mixture, stirred 5 h, and then quenched with $NH_4Cl_{(s)}$ (100 mg, 1.9 mmol). The now homogeneous solution was concentrated by Kugelrohr distillation and purified by flash chromatography (3:1 then 1:1 petroleum ether/EtOAc) affording pure **32** (1.02 g, 62%) as a white solid: mp softens at 176 °C, at 184–185 °C; IR (KBr) 3429, 3178, 3051, 2956, 1744, 1721, 1697, 1689, 1466, 1438, 1373, 1298, 1275, 1230, 1206, 1090, 791, 759 cm^{-1}; ^1H NMR ($CDCl_3$) δ 9.44 (br, 1 H), 7.23 (s, 1 H), 4.64 (d, J = 10.5 Hz, 1 H), 3.83 (t, J = 10 Hz, 1 H), 3.76 (s, 3 H), 3.74 (s, 3 H), 2.97 (d, J = 9.5 Hz, 1 H), 1.97 (s, 3 H), 1.46 (s, 3 H), 0.84 (s, 3 H); ^{13}C NMR ($CDCl_3$) δ 171.02, 170.98, 163.83, 151.27, 136.51, 110.21, 59.82, 52.56, 52.16, 45.10, 44.03, 38.50, 28.94, 17.00, 12.63; NOE ($CDCl_3$) irradiation of β-CH_3 (δ 1.5) gave 4.5% enhancement of H-3′ (δ 2.97) and 4.0% enhancement of H-1′ (δ 4.64), irradiation of α-CH_3 (δ 1.97) gave 1.3% enhancement of H-2′ (δ 3.83), irradiation of H-2′ (δ 3.83) gave 6.8% enhancement of H-6 (δ 7.23), confirming that addition occurs with N-1 rather than N-3; MS (EI) m/z (relative intensity) 324 ($M^{\bullet+}$, 6.8), 293 (11.7), 210 (100), 180 (6.7), 151 (68.1), 59 (21.3); HRMS calcd for $C_{15}H_{20}N_2O_6$ 324.1321 ($M^{\bullet+}$), found 324.1322. Anal. Calcd for $C_{15}H_{20}N_2O_6$: C, 55.55; H, 6.22; N, 8.64. Found: C, 55.59; H, 6.15; N, 8.64.

A mixture of **32** and the isomer **33** (500 mg, 2:1 ratio **32:33**) was also isolated giving an overall yield of 93% for the thymine addition. Pure **32** could be obtained by recrystallization from H_2O or by equilibration. A solution of **32** and **33** (500 mg) in CH_3OH was treated with K_2CO_3, refluxed for 24 h, and concentrated by rotary evaporation. The residue was taken up in H_2O and extracted with CH_2Cl_2, dried ($MgSO_4$), and concentrated by rotary evaporation to afford **32** (251 mg, 50%) as a white solid. Equilibration in H_2O/K_2CO_3, however, gave the isomer **33**. A mixture of **32** and **33** (1.02 g, 3.14 mmol) in H_2O (50 mL) and K_2CO_3 (~300 mg) was refluxed for 36 h. Upon cooling, fine, white crystals precipitated and were filtered off giving pure **33** (382 mg, 37%): ^1H NMR ($CDCl_3$) δ 9.01 (br, 1 H), 6.97 (s, 1 H), 4.98 (d, J = 10 Hz, 1 H), 3.83 (t, J = 10 Hz, 1 H), 3.11 (d, J = 10 Hz, 1 H), 1.93 (s, 3 H), 1.24 (s, 3 H), 1.14 (s, 3 H); ^{13}C NMR ($CDCl_3$) δ 171.62, 171.02, 163.73, 151.12, 137.03, 110.14, 60.68, 52.26, 51.98, 48.14, 42.40, 38.91, 24.36, 22.85, 12.61; NOE ($CDCl_3$) irradiation of β-CH_3 (δ 1.14) gave 3.5% enhancement of H-3′ (δ 3.11) and 1.8% enhancement of H-2′ (δ 3.38), irradiation of α-CH_3 gave 2.8% enhancement of H-1′ (δ 4.98).

9-[4,4-Dimethyl-*trans*-2,*cis*-3-bis(hydroxymethyl)cyclobut-*r*-1-yl]thymine (Dimethyl-C-Oxt-T) (35). To a solution of $LiBEt_3H_{24}$ (0.18 mmol) in THF (5 mL) was added $LiBH_4$ (101 mg, 4.62 mmol) and then diester **32** (300 mg, 0.92 mmol).[25] After stirring 24 h 0.3 mmol more $LiBEt_3H$ was added, and the reaction mixture stirred 24 h longer. The reaction was quenched with acetone and then 1 M H_2SO_4 (2 mL). The pH of the solution was adjusted to 7 with 6 N NaOH, combined with silica gel (1 g) and concentrated in vacuo. The white solid was applied to a silica gel column packed with 30:1 CH_2Cl_2/CH_3OH, and eluted with 30:1 then 15:1 then 8:1 CH_2Cl_2/CH_3OH to yield **35** (163 mg, 61%) as a white solid: mp 224–225 °C; IR (KBr) 3356, 3093, 3033, 2958, 2926, 2869, 1688, 1654, 1474, 1390, 1296, 1286, 1065, 1022 cm^{-1}; ^1H NMR (CD_3OD) δ 7.55 (s, 1 H), 4.20 (d, J = 10 Hz, 1 H), 3.55–3.69 (m, 4 H), 2.58–2.73 (m, 1 H), 1.88 (s, 3 H), 1.79 (dt, J_1 = 9 Hz, J_2 = 7.5 Hz, 1 H), 1.27 (s, 3 H), 0.86 (s, 3 H); ^{13}C NMR (CD_3OD) δ 153.45, 140.22, 109.84, 63.97, 62.24, 61.84, 44.46, 42.67, 41.29, 16.66, 12.39; MS (FAB$^+$) m/z (relative intensity) 269 ($M^{\bullet+}$ + 61.7), 126 (26.8). Anal. Calcd for $C_{13}H_{20}N_2O_4$: C, 58.19; H, 7.51; N, 10.44. Found: C, 58.37; H, 7.71; N, 10.37.

Also isolated was monoester **34** (105 mg, 36%): mp 189–190 °C; IR (KBr) 3490, 3397, 3159, 3032, 2959, 1732, 1715, 1693, 1652, 1479, 1464, 1438, 1369, 1298, 1272, 1243, 1220, 1152, 1003, 907, 874, 762 cm^{-1}; ^1H NMR (CD_3OD) δ 7.59 (s, 1 H), 4.34 (d, J = 10 Hz, 1 H), 3.69 (s, 3 H), 3.65/3.56 (AB of ABX, J_{AB} = 12 Hz, J_{AX} = 4 Hz, J_{BX} = 5.5 Hz, 2 H), 3.20–3.30 (m, J = 5 Hz, 1 H), 2.59 (d, J = 10 Hz, 1 H), 1.89 (s, 3 H), 1.36 (s, 3 H), 0.81 (s, 3 H); ^{13}C NMR (CD_3OD) δ 173.68, 139.93, 110.24, 62.14, 60.45, 52.11, 45.09, 44.84, 39.21, 29.47, 17.63, 12.34; MS (EI) m/z (relative intensity) 296 ($M^{\bullet+}$, 2.0), 265 (3.6), 182 (26.7), 153 (7.8), 127 (17.0), 126 (100); HRMS calcd for $C_{14}H_{23}N_2O_5$ 296.1372, found 296.1368.

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Photoinduced Molecular Transformations. 128.[1] Regioselective [2 + 2] Photocycloaddition of 3-Acetoxyquinolin-2(1*H*)-one with Alkenes and Formation of Furo[2,3-*c*]quinolin-4(5*H*)-ones, 1-Benzazocine-2,3-diones, and Cyclopropa[*d*]benz[1]azepine-2,3-diones via a β-Scission of Cyclobutanoxyl Radicals Generated from the Resulting [2 + 2] Photoadducts

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We have found that [2 + 2]π photoadducts can be obtained by the photoaddition of 3-acetoxyquinolin-2(1*H*)-one with acyclic and cyclic alkenes. The photoaddition of 3-acetoxy-2-quinolin-2(1*H*)-one with 2-methylpropene, 2,3-dimethyl-2-butene, and 2-methoxypropene thus afforded regioselective head-to-tail adducts in 59–97% yields. The photoaddition of 3-acetoxy-2-quinolin-2(1*H*)-one with cyclopentene and cyclohexene resulted in the formation of sterically disfavored cis-cisoid-cis photoadducts as the major products, with the accompanying formation of cis-transoid-cis photoadducts as the minor products in combined yields of 87 and 66%, respectively. The photolysis of the hypoiodites generated in situ from cyclobutanols derived from all of the photoadducts induced β-scissions at the outer bonds of the corresponding cyclobutanoxyl radicals to give furo[2,3-*c*]quinolin-4(5*H*)-ones in 15–50% yields with an accompanying formation of 7- and 8-membered lactams arising from β-scissions at the catacondensed bonds of the cyclobutanoxyl radicals in 2–62% yields. The molecular structure of one of the novel 7-membered lactams that successively fused with cyclopropane and cyclopentane rings was established to be *trans*-5,8,9,10,10a,10b-hexahydro-5-methylcyclopenta[3,4]cyclopropa[1,2-*d*]benzazepine-6,7-dione by X-ray crystallographic analysis. The pathways leading to the formation of all of these products arising from β-scissions are discussed.

In previous papers,[2,3] we reported that the [2 + 2] photoaddition of an alkene to an enolyzed 1,3-dicarbonyl compound or its acetate to form β-ketocyclobutanol or its acetate, followed by a regioselective β-scission of the cy-

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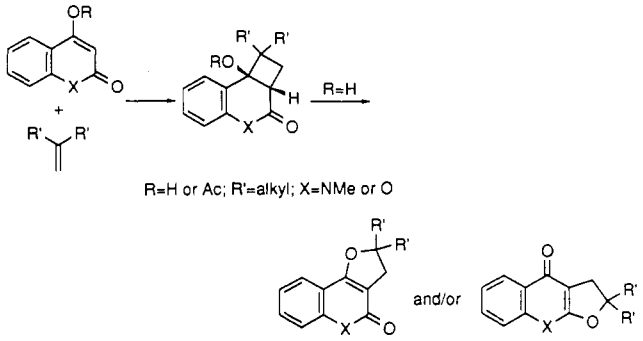
Scheme I

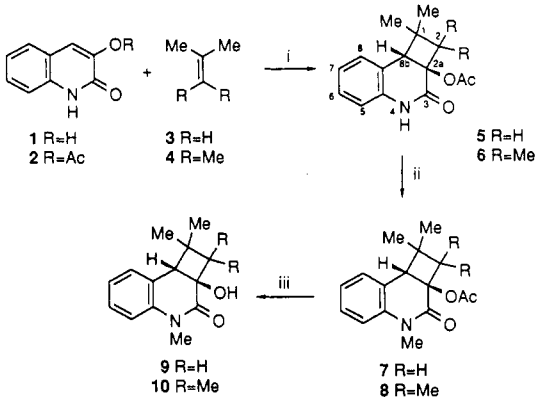


clobutanoxyl radical derived from the resulting cyclobutanol, is a useful process for annelation with a furan ring. For example, 4-hydroxycoumarin and 4-hydroxyquinolin-2(1*H*)-one can be transformed into several furocoumarins, furochromones, furo[3,2-*c*]quinolin-4(5*H*)-ones and furo-[2,3-*b*]quinolin-4(9*H*)-ones, as outlined by the principal sequence of the process given in Scheme I. As part of our continuing investigation[4] to explore the potential of the β-scission of alkoxyl radicals for organic synthesis, we have recently studied the products formed via β-scissions of the cyclobutanoxyl radicals generated from [2 + 2] photoadducts prepared by the photoaddition of 3-acetoxyquinolin-2(1*H*)-one (**2**) with cyclic and acyclic alkenes. There has been no investigation concerning the photoaddition of 3-acetoxyquinolin-2(1*H*)-one (**2**) with alkenes. We found that the photoaddition of the 3-acetoxyquinolinone with unsymmetric alkenes took place in a regioselective manner. We also found that while furo[2,3-*c*]quinolin-4-(5*H*)-ones arising from β-scission of the outer bonds of the cyclobutanoxyl radicals are products in all β-scission reactions (15–50%), novel cyclopropa[*d*]benz[1]azepine-2,3-diones, benzazocine-2,3-diones, and related products arising from β-scissions of the catacondensed bonds of the cyclobutanoxyl radicals comprise substantial accompanying products (combined yields of 14–62%) in these photolysis.

Results

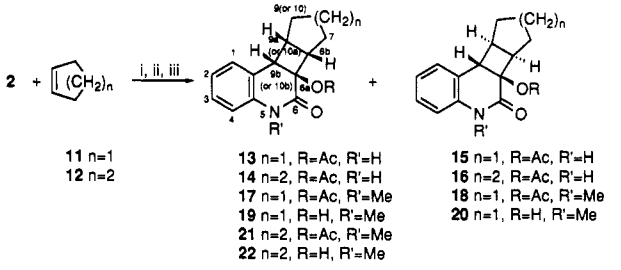
[2 + 2] Photoadditions of 3-Acetoxyquinolin-2-(1*H*)-one (2) with Acyclic or Cyclic Alkenes and Preparation of Cyclobutanols 9, 10, 19, 20, 22, and 27 from the [2 + 2] Photoadducts (Schemes II–IV).

Scheme II[a]



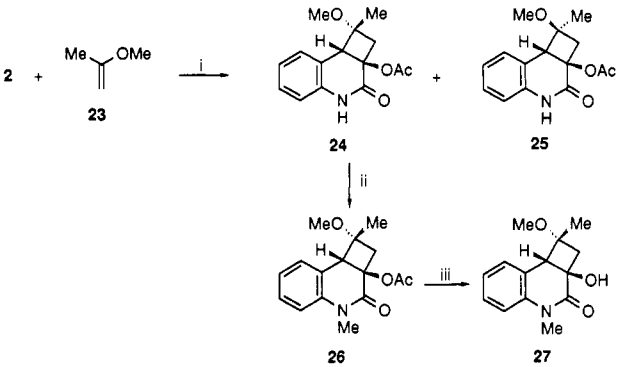
[a] Reagents and conditions: i, *hν*-MeOH; ii, NaH–MeI–DMF, 0 °C–rt; iii, K_2CO_3–MeOH–H_2O.

Scheme III[a]



[a] Reagents and conditions: i, *hν*-MeOH; ii, NaH–MeI–DMF, 0 °C–rt; iii, K_2CO_3–MeOH–H_2O.

Scheme IV[a]



[a] Reagents and conditions: i, *hν*-MeOH; ii, NaH–MeI–DMF, 0 °C–rt; iii, K_2CO_3–MeOH–H_2O.

Acetylation of 3-hydroxyquinolin-2(1*H*)-one (**1**), prepared by the reaction of isatin with diazomethane according to the procedure of Eistert and Selzer,[5] gave the acetate **2**. 2-Methylpropene (**3**), 2,3-dimethyl-2-butene (**4**), cyclopentene (**11**), cyclohexene (**12**), and 2-methoxypropene (**23**) were used as alkenes for the present photoadditions. These photoadditions were carried out, essentially, according to a procedure reported by us[3] for the preparation of the photoadducts of 4-acetoxyquinolin-2(1*H*)-one with alkenes.

Thus, the irradiation of 3-acetoxyquinolin-2(1*H*)-one (**2**) and 10 equiv of 2-methylpropene (**3**) in methanol for 3.5 h under a nitrogen atmosphere with a 500-W high-pressure mercury arc through a Pyrex filter gave, exclusively, a single crystalline [2 + 2] adduct **5** in 97% yield (Scheme II). The ^1H NMR spectrum exhibited a singlet at δ 3.41 that is assignable to 8b-H. The results have proved that

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(2) Suginome, H.; Liu, C. F.; Seko, S.; Kobayashi, K.; Furusaki, A. *J. Org. Chem.* **1988**, *53*, 5952.

(3) Suginome, H.; Kobayashi, K.; Itoh, M.; Seko, S.; Furusaki, A. *J. Org. Chem.* **1990**, *55*, 4933.

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(5) Eistert, B.; Selzer, H. *Chem. Ber.* **1963**, *96*, 1234.

adduct **5** was *cis*-2,2a,4,8b-tetrahydro-2a-acetoxy-1,1-dimethylcyclobuta[*c*]quinolin-3(1*H*)-one (**5**). A similar irradiation of quinolone **2** and 10 equiv of 2,3-dimethyl-2-butene (**4**) in methanol for 13 h also gave a single crystalline adduct **6** in 59% yield (Scheme II).

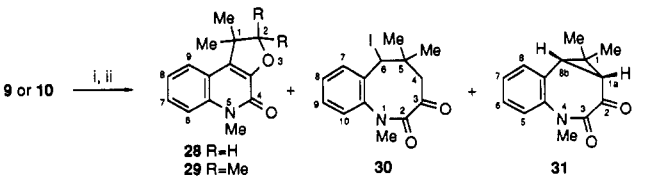
The photoaddition of quinolone **2** with cyclopentene (**11**) (10 equiv) under the above-mentioned conditions gave a 7:3 mixture of cis-cisoid-cis and cis-transoid-cis photoadducts **17** and **18** in 83% yield (Scheme III). Separation of this mixture was achieved after being transformed into the corresponding *N*-methylcyclobutanols **19** and **20** (vide infra) by N-methylation followed by hydrolysis. The configurations of these two isomers, **19** and **20**, were assigned to be cis-cisoid-cis and cis-transoid-cis on the basis of the results of NOE measurements of the NMR spectra. Thus, irradiation of the signal due to 9b-H (δ 3.11) of isomer **19** resulted in an enhancement of the signal due to the 9a-H (δ 2.29); irradiation of the latter signal resulted in an enhancement of the signals due to 9b-H and 6b-H (δ 2.96). Irradiation of the signal due to 6b-H caused an enhancement of the 9a-H signal. These results proved that the isomer had a cis-cisoid-cis configuration. On the other hand, no NOE was observed for the signal due to 9a-H (δ 2.29) when the signal due to 9b-H (δ 3.11) of isomer **20** was irradiated, and vice versa, indicating that 9b-H and 9a-H are disposed trans. Moreover, irradiation of the 9a-H signal resulted in an enhancement of the signal area at δ 2.96 (6b-H). These results indicated that the isomer **20** had a cis-transoid-cis configuration.

The photoaddition of quinolone **2** with cyclohexene (**12**) gave a crystalline [2 + 2] photoadduct **14** (63%) as the major product with a small amount of a minor adduct **16** (3%). The stereochemistry of these two adducts were deduced to be cis-cisoid-cis for the major adduct **14** and cis-transoid-cis for the minor one **16** on the basis of the results of NOE measurements of the ^{1}H NMR spectra. Thus, irradiation of the signal at δ 3.80 due to 10b-H resulted in an enhancement of the area of the signal at δ 2.75 due to 10a-H, while irradiation of the signal due to 10a-H resulted in enhancements of the areas of the signals due to 10b-H and 6b-H (δ 3.04). Moreover, irradiation of the signal due to 6b-H caused an enhancement of the signal area due to 10a-H. These results indicated that isomer **14** had a cis-cisoid-cis stereochemistry.

On the other hand, while irradiation of the signal due to 6b-H at δ 2.94 resulted in an enhancement of the signal due to 10a-H, no enhancement of the signal area due to 10a-H (δ 2.2–2.3) of isomer **16** was observed when the signal at δ 3.55 due to 10b-H was irradiated. These NOE proved that isomer **16** had a cis-transoid-cis stereochemistry.

It is worth noting that the predominant formation of these sterically disfavored cis-cisoid-cis adducts is parallel to the photocycloadditions of the excited 4-hydroxycoumarin and 4-acetoxy-2-quinolone with cyclic alkenes.[2,3] Finally, the photoaddition of quinolone **2** and 2-methoxypropene (**23**) in methanol gave a major photoadduct **24** (55%) as well as a minor one **25** (8%). The ^{1}H NMR spectra of **24** and **25** showed singlets at δ 3.58 and 3.75 attributable to their 8b-H. These results indicated that the two photoadducts are stereoisomers of 2a-acetoxy-2,2a,4,8b-tetrahydro-1-methoxy-1-methylcyclobuta[*c*]-quinolin-3(1*H*)-one. The NOE studies indicated that the OMe groups of **24** and **25** are oriented trans and cis, respectively, to their angular hydrogen (8b-H); irradiation of the signal at δ 3.58 in the ^{1}H NMR of **24** resulted in an enhancement of the signal area due to the 1-Me, while irradiation of the signal at δ 3.75 in the ^{1}H NMR of **25** resulted in an enhancement of the signal area of OMe.

Scheme V[a]



[a] Reagents and conditions: i, HgO–I_2–benzene; ii, *hν*.

The cycloadducts **5**, **6**, **14**, and **24** and a mixture of **13** and **15** were then transformed into their *N*-methyl derivatives **7** and **8** (a mixture of **17**/**18**, **21**, and **26**) with methyl iodide–sodium hydride in DMF. The hydrolysis of the acetoxy group of the *N*-methyl derivatives with potassium carbonate in aqueous methanol at room temperature afforded the corresponding cyclobutanols **9**, **10**, **22**, **27**, and a mixture of **19** and **20** in good yields. At this stage, the mixture of **19** and **20** was subjected to preparative TLC to give two pure isomers, **19** and **20**, as mentioned previously.

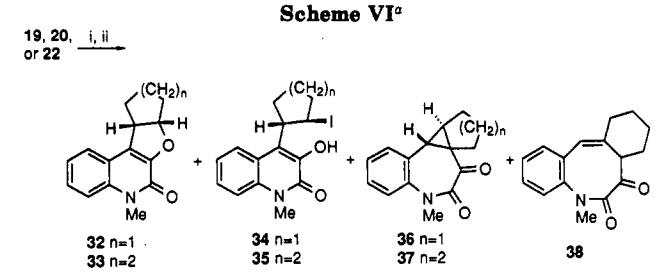
Products in the Photoreactions of Hypoiodites of Fused Cyclobutanols 9, 10, 19, 20, and 27 (Schemes V and VI). The irradiation of cyclobutanol **9** in benzene containing mercury(II) oxide and iodine (each 3 equiv) placed in a Pyrex vessel with a 100-W high-pressure mercury arc under a nitrogen atmosphere while being stirred for 4 h afforded a mixture of products: **28**, **30**, and **31**. This mixture was subjected to preparative TLC to give the three products in 28, 55, and 7% yields, respectively. The molecular formula of crystalline product **28** was established to be $C_{14}H_{15}NO_2$ by high-resolution mass spectrometry. The IR spectrum exhibited two bands at 1624 and 1669 cm^{-1} attributable to an α,β-unsaturated carbonyl group having the quinolinone structure. The ^{1}H NMR spectrum exhibited a singlet (6 H) at δ 1.60 assignable to *gem* dimethyl and a singlet (2 H) at δ 4.39 assignable to $-OCH_2-$ group, in addition to the signals due to aromatic protons and N–Me. These spectral results have indicated that the structure of product **28** was 1,2-dihydro-1,1,5-trimethylfuro[2,3-*c*]quinolin-4(5*H*)-one (Scheme V).

The molecular formula of the major crystalline product **30** was determined to be $C_{14}H_{16}INO$ by high-resolution mass spectrometry. The IR spectrum exhibited two bands at 1654 and 1716 cm^{-1} attributable to the lactam carbonyl and the unstrained cyclic ketone. The ^{1}H NMR spectrum exhibited a double doublet at δ 2.12 (J = 10.99 and 1.10 Hz) and a doublet at δ 3.34 (J = 10.99 Hz) attributable to $-CH_2C{=}O$. It also exhibited a doublet at δ 5.14 (J = 1.10 Hz) assignable to –CHI group. These spectral results, together with a consideration of the formation path, indicated that the structure of product **30** was 1,4,5,6-tetrahydro-6-iodo-1,5,5-trimethyl-1-benzazocine-2,3-dione; the above-mentioned signals are thus assignable to the 4-H, 4-H cis to I, and the 6-H (Scheme VII).

The molecular formula of the minor crystalline product **31** was established to be $C_{14}H_{15}NO_2$ by high-resolution mass spectrometry. The IR spectrum exhibited two bands at 1657 and 1715 cm^{-1} assignable to the lactam carbonyl and the unstrained cyclic ketone. The ^{1}H NMR spectrum exhibited two doublets at δ 2.34 (J = 10.26 Hz) and 2.38 (J = 10.26 Hz). These spectral results, in conjunction with the formation path, indicated that the structure was 1,1a,4,8b-tetrahydro-1,1-dimethylcyclopropa[*d*][1]benzazepine-2,3-dione and that the two doublets are assignable to 1a-H and 8b-H, or vice versa.

The photoreaction of the hypoiodite of cyclobutanol **10** in the presence of mercury(II) oxide and iodine in benzene under conditions similar to those of the reaction of cy-

Scheme VI[a]



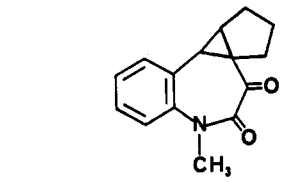
[a] Reagents and conditions: i, HgO–I_2–benzene; ii, *hν*.

clobutanol **9** gave a sole isolable product **29**. Mass spectrometry and combustion analysis established that it had the molecular formula $C_{16}H_{19}NO_2$. The IR spectrum exhibited two bands at 1620 and 1657 cm^{-1} ascribable to the α,β-unsaturated carbonyl function in the quinolinone system. The ^1H NMR spectrum showed a singlet at δ 1.43 (12 H) assignable to a pair of *gem*-dimethyl groups in additions to the signals due to NMe and aromatic protons. These spectral results, along with a consideration of the formation path, indicated that the product was a 2-dimethyl derivative of product **28**.

The photoreaction of the hypoiodite of cyclobutanol **19** in a similar fashion gave a mixture of products **32**, **34**, and **36** in 17, 8, and 20% yields. The molecular formula of crystalline product **32** was established to be $C_{15}H_{15}NO_2$ by mass spectrometric and combustion analyses. The IR spectrum exhibited two bands at 1663 and 1629 cm^{-1} due to the α,β-unsaturated carbonyl group in the quinolinone structure. The ^1H NMR spectrum showed a signal (ddd) centered at δ 4.09 and another signal (ddd) centered at δ 5.48. These spectra indicated that the structure of product **32** was 1,2,3,3a,6,10b-hexahydro-6-methyl-5*H*-cyclopenta-[4,5]furo[2,3-*c*]quinolin-5-one; the above-mentioned signals are thus assignable to 10b-H and 3a-H (Scheme VI). The cyclopentane ring is fused cis with the dihydrofuran ring, since NOE was observed between the signals at δ 4.09 and 5.48.

The molecular formula, $C_{15}H_{16}INO_2$, was established for product **34** by high-resolution mass spectrometry. The IR spectrum exhibited a band at 3270 cm^{-1} attributable to the hydroxy and two bands at 1628 and 1617 cm^{-1} ascribable to the quinolinone structure. The ^1H NMR spectrum exhibited a signal at δ 4.97 (1 H, dt) ascribable to a proton attached to a carbon carrying an iodine atom. The mass spectrum indicated a base peak at an m/z of 242 due to the $(M - I)^+$ ion. These spectral results, together with a consideration of the formation path, indicated that the structure of product **34** was 3-hydroxy-4-(*trans*-2-iodocyclopentyl)-1-methylquinolin-2(1*H*)-one. The two substituents attached to the cyclopentane ring should be trans oriented, based on the coupling constants of the 2′-H signal (dt, J = 5.49 and 5.13 Hz).

The molecular formula of product **36** was established to be $C_{15}H_{15}NO_2$ by mass spectrometric and combustion analyses. The IR spectrum exhibited two bands at 1687 and 1654 cm^{-1} attributable to the COCONMe– group. The ^1H NMR spectrum exhibited three signals (each 1 H) at δ 2.25 (ddd), 2.46 (d), and 2.93 (t). Although these spectral results, coupled with the formation path, suggested a tetracyclic structure **36** with a cyclopropane ring in it, the assignment of the signals of the ^1H NMR spectrum in terms of structure **36** was not unambiguous. The molecular structure of product **36**—*trans*-5,8,9,10,10a,10b-hexahydro-5-methylcyclopenta[3,4]cyclopropa[1,2-*d*][1]benzazepine-6,7-dione—was, therefore, determined by an X-ray crystallographic analysis (Figure 1). Details concerning



36

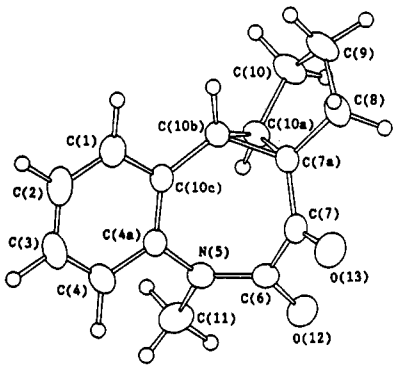


Figure 1.

the analysis are recorded in the Experimental Section.

The photoreaction of the hypoiodite of cis-transoid-cis cyclobutanol **20** under the conditions mentioned above similarly gave products **32**, **34**, and **36** in 50, 12, and 2% yields. It should be noted that the yield of furanoquinolone **32** was appreciably higher than in the case of cis-cisoid-cis cyclobutanol **19**.

The irradiation of cyclobutanol **22** in benzene containing mercury(II) oxide and iodine under the conditions mentioned above gave four products, (**33**, **35**, **37**, and **38**) in 15, 16, 8, and 16% yields. The IR spectrum of product **33**, $C_{16}H_{17}NO_2$, determined by combustion and mass spectrometric analyses, exhibited two bands at 1661 and 1624 cm^{-1} ascribable to the quinolinone group. The ^1H NMR spectrum of product **33** revealed two characteristic one-proton signals at δ 3.37 (dt) and 4.81 (ddd) which are assignable to an allylic and a proton attached to the carbon carrying an oxygen atom, respectively. These spectral results indicated that the structure was 1,2,3,4,4a,11b-tetrahydro-7-methylcyclohexa[4,5]furo[2,3-*c*]quinolin-6-(7*H*)-one and that the above-mentioned two signals are ascribable to 11a-H and 7a-H, which are disposed cis ($J_{11a\text{-}H\text{-}7a\text{-}H}$ = 6.84 Hz).

The IR spectrum of product **35**, $C_{16}H_{18}INO_2$, indicated the presence of the –C=C(OH)CO– group. The ^1H NMR spectrum exhibited a one-proton signal at δ 5.07 (br s) assignable to the –CHI group. These spectral results, together with a consideration of the formation path, indicated that the structure was 3-hydroxy-4-(*trans*-2-iodocyclohexyl)-1-methylquinolin-2(1*H*)-one (**35**).

The molecular formula of product **37** was established to be $C_{16}H_{17}NO_2$ by combustion and mass spectrometric analyses. The IR spectrum showed the presence of two carbonyl group at 1688 and 1650 cm^{-1}. The ^1H NMR spectrum exhibited one-proton doublet at δ 2.36 (J = 6.96 Hz). These and other spectral results were very similar to those of product **36** with a cyclopropane ring, indicating that the structure was *trans*-8,9,10,11,11a,11b-hexahydro-5-methylcyclohexa[3,4]cyclopropa[1,2-*d*][1]benzazepine-6,7-dione (**37**), homologous to the product **36**. The doublet at δ 2.36 in the ^1H NMR is assignable to 11b-H, and the coupling constant indicated that 11a-H and 11b-H are oriented trans.

The molecular formula of product **38** was established to be $C_{16}H_{17}NO_2$ by means of combustion and mass

spectrometric analyses. The IR spectrum exhibited two bands at 1711 and 1648 cm^{-1}, assignable to the -N(Me)-COCO- group. The ^1H NMR spectrum revealed a set of one-proton signals at δ 2.71 (dddd), 3.61 (dd), and 5.69 (s), apart from those of the aromatic protons. These spectral results, in conjunction with the pathway of its formation, indicated that the structure was 5,7a-dihydro-5-methylcyclohexa[*d*][1]benzazocine-6,7-dione (**38**). The above-mentioned three signals are assignable to 11-H, 7a-H, and 12-H, respectively.

The photoreaction of cyclobutanol **27** under conditions similar to those of the photoreaction of the above-mentioned cyclobutanols gave only an intractable mixture.

Discussion

The above-mentioned experiments have indicated that [2 + 2] photoadducts are formed in 59–97% yields by the photoaddition of 3-acetoxyquinolin-2(1*H*)-one (**2**) with alkenes, analogous to the photoaddition of 4-acetoxyquinolin-2(1*H*)-one.[3] The photoaddition of quinolinone **2** with 2-methylpropene (**3**) and 2-methoxypropene (**23**) was regioselective, indicating adduct formation through biradical intermediates.[6]

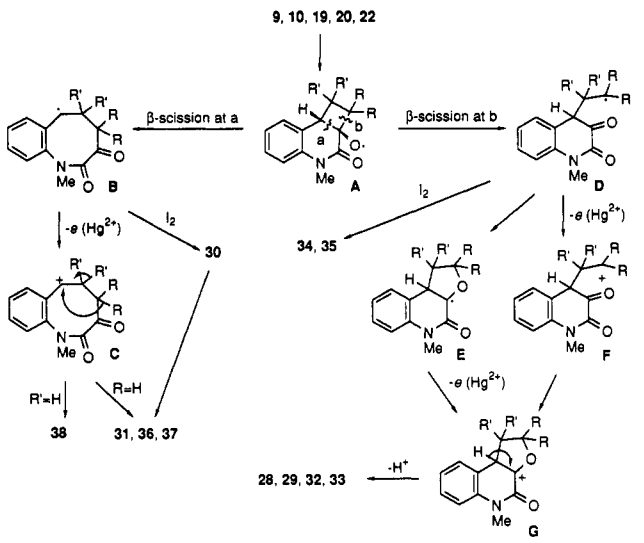
It is worth noting here that the direction in the regiochemistry of the addition of 2-methylpropene (**3**) and 2-methoxypropene (**23**) to quinolinone **2** is parallel to the photoaddition of 4-acetoxyquinolin-2(1*H*)-one, as previously reported by us.[3] This result implies that the acetoxy group attached to the 3- or 4-position of the quinolinones has very little directing power in determining the regiochemistry in this photoaddition.

The aforementioned experiments have also shown that the photoaddition of quinolinone **2** with cyclopentene and cyclohexene resulted in a highly predominant formation of the cis-cisoid-cis adducts. The preferential formation of these sterically disfavored adducts is again parallel to the behavior of the excited 4-hydroxycoumarin and 4-acetoxyquinolin-2(1*H*)-one, as reported by us,[2,3] and is in contrast to the behavior of the excited cyclic enones in which sterically favored cis-transoid-cis adducts have always been the preferential addition products.[6] Although no study concerning the mechanistic details of the present photoaddition has been undertaken, the preferential formation of a sterically disfavored isomer, which may well involve a singlet exciplex, is of considerable mechanistic interest.

The aforementioned experiments have also disclosed that all of the alkoxyl radicals generated from cyclobutanols **9**, **10**, **19**, **20**, and **22** by the photolysis of their hypoiodites prepared in situ with mercury(II) oxide and iodine cleave to give furo[2,3-*c*]quinolin-4(5*H*)-ones **28**, **29**, **32**, and **33** in 15–50% yields with an accompanying formation of 7- and 8-membered lactams arising from ring expansion in up to 62% yields.

The probable paths leading to products **28**–**38** in β-scissions of the alkoxyl radicals generated from the cyclobutanols derived from the photoadducts **9**, **10**, **19**, **20**, and **22** are shown in Scheme VII. Thus, photolysis of the hypoiodites generated in situ by the reaction of iodine oxide and cyclobutanols generates an alkoxyl radical A. A β-scission at either bond a or b affords a carbon-centered radical B or D. The extraction of an iodine atom by either radical B or D gives rise to product **30**, **34**, or **35**. Eight-membered lactam **38** should be formed by the one-electron

Scheme VII



oxidation of the carbon radical B followed by the loss of a proton from the resulting carbocation C. Novel cyclopropabenzazepine diones **31**, **36**, and **37** may be formed through iodides such as product **30**, reported previously by us,[7] since iodide **30** can be transformed into the cyclopropane derivative **31** with a base, such as DBU. The alternative path involving an acid-catalyzed cyclization of the carbocation C for the formation of these products, however, can not be excluded, since no iodides can be isolated from the reactions of cyclobutanols **19**, **20**, or **22**.

Furo[2,3-*c*]quinolin-4(5*H*)-ones **28**, **29**, **32**, **33** produced in 15–50% in all of the foregoing β-scission of the alkoxyl radicals, on the other hand, may be formed by a loss of a proton from cationic intermediates G generated from a radical E or a cationic intermediate F. We have already reported a number of examples of this type of the reaction.[2,3,4a,f,i,q,l]

There have been only sporadic studies concerning the formation of furo[2,3-*c*]quinolin-4(5*H*)-ones.[8] The formation of such heterocycles via the [2 + 2] photoaddition of 3-acetoxyquinolin-2(1*H*)-one with alkenes followed by a β-scission of the alkoxyl radicals generated from the resulting cyclobutanols described here may have potential in the synthesis of molecules of this class. Moreover, whatever is the mechanism of their formation, the β-scission reaction described here has been disclosed to result in the formation of unprecedented novel fused benzazepinediones.

Experimental Section

General Method. Mp's were determined with a Yanagimoto mp apparatus and are uncorrected. IR spectra were determined for Nujol mulls with a JASCO IR-810 spectrophotometer unless stated otherwise. ^1H NMR spectra were determined in $CDCl_3$ ($SiMe_4$ as internal reference) with a Hitachi R-90H operating at 90 MHz, a JEOL JNM-GX 270 operating at 270 MHz, or a JEOL JNM-EX 400 operating at 400 MHz. High- and low-resolution mass spectra were recorded with a JEOL JMS-DX 303 spectrometer. TLC was carried out on Merck Kieselgel 60 PF_{254}. The

(6) For reviews of enone photochemical cycloaddition, see: (a) Baldwin, S. W. In *Organic Photochemistry*; Padwa, A., Ed.; Marcel Dekker: New York, 1981; Vol. 5, p 123. (b) Weedon, A. C. In *Synthetic Organic Photochemistry*; Horspool, W. M., Ed.; Plenum: New York, 1980; p 91.

(7) Suginome, H.; Itoh, M.; Kobayashi, K. *J. Chem. Soc., Perkin Trans. 1* **1988**, 491.

(8) (a) Kanaoka, Y.; Itoh, K. *Synthesis* **1972**, *36*. (b) Ninomiya, I.; Kiguchi, T.; Naito, T. *Heterocycles* **1978**, *9*, 1023. (c) Forrester, A. R.; Johansson, E. M.; Thomson, R. H. *J. Chem. Soc., Perkin Trans 1* **1979**, 1112. (d) Kanaoka, Y.; San-nohe, K. *Tetrahedron Lett.* **1980**, *21*, 3893. (e) Bates, R. B.; Kane, U. V.; Martin, A. R.; Mujumdar, R. B.; Ortega, R.; Hatanaka, Y.; San-nohe, K.; Kanaoka, Y. *J. Org. Chem.* **1987**, *52*, 3178. (f) Yamaguchi, S.; Ohhira, Y.; Yamada, M.; Michitani, H.; Kawase, Y. *Bull. Chem. Soc. Jpn.* **1990**, *63*, 952.

photoadditions were carried out with a 500-W high-pressure Hg arc lamp (Eikosha, EHB-WI-500), and the photolysis of the hypoiodites was carried out with a 100-W high-pressure Hg arc lamp (Eikosha, EHB-WU-100).

3-Hydroxyquinolin-2(1*H*)-one (1). This compound was prepared according to the procedure of Eistert and Selzer.[5]

3-(Acetyloxy)quinolin-2(1*H*)-one (2). A mixture of **1** (4.16 g, 25.8 mmol), acetic anhydride (7.52 g, 77.4 mmol), and pyridine (100 mL) was stirred for 27 h at room temperature. The resulting mixture was then poured into ice–water (400 mL), and the precipitate was filtered off. The crude product was recrystallized from ethanol to give a pure *O*-acetyl derivative **2** (3.41 g, 65%): mp 223–225 °C; IR 3600–2500 (OH), 1767 (ester C=O), 1682 (lactam C=O) cm^{-1}; ^1H NMR (90 MHz) δ 2.42 (3 H, s, OAc), 7.1–7.7 (5 H, m), 11.4–11.7 (1 H, br, NH); MS *m/z* 203 (M^+, 7.9), 161 [$(M - CH_2CO)^+$, 100]. Anal. Calcd for $C_{11}H_9NO_3$: C, 65.02; H, 4.46; N, 6.89. Found: C, 64.84; H, 4.39; N, 6.77.

***cis*-2a-(Acetyloxy)-2,2a,4,8b-tetrahydro-1,1-dimethylcyclobuta[*c*]quinolin-3(1*H*)-one (5).** A solution of **2** (300 mg, 1.48 mL) and 2-methylpropene (830 mg, 14.8 mmol) in methanol (100 mL) was irradiated for 3.5 h under a nitrogen atmosphere through Pyrex with a 500-W high-pressure mercury arc. Removing the solvent and excess 2-methylpropene gave an oil which was subjected to preparative TLC on silica gel with 1:2 ethyl acetate–dichloromethane to afford **5** (371 mg, 97%): mp 174–188 °C (diethyl ether–hexane); IR 3210 (NH), 1737 (OAc), 1684 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.77 (3 H, s, 1-Me), 1.34 (3 H, s, 1-Me), 2.10 (3 H, s, OAc), 2.52 (1 H, d, J = 13.92 Hz, 2-H), 2.58 (1 H, d, J = 13.92 Hz, 2-H), 3.41 (1 H, s, 8b-H), 6.74 (1 H, d, J = 8.06 Hz, 5-H), 6.9–7.05 (2 H, m, 7- and 8-H), 7.17 (1 H, t, J = 7.69 Hz, 6-H), 7.82 (1 H, br s, NH); MS *m/z* 259 (M^+, 2.0), 203 [$(M - CH_2CMe_2)^+$, 9.0], 161 [$(M - CH_2CMe_2 - CH_2CO)^+$, 100]. Anal. Calcd for $C_{15}H_{17}NO_3$: C, 69.48; H, 6.61; N, 5.40. Found: C, 69.44; H, 6.59; N, 5.41.

***cis*-2a-(Acetyloxy)-2,2a,4,8b-tetrahydro-1,1,2,2-tetramethylcyclobuta[*c*]quinolin-3(1*H*)-one (6).** A solution of **2** (400 mg, 1.97 mmol) and 2,3-dimethyl-2-butene (1.66 g, 19.4 mmol) in methanol (130 mL) was irradiated for 13 h, as described above, to give **6** (336 mg, 59%): mp 197–198 °C (diethyl ether–hexane); IR 3188 (NH), 1741 (OAc), 1684 (lactam C=O) cm^{-1}; ^1H NMR (90 MHz) δ 0.67 (3 H, s), 1.09 (3 H, s), 1.22 (3 H, s), 1.31 (3 H, s), 2.08 (3 H, s, Ac), 3.35 (1 H, s, 8b-H), 6.69 (1 H, d, J = 7.59 Hz, 5-H), 6.8–7.25 (3 H, m), 7.6–7.9 (1 H, br, NH); MS *m/z* 287 (M^+, 6.1), 228 [$(M - OAc)^+$, 6.2], 161 [$(M - Me_2CCMe_2 - CH_2CO)^+$, 100]. Anal. Calcd for $C_{17}H_{21}NO_3$: C, 71.06; H, 7.37; N, 4.87. Found: C, 71.04; H, 7.30; N, 4.89.

(±)-(6aα,6bα,9aα,9bα)- (13) and (±)-(6aα,6bβ,9aβ,9bα)-6a-(Acetyloxy)-5,6a,6b,7,8,9,9a,9b-octahydro-6*H*-cyclopenta[3,4]cyclobuta[1,2-*c*]quinolin-6-one (15). A solution of **2** (849 mg, 4.18 mmol) and cyclopentene (2.81 g, 41.8 mmol) in methanol (270 mL) was irradiated for 6 h, as described above, to give a mixture of stereoisomers **13** and **15** (937 mg, 83%): mp 236–250 °C (after purification by preparative TLC with a 1:1 ethyl acetate–dichloromethane); IR 3202 (NH), 1742 (OAc), 1684 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 2.09, 2.13 and 1.25–2.2 (9 H, 2s and m), 2.3–2.45 (1 H, m), 3.08 (0.3 H, d, J = 5.87 Hz, 9b-H of **15**), 3.15–3.35 (2 H, m), 3.82 (0.7 H, d, J = 8.43 Hz, 9b-H of **13**), 6.67 (0.7 H, dd, J = 7.69, 1.10 Hz, 4-H of **13**), 6.74 (0.3 H, d, J = 8.43 Hz, 4-H of **15**), 6.9–7.2 (3 H, m), 7.77 and 7.84 (1 H, 2bs, s, NH); MS *m/z* 271 (M^+, 4.8), 203 [$(M - C_5H_8)^+$, 5.6], 161 [$(M - C_5H_8 - CH_2CO)^+$, 100]. Anal. Calcd for $C_{16}H_{17}NO_3$ C, 70.86; H, 6.28; N, 5.16. Found: C, 70.62; H, 6.28; N, 5.19.

(±)-(6aα,6bα,10aα,10bα)- (14) and (±)-(6aα,6bβ,10aβ,10bα)-6a-(Acetyloxy)-6a,6b,7,8,9,10,10a,10b-octahydrobenzo[3,4]cyclobuta[1,2-*c*]quinolin-6(5*H*)-one (16). A solution of **2** (400 mg, 1.97 mmol) and cyclohexene (1.66 g, 19.7 mmol) in methanol (130 mL) was irradiated for 8 h, as described above, to give **14** (recrystallization from diethyl ether, 355 mg, 63%) and **16** (recrystallization from ethyl acetate, 15 mg, 3%). **14**: mp 169–172 °C; IR 3100 (br, NH), 1747 (Ac), 1681 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.95–1.7 (7 H, m), 1.95–2.1 and 2.06 (4 H, m and s), 2.75 (1 H, br q, J = 9.3 Hz, 10a-H) 3.04 (1 H, ddd, J = 10.26, 8.06, 4.40 Hz, 6b-H), 3.80 (1 H, dd, J = 9.53 Hz, 10b-H), 6.78 (1 H, d, J = 7.70 Hz, 4-H), 6.95–7.2 (3 H, m), 8.32, (1 H, br s, NH); MS *m/z* 285 (M^+, 3.1), 225 [$(M - AcOH)^+$, 4.8], 203 [$(M - C_6H_{10})^+$, 6.5], 161 [$(M - C_6H_{10} - CH_2CO)^+$, 100]. Anal. Calcd for $C_{17}H_{19}NO_3$: C, 71.56; H, 6.71; N, 4.91. Found: C, 71.58; H, 6.77; N, 4.93. **16**: mp 240 °C dec; IR 3180 (NH), 1740 (Ac), 1680 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz), δ 1.05–1.25 (1 H, m), 1.4–1.85 (6 H, m), 1.95–2.1 and 2.05 (4 H, m and s), 2.2–2.3 (1 H, m, 10a-H), 2.94 (1 H, br q, J = 9.0 Hz, 6b-H), 3.55 (1 H, d, J = 9.53 Hz, 10b-H), 6.76 (1 H, d, J = 7.69 Hz, 4-H), 6.95–7.2 (3 H, m), 7.87 (1 H, br s, NH); MS *m/z* 285 (M^+, 4.0), 226 [$(M - AcO)^+$, 4.3], 203 [$(M - C_6H_{10})^+$, 6.4], 161 [$(M - C_6H_{10} - CH_2CO)^+$, 100]. Anal. Calcd for $C_{17}H_{19}NO_3$: C, 71.56; H, 6.71; N, 4.91. Found: C, 71.42; H, 6.72; N, 4.93.

1,2a,8b-*trans*,*cis*- (24) and 1,2a,8b-*cis*,*cis*-2a-(Acetyloxy)-2,2a,4,8b-tetrahydro-1-methoxy-1-methylcyclobuta[*c*]quinolin-3(1*H*)-one (25). A solution of **2** (720 mg, 3.55 mmol) and 2-methoxypropene (2.56 g, 35.5 mmol) in methanol (235 mL) was irradiated for 4 h, as described above, to give **24** (535 mg, 55%) and **25** (77 mg, 8%). **24**: mp 215–216 °C (diethyl ether–hexane); IR 3196 (NH), 1745 (OAc), 1683 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 1.50 (3 H, s, 1-Me), 2.09 (3 H, s, Ac), 2.54 (1 H, dd, J = 13.93, 1.83 Hz, 2-H cis to 1-Me), 2.87 (1 H, d, J = 13.93 Hz, 2-H cis to 1-OMe), 2.95 (3 H, s, OMe), 3.58 (1 H, br s, 8b-H), 6.73 (1 H, d, J = 8.43 Hz, 5-H), 6.9–7.2 (3 H, m), 8.10 (1 H, br s, NH); MS *m/z* 275 (M^+, 0.09), 216 [$(M - OAc)^+$, 31], 161[$(M - CH_2C(OMe)Me - CH_2CO)^+$, 100]. Anal. Calcd for $C_{15}H_{17}NO_4$: C, 65.44; H, 6.22; N, 5.09. Found: C, 65.35; H, 6.20; N, 4.97. **25**: mp 222–224 °C (diethyl ether); IR 3194 (NH), 1745 (OAc), 1677 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.99 (3 H, s, 1-Me), 2.09 (3 H, s, Ac), 2.72 (1 H, dd, J = 13.92, 1.10 Hz, 2-H cis to 1-OMe), 2.88 (1 H, dd, J = 13.92 Hz, 2-H cis to 1-Me), 3.28 (3 H, s, OMe), 3.75 (1 H, br s, 8b-H), 6.80 (1 H, d, J = 7.32 Hz, 5-H), 7.0–7.2 (3 H, m), 8.59 (1 H, br s, NH); MS *m/z* 276 [$(M + 1)^+$, 0.27], 275 (M^+, 0.07), 216 [$(M - OAc)^+$, 42], 161 [$(M - CH_2C(OMe)Me - CH_2CO)^+$, 100]. Anal. Calcd for $C_{15}H_{17}NO_4$: C, 65.44; H, 6.22; N, 5.09. Found: C, 65.15; H, 6.15; N, 5.03.

***cis*-2a-(Acetyloxy)-2,2a,4,8b-tetrahydro-1,1,4-trimethylcyclobuta[*c*]quinolin-3(1*H*)-one (7).** Photoadduct **5** (355 mg, 1.37 mmol) dissolved in DMF (4 mL) was added to a stirred suspension of NaH (76 mg, 50% oil suspension, 1.58 mmol) in DMF (2.6 mL) at 0 °C; the solution was stirred for 15 min. Methyl iodide (226 mg, 1.59 mmol) was then added, and the solution was stirred for another 30 min at room temperature. The reaction was quenched with aqueous NH_4Cl, and the product was extracted with diethyl ether. The extract was washed with water and brine and dried over anhydrous $MgSO_4$. Evaporation of the solvent gave a residue, which was purified by preparative TLC on silica gel with 1:5 ethyl acetate–dichloromethane to afford **7** (313 mg, 84%): mp 110–112 °C (hexane–diethyl ether); IR 1738 (OAc), 1670 (lactam C=O) cm^{-1}; ^1H NMR (90 MHz) δ 0.71 (3 H, s, 1-Me), 1.32 (3 H, s, 1-Me), 2.05 (3 H, s, Ac), 2.42 (1 H, d, J = 13.75 Hz, 2-H), 2.60 (1 H, d, J = 13.75 Hz, 2-H), 3.40 (3 H, s, NMe), 6.8–7.3 (4 H, m); MS *m/z* 273 (M^+, 4.6), 214 [$(M - OAc)^+$, 7.1], 175 [$(M - Me_2CCH_2 - CH_2CO)^+$, 100]. Anal. Calcd for $C_{16}H_{19}NO_3$: C, 70.31; H, 7.01; N, 5.12. Found: C, 70.34; H, 7.08; N, 5.25.

***cis*-2a-(Acetyloxy)-2,2a,4,8b-tetrahydro-1,1,2,2,4-pentamethylcyclobuta[*c*]quinolin-3(1*H*)-one (8).** Photoadduct **6** (203 mg, 0.71 mmol) in DMF (2 mL) was treated with NaH (38 mg, 50% oil suspension, 0.79 mmol), then methyl iodide (114 mg, 0.80 mmol) in DMF (3 mL) as described above to give **8** (191 mg, 89%): mp 125–127 °C (hexane–diethyl ether); IR 1735 (OAc), 1662 (lactam C=O) cm^{-1}; ^1H NMR (90 MHz) δ 0.61 (3 H, s), 1.00 (3 H, s), 1.20 (3 H, s), 1.31 (3 H, s), 2.04 (3 H, s, Ac), 3.34 (1 H, s, 8b-H), 3.39 (3 H, s, NMe), 6.75–7.3 (4 H, m); MS *m/z* 301 (M^+, 4.8), 175 [$(M - Me_2CCMe_2 - CH_2CO)^+$, 100]. Anal. Calcd for $C_{18}H_{23}NO_3$: C, 71.73; H, 7.69; N, 4.65. Found: C, 71.59; H, 7.87; N, 4.59.

(±)-(6aα,6bα,9aα,9bα)- (17) and (±)-(6aα,6bβ,9aβ,9bα)-6a-(Acetyloxy)-5,6a,6b,7,8,9,9a,9b-octahydro-5-methyl-6*H*-cyclopenta[3,4]cyclobuta[1,2-*c*]quinolin-6-one (18). A mixture of **13** and **15** (ca. 7:3, 104 mg, 0.384 mmol) in DMF (1.3 mL) was treated with NaH (20 mg, 50% oil suspension, 0.417 mmol) and then methyl iodide (60 mg, 0.423 mmol) in DMF (2 mL), as described above, to give a mixture of **17** and **18** (ca. 7:3, 96 mg, 81%): mp 170–241 °C (after purification by preparative TLC on silica gel); IR 1738 (OAc), 1667 (lactam C=O) cm^{-1}; ^1H NMR (400 MHz) δ 0.75–1.95 (6 H, m), 2.06 (0.9 H, s, Ac of **18**), 2.10 (2.1 H, s, Ac of **17**), 2.2–2.45 (1 H, m), 3.06 (0.3 H, d, J = 5.86 Hz, 9b-H of **18**), 3.1–3.3 (1 H, m), 3.81 (0.7 H, dd, J = 10.25, 1.95 Hz, 9b-H

of 17), 6.85–7.05 (3 H, m), 7.2–7.3 (1 H, m); MS *m/z* 285 (M^+, 9.8), 175 [$(M - C_5H_8 - CH_2CO)^+$, 100]. Anal. Calcd for $C_{17}H_{19}NO_3$: C, 71.56; H, 6.71; N, 4.91. Found: C, 71.69; H, 6.77; N, 4.88.

(±)-(6aα,6bα,10aα,10bα)-6a-(Acetyloxy)-6a,6b,7,8,9,10,10a,10b-octahydro-5-methylbenzo[3,4]cyclobuta[1,2-*c*]quinolin-6(5*H*)-one (21). Photoadduct **14** (137 mg, 0.481 mmol) in DMF (2.5 mL) was treated with NaH (26 mg, 50% oil suspension, 0.542 mmol) and then methyl iodide (75 mg, 0.530 mmol) in DMF (1.6 mL), as described above, to give **21** (125 mg, 0.418 mmol): mp 152–154 °C (hexane–diethyl ether); IR 1739 (OAc), 1662 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.8–1.1 (3 H, m), 1.3–1.6 (4 H, m), 1.95–2.1 and 2.02 (4 H, m and s), 2.72 (1 H, quintet, *J* = 9.8 Hz, 10a-H), 3.03 (1 H, ddd, *J* = 10.63, 8.06, 3.67 Hz, 6b-H), 3.41 (3 H, s, NMe), 3.77 (1 H, d, *J* = 9.53 Hz, 10b-H), 6.95–7.3 (4 H, m); MS *m/z* 299 (M^+, 2.9), 175 [$(M - C_6H_{10} - CH_2CO)^+$, 100]. Anal. Calcd for $C_{18}H_{21}NO_3$: C, 72.22; H, 7.07; N, 4.68. Found: C, 72.14; H, 7.19; N, 4.62.

1,2a,8b-*trans*,*cis*-2a-(Acetyloxy)-2,2a,4,8b-tetrahydro-1-methoxy-1,4-dimethylcyclobuta[*c*]quinolin-3(1*H*)-one (26). Photoadduct **24** (380 mg, 1.38 mmol) in DMF (2.3 mL) was treated with NaH (73 mg, 50% suspension, 1.52 mmol) and then methyl iodide (216 mg, 1.52 mmol) in DMF (3.5 mL) to give **26** (300 mg, 75%): mp 120–123 °C (hexane–diethyl ether); IR 1743 (OAc), 1656 (lactam C=O) cm^{-1}; ^1H NMR (400 MHz) δ 1.49 (3 H, s, 1-Me), 2.05 (3 H, s, Ac), 2.54 (1 H, dd, *J* = 13.67 Hz, 1.95 Hz, 2-H cis to 1-Me), 2.83 (1 H, d, *J* = 13.67 Hz, cis to 1-OMe), 2.93 (3 H, s, OMe), 3.41 (3 H, s, NMe), 6.95–7.05 (3 H, m), 7.29 (1 H, dd, *J* = 8.31, 1.47 Hz, 8-H); MS *m/z* 290 [$(M + 1)^+$, 0.33], 230 [$(M - AcO)^+$, 61), 175 [$(M - CH_2C(OMe)Me - CH_2CO)^+$, 100]. Anal. Calcd for $C_{16}H_{19}NO_4$: C, 66.42; H, 6.62; N, 4.84. Found: C, 66.03; H, 6.68; N, 4.79.

***cis*-2,2a,4,8b-Tetrahydro-2a-hydroxy-1,2,4-trimethylcyclobuta[*c*]quinolin-3(1*H*)-one (9).** A solution of **7** (296 mg, 1.08 mmol) in 80% aqueous methanol (9 mL) containing K_2CO_3 (90 mg, 0.65 mmol) was stirred for 68 h at room temperature. The solvent was then removed under reduced pressure; the residue was extracted with diethyl ether. The extract was washed with brine and dried over anhydrous $MgSO_4$. Evaporation of the solvent gave a residue which was purified by preparative TLC on silica gel with 10:1 dichloromethane–ethyl acetate to afford **9** (193 mg, 77%): mp 154–155 °C (diethyl ether–hexane); IR 3280 (OH), 1643 (C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.68 (3 H, s), 1.35 (3 H, s), 2.25 (1 H, d, *J* = 12.46 Hz, 2-H), 2.45 (1 H, d, *J* = 12.46 Hz, 2-H), 3.40 (3 H, s, NMe), 3.44 (1 H, s, 8b-H), 7.0–7.3 (4 H, m); MS *m/z* 231 (M^+, 3.4), 214 [$(M - OH)^+$, 5.6], 175 [$(M - CH_2CMe_2)^+$, 100]. Anal. Calcd for $C_{14}H_{17}NO_2$: C, 72.70; H, 7.41; N, 6.01. Found: C, 72.95; H, 7.53; N, 6.01.

***cis*-2,2a,4,8b-Tetrahydro-2a-hydroxy-1,1,2,2,4-pentamethylcyclobuta[*c*]quinolin-3(1*H*)-one (10).** The hydrolysis of **8** (186 mg, 0.621 mmol) was carried out with K_2CO_3 (52 mg, 0.377 mmol) in 80% aqueous methanol (10 mL) for 24 h, as described above, to give **10** (154 mg, 96%): mp 206–209 °C (diethyl ether); IR 3370 (OH), 1650 (C=O) cm^{-1}; ^1H NMR (90 MHz) δ 0.62 (3 H, s), 0.93 (3 H, s), 1.16 (3 H, s), 1.24 (3 H, s), 3.34 (1 H, s, 8b-H), 3.54 (3 H, s, NMe), 6.95–7.3 (4 H, m); MS *m/z* 259 (M^+, 0.17), 241 [$(M - H_2O)^+$, 0.36], 226 [$(M - H_2O - Me)^+$, 0.74], 175 [$(M - Me_2CCMe_2)^+$, 100]. Anal. Calcd for $C_{16}H_{21}NO_2$: C, 73.96; H, 8.16; N, 5.40. Found: C, 73.76; H, 8.08; N, 5.44.

(±)-(6aα,6bα,9aα,9bα)- (19) and (±)-(6aα,6bβ,9aβ,9bα)-5,6a,6b,7,8,9,9a,9b-Octahydro-6a-hydroxy-5-methyl-6*H*-cyclopenta[3,4]cyclobuta[1,2-*c*]quinolin-6-one (20). The hydrolysis of a mixture of **17** and **18** (611 mg, 2.14 mmol, ca. 7:3) was carried out with K_2CO_3 (179 mg, 1.30 mmol) in 80% aqueous methanol (18 mL) for 16 h, as described above, to give **19** (335 mg, 64%) and **20** (122 mg, 23%). **19**: mp 142–144 °C (hexane–diethyl ether); IR 3370 (OH), 1645 (C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.95–1.1 (1 H, m), 1.2–1.65 (4 H, m), 1.7–1.8 (m, 1 H), 3.01 (1 H, dd, *J* = 8.06, 6.23 Hz, 6b-H), 3.25 (1 H, br q, *J* = 8.4 Hz, 9a-H), 3.37 (3 H, s, NMe), 3.86 (1 H, d, *J* = 9.53 Hz, 9b-H), 6.9–7.0 (2 H, m), 7.06 (1 H, ddd, *J* = 7.70, 7.33, 1.10 Hz, 3-H), 7.22 (1 H, dd, *J* = 8.06, 1.10 Hz, 1-H); MS *m/z* 243 (M^+, 2.9), 175 [$(M - C_5H_8)^+$, 100]. Anal. Calcd for $C_{15}H_{17}NO_2$: C, 74.05; H, 7.04; N, 5.76. Found: C, 73.86; H, 7.10; N, 5.70. **20**: mp 178–180 °C, IR 3390 (OH), 1643 (C=O) cm^{-1}; ^1H NMR (270 MHz) δ 1.45–1.65 (2 H, m), 1.8–2.05 (3 H, m), 2.17 (1 H, dd, *J* = 13.19, 6.59 Hz), 2.29 (1 H, dt, *J* = 7.69, 5.86 Hz, 9a-H), 2.96 (1 H, dd, *J* = 8.43, 7.69 Hz, 6b-H), 3.11 (1 H, d, *J* = 5.86 Hz, 9b-H), 3.41 (3 H, s, NMe), 6.95–7.3 (4 H, m); MS *m/z* 243 (M^+, 4.3), 215 [$(M - CO)^+$, 4.6], 175 [$(M - C_5H_8)^+$, 100]. Anal. Calcd for $C_{15}H_{17}NO_2$: C, 74.05; H, 7.04; N, 5.76. Found: C, 74.18; H, 7.08; N, 5.89.

(±)-(6aα,6bα,10aα,10bα)-6a,6b,7,8,9,10,10a,10b-Octahydro-6a-hydroxy-5-methylbenzo[3,4]cyclobuta[1,2-*c*]quinolin-6-(5*H*)-one (22). The hydrolysis of **21** (79 mg, 0.26 mmol) was carried out with K_2CO_3 (22 mg, 0.16 mmol) in 80% aqueous methanol (5 mL) for 20 h, as described above, to give **22** (61 mg, 90%): mp 142–145 °C (hexane–diethyl ether); IR 3360 (OH), 1636 (C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.8–1.1 (3 H, m), 1.25–1.6 (4 H, m), 1.85–2.0 (1 H, m), 2.6–2.8 (2 H, m, 6b-H, 10a-H), 3.41 (3 H, s, NCH_3), 3.78 (1 H, dd, *J* = 9.15, 1.10 Hz, 10b-H), 7.0–7.4 (4 H, m); MS *m/z* 257 (M^+, 1.6), 239 [$(M - OH)^+$, 2.7], 175 [$(M - C_6H_{10})^+$, 100]. Anal. Calcd for $C_{16}H_{19}NO_2$: C, 74.68; H, 7.44; N, 5.44. Found: C, 74.67; H, 7.53; N, 5.44.

1,2a,8b-*trans*,*cis*-2,2a,4,8b-Tetrahydro-2a-hydroxy-1-methoxy-1,4-dimethylcyclobuta[*c*]quinolin-3(1*H*)-one (27). The hydrolysis of **26** (232 mg, 0.803 mmol) was carried out with K_2CO_3 (67 mg, 0.49 mmol) in 80% aqueous methanol (7 mL) for 15 h, as described above, to give **27** (175 mg, 88%): mp 123–124 °C (hexane–diethyl ether); IR 3300 (OH), 1633 (C=O) cm^{-1}; ^1H NMR (400 MHz) δ 1.53 (3 H, s, 1-Me), 2.19 (1 H, dd, *J* = 12.70, 1.95 Hz, 2-H cis to 1-Me), 2.77 (1 H, d, *J* = 12.70 Hz, 2-H cis to 1-OMe), 2.94 (3 H, s, OMe), 3.40 (3 H, s, NMe), 3.55 (1 H, d, *J* = 1.95 Hz, 8b-H), 7.00 (1 H, d, *J* = 8.30 Hz, 1-H, 5-H), 7.05–7.10 (2 H, m), 7.29 (1 H, dd, *J* = 8.30, 1.46 Hz, 8-H); MS *m/z* 248 [$(M + 1)^+$, 0.23], 247 (M^+, 0.05), 230 [$(M - H_2O)^+$, 0.58], 175 [$(M - CH_2C(OMe)Me)^+$, 100]. Anal. Calcd for $C_{14}H_{19}NO_2$: C, 68.00; H, 6.93; N, 5.66. Found: C, 67.82; H, 6.95; N, 5.56.

1,2-Dihydro-1,1,5-trimethylfuro[2,3-*c*]quinolin-4(5*H*)-one (28), 1,4,5,6-Tetrahydro-6-iodo-1,5,5-trimethyl-1-benzazocine-2,3-dione (30) and 1,1a,4,8b-Tetrahydro-1,1-dimethylcyclopropa[*d*][1]benzazepine-2,3-dione (31). Cyclobutanol **9** (178 mg, 0.771 mmol) in benzene (86 mL) containing mercury(II) oxide (500 mg, 2.31 mmol) and iodine (586 mg, 2.31 mmol) in a Pyrex vessel was irradiated with a 100-W high-pressure Hg arc while being stirred for 4 h under a nitrogen atmosphere. The solution was filtered through a Celite pad, and the filtrate was washed with a 5% sodium thiosulfate solution and water. After drying over anhydrous sodium sulfate, evaporation of the solvent gave a residue which was purified by preparative TLC (1:20 ethyl acetate–dichloromethane) to afford **28** (49 mg, 28%), **30** (151 mg, 55%) and **31** (12 mg, 7%). **28**: mp 160–163 °C (hexane); IR 1669 (C=O), 1624 C=CO cm^{-1}; ^1H NMR (90 MHz) δ 1.60 (6 H, s, 1-Me), 3.77 (3 H, s, NMe), 4.39 (2 H, s, 2-H), 7.1–7.5 (3 H, m), 7.72 (1 H, d, *J* = 7.47 Hz, 9-H); MS *m/z* 229 (M^+, 86), 214 [$(M - Me)^+$, 100]. Anal. Calcd for $C_{14}H_{15}NO_2$: C, 73.34; H, 6.59; N, 6.11. Found: C, 73.17; H, 6.61; N, 6.24. **30**: mp 163–165 °C (hexane); IR 1716 (ketone C=O), 1654 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.97 (3 H, s, 5-Me), 1.36 (3 H, s, 5-Me), 2.12 (1 H, dd, *J* = 10.99, 1.10 Hz, 4-H), 3.34 (1 H, d, *J* = 10.99 Hz, 4-H cis to I), 3.39 (3 H, s, NMe), 5.14 (1 H, d, *J* = 1.10 Hz, 6-H), 7.05–7.45 (4 H, m); MS *m/z* 358 [$(M + 1)^+$ 0.7], 357 (M^+, 0.7), 230 [$(M - I)^+$, 19], 202 [$(M - I - CO)^+$, 49], 160 (100); HR-MS calcd for $C_{14}H_{16}NO_2I$ M, 357.0226, found *m/z* 357.0248. **31**: mp 111–113 °C (hexane); IR 1715 (ketone C=O), 1657 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.98 (3 H, s, 1-Me), 1.30 (3 H, s, 1-Me), 2.34 (1 H, d, *J* = 10.26 Hz, 1a-H or 8b-H), 2.38 (1 H, d, *J* = 10.26 Hz, 8b-H or 1a-H), 3.38 (3 H, s, NCH_3), 7.1–7.2 (2 H, m), 7.25–7.35 (2 H, m); MS *m/z* 229 (M^+, 6.9), 201 [$(M - CO)^+$, 100]. Anal. Calcd for $C_{14}H_{15}NO_2$: C, 73.34; H, 6.59; N, 6.11. Found: C, 73.60; H, 6.60; N, 5.90.

1,2-Dihydro-1,1,2,2,5-pentamethylfuro[2,3-*c*]quinolin-4-(5*H*)-one (29). Irradiation of cyclobutanol **10** (38 mg, 0.15 mmol) in benzene (15 mL) containing mercury(II) oxide (106 mg, 0.487 mmol) and iodine (124 mg, 0.487 mmol) for 6 h as described above gave **29** (19 mg, 50%): mp 105–108 °C (hexane); IR 1657 (C=O), 1620 (C=O-O) cm^{-1}; ^1H NMR (90 MHz) δ 1.43 (12 H, s, 1- and 2-Me), 3.77 (3 H, s, NMe), 7.1–7.4 (3 H, m), 7.73 (1 H, d, *J* = 7.47 Hz, 9-H); MS *m/z* 257 (M^+, 83), 242 [$(M - Me)^+$, 100]. Anal. Calcd for $C_{16}H_{19}NO_2$: C, 74.68; H, 7.44; N, 5.44. Found: C, 74.62; H, 7.50; N, 5.39.

***cis*-1,2,3,3a,6,10b-Hexahydro-6-methyl-6*H*-cyclopenta-[4,5]furo[2,3-*c*]quinolin-5-one (32), 3-Hydroxy-4-(*trans*-2-iodocyclopentyl)-1-methylquinolin-2(1*H*)-one (34), and**

***trans*-5,8,9,10,10a,10b-Hexahydro-5-methylcyclopenta[3,4]-cyclopropa[1,2-*d*][1]benzazepine-6,7-dione (36).** Irradiation of cyclobutanol **19** (109 mg, 0.449 mmol) in benzene (50 mL) containing mercury(II) oxide (292 mg, 1.35 mmol) and iodine (343 mg, 1.35 mmol) for 5 h, as described above, gave **32** (18 mg, 17%), **34** (13 mg, 8%), and **36** (22 mg, 20%). **32**: mp 106–108 °C (hexane); IR 1663 (C=O), 1629 (C=CO) cm^{-1}; ^1H NMR (270 MHz) δ 1.55–2.05 (5 H, m), 2.0–2.35 (1 H, m), 3.76 (3 H, s, NMe), 4.09 (1 H, ddd, J = 8.06, 5.81, 4.03 Hz, 10b-H), 5.48 (1 H, ddd, J = 8.06, 6.23, 1.10 Hz, 3a-H), 7.2–7.5 (4 H, m); MS m/z 241 (M^+, 100). Anal. Calcd for $C_{15}H_{15}NO_2$: C, 74.67; H, 6.27; N, 5.80. Found: C, 74.65; H, 6.22; N, 6.07. **34**: mp 153–155 °C (hexane–diethyl ether); IR 3270 (OH), 1628 (C=O), 1617 (C=CO) cm^{-1}; ^1H NMR (270 MHz) δ 1.7–1.85 (1 H, m), 2.0–2.25 (2 H, m), 2.55–2.65 (2 H, m), 3.05–3.35 (2 H, m), 3.84 (3 H, s, NMe), 4.97 (1 H, dt, J = 5.49, 5.13 Hz, 2′-H), 7.25–7.5 (3 H, m), 7.69 (1 H, d, J = 6.96 Hz, 5-H); MS m/z 369 (M^+, 15), 242 [$(M - I)^+$, 100]; HR-MS calcd for $C_{15}H_{16}INO_2$ M, 369.0225, found m/z 369.0234. **36**: mp 182–185 °C (hexane); IR 1687 (C=O), 1654 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 1.4–1.6 (1 H, m), 1.8–2.15 (4 H, m), 2.25 (1 H, ddd, J = 13.56, 8.80, 1.10 Hz), 2.46 (1 H, d, J = 5.49 Hz, 10b-H), 2.93 (1 H, t, J = 5 Hz, 10aH), 3.40 (3 H, s, NMe), 7.1–7.3 (4 H, m); MS m/z 241 (M^+, 37), 213 [$(M - CO)^+$, 48], 157 (56), 95 (100). Anal. Calcd for $C_{15}H_{15}NO_2$: C, 74.67; H, 6.27; N, 5.80. Found: C, 74.67; H, 6.13; N, 5.97.

Irradiation of cyclobutanol **20** (122 mg, 0.502 mmol) in benzene (57 mL) containing mercury(II) oxide (325 mg, 1.50 mmol) and iodine (381 mg, 1.50 mmol) for 5 h gave **32** (61 mg, 50%), **34** (23 mg, 12%) and **36** (2 mg, 2%).

X-ray Crystallographic Analysis of the Structure of *trans*-5,8,9,10,10a,10b-Hexahydro-5-methylcyclopenta[3,4]-cyclopropa[1,2-*d*][1]benzazepine-6,7-dione (36). The crystal data for **36** were as follows: $C_{15}H_{15}NO_2$, mol weight 241.29, monoclinic, space group $P2_1/c$, a = 9.711 (2) Å, b = 8.882 (1) Å, c = 14.482 (2) Å, β = 105.21 (2) Å, V = 1205. 4 (3) Å^3, Z = 4, D_{calcd} = 1.330 g/cm^3.

The diffraction intensities were measured on a Rigaku AF-C5 automated X-ray diffractometer with graphite-monochromated Mo-Kα radiation (λ = 0.71069 Å) using the scanning mode ω ($2\theta \leq 30°$), $2\theta - \omega$ ($2\theta > 30°$). The crystal structure was solved using MULTAN 78,[9] and refinements were carried out with an anisotropic full-matrix least-square method using XTAL.[10] The total number of reflections and observed reflections were 2276 and 1807. The final R value was 0.062.

***cis*-1,2,3,4,4a,11b-Hexahydro-7-methylcyclohexa[4,5]-furo[2,3-*c*]quinolin-6(7*H*)-one (33), 3-Hydroxy-4-(*trans*-2-iodocyclohexyl)-1-methylquinolin-2(1*H*)-one (35), *trans*-8,9,10,11,11a,11b-Hexahydro-5-methylcyclohexa[3,4]cyclopropa[1,2-*d*][1]benzazepine-6,7-dione (37), and 5,7-Dihydro-5-methylcyclohexa[*d*][1]benzazocine-6,7-dione (38).** Photolysis of cyclobutanol **22** (215 mg, 0.837 mmol) in benzene (80 mL) containing mercury(II) oxide (637 mg, 2.94 mmol) and iodine (650 mg, 2.56 mmol) for 1.5 h, as described above, gave **33** (32 mg, 15%), **35** (50 mg, 16%), **37** (17 mg, 8%) and **38** (35 mg, 16%). **33**: mp 115–116 °C (hexane); IR 1661 (C=O), 1624 cm^{-1} (C=CO); ^1H NMR (400 MHz) δ 1.2–1.35 (2 H, m), 1.55–1.9 (4 H, m), 2.1–2.2 (1 H, m), 2.4–2.5 (1 H, m), 3.37 (1 H, dt, J = 10.25, 6.84 Hz, 11a-H), 3.79 (3 H, s, NMe), 4.81 (1 H, ddd, J = 6.84, 3.91, 2.93 Hz, 7a-H), 7.2–7.5 (4 H, m); MS m/z 255 (M^+, 100). Anal. Calcd for $C_{16}H_{17}NO_2$: C, 75.27; H, 6.71; N, 5.49. Found: C, 75.09; H, 6.66; N, 5.57. **35**: mp 176–179 °C (hexane); IR 3320 (OH), 1620 (C=O, C=CO) cm^{-1}; ^1H NMR (270 MHz) δ 1.4–1.6 (1 H, m), 1.65–1.8 (1 H, m), 1.9–2.1 (4 H, m), 2.2–2.35 (1 H, m), 2.8–3.0 (2 H, m), 3.83 (3 H, s, NMe), 5.07 (1 H, br s, 2′-H), 7.25–7.5 (3 H, m), 7.64 (1 H, d, J = 8.06 Hz, 5-H); MS m/z 383 (M^+, 21), 256 [$(M - I)^+$, 100]; HR-MS calcd for $C_{16}H_{18}INO_2$ M, 383.0383, found m/z 383.0388. **37**: mp 167–169 °C (hexane–diethyl ether); IR 1688 (C=O), 1650 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 1.25–1.55 (4 H, m), 1.85–1.95 (2 H, m), 2.0–2.1 (1 H, m), 2.36 (1 H, d, J = 6.96 Hz, 11b-H), 2.45–2.55 (1 H, m), 2.86 (1 H, b t, J = 6.60 Hz), 3.40 (3 H, s), 7.1–7.3 (4 H, m); MS m/z 255 (M^+, 38), 147 (49), 109 (100). Anal. Calcd for $C_{16}H_{17}NO_2$: C, 75.27; H, 6.71; N, 5.49. Found: C, 75.27; H, 6.64; N, 5.42. **38**: mp 179–181 °C (hexane–diethyl ether); IR 1711 (C=O), 1648 (lactam C=O) cm^{-1}; ^1H NMR (270 MHz) δ 0.76 (1 H, dq, J = 3.30, 12.83 Hz), 1.22 (1 H, tq, J = 3.30, 12.83 Hz), 1.35–2.2 (5 H, m), 2.71 (1 H, dddd, J = 12.83, 6.96, 3.30, 1.47 Hz, 11-H), 3.61 (1 H, dd, J = 6.59, 4.40 Hz, 7a-H), 5.69 (1 H, s, 12-H), 7.14 (1 H, dd, J = 7.70, 1.46 Hz, 8-H), 7.2–7.35 (2 H, m), 7.80 (1 H, dd, J = 7.70, 1.46 Hz, 1-H); MS m/z 256 [$(M + 1)^+$, 61], 255 (M^+, 1.1), 228 (79), 146 (100). Anal. Calcd for $C_{16}H_{17}NO_2$: C, 75.27; H, 6.71; N, 5.49. Found: C, 75.42; H, 6.79; N, 5.45.

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Supplementary Material Available: Atomic coordinates of both non-hydrogen atoms and hydrogen atoms as well as the thermal parameters of the non-hydrogen atoms, bond lengths and angles, torsion angles, and distances of atoms from least-squares planes for compound **36** (Table I–VII) (5 pages). This material is contained in many libraries on microfiche, immediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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