Fig. 3. Bond lengths (Å) and angles (°) for (I). The e.s.d.'s are 0·002–0·003 Å for bond lengths and 0·1–0·2° for bond angles.

of −170·5 and 35·2° with respect to the central ketone bridge.

The *anti* position of the pyrrole ring is a necessary consequence of the dimerization of (I) through N(1)—H···O(6) hydrogen bonds of length 2·87 Å between equivalent molecules related by a centre of symmetry. A similar interplanar angle of −167·3° and N—H···O distance of 2·87 Å is observed for that pyrrole ring in (II) which is involved in equivalent hydrogen bonding. Steric contact between the protons

on C(3) and C(2') of (I) must lead to twisting of the thiophene ring with respect to the trigonal coordination plane at C(6). In fact, an interplanar angle of 35·2° is observed for an H(3)···H(2') intramolecular distance of 2·55 Å. The degree of twist in (III) is less pronounced with interplanar angles of 20·9 and 20·1° for a very much shorter intramolecular H···H distance of 2·28 Å. Packing forces must presumably be responsible for this average difference of 14·7° between (I) and (III) in the conformation of the thiophene ring relative to the central ketone bridge.

The bond-length distribution is indicative of a significant mesomeric contribution from the dipolar structure (I). Thus, the N(1)—C(5) and C(2)—C(6) lengths, 1·345 (3) and 1·430 (3) Å, are much shorter than the 1·375 (3) and 1·477 (3) Å for N(1)—C(2) and C(6)—C(3').

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1,7-Diazaspiro[4.4]nonane-2,6-dione

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Abstract. $C_7H_{10}N_2O_2$, M_r = 142·16, monoclinic, $P2_1/c$, a = 9·601 (2), b = 7·815 (1), c = 9·498 (2) Å, β = 91·20 (3)°, Z = 4, V = 712·50 Å³, μ(Cu $K\alpha$) = 0·80 mm⁻¹. The envelope-shaped pyrrolidone rings are nearly perpendicular to each other. The crystal structure consists of infinite hydrogen-bonded layers linked together by normal van der Waals forces.

Introduction. This structure determination is part of a research programme to study the conformation of the pyrrolidone ring in some isomers of spiro-bis-pyrrolidones {*i.e.* diazaspiro[4.4]nonane-diones}, which show interesting chiroptical properties. The title compound was prepared at the Institute of Organic Chemistry, L. Eötvös University, Budapest (Kajtár, Hollóssi & Kinsky, 1975).

The colourless crystals, generally in the shape of hexagonal plates, were preliminarily examined using X-ray photographic methods, such as the Guinier,

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oscillation and De Jong–Bouman techniques. The unit-cell dimensions were refined by least-squares fitting of the cell parameters to powder data from a Guinier photograph, taken at 298 K with KCl ($a = 6.2930$ Å) as an internal standard. Three-dimensional diffraction data were collected from a single crystal, of approximate dimensions $0.28 \times 0.15 \times 0.05$ mm, using a computer-controlled PW 1100 diffractometer with graphite-monochromatized Cu $K\alpha$ radiation. The intensities of 1202 independent reflexions were measured, with $\theta \le 65°$. The net intensities were corrected for Lorentz and polarization effects.

The approximate positions of the non-hydrogen atoms were calculated by direct methods (*MULTAN*; Main, Lessinger, Woolfson, Germain & Declercq, 1974). 180 E values ($E \ge 1.50$) and 1708 triple relations among them were used to generate 16 trial phase sets. The H atom positions were calculated geometrically. The refinement was carried out by the full-matrix least-squares method, using the *SHELX* 76 (Sheldrick, 1976) program system. The positions of the non-hydrogen atoms were refined together with their anisotropic thermal parameters. The H atoms, each with the same isotropic temperature factor as the non-hydrogen atom to which it is bonded, had only their positional parameters refined. The final R value became 0.056 for 970 significant observed reflexions with $F_o >$

$5\sigma(F_o)$. The atomic scattering factors for H atoms were taken from Stewart, Davidson & Simpson (1965) and, for the non-hydrogen atoms, from Cromer & Mann (1968) and Cromer & Liberman (1970). The final atomic coordinates are listed in Tables 1 and 2.* The atomic labelling used is shown in Fig. 1.

Discussion. The pyrrolidone rings are non-planar (Fig. 2). Atoms C(4) and C(9) are significantly out of the best planes calculated for the other five atoms of each moiety (*cf.* Table 3), indicating envelope conformations for these rings. This can best be checked by the use of the pseudo-rotation parameter Δ (Altona, Geise &

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* Lists of structure factors, anisotropic thermal parameters, bond lengths and bond angles have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 34216 (9 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Fractional atomic coordinates* ($\times 10^4$) *for the non-hydrogen atoms, with estimated standard deviations in parentheses*

	x	y	z
N(1)	1442 (3)	5911 (3)	6785 (3)
C(2)	757 (3)	7393 (4)	6831 (3)
O(2)	−320 (2)	7641 (3)	7490 (3)
C(3)	1492 (3)	8664 (4)	5928 (4)
C(4)	2907 (3)	7850 (4)	5660 (4)
C(5)	2656 (3)	5922 (4)	5870 (3)
C(6)	3848 (3)	4969 (4)	6650 (3)
O(6)	4574 (2)	5550 (3)	7616 (2)
N(7)	3901 (3)	3395 (3)	6129 (3)
C(8)	2894 (4)	3049 (4)	4997 (4)
C(9)	2441 (3)	4836 (4)	4553 (3)

Table 2. *Fractional atomic coordinates* ($\times 10^3$) *for the hydrogen atoms, with estimated standard deviations in parentheses*

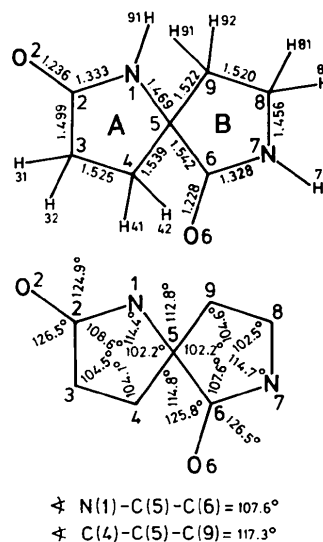|	x	y	z
H(1)	110 (4)	491 (5)	711 (4)
H(31)	98 (4)	872 (5)	510 (4)
H(32)	154 (4)	974 (5)	636 (4)
H(41)	312 (4)	816 (5)	476 (4)
H(42)	358 (4)	824 (5)	636 (4)
H(7)	442 (4)	258 (5)	655 (4)
H(81)	332 (4)	237 (5)	423 (4)
H(82)	210 (4)	245 (5)	540 (4)
H(91)	312 (4)	523 (5)	383 (4)
H(92)	151 (4)	487 (5)	420 (4)

$\neq$ N(1)–C(5)–C(6) = 107.6°
$\neq$ C(4)–C(5)–C(9) = 117.3°

Fig. 1. Intramolecular distances (Å) and bond angles (°) involving the non-hydrogen atoms of the molecule. The e.s.d.'s of the bond lengths are in the range 0.003–0.005 Å, and those of the bond angles are less than 0.3°.
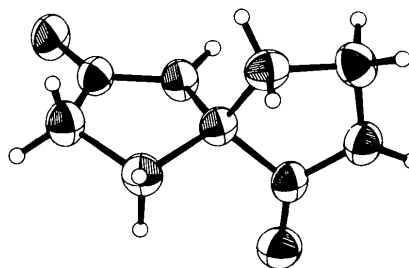

Fig. 2. View of the molecule. The non-hydrogen atoms are represented by their thermal ellipsoids, while the H atoms are drawn as spheres.

Table 3. Equations of the least-squares planes, and deviations (Å) of the atoms from the planes

The planes are expressed as $Ax + By + Cz = D$, where x, y and z are in Å relative to the axes a^*, b and c. The atoms indicated with asterisks were omitted from the calculations of the least-squares planes. A minus sign means that the atom lies between the plane and the origin.

Plane 1: ring A

$A =$	0·565				
$B =$	0·276	N(1)	0·018	C(3)	0·004
$C =$	0·778	O(2)	−0·016	C(4)*	0·376
$D =$	7·028	C(2)	0·010	C(5)	−0·017

The r.m.s. deviation from the plane of the atoms without asterisks is 0·014 Å.

Plane 2: ring B

$A =$	−0·654	C(5)	0·004	N(7)	0·005
$B =$	−0·319	O(6)	0·005	C(8)	−0·001
$C =$	0·686	C(6)	−0·014	C(9)*	−0·445
$D =$	0·639				

The r.m.s. deviation from the plane of the atoms without asterisks is 0·007 Å.

Fig. 3. Torsion angles (°). The e.s.d.'s are less than 0·4°.

Fig. 4. Stereoscopic packing diagram of the crystal structure (y is vertical, x is to the right and z is out of the paper).

Table 4. Intermolecular distances (Å) less than 3·6 Å between the non-hydrogen atoms

The estimated standard deviations are given in parentheses. Atom (2) is generated from the coordinates of Table 1 by using the unit-cell translation shown together with the following symmetry operations:

Superscript: None x,y,z; (i) $-x,-y,-z$; (ii) $-x, \frac{1}{2} + y, \frac{1}{2} - z$; (iii) $x, \frac{1}{2} - y, \frac{1}{2} + z$

		Translation along the axes of the unit cell			
(1)	(2)	a	b	c	
N(1)	O(2^{ii})	0	−1	1	2·863 (3)
O(6)	N(7^{ii})	1	0	1	2·904 (3)
O(2)	C(9^{i})	0	1	1	3·390 (4)
O(2)	C(8^{i})	0	1	1	3·425 (5)
O(2)	C(8^{ii})	0	0	1	3·486 (5)
C(4)	O(6^{iii})	0	1	−1	3·560 (4)
O(6)	C(9^{i})	1	1	1	3·578 (4)
C(4)	O(6^{ii})	1	0	1	3·580 (4)

Romers, 1968). The ideal values for the specific phase angle, $\varDelta$, are ±36° in the envelope (C_s) conformation and 0° in the half-chair or twist (C_2) conformation. The actual values for the present structure, calculated from the torsion angles, presented in Fig. 3, are −30·8 and −30·1° for the rings A and B, respectively. [The maximum torsion angles, φ_m, are 24·6° (ring A) and 28·9° (ring B).] It means that the rings have a 'near-envelope' conformation. The angle between the normals to the two least-squares planes is 86°.

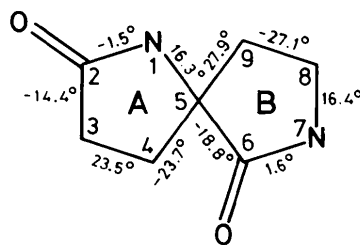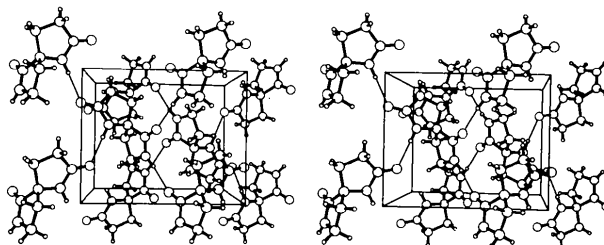The corresponding bond lengths and angles involving the non-hydrogen atoms in the two independent pyrrolidone rings (cf. Fig. 2) are compatible with each other and are in agreement with those found in other X-ray investigations of related compounds (Czugler, Kálmán & Kajtár, 1976, and references therein; Ealick & van der Helm, 1975; Ealick, Washecheck & van der Helm, 1976; Sheldrick & Borkenstein, 1978). The calculated C—H distances, ranging from 0·92 to 1·10 Å with e.s.d.'s of 0·03–0·04 Å, are compatible with X-ray-determined C—H bond lengths (Stewart, Davidson & Simpson, 1965).

The packing is illustrated in Fig. 4. The molecules, related by the symmetry operation $-x, \frac{1}{2} + y, \frac{1}{2} - z$, are linked together by hydrogen bonds to form infinite layers parallel to the xy plane. There are two different hydrogen bonds; one of them [N(1)···O(2) = 2·863 (3), N(1)—H(1) = 0·91 (4), H(1)···O(2) = 1·96 (4) Å, ∠N(1)—H(1)···O(2) = 171 (4)°] links together the A rings, while the other [N(7)···O(6) = 2·904 (3), N(7)—H(7) = 0·90 (4), H(7)···O(6) = 2·01 (4) Å, ∠N(7)—H(7)···O(6) = 173 (4)°] exists between the B rings. The shortest contact distances between these layers have the same magnitude as normal van der Waals interactions (cf. Table 4).

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(9S,11S)-9-Acetoxydihydrocostunolide (Herbolide A)

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Abstract. $C_{17}H_{24}O_4$, $M_r = 292.38$, monoclinic, $P2_1$, $a = 11.279$ (4), $b = 7.703$ (2), $c = 9.499$ (3) Å, $\beta = 93.61$ (2)°, $U = 823.7$ Å^3, $Z = 2$, $D_x = 1.179$ Mg m^{-3}, μ(Mo $K\alpha$) = 0.05 mm^{-1}. The structure was refined to $R = 0.059$ for 1468 unique reflexions; it provides confirmation of the structure deduced by chemical and spectroscopic methods.

Introduction. The title compound (I; trivial name herbolide A) is one of three sesquiterpene lactones isolated from the composite plant *Artemisia herba alba* (Segal, Sokoloff, Haran, Zaitschek & Lichtenberg, 1977). We have already reported the structure of herbolide B (Hull & Kennard, 1978). The crystal structure determination of (I) was undertaken (*a*) to confirm the structure earlier deduced from chemical and spectroscopic results (Segal *et al.*, 1977), in particular the position of the two double bonds; and (*b*) to allow calculation of the sign of the circular-dichroism couplet from exciton theory.

Large colourless crystals in the form of flat blocks were obtained from ethanol. Intensities were measured on a Syntex $P2_1$ diffractometer with monochromated Mo $K\alpha$ radiation and a crystal 0.75 × 0.75 × 0.25 mm. Cell dimensions were obtained by least-squares methods from 15 strong reflexions. Systematic absences $0k0$, k odd, indicated space group $P2_1$. 2138

(I)

reflexions were measured in the range $3° < 2\theta < 55°$; after application of Lp corrections, averaging equivalent reflexions gave 1468 unique reflexions with $F > 4\sigma(F)$.

The structure was solved by multisolution tangent refinement with *SHELX*. A starting set of seven reflexions (chosen from a convergence map) gave 2^7 phase permutations. The best E map showed all non-hydrogen atoms. Least-squares refinement proceeded to $R = 0.14$ (isotropic), 0.09 (anisotropic); a difference synthesis then revealed all non-methyl H atoms. In the final stages of refinement C—H distances and H—C—H angles were fixed at 1.08 Å and 109.5°, and common isotropic temperature factors were assigned to methyl and non-methyl H atoms. High H-atom temperature factors and slow convergence of the methyl groups probably indicate some rotational disorder, as would be expected for methyl groups attached to sp^2 C atoms. The final $R' = \sum w^{1/2}\Delta/\sum w^{1/2}|F_o|$ was 0.063, with a corresponding R of 0.059; the weighting scheme was

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