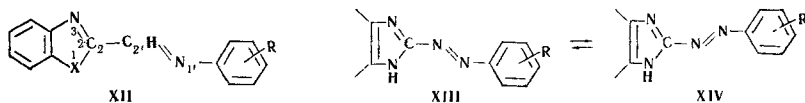
A comparison of the experimentally obtained dipole moments with the values calculated via a vectorial scheme for *s-trans* and *s-cis* conformers of 2-carbonylazoles (VIII and IX) made it possible to establish that predominantly a planar structure with *s-trans*-oriented C=N and C=O bonds [45] is realized for these compounds rather than an equilibrium mixture of two planar conformers, as demonstrated in [46] for analogous imidazole derivatives.

The results of quantum-mechanical calculations also speak in favor of the realization of the *s-trans* form: a considerable gain in energy is observed for VIII [45, 47].



The investigation of N-acylpyrazoles (X) [10] by the method of dipole moments indicates that a structure with C=N and C=O groups situated on different sides of the N₁-C₁ bond is also characteristic for these systems. In addition, the dipole moments found for 4-carbonyl derivatives of pyrazole (XI) can be associated with a configuration in which rotation of the keto group relative to its bond to the ring is observed [45].

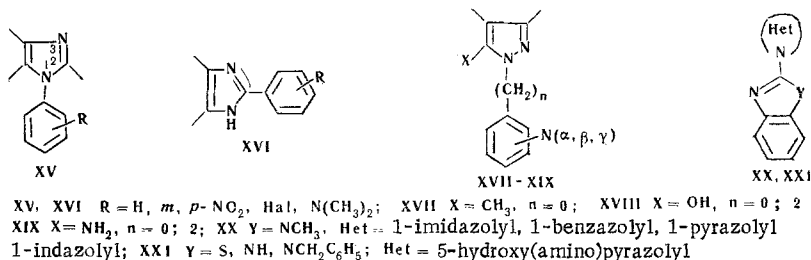
The determination of the dipole moments of the azomethines of the benzazole [48] and pyridine [49, 50] series attests to the realization, for this type of compound, of the *s-trans-trans* configuration (XII) depicted below*:



At the same time, according to the results obtained on comparison of the experimental and calculated (via a vectorial scheme) dipole moments, forms XIII and XIV are equally probable in the case of 2-areneazo derivatives of the imidazole series [49, 51]. Quantum-mechanical calculation leads to the same conclusion. The μ values obtained from the π and σ components of the moment are 4.0 and 4.3 D, respectively, for forms XIII and XIV, while the experimental dipole moments for 2-azoimidazoles with various substituents in the aryl ring (R) vary from 3.8 to 4.5 D [48, 51]. However, calculation of the total energies of the conformational forms XIII and XIV shows that the *s-cis-trans* isomer should have somewhat greater stability.

*The *s-trans-trans* or *s-cis-trans* configurations take into account the different positions of the N₃ and N₁ atoms relative to the C₂-C_{2'} bond (*s-cis-trans* isomerism [4]) and of the rings relative to the azomethine C=N bond (*cis-trans* isomerism). Other possible *s-trans-cis* and *s-cis-cis* forms are not realizable [4].

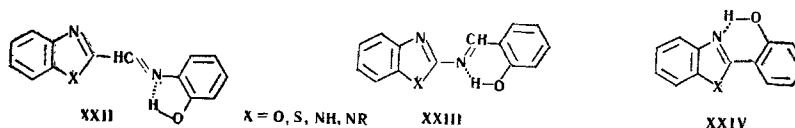
In order to investigate the steric and electronic effects in the azole series by the method of dipole moments, we studied the configurations of N- and C-aryl-substituted imidazoles and benzimidazoles (XV-XVI) [52], 1-(α , β - or γ -pyridyl)pyrazoles (XVII), and their 5-hydroxy- and 5-amino derivatives (XVIII and XIX, respectively) [11, 14, 53-55], 1-azolylbenzimidazoles (XX) [56-58], and 5-hydroxy (amino)-1-benzazolyl-pyrazoles (XXI) [59].



A comparison of the experimentally obtained dipole moments with the values calculated for the cis and trans planar configurations of XV-XXI, as in the case of the data presented earlier, makes it possible to draw the following conclusions:

1. Nonplanar structures are realized for XV-XIX ($n = 0$; $N = \beta$ or γ). In the case of N-benzimidazolyl-2-benzimidazole, as follows from its μ_{exp} (4.38 D) and the results of quantum-mechanical calculations [58], a configuration with rotation of the azole rings by 70° relative to one another is characteristic. For other systems of the XV-XX type, the angles of rotation range from 50 to 80° [11, 53, 54].
2. Bisheterocyclic systems [XVIII and XIX ($n = 0$), XXI] that contain substituents that are capable of forming intramolecular hydrogen bonds (intra HB) have exclusively planar configurations fixed by the intra HB [11, 53, 54, 59]. This conclusion is confirmed not only by the results obtained in the determination of the dipole moments but also by the results of an investigation of the IR and UV spectra [53, 54, 59]. Of particular interest in this connection is the eight-membered intra HB ring that we observed in [55], which fixes the planar structure for XIX ($X = \text{NH}_2$, $n = 2$).
3. The type of conformation (*s*-cis or *s*-trans) in the bisazole systems (XVII-XXI), in C- and N-aryl- (XV-XVI) and carbonylazoles (VIII-XI), and in azo- (XIII) and azomethine (XII) derivatives of the azole series depends substantially on the steric ortho effect as well as on the electrostatic interaction of the heteroatoms in the azole rings. In the absence of appreciable steric hindrance, the latter factor may become the determining one. It is precisely this effect that leads to the predominant realization of the *s*-trans configuration of carbonylazoles (VIII) and azomethines of the azole series (XII) [45, 48-51].
4. When the steric conditions are similar, the electronic nature of the substituent proves to have an appreciable effect on the conformation of the molecule. We recorded this effect particularly distinctly in a study of the structures of 2-(*p*- and *m*-nitrophenyl)benzimidazole and 4,5-diphenylimidazole (XVI) [52]. Despite the absence of appreciable steric hindrance, these molecules do not exist as planar isomers in solution. The insufficient energy of stabilization of the planar forms is apparently associated with the low degree of conjugation between the rings. The latter becomes understandable if one considers the noncompleteness of the para substituents in this case: both the nitro group and the 2-benzimidazolyl grouping have clearly expressed electron-acceptor character [57, 58].

In conclusion, we note that the method of dipole moments makes it possible not only to establish the presence of intra HB in azole molecules, but also to make a judgment regarding their types [48-50, 60]. On the basis of the use of a computational method [13] in conjunction with IR spectroscopic data, it has been shown that intra HB are realized in *o*-hydroxyanils of heterocyclic aldehydes (XXII) with the formation of a five-membered ring without delocalization of the π electrons through the hydrogen bridge. On the other hand, a six-membered, quasi-aromatic H bond ring is realized for salicylal-2-aminobenzazoles (XXIII) [48]. The intra HB in 2-hydroxy-phenylbenzazoles (XXIV) are similar in character [60, 61].



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