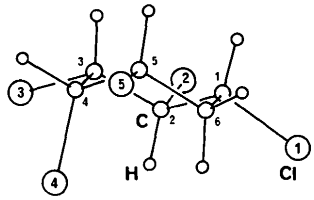

Fig. 1. Perspective view of the molecule.

that the Cl atoms adopt the (i) conformation in the (II) crystal obtained from an *n*-hexane solution (Nakai, Shiro & Hamada, 1978).

Bond lengths, bond angles and torsion angles are given in Fig. 2. The mean e.s.d. values are 0·003 Å for Cl—C, 0·005 Å for C—C, and 0·2° for angles. The mean corrections of bond lengths for the effects of rigid-body libration are 0·010 (ranging from 0·009 to 0·011 Å) for Cl—C and 0·009 (0·008–0·010 Å) for C—C. The shortest intramolecular Cl—Cl distance is 3·228 Å between Cl(3) and Cl(4).

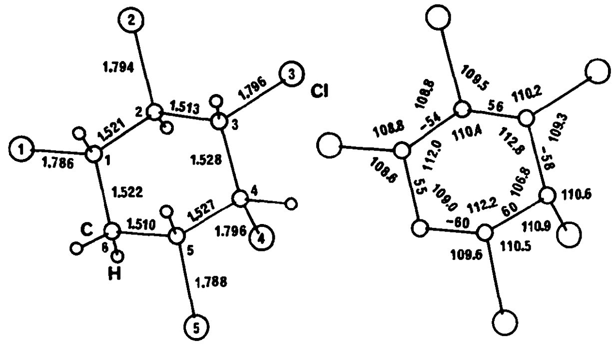

Fig. 2. Bond lengths (Å), bond angles and torsion angles (°).

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Ethyl 3-Methyl-4-oxo-1-phenyl-2-phenylimino-1,3,7,8-tetraazaspiro[4.5]deca-6,9-diene-10-carboxylate–0·5 Benzene

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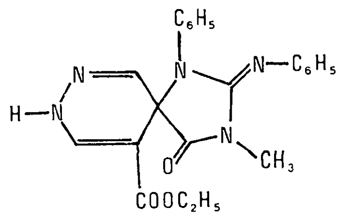
Abstract. $C_{22}H_{21}N_5O_3.\frac{1}{2}C_6H_6$, monoclinic, $P2_1/c$, a = 9·388 (2), b = 28·140 (3), c = 9·017 (2) Å, β = 103·4 (1)°, Z = 4, D_x = 1·27 g cm^{-3}, μ(Mo $K\alpha$) = 0·918 cm^{-1}. Counter technique, direct methods, full-matrix least-squares refinement. R = 0·056 for 1304 observed reflexions measured at room temperature (20 °C). The main geometrical molecular features and the thermal motion were analysed.

Introduction. The chemical preparation of the title compound and other spiro compounds has been described by Adembri, Chimichi, De Sio, Nesi & Scotton (1976). The reaction between the diethylic ester of 4,5-pyridazinedicarboxylic acid with 1,3-diphenylguanidine and NaOH in the presence of tetrahydrofuran at room temperature gives, with good yield, a product (I) of elementary composition $C_{21}H_{19}N_5O_3$. Spectroscopic data and chemical evidence suggested a spiranic structure for this compound (Nesi, 1977). An early attempt to determine the molecular geometry by crystal-structure analysis failed because the crystals decompose too rapidly to allow the collection of reliable data. However, the unit-cell dimensions were determined to be: a = 17·87 (1), b = 19·08 (1), c = 12·19 (1) Å, and β = 100·7 (5)°; space group $P2_1/a$, Z = 8. Eventually a stable mono-methyl derivative $C_{22}H_{21}N_5O_3$ (II) was prepared by treating (I) with diazomethane. Crystals of (II), obtained from benzene as white needles with m.p. 213–215 °C, were kindly supplied by Professor R. Nesi. Intensity data were recorded on a PW 1100 automatic diffractometer (ω–2θ scan, $\theta \leq 20°$, graphite monochromator, Mo $K\alpha$ radiation, λ = 0·7107 Å). The unit-cell dimensions were determined by a least-squares procedure carried out on a selected group of reflexions. 4750 reflexions of the type *hkl* and $\bar{h}k\bar{l}$ were collected and averaged. 1304 out of the 2375 independent and averaged intensities were considered observed [$I \geq 3\sigma(I)$] and used in the structure determination. Intensities were corrected for Lorentz and polarization factors but not for absorption [μ(Mo $K\alpha$) = 0·918 cm^{-1}].

An initial set of signs for the structure factors was obtained with the aid of the program *LSAM* (Main, Woolfson & Germain, 1972). An *E* map calculated with this set of signs gave initial coordinates for most of the atoms in the molecule. The model was completed by means of successive Fourier syntheses that also revealed the presence of a solvent molecule (benzene) located in a special position on the inversion centre at the origin of the unit cell. After some isotropic cycles of least-squares refinement, H atoms were generated at the expected positions, and their contribution to the calculated structure factors was taken into account. The atomic positions were then refined anisotropically for the non-hydrogen atoms by a full-matrix least-squares method, the quantity minimized being $\sum w(\Delta F)^2$ with weights chosen as $w = 4F_o^2/\sigma^2(F_o^2)$. The atomic scattering factors were taken from *International Tables for X-ray Crystallography* (1962).

The final reliability index $R = \sum|\Delta F|/\sum|F_o|$ was 0·056. Atomic coordinates are listed in Table 1.*

Discussion. A schematic drawing of the molecule showing atomic numbering, bond lengths, bond angles and some relevant torsion angles, is given in Fig. 1. The tetrahedral C(1) atom has distances and angles to neighbouring atoms similar to those found in related spiro compounds (Smith-Verdier, Florencio & Garcia-Blanco, 1977; Bordeaux & Lajzérowicz-Bonneteau, 1974). The bond lengths and angles in the rest of the six-membered ring are quite normal, as are those found in the hydantoin ring (Smith-Verdier *et al.*, 1977). Interatomic distances are consistent with the following structural formula:



Some least-squares planes were calculated and are reported in Table 2. These show that, in our case, the five-membered ring is slightly puckered, with the shape roughly approximating the *m* symmetry of the envelope form. The six- (and five-) membered rings do not deviate considerably from planarity. Bond lengths and angles are normal in the phenyl rings and the corresponding least-squares planes (Table 2) show that these groups are planar within the experimental error.

* Lists of structure factors, anisotropic thermal parameters and calculated hydrogen-atom positions have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 33831 (5 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Atomic positional parameters of non-hydrogen atoms with their e.s.d.'s in parentheses*

	x	*y*	*z*
O(1)	−0·6291 (5)	0·1857 (2)	0·3460 (5)
O(2)	−0·4282 (5)	0·0404 (2)	0·2377 (5)
O(3)	−0·4691 (6)	0·0777 (2)	0·4468 (5)
N(1)	−0·3377 (6)	0·1660 (2)	0·0538 (5)
N(2)	−0·3246 (6)	0·2087 (2)	0·1297 (6)
N(3)	−0·2561 (6)	0·1630 (2)	0·5014 (5)
N(4)	−0·4642 (6)	0·1777 (2)	0·5782 (5)
N(5)	−0·2518 (6)	0·1718 (2)	0·7700 (5)
C(1)	−0·3735 (7)	0·1661 (2)	0·3572 (6)
C(2)	−0·3910 (6)	0·1230 (2)	0·2611 (6)
C(3)	−0·3689 (7)	0·1270 (3)	0·1181 (7)
C(4)	−0·3427 (7)	0·2086 (2)	0·2677 (7)
C(5)	−0·3142 (7)	0·1701 (2)	0·6282 (7)
C(6)	−0·5074 (8)	0·1770 (2)	0·4211 (8)
C(7)	−0·5659 (7)	0·1872 (3)	0·6776 (7)
C(8)	−0·4335 (8)	0·0791 (3)	0·3258 (9)
C(9)	−0·4712 (9)	−0·0045 (3)	0·2989 (9)
C(10)	−0·4515 (12)	−0·0433 (3)	0·1955 (10)
C(11)	−0·1275 (7)	0·1361 (3)	0·5032 (7)
C(12)	−0·1018 (9)	0·0940 (3)	0·5797 (8)
C(13)	0·0250 (12)	0·0686 (3)	0·5735 (9)
C(14)	0·1203 (10)	0·0853 (4)	0·4943 (10)
C(15)	0·0939 (9)	0·1275 (4)	0·4192 (9)
C(16)	−0·0313 (8)	0·1543 (3)	0·4216 (8)
C(17)	−0·0975 (8)	0·1781 (3)	0·8197 (7)
C(18)	−0·0189 (9)	0·1500 (3)	0·9378 (8)
C(19)	0·1292 (10)	0·1595 (4)	0·9973 (9)
C(20)	0·1980 (9)	0·1956 (4)	0·9417 (9)
C(21)	0·1215 (9)	0·2237 (3)	0·8234 (9)
C(22)	−0·0263 (8)	0·2141 (3)	0·7621 (7)
C(23)	−0·0503 (49)	0·0247 (8)	0·1045 (22)
C(24)	−0·1435 (16)	−0·0016 (13)	−0·0039 (43)
C(25)	0·0884 (42)	0·0249 (7)	0·1079 (22)

Table 2. *Deviations* (Å) *of atoms from least-squares planes*

Plane *A*: C(1), C(5), C(6), N(3), N(4)

C(1)	−0·008	C(5)	0·004	C(6)	0·012
N(3)	0·003	N(4)	−0·010		

Plane *B*: C(1), C(2), C(3), C(4), N(1), N(2)

C(1)	−0·023	C(2)	0·019	C(3)	−0·001
C(4)	0·014	N(1)	−0·012	N(2)	0·004

Plane *C*: C(11), C(12), C(13), C(14), C(15), C(16)

C(11)	−0·003	C(12)	0·002	C(13)	0·001
C(14)	−0·002	C(15)	0·001	C(16)	0·002

Plane *D*: C(17), C(18), C(19), C(20), C(21), C(22)

C(17)	0·009	C(18)	−0·002	C(19)	−0·003
C(20)	0·002	C(21)	0·004	C(22)	−0·010

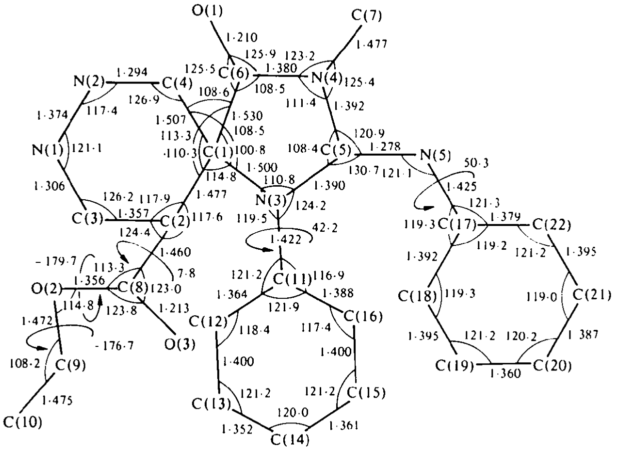


Fig. 1. Atomic numbering, bond distances (Å) and bond angles (°) for non-hydrogen atoms. E.s.d.'s are in the range 0·007–0·014 Å for distances and 0·3–0·5° for angles. Some torsion angles of interest are also given.

No special features were found in the molecular packing, all the intermolecular distances being in the normal range. An *ORTEP* drawing (Fig. 2) shows the stereochemistry of the molecule.

The anisotropic temperature factors of the non-hydrogen atoms were analysed by the method of Schoemaker & Trueblood (1968).

As expected, the whole molecule does not behave as a rigid body but a satisfactory fit was obtained for the phenyl rings and the central ring skeleton considered separately, excluding the side chain. For these fragments the r.m.s. values ΔU_{ij} are of the same order as the average value (0·0043 Å^2) of $\sigma(U_{ij})$.

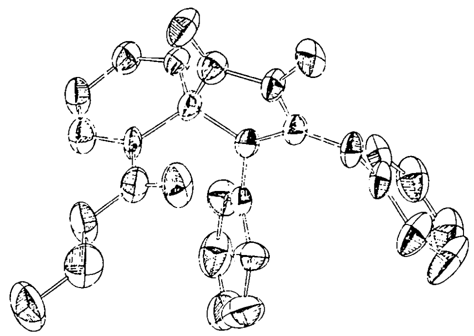


Fig. 2. An *ORTEP* view of the main molecule. Ellipsoids of the non-hydrogen atoms are at 50% probability level.

Bond lengths have not been corrected for librational shortening, the corrections being smaller than the corresponding e.s.d.'s.

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Diethyl 2,6-Dimethyl-4-phenyl-1,4-dihydro-3,5-pyridinedicarboxylate

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Abstract. $C_{19}H_{23}NO_4$, monoclinic, $P2_1/c$, $a = 9{\cdot}73$ (1), $b = 7{\cdot}39$ (1), $c = 24{\cdot}32$ (2) Å, $\beta = 92{\cdot}62$ (5)°, $V = 1747$ Å^3, $Z = 4$, $D_m = 1{\cdot}26$, $D_x = 1{\cdot}25$ g cm^{-3}, $F(000) = 704$. The structure was solved by direct methods and refined by full-matrix least squares to a final $R = 0{\cdot}077$ for 4014 reflections. The dihydropyridine ring adopts a flat-boat conformation. The phenyl ring is approximately perpendicular to the dihydropyridine ring.

Introduction. This study was undertaken to establish the conformational features of the title compound (I). The crystals were yellow and needle-like and were elongated along the *b* axis. The space group was found

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