

Optical Anisotropies of Some Organic Molecules

BY M. A. LASHEEN AND I. H. IBRAHIM

Physics Department, Faculty of Science, Alexandria University, Alexandria, Egypt

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A quantitative knowledge of the refractivities and anisotropies of some organic molecules, aromatic and aliphatic, of different shapes and containing various electronic groupings was obtained. The studied aromatic compounds include three rings and one ring with different substituents while the aliphatic compounds contain six-membered rings, five-membered rings and open chains. The results show that strong magnetic anisotropy is always associated with strong optical anisotropy, and the optical anisotropy of the molecules divided by that of the benzene molecule is smaller than the corresponding value for magnetic anisotropy. Also the optical anisotropy in the case of aromatic compounds is markedly decreased by substitution.

Introduction

The optical properties of crystals and those of the molecules may be used to provide the starting point for trial structures and thus would help in the interpretation of the X-ray diffraction patterns.

In the present work the principal refractive indices of some organic substances, of known crystal structure, have been measured with the polarizing microscope and the 'Becke test' for sodium light at about 22°C. Some of these substances are orthorhombic and the rest are monoclinic.

The gram molecular refractivity R along any of the principal axes of a crystal can be deduced according to Lorentz-Lorenz equation

$$R = \frac{(n^2 - 1)}{(n^2 + 2)} \cdot \frac{M}{D},$$

where n is the refractive index of the crystal along that axis, M the molecular weight and D the density. In the case of orthorhombic crystals the principal refractivities along the crystal axes a , b and c are symbolized by R_a , R_b and R_c . For monoclinic crystals, R_1 , R_2 and R_3 are the principal refractivities of the crystal, where R_3 is parallel to the b axis and R_1 and R_2 are the values in the (010) plane.

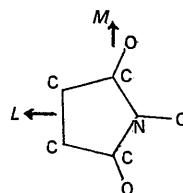
The transition from crystalline to molecular refractivities was carried out by the use of molecular direction cosines and the mathematical relations given by Lonsdale & Krishnan (1936). The molecular refractivities are denoted by r_L and r_M in the molecular plane and r_N normal to it, and the molecular anisotropy $\Delta r = (r_L + r_M)/2 - r_N$; r_L is usually along the length of the molecule.

It was noticed that small experimental errors in the crystal refractive indices could lead in some cases to very large errors in the molecular refractivities and anisotropies. These probable errors were calculated for all the substances investigated by methods similar to

that given for the study of errors in the molecular susceptibilities of orthorhombic crystals by Lasheen & Tadros (1968) and of monoclinic crystals by Walley (1967).

Orthorhombic crystals

(1) *N*-Chlorosuccinimide, $C_4H_4O_2NCl$, and (2) *N*-bromosuccinimide, $C_4H_4O_2NBr$



The crystal structure of *N*-chlorosuccinimide and the unit cell dimensions and space group of *N*-bromosuccinimide were determined by Brown (1961). The two crystals form an isomorphous pair, of unit-cell dimensions:

N-chlorosuccinimide: $a = 6.41$, $b = 7.11$, $c = 11.69$ Å
 $Z = 4$; space group $P2_12_12_1$.

N-bromosuccinimide: $a = 6.48$, $b = 7.25$, $c = 11.86$ Å
 $Z = 4$; space group $P2_12_12_1$.

Both *N*-chlorosuccinimide and *N*-bromosuccinimide contain a five-membered ring and two $\gg C=O$ double bonds.

N-Chlorosuccinimide crystals were obtained from solution in benzene by slow evaporation and cooling from about 40°C. They are fine prismatic large plates with (001) as the plate face and (110), (010) and (011) well developed. *N*-Bromosuccinimide crystals having the above habit were obtained from solution in benzene under the same conditions as those of *N*-chlorosuccinimide.

The principal refractive indices of the two compounds are:

	n_a	n_b	n_c
<i>N</i> -chlorosuccinimide	1.688	1.616	1.661
	± 0.003	± 0.003	± 0.004
<i>N</i> -bromosuccinimide	1.706	1.641	1.681
	± 0.003	± 0.002	± 0.003

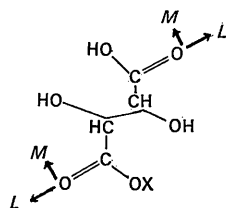
Thus the gram molecular refractivities of the two compounds are:

	M	D	R_a	R_b	R_c
<i>N</i> -chlorosuccinimide	133.54	1.65	30.87	28.33	29.95
			± 0.23	± 0.23	± 0.31
<i>N</i> -bromosuccinimide	178.00	2.109	32.84	30.45	31.93
			± 0.24	± 0.15	± 0.24

Hence the molecular refractivities of the two compounds are:

	r_L	r_M	r_N	Δr
<i>N</i> -chlorosuccinimide	29.42	32.28	27.40	3.46
	± 0.33	± 0.36	± 0.23	± 0.58
<i>N</i> -bromosuccinimide	31.43	34.17	29.58	3.22
	± 0.23	± 0.40	± 0.11	± 0.43

(3) *Ammonium hydrogen d-tartrate*, $C_4H_4O_6(NH_4) \cdot H$, and (4) *potassium hydrogen d-tartrate*, $C_4H_4O_6 \cdot K \cdot H$.



The crystal structure of ammonium hydrogen *d*-tartrate and the cell dimensions and space group of potassium hydrogen *d*-tartrate were determined by van Bommel & Bijvoet (1958). The cell dimensions are:

ammonium hydrogen <i>d</i> -tartrate:	$a = 7.648$, $b = 11.066$, $c = 7.843 \text{ \AA}$; $Z = 4$; space group $P2_12_12_1$;
potassium hydrogen <i>d</i> -tartrate:	$a = 7.64$, $b = 10.62$, $c = 7.75 \text{ \AA}$; $Z = 4$; space group $P2_12_12_1$

As in the crystal structure of all tartrates so far examined, the carbon chain and the oxygen and carbon atoms of each half-CHOH-COOH of the tartaric acid molecule are nearly planar.

Large fine crystals of the ammonium salt and small crystals of the potassium salt were obtained from the solutions of the two salts in water and tartaric acid (after the salt is dissolved in water tartaric acid is added). Both crystals are prismatic plates with (010)

as the plate face and (011) developed only in the ammonium salt crystals.

The principal refractive indices of the two isomorphous crystals are:

	n_a	n_b	n_c
ammonium hydrogen <i>d</i> -tartrate	1.534	1.500	1.510
	± 0.001	± 0.002	± 0.002
potassium hydrogen <i>d</i> -tartrate	1.566	1.534	1.544
	± 0.002	± 0.003	± 0.002

Thus the gram molecular refractivities of the two compounds are:

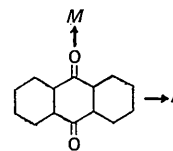
	M	D	R_a	R_b	R_c
ammonium hydrogen <i>d</i> -tartrate	167.07	1.68	30.92	29.25	29.74
			± 0.09	± 0.18	± 0.18
potassium hydrogen <i>d</i> -tartrate	188.18	1.954	31.41	29.94	30.40
			± 0.18	± 0.27	± 0.18

Hence the molecular refractivities of the two compounds for only half of each molecule are:

	r_L	r_M	r_N	Δr
ammonium hydrogen <i>d</i> -tartrate	29.58	31.88	28.46	2.27
	± 0.08	± 0.10	± 0.20	± 0.21
potassium hydrogen <i>d</i> -tartrate	30.37	32.19	29.26	2.02
	± 0.50	± 0.13	± 0.06	± 0.25

Monoclinic crystals

(1) *Anthraquinone*, $C_{14}H_8O_2$



The crystal structure of anthraquinone was refined by Murty (1960) who gave: $a = 15.810$, $b = 3.942$, $c = 7.865 \text{ \AA}$, $\beta = 102^\circ 43'$; $Z = 2$; space group $P2_1/a$.

The different atoms of the anthraquinone molecule were found to conform closely to a planar configuration.

Crystals of anthraquinone were crystallized from glacial acetic acid.

The principal refractive indices of anthraquinone are:

Winchell (1943)

$$n_x = 1.506, \quad n_y = 1.698, \quad n_z = 1.816$$

Present work

$$n_1 = 1.875 \pm 0.003, \quad n_2 = 1.717 \pm 0.003, \quad n_3 = 1.486 \pm 0.002.$$

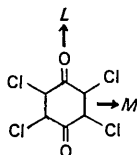
Thus the gram molecular refractivities of anthraquinone, where $M=208.20$ and $D=1.438$, are:

$$R_1 = 66.04 \pm 0.43, R_2 = 57.00 \pm 0.43, R_3 = 41.57 \pm 0.26.$$

Hence the molecular refractivities and the anisotropy of anthraquinone are:

$$r_L = 74.91 \pm 9.62, r_M = 57.16 \pm 8.48, \\ r_N = 32.54 \pm 0.02, \Delta r = 33.49 \pm 9.03.$$

(2) *Tetrachloro-p-benzoquinone (chloranil)*, $C_6Cl_4O_2$



The crystal structure of tetrachloro-*p*-benzoquinone has been determined by Chu, Jeffrey & Sakurai (1962) who gave: $a=8.708$, $b=5.755$, $c=8.603$ Å, $\beta=105^\circ 51'$; $Z=2$; space group $P2_1/a$.

Crystals were grown from acetone as well-developed transparent yellow rhombic plates with (001) as the plate face and [100] and [010] along the diagonals.

The principal refractive indices of tetrachloro-*p*-benzoquinone are:

$$n_1 = 1.522 \pm 0.002, n_2 = 1.621 \pm 0.003, \\ n_3 = 1.589 \pm 0.002.$$

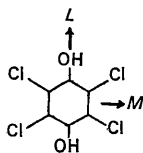
Thus the gram molecular refractivities of tetrachloro-*p*-benzoquinone, where $M=245.89$ and $D=1.712$, are:

$$R_1 = 43.81 \pm 0.26, R_2 = 50.51 \pm 0.41, \\ R_3 = 48.40 \pm 0.26.$$

Hence the molecular refractivities and the anisotropy of tetrachloro-*p*-benzoquinone are:

$$r_L = 52.17 \pm 2.83, r_M = 50.51 \pm 0.77, \\ r_N = 40.04 \pm 1.13, \Delta r = 11.31 \pm 0.67.$$

(3) *Tetrachlorohydroquinone*, $C_6Cl_4(OH)_2$



The crystal structure of tetrachlorohydroquinone has been determined by Sakurai (1962) who gave: $a=8.214$, $b=4.843$, $c=12.441$ Å, $\beta=123^\circ 49'$; $Z=2$; space group $P2_1/c$.

The crystals were grown by the slow evaporation of alcoholic solution. They were light brown in colour and of parallelepipedal shape elongated along [010], (104) being the plate face.

The principal refractive indices of tetrachlorohydroquinone are:

$$n_1 = 1.676 \pm 0.003, n_2 = 1.625 \pm 0.002, \\ n_3 = 1.636 \pm 0.003.$$

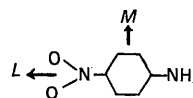
Thus the gram molecular refractivities of tetrachlorohydroquinone, where $M=247.90$ and $D=2.002$, are:

$$R_1 = 46.58 \pm 0.36, R_2 = 43.77 \pm 0.23, \\ R_3 = 44.39 \pm 0.36.$$

Hence the molecular refractivities and the anisotropy of tetrachloro-hydroquinone are:

$$r_L = 45.64 \pm 2.23, r_M = 49.17 \pm 1.82, \\ r_N = 39.94 \pm 1.37, \Delta r = 7.46 \pm 0.66.$$

(4) *4(p)-Nitroaniline*, $C_6H_4(NO_2) \cdot NH_2$



The crystal structure of 4(*p*)-nitroaniline has been determined by Trueblood, Goldish & Donohue (1961) who gave: $a=12.336$, $b=6.07$, $c=8.592$ Å, $\beta=91.45^\circ$; $Z=4$; space group $P2_1/n$.

Crystals of 4-nitroaniline were obtained as thin, bright yellow plates by slow evaporation of a chloroform solution. They are very fragile and cleave easily parallel to (101).

The principal refractive indices of 4-nitroaniline are:

$$\text{Winchell (1943)} \\ n_x = 1.556, \quad n_y = 1.78 \quad n_z = -$$

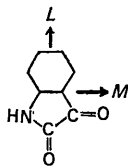
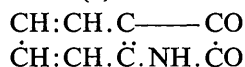
$$\text{Present work} \\ n_1 = 1.788 \pm 0.004, n_2 = 1.525 \pm 0.002, n_3 = 1.756 \pm 0.003.$$

Thus the gram molecular refractivities of 4-nitroaniline, where $M=138.12$ and $D=1.437$, are:

$$R_1 = 40.63 \pm 0.38, R_2 = 29.46 \pm 0.18, \\ R_3 = 39.40 \pm 0.28.$$

Hence the molecular refractivities and the anisotropy of 4-nitroaniline are:

$$r_L = 38.60 \pm 1.91, r_M = 42.54 \pm 2.72, \\ r_N = 28.34 \pm 0.03, \Delta r = 12.23 \pm 2.29.$$

(5) *Isatin* (2,3-indolinedione; *isatic acid lactam*),

Goldschmidt & Llewellyn (1950) determined the structure of isatin and gave the following cell dimensions: $a=6.19$, $b=14.46$, $c=7.17 \text{ \AA}$, $\beta=94^\circ 49'$; $Z=4$; space group $P2_1/c$.

The crystals used in the present work were crystallized from acetone in the form of prismatic needles elongated along [001] with (010) as the predominant face.

The principal refractive indices of isatin are:

Winchell (1943)

$$n_x = 1.46 \pm 0.005, \quad n_y = 1.80 \pm 0.03, \quad n_z = 1.90 \pm 0.03$$

Present work

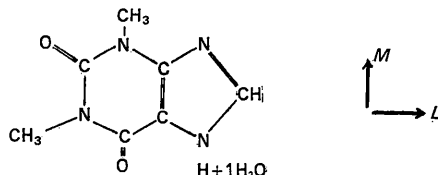
$$n_1 = 1.886 \pm 0.005, \quad n_2 = 1.453 \pm 0.002, \quad n_3 = 1.782 \pm 0.004.$$

Thus the gram molecular refractivities of isatin, where $M=147.05$ and $D=1.527$, are:

$$\begin{aligned} R_1 &= 44.31 \pm 0.48, \quad R_2 = 26.03 \pm 0.17, \\ R_3 &= 40.47 \pm 0.38. \end{aligned}$$

Hence the molecular refractivities and the anisotropy of isatin are:

$$\begin{aligned} r_L &= 40.27 \pm 1.64, \quad r_M = 44.17 \pm 0.73, \\ r_N &= 26.37 \pm 0.01, \quad \Delta r = 15.86 \pm 1.18. \end{aligned}$$

(6) *Theophylline* (1,3-dimethylxanthine monohydrate), $\text{C}_7\text{H}_8\text{N}_4\text{O}_2 \cdot \text{H}_2\text{O}$ 

The crystal structure of theophylline, which contains a six-membered ring (a substituted pyrimidine ring) and a five-membered ring (an imidazole ring) sharing a double C=C bond, was determined by Sutor (1958) who gave: $a=13.3$, $b=15.3$, $c=4.5 \text{ \AA}$, $\beta=99.5^\circ$; $Z=4$; space group $P2_1$.

The crystals used in the present work were crystallized from aqueous solution as needles elongated along [001] with faces (110).

The principal refractive indices of theophylline are:

Winchell (1943)

$$n_1 = 1.528, \quad n_2 = 1.69,$$

Winchell (1943)

$$n_1 = 1.447, \quad n_2 = 1.695, \quad n_3 = 1.733$$

Present work

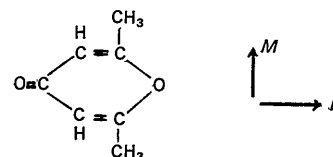
$$n_1 = 1.47 \pm 0.002, \quad n_2 = 1.687 \pm 0.004, \quad n_3 = 1.641 \pm 0.003.$$

Thus the gram molecular refractivities of theophylline, where $M=198.275$ and $D=1.454$, are:

$$\begin{aligned} R_1 &= 36.57 \pm 0.25, \quad R_2 = 52.09 \pm 0.52, \\ R_3 &= 49.19 \pm 0.39. \end{aligned}$$

Hence the molecular refractivities and the anisotropy of theophylline are:

$$\begin{aligned} r_L &= 51.96 \pm 0.09, \quad r_M = 49.54 \pm 2.02, \\ r_N &= 36.36 \pm 0.77, \quad \Delta r = 14.39 \pm 1.83. \end{aligned}$$

(7) *2,6-Dimethyl- γ -pyrone*, $\text{C}_7\text{H}_8\text{O}_2$ 

The crystal structure of 2,6-dimethyl- γ -pyrone, which contains a six-membered ring (a γ -pyrone ring), was determined by Brown, Norment & Levy (1957) who gave: $a=7.672$, $b=7.212$, $c=13.92 \text{ \AA}$, $\beta=120^\circ 59'$; $Z=4$; space group $P2_1/c$.

Large prismatic crystals of 2,6-dimethyl- γ -pyrone were obtained from its aqueous solution with (100) and (20 $\bar{1}$) as the prominent faces; (001) appeared occasionally. After some trials it was possible to grow small platy crystals with (100) as the plate face which are just suitable for optical measurements.

The principal refractive indices of 2,6-dimethyl- γ -pyrone are:

$$n_1 = 1.556 \pm 0.001, \quad n_2 = 1.463 \pm 0.001, \quad n_3 = 1.535 \pm 0.001.$$

Thus the gram molecular refractivities of 2,6-dimethyl- γ -pyrone, where $M=124.06$ and $D=1.234$, are:

$$\begin{aligned} R_1 &= 32.31 \pm 0.09, \quad R_2 = 27.69 \pm 0.09, \\ R_3 &= 31.30 \pm 0.09. \end{aligned}$$

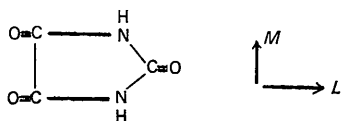
Hence the molecular refractivities and the anisotropy of 2,6-dimethyl- γ -pyrone are:

$$\begin{aligned} r_L &= 31.94 \pm 1.97, \quad r_M = 32.57 \pm 1.74, \\ r_N &= 26.79 \pm 0.02, \quad \Delta r = 5.47 \pm 1.84. \end{aligned}$$

Table 1. *Molecular magnetic anisotropies and optical anisotropies and their ratios with respect to that of one benzene molecule*

	Formula	Molecular susceptibilities				Molecular refractivities				$\frac{\Delta r_{\text{mol.}}}{\Delta r_{\text{benz.}}}$	
		K_L	K_M	K_N	ΔK	$\frac{\Delta K_{\text{mol.}}}{\Delta K_{\text{benz.}}}$	r_L	r_M	r_N		Δr
Orthorhombic crystals											
<i>N</i> -Chlorosuccinimide	$C_4H_4O_2NCl$	-64.50	-51.76	-76.96	18.83 ⁽¹⁾	0.32	29.42 ± 0.33	32.28 ± 0.36	27.40 ± 0.23	3.46 ± 0.58	0.22
<i>N</i> -Bromosuccinimide	$C_4H_4O_2NBr$	-76.03	-61.57	-87.31	18.51 ⁽¹⁾	0.31	31.43 ± 0.23	34.17 ± 0.40	29.58 ± 0.11	3.22 ± 0.43	0.21
Ammonium hydrogen d-tartrate (for half the molecule)	$C_4H_4O_6(NH_4)_2 \cdot H$	-37.41	-38.57	-46.80	8.81 ⁽¹⁾	0.15	29.58 ± 0.08	31.88 ± 0.10	28.46 ± 0.20	2.27 ± 0.21	0.14
Potassium hydrogen d-tartrate (for half the molecule)	$C_4H_4O_6 \cdot K \cdot H$	-38.31	-39.04	-47.26	8.59 ⁽¹⁾	0.14	30.37 ± 0.50	32.19 ± 0.13	29.26 ± 0.06	2.02 ± 0.25	0.13
Monoclinic crystals											
Anthraquinone	$C_{14}H_8O_2$	-76.1	-64.5	-217.9	147.6 ⁽¹⁾	2.47	74.91 ± 9.62	57.16 ± 8.48	32.54 ± 0.02	33.49 ± 9.03	2.13
Tetrachloro- <i>p</i> -benzo- quinone (chloranil)	$C_6Cl_4O_2$	-84.4	-98.5	-138.5	47.0 ⁽¹⁾	0.79	52.17 ± 2.83	50.51 ± 0.77	40.04 ± 1.13	11.31 ± 0.67	0.72
Tetrachlorohydroquinone	$C_6Cl_4(OH)_2$	-103.0	-114.3	-144.9	36.3 ⁽¹⁾	0.61	45.64 ± 2.23	49.17 ± 1.82	39.94 ± 1.37	7.46 ± 0.66	0.48
4(<i>p</i>)-Nitroaniline	$C_6H_4(NO_2) \cdot NH_2$	-52.0	-43.0	-104.8	57.3 ⁽¹⁾	0.96	38.60 ± 1.91	42.54 ± 2.72	28.34 ± 0.03	12.23 ± 2.29	0.79
Isatin	$CH:CH \cdot C \begin{array}{l} \text{---} \\ \text{---} \end{array} \begin{array}{l} \text{CO} \\ \text{CO} \end{array}$	-62.0	-57.3	-124.2	64.6 ⁽¹⁾	1.08	40.27 ± 1.64	44.17 ± 0.73	26.37 ± 0.01	15.86 ± 1.18	1.01
Theophylline	$C_7H_8N_4O_2 \cdot H_2O$	-63.4	-64.7	-124.5	60.4 ⁽²⁾	1.01	51.96 ± 0.09	49.54 ± 2.02	36.36 ± 0.77	14.39 ± 1.83	0.92
2,6-Dimethyl- γ -pyrone	$C_7H_8O_2$	±1.0	±2.1	±1.1	±1.6						
Parabanic acid	$CO(NH \cdot CO)_2$	-62.9	-59.7	-86.6	25.3 ⁽²⁾	0.42	31.94 ± 1.97	32.57 ± 1.74	26.79 ± 0.02	5.47 ± 1.86	0.35
		±3.0	±2.5	±0.7	±2.1						
		-28.6	-27.5	-49.9	21.9 ⁽¹⁾	0.37	23.91 ± 1.63	17.68 ± 0.10	16.19 ± 1.95	4.60 ± 1.08	0.29

(1) Lasheen (1964), (2) Lasheen (1968).

(8) *Parabanic acid (imidazoletrione oxalylurea),*
CO(NH.CO)₂

The crystal structure of parabanic acid, which contains a five-membered ring and three >C=O double bonds, has been determined by Davies & Blum (1955). The cell dimensions are: $a=10.685$, $b=8.194$, $c=5.054$ Å, $\beta=92^\circ 44'$; $Z=4$; space group $P2_1/n$.

Tabular crystals elongated along [001] were obtained from the aqueous solution of parabanic acid with (100) as the plate face and (010) developed. (010) is the cleavage plane.

The principal refractive indices of parabanic acid are:

Winchell (1943)

$$n_1 = 1.43, \quad n_2 = 1.55, \quad n_3 = -$$

Present work

$$n_1 = 1.441 \pm 0.002, \quad n_2 = 1.554 \pm 0.003, \quad n_3 = 1.479 \pm 0.002.$$

Thus the gram molecular refractivities of parabanic acid, where $M=114.03$ and $D=1.713$, are:

$$R_1 = 17.57 \pm 0.12, \quad R_2 = 21.33 \pm 0.19, \\ R_3 = 18.88 \pm 0.12.$$

Hence the molecular refractivities and the anisotropy of parabanic acid are:

$$r_L = 23.91 \pm 1.63, \quad r_M = 17.68 \pm 0.10, \\ r_N = 16.19 \pm 1.95, \quad \Delta r = 4.60 \pm 1.08.$$

Conclusion

When the molecular refractivities are compared with the molecular susceptibilities of the same molecules, both shown in Table 1, it is seen that strong magnetic anisotropy (ΔK) is always associated with strong optical anisotropy (Δr).

It can also be seen that the optical anisotropy of each of the molecules of the crystals investigated di-

vided by that of the benzene molecule shown in Table 1 is smaller than the corresponding ratio for the magnetic anisotropy. This is because the induced magnetic dipoles do not interact on one another appreciably whereas the optically induced dipoles are strong and have a considerable effect on one another.

The molecular refractivities and anisotropy are related to the molecular shape and constitution, and to the bond character; and in the case of aromatic compounds they are strongly influenced by substitution, which, in general, markedly decreases the anisotropy. This decrease varies not only with the nature of the substituents (heavy atom, compact group or aliphatic chain) but with their positions on the nucleus.

In some special cases small experimental errors in the crystal refractive indices may lead to large apparent errors in the molecular refractivities and anisotropies. These probable errors were calculated in this work for all the substances investigated (Table 1).

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