

The Molecular Structure of [4]-Rotane

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A combined X-ray crystallography and gas electron-diffraction study has been carried out on [4]-rotane. The X-ray data were taken at 115 K and the electron-diffraction data at 364 K. The results of the two methods are in generally good agreement. The only apparently significant deviation concerns the central four-membered ring that is found planar in the crystal while the electron-diffraction study indicates either a deviation from the planarity of the four-membered ring or, more probably, a large amplitude motion. This deviation may be due to the fact that the gas study was carried out at a temperature 250 K higher than that of the crystal study. Further, the difference in state of aggregation may be of importance.

The title compound (see Fig. 1) has attracted great general interest due to the valence conditions in three- and four-membered ring systems and, in particular, in spiro arrangements. A series of structure studies using various experimental methods has been carried out.¹⁻⁵ The present study aims at a determination of the geometrical parameters with highest possible precision. The gas electron-diffraction data were taken already in 1977. For the final electron-diffraction refinements calculated vibrational amplitudes (u) and perpendicular correction coefficients (K) were included. The electron-diffraction data and those obtained by X-ray crystallography are in good agreement in spite of difference in temperature and state of aggregation. However, the geometrical data deviate somewhat from those obtained in an earlier study.^{1,2} The result of the present work is summarized in Table 5.

EXPERIMENTAL

(a) *Sample.* The sample of [4]-rotane was prepared by J. P. Barnier and J. M. Conia,²⁻⁴ Laboratoire des Carbocycles, Université de Paris-Sud, Batiment 420, F-91405 Orsay Cédex.

(b) *X-Ray crystallography.* Crystals were formed by sublimation; the specimen used for data collection had dimensions $0.2 \times 0.3 \times 0.4$ mm. Data were collected on a SYNTEX PT diffractometer with a temperature of 115 K at the crystal site, using graphite crystal monochromated $MoK\alpha$ radiation ($\lambda = 0.71069 \text{ \AA}$). Cell parameters were determined by a least-squares fit to the diffractometer settings for 15 general reflections. Intensity data were recorded with the θ - 2θ scan technique, scan speed (2θ) was $2-4^\circ \text{ min}^{-1}$ depending on the intensity, scan width was from 1.0° below $2\theta(\alpha_1)$ to 1.2° above $2\theta(\alpha_2)$; background counts were taken at each of the scan limits for 0.35 times the scan time. Three standard reflections were measured after every 100 reflections; variations up to 4% were observed and the data were accordingly adjusted. All reflections in a quadrant of reciprocal space within $\sin\theta/\lambda = 0.65 \text{ \AA}^{-1}$ were measured; at higher angles up to $\sin\theta/\lambda = 0.99 \text{ \AA}^{-1}$ intensities were measured if higher intensities than a preset value were found for a 2 sec prescan. Of the 2324 independent reflections measured 2184, with $I > 2.5 \sigma(I)$ were retained for the structure determination. The estimate of the standard deviation of the intensity was based on counting statistics with an additional term of 2% of the net intensity. The data were corrected for Lorentz and polarization effects.

A description of the computer programs applied for the structure analysis is given in Ref. 6. Atomic scattering factors were taken from Ref. 7 for carbon and Ref. 8 for hydrogen.

The crystal data are as follows: [4]-Rotane,

Table 1. The final X-ray parameters for [4]-rotane.

Atom	X	Y	Z	U11	U22	U33	U12	U13	U23
C1	-.09950(6)	.08038(6)	.03393(4)	.0179(2)	.0158(1)	.0169(1)	.0022(1)	.0079(1)	.0004(1)
C5	-.22892(11)	.24500(8)	.01326(8)	.0318(3)	.0231(2)	.0328(2)	.0115(2)	.0170(2)	.0044(2)
C6	-.21061(10)	.10693(9)	.13552(7)	.0302(2)	.0302(2)	.0273(2)	.0054(2)	.0191(2)	.0015(2)
C2	.12393(6)	.07413(6)	.07699(4)	.0171(2)	.0161(1)	.0148(1)	-.0011(1)	.0052(1)	-.0003(1)
C7	.26545(9)	.23181(8)	.11033(6)	.0258(2)	.0221(2)	.0236(2)	-.0075(1)	.0073(2)	-.0001(1)
C8	.27976(9)	.09289(9)	.23067(6)	.0254(2)	.0283(2)	.0172(2)	-.0060(2)	.0017(1)	.0008(1)

Atom	X	Y	Z	B	Atom	X	Y	Z	B
H51	-.362(6)	.258(6)	-.091(5)	3.3(5)	H52	-.159(7)	.370(7)	.043(5)	3.5(5)
H61	-.318(7)	.023(7)	.126(5)	3.5(5)	H62	-.113(7)	.144(6)	.256(5)	3.5(5)
H71	.207(10)	.360(10)	.118(7)	4.4(7)	H72	.356(5)	.231(5)	.058(4)	2.9(4)
H81	.224(7)	.124(6)	.315(5)	3.5(5)	H82	.395(6)	-.001(5)	.264(4)	2.9(4)

Table 2. The final X-ray structure data for [4]-rotane.

Bond lengths (Å)	Corr.	Bond angles (°)
C1-C5	1.501	C5-C1-C6
C1-C6	1.499(1)	C2-C1-C5
C5-C6	1.520(1)	C5-C1-C4
C1-C2	1.523	C6-C1-C2
C1-C4	1.521(1)	C6-C1-C4
C1-C7	1.520(1)	C2-C1-C4
C2-C7	1.499(1)	C1-C5-C6
C2-C8	1.497(1)	C5-C6-C1
C7-C8	1.517(1)	
C-H (mean)	1.05(6)	

Bond lengths (Å)	Corr.	Bond angles (°)
C1-C5	1.501	C5-C1-C6
C1-C6	1.499(1)	C2-C1-C5
C5-C6	1.520(1)	C5-C1-C4
C1-C2	1.523	C6-C1-C2
C1-C4	1.521(1)	C6-C1-C4
C1-C7	1.520(1)	C2-C1-C4
C2-C7	1.499(1)	C1-C5-C6
C2-C8	1.497(1)	C5-C6-C1
C7-C8	1.517(1)	
C-H (mean)	1.05(6)	

$C_{12}H_{16}$, subl. Monoclinic, $a=7.282(1)$ Å; $b=7.318(1)$ Å; $c=9.641(1)$ Å; $\beta=112.04(1)^\circ$; $V=476.2(1)$ Å³ ($T=115$ K). $M=160.26$; $Z=2$; $F(000)=176$; $\mu(\text{MoK}\alpha)=0.67$ cm⁻¹; $D_x=1.118$ g cm⁻³. Space group $P2_1/c$ (No. 14).

The structure was solved by direct methods⁹ and refined by Fourier and full-matrix least-squares methods. In order to reduce systematic errors due to the contribution to the structure factors from valence electrons, only high order reflections were used in the least-squares procedure; the best figures of merit were obtained by using only reflections with $\sin\theta/\lambda$ higher than 0.6 Å⁻¹. Introducing anisotropic thermal parameters for hydrogen atoms did not significantly improve the result, and with isotropic hydrogen atoms the refinements converged to a conventional R -value of 0.030 for 1404 reflections, (overall R -value of 0.043) $R_w=0.030$ and $S=(\sum w\Delta^2/(n-m))^2=1.18$. The overdetermination ratio was 16.1.

(c) *Electron diffraction.* The electron-diffraction data were recorded in 1977 by the Oslo Apparatus¹⁰ with nozzle-to-plate distances of 480.72 and 200.72 mm using Kodak Electron Image plates and a nozzle-tip temperature of 364 K. The electron-beam wavelength was 0.06469 Å as calibrated against diffraction patterns of gaseous benzene. The estimated standard deviation in the determination of the wavelength is 0.1 %. For each of the two camera distances six plates were selected for the structure analysis. The intensity data were obtained using a Joyce Loebel densitometer. A series of blackness corrections was tested for reasons to be described later on. A modification function $s/|f_c|^2$ was applied. The computer programs as well as the atomic scattering amplitudes and phases were applied according to the usual routine.¹²⁻¹⁴

The data range covers 1.25 – 19.25 Å⁻¹ for the 48 cm plates and 6.75 – 43.00 Å⁻¹ for the 20 cm plates. The Δs intervals are 0.125 and 0.25 Å⁻¹ for the two camera distances, respectively. Using the terminology from Ref. 13 the weight scheme is characterized by the values s_1 , s_2 , w_1 , and w_2 .

For the 48 cm plates these values are 4, 19, 0.1, and 2 Å², for the 20 cm plates 7, 40, 1 and 0.02 Å².

RESULTS

(a) *X-Ray investigation.* The final parameters are given in Table 1. Structural data are listed in Table 2; the estimated standard deviations given are calculated from the variance-covariance matrix. The structure factor listing may be obtained from the authors.

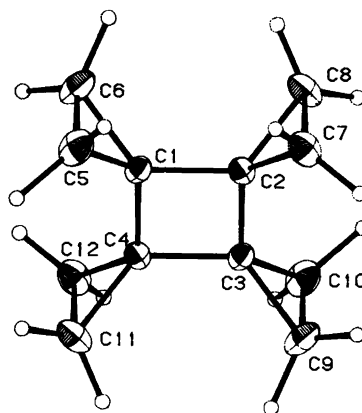


Fig. 1. ORTEP plot of [4]-rotane scaled to enclose 50 % probability.

An ORTEP plot of the molecule is presented in Fig. 1.

The X-ray analysis in general confirms the results of the previous investigation^{1,2} though the C–C bond lengths are not in complete agreement. The molecule possesses a centre of symmetry demanded by the space group symmetry and the central cyclobutane ring should thus be strictly planar in the crystal. The cyclopropane rings are within 1° normal to the central ring and the plane of the cyclobutane ring bisects the bond between the outer carbon atoms of the cyclopropane rings. There is a very good agreement between bond lengths and angles in the chemically equivalent parts of the molecule; mean values are given in Table 5 for comparison with the results from the electron-diffraction investigation.

Non-planarity of the cyclobutane ring was indicated by the electron-diffraction analysis and the thermal motion of the molecules in the crystals was therefore studied in some detail. The thermal parameters of the carbon atoms were analysed in terms of rigid body motion. The r.m.s. ΔU was found to be $9.7 \cdot 10^{-4}$ Å² indicating that the molecule to a good approximation behaves as a rigid body. The eigenvectors of T correspond to fairly isotropic translational vibrations, the r.m.s. amplitudes being 0.13, 0.12 and 0.11 Å, respectively, the smallest amplitude being that along the largest axis of inertia (*i.e.* the pseudo fourfold symmetry axis of the molecule). The eigenvalues of L correspond to oscillations with r.m.s. amplitudes of 3.6, 2.8 and 2.3° ,

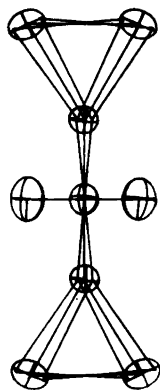


Fig. 2. Projection of [4]-rotane along one of the cyclobutane diagonals.

respectively. The largest libration is about the pseudo fourfold axis; the remaining two axes are parallel to the diagonals of the cyclobutane ring.

A semiempirical calculation¹¹ of the non-bonded potential energy connected with oscillations along and about the inertial axes reproduced the translational oscillations accurately and the librations well. The calculations also gave

the largest libration amplitude about the pseudo fourfold axis (4.6°) and smaller amplitudes (2.9° and 2.8°) along the diagonals of the cyclobutane ring. No double minimum potential curve was indicated.

Fig. 2 shows the projection of the molecule along one of the cyclobutane diagonals. The atomic positions after a twist of 5° about this diagonal are also indicated, simulating a librational motion. It is obvious that the X-ray method cannot differentiate between this libration and a symmetric out-of-plane oscillation of the two cyclopropane moieties; which would correspond to a deviation from planarity as suggested by the electron diffraction investigation. The effect in the crystal at 115 K is obviously expected to be less pronounced, corresponding to a ϕ larger than 174° .

(b) *Electron-diffraction structure studies.* The electron-diffraction intensity data and the corresponding radial distribution curve are presented in Figs. 3 and 4, respectively.

The structure refinements were based upon a few assumptions of symmetry (see Fig. 1): 1. The plane of each of the three-membered rings is perpendicular to the plane of the three adjacent

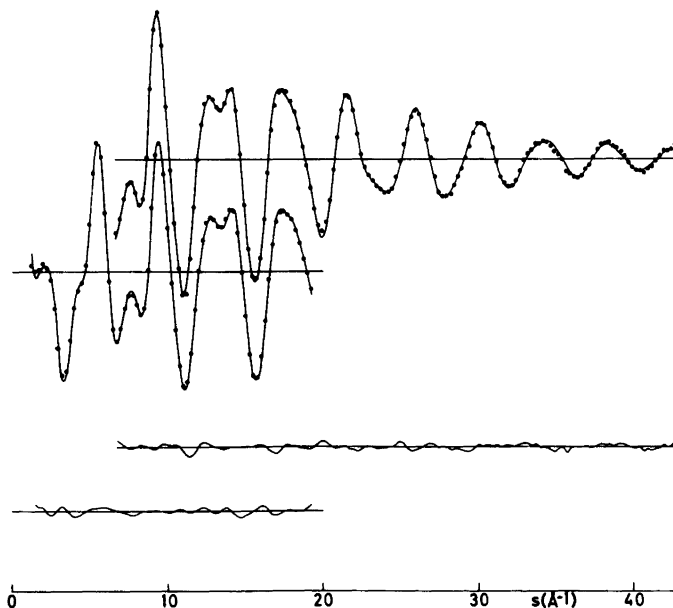


Fig. 3. Observed electron-diffraction molecular intensity data ($^\circ$) for [4]-rotane compared to calculated intensities (full line) of final structure refinement. Upper curve 48 cm and lower curve 20 cm camera distance, respectively. Difference curves between experimental and calculated data are also included.

atoms in the central four-membered ring. 2. The angle C2C1C4 is bisected by the plane of the adjacent three-membered ring. 3. The plane through C2C1C4 bisects the angle C5C1C6. 4. The geometry of the molecule is determined by six parameters: the bond distances C1–C2, C1–C5, C5–C6, C–H, the HCH angle and the angle ϕ describing a possible deviation from planarity in the four-membered ring. $\phi=180^\circ$ for a planar ring.

The most dominating peak in the radial distribution curve of [4]-rotane at about 1.5 Å is composed of contributions from three different C–C bond distances. Most of the other peaks in the radial distribution curve are also composed of contribution from several internuclear distances as may be seen from Fig. 4. In such a case, with serious overlap, the geometry of the molecule may be determined even with great precision if the vibrational amplitudes (u) and preferably, the perpendicular amplitude correction coefficients (K) are known. The u and K values are in

principle obtainable from spectroscopical observations. It is difficult to make quantitative statements as to the accuracy of the geometrical parameters in molecules with badly overlapping distances and the dependence of the accuracy of the u and K values. Apparently reliable and reproducible results have been obtained in a few cases with severe distance overlap.^{14,15} In the mentioned cases attempts have been made to obtain information about the influence of the geometric results with minor variations of the u values. Apparently the obtainable accuracy varies from molecule to molecule, and general statements as to the accuracy of the method are hard to make.

There were reasons to believe that the procedure used for [4]-rotane might be vulnerable to inaccuracies in blackness correction. The measured blackness correction generally used varies somewhat from time to time, probably due to minor changes in the film quality. In the present case four different sets of blackness corrections,

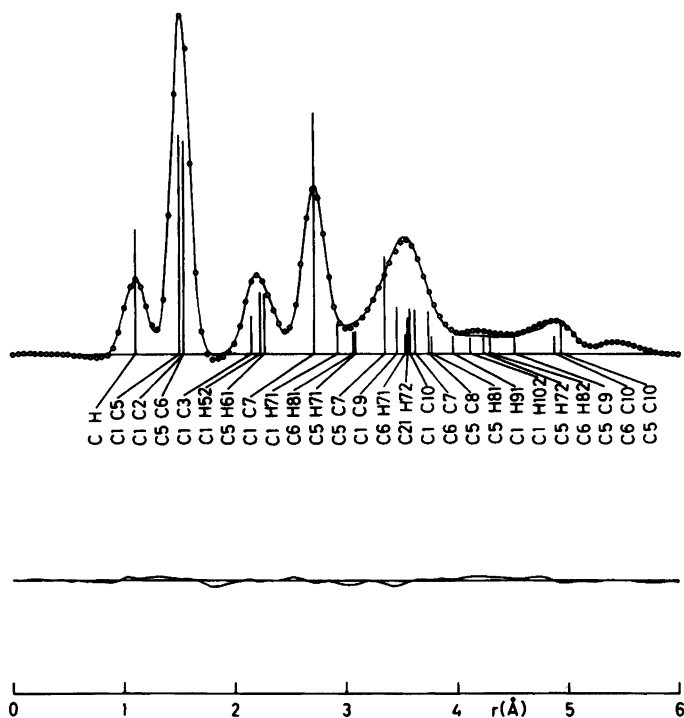


Fig. 4. Experimental (°) and calculated (full line) radial distribution curves for [4]-rotane, including difference curves between experimental and calculated data. The internuclear distances are indicated by vertical lines.

$1+aD+bD^2+cD^3$, were tested, using various combinations of the parameters a, b, c , chosen in a reasonable interval. (D is the optical density.) The initial test was carried out by particularly chosen least squares refinements, partly keeping u values constant and refining the geometry, partly keeping the geometry constant and refining the u values. In general, as expected, the blackness correction seems to have small effect on the geometry but some effect on the u values. The blackness correction chosen for the further work was the one that led to the smallest R factor in the least squares evaluation. At a late stage of the work it appeared that the blackness correction leading to u values in best agreement with those calculated from valence force field estimations was slightly different from that used. The main final refinements were therefore repeated using the new correction. This led to no appreciable change in the geometry of the molecule. We thus feel that the presumed inaccuracy due to blackness correction has been taken appropriate care of in the present study.

For a molecule like [4]-rotane where determination of molecular geometry from gas elec-

tron diffraction is assumed to rest upon rather accurate vibrational data, it is advisable as a first step in the refinement procedure to test what may be obtained using exclusively electron-diffraction data. This is done mainly in order to collect information about the reliability and possible defects of the procedure using fixed vibrational parameter.

Three different sets of u values were used for the geometry refinements. The first set is based upon u values estimated and refined from electron-diffraction data alone before calculated u values were available. The two other sets were based upon calculated u values (Table 3). The calculated data are based upon a diagonal valence force field estimated from related molecules, assuming the central four-membered ring planar and assuming a D_{4h} symmetry for the molecule. The only difference of the applied force field for alternative 1 and 2 lies in the size of the torsional force constant describing the inner ring deformation. For alternative 1 this numerical value has been chosen as 0.5 mdyne/Å and for alternative 2 as 0.1 mdyne/Å, leading to 73 and 38 cm^{-1} for the ring deformation frequency of the two versions, respectively. A preliminary spectroscopical investigation showed alternative 2 to be the most reasonable one. This alternative also leads to the smallest R factor in the electron-diffraction refinements.

Keeping the u values for each of the three sets constant the geometrical parameters were refined. The refinements were carried out (a) assuming the central four membered ring planar, (b) including a possible deviation from planarity. The results of these refinements are quite similar. Data obtained using the two alternative sets of calculated u values are given in Table 4. The refinements were carried out using diagonal weight matrices. The final refinement was carried out with complete matrices in order to obtain more realistic standard deviations.

The apparent deviation from planarity of the four-membered ring may be explained by the shrinkage effect. Refinements were, therefore, carried out including the shrinkage effect involving the perpendicular amplitude correction coefficients (K). This led to only a few degrees increment of the ϕ angle. However, this correction is not able to take proper care of large amplitude motion. (Results also included in Table 4.)

Table 3. Calculated u values in Å for [4]-rotane, alternatives 1 and 2.

	Alt. 1	Alt. 2
C1-C2	0.0489	0.0489
C1-C5	0.0484	0.0484
C5-C6	0.0486	0.0486
C-H	0.0774	0.0774
C1-C3	0.0535	0.0535
C1-C7	0.0749	0.0749
C5-C7	0.1335	0.1335
C1-C9	0.0783	0.0895
C5-C8	0.1105	0.1213
C5-C9	0.1385	0.1986
C5-C10	0.0821	0.0821
C1-H5,1	0.1077	0.1077
C5-H6,1	0.1078	0.1078
C1-H7,1	0.1580	0.1580
C5-H7,1	0.2214	0.2216
C1-H7,2	0.1266	0.1266
C5-H8,1	0.1984	0.2182
C1-H9,1	0.1573	0.1695
C5-H7,2	0.1793	0.1810
C5-H8,2	0.1315	0.1384
C5-H9,1	0.2230	0.2891
C5-H10,1	0.1576	0.1612

Table 4. Final electron diffraction geometrical data for [4]-rotane, distances given in Å and angles in °. Refinements are carried out under several different conditions: (1) The ϕ angle is kept constant equal to 180°, alternatively refined as an independent parameter. (2) Both the two alternative sets of u values are involved. (3) Refinements are carried out with and without spectroscopically estimated shrinkage.

	Without shrinkage		With shrinkage	
	u -values Alt. 1	u -values Alt. 2	u -values Alt. 1	u -values Alt. 2
Planar 4-membered ring				
C1–C2	1.5254(26)	1.5277(21)	1.5282(26)	1.5322(19)
C1–C5	1.4944(23)	1.4929(18)	1.4951(23)	1.4926(16)
C5–C6	1.5318(31)	1.5321(24)	1.5274(32)	1.5282(24)
C–H	1.1012(53)	1.1020(44)	1.1009(55)	1.1021(41)
ϕ	180.00	180.00	180.00	180.00
\angle C4–C1–C2	90.00	90.00	90.00	90.00
\angle C5–C1–C6	61.66(22)	61.74(16)	61.27(23)	61.43(16)
\angle C2–C1–C5	127.39(6)	127.37(4)	127.85(6)	128.68(4)
\angle CCH	120.89(72)	120.89(59)	122.73(76)	123.79(61)
\angle HCH	114.98(1.50)	115.34(1.24)	113.25(1.60)	113.36(1.26)
R%	7.732	6.378	7.918	6.235
Non-planar 4-membered ring				
C1–C2	1.5314(18)	1.5315(16)	1.5341(18)	1.5344(18)
C1–C5	1.4907(13)	1.4908(13)	1.4914(14)	1.4914(14)
C5–C6	1.5321(20)	1.5318(19)	1.5285(22)	1.5281(22)
C–H	1.1029(37)	1.1032(35)	1.1029(39)	1.1031(39)
ϕ	165.08(71)	166.44(83)	164.94(74)	168.21(1.07)
\angle C4–C1–C2	89.51(4)	89.60(4)	89.55(4)	89.74(5)
\angle C5–C1–C6	61.85(13)	61.83(13)	61.49(14)	61.48(14)
\angle C2–C1–C5	127.53(4)	127.50(4)	128.00(4)	128.79(4)
\angle CCH	121.43(49)	121.25(46)	123.23(55)	123.96(55)
\angle HCH	114.55(1.08)	114.87(1.00)	113.10(1.16)	113.20(1.16)
R%	5.505	5.130	5.693	5.645

COMPARISON OF THE RESULTS OBTAINED BY THE TWO METHODS

In Table 5 are presented the geometrical parameters considered to be the best estimates. For the X-ray data average values are given for bond distances that should be equal in an isolated molecule. For the electron-diffraction data those of the last column of Table 4 were elected as the best choice.

There seems to be no scale error, since the average of the C–C bond distances as obtained by the two methods deviates only by about 0.001 Å. The C–H bond distance obtained from X-ray crystallography is found slightly, though not significantly, shorter than that obtained from electron diffraction. This is normal since X-ray

crystallography measures distances between electron density maxima while electron diffraction essentially measures the internuclear distances.

The correspondence between the C–C bond

Table 5. Comparison between best sets of geometrical data obtained from the two experimental methods.

	X-Ray data	E.D. data
C1–C2 (Å)	1.525(1)	1.534(2)
C1–C5 (Å)	1.502(1)	1.491(1)
C5–C6 (Å)	1.521(1)	1.528(2)
C–H (Å)	1.05(6)	1.103(4)
ϕ (°)	180	168.2(11)

distances obtained by the two methods is quite satisfactory, though the differences are generally larger than what should be expected from the standard deviations. At least two explanations may be suggested. Firstly there may be molecular structure differences in free molecules compared to those packed in a crystal. Secondly the standard deviation may in general come out too optimistic, in particular for electron diffraction when u values are kept constant during the refinement.

The most striking difference is the apparent deviation from planarity of the central four-membered ring suggested by electron diffraction compared to the planarity suggested by X-ray crystallography. Several explanations may be considered. As, for example, in biphenyl and in some of its derivatives analogous structural differences are proved to exist in the crystalline and gaseous phase.¹⁶ It should be pointed out that the carbon skeleton in cyclobutane itself and in several of its derivatives deviates from planarity in the gas phase.¹⁶ Further, it is not unlikely that large amplitude motion takes place in the four-membered carbon ring of [4]-rotane. This motion may be hampered in the crystalline phase. The great temperature difference of the two investigations (about 250 K) may contribute to the effect. The amplitude of an intramolecular motion may be considerably larger under the high temperature conditions. The electron diffraction method could hardly discriminate between a large amplitude motion around $\phi=180^\circ$ and a case with an energy minimum different from $\phi=180^\circ$. Large amplitude motion is not taken fully care of by the shrinkage correction calculated in this case from spectroscopic consideration.

The bond distances as presented in Table 5 seem to fit well into the general picture obtained by structure studies of related compounds. The average C–C bond distance in the three-membered ring of [4]-rotane is 1.503 Å as measured by electron diffraction and 1.508 Å as measured by X-ray, *i.e.* approximately equal to or slightly shorter than the C–C bond distance in gaseous cyclopropane of 1.510 Å.¹⁷ The C–C bond distance in the four-membered ring of [4]-rotane is found to be shorter than the corresponding bond in cyclobutane. Two electron-diffraction gas studies of cyclobutane lead to nearly the same value, 1.548¹⁸ and 1.551 Å.¹⁹ It is also interesting to compare the results obtained for [4]-rotane

with the electron diffraction results of spiroentane.^{20,21} Firstly the average C–C bond distance in spiroentane is shorter than the C–C bond distance in cyclopropane. Secondly the central C–C bond distance in spiroentane is shorter than the peripheral one, in correspondence with the findings in [4]-rotane.

As a whole the small, but apparently significant systematic differences in bond distances in the above-mentioned molecules and in a possibly large series of related compounds, call for further study both experimental and theoretical.

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