

Atomic Arrangement in an Addition Compound in which Dinitrogen Tetroxide is the Acceptor Molecule

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The crystal structure of the 1:1 compound formed by dinitrogen tetroxide and 1,4-dioxan has been investigated using three-dimensional methods. The Dirichlet's reduced triclinic unit cell containing one formula unit has the parameters:

$$a = 5.46 \text{ \AA}; b = 6.47 \text{ \AA}; c = 6.81 \text{ \AA}; \alpha = 108.5^\circ; \beta = 91.8^\circ; \gamma = 108.4^\circ$$

The space group is $P\bar{1}$ and endless chains of alternating acceptor and donor molecules are running along the direction of the needle-axis, [111]. The structure is disordered, the dioxan molecules being able to take up at least two alternative orientations, but the dioxan oxygens occupy well defined positions and each of them is bonded to a nitrogen atom of an acceptor molecule with a N-O separation a little shorter than the expected van der Waals distance.

The dinitrogen tetroxide molecule is *planar* and its molecular parameters agree closely with those found in the vapour state¹ and in the (unstable) monoclinic modification.² A summary of our results was published in 1962.³

Atomic arrangements in addition compounds in which nitrogens may be suspected to act as the acceptor atoms appear to be of considerable interest, but so far little has been done to elucidate such structures. For this reason X-ray work was started several years ago in this laboratory on the well established 1:1 compound formed by dinitrogen tetroxide and 1,4-dioxan. Certain experimental difficulties had first to be overcome, however, because of the extreme sensibility of the crystals towards humidity. Even if the compound is prepared from carefully dried component substances the crystals tend to liquify in the sealed capillary tubes in which they have to be kept for X-ray investigation. They therefore had to be transferred into the capillaries without opening of the device in which they had been grown (by sublimation).

The crystals, colourless needles, turned out to be triclinic and to have a repeat period along the needle-axis which is not the shortest lattice period. The Dirichlet's reduced unit cell has the parameters:

$$a = 5.46 \text{ \AA}; b = 6.47 \text{ \AA}; c = 6.81 \text{ \AA}; \alpha = 108.5^\circ; \beta = 91.8^\circ; \gamma = 108.4^\circ$$

and contains one formula unit. The density computed from these figures is 1.30. Statistical criteria very strongly suggest the space group $P\bar{1}$.

Our X-ray material was collected chiefly from Weissenberg diagrams partly using $\text{CuK}\alpha$, partly $\text{MoK}\alpha$ radiation. Integrating Weissenberg cameras and the multiple film method were employed and the intensities measured photometrically.

The two-dimensional analysis was based on diagrams taken at room temperature except for the diagrams with rotation about the needle-axis which was taken below the temperature above which a transition, indicated by NMR measurements, takes place. The three-dimensional material was obtained at room temperature, — the level scale factors were determined with the help of oscillation diagrams.

Attempts were made to tackle the phase problem by applying Harker-Kasper inequalities to the $0kl$ and $h0l$ projections, but were not very successful. Eventually it proved possible, however, to interpret the (sharpened) Patterson chart of the first-mentioned projection and subsequently to refine the y and z coordinates using a least squares procedure which finally lowered the R_{0k} factor to 0.066. The corresponding Fourier map is reproduced in Fig. 1. The z parameters which were thus determined with considerable accuracy made it comparatively simple to interpret even the Patterson map for the $h0l$ projection. A least squares refinement was performed and led to an R_{h0l} -value of 0.075 and to the Fourier map reproduced in Fig. 2.

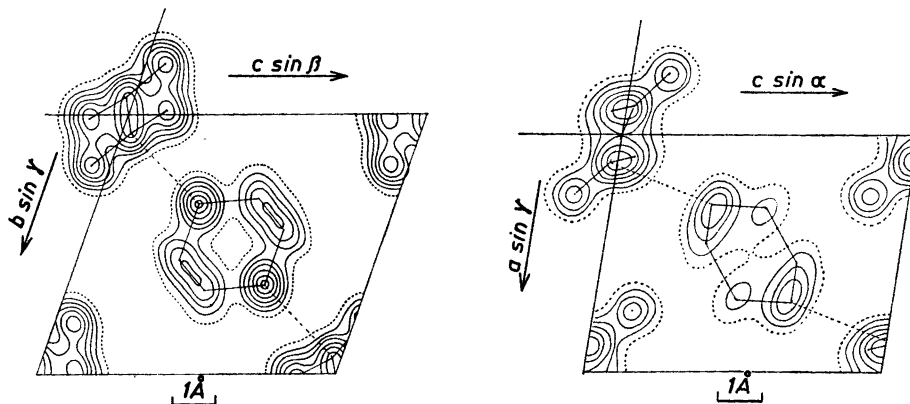


Fig. 1. Fourier projection along the a axis. Fig. 2. Fourier projection along the b axis.

The atomic arrangement deduced from the parameters thus obtained were, in general, very acceptable. The dinitrogen tetroxide molecule closely corresponded to that found in the electron diffraction investigation of its vapour¹ and in the solid state.² Further, the intramolecular distance between the two dioxan oxygen atoms was identical with that found in the "free" molecule.⁴ The localisation of the carbon atoms, was, however, not quite straightforward. During the later part of the least squares procedure the B factors of these atoms became suspiciously large and their images in the Fourier

Table 1. Final atomic parameters.

	<i>x</i>	<i>y</i>	<i>z</i>
O ₁	0.29866	0.34929	0.34256
O ₂	0.74019	0.99492	0.13600
O ₃	0.10155	0.79781	0.05934
N	0.88179	0.06689	0.02812
C ₁	0.41893	0.59656	0.36295
C ₂	0.71944	0.64074	0.43833
C ₃	0.50237	0.53260	0.30439
C ₄	0.81660	0.81660	0.44090

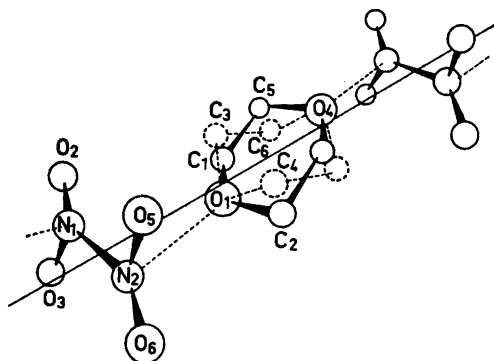


Fig. 3. Schematical drawing of the molecule.

map are somewhat blurred. These findings made us suspect that the structure might in fact be disordered with respect to the positions of the carbon atoms. Dr. phil. Björn Pedersen kindly carried out NMR measurements on the addition compound and found clear evidence of a transition occurring at temperatures above -40°C and connected with some kind of molecular motion. X-Ray diagrams taken at low temperatures do not, however, show great differences from those taken at room temperature and it must therefore be assumed that frozen-in statistical disorder prevails at low temperatures.

Table 2. Anisotropic thermal vibration parameters.

	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
O ₁	0.12637	0.07351	0.04784	0.05039	-0.05379	-0.04438
O ₂	0.09024	0.09854	0.06917	0.08327	0.06645	0.06987
O ₃	0.11877	0.08345	0.08567	0.10620	0.02712	0.08695
N	0.07054	0.05652	0.05034	0.04571	0.01533	0.03726
C ₁	0.12024	0.05164	0.03716	0.04608	-0.00578	0.01665
C ₂	0.05436	0.07582	0.05119	0.05163	0.03844	0.07949
C ₃	0.12312	0.05253	0.04929	0.02909	-0.02644	-0.04641
C ₄	0.11034	0.06895	0.04819	0.06816	-0.02636	-0.03595

Table 3. Observed and calculated structure factors.

$h k l$	$ F_o $	F_c	$h k l$	$ F_o $	F_c
0 0 2	14.6	13.6	0 4 1	0.9	— 0.2
3	11.0	9.7	2	0.6	— 0.6
4	3.5	— 3.5	3	2.1	— 2.1
5	1.9	— 1.3	4	1.1	— 0.9
6	3.1	2.6	0 5 8	1.0	— 1.0
7	0.7	— 0.6	6	0.8	— 0.6
8	0.5	0.3	5	3.2	— 2.9
0 1 8	0.6	0.2	4	4.2	— 4.8
7	1.7	1.7	3	0.9	— 0.7
5	3.3	— 3.2	2	2.5	— 2.6
4	4.2	3.5	1	2.4	— 0.4
3	16.8	— 15.4	0	1.4	— 1.5
2	5.2	5.3	1	1.8	— 1.8
1	37.8	35.2	2	0.9	— 1.2
0	5.2	5.4	3	0.7	— 0.5
1	31.4	30.6	0 6 8	0.6	— 0.5
2	17.1	16.5	7	0.9	— 0.9
3	1.0	— 1.1	6	1.2	— 0.5
5	3.6	2.8	5	2.0	— 2.2
6	0.6	— 0.9	4	1.1	— 1.3
0 2 9	0.5	0.4	3	1.8	— 2.1
8	1.1	1.0	2	1.9	— 2.3
7	2.2	2.0	1	0.8	— 0.2
6	1.4	0.9	0	0.5	— 0.3
5	4.8	4.1	1	0.6	— 0.8
4	10.2	— 8.6	2	0.6	— 0.2
3	2.0	— 1.6	0 7 6	0.7	— 0.6
0 2 2	8.7	— 7.1	5	0.8	— 1.0
0	4.4	4.8	4	0.7	— 0.9
1	10.1	9.8	3	1.4	— 1.5
2	4.1	— 4.2	2	0.9	— 1.1
3	1.0	— 0.4	1	0.7	— 0.7
4	1.0	0.5	0	0.6	— 0.7
5	0.8	— 1.1	1	0.8	— 0.3
0 3 9	0.8	0.6	0 8 4	1.0	— 0.4
8	1.3	1.3	1 7 2	1.3	— 1.2
7	1.7	1.7	7	0.7	— 0.5
6	4.6	4.5	1 6 0	1.4	— 0.8
5	3.9	3.7	1	2.5	— 2.4
3	2.3	1.9	2	1.7	— 1.5
2	3.8	3.2	3	1.6	— 1.5
1	5.8	— 4.1	4	4.6	— 4.8
0	9.0	8.7	6	1.1	— 1.1
1	3.5	— 2.8	1 5 1	0.9	— 0.6
2	3.1	— 2.4	0	2.2	— 2.3
3	1.6	— 1.3	1	2.9	— 2.7
4	2.1	— 2.0	3	4.2	— 4.0
5	0.9	— 0.6	5	4.3	— 4.5
0 4 8	0.8	0.5	6	2.5	— 2.8
7	2.5	2.3	1 4 2	3.3	— 2.7
6	3.0	3.1	1	3.8	— 3.9
5	1.1	— 1.3	0	4.5	— 4.0
4	8.0	8.0	1	3.2	— 2.6
2	1.1	1.0	2	5.5	— 5.7
1	9.4	9.6	3	7.2	— 6.8
0	1.3	0.2	4	0.7	— 0.6

$h k l$	$ F_o $	F_c	$h k l$	$ F_o $	F_c
1 $\bar{3}$ $\bar{3}$	5.0	- 3.8	1 2 2	10.1	7.7
	8.1	- 8.5	1 3 $\bar{7}$	1.7	1.4
	6.6	- 6.3		3.8	4.7
	5.0	- 4.9		4	9.0
	4.0	3.4		$\bar{3}$	- 0.4
	8.9	9.7		2	11.1
	9.5	9.7		$\bar{1}$	- 3.7
	3.6	4.1		0	2.9
	1.7	1.5		1	1.3
1 $\bar{2}$ 4	1.2	- 0.8	1 4 $\bar{5}$	8.8	9.6
	5.9	- 6.5		4	1.1
	8.6	- 8.9		$\bar{3}$	3.1
	10.9	- 9.5		2	4.8
	22.0	-20.6		$\bar{1}$	- 1.8
	4.8	5.0		0	1.7
	8.5	7.9	1 5 $\bar{6}$	2.4	2.3
	5.9	- 5.5		$\bar{5}$	0.9
	7.1	6.8		4	3.5
1 $\bar{1}$ $\bar{5}$	2.0	2.1		$\bar{3}$	3.1
	0.9	- 0.6		$\bar{1}$	0.7
	4.2	3.3	1 6 $\bar{6}$	0.8	0.9
	2.5	- 2.5		4	1.0
	22.4	-25.8	1 7 $\bar{7}$	0.5	0.6
	22.5	26.8	2 $\bar{7}$ 1	1.1	- 1.1
	4.1	- 3.5	2 $\bar{6}$ 0	2.6	- 2.5
	10.0	9.7		1	- 2.2
	1.8	- 1.9		2	- 2.5
	4.5	- 4.7		3	- 2.9
1 0 $\bar{8}$	0.5	- 0.8		4	- 0.9
	0.9	1.0		6	- 1.6
	0.8	- 0.1	2 $\bar{5}$ $\bar{1}$	1.8	- 1.7
	8.5	8.8		0	- 2.0
	1.2	- 0.5		1	- 3.4
	1.9	- 1.5		2	- 4.2
	27.4	26.8		3	- 1.8
	2.7	- 1.2		4	- 2.9
	22.7	24.7		5	- 3.9
	27.2	24.8		7	- 1.7
	4.8	4.2	2 4 $\bar{1}$	1.3	- 1.4
	3.1	- 2.7		0	- 2.2
	4.5	4.5		1	- 3.1
	2.7	- 2.7		2	- 1.4
	1.0	0.5		3	1.5
	4.7	5.0		4	- 1.5
	1.9	- 2.0		5	0.5
	1.7	1.3		6	- 3.0
	3.4	- 2.0	2 $\bar{3}$ $\bar{3}$	2.1	- 1.9
	5.4	5.9		2	- 3.1
	15.3	14.1		$\bar{1}$	- 4.2
	22.4	29.7		0	10.0
	5.5	9.5		1	- 1.3
	8.4	8.0		2	3.7
1 2 $\bar{6}$	2.1	2.3		3	2.3
	2.8	2.5		4	6.1
	3.3	- 3.6		5	1.7
	2.2	- 1.7		6	- 0.1
	12.5	11.3	2 $\bar{2}$ 4	2.2	- 2.0
	4.9	5.0		$\bar{2}$	- 4.9

$h k l$	$ F_o $	F_c	$h k l$	$ F_o $	F_c
2 $\bar{2}$ $\bar{1}$	19.6	-22.0	2 5 $\bar{3}$	2.2	1.6
0	12.0	-11.8	2 $\bar{2}$	2.7	2.8
1	12.4	-13.3	2 6 $\bar{6}$	0.7	-0.6
2	8.2	-7.3	3 $\bar{3}$	1.5	1.2
3	6.7	6.5	3 $\bar{7}$ 5	1.7	1.3
4	5.5	5.8	3 $\bar{6}$ 0	1.2	-1.2
5	1.7	-1.6	2	2.6	-2.4
2 $\bar{1}$ $\bar{5}$	1.6	-1.6	3 $\bar{6}$	0.9	0.9
4	3.7	3.4	3 $\bar{5}$ 1	3.7	-4.8
3	1.7	1.0	2	3.6	-3.6
2	6.7	-6.9	3	4.1	-4.3
1	5.9	-5.9	4	3.9	-3.5
0	20.8	-21.9	5	1.1	-0.6
1	20.5	-21.8	6	1.3	-1.4
4	3.4	-3.8	3 $\bar{4}$ $\bar{2}$	2.3	2.1
2 0 8	0.5	-0.3	1	2.3	2.3
7	1.1	-1.1	0	2.9	-1.1
6	1.6	-1.9	1	1.9	-1.2
5	1.7	1.9	2	4.0	-4.3
4	3.3	3.2	3	4.3	-4.4
3	6.5	6.0	4	2.9	-2.4
2	7.8	8.0	5	1.6	-1.8
1	1.1	-1.1	6	1.2	-1.1
0	12.3	-11.5	3 $\bar{3}$ $\bar{4}$	1.4	-1.7
1	6.4	5.3	3	1.4	0.6
2	5.5	-4.7	1	1.0	0.7
3	1.2	0.4	1	1.4	-1.5
4	5.0	5.4	2	4.0	-3.8
5	1.5	-1.8	3	1.7	-1.2
7	0.6	0.5	4	1.8	1.6
2 1 7	1.5	-1.7	5	2.0	-2.1
6	1.8	-1.4	3 $\bar{2}$ $\bar{5}$	2.1	-2.0
4	1.6	2.2	4	2.8	2.2
3	8.5	8.0	3	2.2	-2.5
2	10.2	9.8	2	3.0	-2.1
1	2.7	-2.5	1	2.1	-1.6
0	8.8	8.3	0	2.5	-3.1
1	1.0	0.1	1	10.8	-10.4
2	5.1	5.5	2	2.8	-2.8
2 2 5	3.4	-2.9	3	0.7	0.1
4	4.6	3.6	4	3.4	-3.6
3	3.5	3.9	3 $\bar{1}$ $\bar{6}$	1.2	-1.2
2	3.7	-3.4	5	2.5	-2.4
1	5.6	5.7	4	2.4	-2.2
1	0.8	-1.2	3	3.4	-3.2
2 3 6	2.1	-2.4	2	2.0	-0.9
5	4.0	3.8	1	2.2	-2.3
3	1.0	0.9	0	13.6	-13.1
2	4.4	4.1	1	4.7	-4.5
1	1.7	-1.0	2	9.1	-9.4
0	6.1	-6.0	3	5.8	-5.6
2 4 6	1.9	2.0	3 0 6	1.3	-1.3
4	3.2	3.4	5	1.6	-1.9
3	4.7	5.3	4	2.8	-3.3
2	1.7	0.9	3	2.0	1.7
1	1.7	-1.1	2	3.4	3.3
2 5 5	0.8	1.2	1	1.2	-1.3
4	3.5	3.4	1	9.3	-8.4

$h k l$	$ F_o $	F_c	$h k l$	$ F_o $	F_c
3 0 2	6.6	- 5.0	4 $\bar{1}$ $\bar{4}$	3.3	- 3.8
3 0 3	3.2	- 2.3	4 0 $\bar{3}$	3.6	- 3.1
3 0 4	0.6	- 0.3	4 0 $\bar{2}$	3.8	- 4.4
3 0 5	0.9	- 0.5	4 0 $\bar{1}$	3.7	- 3.3
3 0 6	1.2	0.9	4 0 1	1.5	- 2.0
3 1 $\bar{6}$	1.7	- 1.8	4 0 2	2.4	- 2.3
3 1 $\bar{5}$	3.7	- 3.5	4 0 $\bar{5}$	1.2	- 0.9
3 1 $\bar{4}$	1.4	- 1.0	4 0 $\bar{4}$	1.9	- 1.8
3 1 $\bar{3}$	1.8	1.7	4 0 $\bar{3}$	3.4	- 3.8
3 1 $\bar{2}$	3.0	2.5	4 0 $\bar{2}$	3.2	- 3.7
3 1 $\bar{1}$	6.1	6.4	4 0 $\bar{1}$	2.2	- 2.1
3 1 0	3.4	3.2	4 0 1	3.4	- 2.6
3 2 $\bar{1}$	2.8	- 2.5	4 0 2	2.6	- 2.2
3 2 $\bar{6}$	2.6	- 2.7	4 0 3	1.6	- 0.6
3 2 $\bar{5}$	2.3	- 2.4	4 0 4	2.9	- 2.5
3 2 $\bar{4}$	2.5	- 2.7	4 1 $\bar{4}$	1.7	- 1.8
3 2 $\bar{3}$	1.4	- 1.5	4 1 $\bar{3}$	3.1	- 3.1
3 2 $\bar{2}$	3.9	3.9	4 1 $\bar{2}$	2.0	- 1.5
3 2 $\bar{1}$	5.3	5.0	4 1 $\bar{1}$	1.7	- 1.2
3 3 0	2.3	- 2.5	4 1 0	2.0	- 2.1
3 3 $\bar{5}$	2.5	- 2.4	4 2 $\bar{4}$	2.7	- 2.8
3 3 $\bar{4}$	2.8	- 2.7	4 2 $\bar{3}$	1.5	- 1.7
3 3 $\bar{3}$	1.0	0.8	4 2 $\bar{2}$	2.2	- 1.9
3 3 $\bar{2}$	0.9	1.0	4 2 $\bar{1}$	1.8	- 1.5
3 3 $\bar{1}$	1.5	- 1.6	4 3 $\bar{5}$	2.2	- 2.1
3 4 $\bar{4}$	0.9	0.8	4 3 $\bar{3}$	1.3	- 1.5
3 4 $\bar{3}$	2.8	1.1	4 4 $\bar{4}$	1.1	- 1.2
3 4 $\bar{2}$	0.7	0.7	4 4 $\bar{3}$	1.4	- 0.5
3 4 $\bar{1}$	1.4	1.4	4 4 $\bar{2}$	1.0	0.5
4 7 2	1.3	0.9	4 4 $\bar{1}$	1.2	- 0.8
4 5 1	1.8	0.4	4 4 4	0.8	- 0.7
4 5 2	1.7	- 2.0	5 4 $\bar{4}$	1.7	1.7
4 5 3	2.3	- 1.6	5 4 $\bar{3}$	0.8	- 0.7
4 5 4	1.2	- 1.8	5 4 $\bar{2}$	1.7	1.7
4 5 5	1.0	- 0.6	5 4 $\bar{1}$	1.7	- 1.5
4 4 $\bar{1}$	1.9	2.2	5 3 1	1.7	- 0.1
4 4 $\bar{2}$	1.1	0.8	5 3 $\bar{2}$	1.7	1.7
4 4 $\bar{3}$	1.9	- 2.0	5 3 $\bar{1}$	1.7	- 0.1
4 4 $\bar{4}$	3.1	- 3.1	5 2 $\bar{2}$	1.7	1.7
4 4 $\bar{5}$	2.1	- 1.9	5 2 $\bar{1}$	1.7	- 1.5
4 4 5	2.4	- 2.4	5 2 0	1.7	1.7
4 3 $\bar{3}$	1.6	- 1.1	5 1 1	1.8	1.5
4 3 0	4.4	4.7	5 1 2	0.9	0.6
4 3 1	1.4	0.5	5 1 $\bar{2}$	2.2	- 1.8
4 3 3	0.6	- 1.0	5 1 $\bar{1}$	2.5	- 2.7
4 3 4	3.3	- 3.4	5 1 1	0.9	- 1.0
4 3 5	1.6	- 0.9	5 0 $\bar{2}$	1.2	- 1.4
4 2 $\bar{4}$	3.2	- 3.4	5 0 $\bar{1}$	2.5	- 2.2
4 2 $\bar{3}$	2.2	- 2.1	5 0 $\bar{2}$	1.9	- 2.1
4 2 $\bar{2}$	1.0	0.9	5 0 $\bar{1}$	2.8	- 2.7
4 2 1	2.9	3.0	5 0 0	1.1	- 0.6
4 2 2	0.8	- 0.5	5 1 3	1.1	- 1.2
4 2 3	2.5	- 2.3	5 1 $\bar{3}$	1.3	- 1.5
			5 1 $\bar{2}$	2.6	- 2.4
			5 1 $\bar{1}$	1.2	- 0.6
			5 2 $\bar{3}$	1.3	- 1.4
			5 2 $\bar{2}$	0.8	- 0.7
			5 3 $\bar{3}$	1.3	0.7
			6 $\bar{6}$ 4	1.1	0.2
			6 5 3	1.1	- 0.8
			6 0 0		

Table 4. Interatomic distances (in Å) and angles (°).

O ₁ -C ₁	1.48	C ₁ -O ₁ -C ₂	103
O ₁ -C ₂	1.48	C ₂ -O ₁ -C ₄	114
O ₁ -C ₃	1.45	O ₁ -C ₁ -C ₅	100
O ₁ -C ₄	1.45	O ₁ -C ₃ -C ₆	112
C ₁ -C ₅	1.61	C ₁ -C ₅ -O ₄	100
C ₃ -C ₆	1.36	C ₃ -C ₆ -O ₄	116
C ₁ -C ₃	0.75	O ₄ -O ₁N ₂	177
C ₂ -C ₄	0.80	O ₄ -O ₁ -N ₁	147
O ₁ -O ₄	2.75	O ₁N ₂ -O ₅	96
O ₁N ₂	2.76	O ₁N ₂ -O ₆	96
O ₁ -N ₁	2.91	O ₁N ₂ -N ₁	76
N ₁ -N ₂	1.75	O ₂ -N ₁ -O ₃	135
N ₁ -O ₂	1.18	O ₂ -N ₁ -N ₂	112
N ₁ -O ₃	1.18	O ₃ -N ₁ -N ₂	112

The fact that the position of the dioxan oxygen atoms are well defined even at room temperature indicates that the "movements" may possibly be described as a "rotation" of the dioxan molecule about an axis drawn between the two oxygens, respectively of an "inversion" of the cyclohexane ring. Such movements would not, of course, alter the position of the centre of symmetry of the dioxan ring.

Complete three-dimensional intensity material was employed when carrying out the final refinement of the structure. The model of the dioxan molecule found in the electron diffraction analysis⁴ was used and two "resting" positions assumed which combined would appear to explain the picture of the nearly planar six-membered ring resulting from the two-dimensional analysis. The symmetry of the space group does not impose any restrictions on the weight factors for the C-atom distribution and we started with the assumptions that the two positions have equal probabilities. Introducing anisotropic damping the *R*-factor decreased during the refinement procedure from 0.27 to 0.11. Block diagonal least squares program written for the Mercury computer by J. S. Rollett was used. The atomic coordinates thus arrived at are listed in Table 1, the vibrational parameters in Table 2. Table 3 contains observed and calculated structure factors. A list of interatomic distances and angles is given in Table 4. Finally a schematical drawing is reproduced in Fig. 3. Attempts to improve the agreement between observed and calculated structure factors by assuming the ratio of the weights of the two resting positions to deviate from 1:1 were not successful.

A three-dimensional Fourier synthesis based on the structure factor signs leading to the value *R* = 0.11 was finally computed. From this synthesis no further conclusions regarding the weights of the two resting positions could be drawn, however.

The present investigation, besides confirming the results obtained from X-ray and electron diffraction investigations regarding the structure of the dinitrogen tetroxide molecule, has thus even confirmed the presence of bonds between nitrogen atoms and dioxan oxygen atoms in the addition compound. The bond distance is, however, not very much shorter than the intermolecular

separation expected for a van der Waals interaction. The distance between the dioxan oxygen atom and the second nitrogen atom, on the other hand, closely corresponds to that of a van der Waals type of interaction.

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