Synthesis of 2-Alkylidenecyclopentanones via Palladium-Catalyzed Carbopalladation/Ring Expansion of 1-(1-Alkynyl)cyclobutanols†

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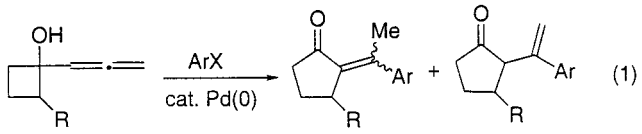
The palladium-catalyzed cross-coupling of aryl halides or vinylic halides or triflates and 1-(1-alkynyl)cyclobutanols affords good yields of stereoisomerically pure 2-arylidene- or 2-(2-alkenylidene)cyclopentanones, respectively. The process involves (1) oxidative addition of the organic halide or triflate to Pd(0), (2) regioselective, intermolecular carbopalladation of the carbon−carbon triple bond of the 1-(1-alkynyl)cyclobutanol to produce a vinylic palladium intermediate, (3) regioselective ring expansion to a palladacycle, and (4) reductive elimination of the 2-alkylidenecyclopentanone with simultaneous regeneration of the Pd(0) catalyst. Generally, the best results are obtained by employing 10 mol % of $Pd(OAc)_2$, 20 mol % of PPh_3, 2 equiv of the aryl or vinylic iodide or vinylic triflate, 2 equiv of diisopropylethylamine, and *n*-Bu_4NCl in DMF as the solvent.

Introduction

The synthesis of 2-alkylidenecyclopentanones has received considerable attention in the literature due to the fact that some of the natural products containing or derived from such alkylidenecyclopentanones show significant biological activity.[1] A number of reactions of palladium reagents and 3- or 4-membered ring alkenes and alcohols have been reported to produce ketones, particularly cycloalkanones. For example, cyclopropanols react with Pd(II) salts to produce 2-alkenones.[2] Silyloxycyclopropanes react with aryl triflates in the presence of a palladium catalyst to afford 3-arylalkanones.[3] Methylenecyclobutanes undergo palladium(II)-catalyzed ring expansion to cyclopentanones.[4] The reaction of cyclobutanols and catalytic amounts of $Pd(OAc)_2$ generates 3-alkenones.[5] The analogous palladium(0)-catalyzed reaction of cyclobutanols and aryl halides affords 4-arylalkanones.[6] Recently, 1-vinylcyclobutanols or the corresponding silyl ethers have been shown to react with palladium(II) salts to produce ring-expanded palladiocyclopentanones, which undergo either hydride elimination to unsaturated cyclopentanones[7] or further cyclization to produce unsaturated bicyclic ketones.[8] 2-(1-Alkynyl)-2-hydroxycyclobutanones undergo palladium(II)-catalyzed ring expansion to 2-alkylidene-1,3-cyclopentanediones.[9] 1-(3-Methoxycarbonyloxy-1-propynyl)cyclobutanols undergo palladium(0)-catalyzed ring expansion in the presence of phenols to produce 2-(1-aryloxyvinyl)cyclopentanones.[10] Finally, 1-allenylcyclobutanols react with aryl or vinylic halides in the presence of a palladium catalyst by a process involving carbopalladation of the allene and subsequent ring expansion of the resulting π-allylpalladium intermediate to generate unsaturated cyclopentanones (eq 1).[11]



(1)

Despite the fact that we have carried out a large number of carbopalladation reactions on 1-(1-alkynyl)-cycloalkanols bearing 5- or 6-membered rings without any involvement of the carbocyclic ring, this latter process involving allenylcyclobutanols encouraged us to examine the possibility that 1-(1-alkynyl)cyclobutanols might undergo an analogous carbopalladation/ring-expansion process to afford 2-alkylidenecyclopentanones. We now wish to report just such a process.[12]

† This paper is dedicated to Professor Herbert C. Brown, a pioneer in organometallic chemistry directed toward organic synthesis, on the occasion of his 90th birthday.

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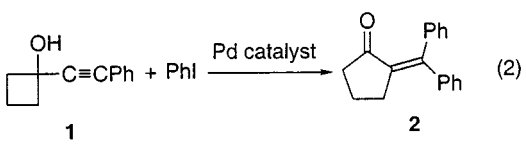
Table 1. Palladium-Catalyzed Carbopalladation/Ring Expansion of 1-(Phenylethynyl)cyclobutanol (1) and Iodobenzene (Eq 2)[a]

entry	Pd catalyst	PPh_3 (%)	base (equiv)	chloride source (equiv)	% isolated yield of **2**
1	$Pd(PPh_3)_4$		K_2CO_3 (2)		58
2	$Pd(dba)_2$		K_2CO_3 (2)		47
3	$Pd(dba)_2$	20	K_2CO_3 (2)		45
4	$Pd(dba)_2$	20	K_2CO_3 (2)	LiCl (2)	46
5	$Pd(OAc)_2$	20	K_2CO_3 (2)		40
6	$Pd(OAc)_2$	20	K_2CO_3 (2)	TBAC (2)	65
7	$Pd(OAc)_2$	20	K_2CO_3 (2)	LiCl (2)	58
8	$Pd(OAc)_2$	20	K_2CO_3 (2)	TBAC (2)	0[b]
9	$Pd(OAc)_2$	10	K_2CO_3 (2)	TBAC (2)	53[c]
10	$Pd(OAc)_2$	20	Na_2CO_3 (2)	TBAC (2)	68
11	$Pd(OAc)_2$	20	$NaHCO_3$ (2)	TBAC (2)	66
12	$Pd(OAc)_2$	20	KOAc (2)	TBAC (2)	46
13	$Pd(OAc)_2$	20	$KHCO_3$ (2)	TBAC (2)	45
14	$Pd(OAc)_2$	20	Na_2CO_3 (1)	TBAC (2)	61
15	$Pd(OAc)_2$	20	Na_2CO_3 (2)	TBAC (1)	61
16	$Pd(OAc)_2$	20	Na_2CO_3 (2)	TBAC (2)	46[d]
17	$Pd(OAc)_2$	20	Na_2CO_3 (2)	TBAC (2)	62[e]
18	$Pd(OAc)_2$	20	Na_2CO_3 (2)	TBAC (2)	57[f]
19	$Pd(OAc)_2$	20	Et_3N (4)	TBAC (2)	69
20	$Pd(PPh_3)_4$		Et_3N (4)	TBAC (2)	10
21	$Pd(OAc)_2$	20	*i*-Pr_2NEt (2)	TBAC (2)	70
22	$Pd(OAc)_2$	20	*i*-Pr_2NEt (1)	TBAC (2)	66
23	$Pd(OAc)_2$	20	*i*-Pr_2NEt (4)	TBAC (2)	58
24	$Pd(OAc)_2$	20	*i*-Pr_2NEt (2)	TBAC (1)	62
25	$Pd(OAc)_2$	20	*i*-Pr_2NEt (1)	TBAC (1)	64
26	$Pd(OAc)_2$	10	*i*-Pr_2NEt (2)	TBAC (2)	54[g]
27	$Pd(PPh_3)_4$		Na_2CO_3 (2)	TBAC (2)	75[h]
28	$Pd(PPh_3)_4$		Na_2CO_3 (2)	TBAC (3)	58[h]
29	$Pd(PPh_3)_4$		*i*-Pr_2NEt (2)	TBAC (2)	0[h]
30	$Pd(PPh_3)_4$		Na_2CO_3 (1)	TBAC (2)	0[h]
31	$Pd(PPh_3)_4$		Na_2CO_3 (2)	TBAC (1)	0[h]

[a] Unless otherwise stated, all reactions were carried out under an argon atmosphere using 1 equiv of **1** (0.5 mmol), 2 equiv of **2** (1.0 mmol), 10 mol % of the Pd catalyst (0.05 mmol), 20 mol % of PPh_3 (0.1 mmol), 2 equiv of *n*-Bu_4NCl (TBAC, 1.0 mmol), 2 equiv of base (1.0 mmol), and DMF (5 mL) at 80 °C for 12 h. [b] The reaction was carried out at 70 °C. [c] The reaction was carried out using 5 mol % of $Pd(OAc)_2$ and 10 mol % of PPh_3 at 80 °C for 12 h. [d] The reaction was carried out using 1.2 equiv of **2**. [e] The reaction was carried out using 10 mL of DMF. [f] The reaction was carried out using 20 mol % of (*o*-$CH_3C_6H_4)_3P$. [g] The reaction was carried out using 5 mol % of $Pd(OAc)_2$ and 10 mol % PPh_3. [h] The reaction was carried out at 80 °C for 24 h.

Results and Discussion

1-(1-Alkynyl)cyclobutanols are easily prepared by metal acetylide addition to the appropriate cyclobutanones, which are in turn readily available by ketene cycloaddition to olefins.[13] With a wide variety of 1-(1-alkynyl)-cyclobutanols readily available, we focused our initial studies on the reaction of 1-(phenylethynyl)cyclobutanol (**1**) and iodobenzene as our model system (eq 2).



1 **2** (2)

The reaction of alkynol **1** with 2 equiv of iodobenzene in the presence of 10 mol % of $Pd(PPh_3)_4$ and 2 equiv of K_2CO_3 as a base in DMF as the solvent at 80 °C resulted in the formation of arylidenecyclopentanone **2** in 58% yield after 12 h (Table 1, entry 1). In an effort to optimize the yield of this process, we have looked at the effect of several different palladium catalysts [$Pd(PPh_3)_4$, $Pd(OAc)_2$, and $Pd(dba)_2$], various inorganic and organic bases, two different chloride sources [LiCl and *n*-Bu_4NCl (TBAC)], and the ligand PPh_3. The results are summarized in Table 1. This investigation led to the following standard reaction procedure: 10 mol % of $Pd(OAc)_2$, 20 mol % of PPh_3, 2 equiv of aryl or vinylic iodide or triflate, 2 equiv of diisopropylethylamine as the base, and 2 equiv of *n*-Bu_4NCl in DMF as the solvent at 80 °C. This procedure afforded a 70% yield of cyclopentanone **2** after 12 h reaction time (Table 1, entry 21). Only much later after encountering difficulties with some synthetic examples did we go back and examine additional variables. It was then observed that a slightly higher yield of cyclopentanone **2** could be obtained by using 10 mol % of $Pd(PPh_3)_4$ and 2 equiv of Na_2CO_3 and *n*-Bu_4NCl (Table 1, entry 27), but this procedure did not provide as high yields on a number of other starting materials as our original procedure above and often afforded products arising by phenyl transfer from the PPh_3 and has thus not been widely employed.

With our standard procedure in hand, we next set out to explore the scope and limitations of this novel route to 2-alkylidenecyclopentanones by examining a wide variety of 1-(1-alkynyl)cyclobutanols and aryl or vinylic iodides or triflates as shown in Table 2. The reaction of 1-(phenylethynyl)cyclobutanol (**1**) and electron-rich methoxy-containing aromatic iodides provided the corresponding 2-arylidenecyclopentanones as single stereoisomers in good yields (Table 2, entries 2 and 3). However, the reaction of 1-(phenylethynyl)cyclobutanol (**1**) and 4-iodoanisole under our standard conditions also produced a side product 2-(diphenylmethylene)cyclopentanone (**2**) in 11% yield (Table 2, entry 3). The phenyl group must be coming from the ligand PPh_3 added to this reaction. This type of aryl exchange between arylpalladium intermediates and triarylphosphines is well documented.[14] Steric hindrance appears to be no problem here, since the more hindered ortho-substituted system actually gave a higher yield. Even *o*-iodoaniline reacted well, although it usually undergoes oxidative addition only slowly and requires an elevated temperature (Table 2, entry 4). Under the usual reaction conditions, the cyclopentanone **5** could be isolated in good yield, but this product proved unstable. When the reaction was run at 150 °C for 12 h, the product was the interesting tricyclic pyridine derivative **6**, apparently formed by olefin inversion in the initially formed cyclopentanone and intramolecular condensation of the amino group and the ketone (Table 2, entry 5). A high yield was also obtained from the electron-poor *o*-iodonitrobenzene (Table 2, entry 6). Under our standard reaction conditions for aryl halides, phenyl triflate failed to produce any of the expected cyclopentanone product, although the starting alkyne was observed to disappear during the reaction.

We were also curious as to whether this process could be applied to vinylic halides. The reaction of 1-(phenylethynyl)cyclobutanol (**1**) and (*E*)-1-iodo-1-hexene (2 equiv) under our standard conditions afforded a single isomeric product **8** in 40% yield (Table 2, entry 7). The product was characterized by 1D and 2D NMR HMQC, COSY, and NOESY spectroscopy. The 2D NOESY spectrum of 2-[(2*E*)-1-phenylhept-2-enylidene]cyclopentanone (**8**) clearly

(12) For a previous communication, see: Larock, R. C.; Reddy, C. K. *Org. Lett.* **2000**, *2*, 3325.

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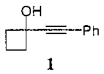
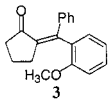
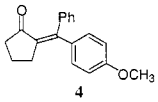
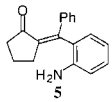
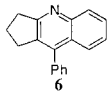
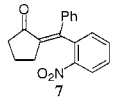
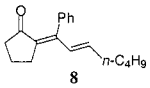
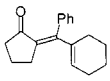
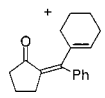
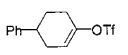
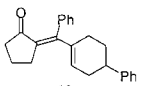
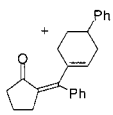
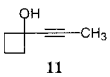
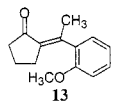
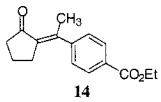
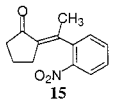
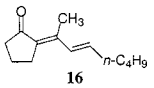
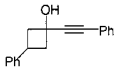
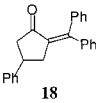
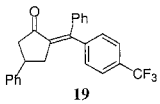
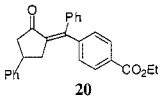
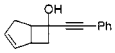
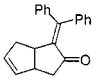
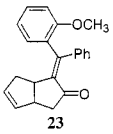
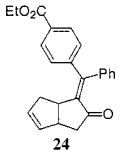
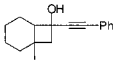
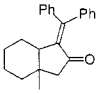
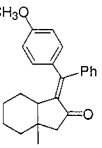
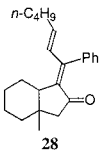
Table 2. Palladium-Catalyzed Carbopalladation/Ring Expansion of 1-(1-Alkynyl)cyclobutanols[a]

entry	1-(1-alkynyl)-cyclobutanol	organic iodide or triflate	product(s)	% isolated yield
1	 **1**	C_6H_5I	 **2**	70
2		*o*-$CH_3OC_6H_4I$	 **3**	74
3		*p*-$CH_3OC_6H_4I$	 **4**	60[b]
4		*o*-$H_2NC_6H_4I$	 **5**	75
5			 **6**	58[c]
6		*o*-$NO_2C_6H_4I$	 **7**	70
7		*E*-*n*-C_4H_9CH=CHI	 **8**	40
8			 **9a**	34
			 **9b**	23
9			 **10a**	36
			 **10b**	35
10	 **11**	PhI	 **12**	60
11		*o*-$CH_3OC_6H_4I$	 **13**	63
12		*p*-$IC_6H_4CO_2Et$	 **14**	33

entry	1-(1-alkynyl)-cyclobutanol	organic iodide or triflate	product(s)	% isolated yield
13		*o*-$NO_2C_6H_4I$	 **15**	47
14		*E*-*n*-C_4H_9CH=CHI	 **16**	35
15	 **17**	PhI	 **18**	71
16		*p*-$CF_3C_6H_4I$	 **19**	54
17		*p*-$IC_6H_4CO_2Et$	 **20**	64
18	 **21**	PhI	 **22**	50[d]
19		*o*-$CH_3OC_6H_4I$	 **23**	52[d]
20		*p*-$IC_6H_4CO_2Et$	 **24**	42[d]
21	 **25**	PhI	 **26**	60
22		*p*-$CH_3OC_6H_4I$	 **27**	44[e]
23		*E*-*n*-C_4H_9CH=CHI	 **28**	43

[a] Unless otherwise stated, all reactions were carried out under an argon atmosphere using 1 equiv of cyclobutanol (0.5 mmol), 2 equiv of organic halide or triflate (1.0 mmol), 10 mol % $Pd(OAc)_2$ (0.05 mmol), 20 mol % of PPh_3 (0.1 mmol), 2 equiv of *n*-Bu_4NCl (TBAC, 1.0 mmol), 2 equiv of *i*-Pr_2NEt (1.0 mmol), and DMF (5 mL) at 80 °C for 12 h. [b] An 11% yield of 2-(diphenylmethylene)cyclopentanone (**2**) was also observed. [c] The reaction was carried out at 150 °C for 12 h. [d] These reactions were carried out for 6 h at 80 °C. [e] A 10% yield of bicyclononanone **26** was also observed.

shows a cross-peak between the protons of the cyclopentanone allylic methylene and the vinylic proton and no interaction between the protons of the phenyl group and the cyclopentanone allylic methylene protons. This is only

consistent with the carbonyl group and the new vinylic moiety being trans to each other.

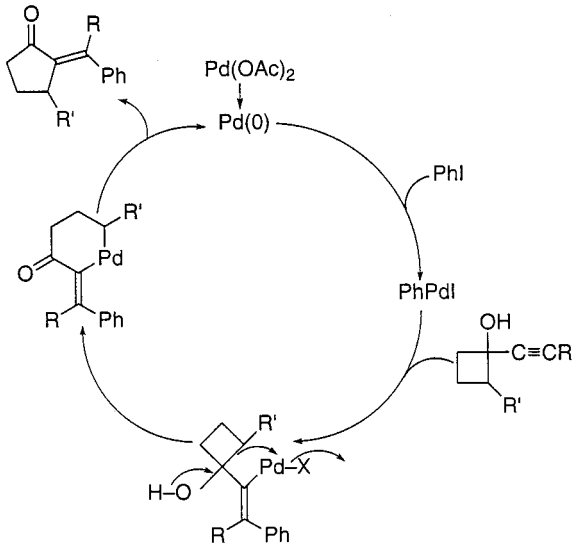
In view of our failure to obtain any cyclopentanone product from phenyl triflate, it was rather surprising that we obtained a decent yield of the anticipated cyclopentanone **9a** when cyclohex-1-enyl triflate was employed as a starting material (Table 2, entry 8). This product was accompanied by substantial amounts of the stereoisomeric dienone **9b**. Unfortunately, neither of these compounds was stable enough for full characterization. It appears that the increased steric bulk of the cyclohexenyl group leads to the low stereoselectivity of this reaction and probably to the instability of the products. 4-Phenylcyclohex-1-enyl triflate afforded a good yield of essentially equal amounts of two stereoisomeric dienones **10a** and **10b**, which also proved to be relatively unstable (Table 2, entry 9). The stereochemistry of the products in entries 8 and 9 of Table 2 is based on the assumption that the vinylic proton trans to the carbonyl should appear further downfield in the ^{1}H NMR spectrum than the corresponding cis proton.

The reaction of 1-(prop-1-ynyl)cyclobutanol (**11**) and iodobenzene under our standard conditions again gave a single isomeric product **12** (Table 2, entry 10). The product was characterized by 1D and 2D NMR HMQC, COSY, and NOESY spectroscopy. The 2D NOESY spectrum of 2-(1-phenylethylidene)cyclopentanone (**12**) (entry 10) clearly shows a cross-peak between the protons of the phenyl group and the allylic methylene protons of the cyclopentanone and no interaction between the methyl protons and the allylic methylene protons of the cyclopentanone. This is consistent with the structure **12** with the phenyl group trans to the carbonyl. Further examples of the reaction of 1-(prop-1-ynyl)cyclobutanol (**11**) and various aryl iodides or one vinylic iodide produced analogous ring expanded products in slightly lower yields than those obtained with 1-(phenylethynyl)cyclobutanol (**1**) (Table 2, entries 11−14). In all cases, a single stereoisomeric product was obtained. Good results were also obtained when reactions between 3-phenyl-1-(phenylethynyl)cyclobutanol (**17**) and several aryl iodides were carried out (Table 2, entries 15−17).

To gain better mechanistic insight into this interesting process, we next carried out a reaction between 6-(phenylethynyl)bicyclo[3.2.0]hept-2-en-6-ol (**21**), an unsymmetrical cyclobutanol, and iodobenzene under our standard reaction conditions. This reaction gave a single regioisomeric product **22** in good yield (Table 2, entry 18). The product was characterized by 1D and 2D NMR HMQC, COSY, and NOESY spectroscopy. The 2D NOESY spectrum of the product 2-(diphenylmethylene)bicyclo[3.3.0]-oct-6-en-3-one (**22**) clearly shows a cross-peak between the protons of the phenyl group and the bicyclic bridgehead proton. This result is consistent with a mechanism in which an electron-deficient intermediate is being generated and the more substituted, electron-rich carbon of the cyclobutanol undergoes subsequent migration. Single stereoisomeric products were also obtained in decent yields from the reactions of *o*-iodoanisole (Table 2, entry 19) and ethyl *p*-iodobenzoate (Table 2, entry 20) and cyclobutanol **21**.

Several reactions have also been successfully carried out with 1-methyl-7-(phenylethynyl)bicyclo[4.2.0]octan-7-ol (**25**) (Table 2, entries 21−23). The reaction of iodobenzene gave a good yield of the bicyclononanone **26** in which the more substituted neighboring carbon of the cyclobutanol has undergone exclusive migration. The reaction of bicyclobutanol **25** and 4-iodoanisole under our standard conditions gave a single stereo- and regioisomer **27** (Table 2, entry 22). Once again, the more highly substituted carbon of the cyclobutanol has undergone migration. In this reaction, we once again observed another minor product, 7-diphenylmethylene-1-methylbicyclo[4.3.0]nonan-8-one (**26**), in 10% yield. The new phenyl group in the product again apparently comes from the PPh_3 ligand.

Scheme 1



Based on the above results and previous work,[10,11] we believe that this novel ring-expansion process proceeds as shown in Scheme 1 by a sequence involving (1) reduction of $Pd(OAc)_2$ to the actual Pd(0) catalyst, (2) oxidative addition of the aryl or vinylic iodide or triflate to Pd(0), (3) vinylic or arylpalladium coordination to the carbon−carbon triple bond of the alkynylcyclobutanol and subsequent regio- and stereoselective insertion of the alkynylcyclobutanol to form a vinylpalladium intermediate, (4) release of the cyclobutane ring strain by migration of the more highly substituted, electron-rich carbon of the cyclobutanol to palladium to produce a palladacyclohexanone, and (5) reductive elimination to afford a single stereoisomeric 2-alkylidenecyclopentanone with simultaneous regeneration of the Pd(0) catalyst. Ring expansion to a palladacycle nicely explains the fact that the aryl or vinylic group arising from the iodide or triflate always ends up trans to the carbonyl carbon. Were the cyclobutanol carbon to migrate so as to effect a direct backside displacement of the vinylic palladium, one would expect the opposite stereochemistry in the final product.

In conclusion, a variety of highly substituted 2-alkylidenecyclopentanones have been prepared by the reaction of aryl or vinylic iodides or triflates and 1-(1-alkynyl)-cyclobutanols in the presence of a palladium catalyst. These products are formed regio- and stereoselectively in moderate yields. The process appears to involve a novel ring expansion to a palladacyclohexanone and subsequent reductive elimination. It would be very hard to prepare such products by any other present methodology.

Experimental Section

General Methods. ^{1}H and ^{13}C NMR spectra were recorded at 300 or 400 and 75.5 or 100 MHz, respectively. Thin-layer

chromatography (TLC) was performed using commercially prepared 60-mesh silica gel plates (Whatman K6F), and visualization was effected with short wavelength UV light (254 nm) and basic $KMnO_4$ solution [3 g of $KMnO_4$ + 20 g of K_2CO_3 + 5 mL of NaOH (5%) + 300 mL of H_2O]. All melting points are uncorrected. High-resolution mass spectra were recorded on a Kratos MS50TC double-focusing magnetic sector mass spectrometer using EI at 70 eV. Elemental analyses were performed at Iowa State University on a Perkin-Elmer 2400 CHNS/O Series II analyzer. Reaction products were purified by flash column chromatography with 40−63 μm SiO_2 (Merck).

Reagents. All reagents were used directly as obtained commercially unless otherwise noted. Anhydrous forms of Na_2CO_3, K_2CO_3, NaOAc, $NaHCO_3$, LiCl, DMF, THF, diethyl ether, and hexanes were purchased from Fisher Scientific. All palladium salts and PPh_3 were donated by Kawaken Fine Chemicals Co., Ltd. Cyclobutanone, phenylacetylene, dicyclopentadiene, 1-methylcyclohexene, styrene, zinc dust, 1-propynylmagnesium bromide, Et_3N, and *i*-Pr_2NEt were purchased from Aldrich Chemical Co., Inc. *n*-Bu_4NCl was purchased from Lancaster Synthesis, Inc.

Representative Synthesis of a 1-(1-Alkynyl)cyclobutanol: 1-(Phenylethynyl)cyclobutanol (1). A three-neck flask with thermometer inlet was charged with diisopropylamine (1.212 g, 12 mmol) in THF (5 mL). The flask was cooled to −78 °C, and *n*-BuLi (12 mmol) in hexanes (2.10 mol L^{-1}) was added dropwise under an argon atmosphere. The reaction mixture was allowed to warm to room temperature for 20 min and then cooled to −78 °C, and phenylacetylene (1.224 g, 12 mmol) in THF (2 mL) was added. The flask was allowed to warm to 0 °C for 20 min and then stirred for 1 h. The flask was again cooled to −78 °C, and cyclobutanone (0.700 g, 10 mmol) in THF (4 mL) was added. The flask was allowed to warm to room temperature for 30 min, further stirred for 2 h, quenched with water, and extracted with ether (3 × 25 mL), and the combined extracts were washed with saturated aqueous NH_4Cl, water, and brine, dried (Na_2SO_4), and filtered. The solvent was removed under reduced pressure, and the resulting oil was purified by flash column chromatography using 60:40 hexanes/diethyl ether to afford 1.566 g (91%) of the desired compound as a white solid: mp 42 °C; ^{1}H NMR ($CDCl_3$) δ 1.82−1.93 (m, 2H), 2.10 (br s, 1H), 2.23−2.40 (m, 2H), 2.50−2.58 (m, 2H), 7.26−7.34 (m, 3H), 7.42−7.46 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 12.3, 38.6, 68.2, 83.4, 92.6, 122.7, 128.2, 131.7 (one carbon missing as a result of overlap). The syntheses of cyclobutanols **11**, **17**, **21**, and **25** are reported in the Supporting Information.

General Procedure for the Palladium-Catalyzed Carbopalladation/Ring Expansion of 1-(1-Alkynyl)cyclobutanols by Organic Halides. DMF (5 mL), $Pd(OAc)_2$ (11 mg, 0.05 mmol), PPh_3 (26 mg, 0.1 mmol), the organic iodide or triflate (1.0 mmol), *i*-Pr_2NEt (130 mg, 1.0 mmol), *n*-Bu_4NCl (277 mg, 1.0 mmol), and the 1-(1-alkynyl)cyclobutanol (0.5 mmol) were placed in a 4 dram vial. The vial was flushed with Ar and heated in an oil bath at 80 °C for the indicated period of time. The reaction was monitored by TLC to establish completion. The reaction mixture was cooled, diluted with 30 mL of ether, washed with 40 mL of saturated NaCl, dried (Na_2SO_4), and filtered. The solvent was evaporated under reduced pressure, and the product was isolated by chromatography on a silica gel column.

Compounds Prepared. 2-(Diphenylmethylene)cyclopentanone (2). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 87 mg (70%) of the indicated compound as a yellow solid: mp 105 °C; ^{1}H NMR ($CDCl_3$) δ 1.93 (m, 2H), 2.38 (t, J = 7.8 Hz, 2H), 2.82 (t, J = 7.0 Hz, 2H), 7.10−7.14 (m, 2H), 7.17−7.20 (m, 2H), 7.30−7.35 (m, 6H); ^{13}C NMR ($CDCl_3$) δ 20.5, 32.9, 39.8, 127.8, 127.9, 128.3, 129.4, 129.6, 134.3, 140.1, 141.8, 148.2, 206.5 (one carbon missing as a result of overlap); IR ($CHCl_3$, cm^{-1}) 1706; MS *m*/*z* (rel intensity) 248 (50, M^+), 247 (100), 191 (30). Anal. Calcd for $C_{18}H_{16}O$: C, 87.06; H, 6.49. Found: C, 86.90; H, 6.80.

(*E*)-2-[(2-Methoxyphenyl)phenylmethylene]cyclopentanone (3). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 103 mg (74%) of the indicated compound as a yellow solid: mp 110 °C; ^{1}H NMR ($CDCl_3$) δ 1.89−1.99 (m, 2H), 2.37 (t, J = 7.8 Hz, 2H), 2.87 (t, J = 6.9 Hz, 2H), 3.82 (s, 3H), 6.85−6.88 (m, 2H), 7.11−7.17 (m, 4H), 7.32−7.35 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 20.6, 33.2, 39.7, 55.2, 113.2, 127.6, 127.7, 129.4, 131.4, 133.2, 134.0, 140.5, 148.0, 159.7, 206.4; IR ($CHCl_3$, cm^{-1}) 1712; HRMS calcd for $C_{19}H_{17}O_2$ 277.1229, found 277.1232.

(*E*)-2-[(4-Methoxyphenyl)phenylmethylene]cyclopentanone (4). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 84 mg (60%) of the indicated compound as a yellow solid: mp 110 °C; ^{1}H NMR ($CDCl_3$) δ 1.85−1.95 (m, 2H), 2.40 (t, J = 7.8 Hz, 2H), 2.54 (t, J = 7.2 Hz, 2H), 3.74 (s, 3H), 6.91−6.96 (m, 2H), 7.04−7.07 (m, 1H), 7.21−7.33 (m, 6H); ^{13}C NMR ($CDCl_3$) δ 19.6, 31.8, 40.3, 55.4, 111.3, 120.4, 127.3, 127.5, 128.9, 129.2, 129.8, 131.2, 135.0, 139.6, 145.2, 156.0, 205.9; IR ($CHCl_3$, cm^{-1}) 1712; MS *m*/*z* (rel intensity) 278 (50, M^+), 247 (100). Anal. Calcd for $C_{19}H_{18}O_2$: C, 81.99; H, 6.52. Found: C, 81.47; H, 6.75.

(*E*)-[(2-Aminophenyl)phenylmethylene]cyclopentanone (5). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 76 mg (75%) of the indicated compound as a yellow solid: ^{1}H NMR ($CDCl_3$) δ 1.88−1.98 (m, 2H), 2.43 (t, J = 7.8 Hz, 2H), 2.64 (t, J = 6.9 Hz, 2H), 3.68 (br s, 2H), 6.70 (dd, J = 1.2, 8.1 Hz, 1H), 6.74 (dt, J = 1.2, 7.2 Hz, 1H), 6.96 (dd, J = 1.5, 7.5 Hz, 1H), 7.13 (ddd, J = 1.8, 7.5, 8.1 Hz, 1H), 7.26−7.32 (m, 5H); ^{13}C NMR ($CDCl_3$) δ 19.7, 32.2, 40.3, 115.9, 117.9, 127.4, 127.7, 128.3, 129.1, 129.3, 129.6, 135.4, 138.3, 143.3, 145.8, 206.1. This compound was too unstable to get further characterization.

9-Phenyl-1,2,3-trihydropenta[1,2-*b*]quinoline (6). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 71 mg (58%) of the indicated compound as a yellow solid: mp 128−130 °C; ^{1}H NMR ($CDCl_3$) δ 2.13−2.20 (m, 2H), 2.89 (t, J = 7.3 Hz, 2H), 3.23 (t, J = 7.6 Hz, 2H), 7.33−7.39 (m, 3H), 7.45−7.54 (m, 3H), 7.59−7.64 (m, 2H), 8.05−8.09 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 23.5, 30.3, 35.2, 125.5, 125.6, 126.2, 127.9, 128.2, 128.5, 128.8, 129.3, 133.6, 136.7, 142.7, 147.9, 167.4; IR ($CHCl_3$, cm^{-1}) 3027, 2976; MS *m*/*z* (rel intensity) 246 (20, M + 1), 245 (100). Anal. Calcd for $C_{18}H_{15}N$: C, 88.13; H, 6.16; N, 5.71. Found: C, 87.90; H, 6.23; N, 5.55.

(*E*)-2-[(2-Nitrophenyl)phenylmethylene]cyclopentanone (7). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 103 mg (70%) of the indicated compound as a yellow viscous oil: ^{1}H NMR ($CDCl_3$) δ 1.88−1.97 (m, 2H), 2.38−2.51 (m, 4H), 7.24−7.29 (m, 5H), 7.39 (dd, J = 1.5, 7.5 Hz, 1H), 7.49 (dt, J = 1.5, 7.5 Hz, 1H), 7.66 (dt, J = 1.2, 7.5 Hz, 1H), 8.00 (dd, J = 1.5, 8.4 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ 19.5, 31.8, 40.1, 124.8, 127.5, 128.4, 128.8, 129.5, 130.7, 133.4, 134.6, 136.5, 136.9, 144.4, 147.5, 205.0; IR ($CHCl_3$, cm^{-1}) 1716, 1524, 1348; HRMS calcd for $C_{18}H_{14}NO_3$ 292.0974, found 292.0968.

(*E*)-2-[(2*E*)-1-Phenylhept-2-enylidene]cyclopentanone (8). The reaction mixture was chromatographed using 70:30 hexanes/ether to afford 50 mg (40%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 0.88 (t, J = 6.9 Hz, 3H), 1.27−1.37 (m, 4H), 1.95−2.03 (m, 2H), 2.17 (q, J = 6.6 Hz, 2H), 2.29 (t, J = 7.8 Hz, 2H), 2.88 (t, J = 7.2 Hz, 2H), 5.55−5.65 (m, 1H), 6.47 (d, J = 15.3 Hz, 1H), 7.02−7.05 (m, 2H), 7.31−7.39 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 13.8, 19.4, 22.2, 29.2, 30.9, 33.3, 40.2, 126.9, 127.6, 128.6, 130.8, 131.1, 138.0, 144.9, 206.0; IR ($CHCl_3$, cm^{-1}) 1708; HRMS calcd for $C_{18}H_{22}O$ 254.1671, found 254.1670.

(*E*)-[(Cyclohex-1-enyl)phenylmethylene]cyclopentanone (9a). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 43 mg (34%) of the indicated compound as a yellow viscous oil: ^{1}H NMR ($CDCl_3$) δ 1.57−1.61 (m, 4H), 1.84−1.97 (m, 4H), 2.16−2.18 (m, 2H), 2.30 (t, J = 7.8 Hz, 2H), 2.79 (t, J = 6.9 Hz, 2H), 5.78−5.81 (m, 1H), 7.11−7.15 (m, 2H), 7.27−7.34 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 20.5, 21.9, 22.7, 25.5, 27.4, 32.4, 40.0, 127.5, 127.7, 128.5, 129.3, 131.7, 138.9, 139.2, 151.3, 206.7. This compound was too unstable for further characterization.

(*Z*)-[(Cyclohex-1-enyl)phenylmethylene]cyclopentanone (9b). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 29 mg (23%) of the

indicated compound as a yellow viscous oil: ^{1}H NMR ($CDCl_3$) δ 1.63−1.70 (m, 4H), 1.81−1.89 (m, 2H), 1.98−2.00 (m, 2H), 2.10−2.13 (m, 2H), 2.35 (t, J = 7.5 Hz, 2H), 2.65 (t, J = 6.9 Hz, 2H), 5.46−5.50 (m, 1H), 7.29−7.37 (m, 5H). This compound was too unstable for further characterization.

(*E*)-[Phenyl(4-phenylcyclohex-1-enyl)methylene]cyclopentanone (10a). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 60 mg (36%) of the indicated compound as a yellow viscous oil: ^{1}H NMR ($CDCl_3$) δ 1.75−1.85 (m, 1H), 1.93−2.11 (m, 5H), 2.33−2.43 (m, 3H), 2.50−2.56 (m, 1H), 2.79−2.90 (m, 3H), 5.95 (s, 1H), 7.19−7.41 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 20.4, 27.9, 29.8, 32.3, 33.5, 39.5, 39.9, 126.1, 126.7, 127.5, 127.7, 128.3, 128.4, 128.6, 131.9, 138.7, 139.0, 146.4, 150.6, 206.5. This compound was too unstable for further characterization.

(*Z*)-[Phenyl(4-phenylcyclohex-1-enyl)methylene]cyclopentanone (10b). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 57.4 mg (35%) of the indicated compound as a yellow viscous oil: ^{1}H NMR ($CDCl_3$) δ 1.83−2.13 (m, 5H), 2.25−2.48 (m, 5H), 2.71 (t, J = 6.9 Hz, 2H), 2.90−3.00 (m, 1H), 5.61−5.62 (m, 1H), 7.19−7.41 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 20.7, 29.1, 30.0, 32.2, 33.8, 39.5, 39.7, 125.7, 125.9, 126.9, 128.0, 128.3, 128.7, 133.3, 138.1, 140.4, 145.0, 147.2, 150.7, 207.2. This compound was too unstable for further characterization.

(*E*)-2-(1-Phenylethylidene)cyclopentanone (12). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 56 mg (60%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 1.74−1.83 (m, 2H), 2.37 (t, J = 7.5 Hz, 2H), 2.52−2.60 (m, 5H), 7.22−7.39 (m, 5H); ^{13}C NMR ($CDCl_3$) δ 19.8, 20.3, 31.7, 40.5, 127.2, 127.8, 128.1, 132.6, 143.5, 147.4, 208.8; IR ($CHCl_3$, cm^{-1}) 1703; HRMS calcd for $C_{13}H_{14}O$ 186.1045, found 186.1039.

(*E*)-2-[(2-Methoxyphenyl)ethylidene]cyclopentanone (13). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 63 mg (63%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 1.73−1.83 (m, 2H), 2.34−2.45 (m, 7H), 3.79 (s, 3H), 6.89−7.04 (m, 3H), 7.24−7.29 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 19.8, 19.9, 30.8, 40.7, 55.4, 111.0, 120.5, 128.1, 128.8, 132.4, 133.3, 145.8, 155.4, 208.7; IR ($CHCl_3$, cm^{-1}) 1701; HRMS calcd for $C_{14}H_{16}O_2$ 216.1150, found 216.1145.

(*E*)-Ethyl 4-[1-(2-oxocyclopentylidene)ethyl]benzoate (14). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 42 mg (33%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 1.39 (t, J = 7.0 Hz, 3H), 1.76−1.85 (m, 2H), 2.39 (t, J = 7.8 Hz, 2H), 2.51−2.56 (m, 5H), 4.39 (q, J = 7.2 Hz, 2H), 7.28−7.32 (m, 2H), 8.02−8.06 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 14.4, 19.7, 20.3, 31.5, 40.5, 61.1, 127.2, 129.6, 129.8, 133.3, 146.2, 148.0, 166.2, 208.7 (one carbon missing as a result of overlap); IR ($CHCl_3$, cm^{-1}) 1717, 1606; HRMS calcd for $C_{16}H_{18}O_3$ 258.1256, found 258.1252.

(*E*)-2-[1-(2-Nitrophenyl)ethylidene]cyclopentanone (15). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 54 mg (47%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 1.73−1.83 (m, 2H), 2.20−2.24 (m, 2H), 2.36 (t, J = 7.8 Hz, 2H), 2.46 (s, 3H), 7.22 (dd, J = 1.5, 7.8 Hz, 1H), 7.48 (dt, J = 1.5, 7.5 Hz, 1H), 7.65 (dt, J = 1.2, 7.5 Hz, 1H), 8.07 (dd, J = 0.9, 8.4 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ 19.6, 19.8, 30.4, 40.5, 124.7, 128.4, 129.1, 132.4, 133.9, 138.9, 144.7, 146.4, 207.8; IR ($CHCl_3$, cm^{-1}) 1709, 1633, 1606, 1526, 1347; HRMS calcd for $C_{13}H_{13}NO_3$ 231.0895, found 231.0892.

(*E*)-2-[(2*E*)-1-Methylhept-2-enylidene]cyclopentanone (16). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 34 mg (35%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 0.91 (t, J = 6.6 Hz, 3H), 1.29−1.45 (m, 4H), 1.85−1.92 (m, 2H), 2.18−2.36 (m, 7H), 2.72 (t, J = 6.3 Hz, 2H), 6.21−6.25 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 13.1, 13.9, 19.4, 22.3, 29.3, 31.3, 33.4, 40.8, 131.0, 131.1, 139.9, 142.7, 208.9; IR ($CHCl_3$, cm^{-1}) 1692, 1585; HRMS calcd for $C_{13}H_{20}O$ 192.1514, found 192.1513.

2-Diphenylmethylene-4-phenylcyclopentanone (18). The reaction mixture was chromatographed using 80:20 hexanes/ether to afford 115 mg (71%) of the indicated compound as a yellow solid: mp 119−120 °C; ^{1}H NMR ($CDCl_3$) δ 2.68 (dd, J = 10.8, 18.0 Hz, 1H), 2.89 (ddd, J = 1.8, 7.5, 18.0 Hz, 1H), 3.05 (dd, J = 10.2, 16.0 Hz, 1H), 3.25 (ddd, J = 1.5, 6.9, 16.0 Hz, 1H), 3.42−3.54 (m, 1H), 7.23−7.43 (m, 15H); ^{13}C NMR ($CDCl_3$) δ 39.5, 41.1, 46.9, 126.6, 126.7, 127.8, 127.9, 128.0, 128.5, 128.6, 129.4, 129.5, 133.9, 139.9, 141.5, 143.1, 148.8, 204.5; IR ($CHCl_3$, cm^{-1}) 1709; HRMS calcd for $C_{24}H_{20}O$ 324.1514, found 324.1511.

(*E*)-4-Phenyl-2-[phenyl[4-(trifluoromethyl)phenyl]methylene]cyclopentanone (19). The reaction mixture was chromatographed using 80:20 hexanes/ether to afford 106 mg (54%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 2.68 (dd, J = 10.8, 18.3 Hz, 1H), 2.85−3.02 (m, 2H), 3.17 (ddd, J = 1.8, 6.9, 16.2 Hz, 1H), 3.42−3.54 (m, 1H), 7.19−7.42 (m, 12H), 7.61−7.64 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 39.4, 40.9, 46.9, 122.1, 125.0, 125.1, 125.1, 125.2, 125.7, 126.7, 126.8, 128.0, 128.3, 128.7, 129.3, 129.7, 130.0, 130.4, 135.1, 139.1, 142.8, 145.1, 147.1, 204.3; IR ($CHCl_3$, cm^{-1}) 1710; HRMS calcd for $C_{25}H_{19}F_3O$ 392.1388, found 392.1383.

(*E*)-Ethyl 4-[(2-Oxo-4-phenylcyclopentylidene)phenylmethyl]benzoate (20). The reaction mixture was chromatographed using 80:20 hexanes/ether to afford 126 mg (64%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 1.40 (t, J = 7.0 Hz, 3H), 2.65 (dd, J = 10.8, 18.0 Hz, 1H), 2.83−3.02 (m, 2H), 3.19 (dd, J = 6.9, 15.9 Hz, 1H), 3.40−3.52 (m, 1H), 4.41 (q, J = 7.0 Hz, 2H), 7.17−7.39 (m, 12H), 8.05 (d, J = 8.4 Hz, 2H); ^{13}C NMR ($CDCl_3$) δ 14.2, 39.2, 40.9, 46.8, 61.0, 126.6, 126.7, 127.9, 128.2, 128.6, 129.2, 129.3, 129.4, 130.2, 134.8, 139.1, 142.8, 145.9, 147.6, 165.9, 204.3 (two carbons missing as a result of overlap); IR ($CHCl_3$, cm^{-1}) 1710; HRMS calcd for $C_{27}H_{24}O_3$ 396.1726, found 396.1720.

2-(Diphenylmethylene)bicyclo[3.3.0]oct-6-en-3-one (22). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 70 mg (50%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 2.20−2.33 (m, 2H), 2.47−2.57 (m, 1H), 2.70 (dd, J = 9.9, 18.6 Hz, 1H), 3.30−3.37 (m, 1H), 3.79−3.87 (m, 1H), 5.66−5.73 (m, 2H), 7.12−7.36 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 41.4, 42.4, 44.0, 44.4, 127.7, 127.8, 128.0, 128.2, 128.9, 129.2, 130.8, 133.9, 139.4, 140.6, 141.9, 150.1, 206.7; IR ($CHCl_3$, cm^{-1}) 1706; HRMS calcd for $C_{21}H_{18}O$ 286.1358, found 286.1352.

(*E*)-2-[(2-Methoxyphenyl)phenylmethylene]bicyclo[3.3.0]oct-6-en-3-one (23). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 82 mg (52%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 2.23−2.31 (m, 3H), 2.69 (dd, J = 9.9, 18.9 Hz, 1H), 3.25−3.33 (m, 1H), 3.51−3.59 (m, 1H), 3.72 (s, 3H), 5.62−5.69 (m, 2H), 6.89−6.98 (m, 2H), 7.22−7.31 (m, 7H); ^{13}C NMR ($CDCl_3$) δ 42.4, 44.1, 45.0, 55.4, 111.4, 120.5, 127.3, 127.5, 128.7, 129.1, 130.7, 133.9, 140.0, 206.4 (six carbons missing as a result of overlap); IR ($CHCl_3$, cm^{-1}) 1706; HRMS calcd for $C_{22}H_{20}O_2$ 316.1463, found 316.1457.

(*E*)-Ethyl 4-[(3-Oxobicyclo[3.3.0]oct-6-en-2-ylidene)-phenylmethyl]benzoate (24). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 75 mg (42%) of the indicated compound as a yellow oil: ^{1}H NMR ($CDCl_3$) δ 1.39 (t, J = 7.2 Hz, 3H), 2.15−2.32 (m, 2H), 2.43−2.53 (m, 1H), 2.69 (dd, J = 9.9, 18.9 Hz, 1H), 3.32−3.36 (m, 1H), 3.73−3.80 (m, 1H), 4.39 (q, J = 7.2 Hz, 2H), 5.63−5.71 (m, 2H), 7.08−7.11 (m, 2H), 7.25−7.31 (m, 5H), 8.01−8.04 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 14.3, 41.4, 42.5, 43.9, 44.4, 61.1, 127.9, 128.1, 128.9, 129.2, 129.5, 130.0, 130.7, 134.0, 139.9, 140.3, 146.5, 148.9, 166.1, 206.5 (one carbon missing as a result of overlap); IR ($CHCl_3$, cm^{-1}) 1709; HRMS calcd for $C_{24}H_{22}O_3$ 358.1569, found 358.1562.

7-Diphenylmethylene-1-methylbicyclo[4.3.0]nonan-8-one (26). The reaction mixture was chromatographed using 60:40 hexanes/ether to afford 94 mg (60%) of the indicated compound as a yellow solid: mp 157−158 °C; ^{1}H NMR ($CDCl_3$) δ 1.07−1.47 (m, 7H), 1.59−1.72 (m, 4H), 1.93 (dd, J = 1.5, 18.0 Hz, 1H), 2.50 (dd, J = 5.7, 10.8 Hz, 1H), 2.68 (d, J = 18.0 Hz, 1H), 7.16−7.36 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 21.7, 24.5, 29.2, 30.2, 34.8, 35.3, 47.2, 49.1, 127.6, 127.7, 128.1, 128.4, 128.9, 140.3, 140.7, 141.9, 148.3, 206.4 (one carbon missing

as a result of overlap); IR ($CHCl_3$, cm^{-1}) 1710; HRMS calcd for $C_{23}H_{23}O$ (M − 1) 315.1749, found 315.1755.

(*E*)-1-Methyl-7-[(4-methoxyphenyl)phenylmethylene]-bicyclo[4.3.0]nonan-8-one (27). The reaction mixture was chromatographed using 70:30 hexanes/ether to afford 74 mg (44%) of the indicated compound as a yellow oil: ^1H NMR ($CDCl_3$) δ 1.10−1.45 (m, 7H), 1.57−1.75 (m, 4H), 1.90 (dd, J = 1.5, 17.7 Hz, 1H), 2.52−2.63 (m, 2H), 3.82 (s, 3H), 6.85−6.88 (m, 2H), 7.10−7.14 (m, 4H), 7.28−7.30 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 21.8, 24.5, 29.2, 30.1, 34.9, 35.5, 47.3, 49.3, 55.1, 113.5, 127.6, 127.7, 129.1, 130.1, 134.3, 140.2, 140.9, 148.1, 159.3, 206.6 (two carbons missing as a result of overlap); IR ($CHCl_3$, cm^{-1}) 1708, 1604; HRMS calcd for $C_{24}H_{26}O_2$ 346.1933, found 346.1929.

(*E*)-1-Methyl-7-[(2*E*)-1-phenylhept-2-enylidene]bicyclo-[4.3.0]nonan-8-one (28). The reaction mixture was chromatographed using 70:30 hexanes/ether to afford 68 mg (43%) of the indicated compound as a yellow oil: ^1H NMR ($CDCl_3$) δ 0.88 (t, J = 6.9 Hz, 3H), 1.06 (s, 3H), 1.23−1.39 (m, 8H), 1.66−1.76 (m, 4H), 1.94−1.99 (m, 1H), 2.12−2.19 (m, 2H), 2.59 (d, J = 17.4 Hz, 1H), 2.68−2.73 (m, 1H), 5.51−5.61 (m, 1H), 6.48 (d, J = 15.3 Hz, 1H), 7.01−7.04 (m, 2H), 7.32−7.35 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 13.8, 22.0, 22.3, 25.0, 29.7, 31.0, 31.1, 33.3, 35.0, 35.2, 47.1, 47.9, 126.8, 127.7, 128.6, 130.3, 138.0, 138.3, 144.4, 144.8, 205.9; IR ($CHCl_3$, cm^{-1}) 1700, 1621, 1583; HRMS calcd for $C_{23}H_{30}O$ 322.2297, found 322.2296.

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Supporting Information Available: ^1H and ^{13}C NMR spectra for all ketone products in Table 2 and preparative procedures and characterization for 1-(1-alkynyl)cyclobutanols **11**, **17**, **21**, and **25**. This material is available free of charge via the Internet at http://pubs.acs.org.

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