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Studies on Fungal Products. XVIII.[1] Isolation and Structures of a New Fungal Depsidone Related to Nidulin and a New Phthalide from *Emericella unguis*

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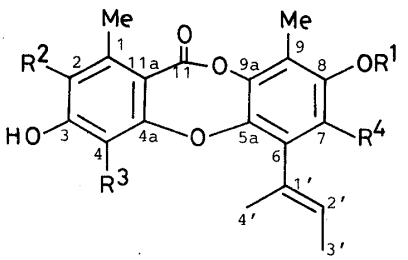
Two new compounds, 2-chlorounguinol (**1**), $C_{19}H_{17}ClO_5$, and 3-ethyl-5,7-dihydroxy-3,6-dimethylphthalide (**5**), $C_{12}H_{14}O_4$, were isolated from the dichloromethane extract of the culture filtrate of *Emericella unguis* (anamorph: *Aspergillus unguis*), together with nidulin (**2**) and unguinol (**4**). The structure of 2-chlorounguinol (**1**) was determined on the basis of spectroscopic investigations and X-ray crystallography, and that of the phthalide (**5**) was confirmed by spectroscopic investigations of several derivatives. 2-Chlorounguinol (**1**) is a fungal depsidone related to nidulin (**2**), originally isolated from *E. nidulans*. This is the first example of the isolation of a 3,3-disubstituted phthalide as a naturally occurring compound.

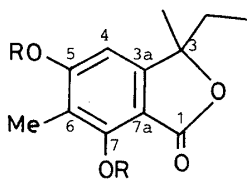
Keywords——*Emericella unguis*; *Aspergillus unguis*; depsidone; nidulin; unguinol; 2-chlorounguinol; phthalide; 3-ethyl-5,7-dihydroxy-3,6-dimethylphthalide

Nidulin (**2**), nornidulin (**3**), unguinol (tridechloronornidulin) (**4**), and related compounds have been isolated from non-ascosporic strains *Emericella unguis* MALLOCH *et* CAIN (anamorph: *Aspergillus unguis* (EMILE-WEIL *et* GAUDIN) THOM *et* RAPER), strains IMI 138767[2] and NRRL 5250.[3] In the course of screening of the monoamine oxidase inhibitory potency in *Emericella* spp., the ethyl acetate extract of *E. unguis*, strain IFM 42017, was found to have an inhibitory ratio of 18.5% at the concentration of 0.1 mg/ml.[4] In order to isolate the active compounds, the dichloromethane extract of the culture filtrate of the above strain was examined, and two new compounds **1** and **5** were isolated along with **2** and **4**. The structural elucidation of **1** and **5** is mainly reported in this paper.

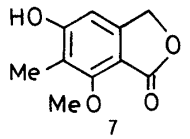
Compound **1**, finally named 2-chlorounguinol, mp 228—230 °C, gave molecular ion



1 : $R^1=R^3=R^4=H$, $R^2=Cl$
2 : $R^1=Me$, $R^2=R^3=R^4=Cl$
3 : $R^1=H$, $R^2=R^3=R^4=Cl$
4 : $R^1=R^2=R^3=R^4=H$



5 : R=H
6 : R=Ac



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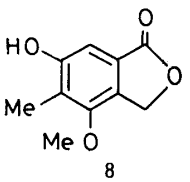

8

Chart 1

TABLE I. ^{1}H-NMR Chemical Shifts of 2-Chlorounguinol (**1**), Nidulin (**2**), and Unguinol (**4**) in $CDCl_3$

Proton No.	**1**	**2**	**4**[a]
1-Me	2.52	2.52	2.39
2-H			6.32[b]
3-OH	6.05[b]	6.38	—
4-H	6.59[c]		6.52[b]
7-H	6.41[c]		6.40
8-OH	4.77[b]		—
8-OMe		3.79	
9-Me	2.21	2.33	2.14
2′-H	5.56	5.42	5.54
3′-H (Me)	1.82	1.82	1.84
4′-H (Me)	2.05	1.96	2.06

a) This compound was measured in CD_3OD. *b*, *c*) The assignments may be reversed.

TABLE II. ^{13}C-NMR Chemical Shifts of 2-Chlorounguinol (**1**) and Nidulin (**2**) in $CDCl_3$

Carbon No.	**1**	**2**
1	141.24	140.24
2	119.17	119.69
3	161.48	157.32
4	105.50	110.34
4a	156.02	151.95
5a	141.21	145.44
6	132.50	129.48
7	111.25	124.44
8	151.98	152.70
9	115.10	123.53
9a	143.35	141.78
11	162.73	161.59
11a	115.21	115.97
1-Me	18.06	18.88
9-Me	9.00	10.46
8-OMe		60.53
1′	135.70	136.09
2′	125.26	128.23
3′	13.57	14.18
4′	17.33	17.52

peaks at *m/z* 360 and 362 in the ratio of 3 : 1 in electron impact ionization (EI) mass (MS) spectrometry, and elemental analysis confirmed the molecular formula of **1** as $C_{19}H_{17}ClO_5$. The presence of the ester carbonyl was shown by the absorption at 1720 cm^{-1} in the infrared (IR) spectrum of **1**, as well as those of **2** and **4**. The proton nuclear magnetic resonance (^{1}H-NMR) spectrum of **1** is similar to that of **4**, except for the disappearance of one of the *meta*-coupled aromatic protons (Table I). The carbon-13 nuclear magnetic resonance (^{13}C-NMR) spectrum of **1** is also similar to that of **2**, except for the multiplicity of two carbons bearing the chlorine atoms in **2** (Table II). The above results indicated that compound **1** was a fungal depsidone which was closely related to nidulin (**2**) and unguinol (**4**), considering the co-occurrence of **2** and **4** in our strain.

A comparison of the molecular formula of 2-chlorounguinol (**1**) with that of unguinol (**4**), $C_{19}H_{18}O_5$, showed that one hydrogen atom in **4** was replaced with a chlorine atom in **1**. The aromatic methyl protons that appeared downfield (δ 2.52) in **1** were at the same chemical shift as those of nidulin (**2**), not of **4** (δ 2.39), which showed that the chlorine atom is attached to the aromatic ring bearing the carbonyl group of the ester. Two aromatic proton signals in **1** appeared at δ 6.41 and 6.59, both as broad singlets. The above results confirmed that compound **1** was chlorinated at the C-2 or C-4 position of unguinol (**4**).

In order to determine the exact structure of **1**, especially the location of the chlorine atom and the stereochemistry of the 1-methyl-1-propenyl group, an X-ray structure analysis of **1** was undertaken. Since crystals of **1** recystallized from benzene were not suitable for X-ray analysis, various solvents were used for crystallization of **1**. Crystals of **1** monohydrate grew as colorless prisms when a drop of water was added to a chloroform solution of **1**. The molecular structure of **1** monohydrate is illustated in Fig. 1. Therefore the structure of 2-chlorounguinol was confirmed as 2-chloro-3,8-dihydroxy-1,9-dimethyl-6-(1-methyl-1-propenyl)-11*H*-dibenzo[*b*,*e*][1,4]-dioxepin-11-one, depicted as **1**. Bond lengths and angles are also shown in Fig. 1. These values are not significantly different from the expected ones. Based on the O(3)–O(8), O(8)–O(w), and O(w)–O(11) distances (2.78, 2.87, and 2.78 Å, re-

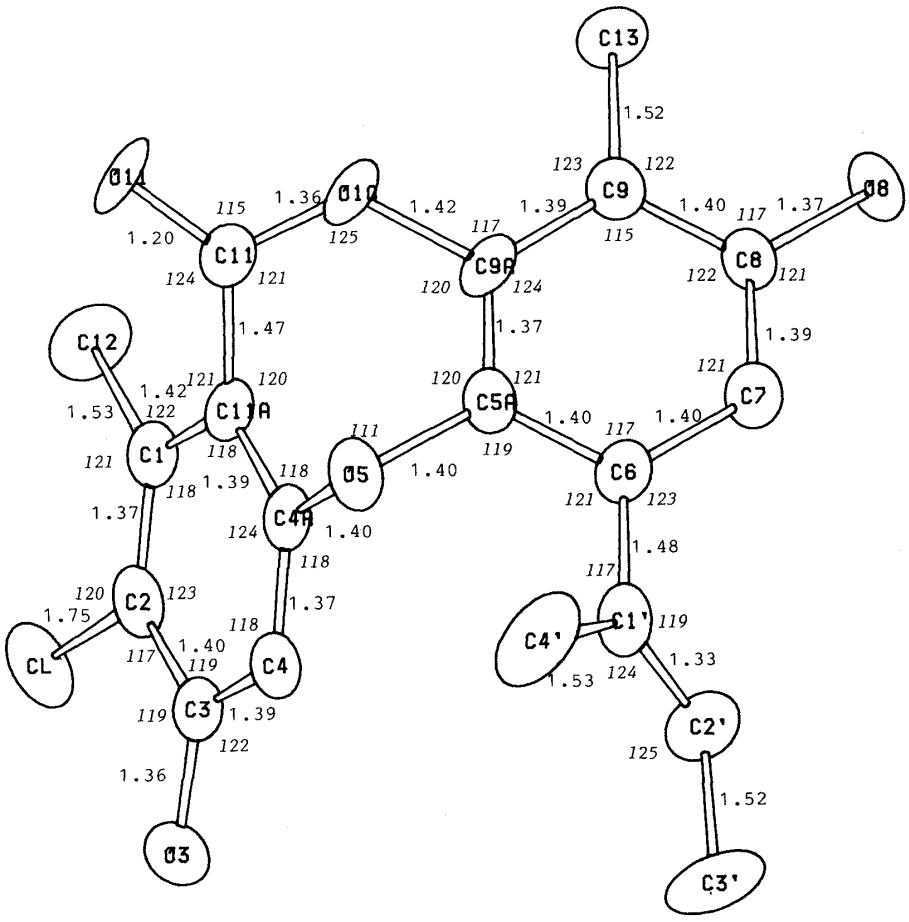

Fig. 1. Perspective View of the Crystal Structure of 2-Chlorounguinol (**1**) Monohydrate with Thermal Ellipsoids at 50% Probability

Bond lengths (Å) and angles (°) are shown.

spectively), O(3)–H–O(8)–H–O(W)–H–O(11) seem to be intermolecular hydrogen bonds. The molecules are packed together mainly through the hydrogen bonding between the molecules of **1** and water in the crystals.

The molecular formula of compound **5**, mp 133—135 °C, was confirmed as $C_{12}H_{14}O_4$ by the EI-MS spectrum and the elemental analysis. The ^{1}H-NMR signals at δ 6.38 and 7.90, which disappeared on addition of D_2O, were assigned to two phenolic hydroxyl protons. On acetylation, **5** afforded a diacetate (**6**), $C_{16}H_{18}O_6$, which showed ^{1}H-NMR signals at δ 2.08 (3H, s) and 2.36 (3H, s) assigned to the methyl protons of two acetoxyl groups. Thus two of the four oxygen functions in **5** have been assigned to two phenols. The remaining two oxygens must be an ester in view of the ^{13}C-NMR signals at δ 172.14 and the IR absorption maximum at 1700 cm^{-1} in **5**.

The aromatic methyl protons and the aromatic proton in **5** appeared at δ 2.14 (3H, s) and 6.34 (1H, s) in the ^{1}H-NMR spectrum and the corresponding carbons at δ 7.35 (Q) and 99.73 (D) in the ^{13}C-NMR spectrum, respectively. The ^{1}H-NMR signals at δ 0.78 (3H, t), 1.95 (2H, m), and 1.59 (3H, s) in **5**, which corresponded to the ^{13}C-NMR signals at δ 7.89 (Qt), 32.89 (Tm), and 25.65 (Qt), respectively, were assigned to an aliphatic ethyl group and an aliphatic methyl group. The other signal at δ 89.73 (Sm) in the ^{13}C-NMR spectrum was assigned to the quaternary carbon attached to the oxygen atom of the ester. The above results indicated that compound **5** was a 3-ethyl-3-methylphthalide bearing two hydroxyl groups and one methyl group in the aromatic ring.

The ^{13}C-NMR signal at δ 125.01 (Sqd) in the diacetate (**6**) was assigned to the aromatic carbon bearing the methyl group at the *meta*-position to the aromatic proton in view of the multiplicity and the coupling constant of this signal. The signals of aromatic carbons bearing the acetoxyl groups at δ 154.96 (Sqd) and 147.81 (Sq) were assigned to the carbons vicinal to the aromatic methyl group; the former was also vicinal to the aromatic proton. The above results confirmed the arrangement of the substituents except for the junction between these substituents and the five-membered lactone. The lactone carbonyl at 1700 cm^{-1} in the IR spectrum of **5** was shifted to 1760 cm^{-1} in that of **6**, and the aromatic proton at δ 6.34 in **5** corresponded well to that of nidulol (**7**) (δ 6.59) rather than silvaticol (**8**) (δ 7.08). These compounds have previously been isolated from *Aspergillus silvaticus* FENNELL *et* RAPER.[5] Furthermore, a 13.5% nuclear Overhauser enhancement of the aromatic proton signal at δ 6.98 in **6** was observed when the methyl protons at δ 1.62 were irradiated. Therefore the aromatic proton must be attached at the C-4 position. From the above results, the structure of compound **5** was confirmed as 3-ethyl-5,7-dihydroxy-3,6-dimethylphthalide.

2-Chlorounguinol (**1**) is a new fungal depsidone related to nidulin (**2**) and unguinol (**4**), already isolated from *Aspergillus unguis*.[3,4] 3-Ethyl-5,7-dihydroxy-3,6-dimethylphthalide (**5**) is the first example of a 3,3-disubstituted phthalide to be isolated from a natural source. The monoamine oxidase inhibitory potency of these compounds was tested according to Kraml's method[6] by using kynuramine as the substrate, but compounds **1**, **2**, **4**, and **5** had inhibitory ratios of only 2.0 to 5.2% even at the concentration of 10^{-4} mol/l. Thus the active compound of *E. unguis* remains to be successfully identified.

Experimental

Melting points were determined on a Yanagimoto micro-melting point apparatus and are uncorrected. IR and ultraviolet (UV) spectra were recorded on a JASCO IR-810 spectrophotometer and a Hitachi 124 spectrophotometer, respectively. EI-MS spectra were obtained on a JEOL JMS-D 300 spectrometer. ^{1}H-NMR spectra were measured with a JEOL JNM-FX 100 spectrometer at 99.60 MHz, whereas ^{13}C-NMR spectra were recorded on a JEOL JNM-GX 400 spectrometer at 100.43 MHz, using tetramethylsilane as an internal standard. The coupling patterns are indicated as follows: singlet = S or s, doublet = D or d, triplet = T or t, quartet = Q or q, multiplet = m, and broad = br. Capital letters refer to the pattern resulting from directly bonded coupling ($^1J_{C,H}$). Column chromatography was performed using Kieselgel 60 (Art. 7734; Merck). Low-pressure liquid chromatography (LPLC) was performed with a Chemco Low-Prep pump (81-M-2) and a glass column (150 × 10 mm) packed with Silica gel CQ-3 (30—50 μm; Wako). Thin layer chromatography (TLC) was conducted on precoated Kieselgel 60 GF_{254} plates (Art. 5715; Merck). Spots on TLC were detected under UV light, and/or by exposure to iodine vapor.

Isolation of 2-Chlorounguinol (1) and 3-Ethyl-5,7-dihydroxy-3,6-dimethylphthalide (5)——*Emericella unguis*, strain IFM 42017, was cultivated at 28 °C for 3 weeks in Czapek-Dox medium supplemented with 0.2% yeast extract. The culture filtrate (15 l) was extracted with dichloromethane at pH 4, and the organic layer was dried over Na_2SO_4 and concentrated under reduced pressure to give the extract (2.75 g). This extract (2.0 g) was purified by column chromatography with chloroform–methanol (100 : 1, v/v) followed by recrystallization from cyclohexane to obtain nidulin (**2**) (210 mg), and with chloroform–methanol (50 : 1, v/v) followed by LPLC to give two fractions. The fraction eluted with chloroform–methanol (100 : 1, v/v) afforded unguinol (**4**) (30 mg). The fraction eluted from chloroform was further purified by LPLC with benzene to give 2-chlorounguinol (**1**) (70 mg) and with benzene–acetone (100 : 1, v/v) to obtain 5,7-dihydroxy-3,6-dimethyl-3-ethylphthalide (40 mg) (**5**).

2-Chlorounguinol (**1**): Colorless needles from benzene, mp 228—230 °C. IR ν_{max}^{KBr} cm^{-1}: 3400 (OH), 1720 (COO). UV λ_{max}^{MeOH} nm (log ε): 280 (3.90), 310 (3.51). EI-MS *m/z*: 362 [M$^+$(^{37}Cl), 2%], 360 [M$^+$(^{35}Cl), 5], 347 [M − Me(^{37}Cl), 3], 345 [M − Me(^{35}Cl), 10], 40 (100). *Anal.* Calcd for $C_{19}H_{17}ClO_5$: C, 63.25; H, 4.75. Found: C, 63.41; H, 5.00. ^{1}H-NMR ($CDCl_3$) δ: 1.82 [3H, dq, J = 6.8, 1.0 Hz, –C(Me)=CH–Me], 2.05 [3H, br s, –C(Me)=CH–Me], 2.21 (3H, s, 9-Me), 2.52 (3H, s, 1-Me), 4.77 (1H, br s, aromatic OH), 5.56 [1H, br q, J = 6.8 Hz, –C(Me)=CH-Me], 6.05 (1H, s, aromatic OH), 6.41 (1H, br s, 7-H), 6.59 (1H, br s, 4-H). ^{13}C-NMR ($CDCl_3$) δ: 9.00 (Qd, J = 129 Hz, 9-Me), 13.57 (Qd, J = 126, 3 Hz, –C(Me)=CH–Me), 17.33 (Qd, J = 129, 9 Hz, –C(Me)=CH–Me), 18.06 (Q, J = 129 Hz, 1-Me), 105.50 (Dd, J = 167, 4 Hz, C-4), 111.25 (D, J = 161 Hz, C-7), 115.10 (Sm, C-11a), 115.21 (Sq, J = 6 Hz, C-9), 119.17 (Sm, C-2), 125.26 (Dm, J = 152 Hz, –C(Me)=CH-Me), 132.50 (Sm, C-6), 135.70 (Sbr, –C(Me)=CH–Me), 141.21 (Sd, J = 6 Hz, C-5a), 141.24 (Sq, J = 3 Hz, C-1), 143.35 (Sq, J = 2 Hz, C-9a), 151.98 (Sq, J = 3 Hz, C-8), 156.02 (S, C-4a), 161.48 (Sd, J = 4 Hz, C-3), 162.73 (S, C-11).

3-Ethyl-5,7-dihydroxy-3,6-dimethylphthalide (**5**): Colorless needles from cyclohexane, mp 133—135 °C. $[\alpha]_D^{15} \pm 0°$ ($c=1.0$, $CHCl_3$). IR ν_{max}^{KBr} cm^{-1}: 3400 (OH), 1700 (COO), 1630, 1620. UV λ_{max}^{MeOH} nm (log ε): 261 (4.25), 283 sh (3.81). EI-MS *m/z*: 222 (M^+). *Anal.* Calcd for $C_{12}H_{14}O_4$: C, 64.85; H, 6.35. Found: C, 65.05; H, 6.37. ^{1}H-NMR ($CDCl_3$) δ:0.78 (3H, t, $J=7.3$ Hz, $-CH_2-C\underline{H}_3$), 1.59 (3H, s, –Me), 1.95 (2H, m, $-C\underline{H}_2-CH_3$), 2.14 (3H, s, aromatic Me), 6.34 (1H, s, aromatic proton), 6.38 (1H, brs, aromatic OH), 7.90 (1H, s, aromatic OH). ^{13}C-NMR ($CDCl_3$) δ:7.35 (Q, $J=129$ Hz, 6-Me), 7.89 (Qt, $J=129$, 5 Hz, $-CH_2-\underline{C}H_3$), 25.65 (Qt, $J=129$, 3 Hz, –Me), 32.89 (Tm, $J=129$ Hz, $-\underline{C}H_2-CH_3$), 89.73 (Sm, C-3), 99.73 (D, $J=161$ Hz, C-4), 104.12 (Sd, $J=7$ Hz, C-7a), 110.69 (Sm, C-6), 152.73 (Sm, C-3a), 155.59 (Sm, C-7), 161.82 (Sm, C-5), 172.14 (S, C-1).

Acetylation of 3-Ethyl-5,7-dihydroxy-3,6-dimethylphthalide (5)——Compound **5** (50 mg) was dissolved in pyridine (2 ml) containing acetic anhydride (1 ml) and the solution was kept at room temperature for 1 d. The reaction mixture was poured into ice-water and extracted with chloroform. The extract was evaporated and the residue was purified by LPLC with benzene to give a diacetyl derivative (40 mg) as a colorless viscous oil. IR ν_{max}^{KBr} cm^{-1}: 1760, 1720 (OAc, COO). UV λ_{max}^{MeOH} nm (log ε): 238 (4.04), 280 (3.45). EI-MS *m/z*: 306 (M^+). *Anal.* Calcd for $C_{16}H_{18}O_6$: C, 62.74; H, 5.92. Found: C, 62.74; H, 5.83. ^{1}H-NMR ($CDCl_3$) δ: 0.78 (3H, t, $J=7.3$ Hz, $-CH_2-C\underline{H}_3$), 1.62 (3H, s, –Me), 1.87 (1H, dq, $J=14.6$, 7.3 Hz, $-C\underline{H}_2-CH_3$), 2.02 (1H, dq, $J=14.6$, 7.3 Hz, $-C\underline{H}_2-CH_3$), 2.08 (3H, s, OAc), 2.36 (3H, s, OAc), 2.44 (3H, s, aromatic Me), 6.98 (1H, s, aromatic proton). ^{13}C-NMR δ:7.85 (Qt, $J=124$, 4 Hz, $-CH_2-\underline{C}H_3$), 9.61 (Q, $J=128$ Hz, 6-Me), 20.55 (Q, $J=129$ Hz, $-OCO\underline{C}H_3$), 20.87 (Q, $J=129$ Hz, $-OCO\underline{C}H_3$), 25.55 (Qt, $J=128$, 2 Hz, –Me), 33.03 (Tm, $J=127$ Hz, $-\underline{C}H_2CH_3$), 87.19 (Sm, C-3), 112.34 (D, $J=162$ Hz, C-4), 115.97 (Sd, $J=7$ Hz, C-7a), 125.01 (Sqd, $J=5$, 7 Hz, C-6), 147.81 (Sq, $J=4$ Hz, C-7), 152.97 (Sm, C-3a), 154.96 (Sqd, $J=4$, 4 Hz, C-5), 166.57 (s, C-1), 168.07 (Sq, $J=5$ Hz, $-O\underline{C}OCH_3$), 168.36 (Sq, $J=5$ Hz, $-O\underline{C}OCH_3$).

X-Ray Structure Analysis of 2-Chlorounguinol (1) Monohydrate——Crystals of **1** were grown from chloroform containing a drop of water to yield **1** monohydrate as colorless prisms, mp 228 °C.

Crystal Data: $C_{19}H_{17}ClO_5 \cdot H_2O$; $M_r=378.8$; monoclinic; $P2_1/c$; $a=9.927$ (12), $b=18.682$ (25), $c=10.988$ (16) Å; $\beta=115.86$ (10)°; $V=1833.7$ (45) Å^3; $Z=4$; $D_c=1.373$ g·cm^{-3}; $F(000)=752$.

The diffraction intensities were collected from a 2-chlorounguinol (**1**) monohydrate crystal with dimensions of $0.6\times0.5\times0.4$ mm on a Rigaku AFC-5 FOS four-circle diffractometer using CuK_α radiation monochromated by means of a graphite plate. A total of 2867 reflections were measured within a 2θ range of 130° as above the 3σ (F)

TABLE III. Final Atomic Parameters ($\times 10^4$) and Equivalent Thermal Parameters, with Estimated Standard Deviations in Parentheses

	x	y	z	B_{eq} (Å^2)
Cl	3456 (2)	2182 (2)	3248 (2)	3.47
O(3)	5038 (4)	1256 (4)	5512 (5)	3.37
O(5)	7196 (4)	3093 (4)	8811 (4)	2.35
O(8)	12323 (4)	4461 (4)	9269 (5)	2.95
O(10)	7069 (4)	4465 (4)	7812 (5)	2.87
O(11)	4692 (4)	4558 (4)	6495 (5)	3.56
O(w)	6780 (5)	379 (4)	7328 (5)	3.68
C(1)	4798 (6)	3202 (6)	5142 (6)	2.40
C(2)	4611 (6)	2481 (6)	4875 (6)	2.52
C(3)	5296 (6)	1954 (6)	5862 (6)	2.43
C(4)	6213 (6)	2165 (6)	7192 (6)	2.33
C(4a)	6377 (5)	2882 (6)	7468 (6)	2.15
C(5a)	8528 (6)	3439 (5)	8993 (6)	2.00
C(6)	9895 (6)	3093 (5)	9701 (5)	1.97
C(7)	11176 (6)	3453 (6)	9806 (6)	2.19
C(8)	11072 (6)	4115 (6)	9206 (6)	2.19
C(9)	9700 (6)	4469 (5)	8503 (6)	2.19
C(9a)	8450 (6)	4100 (5)	8424 (6)	2.21
C(11)	5762 (6)	4164 (6)	6914 (6)	2.44
C(11a)	5706 (6)	3413 (5)	6498 (6)	2.12
C(12)	4073 (7)	3751 (7)	4009 (7)	3.92
C(13)	9605 (7)	5198 (6)	7887 (7)	3.07
C(1′)	9970 (6)	2368 (6)	7887 (5)	2.32
C(2′)	10679 (7)	1845 (6)	9980 (6)	2.97
C(3′)	10842 (9)	1083 (7)	10508 (8)	4.16
C(4′)	9244 (8)	2280 (8)	11251 (7)	3.40

level. These were used in the solution and refinement of the structure.

Determination of the Structure: The structure was solved by the direct method using MULTAN 84[7] and refined by the block-matrix least-squares method. In the final refinement, anisotropic thermal parameters were used for non-hydrogen atoms. The final R factor without hydrogen atoms was 0.076. The final atomic parameters are shown in Table III, and bond lengths and angles in Fig. 1.[8]

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References and Notes

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8) Lists of F_O and F_C values, anisotropic thermal parameters, and bond lengths and angles are available from one of the authors (K. K.) upon request.