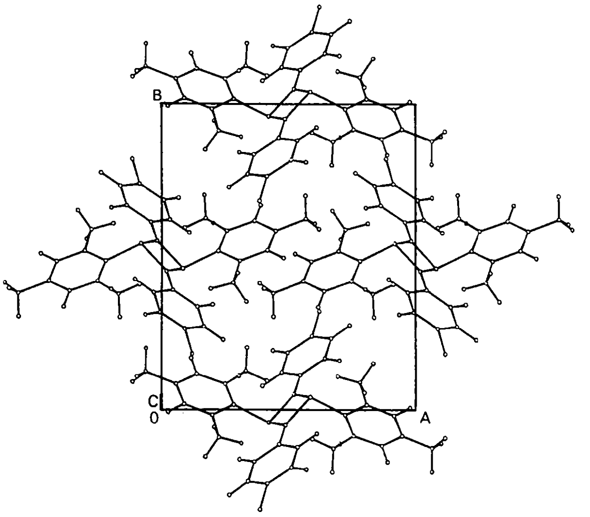

Fig. 2. Projection des molécules dans le plan *xy*.

H(C16^{i}) = 3,00, Cl(11)···H(C14^{ii}) = 2,92 et Cl(11)··· H''(C18^{ii}) = 2,95 Å, avec les symétries (i) 0,5 + x, 0,5 − y, 1 + z et (ii) 1,5 − x, −0,5 + y, −z.

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Crystal Studies of Musk Compounds. IV. Structure of 4-Acetyl-6-*tert*-butyl-1,1-dimethylindan (Musk Celestolide)

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Abstract. $C_{17}H_{24}O$, $M_r = 244.38$, triclinic, $P\bar{1}$, $a = 10.160\,(2)$, $b = 12.230\,(2)$, $c = 12.796\,(1)$ Å, $\alpha = 105.735\,(6)$, $\beta = 99.58\,(2)$, $\gamma = 90.09\,(1)°$, $V = 1507.2\,(5)$ Å^3, $Z = 4$, $D_x = 1.08$ Mg m^{-3}, λ(Cu $K\alpha$) = 1.5418 Å, $\mu = 0.461$ mm^{-1}, $F(000) = 536$, $T = 240$ K, final $R = 0.046$ for 4766 observed reflections. There are two independent molecules in the asymmetric unit, related to each other by a non-crystallographic twofold screw axis. The aromatic ring is essentially planar, whereas the five-membered ring is an almost ideal C(2) envelope. The torsion angle C(9)—C(4)—C(12)—O(1) has an average value of 23°. The geometries of the independent molecules are compared.

Introduction. This paper is the fourth in a series dealing with musk compounds [(I) and (II), De Ridder, Goubitz & Schenk (1990*a*,*b*); (III), De Ridder & Schenk (1991*a*)]. Besides the classification of the synthetic aromatic nitro-free musks given in the previous paper, it is also possible to divide this family by referring to the relative orientation of C atoms containing *gem*-dimethyl substitution relative to the benzene ring (Theimer & Davies, 1967). This results in the so-called *ortho* and *meta* musks as none of the known *para* compounds show muskiness. Musk tonalid (De Ridder, Goubitz & Schenk, 1990*b*) and musk phantolid (De Ridder & Schenk, 1991*a*) both belong to the *ortho* musks, while the compound described in this paper belongs to the *meta* musks.

Musk celestolide was discovered in the late fifties by Beets and his co-workers (Beets, van Essen & Meerburg, 1958). They reported it as colourless leaflets, melting point 350.1–350.4 K, having a very strong musk odour. It is also known as musk DTI.

Experimental. A sample was recrystallized by slow evaporation from methanol. Intensities collected on

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Table 1. *Fractional coordinates of the non-*H *atoms and equivalent isotropic thermal parameters* (Å^2)

$$U_{eq} = (1/3)\textstyle\sum_i\sum_j U_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	U_{eq}
Molecule (A)				
C(1)	0.8535 (2)	−0.0039 (2)	0.2889 (2)	0.039 (1)
C(2)	0.9135 (2)	−0.0450 (2)	0.1820 (2)	0.044 (1)
C(3)	0.8727 (2)	0.0385 (2)	0.1138 (2)	0.039 (1)
C(4)	0.6381 (2)	0.1322 (1)	0.0989 (1)	0.0294 (9)
C(5)	0.5254 (2)	0.1593 (2)	0.1502 (1)	0.0307 (9)
C(6)	0.5155 (2)	0.1384 (2)	0.2501 (1)	0.0302 (9)
C(7)	0.6207 (2)	0.0859 (2)	0.2995 (1)	0.033 (1)
C(8)	0.7321 (2)	0.0555 (2)	0.2481 (1)	0.0321 (9)
C(9)	0.7421 (2)	0.0799 (2)	0.1503 (1)	0.0303 (9)
C(10)	0.8175 (2)	−0.1027 (2)	0.3318 (2)	0.059 (2)
C(11)	0.9496 (2)	0.0833 (2)	0.3778 (2)	0.054 (1)
C(12)	0.6460 (2)	0.1600 (2)	−0.0059 (2)	0.034 (1)
C(13)	0.5193 (2)	0.1741 (2)	−0.0791 (2)	0.043 (1)
C(14)	0.3895 (2)	0.1729 (2)	0.3015 (1)	0.034 (1)
C(15)	0.3906 (2)	0.1369 (2)	0.4071 (2)	0.047 (1)
C(16)	0.3810 (2)	0.3021 (2)	0.3272 (2)	0.047 (1)
C(17)	0.2657 (2)	0.1154 (2)	0.2180 (2)	0.048 (1)
O(1)	0.7545 (1)	0.1707 (1)	−0.0332 (1)	0.0497 (9)
Molecule (B)				
C(1)	1.2310 (2)	0.5057 (2)	−0.2868 (2)	0.042 (1)
C(2)	1.3389 (2)	0.5436 (2)	−0.1819 (2)	0.048 (1)
C(3)	1.3262 (2)	0.4602 (2)	−0.1137 (2)	0.042 (1)
C(4)	1.0980 (2)	0.3671 (2)	−0.0971 (1)	0.0305 (9)
C(5)	0.9641 (2)	0.3393 (2)	−0.1475 (1)	0.0310 (9)
C(6)	0.9114 (2)	0.3598 (2)	−0.2479 (1)	0.0318 (9)
C(7)	0.9957 (2)	0.4132 (2)	0.2969 (1)	0.034 (1)
C(8)	1.1277 (2)	0.4445 (2)	−0.2460 (1)	0.033 (1)
C(9)	1.1802 (2)	0.4201 (2)	−0.1485 (1)	0.0313 (9)
C(10)	1.1760 (2)	0.6073 (2)	−0.3241 (2)	0.059 (2)
C(11)	1.2854 (3)	0.4210 (2)	−0.3801 (2)	0.065 (2)
C(12)	1.1508 (2)	0.3394 (2)	0.0080 (1)	0.033 (1)
C(13)	1.0546 (2)	0.3248 (2)	0.0813 (2)	0.043 (1)
C(14)	0.7644 (2)	0.3244 (2)	−0.2993 (2)	0.036 (1)
C(15)	0.7227 (2)	0.3577 (2)	−0.4064 (2)	0.059 (1)
C(16)	0.7438 (2)	0.1958 (2)	−0.3227 (2)	0.049 (1)
C(17)	0.6749 (2)	0.3827 (2)	−0.2165 (2)	0.053 (1)
O(1)	1.2706 (1)	0.3298 (1)	0.0352 (1)	0.0502 (9)

Table 2. *Bond lengths* (Å) *and bond angles* (°)

	Molecule (A)	Molecule (B)
C(1)—C(2)	1.549 (3)	1.545 (3)
C(1)—C(8)	1.522 (3)	1.525 (3)
C(1)—C(10)	1.523 (4)	1.523 (4)
C(1)—C(11)	1.534 (3)	1.535 (3)
C(2)—C(3)	1.530 (3)	1.529 (4)
C(3)—C(9)	1.523 (3)	1.517 (2)
C(4)—C(5)	1.409 (3)	1.403 (2)
C(4)—C(9)	1.399 (2)	1.400 (3)
C(4)—C(12)	1.484 (3)	1.489 (3)
C(5)—C(6)	1.389 (3)	1.395 (3)
C(6)—C(7)	1.397 (3)	1.398 (3)
C(6)—C(14)	1.543 (3)	1.538 (2)
C(7)—C(8)	1.403 (3)	1.393 (2)
C(8)—C(9)	1.382 (3)	1.383 (3)
C(12)—C(13)	1.501 (3)	1.505 (3)
C(12)—O(1)	1.228 (2)	1.225 (2)
C(14)—C(15)	1.530 (3)	1.528 (3)
C(14)—C(16)	1.530 (3)	1.526 (3)
C(14)—C(17)	1.536 (2)	1.540 (3)
C(2)—C(1)—C(8)	100.7 (2)	101.0 (2)
C(2)—C(1)—C(10)	111.8 (2)	111.4 (2)
C(2)—C(1)—C(11)	110.7 (2)	111.1 (2)
C(8)—C(1)—C(10)	113.2 (2)	112.7 (2)
C(8)—C(1)—C(11)	109.5 (2)	109.5 (2)
C(10)—C(1)—C(11)	110.7 (2)	110.7 (2)
C(1)—C(2)—C(3)	106.7 (2)	107.0 (2)
C(2)—C(3)—C(9)	102.2 (2)	102.4 (2)
C(5)—C(4)—C(9)	118.0 (2)	118.5 (2)
C(5)—C(4)—C(12)	120.5 (2)	120.3 (2)
C(9)—C(4)—C(12)	121.4 (2)	121.2 (1)
C(4)—C(5)—C(6)	122.6 (2)	122.4 (2)
C(5)—C(6)—C(7)	118.2 (2)	117.8 (2)
C(5)—C(6)—C(14)	119.1 (2)	119.4 (2)
C(7)—C(6)—C(14)	122.7 (2)	122.9 (2)
C(6)—C(7)—C(8)	120.0 (2)	120.4 (2)
C(1)—C(8)—C(7)	127.0 (2)	127.0 (2)
C(1)—C(8)—C(9)	111.9 (2)	111.8 (2)
C(7)—C(8)—C(9)	121.1 (2)	121.3 (2)
C(3)—C(9)—C(4)	129.9 (2)	130.2 (2)
C(3)—C(9)—C(8)	110.0 (2)	110.2 (2)
C(4)—C(9)—C(8)	120.1 (2)	119.6 (2)
C(4)—C(12)—C(13)	119.2 (2)	119.0 (2)
C(4)—C(12)—O(1)	120.8 (2)	120.8 (2)
C(13)—C(12)—O(1)	120.0 (2)	120.2 (2)
C(6)—C(14)—C(15)	112.4 (2)	112.1 (2)
C(6)—C(14)—C(16)	108.7 (2)	109.2 (2)
C(6)—C(14)—C(17)	108.9 (1)	108.9 (1)
C(15)—C(14)—C(16)	109.3 (2)	109.1 (2)
C(15)—C(14)—C(17)	108.2 (2)	108.8 (2)
C(16)—C(14)—C(17)	109.3 (2)	108.6 (2)

an Enraf–Nonius CAD-4 diffractometer using graphite-monochromated Cu $K\alpha$ radiation and θ–2θ scans at 240 K. The crystal used for data collection had approximate dimensions $0.6 \times 0.4 \times 0.2$ mm. 5709 unique intensities within the ranges $2.5 \leq \theta \leq 70°$, $0 \leq h \leq 12$, $-14 \leq k \leq 14$, $-15 \leq l \leq 15$ were measured, of which 4766 were above the significance level of $2.5\sigma(I)$. Two standard reflections ($1\bar{1}\bar{2}$ and 110), measured hourly, suffered an 8.4% decrease during 107 h collection time, which was corrected for. Unit-cell parameters were refined by a least-squares algorithm using 23 reflections with $80 < 2\theta < 88°$. Corrections applied for Lorentz and polarization effects, not for absorption. Structure determined by direct methods using *XTAL* crystallographic programs (Hall & Stewart, 1989). The H atoms were initially positioned on the basis of standard geometry but these positions were subsequently refined. Block-diagonal least-squares refinement on F, anisotropic for the non-H atoms and isotropic for the H atoms converged to $R = 0.046$, $wR = 0.045$. Max. $\Delta/\sigma = 0.12$, $w^{-1} = 8.08 + F_{obs} + 0.0037F^2_{obs}$. The isotropic secondary-extinction coefficient refined to $0.244\,(6) \times 10^{-6}$ (Larson, 1967). A final difference synthesis revealed a residual electron density between ± 0.2 e Å^{-3}. All calculations were performed with *XTAL*2.6 (Hall & Stewart, 1989) unless stated otherwise. The scattering factors were taken from Cromer & Mann (1968) and *International Tables for X-ray Crytallography* (1974, Vol. IV, p. 55).

Discussion. Final positional parameters for the non-H atoms of the two independent molecules are listed in Table 1.* Bond lengths and angles are given in Table 2 and have normal values. A *PLUTO* (Motherwell & Clegg, 1978) drawing of molecule (A) showing the atomic numbering scheme is given in Fig. 1; the numbering system of molecule (B) is identical.

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* Lists of structure factors, anisotropic thermal parameters, bond lengths and angles involving H atoms, and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 54442 (39 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: MU0236]

The aromatic ring is essentially planar within the limits of accuracy [max. distance of an atom through the best plane is 0.014 (2) for (*A*) and 0.016 (2) Å for (*B*)]. Following the criteria defined by Altona, Geise & Romers (1968) the non-aromatic ring is classified as an almost ideal C(2) envelope [(*A*): $\varphi_m = 28.2°$, $\Delta = 35.9°$; and (*B*): $\varphi_m = 27.0°$, $\Delta = 30.8°$]. The torsion angle C(9)—C(4)—C(12)—O(1) is 23.1 (2) in (*A*) and 22.8 (2)° in (*B*). The equivalent angle is 22° in musk tonalid (De Ridder, Goubitz & Schenk, 1990*b*) and 21° in musk phantolid (De Ridder & Schenk, 1991*a*).

Although the structure is metrically triclinic, and hence molecules (*A*) and (*B*) are crystallographically independent, it can be observed from Table 1 that for each corresponding atom the relations $y(B) = \frac{1}{2} - y(A)$ and $z(B) = -z(A)$ are approximately valid. However, application of the program *MISSYM* (*NRCVAX* package; Le Page, 1987) with a search on all non-H atoms did not lead to the detection of any extra crystallographic symmetry. During matching of the two independent molecules we derived the matching transformation as a pseudosymmetry operation from molecule (*B*) to molecule (*A*): $x(B) = 1.00x(A) + 0.01y(A) - 0.43z(A) + 0.50$; $y(B) = 0.01x(A) - 1.00y(A) + 0.01z(A) + 0.49$; $z(B) = 0x(A) - 0.01y(A) - 1.00z(A) + 0.00$.

There are thus local non-crystallographic 2_1 axes parallel to [100] at $x = 0$, $y = \frac{1}{4}$, $z = 0$; $x = 0$, $y = \frac{1}{4}$, $z = \frac{1}{2}$; *etc.* This is also evident from the stereoscopic representation of the unit cell given in Fig. 2. No intermolecular distances significantly shorter than expected from the van der Waals contacts are observed. The shortest intermolecular distance is found between the atoms O(1) and C(13) [3.447 (2) Å for O(1*B*)—C(13*A*) and 3.463 (2) Å for C(13*B*)—O(1*A*)]. Matching of the independent molecules resulted in an r.m.s. value of 0.07 Å, which is in agreement with the apparent pseudosymmetry.

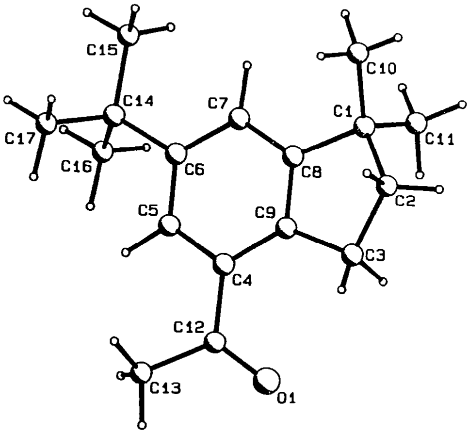

Fig. 1. The numbering system of musk celestolide. The H atoms are shown but not labelled.

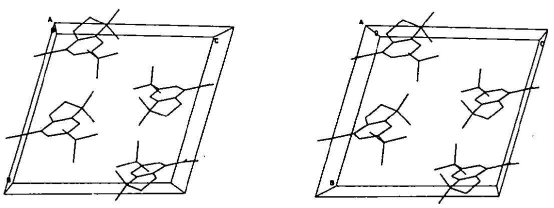

Fig. 2. Stereoscopic representation of the unit cell along [100].

This structure is isomeric with musk phantolid (De Ridder & Schenk, 1991*a*), showing the great variety in homologues and isomers among compounds having a musk odour. Narvaez, Lavine & Jurs (1986) have drawn attention to the correlation between the position of the polar functional group relative to the bulky substituents and musk-odour quality. For a number of indans and tetralins we calculated an average distance between the quaternary C atoms of 2.52 and 3.07 Å respectively (De Ridder & Schenk (1991*b*). The equivalent average distance in musk celestolide of 5.19 Å is significantly larger. This may be an indication that this distance is not a decisive criterion for muskiness.

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