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The Crystal Structure of 3,5-Dinitro-4-methylbenzoic Acid

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3,5-Dinitro-4-methylbenzoic acid, $C_6H_2.CH_3.(NO_2)_2.COOH$, crystallizes in space group $P2_1/a$ with
four molecules per unit cell. The structure comprises hydrogen-bonded dimers with the nitro groups
twisted out of the ring plane by $47.5°$ and $38.9°$ respectively.

1. Introduction

3,5-Dinitro-4-methylbenzoic acid (Fig. 1) was synthe-
sized during studies of Friedel–Crafts reactions by
Leyshon (1952), who gives details of its preparation,
but earlier references to this substance are given by
Claus & Joachim (1891) and Wheeler & Harris (1927).
It was selected for crystallographic study in 1954, the
interest at that time being in the extent of the rotation
of the nitro-groups out of the plane of the benzene ring,
because of the methyl group between them, and also
in the hydrogen bonding. Furthermore, the rather high
density (1·62 g.cm⁻³) indicated that the structure might
well show interesting short non-bonded intermolecular
distances.

Various early attempts at solving the structure were
unsuccessful, including applications of the sign-deter-
mining procedure of Grant, Howells & Rogers (1957)
which was then being developed. Later three-dimen-
sional sign-determining approaches (Grant, Hine &
Richards, 1960) also appeared to fail, and the structure
has now been solved using the $I(\theta, \varphi)$ function of Tollin
& Cochran (1964). In retrospect, a refined version of
the three-dimensional sign-fixing method did produce
a correct set of signs, but these were not recognized

as such at the time. It is intended to publish details
of the sign-determining work later; the object of the
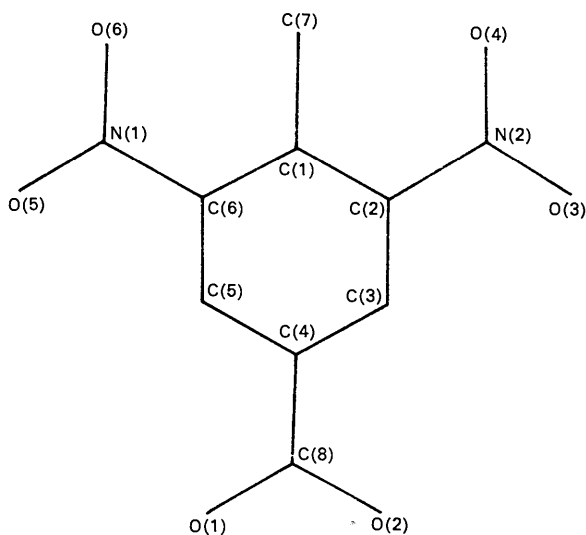present paper is to describe the structure and its solu-
tion.

Fig. 1. The molecule of 3,5-dinitro-4-methylbenzoic acid
showing the numbering scheme.

2. Crystal data and experimental details

The crystals were in the form of pale straw-coloured translucent monoclinic parallelepipeds with well developed (110) and ($\bar{1}$10) faces. The lattice data, determined from oscillation and Weissenberg photographs, are as follows:

$a = 12.34 \pm 0.03$ Å, $b = 8.94 \pm 0.02$ Å, $c = 9.01 \pm 0.02$ Å,

$$\beta = 111.0 \pm 0.2°.$$

$$d_m = 1.62 \text{ g.cm}^{-3},$$

$Z = 4$ molecules of $C_6H_2.CH_3(NO_2)_2.COOH$.

$d_c = 1.62$ g.cm^{-3}. $\mu = 13.0$ cm^{-1} for Cu $K\alpha$.

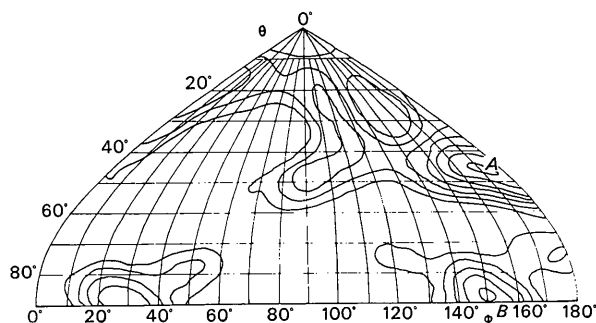Absent spectra $h0l$ for $h = 2n + 1$ and $0k0$ for $k = 2n + 1$. Space group $P2_1/a$.

Fig. 2. $I(\theta, \varphi)$ for $R = 3$ Å and sharpened data. The contours are at arbitrary equal intervals.

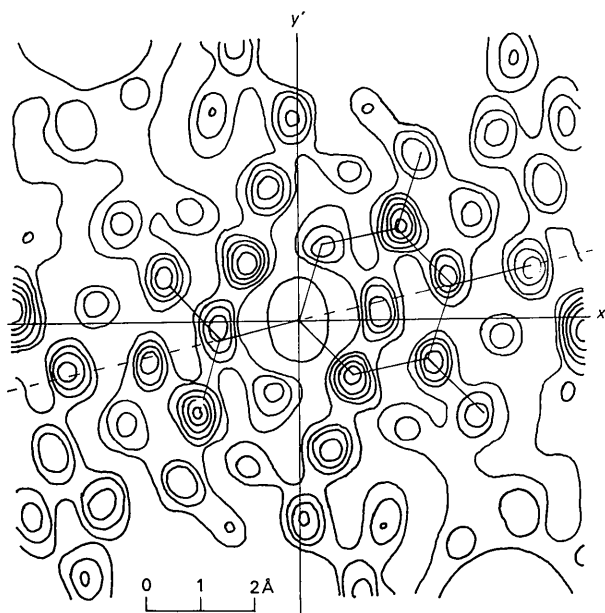Fig. 3. Section of 3-D Patterson through the origin corresponding to $\theta = 88°$, $\varphi = 148°$. The direction cosines of the normal to the plane are $l = \sin\theta\cos\varphi$, $m = \sin\theta\sin\varphi$ and $n = \cos\theta$. The axis Ox' is the intersection of the plane $lx + my + nz = 0$ with $y = 0$, and Oy' is a line in the plane and normal to Ox'. The contours are at arbitrary equal intervals.

Partial three-dimensional data ($k = 0$ to 8) were recorded on Weissenberg equi-inclination photographs from a regular crystal having no dimension greater than 0.2 mm oscillating about [010], and the intensities estimated visually by comparison with a graded step wedge. The data were corrected for Lorentz and polarization effects, and each layer placed approximately on the absolute scale by the method of Wilson (1942). No corrections for absorption were applied.

3. Structure determination

The structure was solved by determining the orientation of the part of the molecule known to be planar – i.e. the ring and the four atoms attached directly to it – by the method of Tollin & Cochran (1964). If θ, φ are the spherical-polar coordinates which define the normal to the planar group then

$$I(\theta, \varphi) = \sum_h |F(\mathbf{h})|^2 2\pi R^2 \frac{J_1(2\pi Rs)}{2\pi Rs},$$

will have a large value when the orientation of a disc of radius R coincides with the θ, φ values defining the plane containing the interatomic vectors of the planar group in the Patterson function.

3,5-Dinitro-4-methylbenzoic acid is a suitable molecule for this method as it has 10 of its 16 atoms lying in one plane. The intensity data were 'sharpened' and the 50 largest values chosen. $I(\theta, \varphi)$ was calculated for $R = 3$ Å and $0 < \theta < 90°$ in intervals of 2° and $0 < \varphi < 180°$ in intervals of $180/\theta$ degrees, and plotted in a Sanson–Flansteed projection (Fig. 2). There are two large peaks, A and B; A is disproportionately high because of the mirror plane along the $\varphi = 180°$ line, and it was expected that B would correspond to the plane containing the atoms C(1) to C(6), C(7), C(8), N(1) and N(2).

The 'sharpened' Patterson section for peak B ($\theta = 88°$, $\varphi = 148°$) was calculated to a distance of 5 Å from the origin (Fig. 3) and contains the vector set for the atoms C(1) to C(6), C(7), N(1) and N(2), and also shows that the atoms O(1) and O(2) lie very close to this plane. Three directions at 60° in the plane give a possible fit of the vector set, but the orientation shown gives the best fit. This determines ψ, the azimuthal angle, giving the orientation of the planar group within this plane. It was later found that the maximum of peak B agrees to within 1° with the position of the normal to the best plane through the 10 atoms.

Peak A (Fig. 2) can be interpreted as a double peak giving the orientation of the planes for atoms O(3), O(4), N(2), and O(5), O(6), N(1). This information was not, however, used in the completion of the solution of the structure.

The orientation of the planar group in the unit cell was now known and there remained the location of the group with respect to the origin of the unit cell. Infrared spectroscopic evidence suggests that the molecules form H-bonded dimers; these must be disposed

about centres of symmetry and, assuming an O–H$\cdots$O distance of 2·64 Å, the coordinates of atoms C(1) to C(8), O(1), O(2), N(1) and N(2) were calculated. Structure factors calculated on this basis give $R=0\cdot64$ and a three-dimensional electron density map immediately gave the positions of O(3), O(4), O(5) and O(6).

4. Refinement

After rescaling the layers, by comparison with the calculated structure factors, two cycles of block-diagonal least-squares refinement with isotropic temperature factors reduced R to 0·23. The weighting scheme adopted was:

$$\sqrt{w} = \left\{1 + \left(\frac{KF_o - 5F_{\min}}{8F_{\min}}\right)^2\right\}^{-1/2},$$

with $F_{\min} = 1\cdot4$ electrons. The scattering factors used were those given in *International Tables for X-ray Crystallography* (1962).

Two further cycles with anisotropic temperature factors reduced R to 0·14 and at this stage a three-dimensional difference map showed the hydrogens at-

tached to atoms C(3) and C(5) and the three methyl hydrogen atoms. It was not possible to locate the hydrogen atom in the hydrogen bond. One further cycle of refinement, including these hydrogen atoms in the structure factors but not refining them, reduced R to 0·13.

The final coordinates and their standard deviations are given in Table 1 and the B_{ij}'s in Table 2. The observed and calculated structure factors are given in Table 3 whilst the bond lengths and angles are given in Table 4.

A representation of the three-dimensional electron-density map projected down **a** is shown in Fig.4.

Description and discussion

The structure comprises pairs of molecules hydrogen-bonded around centres of symmetry forming dimers in a manner typical of carboxylic acids. The hydrogen bond distance 2·647 ± 0·006 Å falls in the well-established range for such bonds. The arrangement of dimers in the crystal is shown in Figs. 5(a) and (b) which are views down [001] and [010] respectively. A layer system is evident with layers parallel to (001). Apart from the

Table 1. *Atomic coordinates and standard deviations*

	x/a	$\sigma(x/a) \times 10^{-5}$	y/b	$\sigma(y/b) \times 10^{-5}$	z/c	$\sigma(z/c) \times 10^{-5}$
C(1)	0·17088	49	−0·15060	71	0·72595	64
C(2)	0·20691	48	−0·02817	74	0·66540	67
C(3)	0·16927	51	0·01045	71	0·50405	65
C(4)	0·09103	48	−0·08452	68	0·39634	68
C(5)	0·04948	51	−0·20922	73	0·44659	67
C(6)	0·09077	52	−0·24240	68	0·60926	65
C(7)	0·20499	61	−0·18555	89	0·90168	68
C(8)	0·04959	50	−0·04402	71	0·22119	66
N(1)	0·04463	55	−0·37861	67	0·65668	64
N(2)	0·28884	47	0·08112	70	0·77380	58
O(1)	−0·02693	42	−0·12959	58	0·12741	50
O(2)	0·08985	42	0·06713	58	0·17912	50
O(3)	0·26695	51	0·21597	61	0·74423	60
O(4)	0·37194	47	0·03238	81	0·88228	65
O(5)	−0·06042	48	−0·40054	69	0·59503	68
O(6)	0·11267	58	−0·46675	68	0·74457	75

Table 2. *Anisotropic temperature factors*

$$\exp\left\{-(B_{11}h^2 + B_{22}k^2 + B_{33}l^2 + B_{23}kl + B_{13}hl + B_{12}hk)\right\}.$$

	B_{11}	B_{22}	B_{33}	B_{23}	B_{13}	B_{12}
C(1)	0·00383	0·00746	0·00501	0·00034	0·00394	0·00464
C(2)	0·00300	0·00834	0·00616	−0·00398	0·00408	0·00026
C(3)	0·00443	0·00712	0·00577	−0·00113	0·00689	−0·00024
C(4)	0·00362	0·00545	0·00674	−0·00076	0·00546	0·00149
C(5)	0·00394	0·00749	0·00598	0·00025	0·00489	0·00018
C(6)	0·00497	0·00507	0·00591	0·00158	0·00605	0·00139
C(7)	0·00640	0·01369	0·00375	0·00418	0·00570	0·00293
C(8)	0·00404	0·00649	0·00578	0·00160	0·00476	0·00141
N(1)	0·00866	0·00713	0·00731	0·00115	0·00751	−0·00231
N(2)	0·00508	0·01324	0·00454	−0·00304	0·00553	−0·00470
O(1)	0·00643	0·01187	0·00545	−0·00056	0·00355	−0·00625
O(2)	0·00718	0·01136	0·00584	0·00499	0·00504	−0·00600
O(3)	0·01056	0·00999	0·01049	−0·00477	0·00980	−0·01078
O(4)	0·00548	0·02309	0·01006	−0·00473	−0·00261	−0·00236
O(5)	0·00740	0·01602	0·01502	0·00715	0·01180	−0·00477
O(6)	0·01235	0·00940	0·01628	−0·01380	0·00231	0·00083

hydrogen bond, already mentioned, the only other intermolecular forces appear to be van der Waals in character. There are several short non-bonded inter-molecular distances (see Table 5). Of these, the N(1)–O(5) separation would be short enough to warrant consideration as a weak hydrogen bond had not the

Table 3. *Observed and calculated structure factors*

Table 3 (*cont.*)

H	K	L	Fo	Fc
-14	1	1	5.7	4.7
-13	1	1	5.7	5.8
-11	1	1	16.3	-13.3
-10	1	1	6.7	-5.0
-9	1	1	9.6	-7.8
-8	1	1	23.0	19.3
-7	1	1	31.6	-30.5
-6	1	1	5.7	3.5
-5	1	1	14.4	10.5
-4	1	1	66.2	83.0
-2	1	1	9.6	5.8
-1	1	1	21.1	-24.3
0	1	1	12.5	-9.0
1	1	1	36.5	42.7
2	1	1	30.7	-32.9
3	1	1	15.3	-14.7
5	1	1	11.5	-9.9
6	1	1	9.6	-7.5
7	1	1	6.7	5.1
8	1	1	10.5	9.4
10	1	1	5.7	-3.1
11	1	1	4.8	4.1
12	1	1	7.7	-7.9
13	1	1	6.7	-6.3
-15	1	2	2.9	-2.7
-12	1	2	3.8	-2.4
-11	1	2	8.6	-7.5
-10	1	2	2.9	-2.0
-9	1	2	8.6	-7.2
-8	1	2	4.8	-3.9
-7	1	2	5.7	5.9
-6	1	2	22.0	17.9
-5	1	2	24.9	19.4
-4	1	2	28.8	-27.2
-3	1	2	40.3	-45.7
-2	1	2	45.1	53.9
-1	1	2	7.7	-9.7
0	1	2	27.8	28.8
1	1	2	16.3	14.3
2	1	2	22.0	20.5
3	1	2	15.3	-14.7
4	1	2	24.0	24.3
6	1	2	6.7	-5.4
9	1	2	18.2	20.0
10	1	2	6.7	-4.3
11	1	2	6.7	6.4
13	1	2	3.8	2.3
-15	1	3	11.3	13.0
-14	1	3	13.4	-13.3
-13	1	3	6.7	6.6
-12	1	3	4.8	-2.9
-11	1	3	3.8	-2.3
-10	1	3	3.8	-2.8
-9	1	3	7.7	6.2
-8	1	3	2.9	1.6
-6	1	3	27.8	24.7
-5	1	3	2.9	1.5
-4	1	3	35.5	-38.5
-2	1	3	25.9	23.5
-1	1	3	48.0	-56.3
0	1	3	25.9	-25.1
1	1	3	25.9	-24.7
2	1	3	47.0	49.8
3	1	3	18.2	15.5
4	1	3	6.7	-3.3
5	1	3	8.6	8.0
6	1	3	18.2	18.2
9	1	3	18.2	22.4
10	1	3	7.7	-6.2
12	1	3	5.7	5.7
-14	1	4	2.9	2.0
-13	1	4	4.8	-2.9
-12	1	4	13.4	14.6
-11	1	4	11.5	-9.7
-9	1	4	24.9	-22.3
-8	1	4	5.8	-2.5
-7	1	4	27.8	25.3
-6	1	4	33.6	-33.0
-4	1	4	12.5	-9.5
-3	1	4	13.4	-9.7
-2	1	4	3.8	1.5
-1	1	4	18.2	-15.4
0	1	4	25.9	-21.5
1	1	4	24.0	20.1
2	1	4	4.8	3.7
3	1	4	6.7	5.8
4	1	4	13.4	-11.5
5	1	4	5.7	-3.8
6	1	4	5.7	4.8
7	1	4	8.6	8.5
8	1	4	13.4	14.2
9	1	4	16.3	-21.0
10	1	4	8.6	3.8
11	1	4	2.9	-2.5
-14	1	5	4.8	-4.6
-13	1	5	5.7	-6.0
-12	1	5	11.5	-12.1
-11	1	5	10.5	9.2
-10	1	5	10.5	-9.4
-9	1	5	21.1	19.6
-8	1	5	4.8	-3.5
-7	1	5	3.8	5.1
-6	1	5	21.1	-18.3
-5	1	5	47.0	-50.0
-3	1	5	22.0	18.4
-1	1	5	10.3	7.0
0	1	5	21.1	19.5
1	1	5	6.7	6.2
2	1	5	6.7	-6.3
3	1	5	4.8	-3.0
4	1	5	21.1	19.5
5	1	5	24.0	-24.6
6	1	5	14.4	-13.8
8	1	5	4.8	-5.5
-14	1	6	7.7	8.2
-13	1	6	9.6	9.4
-12	1	6	4.8	4.0
-11	1	6	2.9	1.8
-10	1	6	6.7	7.8
-9	1	6	16.3	13.4
-8	1	6	18.2	-17.6
-7	1	6	5.7	-4.8
-6	1	6	17.3	15.2
-5	1	6	3.8	1.5
-4	1	6	16.3	-14.5
-3	1	6	2.9	2.1
-2	1	6	6.7	-6.6
-1	1	6	26.9	24.1
0	1	6	16.3	-14.0
1	1	6	4.8	2.2
2	1	6	31.6	-33.6
3	1	6	32.6	-33.1
4	1	6	4.8	3.8
7	1	6	4.8	-4.7
8	1	6	3.8	3.7
-10	1	7	8.6	7.7
-9	1	7	11.5	10.3
-8	1	7	13.4	-14.0
-7	1	7	7.7	5.6
-6	1	7	14.4	13.6
-5	1	7	15.3	13.7
-4	1	7	4.8	-4.6
-3	1	7	17.3	17.5
-2	1	7	4.8	4.0
-1	1	7	6.7	7.3
0	1	7	4.8	-2.5
1	1	7	9.6	-9.1
4	1	7	10.5	-8.3
6	1	7	4.8	-4.0
-14	1	8	4.8	-5.5
-13	1	8	2.9	1.9
-12	1	8	5.7	-3.3
-11	1	8	15.3	18.3
-10	1	8	7.7	7.7
-9	1	8	24.0	-26.3
-8	1	8	24.0	22.8
-7	1	8	9.6	-6.9
-6	1	8	9.6	7.7
-4	1	8	15.3	14.0
-3	1	8	4.8	-5.0
-2	1	8	10.5	-11.2
-1	1	8	6.7	7.8
0	1	8	2.9	-2.9
1	1	8	12.3	-12.3
2	1	8	4.8	2.9
3	1	8	11.5	12.1
5	1	8	9.6	10.2
6	1	8	4.8	-4.6
-13	1	9	9.6	-12.2
-12	1	9	6.7	5.3
-11	1	9	14.4	-14.4
-10	1	9	5.7	-4.5
-6	1	9	17.3	-18.2
-5	1	9	7.7	6.8
-4	1	9	5.7	6.2
-3	1	9	12.5	12.6
-2	1	9	4.8	-4.6
-1	1	9	9.6	-8.3
0	1	9	11.5	10.7
3	1	9	12.5	11.6
4	1	9	5.7	4.5
-11	1	10	5.7	-4.8
-10	1	10	3.8	4.8
-9	1	10	4.8	4.6
-8	1	10	5.7	5.0
-7	1	10	5.7	6.4
-6	1	10	7.7	8.0
-5	1	10	5.7	-4.0
-4	1	10	3.8	3.8
-3	1	10	4.8	-4.7
-0	1	10	12.5	11.3
1	1	10	13.4	11.5
2	1	10	3.8	2.9
-8	1	11	8.6	-7.8
-7	1	11	7.7	-9.7
-6	1	11	4.8	-5.0
-4	1	11	2.9	2.6
-3	1	11	9.6	-9.4
-2	1	11	2.9	2.5
1	3	0	16.1	-18.8
2	3	0	2.3	-0.7
3	3	0	10.3	9.6
4	3	0	35.6	-30.9
5	3	0	10.3	10.0
6	3	0	8.0	-8.4
7	3	0	17.2	-19.7
8	3	0	3.4	-1.8
9	3	0	5.7	-5.0
10	3	0	8.0	7.5
13	3	0	3.4	3.4
14	3	0	3.4	-4.1
-14	3	1	5.7	5.6
-13	3	1	8.0	7.5
-12	3	1	3.4	2.7
-11	3	1	4.6	-3.8
-10	3	1	8.0	7.0
-9	3	1	2.3	-2.1
-8	3	1	4.6	-3.3
-7	3	1	3.4	-3.9
-6	3	1	13.8	12.8
-5	3	1	14.9	-12.0
-4	3	1	5.7	-6.4
-3	3	1	29.9	27.3
-2	3	1	10.3	-8.2
-1	3	1	4.6	6.5
0	3	1	17.2	-19.6
1	3	1	49.4	-56.1
2	3	1	10.3	8.6
3	3	1	10.3	8.8
4	3	1	2.3	2.9
5	3	1	21.8	-21.0
6	3	1	10.3	10.4
7	3	1	3.4	-3.7
8	3	1	3.4	4.1
9	3	1	4.6	-2.7
10	3	1	2.3	2.4
11	3	1	2.3	2.8
12	3	1	10.3	-9.1
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-5	7	8	9.2	7.4
-2	7	8	10.6	-8.5
-1	7	8	7.9	4.9
0	7	8	2.6	-1.6

UNOBSERVEDS

H	K	L		Fc
-14	0	1	0.0	0.0
-12	0	3	0.0	-1.6
2	0	4	0.0	-1.3
0	0	5	0.0	-0.8
-14	0	6	0.0	1.4
-10	0	6	0.0	-1.8
-8	0	6	0.0	-1.9
2	0	6	0.0	-0.9
-14	0	7	0.0	0.5
-14	0	8	0.0	-3.9
-10	0	8	0.0	1.0
-10	0	9	0.0	0.5
-6	0	10	0.0	-0.9
-10	0	11	0.0	1.1
-8	0	11	0.0	3.1
-6	0	11	0.0	-2.6
-4	0	11	0.0	-1.7
1	1	0	0.0	21.9
10	1	0	0.0	1.4
13	1	0	0.0	1.0
14	1	0	0.0	-0.3
-12	1	1	0.0	-2.0
4	1	1	0.0	0.7
9	1	1	0.0	-2.6
14	1	1	0.0	-1.9
-14	1	2	0.0	0.5
-13	1	2	0.0	-1.8
5	1	2	0.0	0.6
7	1	2	0.0	1.0
8	1	2	0.0	0.2
12	1	2	0.0	1.4
-7	1	3	0.0	1.6
7	1	3	0.0	-0.9
8	1	3	0.0	1.0
11	1	3	0.0	-0.1
-10	1	4	0.0	-1.3
-5	1	4	0.0	-2.5
-4	1	5	0.0	2.6
-2	1	5	0.0	-1.6
7	1	5	0.0	2.9
9	1	5	0.0	-0.9
10	1	5	0.0	-0.9
-15	1	6	0.0	+1.3
5	1	6	0.0	+0.8
6	1	6	0.0	+2.2
9	1	6	0.0	+0.8
-15	1	7	0.0	-3.1
-14	1	7	0.0	-2.3
-13	1	7	0.0	-3.4
-12	1	7	0.0	+0.2
-11	1	7	0.0	-0.8
2	1	7	0.0	+0.3
3	1	7	0.0	+0.8
5	1	7	0.0	+0.7
7	1	7	0.0	-2.2

only donor group been unambiguously assigned. There is, in fact, an unusually large number, 20, of intermolecular distances under 3·5 Å, and this is in line with the rather high density. There is little evidence of strain in the neighbourhood of the carboxyl group, which is rotated out of the ring plane by 2·8°. The

Table 3 (*cont.*)

H	K	L		Fc
-5	1	8	0.0	+1.2
4	1	8	0.0	-0.1
-9	1	9	0.0	+0.6
-8	1	9	0.0	-1.2
-7	1	9	0.0	-1.6
1	1	9	0.0	-1.3
2	1	9	0.0	-1.2
-12	1	10	0.0	-6.2
-2	1	10	0.0	+1.9
-1	1	10	0.0	-1.9
-10	1	11	0.0	-2.0
-9	1	11	0.0	+0.4
-5	1	11	0.0	+0.1
1	2	0	0.0	-5.1
13	2	0	0.0	+0.2
14	2	0	0.0	+1.9
-15	2	1	0.0	+0.2
-14	2	1	0.0	+0.4
-12	2	1	0.0	-1.7
10	2	1	0.0	+1.6
14	2	1	0.0	-1.8
-9	2	3	0.0	+1.2
-3	2	3	0.0	+1.9
10	2	3	0.0	-1.6
-12	2	4	0.0	+1.2
11	2	4	0.0	-1.2
-13	2	5	0.0	-0.8
10	2	5	0.0	+1.4
-14	2	6	0.0	-0.8
-10	2	6	0.0	+0.3
9	2	6	0.0	-1.4
-15	2	7	0.0	+1.6
-12	2	7	0.0	+0.5
-7	2	7	0.0	-1.2
-14	2	8	0.0	+2.3
-13	2	8	0.0	-1.9
-12	2	8	0.0	+0.5
2	2	8	0.0	+0.2
3	2	8	0.0	-2.4
-13	2	9	0.0	+1.4
-11	2	9	0.0	-0.1
-9	2	9	0.0	+2.0
-5	2	9	0.0	+2.7
-4	2	9	0.0	+2.1
1	2	9	0.0	+0.7
-12	2	10	0.0	-0.7
-11	2	10	0.0	-0.4
-7	2	10	0.0	-1.1
-4	2	10	0.0	-0.5
-2	2	10	0.0	+1.0
2	2	10	0.0	+2.7
-9	2	11	0.0	+0.3
-8	2	11	0.0	+1.8
-2	2	11	0.0	+14.4
11	3	0	0.0	-1.0
12	3	0	0.0	+0.7
-15	3	2	0.0	-3.0
-13	3	2	0.0	+0.5
-8	3	2	0.0	-2.2
6	3	2	0.0	+0.6
7	3	2	0.0	+0.7
13	3	2	0.0	+4.5
-12	3	4	0.0	+1.0
-8	3	4	0.0	-1.4
-2	3	4	0.0	+0.2
0	3	4	0.0	-2.6
4	3	4	0.0	+0.3
7	3	4	0.0	+0.5
8	3	4	0.0	-0.5
11	3	4	0.0	-0.4
-15	3	5	0.0	+2.0
-9	3	5	0.0	+1.2
-15	3	6	0.0	-6.6
-11	3	6	0.0	+0.5
-10	3	6	0.0	+0.5
-2	3	6	0.0	-1.4
4	3	6	0.0	+1.5
6	3	6	0.0	+1.1
-11	3	7	0.0	-0.8
3	3	7	0.0	-0.8
-14	3	8	0.0	-1.1
-7	3	8	0.0	+0.1
-2	3	8	0.0	+0.7
4	3	8	0.0	-0.2
6	3	8	0.0	-5.7
-13	3	9	0.0	+2.1
-11	3	9	0.0	-1.2
-10	3	9	0.0	-1.0
0	3	9	0.0	+0.5
3	3	9	0.0	-0.5
4	3	9	0.0	-0.6
-8	3	11	0.0	-13.4
-5	3	11	0.0	-0.1
-4	3	11	0.0	+0.7
-3	3	11	0.0	+10.2
12	4	0	0.0	-1.3
13	4	0	0.0	-0.1
14	4	0	0.0	+0.4
-14	4	1	0.0	+0.8
-13	4	1	0.0	-0.7
-12	4	1	0.0	-2.2
5	4	1	0.0	-1.6
9	4	1	0.0	-0.3
12	4	1	0.0	-1.8
13	4	1	0.0	+1.8
-14	4	2	0.0	-1.1
10	4	2	0.0	+0.7
12	4	2	0.0	+0.6
-14	4	3	0.0	+1.5
8	4	3	0.0	-0.4
9	4	5	0.0	-0.2
12	4	3	0.0	-2.1
-15	4	4	0.0	+1.6
-14	4	4	0.0	+0.6
-12	4	4	0.0	+0.2
-7	4	4	0.0	+0.1
-6	4	4	0.0	-0.1
8	4	4	0.0	-1.0
11	4	4	0.0	+0.7
10	4	5	0.0	+4.0
-14	4	6	0.0	-2.2
-12	4	6	0.0	+1.2
-5	4	6	0.0	+0.5
3	4	6	0.0	-0.8
-14	4	7	0.0	+4.0
-13	4	7	0.0	-1.1
-13	4	8	0.0	-0.2
-7	4	8	0.0	+1.1
1	4	8	0.0	-0.4
2	4	8	0.0	-1.1
3	4	8	0.0	-1.2
-12	4	9	0.0	-4.2
-10	4	9	0.0	-1.0
-9	4	9	0.0	+1.0
-8	4	9	0.0	-2.4
-5	4	9	0.0	-0.0
-2	4	9	0.0	+1.5
0	4	9	0.0	-2.8
3	4	9	0.0	+1.9
-10	4	10	0.0	+0.3
12	5	0	0.0	+0.1
-13	5	1	0.0	-0.2
-12	5	1	0.0	-0.4
-14	5	2	0.0	-0.9
-7	5	2	0.0	-0.5
3	5	2	0.0	-1.8
-14	5	3	0.0	-0.3
4	5	4	0.0	-1.9
8	5	4	0.0	+0.1
9	5	4	0.0	-1.0
7	5	5	0.0	-0.6
-14	5	6	0.0	+4.8
1	5	6	0.0	+0.1
8	5	6	0.0	-0.1
-11	5	7	0.0	+0.2
-8	5	7	0.0	-0.1
-10	5	8	0.0	-0.9
-1	5	8	0.0	+0.1
2	5	8	0.0	+0.7
-5	5	9	0.0	+2.3
0	5	9	0.0	-0.5
-13	6	1	0.0	-4.4
-12	6	1	0.0	-0.7
11	6	1	0.0	-0.6
12	6	1	0.0	+0.3
-13	6	2	0.0	-1.4
-7	6	2	0.0	+0.3
8	6	3	0.0	-1.7
-11	6	5	0.0	+0.8
-13	6	6	0.0	+2.1
-5	6	6	0.0	-0.7
-2	6	6	0.0	-0.1
7	6	6	0.0	+1.6
-13	6	6	0.0	+2.1
-7	6	6	0.0	+4.2
5	6	6	0.0	+3.3
6	6	6	0.0	+0.2
7	6	6	0.0	+1.6
-7	6	8	0.0	+2.4
9	7	0	0.0	+0.6
11	7	0	0.0	-0.5
-10	7	2	0.0	-0.3
-7	7	3	0.0	-0.5
-9	7	4	0.0	+1.3
-9	7	5	0.0	+1.1
-4	7	5	0.0	+1.2
5	7	5	0.0	-0.0
-6	7	6	0.0	-1.0
3	7	6	0.0	+0.6
4	7	6	0.0	+0.1
-7	7	8	0.0	-2.1
-6	7	8	0.0	+1.5
-4	7	8	0.0	+0.3
-3	7	8	0.0	-0.4
8	8	0	0.0	+0.4
9	8	0	0.0	-0.9
-10	8	2	0.0	+0.4
5	8	4	0.0	-0.9
7	8	4	0.0	+0.5
-11	8	5	0.0	-4.6
-2	8	5	0.0	+0.4
-7	8	6	0.0	+0.5
-8	8	7	0.0	+0.0
-2	8	7	0.0	-1.3
-7	8	8	0.0	-1.8
-4	8	8	0.0	+0.4

departures of atoms C(4) and C(8) from the plane defined by O(1), O(2) and the origin (Table 6) indicate slight puckering.

Table 4. *Distances and angles with standard deviations*

Numbering scheme shown in Fig. 1.

Bond lengths	
C(1)–C(2)	1·366 ± 0·009 Å
C(2)–C(3)	1·402 ± 0·008
C(3)–C(4)	1·387 ± 0·008
C(4)–C(5)	1·370 ± 0·009
C(5)–C(6)	1·400 ± 0·008
C(6)–C(1)	1·418 ± 0·008
C(1)–C(7)	1·518 ± 0·008
C(2)–N(2)	1·491 ± 0·008
O(4)–N(2)	1·216 ± 0·008
O(3)–N(2)	1·243 ± 0·009
C(6)–N(1)	1·471 ± 0·009
O(5)–N(1)	1·229 ± 0·008
O(6)–N(1)	1·215 ± 0·009
C(4)–C(8)	1·518 ± 0·008
O(1)–C(8)	1·272 ± 0·008
O(2)–C(8)	1·230 ± 0·008
O(5)–C(7)	3·946 ± 0·009
O(6)–C(7)	2·910 ± 0·010
O(4)–C(7)	2·886 ± 0·010
O(3)–C(7)	4·032 ± 0·010
O(1)–O(2′)	2·647 ± 0·006
C(3)–H(1)	1·0
C(5)–H(2)	1·0
C(7)–H(3)	1·0
C(7)–H(4)	1·0
C(7)–H(5)	1·0
O(6)–H(4)	2·7
O(4)–H(3)	2·4
O(4)–H(5)	2·3

Angles	
C(1)–C(2)–C(3)	125·3 ± 0·5°
C(2)–C(3)–C(4)	117·6 ± 0·5
C(3)–C(4)–C(5)	121·1 ± 0·5
C(4)–C(5)–C(6)	118·7 ± 0·5
C(5)–C(6)–C(1)	123·3 ± 0·5
C(6)–C(1)–C(2)	114·1 ± 0·5
C(6)–C(1)–C(7)	120·8 ± 0·5
C(2)–C(1)–C(7)	124·9 ± 0·5
C(1)–C(2)–N(2)	120·3 ± 0·5
C(3)–C(2)–N(2)	114·4 ± 0·5
C(2)–N(2)–O(4)	118·2 ± 0·6
O(4)–N(2)–O(3)	125·1 ± 0·6
O(3)–N(2)–C(2)	116·8 ± 0·5
C(3)–C(4)–C(8)	118·2 ± 0·5
C(5)–C(4)–C(8)	120·7 ± 0·5
C(4)–C(8)–O(2)	119·4 ± 0·5
O(1)–C(8)–O(2)	124·5 ± 0·5
O(1)–C(8)–C(4)	116·1 ± 0·5
C(8)–O(1)–O(2′)	115·9 ± 0·4
C(8)–O(2)–O(1′)	119·5 ± 0·4
C(5)–C(6)–N(1)	116·5 ± 0·5
C(1)–C(6)–N(1)	120·2 ± 0·5
C(6)–N(1)–O(5)	117·1 ± 0·5
O(5)–N(1)–O(6)	124·2 ± 0·6
O(6)–N(1)–C(6)	118·5 ± 0·5
C(2)–C(3)–H(1)	122
C(4)–C(3)–H(1)	121
C(4)–C(5)–H(2)	121
C(6)–C(5)–H(2)	121
C(1)–C(7)–H(3)	106
C(1)–C(7)–H(4)	110
C(1)–C(7)–H(5)	109
H(3)–C(7)–H(4)	109
H(4)–C(7)–H(5)	115
H(3)–C(7)–H(5)	108

O(1′) and O(2′) are related by ($\bar{x}, \bar{y}, \bar{z}$) to the parent molecule.

Table 4 (cont.)

Other angles indicative of strain

$\angle$ C(3)–C(6)	C(6)–N(1)	$3.1 \pm 0.4°$
C(5)–C(2)	C(2)–N(2)	5.0 ± 0.4
C(4)–C(1)	C(1)–C(7)	3.9 ± 0.4
C(4)–C(1)	C(4)–C(8)	1.8 ± 0.4

The two nitro-groups are rotated out of the ring plane by substantial amounts (Table 6) as a result of the presence of the rather bulky methyl group between them, although the angles, $47.5°$ and $38.9°$ respectively,

Table 5. *Short non-bonded intermolecular distances*

The shortest non-bonded distances are:

$$N(1)\text{–}O(5'') = 3.065 \pm 0.008 \text{ Å}$$
$$O(3)\text{–}O(5''') = 3.074 \pm 0.009 \text{ Å}$$
$$O(3)\text{–}C(8''') = 3.053 \pm 0.009 \text{ Å}$$

Altogether, there are 20 non-bonded intermolecular distances of 3.5 Å (these do not include H atoms).

The shortest distances involving H atoms are:

$$O(6)\text{–}H(2'') = 2.7 \text{ Å}$$
$$O(2)\text{–}H(3''') = 2.7 \text{ Å}.$$

Double prime indicates transformation

$$(-x, \quad -1-y, \quad 1-z)$$

Triple prime indicates transformation

$$(\tfrac{1}{2}-x, \quad \tfrac{1}{2}+y, \quad 1-z)$$

Table 6. *Planes*

All planes are referred to orthogonal axes a, b and c^*.

1. Best plane through benzene ring [C(1)–C(6)]:

$$-0.843X + 0.530Y + 0.093Z - 0.051 = 0.$$

Distances from this plane of:

C(1)	$+0.003$ Å
C(2)	-0.003
C(3)	$+0.006$
C(4)	-0.009
C(5)	$+0.009$
C(6)	-0.007
C(7)	$+0.098$
C(8)	-0.001
N(1)	-0.005
N(2)	$+0.040$
O(1)	$+0.062$
O(2)	-0.039
O(3)	$+0.802$
O(4)	-0.673
O(5)	$+0.768$
O(6)	-0.822

2. Plane through O(1), O(2), C(8):

$$-0.818X + 0.562Y + 0.121Z - 0.087 = 0.$$

3. Plane through N(1), O(5), O(6):

$$-0.447X + 0.424Y + 0.788Z - 0.036 = 0.$$

4. Plane through N(2), O(3), O(4):

$$+0.987X + 0.120Y - 0.109Z - 0.431 = 0.$$

Fig. 4. Electron density projected along [100]. The contours are at intervals of 2 e.Å^{-3} starting at 2 e.Å^{-3}.
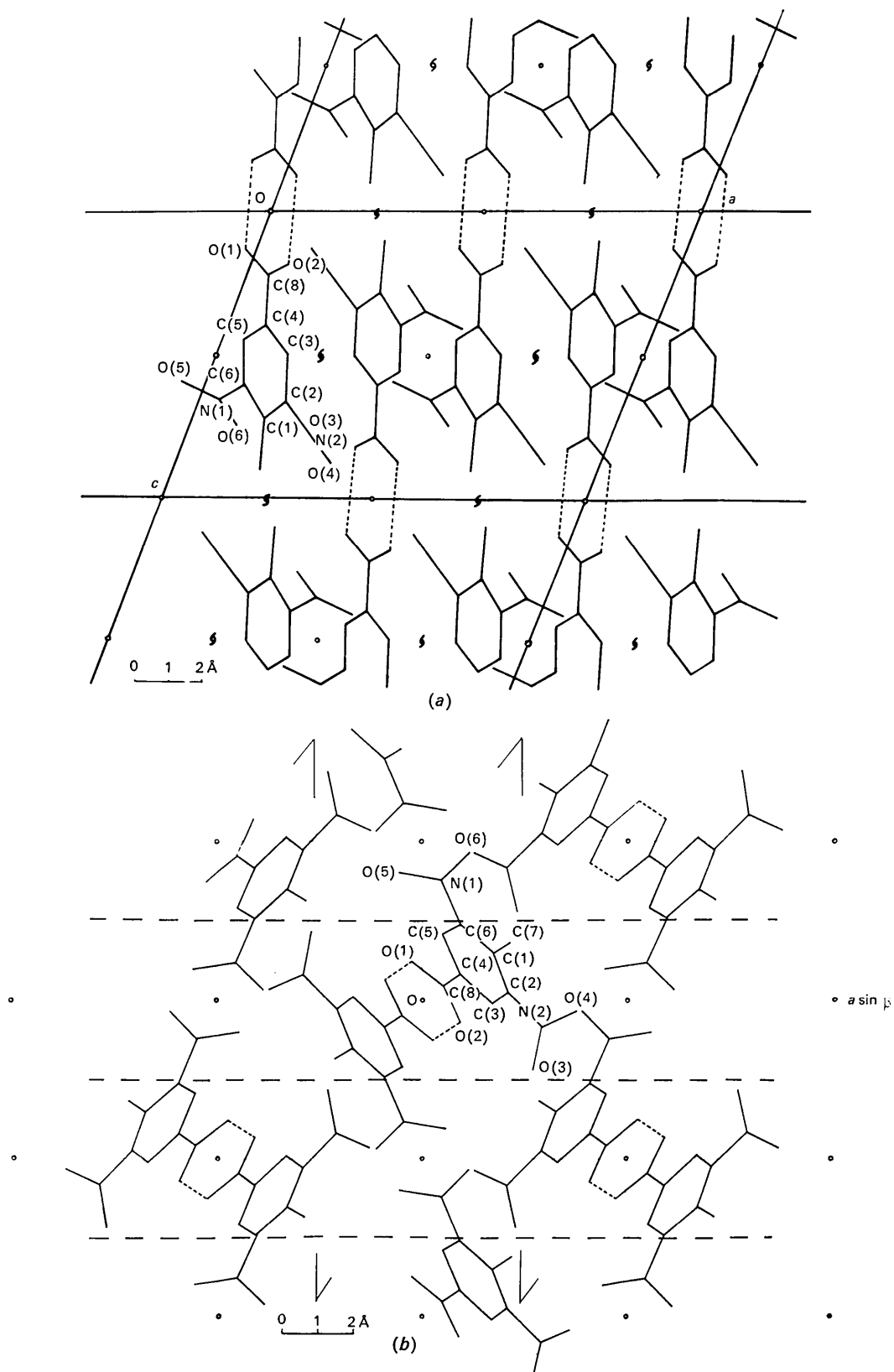
Fig. 5. The molecular packing viewed along (a) [001] and (b) [010] respectively. Broken lines indicate hydrogen bonds.

Table 6 (cont.)

5. Plane through origin, O(1), O(2):

$$+0.815X-0.576Y-0.057Z+0=0 .$$

Distances from this plane of:

C(8)+0.038 Å
C(4)+0.117 Å

6. Angles between plane normals:

1. and 2.	2.8°
1. and 3.	47.5°
1. and 4.	38.9°

are both considerably less than in nitromesitylene (Trotter, 1959) where the angle is 66.4° – also for a nitro group adjacent to a methyl group.

Also, in 4-chloro-2,6-dinitrophenol (Richards, 1960), the nitro group which is not hydrogen bonded is rotated out of the ring plane by 34.3°. In the present structure, there is evidence of overcrowding and accompanying strain in the neighbourhood of the nitro groups; all six short intermolecular contacts listed in Table 5 involve atoms in this part of the structure, and the bonds C(6)–N(1) and C(2)–N(2) have apparently been urged away from the methyl group as evidenced by the angles, 3.1° and 5.0° respectively, that these bonds make with their geometrically ideal directions defined by appropriate pairs of opposite atoms across the ring (see last part of Table 4).

The various bonds and angles within the molecule (Table 4) all appear reasonable and do not call for comment. The only chemically equivalent bonds that differ significantly from one another are C(1)–C(6) and C(1)–C(2).

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The Crystal Structure of Uronium Nitrate (Urea Nitrate) by Neutron Diffraction*

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Uronium nitrate, $(NH_2)_2COH^+NO_3^-$, crystallizes with symmetry $P2_1/c$ and lattice parameters $a=9.5434(6)$, $b=8.2010(5)$, $c=7.4982(4)$ Å, and $\beta=124.246(6)°$. The crystal structure, determined from 1744 independent experimental neutron diffraction intensities, shows a layer arrangement with all atoms approximately at $z=\frac{1}{4}$ or $\frac{3}{4}$. The acidic proton is on the carbonyl oxygen atom with an O–H distance of 1.006(3) Å and forms a hydrogen bond to a nitrate oxygen atom with an O–H···O distance of 2.596(2) Å. Four other hydrogen bonds of the type N–H···O join the uronium and nitrate ions into a two-dimensional network. The only contacts between the layers are of the van der Waals type. The C–O distance, 1.298(2) Å, and C–N distances, 1.312(1) and 1.315(1) Å, are longer and shorter, respectively, than the corresponding distances in urea.

Introduction

The discovery that the acidic proton in acetamide hemihydrochloride (Takei & Hughes, 1959; Peterson

& Worsham, 1959) is centered in a hydrogen bond between the carbonyl oxygen atoms of two adjacent acetamide molecules led to the consideration of similar compounds as further examples of the basic nature of the amide carbonyl oxygen. The prevalence of salts of urea made these compounds obvious choices for consideration and, of these, so-called urea nitrate was