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Structural Studies of Hexa- and Deca-methonium Bromides, $(CH_3)_3N(CH_2)_nN(CH_3)_3 . 2Br . 2H_2O$, in Relation to their Pharmacological Action

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The crystal structures of hexamethonium and decamethonium bromides, both of which are dihydrates, $(CH_3)_3N(CH_2)_nN(CH_3)_3 . 2Br . 2H_2O$, have been determined with the use of three-dimensional data. $n=6$ has $a=7\cdot41$, $b=18\cdot40$, $c=7\cdot20$ Å, $\beta=109°$; ϱcalc. 1·415 g.cm^{-3} for $Z=2$; ϱobs. 1·406(20 °C): $P2_1/c$. $n=10$ has $a=13\cdot49$, $b=10\cdot07$, $c=9\cdot04$ Å, $\beta=109°$; ϱcalc. 1·299 g.cm^{-3} for $Z=2$; ϱobs. 1·305 (20 °C): $P2_1/a$. Both molecules are centrosymmetrical, the chains being fully extended: $n=6$ N $\cdots$ N observed 8·80 Å, calculated 8·78 Å; $n=10$ N $\cdots$ N observed 14·01 Å, calculated 13·79 Å. There is otherwise no similarity between the two structures; that of $n=10$ is of a layer type.

A study of the interatomic distances (excluding those within the chains) and a comparison with those of pentamethonium iodide, indicates that it is not the nitrogen atoms that control the general bonding of the chains with the Br and H_2O, but that there are quite short van der Waals distances connecting all CH_2 and CH_3 groups, both to similar groups in other chains and to Br and H_2O respectively (or to I for the $n=5$ compound). The distances from N to any other atom (except those to which it is directly bonded) are all much too long to have any ionic character.

It is suggested that the activities of ganglionic and neuromuscular blockage, which are so specific with respect to the value of n, are dependent not on the N $\cdots$ N distance (except indirectly); but on the extreme $(CH_3) \cdots (CH_3)$ distances on each side of the chain and on the number of possible van der Waals contacts within those extremes. A quantitative comparison for $n=4$ to 12 supports this suggestion and supplies some geometrical reasons both for the specificity and for the dependence of the duration of activity upon temperature.

Introduction

The object of this study of the structure of certain methonium halides was to provide evidence that might help in the understanding of their curare-like pharmacological action. A review of the properties of the methonium compounds, with numerous references, has been given by Paton & Zaimis (1952); and these authors have published curves for the activities of the methonium salts in respect of ganglionic and neuromuscular blockage which lead to the following approximate relative values (n = number of methylene groups).

Type of block n	4	5	6	7	8	9	10	11	12
Nerve—nerve	0.007	0.80	(1)	0.11	0.022				
Nerve—muscle					0.07	0.67	(1)	0.55	0.22

The vital factor in the mode of action seems to be the chain length; and the extreme sensitivity to, and specificity in respect of n implies some rather specific conditions of 'fit' between the extended molecule and its effect at the site of action. Zaimis, Cannard & Price (1958) have also shown that a lowering of muscle temperature by as little as 3–5 °C markedly lengthened the duration of the neuromuscular block and, to a lesser extent, its magnitude.

A comparison, by the powder method, was first made of the following compounds:

Methonium halides:
$(CH_3)_3N(CH_2)_nN(CH_3)_32Br$ ($n=3, 5, 7, 9$)
$(CH_3)_3N(CH_2)_nN(CH_3)_32Br . 2H_2O$ ($n=4, 6, 8$)
$(CH_3)_3N(CH_2)_{10}N(CH_3)_32Br(?\ 2H_2O)$
$(CH_3)_3N(CH_2)_nN(CH_3)_3 . 2I$ ($n=4, 5, 6, 7, 8, 9, 10$)
$(CH_3)_3N(CH_2)_6N(CH_3)_3 . 2Cl . 2H_2O$
Hexamethonium bitartrate
Hexamethonium embonate.

It was hoped that some evidence might be found of isomorphism or isostructuralism which would assist detailed crystal structure analysis of some members of the series. No clear evidence of any such similarity of structure was, however, found. It seemed probable, therefore, that each structure would have to be separately determined and an attempt was made to grow suitable crystals of the less hygroscopic members of the series. In the end, attention was focused on the pentamethonium iodide and the hexamethonium and decamethonium bromides.

The crystal structure of pentamethonium iodide has been published by Canepa (1962,1963), who argues against any theory of *two-point* attachment of the blocking agent, on the ground that in the solid state the N^+ is coordinated by *six* I^- and that attachment to a variable area of the membrane surface would seem to be a more reasonable hypothesis.

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Hexamethonium bromide

Small twinned crystals of $(CH_3)_3N(CH_2)_6N(CH_3)_3 . 2Br . 2H_2O$ were obtained by evaporation of an ethanol–methanol solution. It was not possible to find a single crystal. Intensity measurements were therefore made on unevenly twinned crystals, which permitted the sorting out of the data for the two individuals. (See Appendix I.) The twin plane was (100). Plates (*b* normal to plate face) and needles (axis *c*) of maximum dimension 0·2 mm, were both used for preliminary studies of all three axial zones by means of oscillation and Weissenberg methods, but for the main collection of three-dimensional intensity data equi-inclination Weissenberg photographs were taken (Cu $K\alpha$). The only systematic absences were $h0l$ for l odd, $0k0$ for k odd (up to $0,14,0$ were observed):

$P2_1/c$

$a=7{\cdot}41\,,\quad b=18{\cdot}40\,,\quad c=7{\cdot}20\ \text{Å};\quad \beta=109°$

M.W. 398·1 ϱ obs. (20 °C) 1·406 g.cm^{-3}

$Z=2$ for ϱ calc. 1·415 g.cm^{-3}.

F obs, F calc are given in Table 8. Fig. 1 shows the numbering of the atoms and their positions relative to the inertia axes $\vec{L}$, $\vec{M}$. Table 1 gives the atomic fractional coordinates at the termination of the refinement. At this stage the C–C distances along the chain varied

Table 1. *Hexamethonium dibromide dihydrate: fractional coordinates of all atoms except hydrogen (see Fig. 1), relative to the unit cell axes a, b, c*

	x	*y*	*z*
Br	0·3182	0·3677	0·1657
$C(H_2)(1)$	−0·0552	0·0363	−0·0042
$C(H_2)(2)$	−0·1167	0·0496	0·1691
$C(H_2)(3)$	−0·2293	0·1173	0·1464
N	−0·3500	0·1233	0·2808
$C(H_3)(2)$	−0·4232	0·2012	0·2705
$C(H_3)(2)$	−0·5105	0·0719	0·2348
$C(H_3)(3)$	−0·2285	0·1118	0·4919
$(H_2)O$	0·0545	0·2723	0·4010

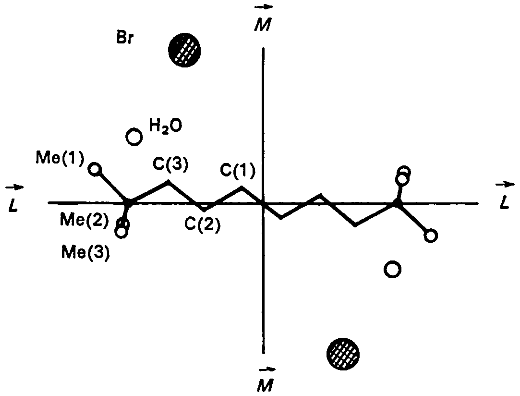

Fig. 1. Projection of the hexamethonium molecule along the $\vec{N}$ inertia axis of the molecule, showing the numbering.

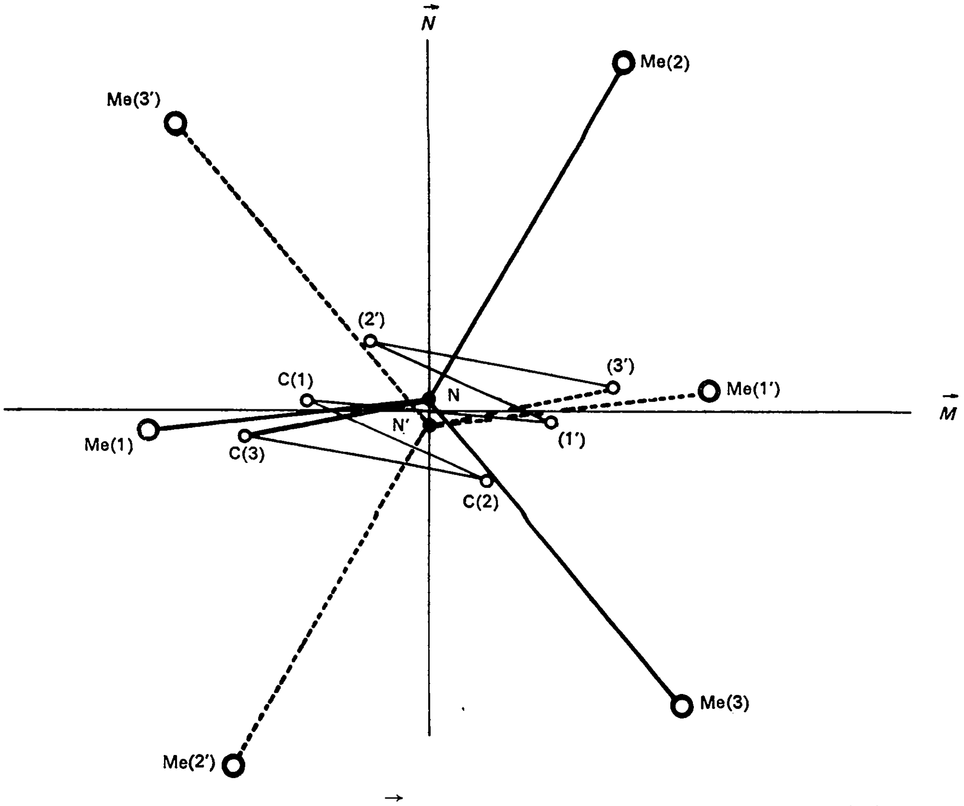

Fig. 2. Projection of hexamethonium chain along the $\vec{L}$ inertia axis. The lack of planarity of the chain is not regarded as significant, but the twist of the methyl groups relative to the mean plane is probably a real effect.

from 1·556 to 1·478 Å. No significance is attached to this variation because of the influence of the heavy atoms. The maximum chain length $(CH_3)(1) \cdots (CH_3)(1')$ is 11·18 Å and the $N \cdots N'$ distance is 8·80 Å. Calculated for a regular chain with tetrahedral bonds C–C 1·54 Å and C–N 1·50 Å, these lengths would be 11·22 Å and 8·78 Å. The average of the four N–C bonds is 1·50(3) Å. Fig. 2, which shows the projection of the chain along its $\vec{L}$ inertia axis, implies that there is a small twist of the $C.N(CH_3)_3$ group out of the average plane of the $(CH_2)_6$.

Figs. 3, 4, 5 show the projection of the structure along the three axial directions, and Fig. 6 shows a possible method of twinning. The precise shift of the structure at the twin plane cannot be ascertained by any known technique.

Table 2 gives the vibration ellipsoid for the 'standard' bromine atom and the individual $\bar{B}$ coefficients derived from the refinement for the other atoms. It is not surprising to find relatively large values for the methyl groups. The larger value of $\bar{B}$ for the water molecule may imply either a large thermal vibration or some relaxation of the water position.

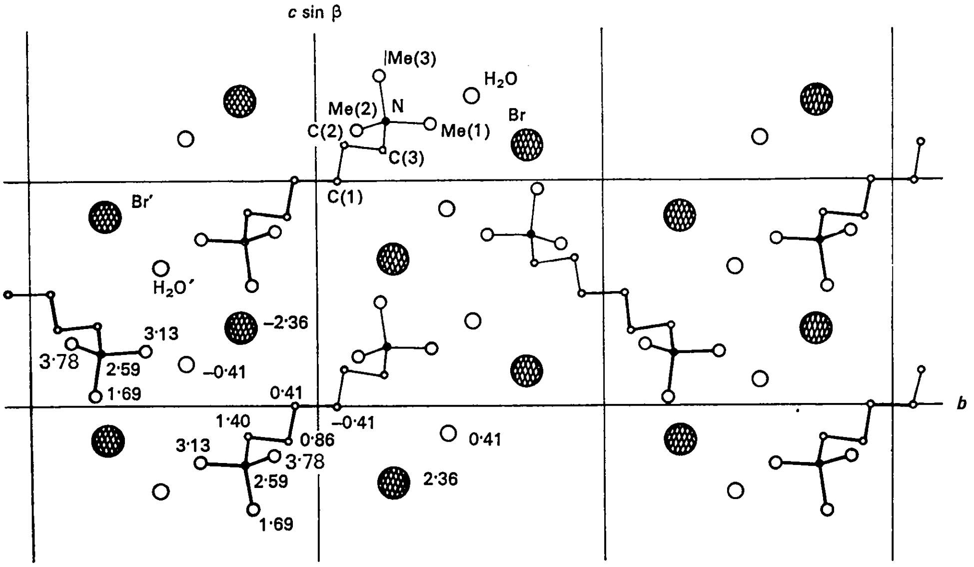

Fig. 3. [100] projection of hexamethonium dibromide dihydrate. The numbers show the distances of atoms above or below the projection.

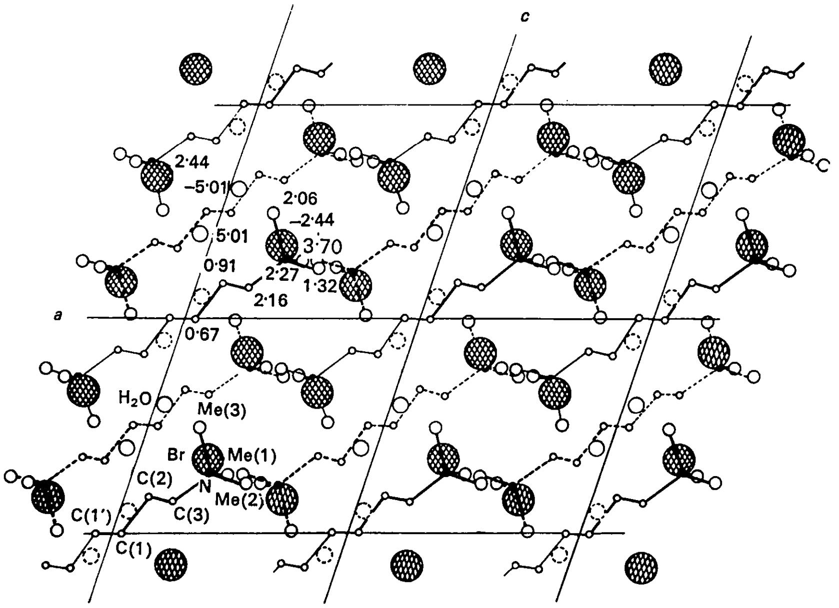

Fig. 4. [010] projection of hexamethonium dibromide dihydrate. The numbers show the distances above or below the projection. The atoms in the dotted molecule are at $x, \frac{1}{2}-y, \frac{1}{2}+z$ relative to the standard molecule.

Table 3 gives all distances between atoms (not in the same chain) that are less than 4 Å. It will be seen that there are a large number of these. In particular, the water molecule is surrounded by twelve neighbours at this relatively short distance. It is particularly significant that there are many van der Waals distances not only between Br, H_2O and chain CH_2 or terminal (CH_3) groups but between neighbouring chains also. The shortest distances from the nitrogen are those to a water molecule, 3·945 Å, and to two bromine atoms, 4·265

Table 2. *Hexamethonium dibromide dihydrate. Magnitudes and direction cosines of* $B_{\max}$, B_{med} *and* $B_{\min}$ *for the standard bromine atom, and individual* $\bar{B}$ *values for the other atoms*

	B	a	b	c^*
$B_{\max}$	2·93	0·1507	−0·1936	0·9694
B_{med}	2·14	0·7444	0·6675	0·0176
$B_{\min}$	1·38	−0·6704	0·7190	0·2447

$C(H_2)(1)$	$C(H_2)(2)$	$C(H_2)(3)$	N	$C(H_3)(1)$	$C(H_3)(2)$	$C(H_3)(3)$	$(H_2)O$
1·26	1·15	1·66	1·08	3·86	2·90	3·82	5·37

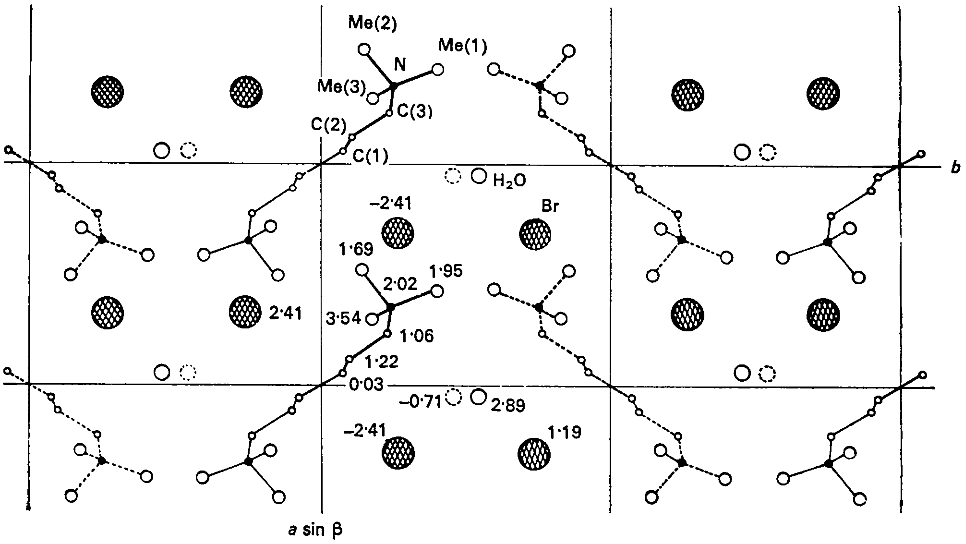

Fig. 5. [001] projection of hexamethonium dibromide dihydrate. The numbers show the distances of the atoms above or below the projection. The atoms in the dotted molecule are at x, $\frac{1}{2}-y$, $\frac{1}{2}+z$ relative to the standard molecule.

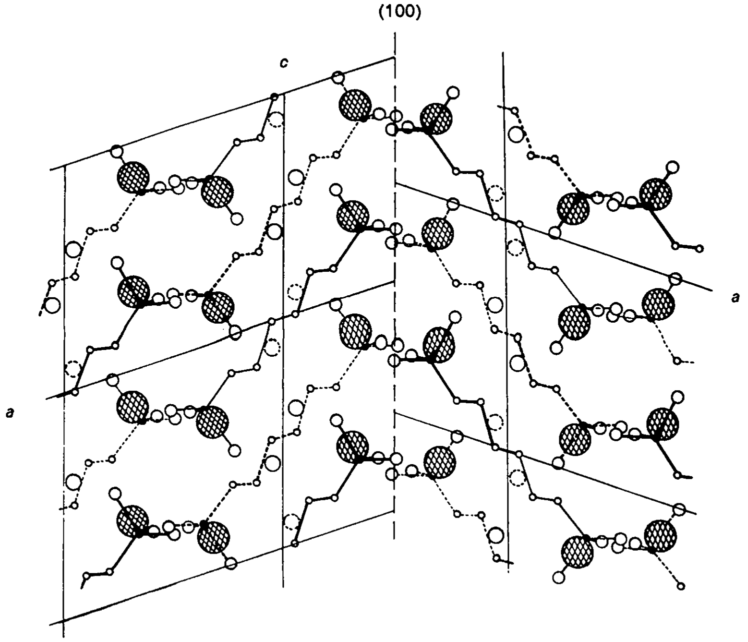

Fig. 6. Possible method of twinning of the hexamethonium compound.

Table 3. *Hexamethonium dibromide dihydrate*: *Distances between atoms that are less than* 4 Å, *excluding those within a single chain*

Molecules may be identified by the coordinates of their centres. A superscript indicates an atom belonging to the centrosymmetrical half of the molecule. C ≡ CH_2, Me ≡ CH_3.

000 Br	$-H_2O$	000	3·451 Å
	$-H_2O$	$0\frac{1}{2}\bar{\frac{1}{2}}$	3·414
	–C(3)	$0\frac{1}{2}\frac{1}{2}$	3·968
	–Me(1)	100	3·564
	–Me(2)	$1\frac{1}{2}\bar{\frac{1}{2}}$	3·881
	–Me(2′)	$0\frac{1}{2}\frac{1}{2}$	3·999
	–Me(3)	$0\frac{1}{2}\bar{\frac{1}{2}}$	3·848
	–Me(3)	$1\frac{1}{2}\frac{1}{2}$	3·969
000 N	$-H_2O$	000	3·945
000 C(l)	$-H_2O$	$0\frac{1}{2}\bar{\frac{1}{2}}$	3·728
	–Me(2)	100	3·900
	–Me(2′)	$\bar{1}00$	3·694
	–Me(3)	$00\bar{1}$	3·706
000 C(2)	–Me(2′)	$\bar{1}00$	3·988
000 C(3)	$-H_2O$	000	3·662
	$-H_2O$	$0\frac{1}{2}\bar{\frac{1}{2}}$	3·756
	–Br	$\bar{1}\frac{1}{2}\frac{1}{2}$	3·968

000 H_2O	–Br	000	3·451 Å
	–Br	$0\frac{1}{2}\frac{1}{2}$	3·414
	$-H_2O$	$0\frac{1}{2}\frac{1}{2}$	3·692
	$-H_2O$	$0\frac{1}{2}\bar{\frac{1}{2}}$	3·692
	–N	000	3·945
	–C(1)	$0\frac{1}{2}\frac{1}{2}$	3·728
	–C(3)	000	3·662
	–C(3)	$0\frac{1}{2}\frac{1}{2}$	3·756
	–Me(1)	000	3·598
	–Me(1)	$1\frac{1}{2}\frac{1}{2}$	3·951
	–Me(3)	000	3·800
	–Me(3)	$0\frac{1}{2}\bar{\frac{1}{2}}$	3·692
000 Me(1)	$-H_2O$	000	3·598
	–Br	$\bar{1}00$	3·564
	$-H_2O$	$\bar{1}\frac{1}{2}\bar{\frac{1}{2}}$	3·951
000 Me(2)	–Br	$\bar{1}\frac{1}{2}\frac{1}{2}$	3·881
	–Br′	$0\frac{1}{2}\frac{1}{2}$	3·999
	–C(l)	$\bar{1}00$	3·900
	–C(l′)	$\bar{1}00$	3·694
	–C(2′)	$\bar{1}00$	3·988
000 Me(3)	$-H_2O$	000	3·800
	$-H_2O$	$0\frac{1}{2}\frac{1}{2}$	3·689
	–Br	$0\frac{1}{2}\frac{1}{2}$	3·848
	–Br	$\bar{1}\frac{1}{2}\frac{1}{2}$	3·969
	–C(l)	001	3·706

Nearest distances (4–5 Å) from N to other atoms, not in the same chain

000 N	–Br	$0\frac{1}{2}\frac{1}{2}$	4·823 Å	000 N	$-H_2O$	$0\frac{1}{2}\frac{1}{2}$	4·887 Å
	–Br	$\bar{1}\frac{1}{2}\frac{1}{2}$	4·265		$-H_2O$	$\bar{1}\frac{1}{2}\bar{\frac{1}{2}}$	4·774
	–Br	$\bar{1}\frac{1}{2}\bar{\frac{1}{2}}$	4·314		–Me(1)	$0\frac{1}{2}\frac{1}{2}$	4·940
	–Br′	$0\frac{1}{2}\frac{1}{2}$	4·718		–Me(1)	$0\frac{1}{2}\bar{\frac{1}{2}}$	4·790

and 4·314 Å, but on the whole it seems clear that *direct* nitrogen 'attachments' are relatively unimportant in structure building. There seems no reason to suppose that they are any more effective in whatever action takes place at the nerve–nerve junction in the body. It does seem possible, however, that water may play some important role.

Decamethonium bromide

This compound was difficult to crystallize, but eventually one single crystal (approximate dimensions 0·05 mm linear) was obtained from an ethanol solution and all measurements were made on this specimen. Weissenberg photographs about [100] and [010] and a complete set of [001] oscillation photographs were used for a preliminary investigation but the main intensity data were obtained from equi-inclination Weissenberg photographs (with Cu $K\alpha$) about [010]. (See Appendix II.) Excluding unobserved reflexions and hydrogen atoms, 673 independent reflexions gave a residual $R=0{\cdot}10$, using anisotropic temperature factors only for bromine (Table 9).

The systematic absences were *h*0*l* for *h* odd, 0*k*0 for *k* odd:

$P2_1/a$.

$a=13{\cdot}49\,,\quad b=10{\cdot}07\,,\quad c=9{\cdot}04\ \text{Å};\quad \beta=109°$.

The material had been supplied as being without water of crystallization, and on this basis ϱ calc. = 1·196 g.cm^{-3} for $Z=2$. The density observed for powdered material was 1·305 g.cm^{-3} at 20°C. On this basis the formula must be

$$(CH_3)_3N(CH_2)_{10}N(CH_3)_3\,.\,2Br\,.\,2H_2O$$

M.W. 454·2; $Z=2$ gives ϱ calc. = 1·299 g.cm^{-3}.

This brings the decamethonium compound into line with the other even salts, all of which are hydrated. Fig. 7 shows the numbering of the atoms and their disposition relative to the inertia axes of the chain. Again there is a noticeable twist of the C . $N(CH_3)_3$ groups relative to the remainder of the chain (a similar small

Table 4. *Decamethonium dibromide dihydrate*: *fractional coordinates of all atoms except hydrogen* (*See* Fig. 4) *relative to the unit cell axes a, b, c*

	x	*y*	*z*
Br	0·5943	0·1459	0·7454
$C(H_2)(1)$	0·0323	0·0484	0·0697
$C(H_2)(2)$	0·0936	−0·0292	0·2061
$C(H_2)(3)$	0·1496	0·0605	0·3553
$C(H_2)(4)$	0·2021	−0·0210	0·5105
$C(H_2)(5)$	0·2584	0·0787	0·6384
N	0·2954	0·0170	0·7968
$C(H_3)(1)$	0·3495	0·1203	0·9179
$C(H_3)(2)$	0·2090	−0·0344	0·8534
$C(H_3)(3)$	0·3724	−0·0953	0·8115
$(H_2)O$	0·4219	0·1823	0·3729

twist occurs for the $n=5$ chain, Fig. 8) but any departure of the chain itself from regularity and 'linearity' is not regarded as significant. The observed overall $(CH_3)(1)$ - - - $(CH_3)(1')$ chain length is 16·38 Å and the N - - - N′ length is 14·01 Å as against 16·24 Å and 13·79 Å calculated for C–C as 1·54 Å and C–N as 1·50 Å, with tetrahedral angles.

Tables 4 and 5 give the fractional atomic coordinates and Debye coefficients, and Table 6 the distances between atoms (not in the same chain) that are less than 4 Å, together with N distances less than 5 Å.

The axial projections of the structure are given in Figs. 9, 10, 11, and it will be seen that these bear no apparent resemblances to those of the hexamethonium

Table 5. *Decamethonium dibromide dihydrate: magnitudes and direction cosines of* B_{max}, B_{med}, B_{min} *for the standard bromine atom, and mean B values for the other atoms.* C ≡ CH_2, Me ≡ CH_3

	B	*a*	*b*	*c**
B_{max}	4·04	0·7610	−0·6485	0·0201
B_{med}	3·29	0·1169	0·1675	0·9789
B_{min}	2·60	0·6382	0·7426	−0·2033

C(1)	C(2)	C(3)	C(4)	C(5)	N	Me(1)	Me(2)	Me(3)	H_2O
2·82	3·92	4·03	2·81	2·45	1·72	3·58	5·16	4·19	4·98

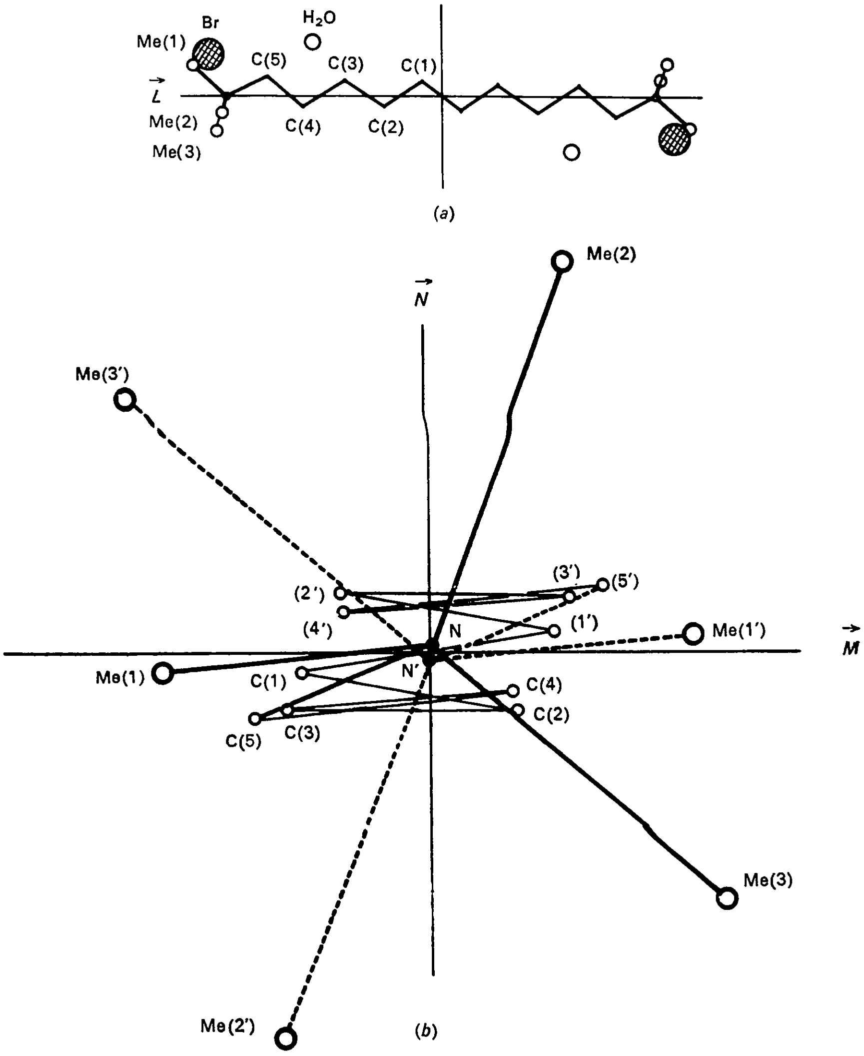

Fig. 7. (*a*) Projection of the decamethonium molecule along the $\vec{N}$ inertia axis of the chain, showing the numbering. (*b*) Projection of the decamethonium chain along the $\vec{L}$ inertia axis. The lack of planarity of the chain is not regarded as significant, but the twist of the methyl groups relative to the mean plane is probably a real effect.

compound in spite of the similarity of the monoclinic angle. The decamethonium structure is of a clear 'layer' type, unlike the hexamethonium structure, which is more of the 'herringbone' type.

Discussion of structure in relation to biological action

The number of intermolecular distances less than 4 Å in length (a limit which is an arbitrary choice, but one which would correspond to a very weak van der Waals bond) is less for the decamethonium layer-type structure than for the hexamethonium. All the atoms except nitrogen do, however, have some 'intermolecular bonds' within this range and, as in the previous case, the water molecule has the largest 'coordination' and the Br to H_2O distances are the shortest (3·433 and 3·458 Å in the deca-, and 3·414 and 3·451 Å in the hexa-compound). No nitrogen 'bond' outside the chain is less than 4·27 Å and only four are less than 4·5 Å. Again, therefore, there is no reason to suppose that the nitrogen plays any particular part except as the carrier of the methyl groups. There is, in fact, no crystallographic evidence of the use of any localized positive charge on the nitrogen atoms, which if used would imply strong coulombic forces in the structure. Such forces do not appear to exist.

On the contrary, the mode of attachment of the chain would appear to be by numerous van der Waals bonds along its length, both directly to other chains and to the bromine and water which surround them.

A re-examination of the data for pentamethonium iodide has shown that the same is true for that compound. There are a number of van der Waals distances (the shortest being $(CH_3)(1)$–$(CH_3)(1')$ and $(CH_2)(6)$–$(CH_2)(6')$, both 3·70 Å, and the shortest I–(CH_3) being 3·80 Å) which are all more structurally significant than the much longer N–I distances quoted by Canepa (1962, 1963).

It does seem highly probable, therefore, that the action of these compounds as blocking agents depends

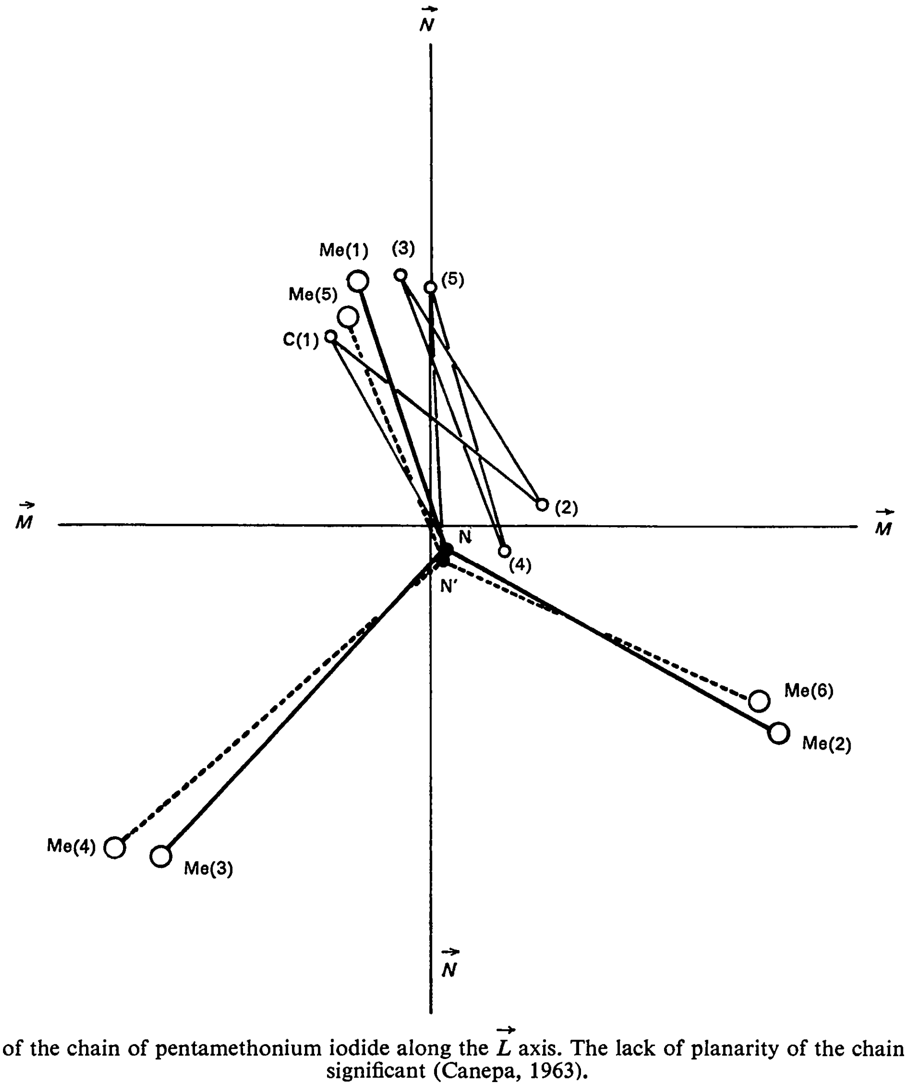

Fig. 8. Projection of the chain of pentamethonium iodide along the $\vec{L}$ axis. The lack of planarity of the chain is not regarded as significant (Canepa, 1963).

neither on 'simultaneous contact with two receptor groups' (Bergman, 1955; Paton & Zaimis, 1951) – the length of the most active compound being a measure of the inter-receptor distance – nor on the existence of two active 'areas' (Canepa, 1962), but rather on the possibility of adhesion along the whole available length of the chain, to an area which will slightly exceed its own projected area. The strength of adhesion, especially of the methyl groups, may of course be increased by transferred charge from the N^+, but only slightly, because the actual bonds are not particularly short, in terms of van der Waals distances.

Using this as a hypothesis, we may calculate the 'active length' of any chain (see Fig. 12) in terms of the limiting methyl–methyl distance on either side of the (ideal) chain and the number of chain atoms to which

Table 6. *Decamethonium dibromide dihydrate: distances between atoms that are less than* 4 Å, *excluding those within a single chain*

000 Br	$-H_2O$	000	3·433 Å
	$-H_2O'$	101	3·458
	–O(5)	$\frac{1}{2}\frac{1}{2}0$	3·864
000 C(1)	–Me(2)	$00\bar{1}$	3·639
	–Me(2′)	001	3·545
	–Me(3′)	$\frac{1}{2}\frac{1}{2}1$	3·845
000 C(2)	–Me(1′)	$\frac{1}{2}\frac{\bar{1}}{2}1$	3·855
	–Me(2)	$00\bar{1}$	3·977
	–Me(2′)	001	3·992
000 C(3)	$-H_2O$	000	3·826
	–Me(3′)	$\frac{1}{2}\frac{1}{2}1$	3·753
000 C(4)	$-H_2O'$	$\frac{1}{2}\frac{\bar{1}}{2}1$	3·739
000 C(5)	$-H_2O$	000	3·893
	–Br	$\frac{\bar{1}}{2}\frac{1}{2}0$	3·864
000 H_2O	–Br	000	3·433 Å
	–Br′	101	3·458
	–C(3)	000	3·826
	–C(4′)	$\frac{1}{2}\frac{1}{2}1$	3·739
	–C(5)	000	3·893
	–Me(1)	$00\bar{1}$	3·956
	–Me(2′)	$\frac{1}{2}\frac{1}{2}1$	3·618
	–Me(3)	101	3·776
000 Me(1)	$-H_2O$	001	3·956
	–C(2′)	$\frac{1}{2}\frac{1}{2}1$	3·855
000 Me(2)	$-H_2O'$	$\frac{1}{2}\frac{\bar{1}}{2}1$	3·618
	–C(1)	001	3·639
	–C(1′)	001	3·545
	–C(2)	001	3·977
	–C(2′)	001	3·992
000 Me(3)	$-H_2O$	101	3·776
	–C(1′)	$\frac{1}{2}\frac{\bar{1}}{2}1$	3·845
	–C(3′)	$\frac{1}{2}\frac{\bar{1}}{2}1$	3·753
Nearest distances (less than 5 Å) from N to other atoms, not in the same chain			
000 N	–Br	000	4·402
	–Br	$\frac{\bar{1}}{2}\frac{1}{2}0$	4·272
	$-H_2O$	000	4·962
	$-H_2O'$	$\frac{1}{2}\frac{1}{2}1$	4·402
	$-H_2O'$	101	4·973
	$-H_2O'$	$\frac{1}{2}\frac{\bar{1}}{2}1$	4·402
000 N	–C(1′)	001	4·990
	–C(2′)	$\frac{1}{2}\frac{1}{2}1$	4·811
	–C(3′)	$\frac{1}{2}\frac{\bar{1}}{2}1$	4·923

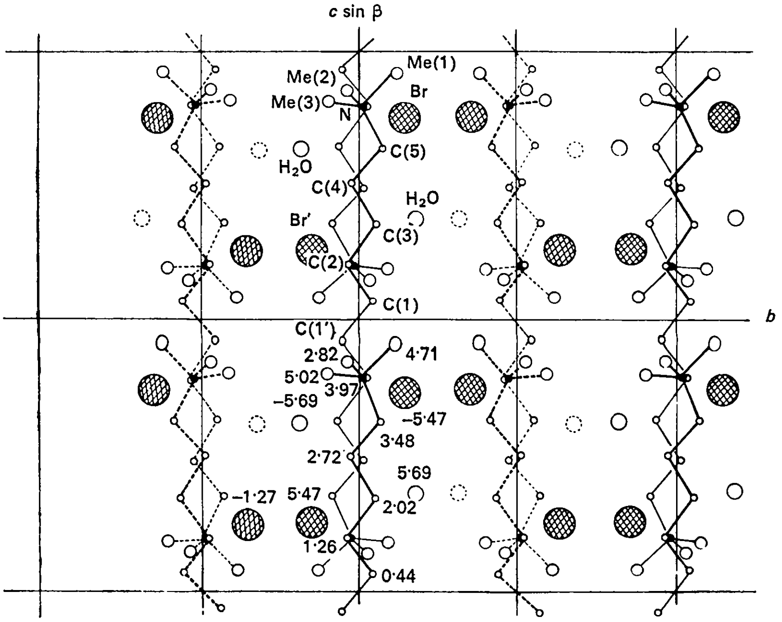

Fig. 9. [100] projection of decamethonium dibromide dihydrate. The numbers show the distances of atoms above or below the projection. The atoms in the dotted molecule are at $\frac{1}{2}+x$, $\frac{1}{2}-y$, z relative to the standard molecule.

attachments may be made. If n is even, there are 3 CH_3 and $\frac{1}{2}n$ CH_2 groups on each side, the extreme length on either side being

$$\{[(n-1)\ 1{\cdot}54+4{\cdot}50]^2 \sin^2\varphi+1{\cdot}50^2\cos^2\varphi\}^{\frac{1}{2}}\ \text{Å}$$

where $\varphi=54°44'$, C–C = 1·54 Å and N–C = 1·50 Å, and the small twist of the methyl groups is neglected. If n is odd, there are 2 CH_3 and $\frac{1}{2}(n+1)$ CH_2 groups on one side, the extreme length on that side being

$$[(n-1)\ 1{\cdot}54+6{\cdot}00]\sin\varphi\ \text{Å}$$

and there are 4 CH_3 and $\frac{1}{2}(n-1)$ CH_2 groups on the other side, the extreme length on that side being

$$[(n-1)\ 1{\cdot}54+3{\cdot}00]\sin\varphi\ \text{Å}\,.$$

We may then list the lengths and number of contacts as in Table 7.

According to Table 7, the maximum ganglionic blockage activity corresponds to 6 possible contacts over a length of 10 Å, two chances being available, one for each side of the 6-methonium chain. The 5-methonium chain with 5 contacts in a length of 9·9 Å on one side,

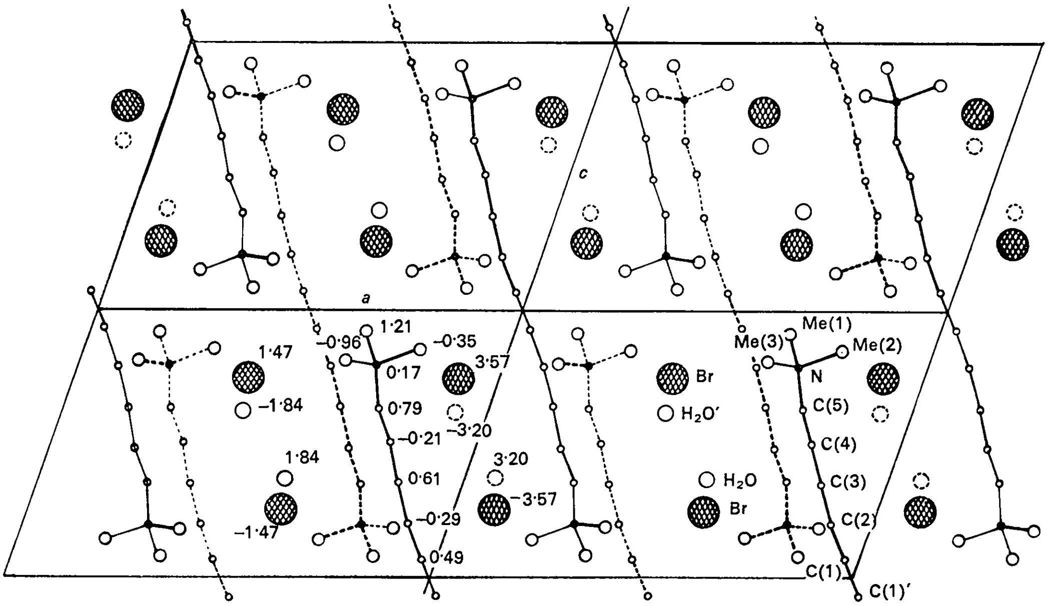

Fig. 10. [010] projection of decamethonium dibromide dihydrate. The small apparent irregularities of the chain are probably caused by the proximity of the heavy bromide atoms. The numbers show distances of atoms above or below the projection. The dotted molecule is reflected in the plane at $y=b/4$ to give atoms at $\frac{1}{2}+x,\ \frac{1}{2}-y,\ z$.

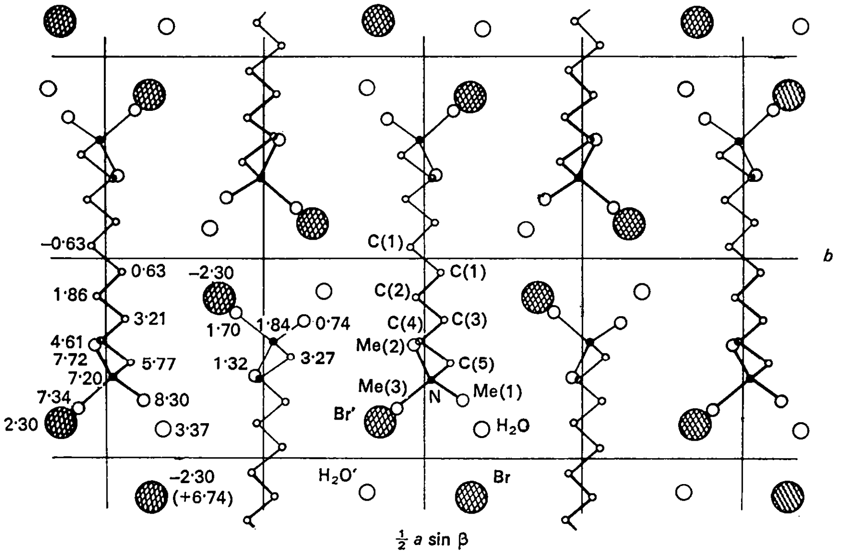

Fig. 11. [001] projection of decamethonium dibromide dihydrate. The numbers show the distances of atoms above or below the the projection.

and 6 contacts in a length of only 7·5 Å on the other is less but not much less efficient. Longer molecules would either have one very mobile free end, or would have to bend round and thus reduce the number of contacts drastically. Either way, they would be easily detached.

The maximum neuromuscular block corresponds to two chances of 8 contacts in a length of 15 Å. Seven contacts along 15 Å or 8 contacts in 12·5 Å give the reduced activity of 9-methonium, but the activity falls off rather less sharply on the high-*n* side because the relative change in length is less for a longer chain. It could be, of course, that some form of incipient one-layer crystallization might occur, on to a suitable substrate. The dissimilarity of the 5-methonium and 6-methonium arrangements, apart from their chain lengths, does not, however, lend any weight to this suggestion, unless the actual mode of crystallization is unimportant so long as coverage is provided.

The increased efficiency of action on cooling is easily understood in terms of numerous van der Waals attachments, since these may well be sensitive to thermal vibration, especially of the more mobile methyl groups. A rise of temperature will be accompanied by a greater tendency for the chains to break away. If the bonds were ionic, temperature would not be expected to affect them much.

Some of the early powder photographs were taken by Mr T. J. P. Richards, and eye estimations of intensity on the Weissenberg photographs of the hexamethonium bromide twinned crystal were made by Mrs Barbara Varwell. We are indebted to Messrs May & Baker, Messrs Allen & Hanbury, Professor W. D. M. Paton F.R.S. and Mr T. D. Whittet for our material. The Medical Research Council and the Wellcome Trust provided grants for the investigation. We are grateful also to various of our colleagues for their interest and for discussions which helped to clarify the problem.

Table 7. *The number of contacts and extreme lengths on each side of the n-methonium chains, in relation to their approximate blocking activity*

n	N_1	L_1	N_2	L_2	Approximate activity of blockage	
	CH_3 CH_2	Å	CH_3 CH_2	Å	Nerve–nerve	Nerve–muscle
4	3+2	7·50	3+2	7·50	0·007	
5	2+3	9·93	4+2	7·48	0·80	
6	3+3	10·00	3+3	10·00	(1)	
7	2+4	12·45	4+3	10·00	0·11	
8	3+4	12·50	3+4	12.50	0·022	0·07
9	2+5	14·96	4+4	12·51		0·67
10	3+5	15·02	3+5	15·02		(1)
11	2+6	17·48	4+5	15·03		0·55
12	3+6	17·53	3+6	17·53		0·22

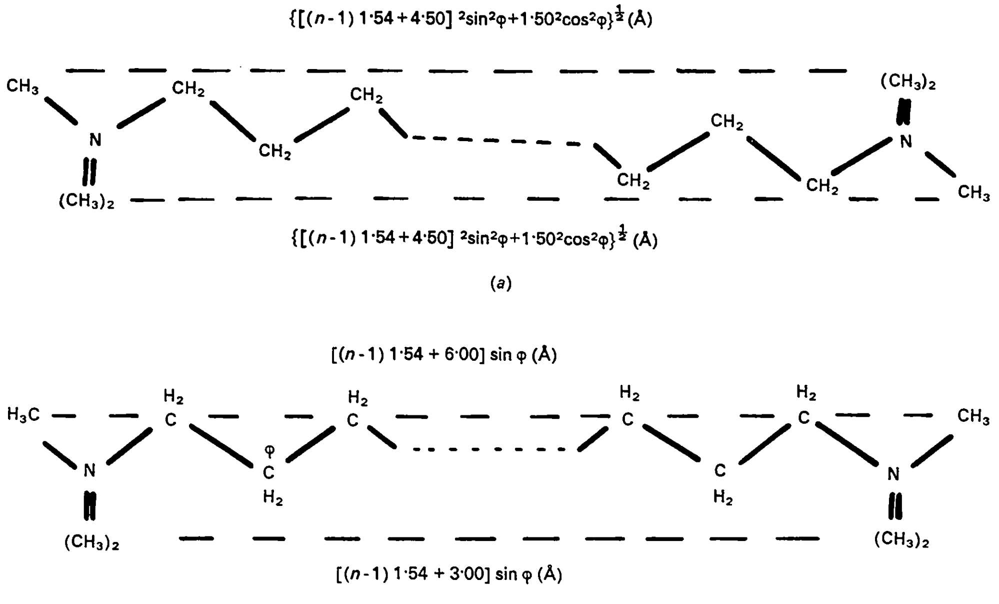

Fig. 12. (*a*) Even chain: dependence upon *n* of extreme length on each side ($\varphi = 54°44'$). (*b*) Odd chain: dependence upon *n* of extreme length on each side ($\varphi = 54°44'$).

Finally, we acknowledge with thanks the help we have had from Ferranti Ltd and International Computers and Tabulators for the gift and servicing of the computer PEGASUS, without which this work would not have been completed.

APPENDIX I
Hexamethonium Bromide

The only crystals of hexamethonium bromide which could be obtained were always twinned, and usually consisted of two individuals of approximately equal size. Inspection of any section of the reciprocal lattice of a twinned crystal normal to [010] (Fig. 13) shows that failure to disentangle the superimposed reflexions would result in the loss of nearly one-third of the 3-D data. However, one crystal consisting of two individuals of *different* volumes (subsequently found to be about 1:5) was obtained and the separate intensities could then be derived as follows:

Let I be the intensity of reflexion from the main crystal (larger individual, M) and $f.I$ be the corresponding intensity from the smaller twin (T) where f is to be determined.

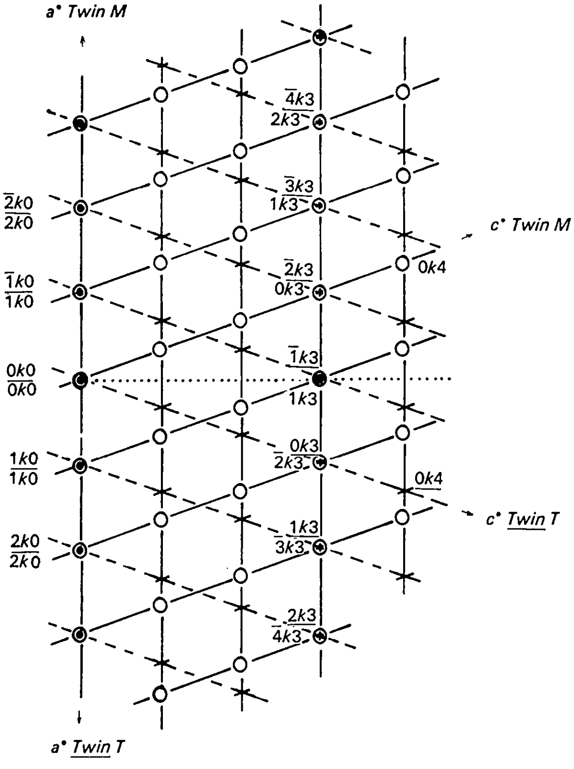

Fig. 13. Reciprocal lattice layer normal to [010], for twinned crystal of hexamethonium bromide, showing coincidences of reflexions from the two individuals, for which [100] of one coincides with [302] of the other. This results in a superposition of reflexions from the two individuals (*a*) for all *hk*0, identical indices. (*b*) for all *hk*3*n*, identical indices if $h=-n$ (*e.g.* $\bar{1}23$; or $\bar{2}16$), non-identical indices if $h\neq -n$ (*e.g.* $\bar{3}13$ and 113; or $\bar{2}23$ and 023). Note that since the space group is $P2_1/c$ all reflexions $h0l$ are absent for l odd, thus the $h0l$ section shows coincidences only for $h00$, $h06n$. Encircled black dots: equivalent hkl coincide. Encircled asterisks: non-equivalent hkl coincide.

If reflexions $(h_1k_1l_1)_M$ and $(h_2k_2l_2)_T(\equiv I_1)$ are superimposed, it follows that $(h_2k_2l_2)_M$ and $(h_1k_1l_1)_T(\equiv I_2)$ are also superimposed. I_1 and I_2 are measured, then

$$Ih_1k_1l_1+f\,.\,Ih_2k_2l_2=I_1$$
$$f\,.\,Ih_1k_1l_1+Ih_2k_2l_2=I_2$$
$$(1-f^2)Ih_1k_1l_1=I_1-f\,.\,I_2$$

and similarly

$$(1-f^2)Ih_2k_2l_2=I_2-f\,.\,I_1$$

For the crystal used, f was found to be about 0·2 from a comparison of main and twin $h0l$ intensities.

The 3-D Cu K Weissenberg data ($0kl-5kl$) had, however, been collected about the a axis, and thus the pairs of reflexions which had to be considered together did not occur on the same photographs. The observed data were therefore corrected in the usual way, using the function $W/2\text{Lp}$ tabulated in *International Tables for X-ray Crystallography* (1962) and the resulting F values were scaled to the $hk0$ and $hk1$ layers by a least-squares process (Walley: to be published). The appropriate pairs of F^2 values were then corrected by the expressions

$$0{\cdot}96\ F^2{}_{h_1k_1l_1}=F_1^2-0{\cdot}2\ F_2^2$$
$$0{\cdot}96\ F^2{}_{h_2k_2l_2}=F_2^2-0{\cdot}2\ F_1^2$$

The resulting pairs of F-values are linked in Table 8. Furthermore, the $hk0$ and $\bar{1}k3$ intensities must be reduced to $1/(1+f)=5/6$ of their observed values, since reflexions of identical indices for both main and twin crystals are superposed. These values are marked with an asterisk in Table 8. The available photographs enabled a complete set of 3-D data to be obtained for h, $l\leq 5$, *i.e.* within a sphere of radius $\sin\theta=0{\cdot}625$. Beyond this value the data were limited in the **a** direction because no further photographs had been obtained; and in the **c** direction by the fact that data for $l=6$ had again to be obtained by sorting out superimposed reflexions, which was not expected to yield very good results. However, 502 reflexions are available within $\sin\theta=0{\cdot}625$, which allows adequate overdetermination in this structure. Data beyond $\sin\theta=0{\cdot}625$ were therefore not used, even though some were available.

The point-Fourier facility in the SFLS program (Milledge & Milledge, 1961), which computes the electron density for groups of up to 80 reflexions at a time, was used to obtain a 3-D Fourier synthesis from the 80 strongest reflexions, using the signs for the bromine atom given by the 2-D Patterson syntheses. This was quite adequate to define the chain; and of two possible peaks for the water molecule one was easily selected.

Least-squares refinement was first carried out on the coordinates and isotropic temperature factors for all atoms except hydrogen, and then including anisotropic temperature factors for bromine. F values for the 15 hydrogen atoms associated with the chain were calc-

Table 8. $|F|_{obs}$ *and* F_{calc} *for hexamethonium bromide dihydrate*

The print-out gives F_{obs} the same sign as F_{calc}

† = REFLEXIONS OMITTED FROM L.S. REFINEMENT

H	K	L	F(OBS)	F(CALC)
0	0	0	-	101.00
1	0	0 •	-10.81	-11.46
2	0	0 •	-13.56	-12.73
3	0	0 •	22.23	22.92
4	0	0 •	-2.14	-2.15
5	0	0 •	-16.21	-14.72
-6	0	2	-15.60	-15.79
-5	0	2	-2.44	-2.70
-4	0	2	22.65	21.72
-3	0	2	-18.14	-15.94
-2	0	2	-23.15	-22.59
-1	0	2 †	34.07	38.88
0	0	2	-19.99	-19.35
1	0	2	3.56	3.68
2	0	2	30.30	26.79
3	0	2	-2.04	-2.36
4	0	2	-7.24	-8.44
-5	0	4	15.09	14.44
-4	0	4	-15.30	-15.42
-3	0	4	-6.42	-5.75
-2	0	4	25.80	26.36
-1	0	4	-15.80	-15.70
0	0	4	-4.08	-5.02
1	0	4	11.73	12.54
2	0	4	-8.15	-5.88
1	1	0 •	-9.58	-7.64
2	1	0 •	11.43	10.06
3	1	0 •	1.43	2.34
4	1	0 •	-18.36	-17.61
5	1	0 •	5.61	5.04
-5	1	1	10.81	10.73
-4	1	1	-12.23	-12.86
-3	1	1	-4.89	-4.78
-2	1	1	20.10	24.13
-1	1	1	-16.31	-15.61
0	1	1	-4.68	-3.97
1	1	1 †	24.68	35.92
2	1	1	-2.04	-0.21
3	1	1	-9.58	-8.41
4	1	1	12.34	11.95
5	1	1	-2.05	-2.91
-5	1	2	10.71	11.62
-4	1	2	-6.73	-5.96
-3	1	2	-1.41	-2.81
-2	1	2	19.89	19.82
-1	1	2	4.80	3.59
0	1	2	-15.19	-14.44
1	1	2	15.09	14.41
2	1	2	12.54	11.07
3	1	2	-13.66	-13.67
4	1	2	6.93	5.64
-5	1	3	-10.40	-11.10
-4	1	3	-2.74	-2.07
-3	1	3	22.84	23.89
-2	1	3	-5.30	-7.80
-1	1	3 •	-5.81	-6.55
0	1	3	12.64	12.16
1	1	3	-4.68	-7.73
2	1	3	-1.83	-2.27
3	1	3	14.69	12.58
-5	1	4	-6.22	-6.66
-4	1	4	-6.93	-7.87
-3	1	4	8.78	7.74
-2	1	4	-2.04	-0.13
-1	1	4	-12.13	-10.83
0	1	4	12.55	12.48
1	1	4	6.63	6.56
2	1	4	-11.42	-10.86
-4	1	5	10.41	9.46
-3	1	5	-10.19	-8.61
-2	1	5	2.05	2.44
-1	1	5	14.58	13.91
0	1	5	-5.09	-4.31
1	1	5	-3.15	-4.15
0	2	0 •	1.93	3.07
1	2	0 •	4.29	4.42
2	2	0 •	2.05	0.31
3	2	0 •	-6.83	-5.87
4	2	0 •	2.05	0.13
5	2	0 •	-2.04	-1.15
-5	2	1	-7.94	-7.45
-4	2	1	-12.75	-12.44
-3	2	1	22.84	24.24
-2	2	1	-9.58	-6.93
-1	2	1	-14.89	-13.69
0	2	1	7.34	6.51
1	2	1	9.79	9.26
2	2	1	-21.72	-22.54
3	2	1	18.15	16.50
4	2	1	8.46	7.98
5	2	1	-13.36	-13.64
-5	2	2	4.80	4.09
-4	2	2	2.05	0.63
-3	2	2	7.25	6.52
-2	2	2	2.65	2.00
-1	2	2	-3.15	-3.02
0	2	2	3.46	2.79
1	2	2	1.93	2.75
2	2	2	-3.56	-3.50
3	2	2	2.05	1.49
4	2	2	-2.04	-0.57
-5	2	3	-8.45	-8.95
-4	2	3	13.16	14.06
-3	2	3	-8.77	-8.40
-2	2	3	-7.34	-8.24
-1	2	3 •	19.48	21.73
0	2	3	8.37	7.00
1	2	3	-21.52	-25.72
2	2	3	12.55	11.62
3	2	3	9.39	8.87
-4	2	4	6.02	5.05
-3	2	4	5.91	4.78
-2	2	4	5.91	4.33
-1	2	4	7.95	6.78
0	2	4	4.59	4.12
1	2	4	-2.75	-0.80
-4	2	5	-5.30	-5.69
-3	2	5	-15.08	-13.91
-2	2	5	12.24	9.57
-1	2	5	5.62	4.42
0	2	5	-12.54	-12.71
1	2	5	9.90	10.73
1	3	0 •	-2.04	-0.23
2	3	0 •	19.59	22.53
3	3	0 •	5.31	4.61
4	3	0 •	-6.83	-6.35
5	3	0 •	6.53	6.32
-5	3	1	-11.82	-12.33
-4	3	1	12.85	11.91
-3	3	1	5.72	5.96
-2	3	1	-23.96	-27.20
-1	3	1	19.88	22.06
0	3	1	18.25	18.21
1	3	1 †	-19.47	-26.43
2	3	1	12.24	11.94
3	3	1	12.54	11.56
4	3	1	-13.76	-12.99
-5	3	2	6.84	6.24
-4	3	2	-5.20	-4.10
-3	3	2	-9.78	-9.82
-2	3	2	12.54	11.12
-1	3	2	-10.59	-9.18
0	3	2	-26.11	-24.32
1	3	2	9.07	8.31
2	3	2	3.78	2.73
3	3	2	-11.01	-8.70
4	3	2	3.68	3.81
-5	3	3	13.16	14.31
-4	3	3	8.37	7.28
-3	3	3	-17.84	-17.51
-2	3	3	16.62	16.50
-1	3	3 •	9.59	10.24
0	3	3	-15.60	-16.20
1	3	3	9.59	10.97
2	3	3	6.73	5.45
3	3	3	-9.79	-10.37
-4	3	4	-7.03	-7.54
-3	3	4	8.56	8.12
-2	3	4	-2.04	-0.66
-1	3	4	-9.89	-9.92
0	3	4	11.01	9.98
1	3	4	2.75	1.45
2	3	4	-9.27	-9.53
-4	3	5	-11.00	-11.03
-3	3	5	13.26	12.64
-2	3	5	2.65	4.45
-1	3	5	-11.51	-10.11
0	3	5	10.41	10.28
1	3	5	3.16	6.18
0	4	0 •	-30.39	-34.72
1	4	0 •	18.97	21.71
2	4	0 •	24.78	30.68
3	4	0 •	-20.09	-20.81
4	4	0 •	2.05	1.53
5	4	0 •	11.83	11.09
-5	4	1	2.76	3.27
-4	4	1	-1.82	-1.93
-3	4	1	-8.46	-6.87
-2	4	1	-7.64	-5.79
-1	4	1	-3.05	-1.73
0	4	1	-12.03	-11.13
1	4	1	-2.04	-1.94
2	4	1	7.03	5.46
3	4	1	6.22	3.94
4	4	1	2.05	0.49
5	4	1	3.46	6.58
-5	4	2	2.05	0.98
-4	4	2	-20.69	-23.63
-3	4	2	15.20	15.44
-2	4	2	11.73	10.72
-1	4	2	-24.68	-24.95
0	4	2	14.69	15.21
1	4	2	13.26	13.96
2	4	2	-17.43	-17.35
3	4	2	11.94	10.89
4	4	2	12.03	12.68
-5	4	3	-2.04	-2.19
-4	4	3	-4.08	-3.65
-3	4	3	-2.14	-0.67
-2	4	3	5.50	4.57
-1	4	3 •	-2.85	-2.84
0	4	3	-1.52	-0.93
1	4	3	-2.04	-1.32
2	4	3	-3.46	-3.07
3	4	3	-2.04	-2.45
-5	4	4	-15.39	-15.35
-4	4	4	15.40	15.01
-3	4	4	5.50	4.17
-2	4	4	-20.49	-20.14
-1	4	4	12.14	10.68
0	4	4	6.12	6.55
1	4	4	-12.44	-14.76
2	4	4	7.24	5.78
-4	4	5	-2.04	-1.31
-3	4	5	-2.04	-0.07
-2	4	5	-2.65	-3.94
-1	4	5	2.05	1.41
0	4	5	3.47	3.09
1	4	5	2.05	0.02
1	5	0 •	16.32	17.76
2	5	0 •	-12.64	-10.25
3	5	0 •	-3.15	-2.71
4	5	0 •	16.52	16.28
5	5	0 •	-8.15	-7.24
-5	5	1	-9.99	-10.48
-4	5	1	11.94	9.92
-3	5	1	-2.04	-0.13
-2	5	1	-13.66	-13.24
-1	5	1	16.32	15.16
0	5	1	14.90	13.94
1	5	1	-10.91	-11.44
2	5	1	6.23	4.95
3	5	1	14.18	12.85
4	5	1	-10.19	-8.87
-5	5	2	-12.33	-12.67
-4	5	2	8.26	7.80
-3	5	2	13.46	13.19
-2	5	2	-17.03	-16.05
-1	5	2	6.53	4.45
0	5	2	20.60	20.00
1	5	2	-12.23	-12.73
2	5	2	-6.21	-5.49
3	5	2	17.24	16.10
4	5	2	-3.86	-3.67
-5	5	3	10.92	10.17
-4	5	3	2.25	1.20
-3	5	3	-14.68	-13.70
-2	5	3	4.49	3.92
-1	5	3 •	6.53	5.45
0	5	3	-8.15	-6.98
1	5	3	2.65	4.97
2	5	3	5.31	4.79
3	5	3	-9.38	-8.64
-4	5	4	3.46	4.82
-3	5	4	-16.11	-15.24
-2	5	4	2.05	2.19
-1	5	4	14.69	14.11
0	5	4	-9.78	-8.80
1	5	4	-6.42	-0.54
2	5	4	13.05	13.75
-3	5	5	6.32	6.05
-2	5	5	2.05	0.61
-1	5	5	-8.77	-8.34
0	5	5	4.69	4.04
0	6	0 •	-2.04	-2.15
1	6	0 •	-4.88	-3.78
2	6	0 •	-8.77	-6.22
3	6	0 •	6.32	6.74
4	6	0 •	-2.24	-2.29
-5	6	1	9.69	8.43
-4	6	1	11.22	10.88
-3	6	1	-16.21	-15.71
-2	6	1	-6.01	-3.84
-1	6	1	20.91	21.41
0	6	1	-20.90	-21.81
1	6	1	1.93	2.51
2	6	1	15.91	16.84
3	6	1	-7.24	-7.11
4	6	1	-5.30	-3.85
-5	6	2	-2.04	-0.41
-4	6	2	6.93	6.37
-3	6	2	-4.58	-4.46
-2	6	2	3.27	2.92
-1	6	2	6.12	5.61
0	6	2	-3.56	-2.86
1	6	2	-5.90	-6.66
2	6	2	6.73	5.37
3	6	2	-2.05	-3.11
4	6	2	-3.96	-3.29
-5	6	3	4.19	2.90
-4	6	3	-12.23	-13.36
-3	6	3	7.04	4.54
-2	6	3	20.40	17.15
-1	6	3 •	-16.31	-18.16
0	6	3	1.23	2.86
1	6	3	17.64	19.67
2	6	3	-7.03	-7.89
-4	6	4	-3.46	-4.31
-3	6	4	2.05	1.74
-2	6	4	6.52	4.95
-1	6	4	-2.04	-0.86
0	6	4	-1.83	-1.68
1	6	4	9.29	6.60
-3	6	5	11.32	9.47
-2	6	5	-16.10	-14.37
-1	6	5	-2.04	-1.41
0	6	5	10.82	11.09
1	7	0 •	7.75	6.15
2	7	0 •	-2.04	-0.70
3	7	0 •	8.26	6.92
4	7	0 •	10.20	10.34
-5	7	1	13.77	14.00
-4	7	1	-13.36	-11.99
-3	7	1	-2.03	-2.29
-2	7	1	21.01	20.90
-1	7	1	-18.86	-17.94
0	7	1	-16.01	-15.41
1	7	1	18.97	19.04
2	7	1	-12.63	-11.65
3	7	1	-15.90	-15.41
4	7	1	13.05	13.20
-5	7	2	-3.26	-4.78
-4	7	2	2.05	1.12
-3	7	2	4.18	3.07
-2	7	2	-13.66	-11.28
-1	7	2	-4.17	-1.97
0	7	2	7.75	7.08
1	7	2	-9.17	-9.51
2	7	2	-6.11	-4.70
3	7	2	7.95	7.81
4	7	2	2.05	1.84
-5	7	3	-14.89	-13.62
-4	7	3	-3.35	-1.16
-3	7	3	18.66	17.97
-2	7	3	-8.56	-9.36
-1	7	3 •	-7.13	-6.11
0	7	3	18.77	18.47
1	7	3	-3.96	-6.69
2	7	3	-8.16	-8.36
3	7	3	11.01	10.78
-4	7	4	2.75	2.51
-3	7	4	-7.94	-6.86
-2	7	4	-2.04	-0.02
-1	7	4	8.37	7.29
0	7	4	-4.58	-4.94
-2	7	5	-2.85	-3.29
-1	7	5	13.26	12.93
0	8	0 •	22.04	25.20
1	8	0 •	-8.05	-7.44
2	8	0 •	-22.94	-21.99
3	8	0 •	22.64	22.45
4	8	0 •	-2.54	-1.32
-5	8	1	-3.46	-4.35
-4	8	1	-4.79	-4.87
-3	8	1	2.15	2.46
-2	8	1	-7.65	-6.22
-1	8	1	-17.64	-16.75
0	8	1	8.97	7.92
1	8	1	-2.04	-1.31
2	8	1	-4.48	-3.94
3	8	1	6.94	6.52
4	8	1	5.71	6.02
-5	8	2	-3.46	-4.93
-4	8	2	19.78	18.65
-3	8	2	-16.61	-15.87
-2	8	2	-4.28	-5.02
-1	8	2	18.66	18.07
0	8	2	-7.85	-7.27
1	8	2	-10.81	-10.89
2	8	2	18.77	18.61
3	8	2	-10.70	-8.83
-4	8	3	3.56	4.93
-3	8	3	-3.15	-1.27
-2	8	3	-9.99	-7.89
-1	8	3 •	6.63	6.01
0	8	3	-0.91	-3.41
1	8	3	-2.74	-4.58
2	8	3	2.15	2.54
-4	8	4	-15.19	-15.73
-3	8	4	-5.30	-4.54
-2	8	4	14.49	12.57
-1	8	4	-9.78	-9.47
0	8	4	-8.15	-8.33
1	8	4	12.24	13.88
1	9	0 •	-14.17	-14.73
2	9	0 •	16.42	15.64
3	9	0 •	5.71	5.90
4	9	0 •	-12.33	-12.08
-4	9	1	-3.67	-3.57
-3	9	1	-2.04	-1.32
-2	9	1	14.39	12.71
-1	9	1	-8.96	-7.75
0	9	1	-6.22	-5.49
1	9	1	13.36	12.06
2	9	1	-6.22	-4.94
3	9	1	-4.99	-3.91
4	9	1	5.91	5.78
-4	9	2	-5.30	-5.88
-3	9	2	-13.76	-12.08
-2	9	2	16.94	17.34
-1	9	2	-2.04	-1.23
0	9	2	-15.60	-15.97
2	9	2	6.23	5.22
3	9	2	-10.59	-10.66
-4	9	3	-7.54	-7.31
-3	9	3	6.73	6.48
-2	9	3	-6.52	-5.59
-1	9	3 •	-4.89	-4.75
0	9	3	5.50	5.45
1	9	3	-2.24	-3.31
2	9	3	2.14	0.84
-3	9	4	11.22	11.45
-2	9	4	-2.05	-2.99
-1	9	4	-10.70	-10.81
0	9	4	11.43	12.03
0	10	0 •	-9.88	-9.68
1	10	0 •	-3.05	-2.16
2	10	0 •	3.87	2.62
3	10	0 •	-7.85	-7.11
-4	10	1	-5.20	-6.77
-3	10	1	12.86	12.62
-2	10	1	2.05	1.91
-1	10	1	-15.19	-13.61
0	10	1	15.60	14.62
2	10	1	-10.40	-10.19
3	10	1	7.54	7.28
-4	10	2	-5.49	-5.41
-3	10	2	7.85	7.11
-2	10	2	2.45	1.02
-1	10	2	-7.43	-7.10
0	10	2	3.97	3.42
1	10	2	2.75	4.65
2	10	2	-10.50	-10.15
-4	10	3	6.02	7.01
-3	10	3	-4.48	-3.23
-2	10	3	-12.64	-10.84
-1	10	3 •	17.04	15.84
0	10	3	0.51	2.75
1	10	3	-10.19	-11.46
2	10	3	7.54	8.43
-2	10	4	-7.43	-7.46
-1	10	4	2.05	2.27
0	10	4	2.04	3.37
1	11	0 •	2.65	1.83
2	11	0 •	2.05	1.30
3	11	0 •	2.05	0.91
-4	11	1	13.05	12.90
-3	11	1	4.59	5.12
-2	11	1	-19.47	-18.33
-1	11	1	11.22	9.74
0	11	1	5.72	4.64
1	11	1	-24.88	-24.76
2	11	1	3.97	2.09
3	11	1	9.19	8.29
-4	11	2	-2.85	-3.76
-3	11	2	-2.54	-1.57
-2	11	2	2.55	2.15
-1	11	2	2.05	0.09
0	11	2	-4.48	-4.12
1	11	2	2.05	2.27
2	11	2	6.53	6.11
-3	11	3	-15.90	-15.66
-2	11	3	11.63	11.21
-1	11	3 •	6.63	7.21
0	11	3	-13.66	-14.62
1	11	3	5.09	6.95
2	11	3	6.33	6.37
0	12	0 •	-22.22	-24.88
1	12	0 •	4.49	4.64
2	12	0 •	11.73	10.72
3	12	0 •	-13.86	-14.41
-3	12	1	-11.10	-11.45
-2	12	1	2.05	1.90
-1	12	1	7.65	6.72
0	12	1	-9.28	-8.98
1	12	1	-2.04	-1.72
2	12	1	11.11	9.47
-3	12	2	9.69	9.88
-2	12	2	6.32	5.88
-1	12	2	-16.11	-15.30
0	12	2	8.26	7.67
1	12	2	2.05	4.77
2	12	2	-15.50	-15.54
-2	12	3	2.75	3.72
-1	12	3 •	-7.94	-7.79
0	12	3	-2.04	-1.41
1	12	3	3.16	5.02
1	13	0 •	13.57	12.95
2	13	0 •	-10.70	-11.09
-2	13	1	-4.89	-5.28
-1	13	1	3.68	3.65
0	13	1	2.05	1.72
1	13	1	-8.56	-7.45
2	13	1	2.86	2.09
-2	13	2	-14.89	-13.79
-1	13	2	2.05	0.23
0	13	2	13.56	13.64
1	13	2	-9.79	-8.69
0	14	0	7.95	7.56
1	14	0 •	-7.75	-7.09
2	14	0 •	-3.67	-3.49
-2	14	1	2.85	3.40
-1	14	1	10.50	10.77
0	14	1	-4.89	-4.66
1	14	1	-2.05	-2.29
-1	14	2	10.71	9.90

• = REFLEXIONS FOR WHICH EQUIVALENT REFLEXIONS COINCIDE FOR BOTH INDIVIDUALS OF TWIN

DERIVED VALUES OF THE NON-EQUIVALENT REFLEXIONS WHICH ARE SUPERPOSED ON THE DIFFRACTION PATTERN OF THE TWINNED CRYSTAL ARE LINKED IN PAIRS ulated for reasonable positions and included in the structure-factor calculations, but were not altered as the refinement progressed. When all shifts were small compared with the e.s.d. of the parameters, R was 0·097. At this stage the standard deviations of the bond lengths involving two light atoms were approx 0·04 Å, so that irregularities in the chain are not likely to be significant.

No absorption correction was applied, because the linear absorption coefficient for Cu $K\alpha$ is 56 cm^{-1}, and the crystal measured $0{\cdot}15 \times 0{\cdot}15 \times 0{\cdot}23$ mm, so that it approximated to a cylinder of radius 0·01 cm, for which the absorption correction A varies from 0·269 at $\sin\theta = 0$ to 0·264 at the limit of the data used.

It is interesting to note that the separation of the superimposed intensities was reasonably successful, as

Table 9. $|F|_{\text{obs}}$ *and* F_{calc} *for decamethonium bromide dihydrate*

H	K	L	F(OBS)	F(CALC)
0	0	0	-	117.00
4	0	0	-23.06	-22.98
6	0	0	-22.19	-22.60
1	1	0	-11.13	-9.43
2	1	0	-15.53	-17.21
3	1	0	6.72	5.30
4	1	0	-13.45	-13.59
5	1	0	18.98	17.90
6	1	0	10.20	8.63
7	1	0	6.46	6.35
8	1	0	11.23	9.98
9	1	0	-6.23	-5.74
0	2	0	21.39	22.24
1	2	0	9.76	9.20
2	2	0	-16.32	-14.68
3	2	0	22.21	24.13
4	2	0	8.64	8.25
6	2	0	7.62	6.83
7	2	0	-15.20	-15.52
9	2	0	-11.17	-11.74
1	3	0	31.31	34.00
2	3	0	-11.31	-10.60
3	3	0	-9.79	-8.32
4	3	0	-11.65	-11.73
5	3	0	-21.54	-22.21
6	3	0	8.72	5.80
7	3	0	-9.22	-9.45
11	3	0	7.31	9.13
0	4	0	-20.75	-19.71
1	4	0	-5.09	-4.44
2	4	0	-10.08	-9.77
4	4	0	18.12	17.06
6	4	0	12.17	12.32
2	5	0	15.95	17.47
4	5	0	7.99	8.70
5	5	0	-3.66	-1.97
6	5	0	-4.98	-3.63
12	5	0	4.41	5.32
1	6	0	-7.31	-6.21
2	6	0	5.70	4.56
3	6	0	-12.53	-12.31
6	6	0	-7.81	-9.73
8	6	0	-5.58	-4.22
5	7	0	9.82	10.21
3	8	0	8.36	8.62
4	8	0	-5.92	-4.28
7	8	0	-6.94	-6.93
4	9	0	-7.57	-8.12
7	9	0	-5.25	-3.03
6	10	0	5.73	5.01
5	11	0	-3.79	-4.03
3	12	0	-3.97	-4.30
-12	0	1	-7.92	-8.66
-10	0	1	7.33	6.71
-8	0	1	18.46	19.48
-6	0	1	12.09	11.22
-4	0	1	-23.29	-23.24
-2	0	1	-7.14	-6.20
2	0	1	17.37	18.81
4	0	1	19.70	20.51
8	0	1	-14.61	-14.69
10	0	1	-5.89	-6.83
-13	1	1	6.91	6.14
-12	1	1	6.05	5.23
-10	1	1	8.64	8.80
-9	1	1	-6.08	-6.71
-7	1	1	-13.89	-12.44
-6	1	1	-14.33	-12.54
-5	1	1	-3.09	-0.32
-4	1	1	-6.03	-5.69
-2	1	1	5.87	4.57
0	1	1	18.59	19.41
1	1	1	4.20	3.02
2	1	1	4.83	5.55
3	1	1	-18.56	-17.75
4	1	1	-16.26	-15.16
5	1	1	-4.70	-4.03
6	1	1	-15.52	-14.93
7	1	1	8.03	6.93
9	1	1	6.36	6.41
10	1	1	7.20	7.88
12	1	1	3.93	4.13
13	1	1	-3.48	-5.27
-15	2	1	3.34	4.78
-11	2	1	-7.82	-7.47
-9	2	1	-6.66	-6.40
-8	2	1	-7.29	-7.24
-7	2	1	11.72	11.65
-6	2	1	-1.62	-2.99
-5	2	1	11.49	11.46
-4	2	1	9.14	7.06
-3	2	1	9.39	9.82
-2	2	1	25.36	27.00
-1	2	1	-21.62	-23.98
0	2	1	6.06	4.96
1	2	1	-17.87	-18.91
2	2	1	-9.88	-10.39
3	2	1	8.21	7.16
5	2	1	14.87	16.25
7	2	1	5.47	5.12
8	2	1	4.65	4.65
9	2	1	-6.34	-6.80
10	2	1	0.01	1.53
11	2	1	-6.83	-8.23
-13	3	1	-4.14	-4.97
-12	3	1	4.16	4.28
-11	3	1	-3.99	-3.84
-10	3	1	5.59	6.19
-9	3	1	12.33	12.39
-7	3	1	15.31	16.01
-6	3	1	-8.26	-7.92
-5	3	1	-7.24	-6.84
-3	3	1	-15.98	-16.19
-1	3	1	-19.30	-18.04
0	3	1	9.48	8.86
1	3	1	14.85	15.99
2	3	1	5.58	5.47
3	3	1	17.29	18.98
4	3	1	-5.63	-4.72
5	3	1	8.21	7.31
6	3	1	-10.58	-11.45
7	3	1	-8.01	-7.97
9	3	1	-11.18	-11.81
10	3	1	2.95	2.62
13	3	1	3.76	4.15
-12	4	1	6.15	6.03
-11	4	1	6.63	6.35
-8	4	1	-14.92	-14.17
-6	4	1	-5.81	-5.74
-5	4	1	-12.12	-10.04
-4	4	1	15.18	15.40
-3	4	1	6.32	3.98
-2	4	1	18.02	19.02
-1	4	1	6.50	6.28
1	4	1	9.01	7.45
2	4	1	-14.72	-15.65
3	4	1	-3.76	-3.69
4	4	1	-10.58	-10.43
5	4	1	-13.55	-13.58
7	4	1	-3.90	-4.73
8	4	1	11.16	12.74
9	4	1	4.29	4.35
10	4	1	3.64	3.32
11	4	1	3.14	3.53
12	4	1	-5.20	-4.76
-11	5	1	-3.97	-2.37
-10	5	1	-5.40	-6.23
-6	5	1	11.05	11.16
-5	5	1	-2.90	-2.71
-4	5	1	12.46	13.54
-3	5	1	3.75	1.83
-2	5	1	-8.24	-7.02
-1	5	1	3.14	2.32
0	5	1	-18.31	-18.78
1	5	1	4.60	3.65
2	5	1	-8.40	-7.65
3	5	1	-3.18	-2.71
4	5	1	12.26	12.49
5	5	1	-4.57	-3.70
6	5	1	7.19	7.98
7	5	1	2.28	2.28
8	5	1	-2.64	-1.83
9	5	1	-3.19	-2.56
10	5	1	-7.57	-7.11
11	5	1	1.98	2.00
12	5	1	-3.45	-4.03
-11	6	1	7.49	8.18
-9	6	1	3.83	3.80
-8	6	1	6.94	7.74
-6	6	1	4.83	5.26
-5	6	1	-12.12	-11.72
-4	6	1	-8.38	-7.69
-3	6	1	-3.14	-2.63
-2	6	1	-15.46	-15.66
-1	6	1	10.02	9.34
1	6	1	11.65	11.48
2	6	1	10.53	10.56
4	6	1	10.25	9.85
5	6	1	-12.95	-14.30
6	6	1	-4.14	-4.61
7	6	1	-4.89	-5.48
8	6	1	-4.66	-4.18
9	6	1	3.40	2.90
10	6	1	-1.99	-3.20
-7	7	1	-10.11	-9.41
-3	7	1	13.28	12.89
-2	7	1	2.16	1.81
-1	7	1	6.22	5.68
1	7	1	-7.37	-6.94
3	7	1	-12.26	-12.18
4	7	1	-2.30	-2.27
5	7	1	-5.26	-4.95
6	7	1	-3.17	-2.95
7	7	1	9.76	10.40
8	7	1	-2.07	-1.34
9	7	1	4.48	5.24
-8	8	1	3.12	2.91
-7	8	1	5.11	5.00
-6	8	1	3.64	2.51
-5	8	1	8.06	7.31
-4	8	1	-5.32	-4.57
-2	8	1	-5.83	-5.41
-1	8	1	-7.15	-7.26
0	8	1	2.32	1.58
1	8	1	-10.16	-9.93
2	8	1	3.72	3.18
3	8	1	3.57	3.34
5	8	1	4.36	5.18
6	8	1	-3.61	-3.19
7	8	1	2.37	3.71
9	8	1	-3.18	-3.41
-10	9	1	5.49	5.75
-6	9	1	-5.53	-5.99
-4	9	1	-8.79	-8.28
-3	9	1	-5.12	-4.52
-2	9	1	2.41	2.60
2	9	1	4.99	4.48
3	9	1	4.97	4.55
4	9	1	-7.94	-6.51
6	9	1	-6.27	-6.71
7	9	1	-4.87	-0.83
10	9	1	3.33	4.49
-8	10	1	-4.34	-5.17
-2	10	1	7.24	5.97
1	10	1	-4.83	-3.85
2	10	1	-7.61	-7.19
4	10	1	-4.44	-4.57
-8	11	1	-2.74	-0.35
-3	11	1	-4.56	-4.63
3	11	1	2.96	3.73
-5	12	1	-3.58	-5.56
5	12	1	-3.11	-4.40
4	2	1	-8.18	-7.35
-12	0	2	-5.04	-5.20
-10	0	2	-16.52	-15.21
-6	0	2	22.11	22.58
-4	0	2	35.36	34.58
-2	0	2	6.37	3.88
0	0	2	-34.14	-35.35
2	0	2	-19.25	-19.53
4	0	2	12.77	11.68
6	0	2	19.28	18.45
10	0	2	-7.38	-9.24
-12	1	2	-4.47	-3.81
-11	1	2	4.35	5.26
-9	1	2	10.64	9.36
-8	1	2	11.48	10.06
-7	1	2	-12.60	-10.90
-6	1	2	10.53	9.11
-5	1	2	2.36	0.07
-4	1	2	-17.22	-18.53
-3	1	2	-9.54	-6.50
-2	1	2	-20.58	-21.29
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1	1	2	23.48	25.36
2	1	2	23.01	24.45
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7	1	2	-6.85	-6.51
8	1	2	-11.57	-11.48
-13	2	2	7.95	7.51
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-11	2	2	2.65	2.36
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-7	2	2	-13.03	-13.26
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4	2	2	-1.77	-1.65
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6	2	2	-3.22	-3.14
7	2	2	13.60	12.87
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7	3	2	7.58	7.07
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1	4	2	11.19	10.40
2	4	2	11.79	10.03
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6	4	2	-13.36	-12.34
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3	8	2	-9.51	-9.64
4	8	2	5.57	5.10
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-4	11	2	3.57	3.11
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5	11	2	3.10	3.88
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-12	0	3	9.49	10.42
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-8	0	3	-7.98	-7.85
-6	0	3	-15.88	-12.59
-4	0	3	13.56	12.84
2	0	3	-12.46	-11.98
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6	1	3	11.95	12.58
7	1	3	-5.45	-5.32
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10	4	3	-4.45	-4.32
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-10	6	3	-2.72	-2.13
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-1	6	3	-6.46	-5.60
0	6	3	-3.31	-1.99
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3	6	3	-2.84	-1.72
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-6	8	3	-4.94	-3.48
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0	8	3	-3.60	-2.82
1	8	3	6.57	7.12
2	8	3	-3.63	-3.88
3	8	3	-2.37	-2.35
4	8	3	-2.72	-3.45
5	8	3	-6.39	-6.19
6	8	3	2.57	2.78
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-7	9	3	-3.81	-4.15
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0	9	3	-10.11	-8.90
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1	3	4	7.96	8.09
2	3	4	-6.80	-5.37
3	3	4	3.38	2.20
4	3	4	-6.19	-4.87
5	3	4	-9.98	-10.07
7	3	4	-8.60	-7.50
8	3	4	4.38	3.89
9	3	4	4.15	2.94
-12	4	4	-3.20	-3.34
-10	4	4	-7.59	-7.22
-9	4	4	-7.97	-7.31
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-6	4	4	9.98	10.18
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-2	4	4	-9.63	-7.72
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2	4	4	-7.90	-6.58
3	4	4	-9.52	-9.75
4	4	4	6.76	5.50
6	4	4	8.07	8.66
9	4	4	5.21	5.17
-12	5	4	-6.48	-5.28
-10	5	4	3.66	3.18
-8	5	4	9.68	11.25
-6	5	4	7.64	6.29
-5	5	4	-3.91	-3.18
-4	5	4	-10.14	-8.72
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-2	5	4	-18.22	-19.03
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0	5	4	-6.64	-5.85
2	5	4	14.71	12.91
3	5	4	3.01	2.35
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5	5	4	-4.84	-2.80
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-12	6	4	3.71	2.69
-10	6	4	6.04	4.98
-9	6	4	-3.65	-3.84
-7	6	4	-7.97	-6.80
-6	6	4	-9.72	-7.97
-4	6	4	-4.92	-4.50
-3	6	4	5.47	4.31
-1	6	4	4.39	4.53
0	6	4	11.27	10.60
1	6	4	-5.45	-3.92
3	6	4	-9.41	-8.70
4	6	4	-3.63	-3.59
6	6	4	-3.22	-4.28
-11	7	4	-6.03	-5.02
-9	7	4	-8.41	-7.11
-7	7	4	6.35	4.43
-5	7	4	12.07	11.63
-4	7	4	6.18	4.19
-1	7	4	-5.48	-7.83
0	7	4	-4.49	-3.63
1	7	4	-9.38	-8.86
4	7	4	-6.71	-1.87
5	7	4	6.39	6.70
-9	8	4	7.26	5.82
-7	8	4	6.16	5.28
-6	8	4	-6.67	-5.77
-1	8	4	-5.90	-5.16
0	8	4	9.40	7.01
3	8	4	6.58	6.19
-8	9	4	-3.97	-4.04
-6	10	4	3.64	4.46
-4	10	4	4.04	4.81

judged by the fact that the final R was 0·095 for the reflexions $hk0$ and $\bar{1}k3$ common to both individuals whereas R was 0·137 for the F-values derived for other $hk3$ reflexions, the overall R-factor for all reflexions being 0·097. Small scaling errors indicate that the volume of the smaller twin crystal was in fact slightly less than one-fifth of that of the main individual, so that the common reflexions (which tend to equivalence through twinning) have been slightly over-corrected.

APPENDIX II

Decamethonium bromide

The bromine atoms were located from Patterson projections, and after attempts to solve the structure from the projections alone had failed, the chains were located from a 3-D Fourier synthesis. The refinement was carried out in exactly the same way as for the hexamethonium compound.

References

BERGMAN, F. (1955). *The Physical Chemistry of Enzymes.* Publ. no. 20, p. 126. London: Faraday Society.

CANEPA, F. G. (1962). *Nature, Lond.* **195**, 573.

CANEPA, F. G. (1963). *Acta Cryst.* **16**, 145.

International Tables for X-ray Crystallography (1962). Vol. III. p. 136. Birmingham: Kynoch Press.

MILLEDGE, H. J. & MILLEDGE, D. (1961). In *Computing Methods and the Phase Problem in X-ray Crystal Analysis.* Ed. R. Pepinsky, J. M. Robertson and J. C. Speakman, p. 79. London: Pergamon Press.

PATON, W. D. M. & ZAIMIS, E. J. (1951). *Brit. J. Pharmacol.* **6**, 155.

PATON, W. D. M. & ZAIMIS, E. J. (1952). *Pharmacol. Rev.* **4**, 219.

ZAIMIS, E. J., CANNARD, T. H. & PRICE, H. L. (1957). *Science,* **128**, 34.

Acta Cryst. (1965). **19**, 840

The Crystal Structure of Bicyclohexylidene, $C_{12}H_{20}$

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Bicyclohexylidene crystallizes in the triclinic space group $P\bar{1}$ with the unit-cell dimensions $a=5·32$, $b=6·25$, $c=8·36$ Å, $\alpha=107·2°$, $\beta=79·1°$, $\gamma=105·6°$. $D_x=1·073$ g.cm^{-3}, $D_m=1·066$ g.cm^{-3}, $Z=1$.

The phase problem was solved from the Patterson function. The final anisotropic parameters for the C atoms and positional parameters of the H atoms were obtained by three-dimensional least-squares refinement. For the calculation of interatomic distances the atomic coordinates were corrected for rigid-body thermal libration of the molecule.

The molecule consist of two cyclohexane rings in the chair conformation with a center of symmetry at the mid-point of the double bond linking them. The six carbon atoms contiguous with the double bond are planar. The whole molecule has a non-crystallographic mirror plane perpendicular to this plane and passing through the center of symmetry and the center of the two cyclohexane rings.

The length of the C=C double bond is 1·332 Å and the single bonds in the ring vary between 1·509 Å and 1·533 Å with e.s.d. 0·004 Å. The mean length of the ten independent C–H bonds is 1·01 Å, with individual deviations from the mean averaging 0·03 Å. The shortest distance between hydrogen atoms belonging to neighbouring carbon atoms of the molecule is 1·944 Å. The angles in the ring vary between 110·4° and 111·9° but conform to the mirror-plane symmetry of the molecule. The intermolecular atomic radii for hydrogen and carbon are 1·27 Å and 1·87 Å, respectively.

Introduction

The compound $C_{12}H_{20}$ was first prepared synthetically by Criegee, Vogel & Horger (1952) by zinc powder dehalogenization of 1,1′-dibromobicyclohexyl. The molecule was shown to consist of two cyclohexane rings linked by a double bond. The conformation and detailed stereochemistry of this molecule are of interest because of the non-bonding hydrogen atom interactions which might distort the cyclohexane rings from the customary strain-free chair conformation. In fact, Tömösközi (1959) has proposed that the boat form would be the more stable for this reason.

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