sur le plan $y=\frac{1}{4}$ en une région inoccupée (Fig. 6) et que l'assemblage des piles, dans le plan $y=\frac{1}{4}$, n'est pas le plus compact possible (Fig. 5).

Références

ALMENNINGEN, A., BASTIANSEN, O. & HANSEN, L. (1955). *Acta Chemica Scandinavia*, **9**, 1306.

BAK, B., HANSEN, L. & RASTRUP-ANDERSEN, J. (1954). *J. Chem. Phys.* **22**, 2013.

COCHRAN, W. (1948). *J. Sci. Instrum.* **25**, 253.

DONOHUE, J. (1952). *J. Phys. Chem.* **56**, 502.

ELLER, G. VON (1955). *Bull. Soc. franç. Minér. Crist.* **78**, 157.

FRIEDLANDER, P. H., LOVE, W. & SAYRE, O. (1955). *Acta Cryst.* **8**, 732.

International Tables for X-ray Crystallography (1959), vol. 2, p. 326. Birmingham: Kynoch Press.

KUPFER, D. (1961). Thèse, Faculté de Médecine, Paris.

PAULING, L. (1960). *The Nature of the Chemical Bond*, p. 140. New York: Wiley.

RERAT, C. (1960*a*). *Acta Cryst.* **13**, 72.

RERAT, C. (1960*b*). *Acta Cryst.* **13**, 459.

ROLLETT, J. S. & SPARKS, R. A. (1960). *Acta Cryst.* **13**, 273.

SCHOMAKER, V. & PAULING, L. (1939). *J. Amer. Chem. Soc.* **61**, 1769.

Acta Cryst. (1962). **15**, 433

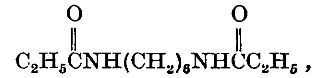
Refinement of the Structure of N,N'-Hexamethylenebispropionamide

BY L. H. JENSEN

Department of Anatomy, University of Washington, Seattle, Washington, U.S.A.

(*Received* 18 *November* 1960 *and in revised form* 11 *August* 1961)

The structure of N,N'-hexamethylenebispropionamide,



$$C_2H_5\overset{O}{\overset{\|}{C}}NH(CH_2)_6NH\overset{O}{\overset{\|}{C}}C_2H_5\,,$$

has been refined by three-dimensional ΔF syntheses. The following average bond lengths and angles with the indicated standard deviations were determined:

$$\text{C–C, } 1{\cdot}529\pm0{\cdot}002\text{ Å; } \angle\text{ C–C–C, } 112{\cdot}8\pm0{\cdot}7°;\ \angle\text{ H–C–H, } 108{\cdot}9\pm1{\cdot}2°.$$

Electron densities at the H atom sites range from 0·32–0·42 e.Å^{-3}, average 0·38 e.Å^{-3}.

There is pronounced thermal anisotropy with the direction of maximum vibration almost perpendicular to the plane of the molecule. It was not possible to determine with certainty what part of the thermal anisotropy is due to rotary oscillation about the long axis of the molecule; it is not large but, if present and corrected for, would increase slightly the average C–C bond length and decrease the average C–C–C bond angle.

Introduction

The determination of the crystal structure of N,N'-hexamethylenebispropionamide (Fig. 1) has been reported by Jensen, Krimm, Parrish & Wood (1957). The work originally was undertaken primarily to ascertain the direction of the bonds in the peptide group. Along with polarized infra-red spectroscopic studies, this information permits the location of the transition moments with respect to the bond directions.

Refinement of the two-dimensional data was carried to the point where standard deviations in the bond lengths could be expected to lie in the range 0·01–0·02 Å. All the C–C bond lengths in the chain were appreciably shorter than the bond length in diamond, 1·544 Å, and the five C–C bonds in the chain averaged almost 0·03 Å less than this value. The present work was carried out to determine more accurate bond lengths and to study the electron density in the crystal in greater detail, particularly the electronic anisotropy due to thermal motion and the electron density due to hydrogen atoms.

Experimental

The unit-cell parameters used were the same as reported earlier:

$$a=18{\cdot}60,\ b=4{\cdot}96,\ c=7{\cdot}49\text{ Å};\ \beta=97°\ 15'$$
$$(\text{based on Cu } K\alpha,\ \lambda=1{\cdot}5418\text{ Å})\,.$$

The space group is $P2_1/a$ with two molecules per unit cell.

An ordinary histological microtome fitted with a razor blade was used to cut a cylindrical crystal about 0·15 mm. in diameter, cylinder axis coincident with b. Zero and upper level unidimensionally integrated

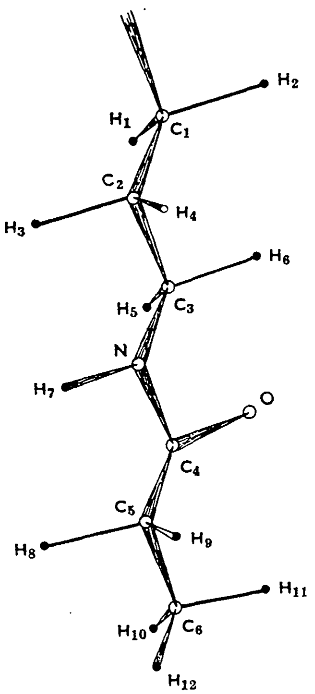


Fig. 1. Numbering of the atoms in N,N'-hexamethylenebispropionamide. The half molecule shown is the asymmetric unit in the crystal structure.

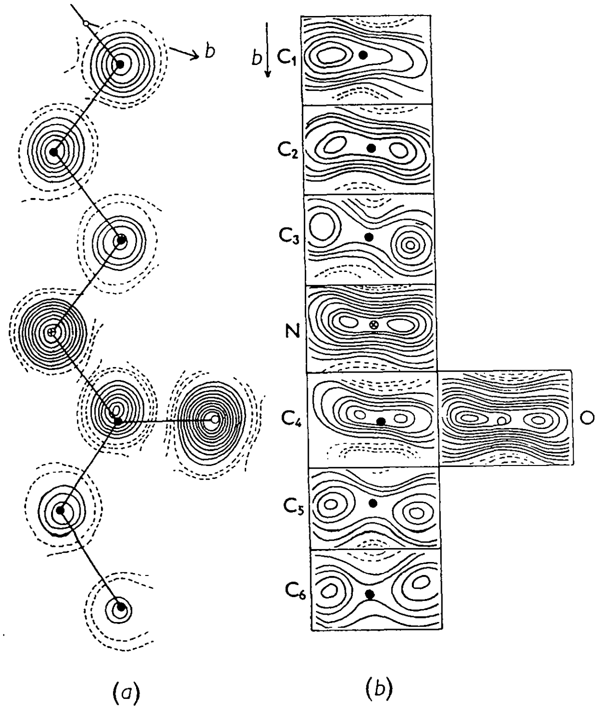


Fig. 2. Difference synthesis showing electronic anisotropy. Differences are from electron densities corresponding to B values of 4·0 for C_1, C_2, C_3, C_4, N; 5·5 for C_5 and 6·0 for C_6 and O. Contours at 0·1 e.Å^{-3}, zero contour omitted, negative contours broken. (*a*) Composite map of sections close to the plane of the molecule. (*b*) Sections essentially perpendicular to the plane of the molecule.

Weissenberg data were collected on a Wiebenga-type (Wiebenga & Smits, 1950) integrating camera. Intensities were determined with a recording microdensitometer (Jensen, 1954) and were measured on both top and bottom halves of the film. For all but the fourth level, the measurements were done by different technicians. Comparison of the two sets of results indicated random errors of less than 2% in the structure factors.

A total of 1341 reflections with $0 \leq k \leq 4$ were accessible with the camera used. Of these, 851 (63·5%) were observed.

Refinement of the structure

Calculation of the observed structure factors with the coordinates and scale factor from the two-dimensional work and isotropic temperature factors with $B=4{\cdot}0$ Å^2 for the C_1–C_4 and N atoms; 5·5 Å^2 for the C_5 atom and 6·0 Å^2 for the C_6 and O atoms resulted in a reliability index R of 19·9%. This rather high value of R does not reflect large errors in the positional parameters, but is the result of pronounced electronic anisotropy as is apparent from the ΔF synthesis shown in Figs. 2(*a*), (*b*). The deviation of the electron density from that assumed for spherical atoms is sufficient to mask the atomic shifts for most of the atoms. The direction of maximum thermal motion is in a plane essentially at right angles to the plane of

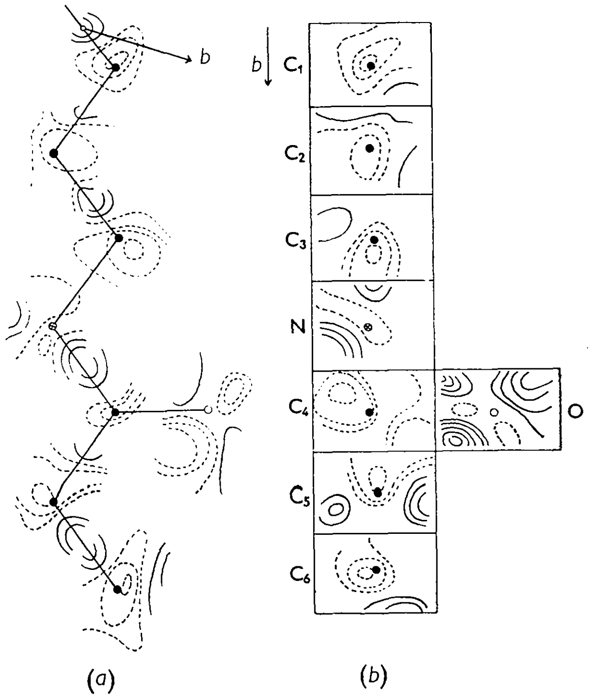


Fig. 3. Difference synthesis from which final parameter changes were derived. Contours at 0·05 e.Å^{-3}, zero contour omitted, negative contours broken. (*a*) Composite map of sections close to the plane of the molecule. (*b*) Sections essentially perpendicular to the plane of the molecule.

the molecule and, except for C_6, makes an angle of approximately 5° with (010).

Three refinement cycles by ΔF syntheses (Cochran, 1951), based on structure factors calculated with individual atom anisotropic temperature factors, reduced R to 9·5%. The ΔF synthesis at this stage showed a ridge of positive electron density running down the backbone of the molecule, and it was clear that errors in the temperature-factor parameters alone could not account for it. For these three cycles the

Table 1. *Observed and calculated structure factor*

h	$10\|F_o\|$	$10F_c$
	h00	
2	173	170
4	316	-314
6	94	78
8	128	140
10	<14	-3
12	121	-115
14	80	80
16	91	91
18	<19	8
20	24	15
22	<16	1
	h01	
-22	<17	-4
-20	28	-21
-18	<19	11
-16	<19	2
-14	<18	-9
-12	173	166
-10	192	172
-8	245	-235
-6	47	43
-4	61	48
-2	225	218
0	470	-466
2	94	74
4	1289	1459
6	118	110
8	175	-179
10	22	6
12	54	64
14	<18	5
16	44	-47
18	39	42
20	35	42
22	<15	7
	h02	
-22	21	-18
-20	30	30
-18	23	-18
-16	29	-25
-14	39	37
-12	20	-23
-10	35	-44
-8	361	336
-6	374	352
-4	287	-271
-2	49	3
0	136	143
2	206	200
4	384	-390
6	<11	-15
8	519	523
10	69	71
12	44	-46
14	<19	-3
16	<19	16
18	<18	4
20	<15	-17
22	17	16
	h03	
-22	<15	-2
-20	44	-50
-18	49	-54
-16	59	57
-14	54	-49
-12	40	-26
-10	66	66
-8	61	-49
-6	54	-69
-4	323	304
-2	300	283
0	216	-210
2	80	-89
4	156	164
6	116	111
8	177	-175
10	<17	-6
12	193	193
14	36	37
16	<19	-8
18	<17	-2
20	<16	7
	h04	
-22	<13	13
-20	<15	8
-18	<18	1
-16	97	-104
-14	88	-98
-12	93	89
-10	95	-94
-8	<16	-10
-6	80	89
-4	83	-76
-2	71	-77
0	95	94
2	122	121
4	104	-108
6	58	-61
8	67	69
10	47	49
12	60	-65
14	<19	-2
16	60	59
18	14	15
20	<11	-3
	h05	
-20	34	-30
-18	27	26
-16	<18	10
-14	22	16
-12	160	-160
-10	136	-139
-8	91	92
-6	106	-117
-4	23	14
-2	67	76
0	39	-39
2	51	-52
4	20	21
6	43	45
8	44	-44
10	35	-34
12	27	31
14	17	18
16	18	-21
18	<12	-1
	h06	
-20	16	11
-18	<16	0
-16	51	-42
-14	44	35
-12	<19	12
-10	37	36
-8	187	-184
-6	143	-142
-4	43	48
-2	72	-81
0	20	8
2	36	40
4	<19	-12
6	24	-21
8	<19	-10
10	<18	10
12	20	-21
14	<14	-16
16	<12	11
	h07	
-18	69	-69
-16	24	15
-14	<19	-5
-12	47	-42
-10	31	26
-8	23	21
-6	39	40
-4	131	-138
-2	101	-95
0	<19	13
2	40	-44
4	<19	6
6	<18	17
8	<17	-2
10	<15	-7
12	<13	-10
	h08	
-14	81	-73
-12	<16	12
-10	<18	-3
-8	33	-31
-6	<17	4
-4	26	24
-2	31	31
0	75	-78
2	60	-53
4	<16	-3
6	20	-22
8	<15	3
10	<11	6
	h09	
-10	63	-55
-8	<14	5
-6	<15	2
-4	19	-18
-2	<16	-7
0	18	15
2	19	18
4	35	-36
	h10	
1	151	144
2	488	-525
3	518	531
4	38	-41
5	16	-13
6	133	-133
7	72	-63
8	69	76
9	44	-47
10	40	40
11	105	100
12	46	-46
13	23	-20
14	23	21
15	34	-32
16	23	-14
17	69	66
18	26	-27
19	34	34
20	<16	-17
21	<15	2
22	19	-17
23	<11	-4
	h11	
-23	<10	-2
-22	16	-16
-21	<15	6
-20	<16	-13
-19	19	-15
-18	33	29
-17	<19	0
-16	37	40
-15	73	73
-14	63	65
-13	143	140
-12	17	15
-11	91	-88
-10	22	-17
-9	69	-58
-8	134	128
-7	207	193
-6	107	-102
-5	121	-106
-4	101	-94
-3	48	-46
-2	151	128
-1	31	-26
0	48	52
1	535	561
2	538	561
3	127	123
4	18	21
5	74	73
6	235	-243
7	258	262
8	37	-39
9	17	22
10	59	-65
11	36	-30
12	23	32
13	20	-21
14	<18	10
15	41	44
16	<19	-8
17	<19	-1
18	<18	18
19	<17	-8
20	<16	-5
21	26	26
22	<13	-7
	h12	
-23	<12	-3
-22	22	27
-21	33	-35
-20	37	40
-19	<17	-3
-18	34	-30
-17	19	17
-16	24	-30
-15	42	-39
-14	39	33
-13	<18	-15
-12	77	74
-11	121	121
-10	146	136
-9	293	276
-8	<13	12
-7	173	-171
-6	54	48
-5	157	-154
-4	245	236
-3	218	216
-2	107	-104
-1	144	-133
0	50	-46
1	55	-59
2	12	1
3	20	-23
4	34	24
5	280	279
6	274	259
7	47	33
8	<14	9
9	42	46
10	102	-94
11	109	103
12	<17	-19
13	<18	8
14	23	-25
15	<17	-16
16	<17	12
17	<17	-3
18	<16	-2
19	<15	16
20	<13	2
21	<12	2
22	<10	8
	h13	
-23	<10	4
-22	<11	-1
-21	<14	3
-20	22	-20
-19	<17	-10
-18	53	55
-17	81	-77
-16	84	83
-15	<19	-4
-14	46	-39
-13	39	35
-12	50	-48
-11	75	-69
-10	18	20
-9	24	-22
-8	114	98
-7	150	139
-6	236	221
-5	377	372
-4	<12	-1
-3	198	-200
-2	125	124
-1	193	-193
0	179	171
1	122	129
2	<12	-1
3	30	-16
4	13	-24
5	59	-68
6	44	-26
7	<15	-15
8	<15	4
9	109	114
10	123	121
11	28	26
12	<18	14
13	23	16
14	28	-19
15	38	36
16	<18	-5
17	<17	-1
18	<16	-4
19	<14	-9
20	<12	3
21	<10	0
	h14	
-22	<11	3
-21	<12	-5
-20	<14	7
-19	<16	6
-18	<17	-4
-17	<18	4
-16	53	-53
-15	<19	-15
-14	88	89
-13	143	-142
-12	141	141
-11	<18	-6
-10	35	-35
-9	38	39
-8	44	-39
-7	83	-82
-6	39	-30
-5	34	-34
-4	97	92
-3	106	100
-2	150	134
-1	241	237
0	<14	-31
1	123	-114
2	50	54
3	110	-111
4	66	66
5	43	50
6	27	20
7	<17	-1
8	<17	4
9	21	-24
10	34	-28
11	<19	-2
12	<19	3
13	42	45
14	50	52
15	<17	14
16	<16	10
17	<15	4
18	<13	-3
19	<11	11
	h15	
-21	<11	4
-20	<12	0
-19	<14	0
-18	<15	-4
-17	<17	-12
-16	<17	14
-15	<18	4
-14	<19	-7
-13	<19	9
-12	88	-87
-11	<19	-16
-10	117	116
-9	196	-193
-8	171	162
-7	30	-23
-6	<17	-2
-5	46	47
-4	25	-15
-3	57	-61
-2	53	-50
-1	50	-52
0	49	56
1	43	46
2	51	44
3	112	109
4	<17	-14
5	50	-49
6	24	28
7	54	-56
8	29	28
9	<19	7
10	26	19
11	<19	-10
12	<18	12
13	<17	-8
14	<16	-17
15	<15	1
16	<14	0
17	<12	15
18	<11	16
	h16	
-19	38	-27
-18	<13	-5
-17	<14	11
-16	<15	4
-15	<17	0
-14	21	-12
-13	<18	-17
-12	35	23
-11	<19	-1
-10	<19	-16
-9	<19	7
-8	100	-107
-7	<19	-16
-6	87	94
-5	166	-164
-4	108	107
-3	32	-29
-2	25	21
-1	41	43
0	<18	-2
1	25	-28
2	29	-30
3	34	-33
4	20	23
5	28	17
6	28	18
7	49	50
8	<18	1
9	<18	-21
10	<17	17
11	22	-26
12	<16	9
13	<15	-2
14	<13	13
15	<11	-4
16	<10	7
	h17	
-17	<12	-8
-16	30	27
-15	42	-35
-14	<15	-1
-13	23	16
-12	<16	6
-11	<17	3
-10	21	-16
-9	22	-20
-8	30	30
-7	<18	-10
-6	33	-29
-5	<19	-1
-4	85	-90
-3	<19	-13
-2	44	49
-1	99	-99
0	51	52
1	19	-24
2	20	19
3	21	19
4	<18	5
5	<18	-9
6	<17	-13
7	<17	-14
8	<16	7
9	<15	6
10	<14	4
11	<13	17
12	<11	2
13	<11	-8
	h18	
-14	<12	4
-13	<11	-6
-12	20	17
-11	38	-32
-10	<15	3
-9	21	16
-8	<16	1
-7	<16	7
-6	<16	-10
-5	<16	-15
-4	19	27
-3	<17	-13
-2	21	-26
-1	<17	-2
0	51	-51
1	<16	-8
2	21	21
3	52	-49
4	20	21
5	19	-16
6	<14	11
7	<13	6
8	<11	4
9	<12	-2
10	<9	-4
	h19	
-10	<9	-2
-9	<11	-3
-8	<12	6
-7	<10	-21
-6	<11	2
-5	<11	11
-4	<11	-5
-3	<11	8
-2	<11	-3
-1	<11	-7
0	<11	18
1	<11	-9
2	<12	-16
3	<11	-2
4	<10	-23
5	<7	-4
	h20	
0	104	-77
1	84	-88
2	<6	-8
3	189	-176
4	100	90
5	74	70
6	<16	20
7	88	90
8	25	30
9	65	-75
10	47	-48
11	66	-58
12	41	-39
13	21	-20
14	28	29
15	43	42
16	23	-22
17	<17	-7
18	19	18
19	36	-34
20	<14	18
21	<12	-12
22	<11	9
	h21	
-22	23	-24
-21	<12	13
-20	21	19
-19	<15	11
-18	<16	18
-17	22	22
-16	47	51
-15	79	80
-14	37	39
-13	34	29
-12	39	-42
-11	81	-74
-10	73	68
-9	14	15
-8	114	-112
-7	149	144
-6	111	-103
-5	156	154
-4	9	-15
-3	60	-49
-2	23	-10
-1	110	-110
0	233	245
1	195	200
2	18	7
3	105	96
4	54	-53
5	18	-14
6	17	13
7	141	-155
8	45	35
9	<14	8
10	19	15
11	48	46
12	19	17
13	<16	-17
14	22	-23
15	32	-32
16	<17	-10
17	18	-16
18	<15	10
19	21	19
20	<13	-7
21	<11	1
	h22	
-22	<11	3
-21	<12	3
-20	26	-32
-19	27	27
-18	50	-55
-17	28	28
-16	38	35
-15	21	27
-14	22	27
-13	27	27
-12	101	102
-11	120	118
-10	46	48
-9	80	71
-8	69	-67
-7	110	-102
-6	121	118
-5	50	42
-4	249	-243
-3	220	216
-2	148	-144
-1	229	218
0	45	-42
1	<10	12
2	<14	9
3	29	-33
4	213	225
5	128	145
6	31	25
7	65	60
8	28	-24
9	16	15
10	<15	7
11	61	-64
12	<18	8
13	<17	-2
14	<17	2
15	20	22
16	<17	-1
17	<16	7
18	<15	-9
19	<13	-13
20	<11	2
21	<10	-8

Table 1 (*cont.*)

h	$10\lvert F_o\rvert$	$10F_c$
	h23	
-22	8	10
-21	12	11
-20	<13	4
-19	<14	4
-18	<15	15
-17	<16	2
-16	52	-57
-15	42	43
-14	94	-98
-13	47	46
-12	41	37
-11	42	48
-10	20	14
-9	24	22
-8	168	167
-7	132	126
-6	53	50
-5	107	103
-4	60	-63
-3	98	-98
-2	112	124
-1	40	32
0	207	-215
1	141	135
2	93	-99
3	138	150
4	46	-47
5	<13	14
6	<14	10
7	<15	0
8	114	115
9	51	52
10	21	18
11	24	26
12	<17	-5
13	20	13
14	<17	-1
15	<16	-15
16	<15	-4
17	<14	-4
18	<13	0
19	<11	9
20	<10	-4
	h24	
-21	<10	10
-20	<11	10
-19	<13	-6
-18	<14	9
-17	21	17
-16	<16	7
-15	<17	8
-14	36	34
-13	<17	3
-12	77	-81
-11	56	61
-10	132	-134
-9	66	63
-8	24	16
-7	61	65
-6	<15	1
-5	19	23
-4	175	182
-3	113	117
-2	39	36
-1	80	87
0	17	-23
1	62	-63
2	61	71
3	22	18
4	103	-107
5	71	71
6	44	-52
7	59	69
8	41	-42
9	<17	19
10	<17	-1
11	<17	10
12	50	53
13	<17	17
14	<16	11
15	<15	8
16	<14	1
17	<12	8
18	<10	-2
19	<8	-3
	h25	
-20	<11	0
-19	21	17
-18	<14	9
-17	<14	15
-16	<15	11
-15	<16	-16
-14	<16	2
-13	22	17
-12	20	11
-11	<17	8
-10	45	46
-9	<17	5
-8	75	-79
-7	56	61
-6	117	-119
-5	54	54
-4	<16	-16
-3	37	46
-2	<16	2
-1	21	19
0	107	117
1	76	71
2	24	13
3	39	46
4	<16	-1
5	33	-34
6	26	30
7	23	17
8	48	-49
9	39	34
10	23	-23
11	20	27
12	25	-24
13	<15	11
14	<14	-2
15	<12	9
16	14	20
17	<9	5
	h26	
-18	<11	0
-17	<11	-1
-16	<13	-5
-15	20	19
-14	22	12
-13	25	16
-12	<17	8
-11	26	-22
-10	<17	-13
-9	<17	12
-8	20	16
-7	<17	2
-6	35	37
-5	<17	-1
-4	46	-50
-3	50	51
-2	73	-70
-1	30	30
0	27	-28
1	<17	21
2	<17	-2
3	<17	11
4	54	58
5	42	40
6	<17	6
7	<17	16
8	<16	5
9	<16	-15
10	<15	9
11	<14	9
12	15	-16
13	20	13
14	<12	-8
15	<8	8
	h27	
-16	<10	-12
-15	28	-27
-14	<12	11
-13	<13	-8
-12	<14	-10
-11	<14	12
-10	26	17
-9	<15	11
-8	<16	0
-7	23	-23
-6	22	-26
-5	<16	5
-4	<17	13
-3	<17	-7
-2	18	21
-1	<17	-2
0	21	-23
1	34	35
2	35	-32
3	<16	16
4	<16	-18
5	<15	3
6	<15	0
7	<14	6
8	19	22
9	19	21
10	<11	3
11	<12	4
12	<9	3
	h28	
-13	<10	6
-12	<12	-6
-11	26	-27
-10	16	19
-9	<13	-12
-8	<14	-8
-7	<14	3
-6	17	17
-5	<15	3
-4	<14	-5
-3	<14	-16
-2	19	-23
-1	<14	2
0	<14	6
1	<13	-9
2	<13	9
3	<13	-2
4	<12	-7
5	<17	16
6	<10	-11
7	<11	7
8	<9	-7
	h30	
1	31	33
2	158	145
3	42	-30
4	8	-1
5	40	-50
6	79	-82
7	21	18
8	20	-19
9	116	-123
10	26	-21
11	84	-90
12	16	9
13	36	38
14	36	-39
15	36	37
16	<15	-5
17	15	-19
18	19	27
19	<12	-16
20	<10	-11
	h31	
-20	25	20
-19	<12	11
-18	25	29
-17	46	43
-16	41	38
-15	31	-32
-14	57	-61
-13	34	-37
-12	<16	2
-11	54	58
-10	56	57
-9	67	73
	h31	
-8	<12	1
-7	131	-140
-6	58	63
-5	183	-187
-4	18	20
-3	16	18
-2	151	148
-1	49	-65
0	38	36
1	31	-25
2	160	-148
3	42	43
4	<10	-7
5	12	16
6	74	74
7	37	-34
8	17	22
9	20	-25
10	19	-20
11	<16	7
12	16	-11
13	62	-70
14	<16	-2
15	49	-52
16	<15	9
17	15	17
18	18	-20
19	14	18
20	<10	-3
	h32	
-20	<12	-2
-19	12	-14
-18	25	29
-17	28	-34
-16	31	32
-15	28	25
-14	59	60
-13	89	84
-12	84	85
-11	52	-53
-10	98	-97
-9	54	-54
-8	<14	-4
-7	66	69
-6	57	51
-5	95	103
-4	15	15
-3	139	-146
-2	96	96
-1	183	-177
0	18	12
1	29	23
2	178	173
3	13	-27
4	50	52
5	14	-15
6	82	-87
7	34	36
8	16	-11
9	<15	8
10	20	20
11	20	-19
12	17	19
13	19	-18
14	<16	3
15	<15	2
16	<14	-6
17	28	-32
18	<11	3
19	21	-24
	h33	
-20	<10	-12
-19	<11	8
-18	<13	4
-17	<14	13
-16	<14	-5
-15	17	-21
-14	48	53
-13	58	-61
-12	28	34
-11	42	41
-10	92	89
-9	128	119
-8	123	120
-7	60	-61
-6	111	-101
-5	66	-68
-4	<13	1
-3	62	66
-2	12	8
-1	88	82
0	15	18
1	92	-95
2	89	99
3	124	-119
4	30	18
5	20	12
6	113	125
7	<15	-22
8	35	38
9	<16	-3
10	26	-33
11	19	18
12	<16	-7
13	<16	6
14	<15	2
15	<14	-9
16	<13	10
17	<11	-7
18	<11	5
	h34	
-19	<11	10
-18	<11	0
-17	16	-19
-16	17	-16
-15	18	13
-14	<15	-5
-13	20	24
-12	<16	-6
-11	22	-26
-10	57	61
-9	72	-73
	h34	
-8	22	20
-7	49	49
-6	103	100
-5	153	145
-4	119	120
-3	38	-45
-2	69	-65
-1	57	-58
0	<14	13
1	36	40
2	15	-24
3	55	55
4	<15	10
5	53	-59
6	61	65
7	71	-66
8	28	18
9	<16	-1
10	53	57
11	<16	-13
12	15	18
13	<13	-1
14	<12	-6
15	<11	7
16	<9	-4
17	<7	3
	h35	
-18	12	10
-17	41	30
-16	34	31
-15	19	14
-14	<14	8
-13	29	-31
-12	20	-13
-11	19	17
-10	19	-18
-9	26	31
-8	<16	-5
-7	<16	-21
-6	39	50
-5	60	-60
-4	<16	10
-3	45	46
-2	86	85
-1	120	120
0	78	83
1	<16	-18
2	28	-27
3	36	-36
4	<16	15
5	16	17
6	17	-21
7	31	33
8	<16	3
9	26	-27
10	27	27
11	31	-27
12	16	11
13	<12	-1
14	24	21
15	<9	-2
	h36	
-16	21	-19
-15	26	24
-14	<12	4
-13	43	40
-12	28	22
-11	21	19
-10	19	18
-9	37	-38
-8	<15	-4
-7	15	14
-6	21	-23
-5	23	28
-4	<16	-5
-3	<16	-13
-2	<16	30
-1	44	-45
0	<15	8
1	23	28
2	51	49
3	71	67
4	33	35
5	<15	-3
6	<15	-5
7	20	-21
8	<14	10
9	<13	7
10	<12	-10
11	15	14
12	<11	0
	h37	
-13	24	25
-12	31	-27
-11	26	28
-10	<11	-4
-9	45	43
-8	<12	6
-7	22	20
-6	16	20
-5	30	-32
-4	<14	2
-3	<14	6
-2	<14	-16
-1	18	22
0	<14	-3
1	<14	-5
2	<14	9
3	24	-25
4	<13	4
5	<12	13
6	22	20
7	31	34
8	<12	8
9	<10	3
	h38	
-9	18	21
-8	23	-24
-7	24	23
-6	19	-9
	h38	
-5	39	33
-4	<10	-7
-3	11	15
-2	<10	12
-1	16	-20
0	<10	3
1	<12	0
2	<11	-6
3	<10	12
4	<9	-2
	h40	
0	42	-56
1	<5	16
2	9	-10
3	36	-45
4	69	70
5	11	-21
6	89	-92
7	48	-43
8	179	-185
9	32	-31
10	<14	8
11	56	56
12	<14	1
13	31	30
14	31	-31
15	<13	-9
16	<12	10
17	21	-21
18	<10	14
	h41	
-18	15	-20
-17	16	-14
-16	<12	-12
-15	<13	-11
-14	28	27
-13	38	38
-12	25	16
-11	21	18
-10	63	-59
-9	34	-33
-8	32	33
-7	83	-85
-6	34	33
-5	53	51
-4	227	-226
-3	67	62
-2	103	-109
-1	11	20
0	54	66
1	35	32
2	<9	0
3	<10	-5
4	37	-47
5	<11	20
6	13	-14
7	24	-35
8	61	61
9	<14	-16
10	60	-60
11	22	-13
12	110	-106
13	<14	-10
14	<13	-4
15	31	30
16	<11	-8
17	20	19
	h42	
-18	19	17
-17	21	19
-16	<11	5
-15	45	42
-14	26	-30
-13	24	-23
-12	26	-24
-11	30	-26
-10	47	47
-9	55	51
-8	18	10
-7	31	37
-6	91	-87
-5	32	-31
-4	59	59
-3	98	-102
-2	49	48
-1	50	44
0	176	-195
1	68	62
2	94	-97
3	18	20
4	25	33
5	31	23
6	<13	6
7	<14	-4
8	25	-32
9	<14	14
10	<14	-10
11	<14	-17
12	39	39
13	<13	-9
14	28	-30
15	<11	-4
16	49	-47
17	<8	-3
	h43	
-17	18	-16
-16	<13	-5
-15	<12	-5
-14	33	34
-13	30	29
-12	27	22
-11	78	71
-10	28	-31
-9	37	-30
-8	35	-29
-7	50	-48
-6	72	70
-5	51	46
-4	<13	-4
-3	40	53
	h43	
-2	97	-92
-1	22	-17
0	63	63
1	79	-90
2	46	43
3	37	33
4	116	-121
5	57	55
6	64	-63
7	25	19
8	<14	8
9	21	12
10	<14	6
11	<13	-2
12	16	-19
13	<12	8
14	<11	-4
15	<11	-6
16	<7	18
	h44	
-16	28	24
-15	14	15
-14	38	-30
-13	24	-18
-12	<13	-4
-11	<14	-9
-10	46	44
-9	36	30
-8	60	48
-7	91	82
-6	<15	-20
-5	41	-28
-4	33	-24
-3	60	-59
-2	76	76
-1	33	27
0	<14	-12
1	33	47
2	67	-71
3	<14	-1
4	40	46
5	45	-57
6	28	30
7	19	18
8	54	-54
9	28	31
10	32	-31
11	<12	13
12	<11	4
13	<10	3
14	<9	4
	h45	
-15	30	-29
-14	22	18
-13	19	9
-12	28	26
-11	20	22
-10	41	-39
-9	22	-15
-8	<13	-4
-7	<14	-11
-6	37	39
-5	32	21
-4	76	60
-3	80	74
-2	<14	-7
-1	<14	-16
0	15	-12
1	43	-48
2	53	58
3	<14	6
4	<14	-11
5	23	32
6	34	-41
7	<13	2
8	18	21
9	21	-26
10	16	16
11	<11	6
12	<8	-19
	h46	
-12	19	-14
-11	44	-38
-10	32	27
-9	<11	2
-8	24	23
-7	20	22
-6	33	-35
-5	<13	-7
-4	<13	-7
-3	<13	-9
-2	22	26
-1	<13	11
0	59	52
1	46	47
2	<12	1
3	<12	-5
4	<12	0
5	31	-32
6	35	35
7	<10	-3
8	<10	-6
9	<8	17
	h47	
-8	<9	-7
-7	41	-37
-6	26	28
-5	<11	-4
-4	23	17
-3	18	15
-2	23	-21
-1	<12	-1
0	<11	-10
1	<11	-5
2	<10	13
3	<10	4
4	31	32

Table 2. *Atomic parameters*

	x/a	y/b	z/c	$U_{11}\times 10^2$ (Å^2)	$U_{22}\times 10^2$ (Å^2)	$U_{33}\times 10^2$ (Å^2)	$U_{12}\times 10^2$ (Å^2)	$U_{13}\times 10^2$ (Å^2)	$U_{23}\times 10^2$ (Å^2)
O	0·1329	0·6070	0·4490	9·26	2·91	7·79	0·89	3·33	1·04
N	0·1125	0·1805	0·5323	6·89	3·17	5·86	0·42	2·39	0·42
C_1	0·0080	0·1151	0·9378	6·94	3·80	5·95	0·48	2·31	0·55
C_2	0·0551	0·0296	0·7939	6·85	3·93	5·92	0·45	2·24	0·52
C_3	0·0695	0·2652	0·6724	7·42	4·05	6·14	0·68	2·81	0·60
C_4	0·1413	0·3628	0·4314	6·37	3·17	5·73	0·43	1·74	0·54
C_5	0·1885	0·2476	0·2960	8·45	5·19	6·77	0·61	3·52	0·61
C_6	0·2087	0·5420	0·1626	9·91	6·71	8·04	0·00	3·99	0·00
*H_1	0·0437	0·8068	0·1298						
H_2	0·0364	0·2757	0·0198						
H_3	0·0268	0·8687	0·7111						
H_4	0·1070	0·9518	0·8606						
H_5	0·0177	0·3457	0·6081						
H_6	0·0993	0·4227	0·7542						
H_7	0·1209	0·9843	0·5121						
H_8	0·1586	0·0828	0·2216						
H_9	0·2386	0·1678	0·3712						
H_{10}	0·1586	0·5318	0·0872						
H_{11}	0·2386	0·6168	0·2368						
H_{12}	0·2426	0·3695	0·0654						

* H atom coordinates listed were calculated assuming tetrahedral C atoms with C–H bond lengths of 1·1 Å and a trigonal N atom with N–H bond length 1·0 Å. Final F_c were calculated with these coordinates and H atom isotropic temperature factors of $B=4{\cdot}0$ Å^2.

scale factor for $F_{\text{rel.}}$ was held to the value determined from the two-dimensional data. After some minor corrections in the data, two additional refinement cycles, in which the data for each level were scaled independently, reduced R to 7·3%. The positive electron density down the chain has been removed although a positive region remains in most of the bonds. Figs. 3(*a*), (*b*) show sections of the $\varDelta F$ synthesis from which final parameter corrections were taken.

Before the last refinement cycle, two partial cycles were carried out based on a single section through the $\varDelta F$ syntheses coincident with the 401 plane. Since all the heavier atoms lie near the plane, it should be possible with a relatively small amount of calculation to obtain corrections to the parameters having components in the plane. These two partial cycles did not, however, reduce R and have not been numbered as refinement cycles.

Table 3. *Bond lengths and angles*

C_1–C_1'	1·527 Å	C_1'–C_1–C_2	113·3°
C_1–C_2	1·532	C_1–C_2–C_3	111·5
C_2–C_3	1·525	C_2–C_3–N	111·2
C_3–N	1·459	C_3–N–C_4	120·6
N–C_4	1·333	N–C_4–C_5	115·3
C_4–C_5	1·533	C_4–C_5–C_6	113·6
C_5–C_6	1·504	N–C_4–O	122·8
C_4–O	1·230	C_5–C_4–O	121·9
N–H · · · O	2·947	N–H · · · O	176·8
*C_1–H_1	1·128	H_1–C_1–H_2	110·0
C_1–H_2	1·105	H_3–C_2–H_4	111·5
C_2–H_3	1·090	H_5–C_3–H_6	108·0
C_2–H_4	1·069	H_8–C_5–H_9	103·2
C_3–H_5	1·047	H_{10}–C_6–H_{11}	107·2
C_3–H_6	1·022	H_{10}–C_6–H_{12}	110·0
C_5–H_8	1·096	H_{11}–C_6–H_{12}	112·4
C_5–H_9	1·077		
C_6–H_{10}	1·143		
C_6–H_{11}	1·042		
C_6–H_{12}	1·102		
N–H_7	1·047		

* C–H and N–H bond lengths based on H atom positions estimated from $\varDelta F$ synthesis showing H atoms.

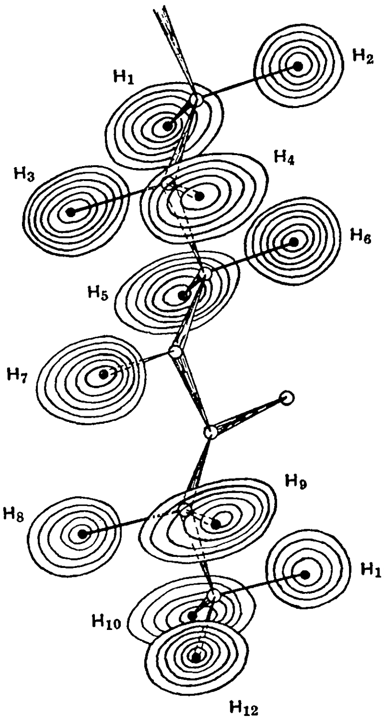


Fig. 4. Composite difference synthesis showing H atoms. Sections through H atoms and parallel to $20,\bar{2},5$ plane. Contours at positive integral values of 0·05 e.Å^{-3} beginning at 0·1 e.Å^{-3}.

The H atoms were located in a $\varDelta F$ synthesis including only terms with $\sin\theta/\lambda \leq 0{\cdot}48$. Fig. 4 is a composite of sections through the H atoms and parallel to the $20,\bar{2},5$ plane. Except for those through H_7 and H_{12}, the sections are roughly either parallel or perpendicular to the H atom–heavy-atom bonds.

All calculations in this work were done on an IBM 650 computer using programs written in these

Laboratories (Brown, Stewart, Lingafelter & Jensen, 1959). In the ΔF syntheses by which refinement was carried out, all reflections, except as noted below, were given unit weight and the coordinates shifted by the full amounts indicated. In calculating structure factors, Amsterdam (Berghuis *et al.*, 1955) scattering factors were used for carbon, nitrogen and oxygen, and McWeeny (1951) values were used for hydrogen. Observed and calculated structure factors appear in Table 1. Parameters used in the final F_c calculation are listed in Table 2 and bond lengths and angles in Table 3.

Precision of the results

Only one of the 851 observed reflections, 401, was seriously in error due to secondary extinction. Accordingly, this reflection was given zero weight in all ΔF syntheses. From the 850 remaining reflections, 5 scale, 24 positional and 35 temperature factor parameters were determined.

To evaluate the standard deviation, σ, of the coordinates, it is necessary to know the curvature, $\partial^2\varrho/\partial x_i^2$, of ϱ along each axis for each atom (Cruickshank, 1949). From the ΔF synthesis shown in Fig. 2, it is clear that for N,N'-hexamethylenebispropionamide, curvatures differ from one atom to the next for the same atom type and for any atom vary appreciably with direction.

'Average' curvatures may be calculated by assuming ϱ to obey the relation $\varrho=\varrho_0\exp[-pr^2]$ (Costain, 1941). Setting the integral of this expression over all space equal to the number of electrons in the atom gives a relation for p as a function of ϱ_0 and n (Booth, 1947). This leads directly to the following expression for the central curvature in three dimensions:

$$(d^2\varrho/dr^2)_{r=0} = -2\pi\varrho_0{}^{5/3}/n^{2/3}$$

where ϱ_0 is the peak density of, and n the number of electrons associated with the atom.

No directly measured ϱ_0 are available for N,N'-hexamethylenebispropionamide since no F_o synthesis was calculated. Nevertheless, comparison of the range of thermal parameters with those of n-nonanoic acid hydrazide (Jensen & Lingafelter, 1961) indicates that without great error $\varrho_0=7$, 8·5 and 10 e.Å^{-3} may be assumed for C, N and O atoms respectively. Curvature calculated for these ϱ_0 are given in Table 4. These values should be underestimates of the curvatures along the bond directions since maximum thermal motion is almost perpendicular to the plane of the chain.

Table 4. *Peak densities, curvatures and standard deviations*

Atom type	Peak density	Curvature	σ
O	10·0 e.Å^{-3}	−72·9 e.Å^{-5}	0·0021 Å
N	8·5	−60·9	0·0025
C	7·0	−48·7	0·0031
H	0·38	−3·0	0·051

Table 5. *Standard deviations in bond lengths*

Bond	σ
C–O	0·0037 Å
C–N	0·0038
C–C	0·0044
C–H	0·051
N–H	0·051

The curvature to be used for the H atoms was calculated in the usual way from the peaks in the ΔF synthesis, Fig. 4: ϱ_0 was taken as the average peak density and p was evaluated along the bond direction for one of the atoms, H_6, by plotting log ϱ versus r. The result also appears in Table 4.

From σ_x for each atom type, standard deviations in the bond lengths were calculated and are listed in Table 5.

Discussion

The maximum difference in any coordinate between the two- and three-dimensional results is 0·028 Å in the y coordinate for C_3. The average difference in coordinates for all atoms is 0·011 Å.

The hexamethylene chain is accurately planar. The equation for the best plane, as determined by least-squares, through the hexamethylene chain atoms is

$$0{\cdot}19276x+0{\cdot}05946y+0{\cdot}15911z=1$$

referred to a, b and c^* as coordinate axes. No one of the six atoms deviates from this plane by more than 0·0017 Å and the average deviation is 0·0011 Å.

The bond lengths and angles found for the peptide group are in remarkable agreement with the values deduced by Corey & Donohue (1950) from the structures of some amino acids and peptides available a decade ago and used by Pauling, Corey & Branson (1951) in their study of the possible helical arrangements of polypeptide chains. Except for C–N, the bond lengths are also in good agreement with those reported in the recent study of Katz & Post (1960) and bond angles are close to their values. In view of the different packing from compound to compound, somewhat larger deviations in the angles might have been expected.

The two-dimensional work gave C–C bond lengths appreciably less than the 1·544 Å C–C bonds in diamond. Brief reference was made to this difference in the report of the two-dimensional work, and a possible explanation in terms of the approximations used in treating the temperature factors was suggested to account in part for the results. From the present work, the five C–C bond lengths average almost 0·01 Å greater than those resulting from the two-dimensional work.

Although the C_4–C_5 bond is adjacent to a resonating system, there is no indication that its length is different from the three C–C bonds in the hexamethylene chain. The average length of these four bonds is 1·529 Å; and the C_5–C_6 bond is 0·025 Å less than this.

Table 6. *Data on C–H bonds*

C atom valence state	Length	Estimated mean	No. bonds averaged	Method	Reference
sp^2	1·08 Å	0·015 Å(¹)	9	*XRD*	Drenth & Wiebenga, 1955
sp^3	0·91	0·06	2	*XRD*	Marsh, 1958
sp^3	0·94	0·05(¹)	3	*XRD*	Reddy & Lipscomb, 1959
sp^3	0·94	0·01	2	*XRD*	Degeilh & Marsh, 1959
sp^3	0·94	0·08	2	*XRD*	Shintani, 1960
sp^3	1·090	0·018(¹)	14	*XRD*	Jensen & Lingafelter, 1961
sp^3	1·084	0·011(¹)	11	*XRD*	This study
sp^2	1·090	0·005	8	e.d.	Bastiansen, Hedberg & Hedberg, 1957
sp^2	1·084	0·003	4	e.d.	Bartell & Bonham, 1959
sp^3	1·108	0·004	10	e.d.	Bonham & Bartell, 1959
sp^3	1·118	0·004	12	e.d.	Bonham, Bartell & Kohl, 1959
sp^3	1·118	0·006	14	e.d.	Bonham, Bartell & Kohl, 1959
sp^3	1·121	0·007	16	e.d.	Bonham, Bartell & Kohl, 1959
sp^2	1·08	0·04	4	n.d.	Bacon & Currie, 1956
sp^2	1·05	0·013(¹)	4	n.d.	Bacon & Currie, 1960*a*
sp^2	1·12 (T = 293 °K.)	0·02	5	n.d.	Bacon & Currie, 1960*b*
sp^2	1·06 (T = 120 °K.)	0·02	5	n.d.	Bacon & Currie, 1960*b*

(¹) From r.m.s. deviations from the mean.

Such a difference would be significant by the ordinary criteria. However, terminal atoms such as C_6 and O in N,N′-hexamethylenebispropionamide, unless restricted by packing or hydrogen bonds, frequently have relatively large motions at right angles to the bonds, with a resultant apparent decrease in bond length (Cruickshank, 1956). The short C_5–C_6 bond is probably to be attributed largely to this cause.

If the thermal motion at right angles to the plane of the chain were due entirely to angular oscillation about a longitudinal axis, bonds in the chain would be short by 0·03 Å or more. It is doubtful whether systematic errors of this magnitude are in fact present since the average C–C bond length excepting C_5–C_6 does not differ appreciably from that found in other hydrocarbon chains. This implies that the excess thermal motion perpendicular to the chain is due to linear rather than rotary oscillation. It seems likely, therefore, that corrections due to angular oscillation are large in N,N′-hexamethylenebispropionamide only for the terminal C_5–C_6 and C_4–O bonds. To give a definite answer will require low-temperature data.

It is evident from Fig. 4 that for H atoms the spread in electron density in a direction at right angles to the bond and the chain axis is considerably greater than in the direction of the bond. Presumably this effect is due to thermal motion involving bending of the C–H bonds since the molecule does not appear to be undergoing appreciable rotary oscillation. The peak densities of the H atoms range from 0·32–0·42 e.Å^{-3}, average 0·38 e.Å^{-3}.

C-H bond lengths

Highly accurate values for C–H bond lengths are now available not only from spectroscopic measurements but also from electron diffraction (Bartell, 1960). Although the precision of the X-ray results is much less, they are nevertheless important. Since X-rays are scattered almost wholly by the planetary electrons, the C–H bond lengths from X-ray diffraction will not be identical with results from techniques which measure internuclear distances. If X-ray results of sufficient accuracy can be obtained, they will be useful in theoretical studies of electron distributions in hydrogen-containing molecules. Moreover, the accuracy of C–H bond lengths may serve as a sensitive indicator of errors in the data or refinement techniques.

The positions of H atoms in hexamethylenebispropionamide were determined from a ΔF synthesis based on structure factors omitting H-atom contributions. The eleven C–H bond lengths range from 1·022–1·144 Å, average 1·084 Å. The value of σ_{mean} is 0·011 Å as calculated from the r.m.s. deviations of the individual bond lengths from the mean, and 0·015 Å as calculated from $\Sigma\Delta F^2$.

The average C–H bond length from this study is listed in Table 6 along with other recent values found in compounds with no atoms heavier than oxygen. Only results which are averages of two or more bond lengths are included, and those from X-ray diffraction are all based on partial or complete three-dimensional data.* Standard deviations in the average values are those of the authors if given or are estimates from other data as indicated.

The results for electron and neutron diffraction indicate no serious differences, either within or between the groups, and are in essential agreement with spectroscopic values (see, for example, Herzberg &

---

* Tomiie (1958) has tabulated a number of additional results from two- and three-dimensional X-ray diffraction data.

Stoicheff, 1955). The results from X-ray diffraction, on the other hand, show no such agreement. Comparison of the estimated σ_{mean} with the differences between different determinations indicates the probable operation of systematic errors, which are more likely to be in the refinement than in the data.

Tomiie (1958) has estimated a C–H bond length of 0·92 Å for a B value of 2·2 and a reasonable value of another parameter, λ. Over the range of these parameters likely to be met with in practice, this study indicates a shortening of 0·1–0·3 Å. Furthermore, Cochran (1956) has evaluated the electron density in the benzene ring, but his study indicates a shortening of only 0·03 Å.

From the variable nature of both the experimental and theoretical results for the C–H bond length, it is difficult to be definite about the value or range of values to be expected from X-ray diffraction. But elimination of systematic errors in the refinement technique coupled with diffraction data taken at or below the temperature of liquid air (Cruickshank, 1960) should result in C–H bond lengths of some significance.

From data collected at liquid air temperature, we may expect central densities of H atoms to increase by a factor of about 2 over values obtained from diffraction data collected at room temperature. Since curvature of electron density varies approximately as the 5/3 power of the central density, standard deviations in the mean C–H bond lengths in the range 0·003–0·005 Å should be obtained when averaging over ten or more bonds in molecules such as N,N′-hexamethylenebispropionamide.

The author wishes to express his thanks to Miss Kate Metheny and Miss Lassia Radomsky who measured most of the intensities, and to acknowledge financial support from the Fund for Biological and Medical Research of the University of Washington and from the National Institutes of Health under Grant A-858.

References

Bacon, G. E. & Curry, N. A. (1955). *Proc. Roy. Soc.* A, **235**, 552.

Bacon, G. E. & Curry, N. A. (1960*a*. *Acta Cryst.*) **13**, 10.

Bacon, G. E. & Curry, N. A. (1960*b*). *Acta Cryst.* **13**, 717.

Bartell, L. S. (1960). Private Communication.

Bartell, L. S. & Bonham, R. A. (1959). *J. Chem. Phys.* **31**, 400.

Bastiansen, O., Hedberg, L. & Hedberg, K. (1957). *J. Chem. Phys.* **27**, 1311.

Berghuis, J., Haanappel, IJ. M., Potters, M., Loopstra, B. O., MacGillavry, C. H. & Veenendaal, A. L. (1955). *Acta Cryst.* **8**, 478.

Bonham, R. A. & Bartell, W. S. (1959). *J. Amer. Chem. Soc.* **81**, 3491.

Bonham, R. A., Bartell, L. S. & Kohl, D. A. (1959). *J. Amer. Chem. Soc.* **81**, 4765.

Booth, A. D. (1947). *Proc. Roy. Soc.* A, **188**, 77.

Brown, B. W., Lingafelter, E. C., Stewart, J. M. & Jensen, L. H. (1959). *Acta Cryst.* **12**, 350.

Cochran, W. (1951). *Acta Cryst.* **4**, 408.

Cochran, W. (1956). *Acta Cryst.* **9**, 924.

Corey, R. B. & Donohue, J. (1950). *J. Amer. Chem. Soc.* **72**, 2899.

Costain, W. (1941). Ph.D. Thesis, University of Birmingham.

Cruickshank, D. W. J. (1949). *Acta Cryst.* **2**, 65.

Cruickshank, D. W. J. (1956). *Acta Cryst.* **9**, 757.

Cruickshank, D. W. J. (1960). *Acta Cryst.* **13**, 774.

Degeilh, R. & Marsh, R. E. (1959). *Acta Cryst.* **12**, 1007.

Drenth, W. & Wiebenga, E. H. (1955). *Acta Cryst.* **8**, 755.

Herzberg, G. & Stoicheff, B. P. (1955). *Nature, Lond.* **175**, 79.

Jensen, L. H. (1954). *J. Amer. Chem. Soc.* **76**, 4663.

Jensen, L. H., Krimm, S., Parrish, R. G. & Wood, D. L. (1957). *Acta Cryst.* **10**, 528.

Jensen, L. H. & Lingafelter, E. C. (1961). *Acta Cryst.* **14**, 507.

Katz, J. L. & Post, B. (1960). *Acta Cryst.* **13**, 624.

Marsh, R. E. (1958). *Acta Cryst.* **11**, 654.

McWeeny, R. (1951). *Acta Cryst.* **4**, 513.

Pauling, L., Corey, R. B. & Branson, H. R. (1951). *Proc. Nat. Acad. Sci. Wash.* **37**, 205.

Reddy, J. & Lipscomb, W. N. (1959). *J. Chem. Phys.* **31**, 610.

Shintani, R. (1960). *Acta Cryst.* **13**, 609.

Tomiie, Y. (1958). *J. Phys. Soc. Japan*, **13**, 1030.

Wiebenga, E. H. & Smits, D. W. (1950). *Acta Cryst.* **3**, 265.