

## Conformational Study on 4-(Dimethylamino)methanesulfonanilides. 1. Structures of 2-Methanesulfonyl- (I), 3-Methanesulfonyl- (II) and 2-Methanesulfonyl-3-methyl-4-(dimethylamino)methanesulfonanilide (III)

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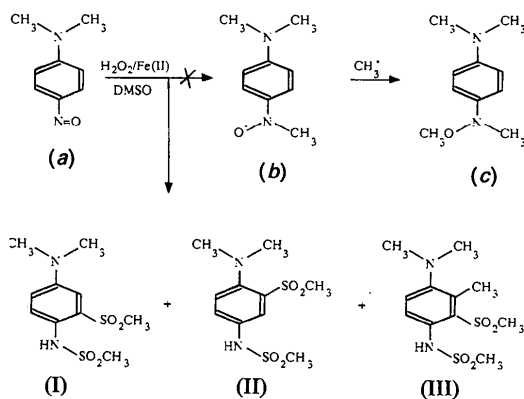
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**Abstract.** (I)  $C_{10}H_{16}N_2O_4S_2$ ,  $M_r = 292.4$ , triclinic,  $P\bar{1}$ ,  $a = 9.464$  (2),  $b = 8.893$  (2),  $c = 9.275$  (2) Å,  $\alpha = 105.5$  (1),  $\beta = 112.0$  (1),  $\gamma = 93.1$  (1)°,  $V = 687.0$  (8) Å<sup>3</sup>,  $Z = 2$ ,  $D_x = 1.41$  g cm<sup>-3</sup>,  $\lambda(\text{Cu } K\alpha) = 1.5418$  Å,  $\mu = 35.5$  cm<sup>-1</sup>,  $F(000) = 308$ ,  $T = 293$  K, final conventional  $R = 0.072$  for 2107 symmetry-independent observed reflections. (II)  $C_{10}H_{16}N_2O_4S_2$ ,  $M_r = 292.4$ , orthorhombic,  $P2_12_12_1$ ,  $a = 23.633$  (5),  $b = 10.430$  (3),  $c = 5.590$  (2) Å,  $V = 1377.9$  (7) Å<sup>3</sup>,  $Z = 4$ ,  $D_x = 1.41$  g cm<sup>-3</sup>,  $\lambda(\text{Cu } K\alpha) = 1.5418$  Å,  $\mu = 35.5$  cm<sup>-1</sup>,  $F(000) = 616$ ,  $T = 293$  K, final conventional  $R = 0.055$  for 1470 symmetry-independent observed reflections. (III)  $C_{11}H_{18}N_2O_4S_2$ ,  $M_r = 306.4$ , monoclinic,  $P2_1/c$ ,  $a = 16.541$  (3),  $b = 8.359$  (2),  $c = 11.089$  (2) Å,  $\beta = 105.5$  (1)°,  $V = 1477.5$  (9) Å<sup>3</sup>,  $Z = 4$ ,  $D_x = 1.38$  g cm<sup>-3</sup>,  $\lambda(\text{Cu } K\alpha) = 1.5418$  Å,  $\mu = 33.3$  cm<sup>-1</sup>,  $F(000) = 648$ ,  $T = 293$  K, final conventional  $R = 0.064$  for 1979 symmetry-independent observed reflections. Deviations from planar trigonal geometry of the dimethylamino group and from coplanarity of the benzene ring are observed in (II) and (III), where the steric hindrance between the substituents on the phenyl group is considerable.

**Introduction.** Fenton's reagent in Me<sub>2</sub>SO (Rudqvist & Torssell, 1971) is one of the most used methods for radical methylation (Giordano, Minisci, Fortelli & Vismara, 1984). The synthesis of the *N,O*-dimethylated hydroxylamine (c) from *N,N*-dimethyl-4-nitrosoaniline (a) was attempted by this method *via* the intermediate nitroxide (b), but no traces of the expected product (c) were obtained. Instead, the products isolated were 4,4'-bis(dimethylamino)-azoxybenzene, traces of *N,N*-dimethyl-4-nitroaniline, and the title compounds (I), (II) and (III). The X-ray analyses of these three compounds have been carried

out in order to understand the structure of these unexpected products and, above all, to establish the correct position of the methanesulfonyl group in (I) and (II), and that of the two new nuclear groups in (III).



**Experimental.** Crystals of the three compounds were colourless prisms. The specimen dimensions were 0.24 × 0.33 × 0.48, 0.47 × 0.38 × 0.57, 0.24 × 0.29 × 0.43 mm respectively. A Siemens AED diffractometer on line to a General Automation Jumbo 220 microcomputer was used with Ni-filtered Cu  $K\alpha$  radiation,  $\theta$ - $2\theta$  scan, scan width from  $(\theta - 0.60)$  to  $(\theta + 0.60 + 0.142 \tan \theta)$ °; scan speed 3.0° min<sup>-1</sup>. Cell dimensions were based on 29  $(\theta, \chi, \varphi)_{hkl}$  ( $20 \leq \theta \leq 30^\circ$ ) setting angles. For (I) 2578 ( $-11 \leq h \leq 10$ ,  $-10 \leq k \leq 10$ ,  $0 \leq l \leq 9$ ), (II) 1554 ( $0 \leq h \leq 28$ ,  $0 \leq k \leq 12$ ,  $0 \leq l \leq 6$ ) and (III) 2792 ( $-20 \leq h \leq 19$ ,  $0 \leq k \leq 10$ ,  $0 \leq l \leq 13$ ) symmetry-independent reflections ( $3 < \theta < 70^\circ$ ) were collected at  $T = 293$  K with a modified version (Belletti, Ugozzoli, Cantoni & Pasquinnelli, 1979) of the Lehmann & Larsen (1974) procedure. One standard reflection measured every

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Table 1. Fractional atomic coordinates ( $\times 10^4$ ) and equivalent isotropic thermal parameters ( $\text{\AA}^2 \times 10^4$ ) for non-H atoms with e.s.d.'s in parentheses

	$U_{\text{eq}} = (1/3)\sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i \cdot \mathbf{a}_j$			
	x	y	z	$U_{\text{eq}}$
(I)				
S(3)	3276 (1)	2708 (1)	4562 (1)	382 (3)
S(4)	5421 (1)	2579 (1)	9446 (1)	373 (3)
O(31)	3197 (3)	4180 (3)	5631 (3)	502 (10)
O(32)	2528 (3)	2429 (3)	2820 (3)	578 (11)
O(41)	5964 (3)	1135 (3)	8956 (3)	565 (10)
O(42)	6365 (3)	4076 (3)	9853 (3)	570 (10)
N(1)	183 (4)	-2867 (3)	2700 (3)	541 (11)
N(4)	3792 (3)	2547 (3)	7996 (3)	413 (9)
C(1)	1082 (3)	-1562 (3)	3989 (4)	391 (11)
C(2)	1653 (3)	-210 (3)	3726 (4)	374 (12)
C(3)	2547 (3)	1117 (3)	5041 (3)	332 (10)
C(4)	2897 (3)	1178 (3)	6658 (3)	326 (10)
C(5)	2325 (3)	-150 (3)	6919 (4)	384 (11)
C(6)	1441 (3)	-1490 (4)	5609 (4)	396 (11)
C(11)	-92 (5)	-4352 (4)	2992 (5)	625 (16)
C(12)	-248 (5)	-2844 (6)	1039 (5)	670 (17)
C(31)	5241 (4)	2554 (5)	5064 (6)	555 (18)
C(41)	4985 (5)	2628 (5)	11134 (4)	565 (15)
(II)				
S(3)	7190 (0)	2439 (1)	940 (2)	355 (3)
S(4)	6051 (0)	-2808 (1)	-1625 (2)	386 (3)
O(31)	7225 (1)	2760 (5)	3440 (7)	604 (13)
O(32)	7652 (1)	1690 (3)	11 (7)	494 (10)
O(41)	5463 (1)	-2748 (4)	-1106 (10)	668 (15)
O(42)	6263 (2)	-3787 (3)	-3181 (8)	624 (12)
N(1)	5959 (2)	3239 (3)	2078 (7)	386 (10)
N(4)	6236 (1)	-1455 (4)	-2897 (7)	389 (9)
C(1)	6016 (2)	2054 (4)	836 (7)	328 (9)
C(2)	6559 (1)	1585 (4)	296 (7)	314 (9)
C(3)	6633 (2)	421 (4)	-916 (9)	368 (12)
C(4)	6159 (2)	-276 (4)	-1622 (8)	352 (11)
C(5)	5623 (2)	186 (4)	-1141 (10)	440 (13)
C(6)	5556 (2)	1346 (4)	81 (9)	396 (11)
C(11)	5472 (2)	3997 (5)	1298 (12)	561 (17)
C(12)	5959 (3)	3063 (6)	4677 (9)	537 (16)
C(31)	7145 (2)	3851 (5)	-742 (12)	462 (14)
C(41)	6434 (3)	-2882 (6)	1071 (10)	571 (16)
(III)				
S(3)	2338 (1)	1614 (1)	4627 (1)	438 (3)
S(4)	838 (1)	-2330 (1)	5875 (1)	573 (3)
O(31)	1946 (2)	1796 (3)	5641 (3)	671 (11)
O(32)	3076 (2)	2533 (3)	4721 (3)	694 (11)
O(41)	1112 (2)	-3849 (4)	6405 (4)	852 (14)
O(42)	396 (2)	-1289 (5)	6526 (3)	805 (13)
N(1)	4015 (2)	-2863 (4)	3159 (3)	609 (12)
N(4)	1633 (2)	-1250 (4)	5774 (3)	581 (12)
C(1)	3402 (2)	-2454 (4)	3788 (3)	454 (12)
C(2)	3158 (2)	-836 (4)	3811 (3)	414 (11)
C(3)	2577 (2)	-436 (4)	4493 (3)	384 (9)
C(4)	2215 (2)	-1636 (4)	5075 (3)	458 (11)
C(5)	2451 (3)	-3212 (4)	4994 (5)	600 (14)
C(6)	3039 (3)	-3621 (4)	4378 (4)	594 (14)
C(11)	3972 (4)	-4522 (7)	2689 (5)	843 (22)
C(12)	4870 (3)	-2424 (8)	3851 (7)	888 (26)
C(21)	3499 (3)	364 (5)	3040 (4)	606 (15)
C(31)	1573 (3)	2107 (6)	3244 (4)	598 (15)
C(41)	226 (4)	-2625 (8)	4342 (5)	760 (19)

50 reflections, no significant variation. Corrections for Lorentz and polarization effects but not for absorption. Structures solved by direct methods (*SHELXS86*; Sheldrick, 1986). (I) 2107, (II) 1470 and (III) 1979 symmetry-independent observed reflections [ $I > 2\sigma(I)$ ] used in full-matrix least-squares refinement of 227 [(I) and (II)] and 244 (III) parameters; all H atoms located in the difference Fourier maps and refined isotropically.  $\sum w(F_o - F_c)^2$  minimized with  $w = [\sigma^2 F_o + g F_o^2]^{-1}$  where  $g$  was

Table 2. Bond distances ( $\text{\AA}$ ), angles ( $^\circ$ ) and selected torsion angles ( $^\circ$ ) with e.s.d.'s in parentheses

	(I)	(II)	(III)
S(3)—O(31)	1.440 (3)	1.439 (4)	1.448 (4)
S(3)—O(32)	1.443 (3)	1.440 (3)	1.422 (4)
S(3)—C(2)		1.774 (3)	
S(3)—C(3)	1.771 (4)		1.774 (4)
S(3)—C(31)	1.763 (4)	1.751 (6)	1.757 (5)
S(4)—O(41)	1.433 (3)	1.421 (3)	1.422 (4)
S(4)—O(42)	1.436 (3)	1.432 (4)	1.447 (4)
S(4)—N(4)	1.615 (3)	1.640 (4)	1.623 (4)
S(4)—C(41)	1.753 (5)	1.760 (6)	1.748 (6)
N(1)—C(1)	1.375 (4)	1.424 (5)	1.418 (5)
N(1)—C(11)	1.446 (5)	1.463 (7)	1.477 (7)
N(1)—C(12)	1.445 (6)	1.464 (6)	1.464 (6)
N(4)—C(4)	1.426 (4)	1.433 (6)	1.425 (5)
C(1)—C(2)	1.408 (5)	1.406 (5)	1.414 (5)
C(1)—C(6)	1.393 (5)	1.380 (6)	1.397 (6)
C(2)—C(3)	1.395 (4)	1.401 (6)	1.413 (5)
C(2)—C(21)			1.521 (6)
C(3)—C(4)	1.394 (4)	1.393 (7)	1.410 (5)
C(4)—C(5)	1.389 (5)	1.382 (7)	1.384 (5)
C(5)—C(6)	1.398 (4)	1.398 (6)	1.373 (8)
O(31)—S(3)—O(32)	118.0 (4)	115.7 (2)	116.1 (2)
O(31)—S(3)—C(2)		111.2 (2)	
O(31)—S(3)—C(3)	109.0 (3)		108.8 (2)
O(31)—S(3)—C(31)	108.2 (3)	109.2 (3)	106.6 (3)
O(32)—S(3)—C(2)		107.0 (2)	
O(32)—S(3)—C(3)	108.8 (3)		108.8 (2)
O(32)—S(3)—C(31)	108.2 (4)	108.0 (2)	110.0 (2)
C(2)—S(3)—C(31)		105.2 (2)	
C(3)—S(3)—C(31)	107.8 (4)		106.2 (2)
O(41)—S(4)—O(42)	119.9 (5)	119.8 (2)	118.6 (2)
O(41)—S(4)—N(4)	108.1 (3)	108.1 (2)	110.5 (2)
O(41)—S(4)—C(41)	109.5 (4)	109.3 (3)	108.3 (3)
O(42)—S(4)—N(4)	106.6 (4)	104.9 (2)	103.1 (2)
O(42)—S(4)—C(41)	106.6 (3)	108.0 (3)	109.4 (3)
N(4)—S(4)—C(41)	105.2 (4)	105.8 (2)	106.3 (2)
C(1)—N(1)—C(11)	119.7 (4)	113.5 (4)	115.3 (4)
C(1)—N(1)—C(12)	120.1 (5)	112.0 (4)	113.5 (4)
C(11)—N(1)—C(12)	119.3 (4)	111.3 (4)	111.7 (4)
S(4)—N(4)—C(4)	124.9 (4)	126.1 (3)	126.1 (3)
N(1)—C(1)—C(2)	121.2 (4)	119.5 (4)	119.0 (3)
N(1)—C(1)—C(6)	122.1 (5)	122.6 (4)	121.0 (3)
C(2)—C(1)—C(6)	116.7 (5)	117.8 (4)	120.0 (4)
S(3)—C(2)—C(1)		123.3 (3)	
S(3)—C(2)—C(3)		115.4 (2)	
C(1)—C(2)—C(3)	120.8 (4)	121.3 (3)	118.3 (3)
C(1)—C(2)—C(21)			118.1 (3)
C(3)—C(2)—C(21)			123.5 (3)
S(3)—C(3)—C(2)	116.6 (3)		118.2 (3)
S(3)—C(3)—C(4)	121.3 (4)		121.2 (3)
C(2)—C(3)—C(4)	122.1 (5)	119.3 (4)	120.6 (3)
N(4)—C(4)—C(3)	121.8 (5)	119.1 (4)	121.3 (3)
N(4)—C(4)—C(5)	121.0 (3)	120.8 (4)	119.6 (3)
C(3)—C(4)—C(5)	117.2 (4)	120.0 (4)	119.1 (4)
C(4)—C(5)—C(6)	121.1 (4)	120.0 (4)	121.3 (3)
C(1)—C(6)—C(5)	122.2 (5)	121.5 (4)	120.6 (3)
O(31)—S(3)—C(2)—C(1)		56.0 (4)	
O(32)—S(3)—C(2)—C(3)		0.0 (4)	
C(31)—S(3)—C(2)—C(1)		-62.2 (4)	
O(31)—S(3)—C(3)—C(4)	-40.8 (6)		15.5 (4)
O(32)—S(3)—C(3)—C(2)	11.3 (6)		-35.7 (4)
C(31)—S(3)—C(3)—C(4)	74.2 (5)		-98.9 (4)
O(41)—S(4)—N(4)—C(4)	13.3 (7)	59.9 (4)	60.7 (4)
O(42)—S(4)—N(4)—C(4)	143.4 (5)	-171.1 (3)	-171.6 (3)
C(41)—S(4)—N(4)—C(4)	-103.7 (5)	-57.1 (4)	-56.7 (4)
C(11)—N(1)—C(1)—C(6)	-15.7 (8)	-34.6 (6)	25.9 (6)
C(12)—N(1)—C(1)—C(2)	-2.7 (8)	-88.9 (5)	75.4 (5)
S(4)—N(4)—C(4)—C(5)	60.7 (7)	-68.1 (5)	-36.0 (5)
N(1)—C(1)—C(2)—C(21)			6.4 (5)
S(3)—C(3)—C(2)—C(21)			-8.6 (5)

0.073, 0.008 and 0.077 in (I), (II) and (III) respectively (*SHELX76* system; Sheldrick, 1976). Maximum shift of parameters,  $\Delta\rho_{\text{max}}$ ,  $\Delta\rho_{\text{min}}$ : (I) 0.8 $\sigma$ , 0.39, -1.1 e  $\text{\AA}^{-3}$ ; (II) 0.7 $\sigma$ , 0.53, -0.54 e  $\text{\AA}^{-3}$ ; (III) 0.6 $\sigma$ , 0.41, -0.90 e  $\text{\AA}^{-3}$ . Atomic scattering factors were from *International Tables for X-ray Crystallography* (1974).

**Discussion.** Final atomic coordinates and equivalent isotropic thermal parameters are given in Table 1.\* The arbitrary numbering scheme used in the crystal

\* Lists of structure factors, anisotropic thermal parameters and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 52957 (39 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

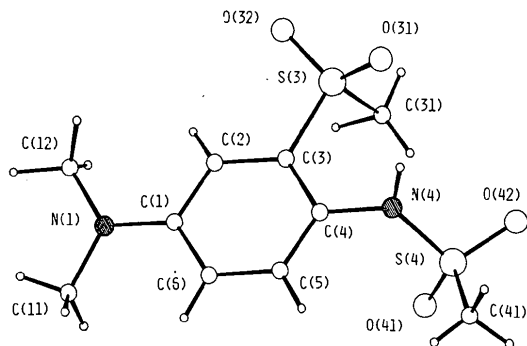


Fig. 1. Perspective view of 2-methanesulfonyl-4-(dimethylamino)methanesulfonanilide with the atomic numbering scheme.

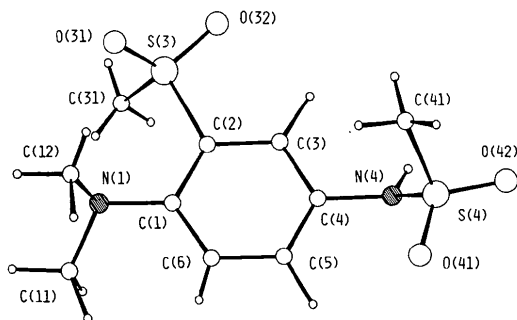


Fig. 2. Perspective view of 3-methanesulfonyl-4-(dimethylamino)methanesulfonanilide with the atomic numbering scheme.

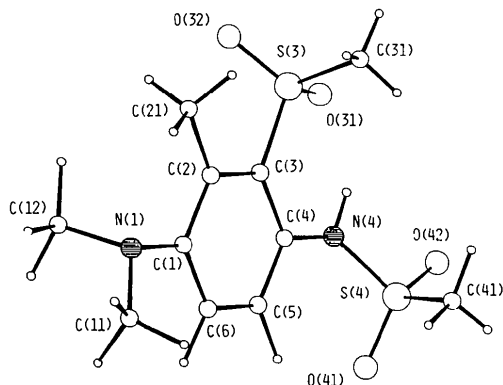


Fig. 3. Perspective view of 2-methanesulfonyl-3-methyl-4-(dimethylamino)methanesulfonanilide with the atomic numbering scheme.

Table 3. Distances ( $\text{\AA} \times 10^3$ ) of relevant atoms from the mean plane with *e.s.d.*'s in parentheses, and angles ( $^\circ$ ) between planes

Starred atoms were not used to define the plane.

Plane A	C(1)—C(6)	(I)	(II)	(III)
	C(1)	-2 (6)	-8 (4)	-11 (4)
	C(2)	5 (6)	6 (4)	19 (4)
	C(3)	-4 (6)	0 (5)	-14 (4)
	C(4)	0 (6)	-6 (4)	1 (4)
	C(5)	3 (6)	5 (5)	17 (5)
	C(6)	-2 (6)	5 (5)	-4 (5)
	S(3)*	-74 (1)	120 (1)	-116 (2)
	N(1)*	24 (7)	-5 (4)	-44 (4)
	N(4)*	12 (6)	11 (4)	-42 (4)
	C(21)*			161 (5)
Plane B	N(1), C(11), C(12); plane C	S(3), C(31), C(2) or C(3); plane D	C(4), N(4), S(4)	
	A-B	12.7 (5)	71.6 (3)	63.5 (3)
	A-C	75.5 (2)	63.9 (2)	82.4 (2)
	A-D	60.3 (4)	67.2 (3)	35.8 (3)

analysis is shown in Figs. 1, 2 and 3, which represent perspective views of the 2-methanesulfonyl-, 3-methanesulfonyl- and 2-methanesulfonyl-3-methyl-4-(dimethylamino)methanesulfonanilide molecules, respectively. Bond distances and angles and selected torsion angles are reported in Table 2.

The bond distances in the three derivatives are quite similar and in agreement with those expected from the hybridization of the atoms, but significant differences are observed in the dimethylamino group concerning both the N(1)—C(1) bond [in (I) 1.375 (4)  $\text{\AA}$ , significantly shorter than those found in (II) and (III), 1.424 (5) and 1.418 (5)  $\text{\AA}$  respectively] and the configuration at the N atom; in fact the angles C(1)—N(1)—C(11), C(1)—N(1)—C(12) and C(11)—N(1)—C(12) [(I) 119.7 (4), 120.1 (5), 119.3 (4) $^\circ$ ; (II) 113.5 (4), 112.0 (4), 111.3 (4) $^\circ$ ; (III) 115.3 (4), 113.5 (4), 111.7 (4) $^\circ$ ] show that only in compound (I) is a planar trigonal arrangement of the methyl substituents C(11) and C(12) present. So a comparison in terms of pyramidalicity of N(1) confirms that in (I) the N atom lies almost in the plane containing the three atoms attached to it, its distance from this plane being 0.081 (7)  $\text{\AA}$ , while in the other two compounds this value is 0.412 (4) and 0.377 (3)  $\text{\AA}$ , respectively. The orientation of the C—N(1)—C group with respect to the benzene ring is significantly different in the three derivatives, the dihedral angles being 12.7 (5), 71.6 (3) and 63.5 (3) $^\circ$ , respectively (see the analysis of the planarity reported in Table 3). We interpret this feature to be determined by steric interactions between the dimethylamino group and the substituents on the phenyl ring at C(2). Steric hindrance also appears to be responsible for the significant deviations from planarity exhibited by the benzene ring in (II) and (III), the deviation being more noticeable in (III)

where the interaction between the substituent groups seems to be stronger.

The orientation of the methanesulfonamide group with respect to the benzene ring, deduced from the torsion angles reported in Table 2, is not different for the three derivatives; the position adopted by the SO<sub>2</sub> group with respect to the N(4) atom is comparable with the N lone pair bisecting the O—S—O angle.

Short N(4)⋯O(31) contacts [(I) N(4)⋯O(31) 2.847 (5), H(4)⋯O(31) 2.26 (5) Å, N(4)—H(4)⋯O(31) 125 (4)°; (III) N(4)⋯O(31) 2.610 (4), H(4)⋯O(31) 1.86 (5) Å, N(4)—H(4)⋯O(31) 137 (4)°] suggest the presence of an intramolecular hydrogen bond between the sulfonamide N and a sulfonyl O atom of the vicinal methanesulfonyl group.

The molecular packing is determined by hydrogen bonds of the type N—H⋯O [(I) N(4)⋯O(42<sup>i</sup>) 3.176 (5), H(4)⋯O(42<sup>i</sup>) 2.62 (4) Å, N(4)—H(4)⋯O(42<sup>i</sup>) 123 (4)°, (i) = 1 - x, 1 - y, 2 - z; (II) N(4)⋯O(32<sup>ii</sup>) 2.887 (4), H(4)⋯O(32<sup>ii</sup>) 2.01 (4) Å, N(4)—H(4)⋯O(32<sup>ii</sup>) 166 (4)°, (ii) =  $\frac{3}{2} - x, -y, -\frac{1}{2} +$

z]. Other contacts are consistent with van der Waals interactions.

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## Crystallographic Studies and Physicochemical Properties of $\pi$ -Electron Systems.

### 21. Structure of *N,N'*-Bis(*p*-chlorophenyl)formamidine

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**Abstract.** C<sub>13</sub>H<sub>10</sub>Cl<sub>2</sub>N<sub>2</sub>, *M<sub>r</sub>* = 265.0, monoclinic, *C2/c* or *Cc*, *a* = 11.450 (2), *b* = 22.992 (7), *c* = 9.720 (3) Å,  $\beta$  = 93.81 (2)°, *V* = 2553.2 Å<sup>3</sup>, *Z* = 8, *D<sub>x</sub>* = 1.378 g cm<sup>-3</sup>,  $\lambda(\text{Cu } K\alpha)$  = 1.54178 Å,  $\mu$  = 42.37 cm<sup>-1</sup>, *F*(000) = 1088, *T* = 293 K, *R* = 0.0628, *wR* = 0.0687 for *C2/c*, for 1934 observed reflections. The crystal structure contains dimers of the title compound with two intermolecular N—H⋯N hydrogen bonds, 2.958 (3) Å, linking the two molecules in the dimer. The planes defining the amidine moieties in the dimer are inclined at a dihedral angle of 27.3 (2)°.

**Introduction.** Amidine derivatives have recently been extensively studied both chemically (Oszczapowicz & Ciszowski, 1987; Gilli & Bertolasi, 1979; Häfelinger, 1975) and crystallographically (Bellucci,

Bertolasi, Ferretii & Gilli, 1985; Norrestam, 1984*a,b*; Alcock, Barker & Kilner, 1988; Kratochvil, Ondracek, Krechl & Hasek, 1987; Ciszak, Gdaniec & Kosturkiewicz, 1987; Tykarska, Jaskólski & Kosturkiewicz, 1986*a,b*; Oszczapowicz, Tykarska, Jaskólski & Kosturkiewicz, 1986) to investigate their chemical (Shriner & Neumann, 1944; Schwenker & Bösl, 1969) and in particular biological (Grout, 1975) properties. One particular feature of some amidine derivatives is their tautomerism (Oszczapowicz & Ciszowski, 1987, 1989; Häfelinger, 1975) and this paper is a continuation of our studies (Anulewicz, Krygowski & Pniewska, 1987; Anulewicz, Krygowski, Jaroszevska-Manaj & Pniewska, 1989). Geometrical analysis of the amidine skeleton seems to be of great interest because of its dependence on the hydrogen bonding in dimers of formamidine