verwhelming evidence for very similar conformations.

(d) Protons $H_{7\alpha}$, H_8, H_9, and $H_{12\alpha}$ in **1** all fall within 0.1 ppm and clearly occupy positions symmetrically disposed with respect to a pseudosymmetry axis running from C_3 to C_{16}. In **2**, $H_{12\alpha}$ is shifted 0.5 ppm downfield by the adjacent hydroxyl group, but the other three protons are almost unshifted from the positions in **1**.

(e) $J_{12\alpha,18}$ of ca. 1 Hz are well-known in 11-keto steroids, and some $J_{1\alpha,19}$ have also been reported,[17] but our results suggest that these may in fact be general phenomena. The lack of similar couplings from $CH_3(18)$ to H_{14} is remarkable.

We do not offer detailed speculation as to the sources of these effects, except to note the substantial deviations from ideal geometry found in the crystal of **1**.[18]

Experimental Section

11β-Hydroxyprogesterone (**2**) was obtained from Sigma Chemical Co. and used without further purification. Solutions (0.1 M) were made up in $CDCl_3$ solution and, for reasons discussed previously,[2] were not degassed.

Details of most spectroscopic procedures were given in ref 2. A brief summary is presented here. All one-dimensional spectroscopy was carried out at 400 MHz, but two-dimensional spectra were obtained at both 270 and 400 MHz. One-dimensional difference spectra were obtained by irradiation at the first frequency for four transients, storage of the FID, repetition of the sequence for each of the remaining frequencies, and then repetition of the whole cycle, under computer control, up to 300 time for NOE experiments and up to 50 times for decoupling experiments. Detailed acquisition microprograms are given in the supplementary material.

Transient NOE's were generated either by a selective pulse from the decoupler or by DANTE.

After submission of the preliminary communications describing some of this work,[3] the original two-dimensional data sets were reprocessed with sine-bell resolution enhancement,[2] and a 270-MHz, two-dimensional, J spectrum was obtained from 0.4 mL of a $CDCl_3$ solution containing 2 drops of benzene-d_6. As a result, virtually all the missing[3] coupling constants were determined, and some chemical shifts have been refined by up to 0.02 ppm.

The total spectrometer time used in this study was 80–100 h, of which 25 h was needed for the transient NOE experiment shown in Figures 5 and 6. The two-dimensional acquisition, processing, and plotting required less than 10 h.

Acknowledgment. We are indebted to S. Sukumar and Dr. B. K. Hunter for many valuable and stimulating discussions on the two-dimensional aspects of this work. This work was carried out by J.K.M.S. during the tenure of an MRC (UK) Travelling Fellowship and was supported by an operating grant to L.D.H. from NSERC of Canada.

Registry No. **2**, 600-57-7.

Supplementary Material Available: Appendix containing detailed aquisition microprograms (3 pages). Ordering information is given on any current masthead page.

(17) Reference 9, pp 115–121.
(18) Duax, W. L.; Norton, D. A.; Pokrywiecki, S.; Eger, C. *Steroids* 1971, *18*, 525.

Antitumor Agents. 43. Conversion of Bruceoside-A into Bruceantin[1]

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Bruceoside-A (**1**) has been converted by two methods to bruceantin (**5**), a potent antileukemic agent. The first method involved the hydrolysis of **1** with potassium hydroxide followed by *p*-toluenesulfonic acid in methanol to afford 49% bruceolide (**2**). Esterification of **2** with 3,4-dimethyl-2-pentenoyl chloride (**9**) yielded the corresponding 3,15-diester (**3**) (47%) and the 3-monoester (**4**) (31%). Compound **4** was reconverted to **3** (77%) by further esterification. Selective hydrolysis of **3** with *p*-toluenesulfonic acid afforded **5** in 40% yield. The second method included the hydrolysis of **1** with potassium hydroxide to yield 57% 15-desenecioyl bruceoside-A (**6**). Boron trifluoride etherate hydrolysis of the 3,4-dimethyl-2-pentenoyl ester (**7**), prepared by esterification of **6** with the corresponding acid chloride, gave **5** in 58% yield.

Bruceantin (**5**), a potent antileukemic quassinoid isolated from the Ethiopian *Brucea antidysenterica*,[2] is currently under clinical trial as an anticancer agent by the National Cancer Institute.[3] It is important to establish an alternate source of **5** to ensure a continuing supply for clinical trials.[4] In connection with our recent isolation of novel antileukemic quassinoid glycosides, bruceoside-A (**1**) and -B from the Chinese *Brucea javanica* in good yield,[5,6] we report the first two methods (methods A and B) leading to the conversion of **1** to **5** in an overall yield of 14 or 33% (methods A and B, respectively). These two methods may be useful in planning the total synthesis of bruceantin and related active analogues. Attempted total synthesis of **5** is currently carried out by many laboratories.[7-11]

(1) For part 42, see: Hall, I. H.; Lee, K. H.; Okano, M.; Sims, D.; Ibuka, T.; Liou, Y. F.; Imakura, Y. submitted for publication in *J. Pharm. Sci.*
(2) Kupchan, S. M.; Britton, R. W.; Lacadie, J. A.; Ziegler, M. F.; Sigel, C. W. *J. Org. Chem.* **1975**, *40*, 648.
(3) "DCT Bulletin"; National Cancer Institute, 1979; Vol. 3.
(4) Private communication from Dr. Matthew Suffness of the Division of Cancer Treatment, National Cancer Institute, Aug 1977.
(5) Lee, K. H.; Imakura, Y.; Huang, H. C. *J. Chem. Soc., Chem. Commun.* **1977**, 69.
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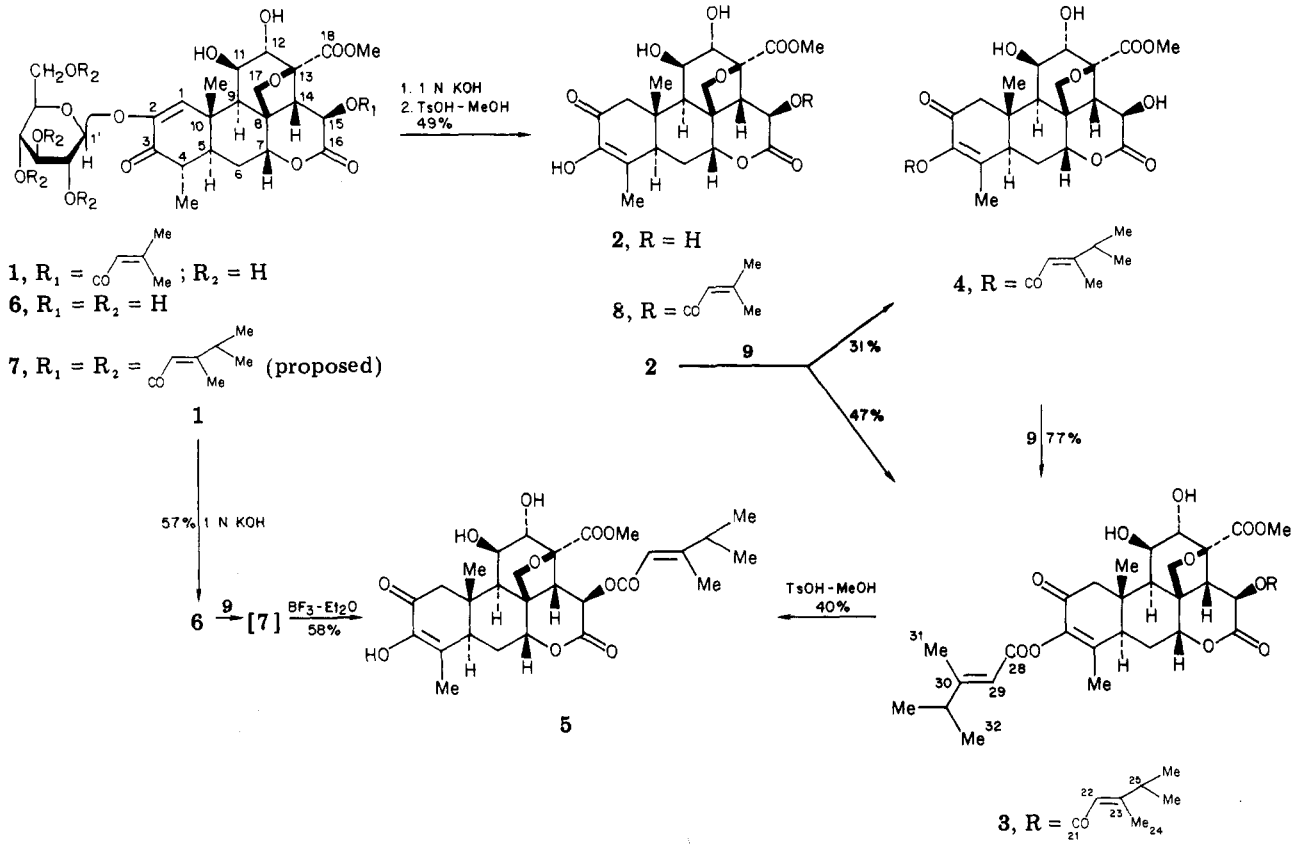
(11) Stojanac, N.; Sood, A.; Stojanac, Z.; Valenta, A. *Can. J. Chem.* **1975**, *53*, 619; *Ibid.* **1979**, *57*, 3346.

0022-3263/81/1946-1138$01.25/0

Scheme I



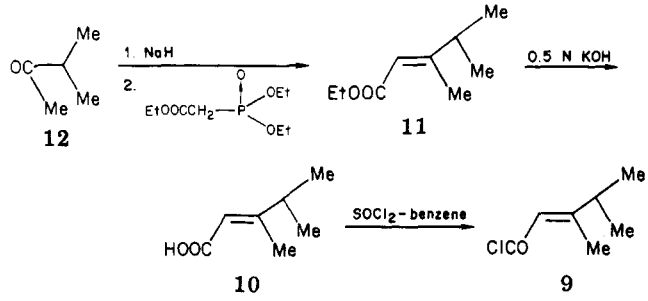
Results and Discussion

We initially planned to use brusatol (8) as the starting material for this study as 8 is readily available from an acid hydrolysis of bruceoside-A (1)[6] (Scheme I). Furthermore, the structure and stereochemistry of 8 is exactly identical with those of bruceantin (5) except for the slight difference in the C-15 ester side chain. Thus, a procedure leading to the simple replacement of the C-15 senecioate moiety in 8 with the corresponding 3,4-dimethyl-2-pentenoate as found in 5 should yield 5. Conversion of 8 into 5 by use of such a procedure involving an initial protection of hydroxyl groups at C-3 and C-12[12] followed by alkaline hydrolysis of the C-15 senecioate, reesterification with 3,4-dimethyl-2-pentenoyl chloride, and removal of the protecting groups to yield 5 was not promising.

In view of the reactivity of the four hydroxyl groups at C-3, C-11, C-12, and C-15 in which C-3 and C-15 are more susceptible toward esterification, the following two methods leading to the facile conversion of either 1 or bruceolide (2) to 5 have been developed, based upon their selective esterification and hydrolysis.

Method A. Bruceolide (2) was obtained from bruceoside-A (1) in 49% yield by a two-step reaction: hydrolysis of the side chain by treatment of 1 with 1 N KOH–methanol solution and hydrolysis of the glucoside by refluxing the hydrolized product with *p*-toluenesulfonic acid in methanol.

3,15-Diester (3) and 3-monoester (4) were obtained in 47% and 31% yield, respectively, by esterification of 2 with 3,4-dimethyl-2-pentenoyl chloride (9) in chloroform–pyridine (1:1). Compound 4 was converted to 3 in 77% yield by further esterification with 9 (50 °C, 23 h). Compound 9 was prepared by reaction of thionyl chloride with 3,4-dimethyl-2-pentenoic acid (10), which was obtained by hydrolysis of ethyl 3,4-dimethyl-2-pentenoate (11). Compound 11 was acquired by Wittig reaction of 3-methyl-2-butanone (12), triethyl phosphonoacetate, and sodium hydride in diglyme (total yield 69%).



Compound 3 was refluxed with *p*-toluenesulfonic acid in methanol to give bruceantin (5) in 40% yield after purification by high-performance liquid chromatography. In addition to 5, unreacted triester (3, 14%) and bruceolide (2, 13%) were isolated by preparative TLC. Alternate hydrolysis of 3 with 3 N sulfuric acid in methanol gave 5 in 15% yield.

Compound 2 was identified by a high-resolution mass spectrum which showed m/z 438.1524 (M^+, calcd for $C_{21}H_{26}O_{10}$ 438.1524). The identity of 5 with an authentic sample of bruceantin was established by mixture melting point determination, TLC, IR, ^1H NMR, and mass (Table I) spectroscopic comparison as well as specific rotation comparison.

Compounds 3 and 4 were characterized by comparing their ^1H NMR and mass spectra (Table I) with those of bruceantin (5). For example, compounds 3 and 4 showed molecular ion peaks at m/z 658 ($C_{35}H_{46}O_{12}$) and 548 ($C_{28}H_{36}O_{11}$), respectively, although their peak matches could not be carried out due to the low intensity. The fact

(12) The C-11 hydroxyl group is highly sterically hindered and need not be protected.

Table I. Relevant ^{1}H NMR[a] and Mass[b] Spectral Data of the Products

signal	3	4	5
H-11	4.24 (m)	4.20 (m)	4.26 (m)
H-12	4.20 (m)	4.20 (m)	4.22 (m)
H-15	6.21 (d, 13[c])	5.29 (d, 13[c])	6.26 (d, 13[c])
H-22	5.64 (m)		5.65 (m)
Me-23	2.15 (br s)		2.16 (br s)
Me-25	1.06 (d, 6.5[c])		1.07 (d, 6.5[c])
H-29	5.90 (m)	5.88 (m)	
Me-30	2.15 (br s)	2.12 (br s)	
Me-32	1.09 (d, 6.5[c])	1.09 (d, 6.5[c])	
m/z 658	0.6% (M^+)		
m/z 548	0.9%	0.5% (M^+)	3.7% (M^+)[d]
m/z 111	100%	100%	100%

[a] δ values (parts per million) relative to Me_4Si in $CDCl_3$ solution. [b] *m/z* values and their relative intensity in EI mass spectra. [c] Coupling constant in hertz. [d] Observed *m/z* 548.2250 (calcd for $C_{28}H_{36}O_{11}$ 548.2256) and 111.0807 (calcd for C_7H_{11} 111.0809).

that the chemical shifts of H-11 and H-12 in **3**, **4**, and **5** coincided with each other indicated the presence of free hydroxyl groups at the **11** and **12** positions. The assignment of a free hydroxyl group at C-15 in **4** was based upon the observation of a higher field shift of H-15 at δ 5.29 in **4**, compared to those of **3** and **5** which appeared at nearly the same chemical shifts at δ 6.21 and 6.26, respectively. Compound **3** had all signals of H-22, Me-23, and Me-25, which were found in **5**, and of H-29, Me-30 and Me-32, which were found in **4**. These evidences led to the conclusion that **3** was a 3,15-diester and **4** was a 3-monoester of bruceolide (**2**).

Method B. 15-Desenecioyl bruceoside-A (**6**) was obtained from bruceoside-A (**1**) in 57% yield by a two-step reaction: hydrolysis of the side chain by treatment of **1** with 1 N KOH–methanol solution at room temperature for 6 h and methylation of the free carboxyl group of C-13 of **6**, which might have been obtained by partial hydrolysis in the first reaction step, with diazomethane in methanol solution at 0 °C for 2 h after the reaction mixture was neutralized by cation-exchange resin (Dowex 50 W-X2).

Esterification of **6** with 3,4-dimethyl-2-pentenoyl chloride in chloroform–pyridine (1:1 v/v) at room temperature for 30 h followed by a subsequent BF_3 etherate hydrolysis in dichloromethane at room temperature for 4 days of the resulting ester (**7**) afforded bruceantin (**5**) in 58% yield.

Compound **6** was obtained as an amorphous substance which decomposed at ca. 200 °C and did not give a sharp melting point. The presence of a glucose moiety in **6** was revealed by IR bands at 3400, 1065, and 1040 cm^{-1}, an anomeric carbon at δ 102.0 (C-1′) in addition to signals at δ 195.1 (C-3, C=O), 173.6 and 172.7 (C-16 and C-18 C=O), and 130.1 (C-1, C=C) in the ^{13}C NMR spectrum, and by the characteristic ions of the trimethylsilyl ether of the sugar moiety at *m/z* 271.1184 (calcd for $C_{12}H_{23}O_3Si_2$ 271.1184) and 361 ($C_{15}H_{33}O_4Si_3$) in the mass spectrum of a trimethylsilyl ether of **6**. Acid hydrolysis of **6** with 3 N sulfuric acid–methanol (1:1 v/v) yielded D-glucose, identified by gas–liquid chromatography as its trimethylsilyl derivative, and an aglycon which showed a molecular ion peak at *m/z* 438.1524 (calcd for $C_{21}H_{26}O_{10}$ 438.1524) and was identified by a mixture melting point determination and comparable IR and ^{1}H NMR spectra with those of the authentic bruceolide (**2**). Compound **5** [*m/z* 548.2250 (M^+, calcd for $C_{28}H_{36}O_{11}$ 548.2256)] showed $[\alpha]_D$ −32° (*c* 0.5, pyridine) and was purified by preparative thin-layer chromatography [silica gel, chloroform–acetone (3:1)]. The identity of **5** with an authentic sample of bruceantin was established by mixture melting point, TLC, specific rotation, IR, and ^{1}H NMR comparison.

Experimental Section

General Procedures. Melting points were determined on a Thomas-Hoover melting-point apparatus and were uncorrected. Specific rotations were obtained on a Rudolph Autopol III automatic polarimeter (*l* = 0.5 dm). Infrared (IR) spectra were recorded with a Perkin-Elmer 257 grating IR spectrometer. Proton nuclear magnetic resonance (^{1}H NMR) spectra were determined with a Varian XL-100 NMR spectrometer (Me_4Si as an internal standard). ^{13}C NMR spectra were recorded on a Varian XL100 spectrometer functioning at 25.20 MHz. All NMR spectra were obtained with the use of the Fourier transform technique. Mass spectra were determined on an AEI MS-902 instrument at 70 eV, using a direct inlet system. Gas–liquid chromatography (GLC) was performed on a Varian Model 3700 gas chromatograph. Silica gel (Mallinckrodt CC7, Special) was used for column chromatography, and precoated silica gel (Merck silica gel 60F-254, 2 mm) was used for thin-layer chromatography (TLC). Detection of components was made by use of a UV lamp. High-performance liquid chromatography (LC) was performed on a Waters Associates analytical instrument.

Bruceolide (2) from Bruceoside-A (1). Bruceoside-A (**1**, 4.1 g, 6.01 mmol)[5] was dissolved in 1 N KOH–methanol solution (100 mL) under cooling at 0 °C and stirred at the same temperature for 15 min. The reaction mixture was acidified by adding 12 N H_2SO_4 under cooling and the salt formed by the procedure was removed by filtration. The mother liquor was concentrated under reduced pressure and the residue was dissolved again by addition of methanol (25 mL). Thereafter, *p*-toluenesulfonic acid (1 g, 5.81 mmol) was added to the solution and the mixture was heated at reflux for 19 h. The reaction mixture was subjected to preparative TLC [chloroform–acetone (1:1 v/v)] to give bruceolide (**2**; 1.33 g, 49% yield, white crystals): mp 308–309 °C (lit.[13] mp 300–302 °C); $[\alpha]^{25}_D$ −78.3° (*c* 0.45, pyridine) (lit.[1] $[\alpha]^{25}_D$ −92.5° (*c* 0.18, pyridine). The spectral (IR, NMR, and mass) data of **2** were in accordance with those reported in the literature[1] for bruceolide.

Ethyl 3,4-Dimethyl-2-pentenoate (11). To a suspension of sodium hydride (2.6 g) in diglyme (10 mL) was added dropwise triethyl phosphonoacetate (24 g, 107 mmol) under nitrogen in an ice bath (ca. 0 °C). The mixture was stirred for 30 min and then 3-methyl-2-butanone (**12**; 4.3 g, 50 mmol) was added dropwise. After 3 h the mixture was cooled, diluted cautiously with large excess amount of water, and extracted with ether. The ethereal extract was dried over anhydrous magnesium sulfate, filtered, and evaporated in vacuo to yield **11** (7.1 g, 91%): NMR (JEOL C 60 HL, $CDCl_3$) δ 1.08 (6 H, d, *J* = 7 Hz, $(CH_3)_2CH$), 1.30 (3 H, t, *J* = 7 Hz, CH_3CH_2O), 2.15 (3 H, d, *J* = 1.5 Hz, CH_3C=), 4.15 (2 H, q, *J* = 7 Hz, CH_3CH_2O), 5.68 (1 H, br s, CH=C); IR (neat) 1705 (C=O), 1640 (C=C), 1385 and 1365 [$(CH_3)_2C$] cm^{-1}; GLC (2% OV-17, Chrom W 80/100, 2 mm × 200 cm, 60 °C, N_2 30 cm^3/m) retention time 9 min.

3,4-Dimethyl-2-pentenoate (10) and 3,4-Dimethyl-2-pentenoyl Chloride (9). A mixture of 0.5 N KOH aqueous solution (92 mL) and **11** (5.5 g) was stirred at 80 °C for 17 h until the oily layer disappeared. The reaction mixture was cooled, acidified with 0.5 N H_2SO_4 (ca. 100 mL), and extracted with ether. The ethereal layer was washed with water, dried ($MgSO_4$), filtered, and evaporated under reduced pressure to afford a yellow liquid (**10**, 4.1 g, 91%). This liquid (**11**, 4.1 g) was then heated to a gentle boil with thionyl chloride (50 g) in benzene (50 mL) until the generation of HCl gas ceased. Removal of the excess thionyl chloride and benzene by evaporation in vacuo followed by a distillation of the reaction product gave pure **9**, bp 64 °C (9.5 mm).

Esterification of 2. A solution of **9** (329 mg, 2.23 mmol) in dry chloroform (4 mL) was added dropwise to a solution of **2** (207 mg, 0.47 mmol) in dry pyridine (4 mL) at 0 °C. The mixture was stirred at 56 °C for 24 h. After cooling, the reaction mixture was acidified with dilute H_2SO_4 and the product was extracted with chloroform. The chloroform layer was dried over anhydrous magnesium sulfate, filtered, and evaporated under reduced

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pressure to give a brown viscous oil. This oil was purified by preparative TLC [chloroform-acetone (10:1 v/v)] to yield 3,15-bis(3,4-dimethyl-2-pentenoyl)bruceolide (**3**, $[\alpha]^{25}_D$ -25° (c 0.45, pyridine), yield 47%] and 3-(3,4-dimethyl-2-pentenoyl)bruceolide (**4**, white crystals, 79.91 mg, yield 31%). Compound **4** could be converted to **3** in 77% yield by further esterification using **9** in an extract procedure described above. The relevant NMR and mass spectral data of **3** and **4** have been described in Table I.

Acid Hydrolysis of 3 to Bruceantin (5). A solution of compound **3** (78.3 mg, 0.119 mmol) in methanol (16 mL) was added to *p*-toluenesulfonic acid (240 mg, 1.26 mmol). The mixture was refluxed and examined by TLC (chloroform-acetone, 1:1). After 92 h, it was purified by preparative TLC (chloroform-acetone, 1:1) to yield **5** (26.9 mg, 41.3%) as white crystals. Further purification of these white crystals by high-performance LC [chloroform-ethyl acetate (1:1), Whatman partisil M 9 10/50] gave 98% pure **5**: mp 220-223 °C (lit.[1] mp 225-226 °C); $[\alpha]^{25}_D$ -31.6° (c 0.5, pyridine) [lit.[1] $[\alpha]^{25}_D$ -43° (c 0.31, pyridine)]. The identity of **5** was confirmed by a direct comparison (mixture melting point, TLC, IR, NMR, and mass spectra) with an authentic sample of bruceantin. In addition to **5**, unreacted triester (10.9 mg, 14%) and bruceolide (**2**, 6.5 mg, 13%) were also isolated from this reaction product by preparative TLC (chloroform-acetone, 1:1).

An alternate hydrolysis of **3** to **5** resulted in only 15% yield. This procedure was carried out by use of a solution of **3** (59.2 mg, 0.09 mmol) in 3 N H_2SO_4-MeOH (1:2, 6 mL) which was heated at reflux for 46 h. The reaction product was purified by preparative TLC (chloroform-acetone, 1:1) to afford pure **5** (7.2 mg) as white crystals.

15-Desenecioyl Bruceoside-A (6). A mixture of **1** (692.3 mg, 1.16 mmol) and 1 N KOH-MeOH (21 mL) was stirred at room temperature for 6 h. The mixture was neutralized with cation-exchange resin (Dowex 50 W-X2) and filtered. The filtrate was methylated with diazomethane[14] in the usual manner. The methylated product was evaporated in vacuo and purified by preparative TLC (chloroform-methanol-water, 50:14:3) to yield **6** (184.2 mg, 57% yield) as an amorphous substance which decomposed at ca. 200 °C. The relevant IR, ^{13}C NMR, and mass spectral data of **6** have been described in the text.

(14) Further methylation of the C-13 $COOCH_3$ was needed as it had been partially hydrolyzed.

Acid Hydrolysis of 6. A solution of **6** (306 mg) in 3 N H_2SO_4-MeOH (1:1, 40 mL) was refluxed for 7 h and then extracted with chloroform. The chloroform extract was dried ($MgSO_4$), filtered, and evaporated in vacuo to give a product which was subjected to preparative TLC (chloroform-acetone, 1:1) to yield pure **2** (43.5 mg).

The aqueous layer was neutralized with cation-exchange resin (Amberlite IR-45), filtered, dried, and evaporated to give a residue which was identified as the trimethylsilyl derivative of D-glucose by GLC [3% OV-17 on Chromosorb (80-100 mesh), 3 mm × 2 m, 170 °C, N_2, 15 mL/min, injection temperature 180 °C, detector temperature 180 °C].

Esterification of 6 and Hydrolysis of 3,5-Dimethyl-2-pentenoyl Ester of 6. A solution of **6** (89.9 mg, 0.15 mmol) in dry pyridine (2 mL) was added dropwise to a solution of **9** (330 mg, 2.25 mmol) in dry chloroform (2 mL). The mixture was stirred at room temperature for 20 h until the TLC (chloroform-methanol-water, 50:14:3) showed the disappearance of **6** and then water was added to decompose the unreacted acid chloride. The reaction product (**7**, proposed[15]), without further purification and isolation, was dissolved in dichloromethane (10 mL) and then 6 drops of BF_3 etherate was added. The reaction mixture was stirred at room temperature and examined by TLC (chloroform-acetone, 1:1). After 4 days, the product was subjected to preparative TLC (chloroform-acetone, 10:1) to yield pure **5** (47.7 mg, 58% yield).

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Registry No. **1**, 63306-30-9; **2**, 25514-28-7; **3**, 76215-40-2; **4**, 76215-41-3; **5**, 41451-75-6; **6**, 76215-42-4; **7**, 76232-15-0; (*E*)-**9**, 76215-43-5; (*E*)-**10**, 38972-59-7; (*E*)-**11**, 21016-44-4; **12**, 563-80-4.

(15) Structure **7** was proposed for this reaction product based upon the fact that in an analogous study of the esterification of brusatol (**8**), bruceantin (**5**), and bruceoside-A (**1**), both hydroxyl groups at C-11 and C-12 of these compounds were resistant to this kind of esterification.

Allergenic α-Methylene-γ-lactones. General Method for the Preparation of β-Acetoxy- and β-Hydroxy-α-methylene-γ-butyrolactones from Sulfoxides. Application to the Synthesis of a Tuliposide B Derivative

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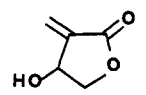
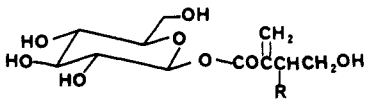
A general synthesis of β-hydroxy-α-methylene-γ-butyrolactones, which is based on the sulfoxide-sulfenate rearrangement, is presented. Several β-acetoxy-α-methylene-γ-butyrolactones have been prepared and transformed into the β-hydroxy derivatives through base hydrolysis. This synthesis has been applied to the first preparation of (tetraacetoxybenzyl)tuliposide B (**22**).

Many natural compounds contain the β-hydroxy-α-methylene-γ-butyrolactone unit **1**.[1] Several of these



1



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substances show bactericidal or fungicidal activity. Among them are β-hydroxy-α-methylene-γ-butyrolactone precursors such as tuliposide B (**2**, R = OH), which is found in tulip bulbs,[2] along with tuliposide A (**2**, R = H), re-

(1) C. J. Cavallito and J. H. Haskell, *J. Am. Chem. Soc.*, **68**, 2332-2334 (1946); M. Niwa, M. Iguchi, and S. Yamamura, *Chem. Lett.*, 655-658 (1975); K. Takeda, K. Sakwawi, H. Ishū, *Tetrahedron*, **28**, 3757-3766 (1972); J. C. Martinez, V. M. Yoshida, and O. R. Gottlieb, *Tetrahedron Lett.*, 1021-1024 (1979).

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