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The Crystal and Molecular Structure of 7,8-Epoxyhalochamigrene

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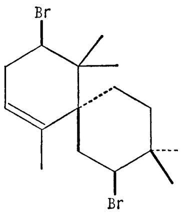
The molecular structure of 7,8-epoxyhalochamigrene, $C_{15}H_{23}OBr_2Cl$, has been confirmed by means of X-ray crystallographic analysis. The crystals are orthorhombic, with sixteen molecules in a unit cell with dimensions of $a=10.303$, $b=71.31$, and $c=9.061$ Å; the space group is $P2_12_12_1$. 4241 independent intensity data were collected on a four-circle diffractometer with graphite-monochromated Cu $K\alpha$ radiation. The structure was solved by the Monte Carlo direct method, using 20 reflections as a starting set, and was refined by the block-diagonal least-squares method. The final R value was 0.072. The molecular structure thus obtained corresponds to that proposed by Howard and Fenical on the basis of chemical and spectroscopic evidence. The four crystallographically-independent molecules have almost the same geometries. Of the two spiro-linked six-membered rings, the one having the epoxy group takes a half-chair conformation, while the other adopts a chair conformation. The crystal consists of alternating single and triple molecular layers parallel to the (010) plane; the latter layer has a molecular arrangement approximately of $P2_12_12_1$ symmetry.

A number of halogenated chamigrene derivatives[1] such as halochamigrene[2] have been isolated from the red algae of the genus *Laurencia*. 7,8-Epoxyhalochamigrene, which had already been obtained by oxidation of halochamigrene,[2a] was recently reported to exist also in nature, *e.g.*, in the red algae *L. nipponica*[3] and *L. okamurai*.[4] On the basis of the chemical and spectroscopic evidence, the **1** and **2** structures were proposed for halochamigrene and 7,8-epoxyhalochamigrene respectively;[2] however, there was no conclusive proof as to these structures. Therefore, in order to establish **1** and **2**, we have now undertaken an X-ray crystallographic study of 7,8-epoxyhalochamigrene.



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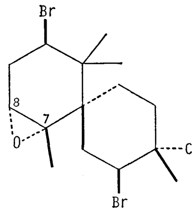

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Experimental

A colorless single crystal cut into a cube with an edge of about 0.2 mm was used for the X-ray experiment. The crystal data are summarized in Table 1. The unit-cell dimensions and diffraction intensities were measured on a Rigaku four-circle diffractometer with graphite-monochromated Cu $K\alpha$ radiation ($\lambda=1.54178$ Å). Because of the long b unit-cell edge, it was necessary to obtain high resolution of the diffraction peaks. For this purpose, a Rigaku RU-200 X-ray tube with a focal size of 0.3×0.3 mm² was employed (50 kV, 60 mA). The θ-2θ continuous-scan technique was applied at a θ scan rate of $10°$ min^{-1}; the scan width in θ was $(0.6+0.142\tan\theta)°$. The background was measured for 5 s at each end of the scan range. Three standard reflections, measured at intervals of every 200 reflections, showed no significant decrease in intensity during the course of the data collection. The intensities were corrected for the Lorentz and polarization factors, but not for absorption or extinction effect. In the range of 2θ values up to 120°, 4241 independent structure factors above the $3\sigma(F)$ level were selected for the structure determination.

Table 1. The crystal data

$C_{15}H_{23}OBr_2Cl$	Mol wt 414.61
Crystal system	Orthorhombic
Space group	$P2_12_12_1$
Cell dimensions	$a=10.303(2)$ Å
	$b=71.31(1)$ Å
	$c=9.061(8)$ Å
V	6658 Å³
Z	16
D_c	1.655 g cm^{-3}
μ(Cu $K\alpha$)	76.8 cm^{-1}

Structure Determination

Because of the presence of eight independent bromine atoms, the heavy-atom method seemed somewhat troublesome to apply to the present structure; hence, the Monte Carlo direct method[5] was tried. The 20 reflections with the greatest $|E|$ values were chosen as a starting set. In order to extend each of the tentative-phase sets derived from random numbers, 12 cycles of the tangent procedure were performed using 1111 $|E|$ values above 1.30. Since the phase set extended from the 66thly-generated random set showed a low R_K value of 0.258 ($R_K=\Sigma||E_o|-k|E_c||/\Sigma|E_o|$), 10 additional cycles of the tangent procedure were carried out by the use of 1347 $|E|$ values above 1.20; the R_K value became 0.268. An E-map based on 1286 phases afforded all the 76 non-hydrogen atoms.

After the structure had been refined by the block-diagonal least-squares method with anisotropic temperature factors, the absolute configuration was determined by taking account of the anomalous dispersion of bromine and chlorine atoms for Cu $K\alpha$ radiation. The R ratio for the two enantiomeric structures, 1.018, rejected one of them at the 99.5% confidence level.[6] A difference Fourier map revealed all the 92 hydrogen atoms. Further least-squares refinements were performed including the hydrogen atoms and the anomalous dispersion effects. The weighting scheme used was as follows:

$$w = 1/\{\sigma(F)^2 \exp(c_1X^2+c_2Y^2+c_3XY+c_4X+c_5Y)\},$$

where $X=|F_o|$ and $Y=\sin\theta/\lambda$. The c_1, c_2, c_3, c_4, and c_5 coefficients were evaluated from the $(\Delta F)^2$ distribution; $c_1=0.7702\times10^{-6}$, $c_2=8.060$, $c_3=0.01900$, $c_4=-0.4314\times10^{-2}$, and $c_5=-8.129$. In this manner, the R value reached 0.072. The final atomic parameters are listed in Table 2.[7]

The calculations were done on a HITAC M-200H computer at the Hokkaido University Computing Center, using our own programs. The atomic scattering factors and dispersion corrections were taken from the International Tables for X-Ray Crystallography.[8]

Results and Discussion

Molecular Structure. The molecular skeleton obtained is shown in Fig. 1, while the bond distances and angles and the torsion angles for the spiro[5.5]-undecane systems are given in Tables 3 and 4 respectively. From these results, it is concluded that the structure of 7,8-epoxyhalochamigrene, including the absolute configuration, corresponds to **2**; consequently the structure of halochamigrene is **1**. The four crystallographically-independent molecules have almost the same geometries. The average bond distances and angles for these four molecules are given in the last column of Table 3.

Table 2. The final atomic parameters and estimated standard deviations

The e.s.d.'s given in the parentheses refer to the last decimal position.

Atom	$x\times10^3$	$y\times10^4$	$z\times10^3$	B_{eq}[a]/Å^2	Atom	$x\times10^3$	$y\times10^4$	$z\times10^3$	B_{eq}[a]/Å^2
Br(A1)	340.3(3)	581.3(2)	213.1(2)	6.14	Br(C1)	249.7(2)	923.6(2)	920.9(2)	5.23
Br(A2)	756.7(3)	−374.1(4)	382.0(3)	9.76	Br(C2)	466.4(3)	1976.4(2)	643.3(3)	7.14
Cl(A)	287.6(5)	620.9(5)	588.0(6)	5.56	Cl(C)	574.0(6)	797.6(6)	909.3(6)	6.87
O(A)	348(1)	−176(1)	576(1)	5.12	O(C)	598(1)	1362(1)	477(1)	5.31
C(A1)	430(2)	222(2)	302(2)	4.04	C(C1)	350(2)	1248(2)	774(2)	3.75
C(A2)	351(2)	387(2)	371(2)	4.59	C(C2)	390(2)	1046(2)	816(2)	4.22
C(A3)	409(2)	463(2)	508(2)	4.22	C(C3)	516(2)	1042(2)	897(2)	4.56
C(A4)	416(2)	296(2)	617(1)	4.18	C(C4)	623(2)	1147(2)	809(2)	4.63
C(A5)	511(2)	143(2)	557(2)	4.15	C(C5)	584(2)	1354(2)	783(2)	3.93
C(A6)	467(2)	63(2)	409(2)	3.69	C(C6)	453(2)	1374(2)	699(2)	4.04
C(A7)	346(2)	−72(2)	432(2)	4.08	C(C7)	468(2)	1319(2)	539(2)	3.85
C(A8)	354(2)	−278(2)	437(2)	4.63	C(C8)	487(2)	1463(2)	416(2)	5.17
C(A9)	490(3)	−375(2)	417(2)	6.91	C(C9)	485(2)	1671(2)	453(2)	5.98
C(A10)	592(2)	−239(2)	430(2)	5.89	C(C10)	496(2)	1704(2)	614(2)	4.82
C(A11)	585(2)	−54(2)	336(2)	5.10	C(C11)	403(2)	1583(2)	711(2)	4.67
C(A12)	708(2)	58(2)	333(2)	5.63	C(C12)	406(2)	1654(2)	872(2)	5.55
C(A13)	550(2)	−110(2)	177(2)	5.21	C(C13)	266(2)	1600(2)	651(2)	5.34
C(A14)	209(2)	−1(2)	402(2)	5.17	C(C14)	426(2)	1128(2)	480(2)	3.73
C(A15)	531(2)	567(2)	499(3)	6.11	C(C15)	509(2)	1113(2)	1058(2)	5.43
Br(B1)	−230.8(3)	1570.0(2)	153.5(2)	6.06	Br(D1)	596.9(3)	2165.2(2)	134.8(3)	6.51
Br(B2)	−53.4(3)	470.3(2)	338.2(3)	6.87	Br(D2)	377.5(3)	3253.4(2)	307.2(4)	9.18
Cl(B)	92.3(7)	1678.0(6)	194.4(9)	9.58	Cl(D)	275.8(7)	2046.1(6)	149.7(8)	8.98
O(B)	92(1)	1039(2)	569(1)	5.20	O(D)	240(1)	2672(2)	526(1)	5.68
C(B1)	−144(2)	1223(2)	274(1)	3.73	C(D1)	500(2)	2508(2)	250(2)	3.64
C(B2)	−97(2)	1426(2)	255(2)	4.19	C(D2)	454(2)	2303(2)	228(2)	4.58
C(B3)	37(2)	1436(2)	172(2)	6.88	C(D3)	329(2)	2291(2)	140(2)	5.47
C(B4)	131(2)	1319(2)	261(2)	4.83	C(D4)	227(2)	2403(2)	222(2)	5.33
C(B5)	84(2)	1113(2)	265(2)	4.37	C(D5)	269(2)	2614(2)	226(2)	4.44
C(B6)	−45(2)	1083(2)	342(2)	3.54	C(D6)	393(2)	2647(2)	308(2)	4.71
C(B7)	−33(2)	1106(2)	508(2)	4.39	C(D7)	374(2)	2615(2)	480(2)	4.12
C(B8)	−25(2)	943(2)	611(2)	4.94	C(D8)	347(2)	2783(2)	579(2)	5.39
C(B9)	−38(2)	753(2)	561(2)	5.57	C(D9)	357(2)	2976(2)	524(2)	5.91
C(B10)	−16(2)	740(2)	391(2)	4.45	C(D10)	352(2)	2989(2)	356(2)	5.90
C(B11)	−102(2)	876(2)	306(2)	4.45	C(D11)	445(2)	2857(2)	279(2)	4.56
C(B12)	−93(2)	828(2)	140(2)	5.08	C(D12)	446(2)	2899(2)	113(2)	6.55
C(B13)	−244(2)	859(2)	350(2)	5.37	C(D13)	583(2)	2875(2)	339(2)	5.20
C(B14)	−69(2)	1283(2)	590(2)	5.80	C(D14)	414(2)	2442(2)	556(2)	5.91
C(B15)	24(2)	1389(2)	11(2)	5.68	C(D15)	347(3)	2335(3)	−26(2)	6.98

a) $B_{eq} = 8\pi^2(u_1^2+u_2^2+u_3^2)/3$, where u_i is the root-mean-square deviation in the ith principal axis of the thermal ellipsoid.

TABLE 3. THE BOND DISTANCES AND ANGLES, WITH THEIR STANDARD DEVIATIONS

	Mol. A	Mol. B	Mol. C	Mol. D	Mean
Bond distance	*l*/Å	*l*/Å	*l*/Å	*l*/Å	*l*/Å
C(1)-C(2)	1.56(2)	1.53(2)	1.55(2)	1.55(2)	1.55
C(1)-C(6)	1.54(2)	1.55(2)	1.54(2)	1.58(2)	1.55
C(2)-C(3)	1.48(2)	1.58(3)	1.49(3)	1.52(3)	1.52
C(2)-Br(1)	2.00(1)	1.95(2)	1.94(2)	1.96(2)	1.96
C(3)-C(4)	1.55(2)	1.51(3)	1.55(3)	1.51(3)	1.53
C(3)-C(15)	1.46(3)	1.51(3)	1.55(2)	1.55(3)	1.52
C(3)-Cl	1.83(2)	1.83(2)	1.85(2)	1.83(2)	1.84
C(4)-C(5)	1.56(2)	1.54(2)	1.55(2)	1.56(2)	1.55
C(5)-C(6)	1.52(2)	1.52(2)	1.56(3)	1.50(3)	1.53
C(6)-C(7)	1.59(2)	1.52(2)	1.51(2)	1.58(2)	1.55
C(6)-C(11)	1.62(3)	1.61(2)	1.58(2)	1.62(2)	1.61
C(7)-C(8)	1.47(2)	1.50(2)	1.52(2)	1.52(2)	1.50
C(7)-C(14)	1.52(3)	1.51(2)	1.52(2)	1.48(2)	1.51
C(7)-O	1.50(2)	1.48(2)	1.49(2)	1.49(2)	1.49
C(8)-C(9)	1.57(3)	1.43(2)	1.52(2)	1.46(2)	1.50
C(8)-O	1.46(2)	1.44(2)	1.46(2)	1.44(2)	1.45
C(9)-C(10)	1.44(3)	1.56(3)	1.48(3)	1.53(3)	1.50
C(10)-C(11)	1.57(2)	1.52(2)	1.56(2)	1.51(3)	1.54
C(10)-Br(2)	2.00(2)	2.02(1)	1.98(1)	1.96(1)	1.99
C(11)-C(12)	1.50(3)	1.54(2)	1.54(2)	1.53(3)	1.53
C(11)-C(13)	1.53(2)	1.52(3)	1.52(3)	1.53(3)	1.53
C-H					1.03
Bond angle	*ϕ*/°	*ϕ*/°	*ϕ*/°	*ϕ*/°	*ϕ*/°
C(2)-C(1)-C(6)	115(1)	117(1)	118(1)	115(1)	116
C(1)-C(2)-C(3)	114(1)	112(1)	112(1)	113(1)	113
C(1)-C(2)-Br(1)	105(1)	109(1)	110(1)	107(1)	108
C(3)-C(2)-Br(1)	112(1)	112(1)	113(1)	113(1)	113
C(2)-C(3)-C(4)	106(1)	106(2)	111(1)	107(1)	108
C(2)-C(3)-C(15)	119(2)	112(2)	115(2)	113(2)	115
C(2)-C(3)-Cl	106(1)	105(1)	109(1)	106(1)	107
C(4)-C(3)-C(15)	113(2)	117(2)	111(1)	117(2)	115
C(4)-C(3)-Cl	105(1)	106(1)	105(1)	106(1)	106
C(15)-C(3)-Cl	107(1)	110(1)	106(1)	106(1)	107
C(3)-C(4)-C(5)	110(1)	110(2)	110(1)	110(2)	110
C(4)-C(5)-C(6)	113(1)	115(1)	113(1)	113(1)	114
C(1)-C(6)-C(5)	111(1)	108(1)	109(1)	109(1)	109
C(1)-C(6)-C(7)	109(1)	112(1)	110(1)	109(1)	110
C(1)-C(6)-C(11)	108(1)	106(1)	107(1)	107(1)	107
C(5)-C(6)-C(7)	110(1)	111(1)	111(1)	111(1)	111
C(5)-C(6)-C(11)	109(1)	111(1)	109(1)	110(1)	110
C(7)-C(6)-C(11)	109(1)	109(1)	110(1)	110(1)	110
C(6)-C(7)-C(8)	124(2)	123(1)	122(1)	119(1)	122
C(6)-C(7)-C(14)	120(1)	124(1)	123(1)	123(1)	123
C(6)-C(7)-O	113(1)	114(1)	113(1)	111(1)	113
C(8)-C(7)-C(14)	113(1)	111(1)	113(1)	116(1)	113
C(8)-C(7)-O	59(1)	58(1)	58(1)	57(1)	58
C(14)-C(7)-O	109(1)	107(1)	108(1)	111(1)	109
C(7)-C(8)-C(9)	119(2)	122(1)	120(1)	122(2)	121
C(7)-C(8)-O	62(1)	61(1)	60(1)	61(1)	61
C(9)-C(8)-O	111(1)	116(2)	114(2)	118(2)	115
C(8)-C(9)-C(10)	110(1)	111(1)	112(1)	113(1)	112
C(9)-C(10)-C(11)	120(2)	112(1)	115(1)	114(2)	115
C(9)-C(10)-Br(2)	106(1)	105(1)	106(1)	106(1)	106
C(11)-C(10)-Br(2)	109(1)	112(1)	112(1)	114(1)	112
C(6)-C(11)-C(10)	104(1)	106(1)	107(1)	107(1)	106
C(6)-C(11)-C(12)	112(1)	112(1)	112(1)	110(1)	112
C(6)-C(11)-C(13)	110(2)	112(1)	111(1)	109(1)	111
C(10)-C(11)-C(12)	115(2)	109(1)	110(1)	109(1)	111
C(10)-C(11)-C(13)	107(1)	112(1)	109(1)	112(1)	110
C(12)-C(11)-C(13)	109(1)	107(2)	109(2)	109(2)	109
C(7)-O-C(8)	59(1)	62(1)	62(1)	63(1)	62

Table 4. The torsion angles for the six-membered rings

	Mol. A	Mol. B	Mol. C	Mol. D
	$\phi/°$	$\phi/°$	$\phi/°$	$\phi/°$
C(6)-C(1)-C(2)-C(3)	50	51	49	50
C(1)-C(2)-C(3)-C(4)	−59	−57	−54	−58
C(2)-C(3)-C(4)-C(5)	64	62	60	64
C(3)-C(4)-C(5)-C(6)	−61	−63	−58	−63
C(4)-C(5)-C(6)-C(1)	47	51	50	50
C(5)-C(6)-C(1)-C(2)	−41	−45	−45	−44
C(11)-C(6)-C(7)-C(8)	20	21	23	26
C(6)-C(7)-C(8)-C(9)	−1	−5	−3	−9
C(7)-C(8)-C(9)-C(10)	13	19	15	19
C(8)-C(9)-C(10)-C(11)	−51	−53	−48	−50
C(9)-C(10)-C(11)-C(6)	70	70	69	68
C(10)-C(11)-C(6)-C(7)	−47	−51	−52	−53

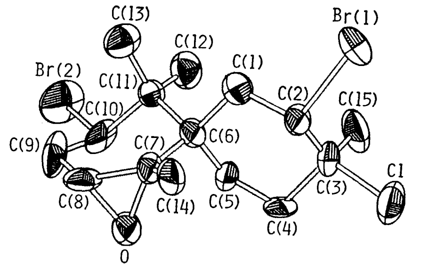

Fig. 1. A perspective view of the 7,8-epoxyhalochamigrene A-molecule, and the crystallographic numbering system.
Non-hydrogen atoms are represented as thermal ellipsoids enclosing 50% probabilities.

Of the two spiro-linked six-membered rings, the cyclohexane ring having the epoxy group takes a half-chair conformation; the Br(2), C(1), and C(12) atoms are equatorially or pseudoequatorially bonded to this ring, while the C(5) and C(13) atoms are axially or pseudoaxially oriented. On the other hand, the remaining six-membered ring has a somewhat deformed chair conformation; the Br(1), Cl, and C(11) atoms take the equatorial orientations, while the C(7) and C(15) atoms occupy the axial positions. Such a conformation of the spiro[5.5]undecane system brings the C(2)H and C(14)H_3 groups very close to each other; the C(2)···C(14) distances for the four molecules are 3.11—3.21 Å. The severe steric repulsion between these groups results in a considerable flattening of the C(5)–C(6)–C(1)–C(2) moiety (see Table 4).

In all four molecules, the C(6)–C(11) bond distance (1.58—1.62 Å) is appreciably longer than the standard C–C single-bond distance.9) This bond lengthening is probably due to the strong steric repulsions between the bulky groups attached to the C(6) and C(11) atoms. The fact that the C(3)–C(2)–Br(1) bond angle is greater than the C(1)–C(2)–Br(1) angle on the average by 5° indicates that the Cl atom and C(15)H_3 group repel the Br(1) atom to a considerable extent. The Br(2) atom also suffers a similar steric hindrance from the C(12)H_3 and C(13)H_3 *gem*-dimethyl groups; the average difference between the C(9)–C(10)–Br(2) and C(11)–C(10)–Br(2) bond angles is 6°.

As to the valence angles at the C(3) atom, an interesting fact is found: the sum of the average C(2)–C(3)–C(4), C(2)–C(3)–C(15), and C(4)–C(3)–C(15) bond angles, 338°, is considerably larger than three times the tetrahedral angle, while all the average C(2)–C(3)–Cl, C(4)–C(3)–Cl, and C(15)–C(3)–Cl bond angles are smaller than the tetrahedral angle. This suggests that the C(3) atomic orbital used for the C(3)–Cl bond may involve a greater amount of p-character than the pure sp^3 hybrid orbital, and that, as the result, the C(3)–Cl bond may be somewhat lengthened. In fact, the average C(3)–Cl bond distance of 1.84 Å is significantly longer than the usual C–Cl distance, 1.77 Å.9)

Crystal Structure. The most interesting point in the present structure is how the four crystallographically-independent A-, B-, C-, and D-molecules are arranged. As Fig. 2 shows, the crystal may be taken as composed of the following three kinds of molecular layers parallel to the (010) plane: (1) A layers consisting of only A-molecules and placed at $y=0$ or 1/2; (2) D layers made up of only D-molecules and situated at $y=1/4$ or 3/4; (3) BC layers containing both B- and C-molecules and lying between the A and D layers. The structure has the sequence of layers, A, BC, D, BC, A, ..., repeating after four layers.

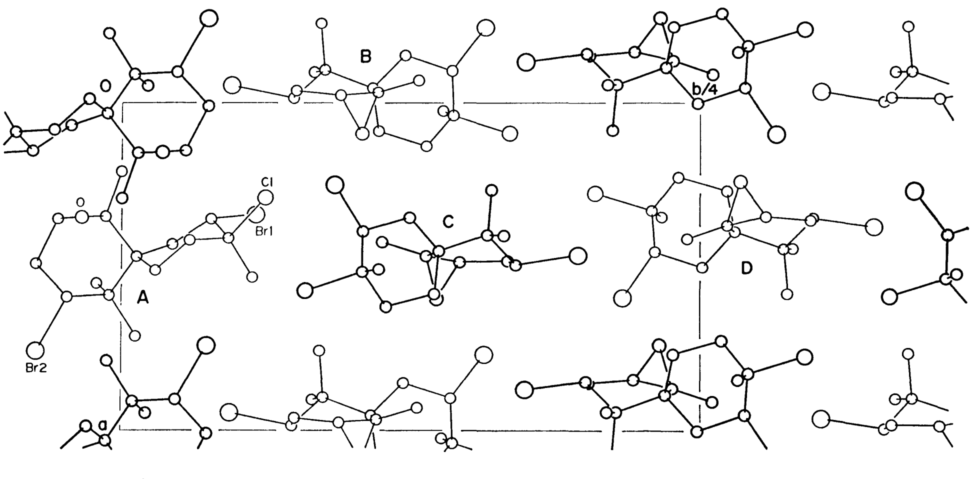

Fig. 2. The molecular arrangement viewed along the c axis.

Although the B- and C-molecules are independent, they are related to each other by an approximate two-fold screw axis along the a axis. These molecules are also related to the D-molecule by an approximate two-fold screw axis (with a translation of $b/8$) along the b axis or by an approximate two-fold rotation axis along the c axis. Consequently, the molecular arrangement in the triple molecular layer formed by adjacent BC, D, and BC layers looks as if it were part of a structure having the $P2_12_12$ space group and unit-cell dimensions of $a'=a$, $b'=b/4$, and $c'=c$. These triple molecular layers and the monomolecular A layers are alternately stacked. The orientation of the A-molecule is substantially different from those of the other three molecules; *e.g.*, the two molecules marked with A and B in Fig. 2 do not take the same orientation until either of them rotates by about 90° around a special axis lying nearly in the $[\bar{8}10]$ direction.

The molecules are bonded mainly by the van der Waals interactions. The halogen atoms are arranged near either surface of the molecular layer to which they belong. The intermolecular halogen-halogen distances, which are shorter than the usual van der Waals distances, are given in Table 5. Of these, the closest Br(A1)···Br(C1), Br(A2)···Cl(C), and Cl(B)···Cl(D) contacts correspond to the apparent reductions, about 0.17 and 0.09 Å respectively, of the chlorine and bromine van der Waals radii.

Table 5. The intermolecular halogen-halogen distances shorter than the usual van der Waals distances

		l/Å
Br(A1)	Br(C1)$^{a)}$	3.720(3)
Br(A2)	Cl(C)$^{b)}$	3.497(5)
Br(C1)	Cl(A)$^{c)}$	3.729(5)
Cl(B)	Cl(D)$^{c)}$	3.260(8)

The symmetry codes are as follows: a) $x, y, -1+z$; b) $3/2-x, -y, -1/2+z$; c) x, y, z.

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