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Three Tetramethylketals of 2,5-Distyryl-[1,4]benzoquinones

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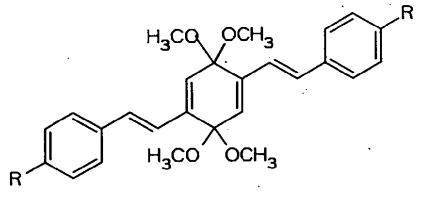
Abstract

The methoxy groups of the title compounds 3,3,6,6-tetramethoxy-1,4-distyrylcyclohexa-1,4-diene, $C_{26}H_{28}O_4$ (I), 1,4-bis[2-(4-chlorophenyl)vinyl]-3,3,-6,6-tetramethoxycyclohexa-1,4-diene, $C_{26}H_{26}Cl_2O_4$ (II), and 3,3,6,6-tetramethoxy-1,4-bis[2-(4-methylphenyl)vinyl]cyclohexa-1,4-diene, $C_{28}H_{32}O_4$ (III), adopt either an *anti–anti* or a *gauche–anti* orientation. In the *anti–anti* conformation the O—C—O bond angle is reduced to 99.2–99.3° because of intramolecular repulsive interactions between the methyl groups. The stacking of the molecules in the crystal is defined by an interlocking pattern.

Comment

In the course of topochemical studies of 2,5-distyryl-[1,4]benzoquinones we synthesized their corresponding ketals 3,3,6,6-tetramethoxy-1,4-distyrylcyclohexa-1,4-diene (I), 1,4-bis[2-(4-chlorophenyl)vinyl]-3,3,6,6-tetramethoxycyclohexa-1,4-diene (II) and 3,3,6,6-tetramethoxy-1,4-bis[2-(4-methylphenyl)-vinyl)cyclohexa-1,4-diene (III) (Irngartinger, Lichtenthäler, Fenske & Baum, 1993).



(I) R = H
(II) R = Cl
(III) R = CH_3

The structures of only two 3,3,6,6-tetramethoxy-cyclohexa-1,4-dienes have been reported, the unsubstituted parent compound (IV) (Liebich, Yvon & Margaretha, 1976) and the tetramethyl derivative (V) (Nørskov-Lauritsen, Larsen, Ettlinger & Jaroszewski, 1982). These two compounds and the derivatives (I), (II) and (III) (Fig. 1) lie on a crystallographic centre of symmetry.

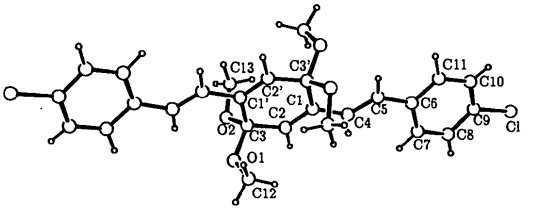


Fig. 1. View of (II) showing the atom labelling.

The cyclohexadiene rings are planar. The methoxy groups show two kinds of conformation. In the derivatives (I), (II) and (V) the methoxy groups adopt an *anti–anti* orientation and in compounds (III) and (IV) a *gauche–anti* conformation (Fig. 2). The *gauche* orientation is defined by the torsion angle O2—C3—O1—C12 of −63.1° in (III). For the *anti* orientation the corresponding torsion angles are in the region of 180° [177.4 (1) in (I), 173.9 (2)° in (II)].

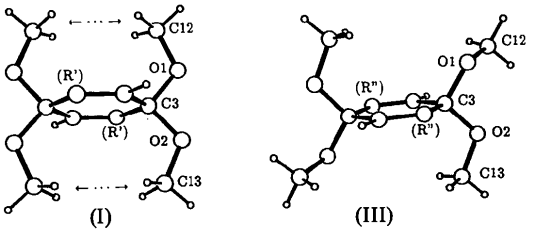


Fig. 2. The orientation of the methoxy groups [R' = styryl, R'' = 2-(4-methylphenyl)vinyl].

The repulsion between the methyl groups [along the double arrow in Fig. 2; 1···8 (C12···C13′) distances 3.718 (2) in (I), 3.731 (4) Å in (II)] generates a reduction of the angle O1—C3—O2 [99.2 (1) in (I), 99.3 (1)° in (II)]. Similar molecular geometry was found in (V) (Nørskov-Lauritsen *et al.*, 1982). In the case of the *gauche–anti* conformation, however, this angle is not affected [106.1 (2)° in (III)], as was also found for (IV) (Liebich *et al.*, 1976). The remaining molecular dimensions (Table 2) are similar to those of related molecules. The stacking and interlocking of the molecules in the packing arrangement are comparable for all the three compounds and are illustrated in Fig. 3 for compound (I).

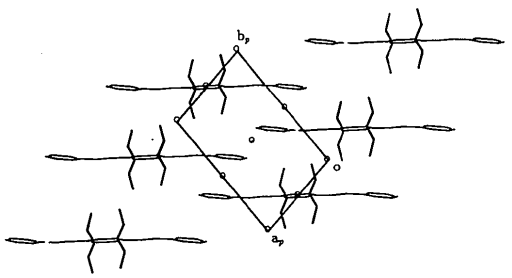


Fig. 3. A view of the molecular packing of (I).

Experimental

Compound (I)

Crystal data

$C_{26}H_{28}O_4$
M_r = 404.4
Triclinic
$P\bar{1}$
a = 7.657 (1) Å
b = 11.535 (2) Å
c = 6.460 (1) Å
α = 95.08 (1)°
β = 101.03 (2)°
γ = 98.18 (1)°
V = 550.3 (3) Å^3
Z = 1
D_x = 1.22 Mg m^{-3}
Mo $K\alpha$ radiation
λ = 0.7107 Å
Cell parameters from 75 reflections
θ = 9.99-20.07°
μ = 0.076 mm^{-1}
T = 293 K
Platelets
0.5 × 0.45 × 0.15 mm
Colourless

Data collection

Enraf-Nonius CAD-4 diffractometer
$\omega/2\theta$ scans
Absorption correction: none
2964 measured reflections
2637 independent reflections
1743 observed reflections [$I > 2.5\sigma(I)$]
R_{int} = 0.017
θ_{max} = 28°
$h = -10 \rightarrow 10$
$k = -15 \rightarrow 15$
$l = 0 \rightarrow 8$
3 standard reflections
frequency: 60 min
intensity variation: 1.7%

Refinement

Refinement on F^2
R = 0.039
wR = 0.050
S = 2.11
1743 reflections
192 parameters
All H-atom parameters refined
$w = 1/\sigma^2(F)$
$(\Delta/\sigma)_{max}$ = 0.01
$\Delta\rho_{max}$ = 0.14 e Å^{-3}
$\Delta\rho_{min}$ = −0.23 e Å^{-3}
Extinction correction: none
Atomic scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV)

Compound (II)

Crystal data

$C_{26}H_{26}Cl_2O_4$
M_r = 473.4
Monoclinic
$P2_1/c$
a = 12.732 (2) Å
b = 10.318 (1) Å
c = 9.014 (1) Å
β = 94.52 (4)°
V = 1180.5 (4) Å^3
Z = 2
D_x = 1.33 Mg m^{-3}
Mo $K\alpha$ radiation
λ = 0.7107 Å
Cell parameters from 48 reflections
θ = 9-18°
μ = 0.303 mm^{-1}
T = 293 K
Platelets
0.5 × 0.5 × 0.4 mm
Colourless

Data collection

Enraf-Nonius CAD-4 diffractometer
$\omega/2\theta$ scans
Absorption correction: analytical by integration from crystal shape
T_{min} = 0.90, T_{max} = 0.92
3105 measured reflections
2818 independent reflections
1774 observed reflections [$I > 2.5\sigma(I)$]
R_{int} = 0.076
θ_{max} = 28°
$h = 0 \rightarrow 16$
$k = 0 \rightarrow 13$
$l = -11 \rightarrow 11$
3 standard reflections
frequency: 60 min
intensity variation: 1.6%

Refinement

Refinement on F^2
R = 0.045
wR = 0.056
S = 2.32
1774 reflections
197 parameters
All H-atom parameters refined
$w = 1/\sigma^2(F)$
$(\Delta/\sigma)_{max}$ = 0.03
$\Delta\rho_{max}$ = 0.28 e Å^{-3}
$\Delta\rho_{min}$ = −0.09 e Å^{-3}
Extinction correction: none
Atomic scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV)

Compound (III)

Crystal data

$C_{28}H_{32}O_4$
M_r = 432
Monoclinic
$P2_1/n$
a = 11.529 (2) Å
b = 11.867 (2) Å
c = 9.592 (2) Å
β = 112.18 (2)°
V = 1215.2 (8) Å^3
Z = 2
D_x = 1.18 Mg m^{-3}
Mo $K\alpha$ radiation
λ = 0.7107 Å
Cell parameters from 49 reflections
θ = 9.98-18.04°
μ = 0.076 mm^{-1}
T = 293 K
Platelets
0.5 × 0.45 × 0.2 mm
Colourless

Data collection

Enraf-Nonius CAD-4 diffractometer
$\omega/2\theta$ scans
Absorption correction: none
3136 measured reflections
2919 independent reflections
1192 observed reflections [$I > 2.5\sigma(I)$]
R_{int} = 0.11
θ_{max} = 28°
$h = 0 \rightarrow 15$
$k = 0 \rightarrow 15$
$l = -12 \rightarrow 12$
3 standard reflections
frequency: 60 min
intensity variation: 17.7%

Refinement

Refinement on F^2
R = 0.051
wR = 0.059
S = 2.30
$(\Delta/\sigma)_{max}$ = 0.01
$\Delta\rho_{max}$ = 0.28 e Å^{-3}
$\Delta\rho_{min}$ = −0.10 e Å^{-3}
Extinction correction: none

1192 reflections
173 parameters
All H-atom parameters refined
$w = 1/\sigma^2(F)$

Atomic scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV)

Table 1. *Fractional atomic coordinates and equivalent isotropic displacement parameters* (Å^2)

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

	x	y	z	U_{eq}
(I)				
O1	0.4181 (1)	−0.1493 (1)	−0.3534 (1)	0.0436 (4)
O2	0.2388 (1)	−0.0199 (1)	−0.3441 (1)	0.0460 (4)
C1	0.6516 (2)	0.0938 (1)	0.0093 (2)	0.0353 (5)
C2	0.5501 (2)	0.0440 (1)	−0.1777 (2)	0.0391 (5)
C3	0.3909 (2)	−0.0535 (1)	−0.2140 (2)	0.0372 (5)
C4	0.8025 (2)	0.1888 (1)	0.0145 (2)	0.0402 (5)
C5	0.9131 (2)	0.2502 (1)	0.1855 (2)	0.0426 (5)
C6	1.0584 (2)	0.3485 (1)	0.1857 (2)	0.0405 (5)
C7	1.1205 (2)	0.3768 (1)	0.0048 (2)	0.0487 (6)
C8	1.2530 (2)	0.4731 (2)	0.0142 (3)	0.0573 (7)
C9	1.3258 (2)	0.5432 (1)	0.2035 (3)	0.0611 (7)
C10	1.2673 (2)	0.5166 (2)	0.3843 (3)	0.0634 (8)
C11	1.1361 (2)	0.4194 (1)	0.3758 (2)	0.0526 (6)
C12	0.5679 (2)	−0.2039 (1)	−0.2794 (3)	0.0641 (7)
C13	0.1666 (2)	0.0734 (2)	−0.2545 (3)	0.0692 (8)
(II)				
Cl	0.0727 (1)	0.1876 (1)	0.6947 (1)	0.0821 (4)
O1	−0.5052 (1)	0.4140 (1)	−0.2780 (1)	0.0495 (7)
O2	−0.6274 (1)	0.3338 (1)	−0.1512 (2)	0.0490 (7)
C1	−0.4200 (2)	0.4393 (2)	0.1022 (2)	0.0362 (9)
C2	−0.4605 (2)	0.3756 (2)	−0.0181 (2)	0.0402 (9)
C3	−0.5425 (2)	0.4235 (2)	−0.1340 (2)	0.0392 (9)
C4	−0.3390 (2)	0.3769 (2)	0.2037 (2)	0.0406 (10)
C5	−0.2871 (2)	0.4285 (2)	0.3226 (2)	0.0428 (10)
C6	−0.2034 (2)	0.3641 (2)	0.4177 (2)	0.0405 (10)
C7	−0.1800 (2)	0.2323 (2)	0.4086 (3)	0.0524 (12)
C8	−0.0971 (2)	0.1773 (2)	0.4948 (3)	0.0580 (12)
C9	−0.0357 (2)	0.2541 (2)	0.5912 (2)	0.0511 (11)
C10	−0.0575 (2)	0.3830 (2)	0.6061 (3)	0.0600 (13)
C11	−0.1418 (2)	0.4369 (2)	0.5211 (2)	0.0559 (12)
C12	−0.4242 (2)	0.5021 (3)	−0.3089 (3)	0.0608 (13)
C13	−0.6876 (2)	0.3191 (3)	−0.0271 (3)	0.0689 (14)
(III)				
O1	0.0120 (2)	−0.2094 (2)	1.0715 (2)	0.074 (1)
O2	0.1189 (2)	−0.0920 (2)	1.2647 (2)	0.068 (1)
C1	0.0795 (2)	0.0367 (2)	0.9226 (3)	0.052 (1)
C2	0.1165 (2)	−0.0472 (3)	1.0209 (3)	0.059 (1)
C3	0.0440 (2)	−0.0946 (2)	1.1077 (3)	0.056 (1)
C4	0.1530 (2)	0.0823 (3)	0.8381 (3)	0.059 (1)
C5	0.2536 (2)	0.0379 (3)	0.8305 (3)	0.060 (1)
C6	0.3325 (2)	0.0831 (2)	0.7524 (3)	0.052 (1)
C7	0.3049 (2)	0.1814 (2)	0.6680 (3)	0.060 (1)
C8	0.3829 (2)	0.2204 (2)	0.5995 (3)	0.059 (1)
C9	0.4901 (2)	0.1632 (2)	0.6105 (3)	0.052 (1)
C10	0.5168 (2)	0.0658 (3)	0.6939 (3)	0.066 (1)
C11	0.4395 (2)	0.0260 (3)	0.7636 (3)	0.066 (1)
C12	0.1097 (3)	−0.2854 (3)	1.0896 (4)	0.102 (2)
C13	0.1500 (3)	0.0178 (3)	1.3261 (4)	0.084 (2)
C14	0.5738 (3)	0.2063 (3)	0.5322 (3)	0.075 (2)

Table 2. *Selected bond lengths* (Å)

	(I)	(II)	(III)
C1—C2	1.336 (2)	1.336 (3)	1.326 (4)
C1—C3′	1.517 (2)	1.528 (3)	1.507 (4)
C2—C3	1.503 (2)	1.501 (3)	1.495 (5)
C1—C4	1.468 (2)	1.472 (3)	1.479 (4)
C4—C5	1.331 (2)	1.325 (3)	1.300 (4)
C5—C6	1.471 (2)	1.472 (3)	1.480 (4)
O1—C3	1.427 (2)	1.420 (2)	1.419 (4)
O2—C3	1.423 (2)	1.422 (2)	1.426 (3)
O1—C12	1.411 (2)	1.419 (3)	1.402 (4)
O2—C13	1.408 (2)	1.413 (3)	1.418 (4)

The structure solutions of (I)–(III) were carried out with *MULTAN* (Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1982) and refinements were performed by full-matrix least-squares techniques. The positions of the H atoms were calculated and refined isotropically. All calculations were performed with the *MolEN* package (Fair, 1990). The figures were plotted using *ORTEP* (Johnson, 1976).

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Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and complete geometry have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 71839 (64 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: SE1044]

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A Spiro-Indole Derivative, $C_{16}H_{10}ClFN_2O_2S$

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Abstract

The spiro-indole derivative, 5-chloro-3′-(4-fluorophenyl)spiro[3*H*-indole-3,2′-thiazolidine]-2,4′(1*H*)-dione, has been synthesized by the reaction of mer-