trans-Reduction of 5-Phenyl-1,2,3,4,5,6-hexahydroazepino[4,5-*b*]indole. *X*-Ray Crystallographic Study of the Geometry of an Intermolecular N–H · · · Phenyl Hydrogen Bond

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The stereochemistries of the two isomeric products (5) and (6) obtained in equal amounts from *trans*-reduction of 3-benzyl-5-phenyl-1,2,3,4,5,6-hexahydroazepino[4,5-*b*]indole (3) have been determined by spectral and single-crystal *X*-ray analyses. Monoclinic crystals of (5) belong to space group $P2_1/c$, with $a = 22.005(10)$, $b = 10.270(5)$, $c = 13.577(7)$ Å, $\beta = 106.42(1)°$, $Z = 8$. The crystal structure was solved by direct methods, and atomic positional and thermal parameters refined by least-squares calculations to R 0.051 over 3 293 reflections measured by diffractometer. The two molecules defining the asymmetric crystal unit have identical conformations and are associated in a like manner in the solid state by N–H · · · N (mean N · · · N 3.26 Å) and N–H · · · phenyl (mean calculated H · · · phenyl 2.32 Å) hydrogen bonds.

As part of a programme aimed at the synthesis of indole derivatives possessing central nervous system activity, the two possible *trans*-indolines (5) and (6), resulting from *trans*-reduction of 5-phenyl-1,2,3,4,5,6-hexahydro[4,5-*b*]indole [1] (1) † were sought. A route to these compounds was suggested by earlier investigations of Berger *et al.*[2] who showed that *trans*-indolines could be prepared by acidic treatment of borane complexes of indoles containing tertiary amine side-chains.

In the present work, benzyl derivative (3) was reacted with a solution of borane in tetrahydrofuran to give the required borane adduct (4) which, on further treatment with refluxing ethanolic hydrochloric acid, furnished a mixture of two products. Separation of these products by chromatography on silica gel, followed by catalytic debenzylation, afforded equal amounts of stereoisomers (5) and (6) in 44% overall yield.

Although the ^{1}H n.m.r. spectra of (5) and (6) in $CDCl_3$ were grossly similar, they showed two major differences. Signals for the pendant phenyl ring protons appeared as a singlet in the spectrum of (5) whereas they occurred as a multiplet in that of (6). Moreover, a rather broad triplet at δ 4.03 in the spectrum of (5), which was replaced in that of (6) by a sharp doublet of doublets at δ 4.40, was assigned to H(5). The magnitudes of the coupling constants derived from extensive decoupling experiments [$J_{5,5a} = J_{5a,10b} = 10.5$ Hz in (5); $J_{5,5a} = 6.5$, $J_{5a,10b} = 10.5$ Hz in (6)] indicated that H(5a) and H(10b) are *trans* in both compounds as are H(5) and H(5a) in (5) while the latter are *cis* in (6). These configurational assignments were verified by the results of a single-crystal *X*-ray analysis of (5).

The crystal structure of (5) was solved by direct methods. Least-squares adjustment of atomic positional and thermal parameters converged to R 0.051 over 3 293 reflections measured by diffractometer. Final non-hydrogen atom positional parameters are in Table 1; bond lengths and angles, together with their means, are listed in Table 2. Endocyclic torsion angles characterizing the five- and seven-membered ring conformations and displacements of selected atoms from least-squares planes are in Tables 3 and 4, respectively.

Table 1. Non-hydrogen atom fractional co-ordinates (× 10^5), with standard deviations in parentheses

Atom	*x*	*y*	*z*
C(1)	51 989(15)	44 066(29)	71 917(21)
C(2)	45 654(15)	44 079(31)	74 158(21)
N(3)	42 988(10)	31 024(24)	74 487(15)
C(4)	38 939(12)	26 704(32)	64 496(19)
C(5)	42 440(11)	21 268(25)	57 090(18)
C(5a)	47 408(11)	30 637(25)	55 348(17)
N(6)	50 575(9)	25 476(22)	47 910(15)
C(6a)	56 820(11)	29 869(26)	50 732(20)
C(7)	61 106(13)	29 774(29)	44 985(21)
C(8)	67 229(14)	34 190(33)	49 681(24)
C(9)	69 022(14)	38 312(34)	59 545(26)
C(10)	64 693(14)	38 430(32)	65 352(23)
C(10a)	58 529(12)	34 249(26)	60 867(19)
C(10b)	52 959(12)	32 867(26)	65 133(18)
C(11)	37 559(11)	17 161(27)	47 279(18)
C(12)	33 843(11)	26 211(31)	40 665(18)
C(13)	29 176(13)	22 260(40)	32 031(21)
C(14)	28 193(16)	9 322(43)	29 872(23)
C(15)	31 828(18)	329(37)	36 243(27)
C(16)	36 505(15)	4 175(31)	44 882(23)
C(1′)	−3 213(12)	5 349(27)	70 794(20)
C(2′)	3 235(14)	5 318(28)	78 832(20)
N(3′)	5 920(9)	18 361(21)	81 234(14)
C(4′)	9 989(10)	22 138(26)	74 828(17)
C(5′)	6 457(10)	27 351(22)	64 140(16)
C(5a′)	1 310(10)	18 222(23)	58 044(16)
N(6′)	−1 916(8)	23 472(22)	47 842(14)
C(6a′)	−8 269(11)	19 686(26)	45 292(19)
C(7′)	−12 727(14)	20 294(32)	35 776(22)
C(8′)	−18 945(14)	16 727(37)	35 445(26)
C(9′)	−20 592(13)	12 758(34)	43 869(28)
C(10′)	−16 062(12)	12 072(30)	53 295(23)
C(10a′)	−9 870(11)	15 496(25)	53 970(19)
C(10b′)	−4 131(10)	16 413(24)	63 064(17)
C(11′)	11 318(9)	31 085(22)	58 664(15)
C(12′)	15 206(10)	21 685(24)	56 053(17)
C(13′)	19 860(11)	25 344(29)	51 548(18)
C(14′)	20 731(11)	38 245(29)	49 625(19)
C(15′)	16 864(12)	47 593(27)	52 020(19)
C(16′)	12 236(11)	43 966(24)	56 543(17)

The asymmetric crystal unit comprises two crystallographically independent molecules of (5) which are related in the solid state as shown in the Figure. Corresponding bond lengths and

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† All compounds reported were obtained as racemic mixtures but for clarity only one enantiomer is shown.

Table 2. Interatomic distances (Å) and angles (°), with standard deviations [a] in parentheses

(a) Bond lengths

	Unprimed	Primed	Mean
C(1)-C(2)	1.509(5)	1.526(4)	1.518(5)
C(1)-C(10b)	1.526(4)	1.521(4)	1.524(4)
C(2)-N(3)	1.469(4)	1.463(4)	1.466(4)
N(3)-C(4)	1.465(3)	1.466(3)	1.466(3)
C(4)-C(5)	1.534(4)	1.536(3)	1.532(4)
C(5)-C(5a)	1.524(4)	1.522(3)	1.521(4)
C(5)-C(11)	1.516(3)	1.514(3)	1.515(3)
C(5a)-N(6)	1.478(3)	1.469(3)	1.474(3)
C(5a)-C(10b)	1.547(4)	1.547(3)	1.547(4)
C(6)-C(6a)	1.393(3)	1.397(3)	1.395(3)
C(6a)-C(7)	1.384(4)	1.385(4)	1.385(4)
C(6a)-C(10a)	1.395(4)	1.390(4)	1.393(4)
C(7)-C(8)	1.394(4)	1.405(5)	1.400(5)
C(8)-C(9)	1.353(5)	1.356(5)	1.355(5)
C(9)-C(10)	1.398(4)	1.384(5)	1.391(5)
C(10)-C(10a)	1.389(4)	1.385(4)	1.387(4)
C(10a)-C(10b)	1.503(4)	1.499(3)	1.502(4)
C(11)-C(12)	1.387(4)	1.400(3)	1.394(4)
C(11)-C(16)	1.377(4)	1.381(3)	1.379(4)
C(12)-C(13)	1.383(4)	1.385(3)	1.384(4)
C(13)-C(14)	1.365(6)	1.374(4)	1.370(6)
C(14)-C(15)	1.360(5)	1.381(4)	1.371(5)
C(15)-C(16)	1.381(5)	1.381(4)	1.381(5)

(b) Bond angles

	Unprimed	Primed	Mean
C(2)-C(1)-C(10b)	114.7(2)	114.2(2)	114.5(2)
C(1)-C(2)-N(3)	113.9(3)	113.1(2)	113.5(3)
C(2)-N(3)-C(4)	113.1(2)	112.7(2)	112.9(2)
N(3)-C(4)-C(5)	115.5(2)	115.0(2)	115.3(2)
C(4)-C(5)-C(5a)	112,8(2)	113.5(2)	113.2(2)
C(4)-C(5)-C(11)	108.4(2)	108.2(2)	108.3(2)
C(5a)-C(5)-C(11)	113.6(2)	114.2(2)	113.9(2)
C(5)-C(5a)-N(6)	112.2(2)	111.9(2)	112.1(2)
C(5)-C(5a)-C(10b)	112.8(2)	112.6(2)	112.7(2)
N(6)-C(5a)-C(10b)	103.3(2)	103.4(2)	103.4(2)
C(5a)-N(6)-C(6a)	108.4(2)	108.3(2)	108.4(2)
N(6)-C(6a)-C(7)	128.3(2)	127.9(2)	128.1(2)
N(6)-C(6a)-C(10a)	110.2(2)	110.6(2)	110.4(2)
C(7)-C(6a)-C(10a)	121.5(2)	121.5(2)	121.5(2)
C(6a)-C(7)-C(8)	117.7(3)	116.4(3)	117.1(3)
C(7)-C(8)-C(9)	121.9(3)	122.8(3)	122.4(3)
C(8)-C(9)-C(10)	120.4(3)	120.0(3)	120.2(3)
C(9)-C(10)-C(10a)	119.1(3)	119.1(3)	119.1(3)
C(6a)-C(10a)-C(10)	119.3(2)	120.2(2)	119.8(2)
C(6a)-C(10a)-C(10b)	109.1(2)	108.8(2)	109.0(2)
C(10)-C(10a)-C(10b)	131.5(2)	130.9(2)	131.2(2)
C(1)-C(10b)-C(5a)	114.8(2)	115.2(2)	115.0(2)
C(1)-C(10b)-C(10a)	115.5(2)	116.0(2)	115.8(2)
C(5a)-C(10b)-C(10a)	102.4(2)	102.8(2)	102.6(2)
C(5)-C(11)-C(12)	121.6(2)	121.1(2)	121.4(2)
C(5)-C(11)-C(16)	120.5(2)	120.7(2)	120.6(2)
C(12)-C(11)-C(16)	117.8(2)	118.2(2)	118.0(2)
C(11)-C(12)-C(13)	120.8(3)	120.4(2)	120.6(3)
C(12)-C(13)-C(14)	120.2(3)	120.4(2)	120.3(3)
C(13)-C(14)-C(15)	119.7(3)	119.8(2)	119.8(3)
C(14)-C(15)-C(16)	120.6(4)	119.9(3)	120.3(4)
C(11)-C(16)-C(15)	120.9(3)	121.4(2)	121.2(3)

[a] The standard deviation quoted for the means is the larger of those for the individual values.

Table 3. Endocyclic torsion angles (°) in (5); $\sigma \pm 0.2$—$0.3°$

	Unprimed	Primed	Mean [a]
(a) In the five-membered ring			
C(10b)-C(5a)-N(6)-C(6a)	−25.4	−24.3	−24.9
C(5a)-N(6)-C(6a)-C(10a)	17.1	16.3	16.7
N(6)-C(6a)-C(10a)-C(10b)	−0.9	−0.6	−0.8
C(6a)-C(10a)-C(10b)-C(5a)	−14.5	−14.2	−14.4
C(10a)-C(10b)-C(5a)-N(6)	23.4	22.7	23.1
(b) In the seven-membered ring			
C(10b)-C(1)-C(2)-N(3) [$\omega_{1,2}$]	30.8	30.2	30.5
C(1)-C(2)-N(3)-C(4) [$\omega_{2,3}$]	−90.6	−91.6	−91.1
C(2)-N(3)-C(4)-C(5) [$\omega_{3,4}$]	80.6	81.8	81.2
N(3)-C(4)-C(5)-C(5a) [$\omega_{4,5}$]	−54.0	−53.5	−53.8
C(4)-C(5)-C(5a)-C(10b) [$\omega_{5,5a}$]	66.0	64.6	65.3
C(5)-C(5a)-C(10b)-C(1) [$\omega_{5a,10b}$]	−89.4	−89.2	−89.4
C(5a)-C(10b)-C(1)-C(2) [$\omega_{10b,1}$]	48.0	49.3	48.7

[a] $\Delta C_2 = |\omega_{1,2} - \omega_{10b,1}| + |\omega_{2,3} - \omega_{5a,10b}| + |\omega_{3,4} - \omega_{5,5a}| = 35.8°$; $\Delta C_s = |\omega_{1,2}| + |\omega_{2,3} + \omega_{10b,1}| + |\omega_{3,4} + \omega_{5a,10b}| + |\omega_{4,5} + \omega_{5,5a}| = 92.6°$.

Table 4. Equations of least-squares planes through selected groups of atoms, in the form $PX + QY + RZ - S = 0$,[a] with, in square brackets, displacements (Å) of atoms from these planes

Plane (i) C(6a)—C(10a)

$$-0.1914X + 0.9308Y - 0.3115Z + 1.2195 = 0$$

[C(5a) 0.313, N(6) −0.067, C(6a) −0.004, C(7) −0.002, C(8) 0.006, C(9) −0.003, C(10) −0.003, C(10a) 0.006, C(10b) −0.033]

Plane (ii) C(11)—C(16)

$$0.8464X + 0.0293Y - 0.5318Z - 2.2414 = 0$$

[C(4) −1.471, C(5) −0.082, C(5a) 1.049, N(6) 2.380, C(11) −0.005, C(12) 0.003, C(13) 0.000, C(14) −0.002, C(15) 0.000, C(16) 0.004, N(3^{I}) 3.331, H(3^{I}) 2.514]

Plane (i′) C(6a′)—C(10a′)

$$0.2647X - 0.9457Y - 0.1888Z + 3.960 = 0$$

[C(5a′) 0.250, N(6′) −0.094, C(6a′) −0.007, C(7′) 0.005, C(8′) 0.000, C(9′) −0.003, C(10′) 0.000, C(10a′) 0.005, C(10b′) −0 066]

Plane (ii′) C(11′)—C(16′)

$$-0.4127X - 0.0970Y - 0.9057Z + 7.323 = 0$$

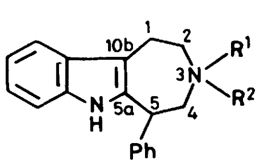
[C(4′) −1.445, C(5′) −0.085, C(5a′) 1.096, N(6′) 2.378, C(11′) −0.005, C(12′) 0.003, C(13′) 0.004, C(14′) −0.008, C(15′) 0.006, C(16′) 0.000, N($3'^{I}$) 3.281, H($3'^{I}$) 2.471]

[a] Cartesian co-ordinates (X, Y, Z) are related to the fractional atomic co-ordinates (x, y, z) in Table 1 by the transformations: $X = xa + zc\cos\beta$, $Y = yb$, $Z = zc\sin\beta$; Roman numeral superscript I refers to equivalent position $x, \frac{1}{2} - y, -\frac{1}{2} + z$.

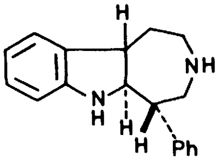
angles within the molecules are in excellent agreement and so the following discussions of the molecular geometry will refer only to the mean values. Bond lengths are close to expected values save for C(8)-C(9) in the fused tricyclic system and those involving C(14) of the phenyl substituent where the apparent foreshortening may be ascribed to lack of correction for the effects of thermal motion.

The five-membered ring lies close to an envelope form with C(5a) as the out-of-plane atom. In the azacycloheptane ring, the mean bond angle is 113.9°, and the ring approximates more closely to a C_2-twist-chair form with C(1) as axis atom than to a C_s-chair form with C(5) and the mid-point of the C(1)-C(2) bond defining the symmetry plane (Table 3). Rotation from an ideal twist-chair form towards a chair form in (5) serves to alleviate repulsive non-bonded transannular interactions between hydrogen atoms bonded to C(2) and

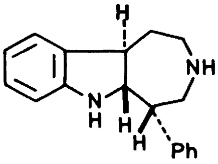
C(5a). Atoms of the phenyl substituent do not depart significantly from coplanarity (Table 4), but C(5) is displaced by 0.083 Å from the least-squares plane through the ring atoms, presumably to optimise the geometry required for the intermolecular N–H · · · phenyl hydrogen-bonded interactions in the crystal. The similar values assumed by torsion angles C(4)–C(5)–C(11)–C(12) and C(5a)–C(5)–C(11)–C(12) at 66.7 and −60.2°, respectively, indicate that the phenyl substituent almost eclipses the hydrogen atom at C(5).



(1) R^1 = H, R^2 = lone pair
(2) R^1 = COPh, R^2 = lone pair
(3) R^1 = CH_2Ph, R^2 = lone pair
(4) R^1 = CH_2Ph, R^2 = BH_3



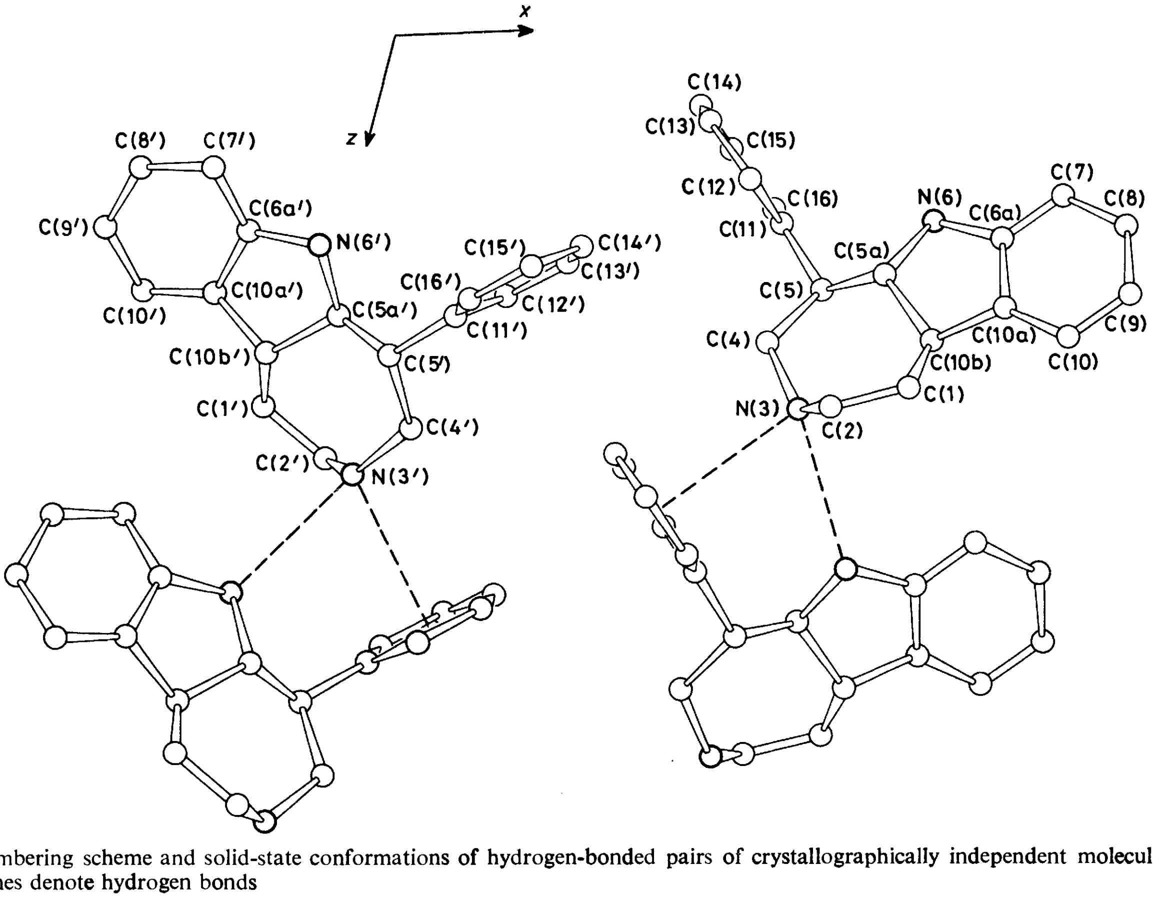
(5)



(6)

Inspection of molecular models reveals that adoption by (6) of an azacycloheptane ring conformation approximating to a twist-chair form with C(10) as the axis atom orients H(5) and H(5a) such that a $J_{5,5a}$ coupling constant of the observed magnitude (6.5 Hz) would result. In addition, consistent with the solution n.m.r. spectral data, rotation of the phenyl substituent about the C(5)–C(11) bond in such a form would be severely restricted by steric interactions involving the phenyl *ortho*-hydrogen atom at C(12) [or symmetry-equivalent C(16)] and atoms of the five-membered ring.

The crystallographically independent molecules of (5) are associated in an almost identical manner in the solid state by N(6)–H(6) · · · N(3) hydrogen bonds [N(6) · · · N(3) 3.216, N(6′) · · · N(3′) 3.310 Å] between molecules related by the *c*-glide plane to produce strands extending along the *c*-direction (Figure). These molecules are further associated by N–H · · · phenyl hydrogen-bonded interactions in which the N(3)–H(3) bond vector lies close to the six-fold axis of the phenyl substituent [N(3) · · · C(Ph) 3.453—3.803 Å, N(3′) · · · C(Ph) 3.551—3.558 Å] with H(3) and H(3′) displaced by 2.51 and 2.47 Å, respectively, from the phenyl ring plane. Since N–H distances derived from *X*-ray studies are generally shorter than those from neutron diffractions studies, calculated positions for these hydrogen atoms were evaluated by use of an N–H bond length of 1.0 Å and tetrahedral geometry at nitrogen. With these calculated positions, the perpendicular distances of H(3) and H(3′) from the phenyl ring planes decrease to 2.34 and 2.30 Å, respectively. These distances are *ca.* 0.2 Å longer than the corresponding values associated with O–H · · · phenyl hydrogen-bonded inter-



Atom numbering scheme and solid-state conformations of hydrogen-bonded pairs of crystallographically independent molecules of (5); broken lines denote hydrogen bonds actions and thus imply weaker interactions, a not unexpected result in view of the lower electronegativity of nitrogen, and the difference is similar to those of *ca.* 0.2—0.3 Å between O–H ··· N and N–H ··· N and between O–H ··· O and N–H ··· O hydrogen bonds.[3,4]

Experimental

M.p.s were determined with a Thomas Hoover m.p. apparatus and are uncorrected. ^{1}H N.m.r. spectra were recorded on a Varian XL-100 spectrometer and are reported in p.p.m. downfield from SiMe$_4$ as internal standard. I.r. spectra were recorded on a Perkin-Elmer 180 spectrophotometer, and mass spectra were determined with a Varian MAT CH5 instrument. Microanalyses were performed by the Physical Analytical Services Department of Schering-Plough Corporation.

*3-Benzoyl-5-phenyl-*1,2,3,4,5,6-*hexahydroazepino*[4,5-b]-*indole* (2).—Benzoyl chloride (4.2 g, 29 mmol) was added dropwise to a stirred two-phase system containing 5-phenyl-1,2,3,4,5,6-hexahydroazepino[4,5-*b*]indole (1) (6.8 g, 26 mmol), ethyl acetate (500 ml), and 2.5M-sodium hydroxide solution (250 ml). After the addition, stirring was continued for 15 min and the organic layer was then separated. The aqueous phase was extracted with ethyl acetate (200 ml), and the combined organic solutions were washed with water (200 ml), dried (MgSO$_4$), and evaporated *in vacuo*. The residue was crystallized from ethanol to give *prisms* (8.0 g, 84%), m.p. 179—180 °C; δ (^{2}H$_6$-DMSO) 3.0—5.0 (m, 7 H), 6.7—7.7 (m, 14 H), and 10.58 (s, NH); *m/e* 366 (80%) (Found: C, 82.1; H, 5.95; N, 7.45. $C_{25}H_{22}N_2O$ requires C, 81.95; H, 6.05; N, 7.65%).

*3-Benzyl-5-phenyl-*1,2,3,4,5,6-*hexahydroazepino*[4,5-b]-*indole* (3).—A solution of (2) (5.3 g, 14.5 mmol) in tetrahydrofuran (THF) (65 ml) was added to a stirred suspension of lithium aluminium hydride (0.65 g) in THF (750 ml) over 15 min. The mixture was boiled under reflux for 1 h, and then allowed to cool. Excess of lithium aluminium hydride was decomposed by the addition of water (1 ml) followed by 20% sodium hydroxide solution (0.5 ml). The precipitate was removed by filtration and the filtrate was evaporated *in vacuo*. The residue was crystallized from ethanol to give *prisms* (4.2 g, 90%), m.p. 121—123 °C; δ (CDCl$_3$) 3.9—3.2 (m, 6 H), 3.82 (s, benzylic CH$_2$), 4.38 [t, H(5)], 6.9—7.4 (m, 9 H), and 7.52 (s, NH); *m/e* 352 (84%) (Found: C, 84.75; H, 7.0; N, 7.9. $C_{25}H_{24}N_2$ requires C, 85.2; H, 6.85; N, 7.95%).

trans-*Reduction of* (3).—Compound (3) (7.2 g, 20.4 mmol) was dissolved in THF (100 ml) and 25 ml of a 1.0M solution of borane in THF was added dropwise. The solution was carefully diluted with water (900 ml), and the aqueous phase was separated from the heavier oily phase by decantation. The oil was dissolved in ethanol (200 ml) containing 6M aqueous hydrochloric acid (50 ml) and the solution was then boiled under reflux for 2 h. Most of the ethanol was evaporated *in vacuo* and the residue was treated with water (300 ml) and basified with 50% sodium hydroxide solution. The mixture was extracted with ethyl acetate (2 × 300 ml) and the combined extracts were washed with water, dried (Na$_2$SO$_4$), and evaporated *in vacuo*. The residual oil consisted of two major components which were separated by chromatography on silica gel (400 g) using chloroform as eluant. Both compounds were obtained as oils and were debenzylated by hydrogenation at 50 lb in^{-2} in ethanol (75 ml) containing concentrated hydrochloric acid (5 ml) using 10% Pd–C (0.3 g). After 4 h the catalyst was removed by filtration. Most of the ethanol was evaporated *in vacuo* and the residue was treated with 2M-sodium hydroxide solution (300 ml). The mixture was extracted with ethyl acetate (2 × 100 ml), and the combined extracts were washed with water (200 ml), dried (Na$_2$SO$_4$), and evaporated *in vacuo*. Isomer (5) (1.1 g, 22%) was obtained as *prisms*, m.p. 134—135 °C (from acetonitrile); δ (CDCl$_3$) 1.80 (m, 2 H), 2.50 (m, 1 H), 2.80—3.60 (m, 7 H), 4.03 (t, 1 H), 6.46 (dd, 1 H), 6.72 (dd, 1 H), 7.00 (t, 2 H), and 7.24 (s, 5 H); $\nu_{max.}$ (Nujol) 3 300 cm^{-1}; *m/e* 264 (6%) (Found: C, 81.45; H, 7.6; N, 10.45. $C_{18}H_{20}N_2$ requires C, 81.75; H, 7.65; N, 10.6%). Isomer (6) (1.1 g, 22%) crystallized from 1-chlorobutane as *prisms*, m.p. 145—148 °C; δ (CDCl$_3$) 1.80 (m, 1 H), 2.40 (m, 2 H), 2.80—3.90 (m, 3 H), 4.40 (dd, 1 H), 6.46 (dd, 1 H), 6.72 (dd, 1 H), 7.00 (t, 2 H), and 7.10—7.30 (m, 5 H): $\nu_{max.}$ (Nujol) 3 330 and 3 260 cm^{-1}; *m/e* 264 (6%) (Found: C, 81.7; H, 7.6; N, 10.7. $C_{18}H_{20}N_2$ requires C, 81.75; H, 7.65; N, 10.6%).

Crystal Data.—$C_{18}H_{20}N_2$ (5), $M = 264.4$. Monoclinic, $a = 22.005(10)$, $b = 10.270(5)$, $c = 13.577(7)$ Å, $\beta = 106.42(1)°$, $U = 2\,943$ Å^3, $Z = 8$, $D_c = 1.193$ g cm^{-3}. Cu-K_α radiation, $\lambda = 1.5418$ Å; μ(Cu-K_α) = 5.5 cm^{-1}. Space group $P2_1/c$ (C_2^5h) uniquely from the systematic absences: $0k0$ when $k \neq 2n$, $h0l$ when $l \neq 2n$.

Crystallographic Measurements.—A crystal of dimensions *ca.* 0.20 × 0.26 × 0.80 mm was oriented on the end of a thin glass fibre with its long dimension approximately parallel to the fibre axis. Oscillation and Weissenberg photographs, taken with Cu-K_α radiation, provided preliminary unit-cell parameters and space-group information. The crystal was then transferred to an Enraf–Nonius CAD-3 automated diffractometer (Ni-filtered Cu-K_α radiation) where the intensities of all unique reflections with $\theta < 67°$ were recorded by means of the θ–2θ scanning procedure as described in detail elsewhere.[5] Refined unit-cell parameters were evaluated by least-squares treatment of the diffractometer setting angles for 40 high-order reflections widely separated in reciprocal space. A total of 3 293 independent reflections with $I > 2.0\sigma(I)$ [$\sigma^2(I)$ = scan count + total background count] were retained for the structure analysis and corrected for the usual Lorentz polarization effects. No absorption corrections were necessary.

Structure Analysis.—The structure was solved by direct methods by use of MULTAN76[6] involving the 350 largest $|E|$ values. All non-hydrogen atoms were located in an E-map computed by use of that set of phases which gave the highest combined figure of merit. Full-matrix least-squares adjustment of non-hydrogen atom positional and isotropic thermal parameters reduced R smoothly to 0.144 from that of 0.276 for the initial model. Hydrogen atoms were then located in a difference Fourier synthesis. Further least-squares iterations, during which hydrogen atom positional and isotropic thermal parameters were varied in addition to non-hydrogen atom positional and anisotropic thermal parameters, converged to R 0.051. Final atomic positional parameters for carbon and nitrogen are in Table 1. Anisotropic thermal parameters for the non-hydrogen atoms, positional and isotropic thermal parameters for the hydrogen atoms, and a list of torsion angles have been deposited along with a Table of observed and calculated structure amplitudes in Supplementary Publication No. SUP 23714 (30 pp.).*

For all structure-factor calculations, atomic scattering factors for carbon and nitrogen were taken from ref. 7, and for hydrogen from ref. 8. In the least-squares iterations, $\Sigma w\Delta^2$ ($\Delta = ||F_o| - |F_c||$) was minimized with weights, w,

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* For details of Supplementary Publications see Instructions for Authors, *J. Chem. Soc., Perkin Trans. 2*, 1983, Issue 1, p. xvii.

assigned according to the scheme: $\sqrt{w} = 1$ for $|F_o| \leqslant 18.5$ and $\sqrt{w} = 18.5/|F_o|$ for $|F_o| > 18.5$.

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Received 5th October 1982; *Paper* 2/1714