Anal. Calcd for $C_{20}H_{20}O_2$: C, 82.15; H, 6.89; sapon equiv, 292. Found: C, 82.04; H, 7.05; sapon equiv,[18] 292.

Basic hydrolysis of acetate **4b** gave recovery of alcohol **2a**.

Tosylation of 2a.—To a solution of 1.48 g (5.9 mmol) of alcohol **2a** in 10 ml of pyridine at 0° was added dropwise (with swirling) a solution of 2.3 g (12 mmol) of *p*-toluenesulfonyl chloride in 10 ml of pyridine. The mixture was kept at −20° for 24 hr, then poured onto ice and processed as in the preceding acetylation. The residue, 2.24 g (94%) of tosylate **4c**, mp 119–120.5°, formed prisms from benzene–hexane: mp 122–123°; pmr δ 1.1–1.7 (m, 3, H-1a plus 2 H-1), 2.43 (s, 3, tosylate CH_3), *ca.* 2.70 (broadened s, 3, H-2 plus 2 H-3), 3.8–4.2 (irregular t, 2, CH_2OTs), 6.6–7.9 (m, 13, aromatic protons); ir (KBr) 1180 and1350 cm^{-1} (sulfonate).

Anal. Calcd for $C_{25}H_{24}O_3S$: C, 74.24; H, 5.96; S, 7.93. Found: C, 74.15; H, 6.02; S, 8.00.

2-Methyl-7b-phenyl-1a,2,3,7b-tetrahydro-1*H*-cyclopropa[*a*]naphthalene (4d).—To a stirred slurry of 0.75 g (20 mmol) of $LiAlH_4$ in 50 ml of tetrahydrofuran at 0° was added dropwise a solution of 1.57 g (3.9 mmol) of tosylate **4c** in 100 ml of the same solvent. The mixture was then stirred at 25° for 30 min, refluxed for 6 hr, treated dropwise with water, brought to pH 1, and extracted with ether. Evaporation of the water-washed, dried extract gave a liquid which formed prisms (0.91 g, 99%) from hexane–ether: mp 70–72° (raised to 72–73° on recrystallization); mass spectrum[15] *m/e* (rel intensity) 234 (M, 100), 219 (M − CH_3, 73), 205 (M − C_2H_5, 33), 192 (M − $CH_3CH{=}CH_2$, 43).

Anal. Calcd for $C_{18}H_{18}$: C, 92.26; H, 7.74. Found: C, 92.49; H, 7.50.

5,6-Dimethoxy-2-hydroxymethyl-7b-(3,4-dimethoxyphenyl)-1a,2,3,7b-tetrahydro-1*H*-cyclopropa[*a*]naphthalene (2c).—In the same manner as used for the synthesis of **2a**, tetramethoxy compound **1b**[3] was reduced to **2c**. The oily product was chromatographed by means of Florisil and (in succession) eluents of benzene and benzene–$CHCl_3$ (1:1, v/v). From the latter eluent was obtained a 36% yield of **2c** (single spot on tlc), converted to light yellow prisms on crystallization from hexane: mp 45–46°; pmr (CCl_4) δ 0.9–1.5 (m, 3, cyclopropane protons), *ca.* 2.6 (broad signal, disappears on shaking with D_2O, OH) which overlaps 2.1–3.0 (complex, 3, H-2 plus 2 H-3), 3.50 (s, 3, OCH_3 at C-6), 3.68, 3.74, 3.77 (3s, other OCH_3 groups) which obscure signals for CH_2OH, 6.30 (s, 1, H-7), 6.52 (s, 1, H-4), 6.57–6.9 (broad s plus m, 3, H-2′, H-5′, and H-6′); ir ($CHCl_3$) 3500 cm^{-1} (broad, OH); mass spectrum[15] *m/e* 370 (M, 69%), 339 (100), 151 (36), 57 (32).

Anal. Calcd for $C_{22}H_{26}O_5$: C, 71.33; H, 7.08. Found: C, 71.61; H, 7.16.

Infrared Spectra.—Spectral examination of samples in the near infrared region of the spectrum was made by means of a Cary model 14 spectrophotometer, with a concentration of *ca.* 50 mg of substrate per milliliter of solvent, CCl_4. Compounds **2a**, **2c**, **4a**, and **4b** (but not deuterated compound **2b**, nor the impure product from catalytic hydrogenolysis of **4d**) showed prominent absorption shoulders or peaks at 1.635–1.645 μ. Extinction coefficients for **2a**, **4a**, and **4b** were 0.35, 0.19, and 0.32, respectively.

The regular infrared spectra (obtained in CS_2 as solvent, by means of a Beckman IR-7 spectrophotometer) of **2a**, **4a**, and **4b** also showed a medium band at 1016–1021 cm^{-1} (ascribed to cyclopropane ring deformation)[19,20] and a weak band at 820–844 cm^{-1} (ascribed to cyclopropane ring CH_2 rocking).[20,21] The latter band was clearly resolved in all compounds, though the former band was sharp only in hydrocarbon **4a**. For **2a** it occurred only as a shoulder on the strong C–O stretching band at 1035 cm^{-1}, but some better resolution was found in the spectrum of **4b**.

Registry No.—**2a**, 34599-28-5; **2b**, 34566-27-3; **2c**, 34566-28-4; **4a**, 34566-29-5; **4b**, 34566-30-8; **4c**, 34566-31-9; **4d**, 34566-32-0.

(18) Analysis by Geller Laboratories, Charleston, W. Va.

(19) J. M. Derfer, E. E. Pickett, and C. E. Boord, *J. Amer. Chem. Soc.*, **71**, 2482 (1949); C. N. R. Rao, "Chemical Applications of Infrared Spectroscopy," Academic Press, New York, N. Y., 1963, p 146, and references cited therein.

(20) S. A. Liebman and B. J. Gudzinowicz, *Anal. Chem.*, **33**, 931 (1961); M. Hanack, H. Eggensperger, and S. Kang, *Chem. Ber.*, **96**, 2532 (1963).

(21) H. A. Szymanski, "Interpreted Infrared Spectra," Vol. 1, Plenum Press, New York, N. Y., 1964, pp 143–162.

Acknowledgment.—The authors are grateful to Professor H. M. Walborsky of Florida State University for providing us with a pmr comparison spectrum of 1-methyl-2,2-diphenylcyclopropane and to Dr. T. M. McGuire (formerly of this laboratory) for obtaining definitive pmr spectra on some of our products.

Structure of a New Fungal Lactone, LL-P880α, from an Unidentified *Penicillium* sp.

G. A. ELLESTAD,* W. J. MCGAHREN, AND M. P. KUNSTMANN

Lederle Laboratories, A Division of American Cyanamid Company, Pearl River, New York 10965

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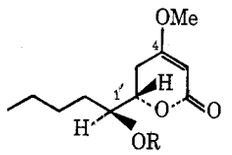
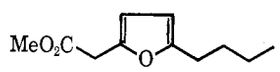
In a continuing program seeking useful pharmacologically active compounds from microorganisms, we had occasion to examine the fermentations of culture P880, an unidentified *Penicillium* species. This report describes the structure, stereochemistry, and some rearrangements of the metabolite LL-P880α.[1] This metabolite, $C_{11}H_{18}O_4$, is characterized by a uv maximum at 235 nm (ε 12,000) and strong ir absorption at 1710 and 1625 cm^{-1} which suggests the presence of the 4-alkoxy-5,6-dihydro-α-pyrone moiety.[2] The nmr spectrum supports this conclusion with a methoxy signal at δ 3.80, the C_3 vinyl proton signal at δ 5.16 ($J_{3,5a}$ = 2 Hz), a 1 H multiplet at δ 4.33 due to the proton of C_6, an eight-line pattern at δ 2.67 (J_{gem} = 18, $J_{3,5a}$ = 2, $J_{5a,6}$ = 11 Hz), and a four-line system at δ 2.23 (J_{gem} = 18, $J_{5e,6}$ = 4 Hz) due to the geminal C_5 protons. In addition, a primary C–Me signal at δ 0.92 as a characteristic 3 H triplet and a 1 H multiplet due to a second



I, R = H
II, R = Ac



III

proton on a carbon bearing an oxygen atom at δ 3.70 are observed. The hydroxy nature of this remaining oxygen is indicated by the formation of acetate II, $C_{13}H_{20}O_5$.

The major fragmentation in the mass spectrum of I results from the loss of the five-carbon side chain, giving the base peak at *m/e* 127. The ion at *m/e* 157 is consistent with cleavage between C_1' and C_2' and expulsion of the *n*-butyl unit. This evidence, in conjunction with the foregoing, unequivocally indicates I as the structure of the metabolite.

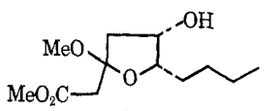
The chemistry of I under acidic or basic conditions is characterized by a marked propensity to rearrange to the furanoid system or derivatives thereof. Thus hydrolysis of I in methanolic hydrochloric acid gave the furan ester III, $C_{11}H_{16}O_3$. Its nmr spectrum shows the two ring-proton signals at δ 6.08 and 5.90 as two dou-

(1) After this work was completed, a note by Y. Kimura, K. Katagiri, and S. Tamura appeared in *Tetrahedron Lett.*, **No. 33**, 3137 (1971), which describes the same compound from *Pestalotia cryptomeriaecola*.

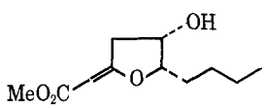
(2) H. B. Henbest and E. R. H. Jones, *J. Chem. Soc.*, 3628 (1950).

blets, J = 3 Hz. A 2 H singlet at δ 3.63 is attributed to the C_2 methylene protons. The methoxy signal resonates at δ 4.71, and the C_6 methylene hydrogens at δ 2.61 as a broad triplet.

Treatment of I with sodium methoxide in dry methanol under reflux with rigorous exclusion of moisture gave IV, $C_{12}H_{22}O_5$, and V, $C_{11}H_{18}O_4$, by ether extraction of the crude concentrate. Acidification during the work-up provided III.



IV



V

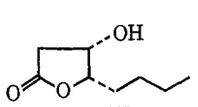
The nmr of IV shows the C_4 geminal hydrogens as a sharp 2 H doublet (J = 4 Hz) at δ 2.29 and the C_2 geminal hydrogens as a 2 H quartet at δ 2.64 and 2.93 (J_{gem} = 16 Hz). The C_5 and C_6 proton signals are buried under the C_3 methoxy signal at δ 3.17 and the ester methoxy signal occurs at δ 3.70.

Although IV is a colorless oil, V is highly crystalline. Two four-line nmr patterns at δ 2.86 and 3.19 ($J_{4,4}$ = 19, $J_{2,4a}$ = 2, $J_{4a,5}$ = 5 Hz) in the spectrum of V are assigned to the pseudoaxial proton signal of C_4. Only one of the signals of the pseudoequatorial C_4 proton is seen at δ 3.45 as a triplet ($J \cong$ 1.5 Hz), since the remaining portion at δ 3.68 is obscured by the methoxy signal. The C_2 olefinic proton is seen at δ 5.37 as a complex multiplet.

An internal Michael addition of the $C_{1'}$ hydroxyl group in I followed by methanolysis of the resulting bicyclic lactone and elimination of the C_4 methoxyl, or other related combinations, will lead to the observed products. A related situation was observed in the chemistry of rubratoxin.[3]

The stereochemistry at C_6 and $C_{1'}$ depicted in I is based on several pieces of evidence, all of which are mutually consistent. The axial nature of the C_6 hydrogen is known from the coupling constant of 11 Hz between the C_{5a} and C_6 hydrogens. A negative Cotton effect ($\Delta\epsilon$ −7.90) at 243 nm in the CD spectrum of I[4] determines the absolute stereochemistry at C_6 as S. Application of the Horeau method[5] to V, in which the lactone ether oxygen is now a secondary alcohol, confirms the CD results. Thus treatment of the latter with (±)-α-phenylbutyric anhydride liberated (−)-α-phenylbutyric acid.

Treatment of I with the racemic anhydride also liberated the (−) acid, suggesting the S configuration at $C_{1'}$. An $[\alpha]_D$ of −71.06° for the lactone VI (obtained by ozonolysis of V) on the basis of the Hudson lactone rule[6] confirms this assignment, as does the $\Delta\epsilon$ of −0.53 at 216 nm in the CD[7] spectrum of VI.



VI

Experimental Section

All melting points were determined on a Fisher-Johns melting point block and are uncorrected. Nmr spectra were recorded with a Varian A-60D in $CDCl_3$; shifts are expressed as δ values (parts per million) from tetramethylsilane as internal standard, and coupling constants are expressed in cycles per second (hertz). Infrared spectra were taken on a Perkin-Elmer Model 137 infracord and ultraviolet spectra on a Cary Model 11. Circular dichroism curves were obtained on a Cary 60 spectropolarimeter with a CD attachment. In nmr descriptions, s = singlet, d = doublet, t = triplet, m = multiplet, dd = double doublet, and q = quartet.

Isolation of I.—The whole mash from a 300-l. fermentation was extracted at pH 6 with a 0.5 volume of ethyl acetate. This was concentrated to an oily residue, which was taken up in methanol. The methanol solution was washed with heptane and then reconcentrated to give 9 g of a dark, semisolid residue. Column chromatography over silica gel (250 g) and elution with methylene chloride gave a crystalline residue which on recrystallization from benzene–hexane gave 3.8 g of I. The analytical sample had mp 84–85°; $[\alpha]_D$ −86.2° (c 0.14, MeOH); λ_{max}^{MeOH} 235 nm (ϵ 12,000); ir (KBr) 1710 and 1625 cm^{-1}; nmr δ 0.92 CMe, t), 2.23 (H_{5e}, dd, $J_{5a,5e}$ = 18, $J_{5e,6}$ = 4 Hz), 2.67 (H_{5a}, dd, $J_{5a,5e}$ = 18, $J_{3,5a}$ = 2, $J_{5a,6}$ = 11 Hz), 3.80 (OMe, s), 4.33 (H_5, m), 5.16 (H_3, d, $J_{3,5a}$ = 2 Hz), 3.70 ($H_{1'}$, m); CD (0.82 mg in 10 cc of MeOH) $\Delta\epsilon_{243}$ −7.90; mass spectrum m/e 214 ($C_{11}H_{18}O_4$).

Anal. Calcd for $C_{11}H_{18}O_4$: C, 61.66; H, 8.47. Found: C, 61.78; H, 8.19.

Acetate II.—A solution containing 100 mg of I in 0.5 ml of acetic anhydride and 0.5 ml of pyridine was allowed to stand at room temperature overnight. The reaction was evaporated to dryness under reduced pressure to give a yellow gum. This was chromatographed on acid-washed silica gel with 25% ether–methylene chloride as the eluent to give 93 mg of a colorless, viscous oil which showed only one spot on silica gel tlc (20% ethyl acetate–benzene); $[\alpha]_D$ −98.5° (c 0.53, MeOH); λ_{max}^{MeOH} 233 nm (ϵ 13,300); ir (smear) 1740, 1710, and 1625 cm^{-1}; nmr δ 1.00 (CMe, t), 2.15 (OAc, s), 2.25 (H_{5e}, dd, $J_{5a,5e}$ = 18, $J_{5e,6}$ = 4 Hz), 2.65 (H_{5a}, dd, $J_{5a,5e}$ = 18, $J_{3,5a}$ = 2, $J_{5a,6}$ = 11 Hz), 3.78 (OMe, s), 4.50 (H_6, m), 5.06 ($H_{1'}$, m), 5.20 (H_3, d, $J_{2,5a}$ = 2 Hz); mass spectrum m/e 256 ($C_{13}H_{20}O_5$).

Conversion of I to III with Methanolic Hydrochloric Acid.—A solution of 20 ml of methanol and five drops of concentrated hydrochloric acid containing 300 mg of I was refluxed overnight. Removal of the solvent and distillation[8] at 100° (80 μ) gave an almost quantitative yield of III: ir (KBr) 1740 cm^{-1}; nmr δ 0.92 (CMe, t), 2.62 (H_7, t), 3.63 (H_2, s), 3.72 (OMe, s), 5.90 and 6.08 (H_4 and H_5, dd, J = 3 Hz); mass spectrum m/e 196 ($C_{11}H_{16}O_3$).

Anal. Calcd for $C_{11}H_{16}O_3$: C, 67.32; H, 8.22. Found: C, 66.90; H, 8.04.

Conversion of I to IV and V.—To a solution of 5 g of I in 100 ml of dry methanol (dried over molecular sieves) was added 2 g of sodium methoxide in 50 ml of dry methanol. The solution was refluxed overnight with the rigorous exclusion of moisture. The methanol was evaporated and the gummy suspension was treated with ether and filtered. The filtrate was evaporated to an oil which was chromatographed over 180 g of silica gel and eluted with 5% ethyl acetate–hexane with fraction volumes of 80–85 ml. Fractions 9–16 gave 2.5 g of a colorless oil which was further purified by partitioning over 220 g of acid-washed Celite using the solvent system heptane–acetonitrile. This provided

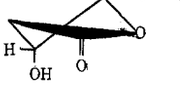
(3) G. Büchi, K. M. Snader, J. D. White, J. Z. Gougouatas, and S. Singh, *J. Amer. Chem. Soc.*, **92**, 6638 (1970).

(4) G. Snatzke, *Angew. Chem.*, **80**, 14 (1968).

(5) A. Horeau and H. B. Kagan, *Tetrahedron*, **20**, 2431 (1964).

(6) C. S. Hudson, *J. Amer. Chem. Soc.*, **32**, 338 (1910); 1525 (1939). The same argument has been used to assign the S configuration to the γ-propylbutyrolactone obtained from oudenone: M. Ohno, M. Okamoto, N. Kawabe, H. Umezawa, T. Takeuchi, H. Iinuma, and S. Takahashi, *J. Amer. Chem. Soc.*, **93**, 1285 (1971).

(7) This is on the basis that VI exists as in the projection; see A. F. Beecham, *Tetrahedron Lett.*, **No. 32**, 3591 (1968); F. I. Carrol, H. Sobti, and R. Meck, *ibid.*, No. 5, 405 (1971), and references cited therein.



(8) Evaporative bulb-to-bulb distillation using a Büchi kugelrohrofen.

1.8 g of pure IV: $[\alpha]D$ −66.3° (*c* 1.09, MeOH); ir (smear) 1740 cm^{-1}; 100-MHz nmr (CCl_4) δ 0.95 (CMe, t), 2.29 (H_4, d, J = 4 Hz), 2.80 (H_2, q, J = 16 Hz), 3.17 (C_3OMe, s), 3.70 (C_1 OMe, s).

Anal. Calcd for $C_{12}H_{22}O_5$: C, 58.51; H, 9.00. Found: C, 58.34; H, 8.80.

Fractions 17–28 from the silica gel column mentioned above gave, on concentration, 350 mg of white solid which was recrystallized from ether–hexane to give the analytical sample of V: mp 105–106°; $[\alpha]D$ −169° (*c* 0.72, MeOH); λ_{max}^{MeOH} 245 nm (ε 22,250); ir (KBr) 3450, 1720, and 1600 cm^{-1}; nmr δ 0.97 (CMe, t), 2.86 and 3.19 (H_{4a}, q, $J_{4,4}$ = 19, $J_{2,4a}$ = 2, $J_{4a,5}$ = 5 Hz), 3.45 (H_{4e}, 1 H, t, $J \cong$ 1.5 Hz), 4.25 (m, H_5 and H_6), 5.37 (H_2, m); mass spectrum *m/e* 214 ($C_{11}H_{18}O_4$).

Anal. Calcd for $C_{11}H_{18}O_4$: C, 61.66; H, 8.47. Found: C, 62.04; H, 8.45.

Conversion of I to III with Sodium Methoxide.—A solution of 1.07 g of I and 270 mg of sodium methoxide in 20 ml of methanol was gently warmed on a steam bath for 30 min. The solution was concentrated and the resultant oil was taken up in ethyl acetate and washed with 4 *N* hydrochloric acid. The ethyl acetate phase was dried and concentrated to an oil which was passed over 80 g of acid-washed silica gel and eluted with 10% ethyl acetate in hexane. This provided 550 mg of a colorless oil from which 150 mg was distilled[8] at 80° (100 μ) to give III. This material was identical in all respects with that obtained above.

Ozonolysis of V.—Ozone was passed through a solution of 400 mg of V in methanol at −70°. The reaction was worked up by the dimethyl sulfide procedure.[9] After removal of the solvent, the residual oil was distilled[8] at 135° (100 μ) to give 100 mg of colorless oil: $[\alpha]D$ −71.1 (*c* 0.73, MeOH); ir (smear) 1770 cm^{-1}; CD (2.44 mg/ml MeOH) $\Delta\epsilon_{216}$ −0.53.

Anal. Calcd for $C_8H_{14}O_3$: C, 60.74; H, 8.92. Found: C, 60.88; H, 8.79.

Application of Horeau's Method to I.—A solution of 59 mg of I and 215 mg of (±)-α-phenylbutyric anhydride in 3 ml of pyridine was allowed to stand over the weekend at ambient temperature. Then 1 ml of water was added with the consequent generation of heat. After 1 hr, 20 ml of water was added and the mixture was extracted three times with ether. The ether extracts were back-extracted twice with 10 ml of 10% sodium carbonate solution. The aqueous alkaline solution was washed with ether and then acidified and extracted once again with ether. This was dried over magnesium sulfate and evaporated to 154 mg of a colorless oil which solidified in the refrigerator. Tlc using benzene–dioxane–acetic acid (50:50:2) showed this material to be α-phenylbutyric acid as did the ir and nmr, $[\alpha]D$ −0.17 ± 0.07° (*c* 2.90, benzene).

Anal. Calcd for $C_{10}H_{12}O_2$: C, 73.14; H, 7.37. Found: C, 73.26; H, 7.20.

Application of Horeau's Method to V.—To 3 ml of pyridine was added 65 mg of V and 218 mg of (±)-α-phenylbutyric anhydride. The solution was allowed to stand over the weekend at room temperature and then worked up as described above to give 145 mg of the acid, $[\alpha]D$ −2.27 ± 0.07° (*c* 2.9, benzene).

Anal. Calcd for $C_{10}H_{12}O_2$: C, 73.14; H, 7.37. Found: C, 73.24; H, 7.46.

Registry No.—I, 34565-32-7; II, 34565-33-8; III, 34565-34-9; IV, 34565-35-0; V, 34565-36-1; VI, 34565-37-2; α-phenylbutyric acid, 938-79-4.

Acknowledgments.—We wish to thank our colleagues for their assistance in particular, Dr. H. Tresner and Miss Jean Hayes for culture isolation and identification, Mr. A. J. Shay for large-scale fermentations, Mr. M. Dann for large-scale work-ups, Mr. L. Brancone for microanalytical data, Mr. W. Fulmor for spectral data and optical rotations, and Miss Pat Mullen of Cyanamid Stamford Laboratories for the CD curves.

(9) J. J. Pappas, W. P. Keaveney, E. Gancher, and M. Berger, *Tetrahedron Lett.*, 4273 (1966).

Photoreduction of Conjugated Cyclopropyl Ketones in Isopropyl Alcohol[1]

WILLIAM G. DAUBEN,* LEONARD SCHUTTE, AND E. JOHN DEVINY

Department of Chemistry, University of California, Berkeley, California 94720

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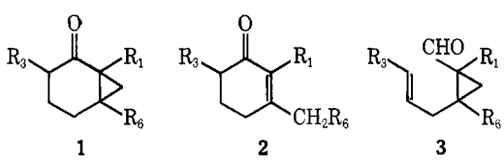
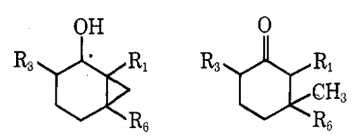
Previous photochemical studies of substituted bicyclo[4.1.0]heptan-2-ones (**1**) under photoisomerization conditions, *i.e.*, *tert*-butyl alcohol used as solvent, have established that the relative efficiencies of three possible reaction pathways from **1** in the triplet state are affected by the pattern of substitution on the ring system.[2] The general photoisomerization of such a conjugated system (**1**, R_3, R_6 = H) to a 3-substituted cyclohex-2-en-1-one (**2**, R_3, R_6 = H) is blocked when R_6 is an alkyl group; in such a case only efficient intersystem crossing from the excited triplet state to the singlet ground state of the starting material occurs. However, with substitution at R_3, the Norrish type I cleavage to an aldehyde **3** becomes the favored primary photoprocess.



1 **2** **3**

On the other hand, irradiation of **1** (R_1, R_3 = H, R_6 = H or CH_3) in isopropyl alcohol, *i.e.*, photoreduction conditions, has been shown to lead to a selective reductive opening of the outside bond of the cyclopropyl ring.[3] In this photoreduction, the intervention of the α-hydroxycyclopropylcarbinyl radical **4** (R_1, R_3 = H, R_6 = H or CH_3) has been established and it is its collapse which leads to a 3-substituted 3-methylcyclohexanone **5** (R_1, R_3 = H). The effect of the pattern of substitution of the ring system on this photoreduction process has now been investigated.



4 **5**

It was found that the disubstituted derivative 1,6-dimethylbicyclo[4.1.0]heptan-2-one (**6**) upon irradiation in isopropyl alcohol was rapidly transformed to isopropyl 5,5-dimethylheptanoate (**8**). When the irradiation was monitored using infrared spectroscopy, it was found that the expected 2,3,3-trimethylcyclohexanone (**7**) was the first photoproduct formed. This latter ketone **7**, prepared from 2,3-dimethylcyclohex-2-en-1-one (**9**) and lithium dimethylcopper, upon ir-

(1) This work was supported by Public Health Service Grant No. 00709, National Institute of Arthritis and Metabolic Diseases, U. S. Public Health Service.

(2) W. G. Dauben, G. W. Shaffer, and E. J. Deviny, *J. Amer. Chem. Soc.*, **92**, 6273 (1970).

(3) W. G. Dauben, L. Schutte, R. E. Wolf, and E. J. Deviny, *J. Org. Chem.*, **34**, 2512 (1969).