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Stereoselectivity in the Michael Reaction. II. Stereochemistry of a Heavily Substituted Alkene

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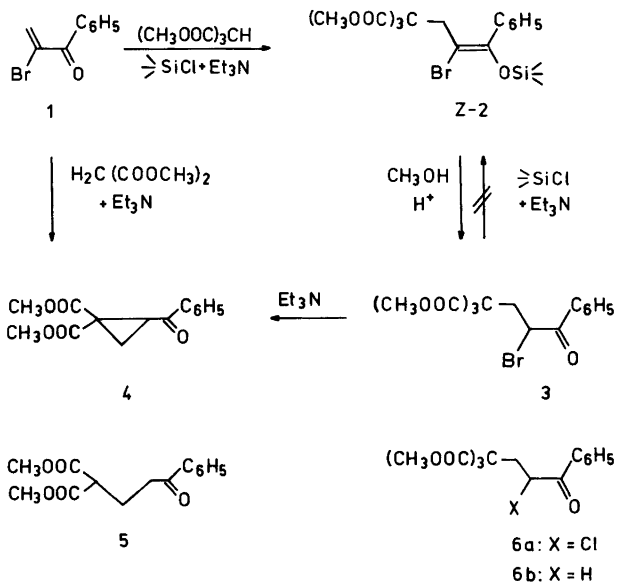
The trimethysilylated primary Michael adduct of tricarbomethoxymethane and 2-bromo-3-oxo-3-phenylpropene-1 is shown by ^{1}H NMR experiments and X-ray diffraction studies to possess the structure *Z-2*. Attempts to prepare the *E* isomer *via* the hydrolysis product of *Z-2* resulted in, *inter alia*, an intramolecular alkylation under decarbomethoxylation to give the cyclopropane ester *4*.

In a previous paper,[1] the observed stereoselectivity of the Michael reaction was attributed to stereoelectronic requirements of the transition state rather than to the bulk of the substituents. Evidence for the *trans* mode of Michael type additions of carbon nucleophiles to α,β-unsaturated aldehydes was based on NMR spectroscopy data.[1] In an attempt to evaluate a possible effect of more bulky substituents on the steric course of the Michael



Scheme 1. Preparation of enolate *Z-2* (see also Fig. 2) and derivatives.

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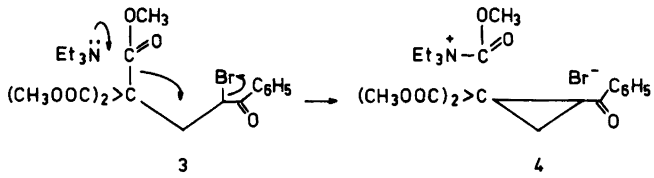

Scheme 2. Rationalization of the intramolecular decarboxylative alkylation giving cyclopropane *4*.

reaction, the bromoketone *1* was reacted with tricarbomethoxymethane under standard conditions[1] (Scheme 1). The crystalline, tetrasubstituted alkene *Z-2* was obtained in a high yield, accompanied by approximately 2 % of the *E* isomer. The structure of *Z-2* followed unequivocally from ^{1}H NMR NOE experiments and X-ray diffraction analyses in keeping with the proposed theory for the formation of enol ethers of type *2*.[1]

Attempts to prepare the isomeric enol ethers *E-* and *Z-2* from bromoketone *3* by the standard method involving triethylamine and trimethylsilyl chloride (Scheme 1) proved unsuccessful. The main product was the halogen exchange product *6a* and the cyclopropane ester *4*, the latter resulting from a base induced intramolecular decarbomethoxylative alkylation (Scheme 2). None of the silylated enol ethers was observed. The reaction was repeated, omitting the trimethylsilyl chloride; in this case only cyclopropane ester *4*, reduction product *6b*, and the reduced and decarbomethoxylated compound *5* were identified. These results illustrate the synthetic advantage of the modified Michael Reaction.[1]

Nuclear Overhauser enchancement measurements on the enol ether Z-2. Irradiation of the methylene group elicited positive signals from the phenyl group, but none from the trimethylsilyl group. Irradiation of the latter resulted in positive signals from the phenyl group, but none from the methylene group. Finally, irradiation of the phenyl group triggered positive signals from both of the other groups. These results are consistent only with the *Z* configuration previously suggested for the enol ether.

Crystal and molecular structure of the enol ether Z-2. The results of an x-ray diffraction study on single crystals of the enol ether *Z-2* and the atomic labels used are shown in Figs. 1 and 2. Details on the structural determination are given in the Experimental section.

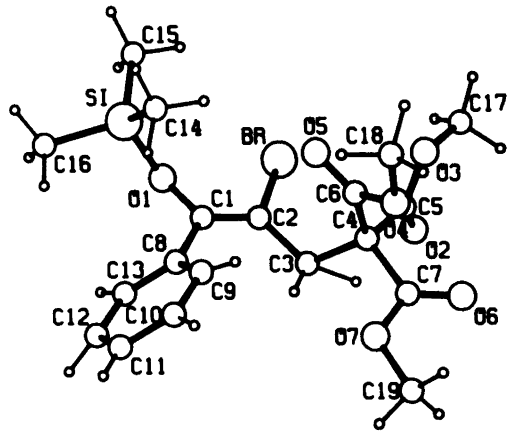

Fig. 1. Diagram showing the atomic lables used for describing the crystal structure.

Table 1. Selected bond distances (Å), angles (°) and torsion angles (°) in methyl(*Z*)-4-bromo-2,2-dicarbomethoxy-5-phenyl-5-trimethyl-silyloxy-pent-4-enoate.

C(1)–C(2)	1.343(8)	C(3)–C(4)	1.563(7)
C(1)–O(1)	1.360(6)	O(1)–Si	1.684(4)
C(1)–C(8)	1.485(8)	Si–C(14)	1.854(6)
C(2)–Br	1.911(5)	Si–C(15)	1.874(7)
C(2)–C(3)	1.503(7)	Si–C(16)	1.852(7)
O(1)–C(1)–C(2)	119.8(5)	C(1)–O(1)–Si	126.7(3)
O(1)–C(1)–C(8)	115.0(5)	O(1)–Si–C(14)	108.8(2)
C(2)–C(1)–C(8)	125.1(5)	O(1)–Si–C(15)	105.5(2)
C(1)–C(2)–Br	117.6(4)	O(1)–Si–C(16)	107.3(3)
C(1)–C(2)–C(3)	126.3(5)	C(14)–Si–C(15)	112.9(3)
C(3)–C(2)–Br	115.8(4)	C(14)–Si–C(16)	111.7(3)
C(2)–C(3)–C(4)	117.2(4)	C(15)–Si–C(16)	110.4(3)
O(1)–C(1)–C(2)–C(3)	172.2(7)	C(1)–C(2)–C(3)–C(4)	123.0(7)
O(1)–C(1)–C(2)–Br	−2.2(7)	C(2)–C(3)–C(4)–C(5)	74.6(7)
C(8)–C(1)–C(2)–C(3)	−4.3(7)	C(2)–C(1)–O(1)–Si	117.4(6)
C(8)–C(1)–C(2)–Br	−178.7(7)	C(1)–O(1)–Si–C(14)	−11.8(6)
		C(2)–C(1)–C(8)–C(1)	−53.7(7)

From Table 1, where the bond distances and angles are listed, it is seen that the C(1)–C(2) distance (1.343(8) Å) is close to that of a typical[2] double bond (*viz.* 1.337 Å). Accordingly, the atoms O(1), C(1), C(2), Br, C(3) and C(8) form a fairly planar ethylene group (*cf.* Fig. 2) with a root mean square (r.m.s.) deviation of 0.034 Å for a least-square plane through the atoms. The maximum deviation 0.049(4) Å for the O(1) atom indicates that the ethylene group is slightly but significantly distorted. As the two substituent atoms O(1) and Br occur on the same side in the double bond, the title compound is evidently the *Z* isomer in agreement with the ^{1}H NMR NOE experiments.

A least-square plane through the nonhydrogen atoms of the phenyl ring indicates planarity as the r.m.s. deviation is only 0.002 Å, compared to the average e.s.d. 0.006 Å of the atoms. The interplanar angle between the phenyl ring and the double bond is 126.8(7)°.

Planes through the nonhydrogen atoms of the three carbomethoxy groups and including the C(4) atom indicate that these are rather coplanar as the r.m.s. deviations are 0.009, 0.034, and 0.007 Å, respectively. The interplanar angles between these three planes range from 93.6 to 104.5(7)°.

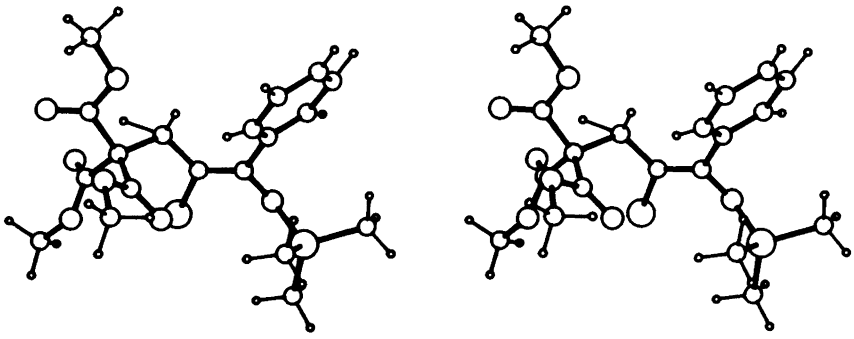

Fig. 2. Stereoplot showing the configuration of the molecule.

The bond distances and angles (Table 1) found in the present study are all close to the values expected.[2]

EXPERIMENTAL

Mass spectra were recorded on a VG Micromass 7070 F instrument, IP 70 eV, and NMR spectra on Bruker WH-90, HXE-90, and HX-270 instruments. Solvent: $CDCl_3$.

The ^1H nuclear Overhauser enhancement experiments were performed on a Bruker HX-270 NMR instrument in the difference mode as published previously.[3] The spectra were obtained on 0.02 M nondegassed samples in deuteriochloroform at 310 K.

2-Bromo-3-oxo-3-phenylpropene-1 1. The hydrochloride of β-dimethylamino-propiophenone[4] (71 g) was pyrolyzed in a flask fitted with a Vigreux column and an ice cooled receiver. Crude phenyl vinyl ketone[5] was collected until 150 °C/1.5 mmHg and shown by NMR to contain about 10 % of acetophenone. Yield 34 g. Steam distillation of the salt[6] gave even higher contents of acetophenone. The crude ketone was dissolved in carbon tetrachloride (25 ml), cooled in an ice bath, and bromine (12.5 ml) followed by triethylamine (36 ml) was added with stirring at 20–30 °C. Ice and water was added to the slurry and the aqueous phase was extracted twice with methylene chloride. The combined organic phases were dried ($MgSO_4$) and distilled rapidly to give the bromo ketone *1*. Yield 33 g (48 %, from the Mannich salt), b.p. 76–92 °C/0.2 mmHg. Redistillation at 78–88 °C/0.05 mmHg gave 25 g of the bromophenone and a large residue due to decomposition. ^1H NMR; δ 6.42 and 6.49 (2 H, AB system, J 2.2 Hz), 7.29–7.87 (5H, m). ^{13}C NMR: δ 128.4 and 129.6 (phenyl *o* and *m*), 134.9 and 129.4 (substituted phenyl carbon and C–Br), 130.0 (methylene), 133.1 (phenyl, *p*), and 190.0 (carbonyl).

Tricarbomethoxymethane[7] was prepared in analogy with the corresponding ethyl ester.[8] The magnesium must be dissolved in methanol (250 ml) before the esters are added, and no other solvents should be used.

Methyl (Z)-4-bromo-2,2-dicarbomethoxy-5-trimethyl-silyloxypent-4-enoate Z-2. To a mixture of the phenone (*1*, 10.6 g, 50 mmol), tricarbomethoxymethane (9.5 g, 50 mmol), and chlorotrimethylsilane (12 ml) in carbon tetrachloride (40 ml), triethylamine (15 ml) was added with stirring. The temperature was kept near 30 °C with an ice bath and stirring was continued at 28–32 °C for a total of 2 h. The slurry was cooled to below −20 °C and ice (20–30 g) added. Methylene chloride may be added if the product crystallizes. The aqueous phase was extracted twice with methylene chloride, the combined organic fractions dried (Na_2SO_4) and concentrated *in vacuo*. Carbon tetrachloride (until a total of about 30 ml) and light petroleum (30 ml) were added and the product was allowed to crystallize for 20 h at −23 °C. The colourless crystals were washed with cold light petroleum and dried at 20 °C/0.1 mmHg. Yields 20.0–20.9 g (85–88 %), m.p. 75–77 °C. Two recrystallizations from diethyl ether gave material with m.p. 75–77 °C. Anal. $C_{19}H_{25}BrO_7Si$: C,H,Br. ^1H NMR: δ 0.027 (9H, s), 3.53 (2H, s), 3.69 (9H, s), 7.20–7.41 (5H,m). ^{13}C NMR: δ 0.2 ($(CH_3)_3Si$), 38.5 (C3), 52.9 (methoxy), 64.1 (C2), 103.0 (C4), 128.1, 128.5, and 128.6 (phenyl), 136.0 (substituted phenyl carbon), 150.5 (C5), 166.0 (Cl). MS [*m/z* (% rel. int.)]: 474 and 472 (1, M), 393 (54), 333 (6), 283 and 285 (16), 131 (20), 105 (44), 103 (18), and 73 (100).

The enol ether is unstable when exposed to the atmosphere due to hydrolysis, even in deuteriochloroform solution (see below).

The synthesis of *2* was repeated employing bromoketone *1* in excess (11.5 g, 54 mmol) to ensure consumption of all of the tricarbomethoxymethane. Yield 21.0 g (88 %). The mother liquor from the crystallization was concentrated *in vacuo* (0.1 mmHg) to give 3.5 g of semicrystalline product. NMR spectroscopical analysis disclosed the presence of approximately 1.8 g of *Z-2*, 0.4 g of ketone *3* (product of hydrolysis), 0.9 g of ketone *1* (starting material used in excess), and 0.4 g of a compound assumed to be *E-2*. Identification of the latter rests on the presence of two resonances of equal intensity, one at δ −0.03 (trimethylsilyl) and one at δ 3.79, the latter assigned to the three ester methyl groups, but coincident with methoxy from any tricarbomethoxymethane present. Lack of any resonance signal at δ 4.43 (H–C), however, precludes the presence of any material of the latter type. Other resonances attributable to the presence of *E-2* were not observed.

Methyl 4-bromo-5-oxo-2-bis(methoxycarbonyl)-5-phenylpentanoate 3. The enol ether *Z-2* (5.0 g) was suspended in methanol (5 ml) and concentrated hydrochloric acid (0.2 ml) was added. The mixture was stirred at room temperature for 2 min. and kept overnight at −20 °C. The crystals were isolated, washed with cold methanol, and recrystallized from a mixture of toluene (10 ml) and ligroin (5 ml, b.p. 80−100 °C) to give colourless crystals (3.5 g, 82 %) m.p. 83−85 °C, unchanged after two additional recrystallizations from ethanol. Anal. $C_{16}H_{17}BrO_7$: C,H,Br. ^{1}H NMR: δ 3.00 (1H, dd, J 3.2 and 15.0 Hz), 3.64 (1H, partly hidden dd, J 8.5 and 15.0 Hz), 3.71 (9H, s), 5.47 (1H, dd, J 3.2 and 8.5 Hz), 7.40−8.02 (5H, m). ^{13}C NMR: δ 36.0 (C3), 41.0 (C4), 53.4 (methoxy), 64.8 (C2), 128.7 (phenyl, *o* and *m*), 133.5 (phenyl, *p*), 134.0 (substituted carbon in phenyl), 166.4 (Cl), 191.5 (C5).

Reactions of bromoketone 3. An ampoule was charged with the ketone *3* (409 mg), triethylamine (1.6 ml), chlorotrimethylsilane (0.6 ml), and carbon tetrachloride (2 ml), evacuated and kept at 90−95 °C for 22 h. The dark coloured product was treated with ice and water, extracted with methylene chloride, dried and concentrated *in vacuo*. NMR analysis showed a 1 to 3 molar ratio of the cyclopropane ester *4* and chloroketone *6a*, respectively. No resonance signals consistent with either *Z-2* or *E-2* were observed. The semicrystalline product was dissolved in ethanol (2 ml), treated with active carbon and crystallized at −70 °C, filtered and washed with cold ethanol. Yield of chloroketone *6a*, 113 mg, m.p. 93−95 °C. Recrystallization from toluene: m.p. 94−96 °C. Anal. $C_{16}H_{17}ClO_7$: C,H,Cl. MS [*m/z* (% rel. int.)]: 358 (0.2), 356 (0.5, M), 281 (0.8), 190 (1.4), 159 (1.2), 106 (9), 105 (100), 77 (19). ^{1}H NMR: δ 2.84 (1H, dd, J 4.8 and 15.0 Hz), 3.50 (1H, dd, J 7.0 and 15.0 Hz), 3.73 (9H, s), 5.51 (1H, dd, J 4.8 and 7.0 Hz), 7.31−7.64 and 7.89−8.11 (5H, m). Preparative TLC (silicagel and a mixture of pentane (100 ml) and ethyl acetate (20 ml)) of the mother liquor gave a slightly impure fraction of the cyclopropane ester *4*, see below.

An ampoule was charged with the ketone *3* (464 mg) and triethylamine (1.0 ml), evacuated, and kept at 92−96 °C for 17 h. Methylene chloride was added, the product washed with dilute hydrochloric acid, dried and concentrated *in vacuo*. Analysis (NMR) showed the presence of an approximately 2 to 1 to 1 molar ratio of the cyclopropane ester *4*, the keto ester *6b*, and the decarbomethoxylated keto ester *5*, see below.

1-Benzoyl-2,2-bis-methoxycarbonyl-cyclopropane 4. Bromoketone *1* (4.22 g), dimethyl malonate (2.5 ml), triethylamine (4 ml) and carbon tetrachloride (8 ml) was kept at about 60 °C for 3 h. Ice water was added, extracted twice with carbon tetrachloride, and the combined fractions distilled, b.p. 155−166 °C/0.6 mmHg. Yield 3.44 g, 65 %. Crystallization from ethanol and recrystallization from a ligroin/toluene mixture gave a product with m.p. 69−71 °C, *cf.* m.p. 74 °C.[9] MS [*m/z* (% rel. int.)]: 262 (1.4, M), 231 (12), 230 (21), 202 (14), 105 (100), 77 (36). ^{1}H NMR: δ 1.78 (1H, dd, J 4.0 and 8.5 Hz), 2,21 (1H, dd, J 4.0 and 6.8 Hz), 3.54 (1H, partly hidden dd, J 6.8 and 8.5 Hz), 3.67 (3H, s), 3.78 (3H, s), 7.27−7.67 and 7.84−8.07 (5H, m). ^{13}C NMR; δ 20.8, 30.9, 38.8 (CH_2, CH, and C of cyclopropane ring, respectively), 52.6 and 53.1 (methoxy of ester), 128.3 and 128.6 (*o* and *m*), 133.4 (*p* phenyl), 136.9 (substituted carbon of phenyl), 166.2 and 169.3 (ester carbonyl), 194.5 (carbonyl). 2,4-Dinitrophenylhydrazone, m.p. variable, depending on the solvent employed, 170−182 °C (lit.[9] 169 °C). ^{1}H NMR: δ 2.11 (2H, m), 3.03 (1H, t, J 8.7 Hz), 3.28 (3H, s), 3.90 (3H, s), 7.27−7.82 (5H, m), 8.04 (1H, d. J 9.5 Hz), 8.31 (1H, dd, J 9.5 and 2.5 Hz), 9.12 (1H, d, J 2.5 Hz), 11.74 (1H, s).

4,4,4-Trismethoxycarbonyl-1-oxo-1-phenylbutane 6b was obtained from phenyl vinyl ketone as for the corresponding bromo compound *3* above.[1] m.p. 71−73 °C. Anal. $C_{16}H_{18}O_7$: CH. ^{1}H NMR: δ 2.47−2.72 and 3.11−3.36 (4 H, m, symmetry centre at δ 2.91), 3.79 (9 H, s), 7.29−7.58 and 7.84−8.07 (5 H, m). ^{13}C NMR: δ 27.3 and 34.1 (CH_2), 53.1 (ester methyl), 64.7 (C4), 127.9 and 128.5 (phenyl, *o* and *m*, 133.0 (phenyl, *p*), 136.6 (substituted carbon in phenyl), 167.2 (ester carbonyl), 198.2 (carbonyl). MS [*m/z* (% rel. int.)]: 322 (3.9, M), 291 (1.7), 231 (1.7), 190 (6.6), 133 (9.4), 105 (100), 77 (20).

4,4-Bismethoxycarbonyl-1-oxo-1-phenylbutane 5. The ketoester *6b* (322 mg) was dissolved in a solution of sodium (36 mg) in methanol (2ml) and kept at 20 °C for 3 h. Water was added and the product extracted with methylene chloride, dried and concentrated *in vacuo*. Yield 173 mg (66 %), m.p. 41−42 °C. Recrystallization from aqueous methanol, 115 mg, m.p. 42−44 °C.[9] ^{1}H NMR: δ 2.33 (2 H, q, J 7 Hz), 3.07 (2 H, t, J 7 Hz), 3.54 (1 H, t, J 7 Hz), 3.74 (6 H, s), 7.27−8.04 (5 H, m).

X-ray structure determination. A crystalline specimen of the enol ether *2* was obtained by

slow evaporation of a diethyl ether solution. A single crystal with the dimensions 0.20×0.36×0.46 mm was selected for data collection on a CAD4 diffractometer, using graphite monochromatized Cu$K\alpha$ (λ=1.54184 Å) radiation. The unit cell parameters at 21(2) and −150(4) °C were determined from the angular settings of 12 very strong reflections with $12.3<\theta<16.3°$. Data were collected at −150(4) °C.

CRYSTAL DATA

$C_{19}H_{25}BrO_7Si$, Mw=473.4
Space group $Pna2_1$ (No. 33), Z=4, μ(Cu$K\alpha$)=34.6 cm^{-1}
a=13.769(2)Å, b=7.632(2)Å, c=20.567(4)Å, D_x=1.455(1) g cm^{-3}
at t=−150(4)°C; a=13.949(4)Å, b=7.652(4)Å, c=20.771(8) Å
D_x=1.418(1) g cm^{-3} at t=21(2) °C.

2255 unique reflections with $\sin\theta/\lambda$ less than 0.625 were collected with the $\omega-2\theta$ scan technique, 2229 reflections had $\sigma(I)/I$ (σ from counter statistics) less than 0.2 and were

Table 2. Fractional atomic coordinates (×10^4) for the nonhydrogen atoms of methyl(*Z*)-4-bromo-2,2-dicarbomethoxy-5-phenyl-5-trimethyl-silyloxy-pent-4-enoate and equivalent isotropic thermal parameters (Å^2×10^4) estimated as one third of the trace of the orthogonalized U_{ij} tensor. The *z* coordinate of Br was fixed to O to define the origin along the planar *c* axis.

Atom	*x*	*y*	*z*	*u*
Br	1489(1)	−2156(1)	0	225(2)
Si	3200(1)	−887(2)	−1566(1)	212(4)
C(1)	1500(3)	243(7)	−1029(3)	167(14)
C(2)	1158(4)	−12(6)	−425(3)	177(12)
C(3)	464(3)	1160(6)	−65(3)	171(13)
C(4)	805(4)	1890(6)	607(3)	158(13)
C(5)	766(4)	456(7)	1124(3)	174(13)
C(6)	1838(4)	2695(7)	571(3)	168(15)
C(7)	88(4)	3314(7)	843(3)	185(13)
O(1)	2034(3)	−1031(5)	−1323(2)	209(10)
O(2)	70(3)	−453(5)	1210(2)	215(11)
O(3)	1576(3)	395(6)	1478(2)	233(12)
C(17)	1568(4)	−905(10)	1983(4)	339(20)
O(4)	1892(3)	4182(5)	907(2)	218(11)
O(5)	2505(3)	2030(5)	295(2)	257(12)
C(18)	2871(4)	4897(9)	948(3)	302(17)
O(6)	−198(3)	3377(6)	1397(2)	266(12)
O(7)	−160(3)	4406(5)	371(2)	206(11)
C(19)	−818(5)	5805(7)	561(3)	288(17)
C(8)	1302(4)	1807(7)	−1438(3)	176(13)
C(9)	1479(3)	3511(8)	−1218(3)	205(16)
C(10)	1308(4)	4930(7)	−1621(3)	231(14)
C(11)	954(4)	4671(7)	−2253(3)	250(15)
C(12)	783(4)	2993(7)	−2471(3)	250(16)
C(13)	950(3)	1556(7)	−2066(3)	217(13)
C(14)	3740(4)	1130(8)	−1217(3)	289(15)
C(15)	3799(6)	−2936(8)	−1265(4)	384(21)
C(16)	3198(5)	−834(9)	−2466(3)	352(19)

considered as significant observations. The reflections were corrected for Lorentz and polarization effects, as well as for absorption effects. The transmission factors ranging from 0.26 to 0.58, were calculated with a 10×10×10 grid used for the Gaussian quadrature.

The structure was solved by fairly straight forward application of conventional heavy atom techniques. The structural parameters were refined by least-squares techniques, minimizing $\Sigma w(\Delta F)^2$, allowing all nonhydrogen atoms to vibrate anisotropically, while the hydrogens were kept isotropic. The methyl groups were constrained to have ideal geometry (bond angles 109.5° and C–H distance 1.08 Å) and with a common isotropic temperature factor for all methyl hydrogens. The remaining hydrogens were constrained to have a common isotropic temperature factor, but their positions were not constrained. The refinement converged to an R-value ($R<\Sigma|\Delta F|/\Sigma F_o$) of 0.071 with the weights estimated as proportional to $1/(\sigma^2 (F_o)+0.0002F_o^2)$. The R-value of a model with the z-axis inverted became 0.083 and this model was discarded. An inspection of the F_o/F_c list showed that the strongest reflections suffered severely from extinction. By omitting the 75 strongest reflections, the refinement converged to an R-value of 0.060 for the remaining 2154 reflections (293 parameters varied). The atomic coordinates are given in Table 2 (Lists of anisotropic temperature factors and structure factors are available from one of the authors (R.N.) on request). The atomic labels used for describing the crystal structure are shown in Fig. 1. The atomic scattering factors used were those of the International Tables for X-ray Crystallography.[10] All the crystallographic calculations were performed with the program systems SHELX[11] and XTAPL.[12] The molecular plots were obtained with the program PLUTO.[13]

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