Discussion

Every P atom in NdP_3O_9 and NdP_5O_{14} is bonded to 4 O atoms to form a PO_4 tetrahedron; every O atom is bonded to at least one P atom, and there is no direct P–Nd bonding. Therefore, the basic structures of these compounds are determined by the requirement that the O atoms must be shared in a manner that reduces the O/P ratio (R) from the value of 4 for isolated tetrahedra, as in $NdPO_4$ (Mooney, 1948), to the observed values of 3 and $\frac{14}{5}$, respectively. In NdP_3O_9 this reduction is accomplished by the formation of one-dimensional –P(2)–P(2)–P(1)– chains in which each PO_4 tetrahedron shares 2 of its O atoms with the adjacent tetrahedra leaving the other 2 unshared, to give $R=2(\frac{1}{2})+2=3$. To decrease R to less than 3 requires cross-linking between chains.

If 2 parallel chains are completely cross-linked by sharing O atoms, each PO_4 tetrahedron shares 3 of its O atoms with the adjacent tetrahedra, leaving 1 unshared, to give $R=3(\frac{1}{2})+1=2{\cdot}5$. If only a fraction x of the tetrahedra are cross-linked, $R=2{\cdot}5x+3(1-x)$. In NdP_5O_{14} the P(3) and P(5) tetrahedra in the 2 chains are linked by sharing P(4) tetrahedra rather than O atoms directly, but the same formula for R can be applied if x is defined as the fraction of the tetrahedra that share 3 O atoms and $(1-x)$ is the fraction that share only 2. Since $R=2{\cdot}8$ for NdP_5O_{14}, x must be 0·4. This value of x is obtained for the structure shown in Fig. 4, where the P(3) and P(5) tetrahedra each share 3 O atoms and the P(1), P(2), and P(4) tetrahedra each share 2 O atoms.

In NdP_5O_{14} the Nd atoms are isolated from each other by PO_4 tetrahedra, since no O atom is common to 2 Nd atoms. This isolation appears to account for the optical observations of Danielmeyer & Weber (1972) indicating that the Nd–Nd pair interaction is small in this compound. This interaction can be expected to be larger in NdP_3O_9, since each Nd atom shares 2 O atoms with one of the neighboring Nd atoms and 2 other O atoms with another. In addition, the minimum Nd–Nd distance in NdP_3O_9 is 4·234 Å, compared with 5·194 Å in NdP_5O_{14}.

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References

BEUCHER, M. (1970). *Les Elements des Terres Rares, Coll.* No. 180, Vol. 1, pp. 331–336. Paris C.N.R.S. (Centre National de la Recherche Scientifique).

CROMER, D. T. (1965). *Acta Cryst.* **18**, 17–23.

CROMER, D. T. & WABER, J. T. (1965). *Acta Cryst.* **18**, 107–109.

DANIELMEYER, H. G. & WEBER, H. P. (1972). *J. Quantum Electron.* **QE-8**, 805–808.

DURIF, A. (1971). *Bull. Soc. Fr. Minér. Crist.* **94**, 314–318.

JAULMES, S. (1969). *C. R. Acad. Sci. Paris, Sér. C*, **268**, 935–937.

MOONEY, R. C. L. (1948). *J. Chem. Phys.* **16**, 1003.

Acta Cryst. (1974). B**30**, 474

Hydrogen Bond Studies. LXXXI.* The Crystal and Molecular Structure of 2-Amino-5-chloropyridine, $C_5H_3N(NH_2)Cl$

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The crystal and molecular structure of 2-amino-5-chloropyridine, $C_5H_3N(NH_2)Cl$, has been determined from four-circle X-ray diffractometer data (Mo $K\alpha$ radiation). The crystals are monoclinic, space group $P2_1/c$, with four formula units in a cell of dimensions $a=13{\cdot}4370$ (6), $b=5{\cdot}7963$ (5), $c=7{\cdot}5123$ (6) Å, $\beta=105{\cdot}512$ (6)°. The hydrogen atoms have been located and full-matrix least-squares refinement gave a final $R(F)$ value of 0·033. The molecules link *via* two N–H···N hydrogen bonds [3·058 (2) Å], to produce (centrosymmetric) dimers which, in turn, pack in a herring-bone formation.

Introduction

This work is part of a systematic investigation of hydrogen bonding in simple organic compounds which can serve as model substances for biologically important molecules. The crystal structures of 6-chloro-2-hydroxypyridine, the addition compound 6-chloro-2-hydroxypyridine–2-pyridone and 5-chloro-2-pyridone have been reported earlier by Kvick & Olovsson (1968), Almlöf, Kvick & Olovsson (1971) and Kvick & Booles

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(1972). We present here the structure determination of 2-amino-5-chloropyridine, $C_5H_3N(NH_2)Cl$, based on single-crystal X-ray diffraction data collected at room temperature. Further investigations of related compounds are in progress at this Institute.

Experimental

Commercial 2-amino-5-chloropyridine was recrystallized several times from chloroform. The final rhomboidal crystals had an experimental melting point of +139 °C.

The diffraction symmetry and systematic absences suggested the monoclinic space group $P2_1/c$ (No. 14, *International Tables for X-ray Crystallography*, 1952).

The cell dimensions were determined from powder photographs taken with a Guinier–Hägg powder camera at +24 °C with monochromated Cr $K\alpha_1$ radiation (λ=2·28962 Å). CoP_3 (a=7·7073 Å) was used as an internal standard. Thirty-six reflexions were used in a least-squares procedure to obtain the cell constants (Table 1).

Table 1. *Crystallographic and physical data*

2-Amino-5-chloropyridine	$C_5H_3N(NH_2)Cl$
F.W.	128·562
Space group	$P2_1/c$
a	13·4370 (6) Å*
b	5·7963 (5)
c	7·5123 (6)
β	105·512 (6)°
U	563·78 Å^3
D_λ	1·514 g cm^{-3}
Z	4
M.p.	139 °C
μ	5·53 cm^{-1}

* Numbers in parentheses here and throughout this paper are the estimated standard deviations in the least significant digits.

A crystal of dimensions 0·18×0·21×0·17 mm was mounted in a glass capillary (wall thickness about 0·02 mm) to prevent sublimation during the experiment. The data collection from this crystal was carried out at +24 °C with a PDP8/I controlled four-circle Stoe X-ray diffractometer and graphite-monochromated molybdenum radiation (λ=0·7107 Å). An ω–2θ scan technique was used to obtain the intensities. Automatic filter selection made it possible to reduce the count rate in the detector by a ratio of up to 8:1. This, in combination with automatic scan-speed selection (×1, ×2 or ×4), resulted in a maximum absolute internal scale factor of 32. Three standard reflexions measured at regular intervals during the experiment showed only those variations expected from counting statistics. No rescaling of the data was therefore necessary. In all, 2312 reflexions were measured with $\sin\theta/\lambda < 0{\cdot}754$ Å^{-1}. This set included some equivalent reflexions which were averaged together after absorption correction. Removal of reflexions of zero intensity due to space group extinction left a total of 2031 reflexions. 736 of these had $I < 3\sigma(I)$.

The data were corrected for background, Lorentz, polarization and absorption effects with the program *DATAPH* (Coppens, Leiserowitz & Rabinovich, 1965). A correction for monochromator polarization was also included according to the formula:

$$p = \frac{1+\cos^2 2\theta_M \cos^2 2\theta}{1+\cos^2 2\theta_M}$$

with $\theta_M = 6{\cdot}1°$ and θ the scattering angle for the measured reflexion. The linear absorption coefficient for Mo $K\alpha$ radiation was calculated to be 5·53 cm^{-1}. Crystal data are given in Table 1.

Structure determination and refinement

The structure was solved by direct methods. The normalized structure factor amplitudes ($|E|$) were obtained from the program *EFAK* (Kvick, 1972) according to the formula:

$$E^2 = \{k \exp[2B(\sin\theta/\lambda)^x]I\}/(\varepsilon \sum_{j=1}^{N} f_j^2)$$

where ε is a symmetry dependent integer, f_j is the scattering factor for the jth atom in the unit cell and N is the number of atoms in the cell. The parameters k, B and x were optimized in a least-squares procedure. The quantity minimized to obtain these parameters is:

$$\sum_{i=1}^{M} [K_i - \ln(k) - 2B(\sin\theta/\lambda)^x]^2$$

where M is the number of $\sin\theta/\lambda$ intervals used and K_i is $\ln(\sum_{j=1}^{N} f_j^2/\langle I\rangle)$ for the ith interval. The statistical distribution of the $|E|$ values is compared in Table 2 with the theoretical values expected for centric and acentric structures. The theoretical values used are those given by Karle, Dragonette & Brenner (1965). The comparison indicated a centric structure, and in agreement with observed extinctions space group $P2_1/c$ was chosen. This was later confirmed by the successful refinement in this space group.

Table 2. *Statistical distribution of the normalized structure amplitudes (E) compared with theoretical values*

	Experimental	Centro-symmetric	Non-centro-symmetric
$\langle\|E\|^2\rangle$	1·00	1·00	1·00
$\langle\|E^2-1\|\rangle$	0·979	0·968	0·736
$\langle\|E\|\rangle$	0·781	0·798	0·886
$\|E\|\% > 3$	0·2	0·3	0·01
$\|E\|\% > 2$	3·1	5·0	1·8
$\|E\|\% > 1$	28·5	32·0	37·0

206 reflexions with $|E| \geq 1{\cdot}5$ were used as input to a multisolution program (Long, 1965) for sign determination by iterative application of Sayre's formula.

Of the 16 solutions obtained, the one with the highest consistency index (0·99) and the fastest convergence gave a useful E map. It was seen later that all of the 206 signs from this solution were correctly determined.

Atomic coordinates obtained for the heavy atoms from the correct E map and a scale factor were refined in the least-squares program *LINUS* (Coppens & Hamilton, 1970). The quantity minimized in all least-squares refinements was $\sum w[|F_o|-|F_c|]^2$. The reflexions were weighted according to the formula:

$$w^{-1}=\sigma_c^2(F^2)/4F^2+(kF)^2=\sigma^2(F)$$

with $k=0{\cdot}022$ in the final cycles of refinements and σ_c based on counting statistics. The quantity kF was introduced to take account of other errors not arising from counting statistics. The heavy atoms were refined for a few cycles with isotropic temperature factors. A few cycles of anisotropic refinement and a subsequent Fourier difference synthesis using data with $\sin\theta/\lambda<0{\cdot}5$ Å^{-1} then revealed all the hydrogen atom positions. The hydrogen atom peaks varied between 0·60 and 0·69 e Å^{-3}. The spurious peaks were all less than 0·2 e Å^{-3}. The validity of the hydrogen atom positions were confirmed from chemical considerations. The presence of residual charge density at the chlorine atom site suggested the inclusion of anomalous scattering for the chlorine. The values used in the subsequent refinement were $\Delta f'=0{\cdot}15$ and $\Delta f''=0{\cdot}19$ (Cromer, 1965).

In the final cycles of refinement with the least-squares program *UPALS* (Lundgren, 1972), one scale factor, the atomic coordinates for all atoms, anisotropic thermal parameters for the heavy atoms and isotropic temperature factors for the hydrogen atoms (93 parameters in all) were allowed to vary. 1295 reflexions had non-zero weight. The final shifts were all smaller than $0{\cdot}5\sigma$. No evidence for secondary extinction effects was observed. The refinement converged at:

$$R=(\sum||F_o|-|F_c||)/\sum|F_o|=0{\cdot}033$$

$$R_w=\{\sum w(|F_o|-|F_c|)^2/\sum w|F_o|^2\}^{1/2}=0{\cdot}060\,.$$

The reflexions with I smaller than $3\sigma(I)$, which were given zero weight in the refinements, all had $|F_o-F_c|/\sigma(F)<3{\cdot}0$ except for 11 reflexions where the quantity varied between 3·0 and 5·0. The atomic parameters from the final refinement are given in Tables 3 and 4. Observed and calculated structures factor amplitudes are listed in Table 5.

Table 3. *Atomic coordinates with standard deviations in parentheses* ($\times 10^5$)

The values for the hydrogen atoms are $\times 10^3$.

	x	y	z
Cl	7757 (3)	18353 (8)	36218 (7)
N(1)	36268 (9)	43245 (24)	46955 (18)
N(2)	44093 (14)	76662 (35)	60237 (29)
C(2)	35415 (12)	63717 (27)	54771 (21)
C(3)	25907 (13)	71564 (28)	57005 (22)
C(4)	17319 (12)	58020 (29)	51096 (22)
C(5)	18287 (11)	36715 (26)	43269 (21)
C(6)	27797 (11)	30083 (26)	41387 (22)
H(1)	493 (2)	715 (4)	582 (3)
H(2)	444 (2)	891 (5)	660 (3)
H(3)	257 (1)	865 (3)	624 (2)
H(4)	108 (2)	632 (4)	524 (3)
H(6)	286 (1)	154 (3)	357 (2)

The atomic scattering factors used were those given by Hanson, Herman, Lea & Skillman (1964), except for the hydrogen atoms for which the spherical scattering factors given by Stewart, Davidson & Simpson (1965) were preferred.

The calculations were performed on the CDC 3600 and IBM 370/155 computers in Uppsala. Smaller calculations were performed using the departmental IBM 1800 computer.

Table 4. *Thermal parameters with estimated standard deviations* ($\times 10^5$)

The anisotropic thermal parameters are of the form $\exp(-\beta_{11}h^2-\ldots-2\beta_{12}hk\ldots)$.

Heavy atoms

	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
Cl	391 (2)	2930 (15)	2412 (11)	−204 (4)	248 (3)	−394 (9)
N(1)	379 (7)	2200 (38)	1778 (27)	15 (13)	211 (11)	−137 (26)
N(2)	562 (10)	3380 (59)	3108 (48)	−453 (21)	491 (18)	−1382 (44)
C(2)	446 (8)	2144 (45)	1451 (28)	−83 (16)	210 (12)	−49 (28)
C(3)	535 (9)	1971 (45)	1571 (30)	43 (17)	323 (14)	−139 (29)
C(4)	439 (9)	2301 (45)	1610 (30)	157 (16)	312 (13)	158 (31)
C(5)	360 (7)	2101 (42)	1424 (26)	−22 (15)	166 (11)	135 (27)
C(6)	402 (7)	1934 (41)	1528 (28)	46 (15)	202 (12)	−130 (29)

Table 4 (*cont.*)

Hydrogen atoms

	B(Å^2)
H(1)	4·3 (5)
H(2)	5·1 (6)
H(3)	3·0 (4)
H(4)	4·7 (5)
H(6)	2·8 (4)

Description of the structure

The 2-amino-5-chloropyridine molecules exist in the amino form and are linked to produce dimers *via* pairs of N–H···N hydrogen bonds [3·058 (2) Å]. Only one of the amino hydrogen atoms participates in hydrogen bonding. The two molecules of the dimer are related

Table 5. *Observed and calculated structure factor amplitudes*

The columns are in order h, l, $10|F_o|$, and $10|F_c|$. Reflexions with $I < 3\sigma(I)$ are not included.

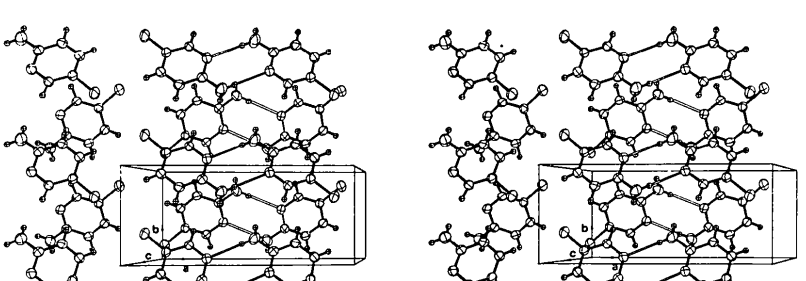

Fig. 1. Stereoscopic drawing of the crystal structure of 2-amino-5-chloropyridine. Covalent bonds are filled and hydrogen bonds are open. The thermal ellipsoids are scaled to enclose 50% probability.

by a centre of symmetry and are both planar and parallel to each other (Table 6). Figs. 1 and 2 give stereoscopic views of the structure. All illustrations in this paper have been prepared with the plotting program *ORTEP*-II (Johnson, 1965).

Table 6. *Deviations of the atoms from the least-squares plane through the six heavy atoms in the ring*

The primed atoms belong to the second molecule in the dimer. The equation of the least-squares plane is:

$$0{\cdot}49261x + 2{\cdot}50253y - 6{\cdot}59750z = -1{\cdot}84060.$$

N(1)	0·004 (1) Å
C(2)	−0·004 (2)
C(3)	−0·002 (2)
C(4)	0·007 (2)
C(5)	−0·005 (2)
C(6)	0·000 (2)
Cl	−0·051 (1)
N(2)	0·002 (2)
H(1)	0·033 (22)
H(2)	−0·067 (23)
H(3)	0·013 (15)
H(4)	0·019 (22)
H(6)	0·015 (15)
N′(1)	0·075 (1)
C′(2)	0·083 (2)
C′(3)	0·081 (2)
C′(4)	0·072 (2)
C′(5)	0·084 (2)
C′(6)	0·079 (2)

In the crystal structure there are alternating zones of hydrogen bond and van der Waals interactions (Fig. 3).

The root-mean-square (r.m.s.) vibrational amplitudes along the principal axes are given in Table 7.

Table 7. *The root-mean-square components* R_i (Å) ($\times 10^3$) *of thermal displacement of the atoms along the ellipsoid axes*

	R_1	R_2	R_3
Cl	174 (1)	222 (1)	261 (1)
N(1)	179 (2)	192 (2)	220 (2)
N(2)	182 (2)	221 (2)	320 (2)
C(2)	184 (2)	197 (2)	201 (2)
C(3)	177 (2)	201 (2)	217 (2)
C(4)	177 (2)	195 (2)	218 (2)
C(5)	174 (2)	184 (2)	202 (2)
C(6)	175 (2)	187 (2)	205 (2)

Packing arrangements

The packing in the structure is illustrated in Figs. 1, 2 and 3. The hydrogen-bonded dimeric units are related by twofold screw axes. Two sets of such units are thus formed (Fig. 2) with an angle of 51·2° between their respective planes. Each of the sets constitute infinite stacks of parallel dimers in the **b** direction. No overlapping occurs within the stacks. The molecules form a herring-bone pattern in projection along the *a* axis as illustrated in Fig. 2.

Fig. 3 illustrates the feature of alternating hydrogen-bonded and van der Waals bonded zones. The shaded area in the figure illustrates one of the zones containing the hydrogen bonds. The chlorine atoms and most of the carbon atoms are included in the intervening layers dominated by van der Waals interactions. The zones are parallel to the crystallographic *bc* plane.

Except for the hydrogen-bond distances there are no intermolecular contacts shorter than the sum of the van der Waals radii (Pauling, 1960). The shortest contacts between the dimeric units are: C(4) $(x,y,z)\cdots$ H(6) $(x,\frac{1}{2}-y,\frac{1}{2}+z)$ 2·96 (2) Å; C(4) $(x,y,z)\cdots$Cl

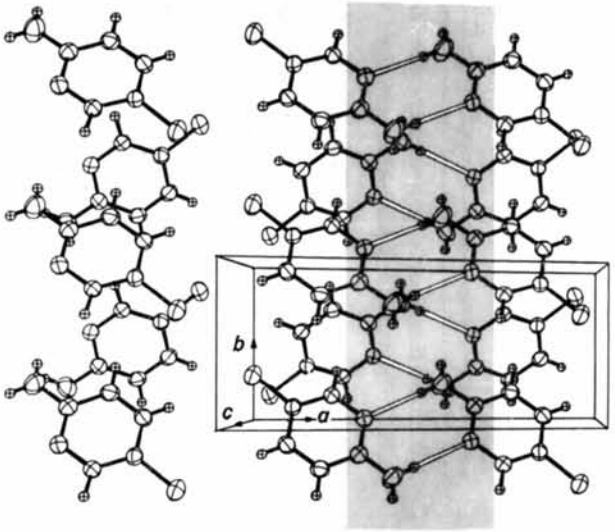

Fig. 3. Drawing showing the alternating hydrogen-bonded and van der Waals-bonded zones in the structure. One hydrogen-bonded zone is shaded.

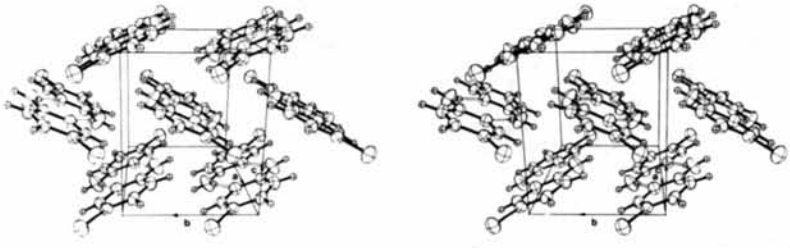

Fig. 2. Stereoscopic drawing showing the structure viewed approximately along the planes of the dimers.

$(x,\frac{1}{2}-y,\frac{1}{2}+z)$ 3·572 (2) Å; C(6) $(x,y,z)\cdots$H(3) $(x,\frac{3}{2}-y,-\frac{1}{2}+z)$ 2·87 (2) Å.

Molecular dimensions

The bond distances and angles, given in Tables 8 and 9 and illustrated in Fig. 4, have been calculated using the program *ORFFE* (Busing, Martin & Levy, 1964). The standard deviations have been estimated from the errors in the atomic coordinates obtained in the form of a variance–covariance matrix from the last cycle of refinement. The effect of the errors in the cell dimensions has also been taken into account.

Table 8. *Bond lengths*

The distances are not corrected for thermal motion.

(*a*) Covalent bonds

Cl—C(5)	1·736 (1) Å
N(1)–C(2)	1·342 (2)
N(1)–C(6)	1·341 (2)
N(2)–C(2)	1·355 (2)
N(2)–H(1)	0·812 (24)
N(2)–H(2)	0·839 (26)
C(2)–C(3)	1·408 (2)
C(3)–C(4)	1·368 (2)
C(3)–H(3)	0·960 (19)
C(4)–C(5)	1·389 (2)
C(4)–H(4)	0·953 (22)
C(5)–C(6)	1·377 (2)
C(6)–H(6)	0·971 (19)

(*b*) Hydrogen bond

N(1)···N(2)	3·058 (2)
N(1)···H(1)	2·246 (25)
N(2)···H(1)	0·812 (24)

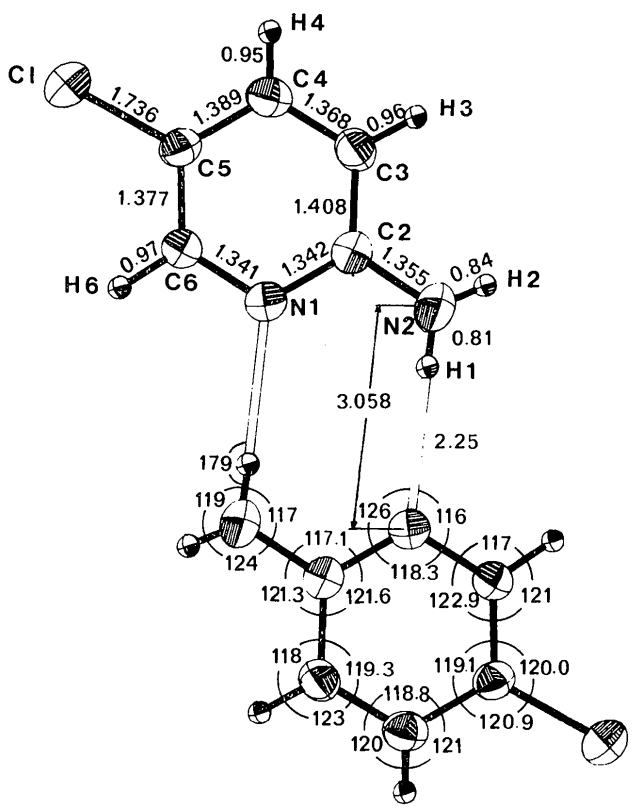

Fig. 4. Bond distances and angles. The values are not corrected for thermal motion.

Table 9. *Bond angles*

(*a*) Covalent bonds

C(2)–N(1)–C(6)	118·3 (1)°
C(2)–N(2)–H(1)	117·0 (16)
C(2)–N(2)–H(2)	123·6 (16)
H(1)–N(2)–H(2)	119·3 (23)
N(1)–C(2)–N(2)	117·1 (2)
N(1)–C(2)–C(3)	121·6 (1)
N(2)–C(2)–C(3)	121·3 (2)
C(2)–C(3)–C(4)	119·3 (1)
C(2)–C(3)–H(3)	117·8 (11)
C(4)–C(3)–H(3)	122·9 (11)
C(3)–C(4)–C(5)	118·8 (1)
C(3)–C(4)–H(4)	120·3 (14)
C(5)–C(4)–H(4)	120·9 (14)
Cl—C(5)–C(4)	120·9 (1)
Cl—C(5)–C(6)	120·0 (1)
C(4)–C(5)–C(6)	119·1 (1)
N(1)–C(6)–C(5)	122·9 (1)
N(1)–C(6)–H(6)	116·6 (11)
C(5)–C(6)–H(6)	120·5 (11)

(*b*) Hydrogen bond

C(2)–N(1)···N(2)	126·1 (1)
C(2)–N(1)···H(1)	126·2 (6)
C(6)–N(1)···N(2)	115·5 (1)
C(6)–N(1)···H(1)	115·5 (6)
N(2)–H(1)···N(1)	178·9 (22)

The skeleton of the pyridine ring is planar; the deviations of the heavy atoms from their least-squares plane are all less than or equal to 0·007 Å. The amino group and the hydrogen atoms also lie in this plane whereas Cl deviates significantly from the plane [0·051(1) Å] (Table 6).

The two molecules of the dimer are related by a centre of symmetry and are parallel. The distance between the least-squares planes of the two molecules is 0·08 Å.

The mean length of the four carbon–carbon bonds in the pyridine ring is 1·386 Å. The double-bond character in C(3)–C(4) [1·368 (2) Å] and C(5)–C(6) [1·377 (2) Å] is slightly larger than in C(2)–C(3) [1·408 (2) Å] and C(4)–C(5) [1·389 (2) Å]. The mean carbon–carbon distance (1·386 Å) can be compared with the mean value of 1·378 Å in 6-chloro-2-hydroxypyridine and 6-bromo-2-hydroxypyridine (Kvick, 1973). The longest carbon–carbon bond is the one closest to the amino group. The carbon–nitrogen distances within the ring are 1·341 (2) and 1·342 (2) Å. These do not differ significantly from the carbon–nitrogen distances involving non-protonated nitrogen atoms encountered earlier in this series of investigations. The carbon–chlorine distance of 1·736 (1) Å is comparable with 1·740 (3) Å in 6-chloro-2-hydroxypyridine and 1·736(3) Å in 5-chloro-2-pyridone.

The hydrogen atom positions

The difference Fourier synthesis clearly indicated that the ring nitrogen atom was not protonated. The angle C(2)–N(1)–C(6) was found to be 118·3 (1)°, as

compared to 124·4 (3)° in 5-chloro-2-pyridone where the ring nitrogen atom is protonated.

The hydrogen positions found from the difference synthesis were refined to give carbon–hydrogen distances between 0·95 (2) and 0·97 (2) Å (Table 8). As often observed the deviations from spherical electron density for the hydrogen atoms cause shifts of the apparent X-ray hydrogen positions (see *e.g.* Almlöf, Kvick & Thomas, 1973).

The hydrogen bond

Dimers are formed by hydrogen bonding between the nitrogen atoms. The N···N distance is 3·058 (2) Å and the bond is almost linear [N(2)–H(1)···N(1)= 179 (2)°]. The distance N(2)–H(1) is 0·81 (2) Å and N(1)···H(1) 2·25 (2) Å. These distances are also affected by the non-spherical charge density for the hydrogen atoms. The accepted N(2)–H(1) nuclear distance is about 0·2 Å longer.

It may be observed that the N···N hydrogen bond, which in this case has an amino nitrogen atom as donor, is weaker than the N···N bond in 6-chloro-2-hydroxypyridine–2-pyridone [N···N 2·904 (4) Å], where the pyridine nitrogen atom serves as hydrogen bond donor. Both bonds are almost linear, so that their N···N distances may be taken as a measure of the relative strengths of the two bonds.

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References

ALMLÖF, J., KVICK, Å. & OLOVSSON, I. (1971). *Acta Cryst.* B**27**, 1201–1208.
ALMLÖF, J., KVICK, Å. & THOMAS, J. O. (1973). *J. Chem. Phys.* In the press.
BUSING, W. R., MARTIN, K. O. & LEVY, H. A. (1964). *ORFFE.* Report ORNL-TM-306, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
COPPENS, P. & HAMILTON, W. C. (1970). *Acta Cryst.* A**26**, 71–83.
COPPENS, P., LEISEROWITZ, L. & RABINOVICH, D. (1965). *Acta Cryst.* **18**, 1035–1038.
CROMER, D. T. (1965). *Acta Cryst.* **18**, 17–23.
HANSON, H. P., HERMAN, F., LEA, J. D. & SKILLMAN, S. (1964). *Acta Cryst.* **17**, 1040–1044.
International Tables for X-ray Crystallography. (1952). Vol. I. Birmingham: Kynoch Press.
JOHNSON, C. K. (1965). *ORTEP.* Report ORNL-3794, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
KARLE, I. L., DRAGONETTE, K. S. & BRENNER, S. A. (1965). *Acta Cryst.* **19**, 713–716.
KVICK, Å. (1972). Unpublished.
KVICK, Å. (1973). To be published.
KVICK, Å. & BOOLES, S. (1972). *Acta Cryst.* B**28**, 3405–3409.
KVICK, Å. & OLOVSSON, I. (1968). *Ark. Kem.* **30**, 71–80.
LONG, R. E. (1965). *The Crystal and Molecular Structures of 7,7,8,8-Tetracyanoquinodimethane and Cyclopropane Carboxamide, and a Program for Phase Determination.* Doctoral Dissertation, Univ. of California, Los Angeles.
LUNDGREN, J.-O. (1972). *UPALS, a Crystallographic Full-Matrix Least-Squares Refinement Program.* UUIC-B13-3, Inst. of Chemistry, Univ. of Uppsala, Uppsala.
PAULING, L. (1960). *The Nature of the Chemical Bond.* Ithaca: Cornell Univ. Press.
STEWART, R. F., DAVIDSON, E. R. & SIMPSON, W. T. (1965). *J. Chem. Phys.* **42**, 3175–3187.

Acta Cryst. (1974). B**30**, 480

The Crystal and Molecular Structure of *cis*-1-(4-Chlorophenyl)-3-methyl-2-methylisoindoline

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The crystal structure of *cis*-1-(4-chlorophenyl)-3-methyl-2-methylisoindoline has been determined by direct methods, from 1851 reflexions collected on a Picker automatic diffractometer. The crystals, which have monoclinic unit-cell dimensions $a=9{\cdot}071$, $b=12{\cdot}443$, $c=11{\cdot}945$ Å and $\beta=93{\cdot}2°$, belong to the space group $P2_1/c$. The final R value is 0·040. The C(3) methyl and the C(1) chlorophenyl groups are *cis* to each other while the N(2) and the C(3) methyl groups are *trans*. The molecules are held in the crystal by van der Waals forces only.

Introduction

Isomers of 1-(4-chlorophenyl)-2,3-dimethylisoindolines were synthesized recently (Freter, Hess, Sygusch & Brisse, 1971). However, the relative orientation of the substituents at C(1) and C(3) could not be determined on the basis of standard n.m.r. techniques. Interpretation of n.m.r. data of symmetrically substituted isoindolines previously led to erroneous assignments (Carpino, 1962), while attempts to assign the stereochem-