

Table 2. Selected interatomic distances (Å), angles (°) and hydrogen bonds (Å, °) with *e.s.d.*'s in parentheses

O(1)—C(1)	1.3751 (30)	C(7)—C(8)	1.3486 (21)
O(1)—C(4)	1.3715 (23)	C(7)—C(14)	1.4292 (22)
C(1)—C(2)	1.3327 (37)	C(8)—C(13)	1.4936 (35)
C(1)—C(5)	1.4266 (47)	C(8)—N(1)	1.3656 (21)
C(2)—C(3)	1.4237 (46)	C(9)—C(10)	1.3518 (21)
C(3)—C(4)	1.3416 (25)	C(9)—C(12)	1.4910 (29)
C(4)—C(6)	1.4962 (21)	C(9)—N(1)	1.3725 (23)
C(5)—N(2)	1.1377 (41)	C(10)—C(11)	1.4219 (25)
C(6)—C(7)	1.5190 (23)	C(11)—N(4)	1.1436 (28)
C(6)—C(10)	1.5192 (23)	C(14)—N(3)	1.1439 (23)

C(1)—O(1)—C(4)	106.3 (2)	C(6)—C(7)—C(8)	122.3 (1)
O(1)—C(1)—C(5)	115.7 (2)	C(8)—C(7)—C(14)	120.1 (1)
O(1)—C(1)—C(2)	110.6 (3)	C(7)—C(8)—N(1)	119.7 (1)
C(2)—C(1)—C(5)	133.7 (3)	C(7)—C(8)—C(13)	124.9 (2)
C(1)—C(2)—C(3)	106.3 (2)	C(13)—C(8)—N(1)	115.4 (1)
C(2)—C(3)—C(4)	107.3 (2)	C(12)—C(9)—N(1)	115.7 (2)
O(1)—C(4)—C(3)	109.6 (2)	C(10)—C(9)—N(1)	119.4 (1)
C(3)—C(4)—C(6)	135.1 (2)	C(10)—C(9)—C(12)	124.9 (2)
O(1)—C(4)—C(6)	115.4 (1)	C(6)—C(10)—C(9)	122.3 (1)
C(1)—C(5)—N(2)	178.9 (3)	C(9)—C(10)—C(11)	120.1 (1)
C(4)—C(6)—C(10)	111.1 (1)	C(6)—C(10)—C(11)	117.5 (1)
C(4)—C(6)—C(7)	111.0 (1)	C(10)—C(11)—N(4)	177.4 (2)
C(7)—C(6)—C(10)	108.8 (1)	C(7)—C(14)—N(3)	178.7 (1)
C(6)—C(7)—C(14)	117.6 (1)	C(8)—N(1)—C(9)	122.9 (1)

<i>D</i> —H... <i>A</i>	<i>D</i> ... <i>A</i>	H... <i>A</i>	<i>D</i> —H... <i>A</i>
N(1)—H(4)...N(4)	2.959 (2)	2.10 (2)	177 (2)
C(13)—H(8)...O(1 ^b)	3.480 (2)	2.72 (2)	138 (2)
C(6)—H(3)...N(3 ⁱⁱⁱ)	3.485 (2)	2.57 (1)	158 (1)

Symmetry code: (i) $x, -\frac{1}{2}-y, -\frac{1}{2}+z$; (ii) $x, \frac{1}{2}-y, -\frac{1}{2}+z$; (iii) $2-x, -\frac{1}{2}+y, \frac{1}{2}-z$.

(Table 2). Double-bond lengths in the furyl ring differ very slightly [C(1)=C(2) = 1.333 (4) and C(3)=C(4) = 1.342 (3) Å] owing to the conjugation in the N(2)(*sp*)-C(5)(*sp*)-C(1)(*sp*²)-C(2)(*sp*²)-C(3)(*sp*²)-C(4)(*sp*²) unit. The replacement of 4-methylpyridine in position 5 of the furan ring (Lokaj, Vrabel, Sivý, Kettmann, Ilavský & Ječný, 1991) by the cyano group has resulted in increased conjugation, leading to a decrease in the C(2)—C(3) bond length from 1.441 (3) to 1.424 (5) Å.

Atomic thermal motion increases significantly with distance from the molecular centre. The U_{eq} values range from <0.042 Å² for dihydropyridine to

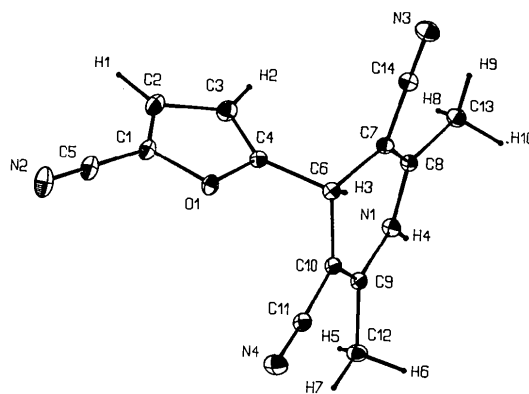


Fig. 1. The molecular structure of the title compound showing the atom numbering.

0.073 Å² in the furyl ring and up to 0.092 (1) Å² for the outer N(2) atom. The ratio of the shortest to longest principal axis increases likewise, with a maximum asymmetry of 2.98 for N(4).

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Structure of 12b-Hydroxy-5-methyl-1,2,3,4,4a,5,6,12b-octahydro-12H-benzo[*a*]xanthen-12-one

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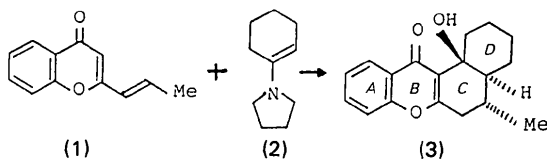
Abstract. C₁₈H₂₀O₃, $M_r = 284.35$, triclinic, $P\bar{1}$, $a = 7.505$ (2), $b = 8.805$ (2), $c = 11.443$ (4) Å, $\alpha =$

101.53 (3), $\beta = 99.57$ (3), $\gamma = 95.43$ (2)°, $V = 724.3$ (9) Å³, $Z = 2$, $D_x = 1.299$ g cm⁻³, $\lambda(\text{Mo } K\alpha) = 0.71073$ Å, $\mu = 0.816$ cm⁻¹, $F(000) = 304$, $T = 296$ K, $R = 0.045$ for 1900 independent reflections.

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Molecules are linked by hydrogen bonding, O(1)—H(1)···O(3') [2.855 (2) Å, 165.4 (2.9)°, symmetry code: (i) $-x, -y, -z$].

Introduction. Reaction of 2-(1-propenyl)-4*H*-chromen-4-one (1) with 1-(*N*-pyrrolidinyl)cyclohexene (2) gave a nitrogen-free hydroxy compound whose structure, and especially stereochemistry, could not be unambiguously established by spectroscopy owing to overlapping aliphatic signals in its ¹H NMR (270 MHz) spectrum. The structure of this compound is significant in that it would be expected to provide useful information on the mechanism of this and analogous reactions (Letcher & Yue, 1992). We have now carried out a single-crystal X-ray structure determination and found the hydroxy compound to have the structure represented by (3).



Experimental. The title compound (3) was obtained in 10% yield from the reaction between (1) and (2) in refluxing ethanol and was crystallized from 95% ethanol.

A colourless crystal of dimensions 0.12 × 0.35 × 0.45 mm was selected for data collection using an Enraf-Nonius CAD-4 diffractometer with graphite-monochromated Mo *K*α radiation and scintillation counter. Cell dimensions were found by least squares from setting angles of 25 reflections ($10 < 2\theta < 13^\circ$) measured on the diffractometer. The intensities of 5084 reflections ($2\theta_{\max} = 50^\circ$), in the range $-8 \leq h \leq 8$, $-10 \leq k \leq 10$, $-13 \leq l \leq 13$, were measured, using the ω - 2θ scan, with ω -scan angle $(1.05 + 0.344 \tan \theta)^\circ$ at 1.27 to 5.49 min⁻¹, extended 25% on each side for background measurement. Three standard reflections measured every 2 h showed no decay. The space group *P* $\bar{1}$ was confirmed in the structure solution. The intensity data were corrected for Lp effects but not for absorption. After equivalent reflections were averaged, the 5084 measured reflections gave 2542 independent ones, of which 1968 had $I > 3\sigma(I)$ [where $\sigma^2(I) = S + 4(B_1 + B_2)$, $S = \text{scan}$, B_1 and $B_2 = \text{background counts}$] and were considered observed. $R_{\text{int}} = 0.023$ for observed and 0.033 for all reflections. The structure was determined by direct methods (*MULTAN*11/82; Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1982) and refined by full-matrix least squares (on *F*) with atomic scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV), using the *SDP* programs (Enraf-Nonius, 1985) on a Micro-

VAX II computer. The C and O atoms were refined anisotropically; the positional parameters of the hydroxy H(1) atom, which was located in the difference Fourier map, were also refined but all other H atoms in calculated positions were not refined. Convergence for 193 variables by least squares with $w = 4F_o^2/\sigma^2(F_o^2)$, where $\sigma(F_o^2) = [\sigma^2(I) + (0.045F_o^2)^2]^{1/2}$, and reflections with $F_o^2 < 3\sigma(F_o^2)$ given negative weights and omitted in the refinement, was reached at $R = 0.045$, $wR = 0.062$ and $S = 1.778$ for 1900 reflections. $(\Delta/\sigma)_{\max} = 0.01$. A final difference Fourier map was featureless, with maximum positive and negative peaks of 0.28 and 0.17 e Å⁻³, respectively.

Discussion. An *ORTEP*II drawing (Johnson, 1976) of two molecules related by the origin, showing the hydrogen bonding and the atomic numbering scheme is presented in Fig. 1. The atomic parameters of the C, O and the hydroxy H(1) atoms are listed in Table 1.* Bond lengths, bond angles and selected torsion angles are given in Table 2.

Rings *A*, *B* and *C* of the xanthene skeleton are nearly coplanar; the dihedral angle between the least-squares planes through the six atoms of ring *A* ($\chi^2 = 50$) and ring *B* ($\chi^2 = 691$) is 1.0°, and that between the six atoms of ring *B* and C(6), C(6a), C(12a)—C(12b) of ring *C* ($\chi^2 = 19$) is 2.7°.

* Lists of structure factors, anisotropic thermal parameters, H-atom coordinates, bond lengths and angles involving H atoms, torsion angles, root-mean-square amplitudes and least-squares planes have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 55668 (25 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: HU1012]

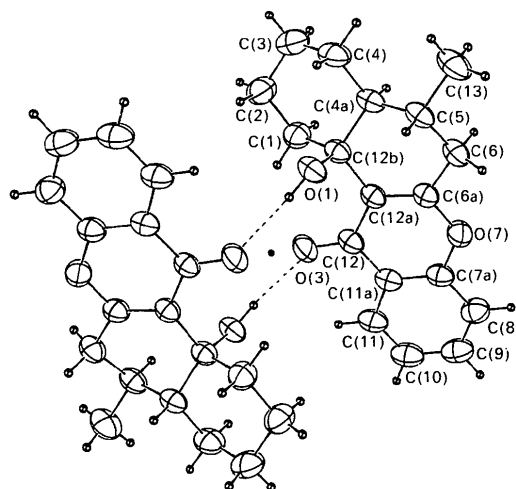


Fig. 1. *ORTEP*II (Johnson, 1976) drawing of two molecules of (3) with the numbering of atoms shown.

Table 1. Atomic coordinates and equivalent isotropic thermal parameters (Å²) with e.s.d.'s in parentheses
$$B_{eq} = (8\pi^2/3) \sum_i \sum_j U_{ij} a_i^* a_j^* a_i a_j$$

	x	y	z	B _{eq}
O(1)	0.0298 (2)	-0.0031 (1)	-0.2310 (1)	3.55 (3)
O(3)	0.1718 (2)	0.1060 (1)	0.0640 (1)	4.38 (3)
O(7)	0.2500 (2)	0.4962 (1)	-0.0683 (1)	3.77 (3)
C(1)	0.3281 (3)	-0.0533 (2)	-0.1364 (2)	3.87 (4)
C(2)	0.3184 (3)	-0.2095 (2)	-0.2260 (2)	4.62 (5)
C(3)	0.3814 (3)	-0.1853 (3)	-0.3405 (2)	5.07 (5)
C(4)	0.2760 (3)	-0.0700 (2)	-0.3971 (2)	4.35 (5)
C(4a)	0.2899 (2)	0.0858 (2)	-0.3065 (2)	3.23 (4)
C(5)	0.2001 (3)	0.2123 (2)	-0.3585 (2)	3.45 (4)
C(6)	0.2559 (3)	0.3668 (2)	-0.2656 (2)	3.71 (4)
C(6a)	0.2450 (2)	0.3519 (2)	-0.1399 (2)	3.09 (4)
C(7a)	0.2368 (3)	0.5101 (2)	0.0512 (2)	3.35 (4)
C(8)	0.2405 (3)	0.6608 (2)	0.1185 (2)	4.15 (5)
C(9)	0.2264 (3)	0.6795 (3)	0.2386 (2)	4.48 (5)
C(10)	0.2110 (3)	0.5517 (3)	0.2912 (2)	4.47 (5)
C(11)	0.2051 (3)	0.4031 (2)	0.2232 (2)	3.88 (4)
C(11a)	0.2179 (2)	0.3800 (2)	0.1007 (2)	3.22 (4)
C(12)	0.2051 (2)	0.2224 (2)	0.0234 (2)	3.11 (4)
C(12a)	0.2299 (2)	0.2174 (2)	-0.1007 (1)	2.92 (4)
C(12b)	0.2188 (2)	0.0613 (2)	-0.1916 (1)	2.98 (4)
C(13)	0.2496 (3)	0.2331 (3)	-0.4793 (2)	5.16 (5)
H(1)	-0.015 (4)	-0.030 (3)	-0.169 (2)	4.6†

† Refined isotropically.

The saturated *D* ring has a chair configuration with C(1) and C(4) deviating -0.67 and 0.67 Å from the mean plane through C(2), C(3), C(4a) and C(12b). The H atom at C(4a) is *trans* to the hydroxy group and is also *trans* to the H atom at C(5). The C(sp³)-C(sp³) bond lengths range from 1.544 (3) for C(4a)-C(12b) to 1.512 (3) Å for C(2)-C(3). Bond angles involving three C(sp³) atoms range from 108.3 (1) to 114.7 (1)°. At the centre of the molecule, the C(6a)-C(12a) bond length is 1.349 (3) Å, which is close to the normal C=C bond length. The bond length of 1.454 (2) Å for C(12)-C(12a) is shorter than that of 1.475 (2) Å for C(12)-C(11a), while the bond lengths for O(7)-C(6a) and O(7)-C(7a) are almost identical, being 1.361 (2) and 1.369 (2) Å, respectively. In the phenyl ring, the C-C bond lengths range from 1.397 (3) to 1.374 (3) Å while the bond angles range from 122.3 (2) at C(7a) to 118.0 (2)° at C(11a).

The ketone O(3)=C(12) bond length is 1.229 (2) Å. The bond length for the hydroxy group [*i.e.* O(1)-C(12b)] is 1.438 (3) Å while the bond length for the fully refined H atom H(1) to O(1) is 0.89 (3) Å. The distances of O(3) at (-*x*, -*y*, -*z*) to O(1) and H(1) are 2.855 (2) and 1.99 (3) Å, respectively, while the O(3)⋯H(1)-O(1) angle is 165.4 (2.9)°, suggesting intermolecular hydrogen bonding. The hydroxy C-O bond length is comparable to 1.425 Å found in 9-(cyclohepta-2,4,6-trien-1-yl)xanthen-9-ol (Badejo, Fry, Karaman, Nadkarni &

Table 2. Bond lengths (Å), bond angles (°) and torsion angles (°) with e.s.d.'s in parentheses

O(1)-C(12b)	1.438 (3)	C(6)-C(6a)	1.485 (3)
O(3)-C(12)	1.229 (2)	C(6a)-C(12a)	1.349 (3)
O(7)-C(6a)	1.361 (2)	C(7a)-C(8)	1.391 (3)
O(7)-C(7a)	1.369 (2)	C(7a)-C(11a)	1.382 (3)
C(1)-C(2)	1.529 (3)	C(8)-C(9)	1.374 (3)
C(1)-C(12b)	1.526 (3)	C(9)-C(10)	1.383 (3)
C(2)-C(3)	1.512 (3)	C(10)-C(11)	1.374 (3)
C(3)-C(4)	1.524 (3)	C(11)-C(11a)	1.397 (3)
C(4)-C(4a)	1.528 (2)	C(11a)-C(12)	1.475 (2)
C(4a)-C(5)	1.525 (3)	C(12)-C(12a)	1.454 (2)
C(4a)-C(12b)	1.544 (3)	C(12a)-C(12b)	1.535 (2)
C(5)-C(6)	1.524 (2)	O(1)-H(1)	0.89 (3)
C(5)-C(13)	1.530 (3)		
C(6a)-O(7)-C(7a)	119.6 (1)	C(8)-C(9)-C(10)	120.8 (2)
C(2)-C(1)-C(12b)	112.6 (1)	C(9)-C(10)-C(11)	120.4 (2)
C(1)-C(2)-C(3)	110.9 (2)	C(10)-C(11)-C(11a)	120.3 (2)
C(2)-C(3)-C(4)	111.2 (3)	C(7a)-C(11a)-C(11)	118.0 (2)
C(3)-C(4)-C(4a)	111.4 (1)	C(7a)-C(11a)-C(12)	119.9 (2)
C(4)-C(4a)-C(5)	114.7 (1)	C(11)-C(11a)-C(12)	122.0 (2)
C(4)-C(4a)-C(12b)	110.9 (2)	O(3)-C(12)-C(11a)	120.2 (2)
C(5)-C(4a)-C(12b)	111.0 (2)	O(3)-C(12)-C(12a)	124.2 (1)
C(4a)-C(5)-C(6)	108.3 (1)	C(11a)-C(12)-C(12a)	115.7 (2)
C(4a)-C(5)-C(13)	114.3 (2)	C(6a)-C(12a)-C(12)	119.2 (1)
C(6)-C(5)-C(13)	108.9 (2)	C(6a)-C(12a)-C(12b)	119.2 (2)
C(5)-C(6)-C(6a)	133.1 (2)	C(12)-C(12a)-C(12b)	121.3 (2)
O(7)-C(6a)-C(6)	109.5 (2)	O(1)-C(12b)-C(1)	110.7 (1)
O(7)-C(6a)-C(12a)	124.2 (2)	O(1)-C(12b)-C(4a)	106.5 (1)
C(6)-C(6a)-C(12a)	126.3 (1)	O(1)-C(12b)-C(12a)	108.0 (1)
O(7)-C(7a)-C(8)	116.6 (2)	C(1)-C(12b)-C(4a)	108.4 (2)
O(7)-C(7a)-C(11a)	121.3 (1)	C(1)-C(12b)-C(12a)	111.9 (1)
C(8)-C(7a)-C(11a)	122.3 (2)	C(4a)-C(12b)-C(12a)	111.2 (1)
C(7a)-C(8)-C(9)	118.2 (3)	C(12b)-O(1)-H(1)	110 (2)
C(12b)-C(1)-C(2)-C(3)	56.3 (3)	O(7)-C(6a)-C(12a)-C(12b)	177.8 (2)
C(2)-C(1)-C(12b)-O(1)	59.4 (2)	C(6)-C(6a)-C(12a)-C(12)	-175.0 (2)
C(2)-C(1)-C(12b)-C(4a)	-57.0 (2)	C(6)-C(6a)-C(12a)-C(12b)	-1.2 (3)
C(1)-C(2)-C(3)-C(4)	-54.2 (3)	O(7)-C(7a)-C(11a)-C(11)	-180.0 (2)
C(2)-C(3)-C(4)-C(4a)	55.7 (2)	O(7)-C(7a)-C(11a)-C(12)	-2.1 (3)
C(3)-C(4)-C(4a)-C(12b)	-57.7 (2)	C(8)-C(7a)-C(11a)-C(11)	-1.1 (3)
C(4)-C(4a)-C(5)-C(13)	-47.9 (2)	C(8)-C(7a)-C(11a)-C(12)	176.8 (2)
C(12b)-C(4a)-C(5)-C(6)	64.0 (2)	C(6a)-C(12a)-C(12b)-O(1)	-97.6 (2)
C(4)-C(4a)-C(12b)-O(1)	-61.8 (2)	C(6a)-C(12a)-C(12b)-C(1)	140.2 (2)
C(4)-C(4a)-C(12b)-C(1)	57.3 (2)	C(6a)-C(12a)-C(12b)-C(4a)	18.8 (2)
C(5)-C(4a)-C(12b)-O(1)	66.9 (2)	C(12)-C(12a)-C(12b)-O(1)	75.8 (2)
C(5)-C(4a)-C(12b)-C(12a)	-30.5 (2)	C(12)-C(12a)-C(12b)-C(1)	-46.3 (2)
C(4a)-C(5)-C(6)-C(6a)	-44.6 (2)	C(12)-C(12a)-C(12b)-C(4a)	-167.7 (2)
O(7)-C(6a)-C(12a)-C(12)	4.3 (3)		

Pinkerton, 1991) in which intermolecular hydrogen bonding was also observed.

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