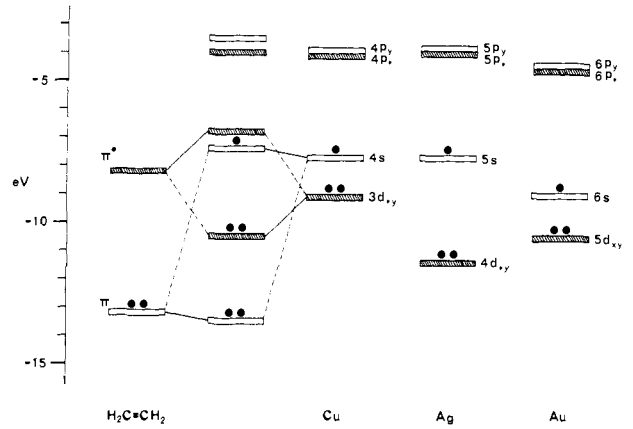

Figure 8. Energy level diagram showing the π and π^* orbitals of ethylene and the valence orbitals of the metal atoms (Cu, Ag, and Au) relevant to π-coordination. The energy levels of copper(0)-mono(ethylene) and the dative interactions responsible for its formation are also indicated.

between the filled π orbital(s) and the vacant s and/or p orbital of the metal atom (eq 3a, 4a, and 4b) are probably insignificant. Thus copper(0)-mono(ethylene) and copper(0)-mono(acetylene) are held primarily by the dative interaction between the d_{xy} and π^* orbitals (eq 3b). The corresponding Ag-monoligand complexes are not formed because of the larger separation between the d_{xy} and π^* orbitals. The formation of bis(ethylene)copper(0) and bis(acetylene)copper(0) is attributed to the dative bond between the d_{xy} and π^* orbitals (eq 4c) and to the one-electron dative bond between the p_x and π^* orbitals (eq 4d). Bis(ethylene)silver(0), on the other hand, is probably held only by the one-electron dative bond of eq 4d.

In a diligand complex of structure II the overlap between the p_x and π^* orbitals would be sensitive to the C-C bond length of the ligands. The shorter C-C bond length of acetylene must adversely affect this overlap; bis(acetylene)silver(0) is not formed. A similar situation has been encountered in the dative interaction between the Al atom ($3s^2,3p^1$) and ethylene and that between Al and acetylene.[27] Aluminum(0)-mono(ethylene) complex is known. Its formation is attributed to the one-electron dative bond between the Al($3p_x$) orbital parallel to the ligand and the π^* orbital of the ligand. The corresponding acetylene complex of Al(0) is not formed.

The complexing ability of the Au atoms is similar to that of Cu in that they can both make the monoligand complex with either ethylene or acetylene. It is similar to that of Ag, however, in that they can both make the diligand complex with ethylene, but not with acetylene.

Registry No. I (M = Au), 87136-54-7; III (M = Au), 84074-14-6; $Au(CaH_4)$, 61943-23-5; gold(0)-bis(ethylene), 87136-55-8; acetylene, 74-86-2.

(27) Kasai, P. H. *J. Am. Chem. Soc.* **1982**, *104*, 1167.

Communications to the Editor

Practical, Cobalt-Mediated, Diastereoselective Synthesis of the 11-(Trimethylsilyl)-3-methoxyestra-1,3,5(10),8(14),9(11)-pentaene Nucleus: A Novel Steroid Intermediate. First Observation of Hindered Rotation in a Vinyltrimethylsilane

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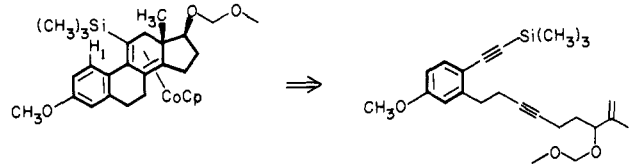
We have recently introduced cobalt-mediated intramolecular achiral enediyne [2 + 2 + 2] cycloadditions as a synthetic method in chiral polycyclic diene construction.[1] We have now found that chiral substrates A (Scheme I) may in some cases [e.g., R_1 = $Si(CH_3)_3$, R_2 = CH_3, $C_6H_5CH_2$, or CH_3OCH_2] undergo the same reaction with remarkable diastereoselectivity, out of the four possible products basically only B (major) and C (minor) being formed.[2]

These transformations may be regarded as suitable models for a diastereoselective synthesis of known and, more importantly, novel hitherto inaccessible steroids.[3] Retrosynthetic analysis of the problem suggests the disconnection **1** → **2**, the ether protecting group chosen such as to maximize the cis stereochemistry in the D ring of **1**.



1 2

A convergent and efficient (42% overall yield) synthetic route to enediyne **2** is shown in Scheme II.[4] It has as its key features an oxazoline-directed ortho-lithiation, methylation, deprotonation sequence,[5] alkylation of the resulting benzyl anion with 6-chloro-4-hexynal ethylene acetal,[6,7] formolysis,[8] Grignard addition,

(1) (a) Sternberg, E. D.; Vollhardt, K. P. C. *J. Am. Chem. Soc.* **1980**, *102*, 4839; *J. Org. Chem.* **1982**, *47*, 3447. (b) Chang, C.; Francisco, C. G.; Gadek, T. R.; King, J. A., Jr.; Sternberg, E. D.; Vollhardt, K. P. C. In "Organic Syntheses: Today and Tomorrow"; Trost, B. M., Hutchinson, C. R., Eds.; Pergamon Press: New York, 1981; p 71. (c) Gadek, T. R.; Vollhardt, K. P. C. *Angew. Chem.* **1981**, *93*, 801; *Angew. Chem., Int. Ed. Engl.* **1981**, *20*, 802.

(2) Stereochemical assignments were based on ^1H NMR spectroscopic analysis,[1] two X-ray structural determinations (see supplementary material), and chemical correlation of ligands by oxidative demetalation.

(3) Akhrem, A. A.; Titov, Y. A. "Total Steroid Synthesis"; Plenum Press: New York, 1970. Blickenstaff, R. T.; Gosh, A. C.; Wolf, G. C. "Total Synthesis of Steroids"; Academic Press: New York, 1974. Lednicer, D. "Contraception: the Chemical Control of Fertility"; Marcel Dekker: New York, 1969.

(4) All new compounds were completely characterized (see ref 11) and gave satisfactory spectral and analytical data.

(5) Gschwend, H. W.; Hamdan, A. *J. Org. Chem.* **1975**, *40*, 2008; *Ibid.* **1982**, *47*, 3652.

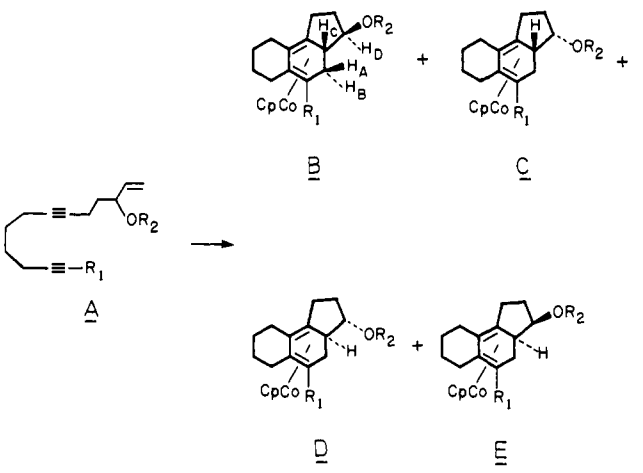
(6) Prepared by alkylation of propargyl alcohol with 3-bromopropanal ethylene acetal[6] [$LiNH_2$, NH_3(l), 4 h, 90%; bp 85 °C (0.1 mm)], followed by chlorination [(a) CH_3SO_2Cl, CH_2Cl_2, $(CH_3CH_2)_3N$, -40 °C, 4 h, room temperature; (b) LiCl, DMF, 60 °C-room temperature, 12 h, 86%; bp 74 °C (0.8mm)].

(7) Büchi, G.; Wüest, H. *J. Org. Chem.* **1969**, *34*, 1122.

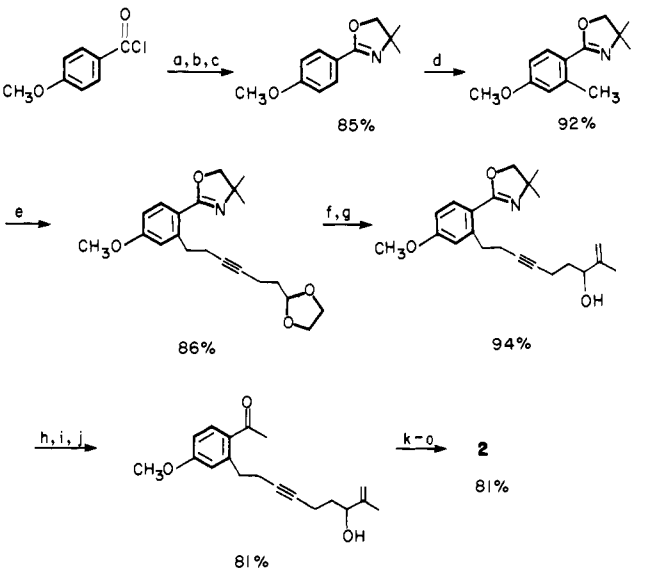
(8) Gorgues, A. *Bull. Soc. Chim. Fr.* **1974**, 529.

0002-7863/83/1505-6710$01.50/0

Scheme I



Scheme II



[a] (a) $H_2NC(CH_3)_2CH_2OH$, CH_2Cl_2, 0 °C–room temperature, 10 h; (b) $SOCl_2$, room temperature, 3 h; (c) NaOH, 0.5 h; (d) *n*-BuLi, ether, −15 °C, 4 h; CH_3I, 5 equiv, room temperature, 3 h; (e) *n*-BuLi, 6-chloro-4-hexynal ethylene acetal, HMPA, −78 °C, 3 h, room temperature; (f) HCOOH, room temperature, 10 h; (g) 2-propenylmagnesium bromide, THF, room temperature, 1 h; (h) CH_3I, 5 equiv, acetone, Δ, 10 h; (i) CH_3MgCl, 3 equiv, THF, Δ, 10 h; (j) 3 N HCl, CH_3OH, room temperature, 4 h; (k) $ClCH_2OCH_3$, 3 equiv, $(CH_3CH_2)_3N$, CH_3CN, 60 °C, 10 h; (l) LDA, THF, −78 °C; (m) $ClP(OCH_2CH_3)_2$, room temperature, 10 h; (n) LDA, 2 equiv, −78 °C, 3 h, room temperature; (o) $(CH_3)_3SiCl$, 3 equiv, room temperature, 3 h.

conversion of the oxazoline unit into a methyl ketone,[5,9] and transformation of the latter into the required phenylalkyne moiety.[10]

Reaction of **2** with $CpCo(CO)_2$ (1.1 equiv, *m*-xylene, Δ, *hν*, 4 h) gave the desired steroid **1** in 72% yield, in addition to the 17-α isomer **3** (20%), *as the only detectable diastereomers*, separated by column chromatography on alumina (Woelm III, hexane–ether, 50:1).[4,11] The efficiency and selectivity of this transformation are remarkable, particularly considering the highly crowded "bay region" generated by the presence of the C_{11}-trimethylsilyl group. Models indicate this substituent to occupy the same area in space as H_1. Indeed, as a result the proton NMR absorption for the trimethylsilyl group in **1** (and also **3**) is very broad at room temperature signaling hindered rotation. Heating to 80 °C sharpens this signal to a singlet. At −35 °C the broad peak separates into three singlets at δ 0.537, 0.520, and 0.029, each integrating for three protons. A temperature-dependence study and computer simulation of the observed spectra give the following activation parameters: $\Delta H^{\ddagger}$ 18.8 kcal mol^{-1}, $\Delta S^{\ddagger} = 6.7$ eu. To our knowledge, this is the first case of an observable restricted rotation of a trimethylsilyl group bound to an sp^2-hybridized carbon.[12]

Oxidative demetalation of **1** gave (81%) the free ligand **4**,[4] in which the trimethylsilyl group is mobile on the NMR time scale even at −75 °C. Clearly, the relative rigidity of the diene moiety, as enforced by complexation in **2**, is responsible for this behavior.

Steroid **4** is air stable, in contrast to its desilylated counterpart.[1a] Since it is now readily obtainable in 24% overall yield from *p*-methoxybenzoyl chloride it should be a useful synthetic intermediate. Thus, the silyldiene function might allow substitution at $C_{8,9,11,14}$ either directly[13] or through the unsaturated 11-keto derivatives.[1a]

Demetalation of **2** ($FeCl_3$, CH_3CN, NEt_3, 0 °C, 0.5 h) in the presence of acid (HCl) resulted (mp 102–104 °C; 82%) in the known[14] 3-methoxy-1,3,5(10),8,14-estrapentaen-17β-ol, which has been converted to estradiol methyl ether.[14] Isomer **3** has been similarly deprotected (75%) to free ligand, and correlated (75%) with the corresponding estrapentaen-17α-ol.[15]

Acknowledgment. This work was supported by NIH GM-22479. K.P.C.V. is a Camille and Henry Dreyfus Teacher-Scholar (1978–1983). E.D. is the recipient of a Fulbright/Ministerio de Universidades e Investigacion (MUI), Spain, Scholarship (1981–1983). J.-C.C. was a C.N.R.S. (France) postdoctoral fellow (1981–1982). The crystal structure analyses were carried out by Dr. F. J. Hollander, staff crystallographer, Dr. C. Orvig, and Messrs. J. M. Boncella, F. Okino, R. P. Planalp, and G. L. Rosenthal on the U.C. Berkeley, Department of Chemistry X-ray facility (CHEXRAY).

Registry No. 1, 87249-12-5; **2**, 87226-48-0; **3**, 87304-29-8; **4**, 87226-49-1; **A** (R_1 = TMS, R_2 = CH_3), 87226-52-6; **A** (R_1 = TMS, R_2 = $C_6H_5CH_2$), 87226-53-7; **A** (R_1 = TMS, R_2 = CH_3OCH_2), 87226-54-8; **A** (R_1 = TMS, R_2 = *t*-BuSi$(CH_3)_2$), 87226-55-9; **A** (R_1 = TMS, R_2 = Si(*i*-Pr)$_3$), 87226-56-0; **A** (R_1 = TMS, R_2 = *t*-BuSi$(C_6H_5)_2$), 87226-57-1; **A** (R_1, R_2 = *t*-BuSi$(CH_3)_2$), 87226-58-2; **B** (R_1 = TMS, R_2 = CH_3), 87249-13-6; **B** (R_1 = TMS, R_2 = $C_6H_5CH_2$), 87249-14-7; **B** (R_1 = TMS, R_2 = CH_3OCH_2), 87249-15-8; **B** (R_1 = TMS, R_2 = *t*-BuSi$(CH_3)_2$), 87249-16-9; **B** (R_1 = TMS, R_2 = Si(*i*-Pr)$_3$), 87249-17-0; **B** (R_1 = TMS, R_2 = *t*-BuSi$(C_{H5})_2$), 87249-18-1; **B** (R_1, R_2 = *t*-BuSi$(CH_3)_2$), 87249-

(9) Meyers, A. I.; Smith, E. *J. Org. Chem.* **1972**, *37*, 4289.

(10) Negishi, E.; King, A. O.; Kline, W. L.; Patterson, W.; Silveira, A., Jr. *J. Org. Chem.* **1980**, *45*, 2526.

(11) For example, **1**: red crystals, mp 95 °C; MS, *m/e* (relative intensity) 522.1987 (calcd 522.2000, M^+, 32), 449 (100); ^1H NMR (250 MHz, C_6D_6) δ 0.33 (s at 80 °C, 9 H), 0.82 (s, 3 H), 0.91 (d, J = 14.8 Hz, H_{12exo}), 1.20–1.40 (m, 2 H), 1.60 (m, 2 H), 1.74 (ddd, J = 16, 8.2, 4.1 Hz, 1 H), 2.12 (ddd, J = 14.6, 4.8, 1.2 Hz, 1 H), 2.20–2.40 (m, 96.26, 2.27 (d, J = 14.8 Hz, H_{12endo}), 2.71 (ddd, J = 14.7, 4.8, 1.7 Hz, 1 H), 3.31 (s, 3 H), 3.39 (s, 3 H), 4.41 (s, 5 H), 4.63 (dd, J = 8.2, 8.2 Hz 1 H), 4.69 (d, J = 6.5 Hz, 1 H), 4.75 (d, J = 6.5 Hz, 1 H), 6.60 (dd, J = 8.5, 2.5 Hz, 1 H), 6.89 (d, J = 2.5 Hz, 1 H), 7.47 (d, J = 8.5 Hz, 1 H); ^{13}C NMR (C_6D_6) 2.04, 21.00, 26.22, 28.18, 29.79, 30.56, 43.26, 43.81, 48.17, 54.83, 55.03, 76.85, 82.17, 87.69, 90.08, 91.68, 96.26, 110.27, 115.04, 129.85, 130.42, 140.72, 159.47. **3**: red crystals, mp 98 °C; MS, *m/e* (relative intensity) 522.1992 (calcd 522.2000, M^+, 56), 73 (100); ^1H NMR (250 MHz, C_6D_6) δ 0.41 (br s, 9 H), 0.49 (s, 3 H), 0.79 (d, J = 14.3 Hz, H_{12exo}), 1.42 (m, 2 H), 1.90 (dddd, J = 16, 8, 8, 2 Hz, 1 H), 2.03 (ddd, J = 16, 8, 8 Hz, 1 H), 2.16 (m, 2 H), 2.25 (ddd, J = 15.0, 4.3, 2.0 Hz, 1 H), 2.66 (d, J = 14.3 Hz, H_{12endo}), 2.77 (ddd, J = 14.3, 4.3, 2.1 Hz, 1 H), 3.36 (s, 3 H), 3.40 (s, 3 H), 3.63 (dd, J = 6.0, 2.0 Hz, 1 H), 4.50 (s, 5 H), 4.59 (d, J = 6.5 Hz, 1 H), 4.76 (d, J = 6.5 Hz, 1 H), 6.63 (dd, J = 8.5, 2.2 Hz, 1 H), 6.90 (d, J = 2.2 Hz, 1 H), 7.51 (d, J = 8.5 Hz, 1 H); ^{13}C NMR (C_6D_6) 2.23, 27.18, 27.67, 30.53, 30.72, 31.05, 38.99, 45.06, 49.15, 54.82, 55.40, 77.99, 82.77, 85.17, 89.03, 91.98, 96.01, 110.27, 114.96, 129.82, 130.34, 140.75, 159.34.

(12) For the very recent observation of rotational isomerism about silicon–carbon single bonds: Nakamura, N.; Kohno, M.; Oki, M. *Chem. Lett.* **1982**, 1809. 1,8-Bis(trimethylsilyl)naphthalene is conformationally mobile at room temperature on the NMR time scale: Soorlyakumaran, R.; Boudjouk, P. *Organometallics* **1982**, *1*, 218.

(13) Koshutin, V. I. *Zh. Obshch. Khim.* **1978**, *48*, 1665.

(14) Rufer, C.; Schröder, E.; Gibian, H. *Justus Liebigs Ann. Chem.* **1967**, *701*, 206.

(15) Haffer, G.; Eder, U.; Sauer, G.; Wiechert, R. *Chem. Ber.* **1975**, *108*, 2665.

19-2; C (R_1 = TMS, R_2 = CH_3), 87304-30-1; C (R_1 = TMS, R_2 = $C_6H_5CH_2$), 87304-31-2; C (R_1 = TMS, R_2 = CH_3OCH_2), 87304-32-3; C (R_1 = TMS, R_2 = t-BuSi$(CH_3)_2$), 87304-33-4; C (R_1 = TMS, R_2 = Si(i-Pr)$_3$), 87304-34-5; C (R_1 = TMS, R_2 = t-BuSi$(C_6H_5)_2$), 87304-35-6; C (R_1, R_2 = t-Bu-Si$(CH_3)_2$), 87304-36-7; D (R_1 = TMS, R_2 = Si(i-Pr)$_3$), 87304-37-8; E (R_1 = TMS, R_2 = CH_3), 87304-38-9; E (R_1 = TMS, R_2 = $C_6H_5CH_2$), 87304-39-0; E (R_1 = TMS, R_2 = CH_3OCH_2), 87304-40-3; E (R_1 = TMS, R_2 = t-BuSi$(CH_3)_2$, 87304-41-4; E (R_1 = TMS, R_2 = Si(i-Pr)$_3$), 87304-42-5; E (R_1 = TMS, R_2 = t-BuSi$(C_6H_5)_2$), 87304-43-6; E (R_1, R_2 = t-BuSi$(CH_3)_2$), 87304-44-7; $CH{=}C(CH_2)_4C{\equiv}C(CH_2)_2C$-H(OH)$CH{=}CH_2$, 87226-61-7; $CH_2{=}CHBr$, 593-60-2; CH_3I, 74-88-4; $C_6H_5CH_2Br$, 100-39-0; $ClCH_2OCH_3$, 107-30-2; $(CH_3)_3SiCl$, 75-77-4; t-BuSi$(CH_3)_2$Cl, 18162-48-6; ClSi(i-Pr)$_3$, 13154-24-0; t-BuSi$(C_6H_5)_2$Cl, 58479-61-1; $H_2NC(CH_3)_2CH_2OH$, 124-68-5; $CpCo(CO)_2$, 12078-25-0; 1,7-octadiyne, 871-84-1; 4-methoxybenzoyl chloride, 100-07-2; 2-bromopropene, 557-93-7; propargyl alcohol, 107-19-7; 3-methoxy-1,3,5-(10),8(14),9(11)-estrapentaen-17β-ol, 87226-50-4; 3-methoxy-1,3,5-(10),8(14),9(11)-estrapentaen-17α-ol, 87226-51-5; 4,10-undecadiynal dimethyl acetal, 87226-59-3; 4,10-undecadiynal, 87226-60-6; 1-bromo-3,3-dimethoxypropane, 36255-44-4; 2-(p-methoxyphenyl)-4,4-dimethyl-2-oxazoline, 53416-46-9; 2-(2-methyl-4-methoxy)-4,4-dimethyl-2-oxazoline, 75817-44-6; 6-chloro-4-hexynal ethylene ketal, 87226-62-8; 7-[2-(4,4-dimethyl-2-oxazolin-2-yl)-5-methoxyphenyl]-4-heptynal ethylene ketal, 87226-63-9; 2-[2-(7-hydroxy-8-methylene-3-nonynyl)-4-methoxyphenyl]-4,4-dimethyl-2-oxazoline, 87226-64-0; 1-(7-hydroxy-8-methylene-3-nonynyl)-2-acetyl-5-methoxybenzene, 87226-65-1; 3-bromopropanal ethylene ketal, 18742-02-4; 7-hydroxy-4-hexynal ethylene ketal, 87226-66-2.

Supplementary Material Available: Results of the preparation and cyclization of **2** (22 complexes), ORTEP drawings, details of the X-ray analyses, a listing of positional and thermal parameters, and table of selected bond lengths and bond angles for B [R_1 = Si$(CH_3)_3$, R_2 = CH_3] and C [R_1 = Si$(CH_3)_3$, R_2 = $C_6H_5CH_2$] (13 pages). Ordering information is given on any current masthead page.

Intramolecular Thermal Cyclization Reactions of Diacryloylamines[1]

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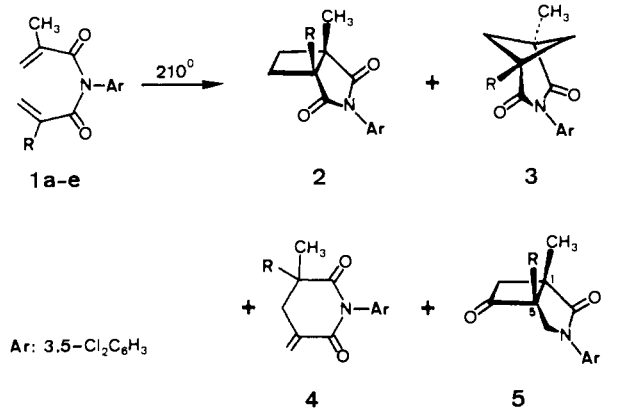
Received May 16, 1983

In the course of screening for antifungal substances against the grey mold fungus on grapes, *Botrytis cinerea*, we have discovered the highly active compound **2b**, which possesses a 3-azabicyclo-[3.2.0]heptane skeleton.[2] As an alternative to our photochemical approach[2] to **2b** we envisaged the synthesis of the cyclobutane moiety of **2** by thermal [2 + 2] cyclodimerization of derivatives of methacrylic acid which are known to proceed regiospecifically yielding exclusively 1,2-disubstituted cyclobutanes.[3] However, in all reported cases the necessary 1,2-cis-disubstituted cyclobutane is formed only as the minor component. It was highly tempting to try to enforce the cis mode of head-to-head [2 + 2] cycloaddition by intramolecular fixing of two acrylic units in diacryloyl amines of type **1** whose thermal behavior has not yet been examined.[4]

Scheme I



Scheme II

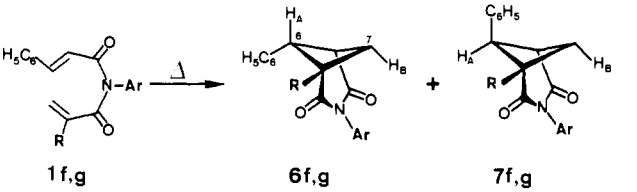


Table I. Substitution Dependence of Product Formation

amide 1	yield, % 2	3	4	5
a, R = H		31	13	6[a]
b, R = CH_3		37	19	7.5
c, R = C_6H_5		50		9
d, R = Si$(CH_3)_3$			21	57[b]
e, R = SCH_3	5[c]	51[c]		

[a] Represents a 1:1 mixture of 5a and the isomer with the position of H and CH_3 exchanged on C(1) and C(5), respectively. [b] Isolation of pure cyclobtanone 5d (mp 127-128 °C) was achieved by crystallization from the crude reaction mixture after thermolysis of 1d (47% yield). In a separate experiment it was observed that upon chromatography of 5d on silica gel (25 °C) the angular trimethylsilyl group on C(5) was removed in a remarkably clean reaction to give 5a in quantitative yield. Thus, the yield given represents the yield of isolated 5a (mp 120-120.5 °C) after treatment of the crude thermolysis mixture with excess of silica gel at room temperature. [c] In this case, 2e and 3e were formed even at 170 °C (5 h).

Table II. Cyclization Experiments with Amides 1f and 1g

amide 1	reaction time, temperature	yield, % 6	7
f, R = CH_3	4 h, 140 °C	45	30
g, R = SCH_3	5 min, 140 °C	46	45
	3 days, 40 °C	51	39

The thermolysis of unsaturated amides **1**[5] was examined in 1,3-dichlorobenzene[10] in sealed tubes (210 °C, 3-16 h). The

(1) Synthesis and Reactivity of Compounds with Cyclobutane Ring(s). 19. For part 18, see: Alder, A.; Bühler, N.; Bellus, D. *Helv. Chim. Acta* **1982**, *65*, 2405.

(2) Bühler, N.; Baumann, M.; Belluš, D.; Sturm, E. (Ciba-Geigy AG), Eur. Pat. Appl. 17994, 1979. Here the cyclobutane moiety in **2b** was obtained by benzophenone-sensitized [2 + 2] photocycloaddition of ethylene to *N*-(3,5-dichlorophenyl)dimethylmaleimide at −78 °C.

(3) (a) Seebach, D. "Houben-Weyl, Methoden der Organischen Chemie"; Georg Thieme Verlag: Stuttgart, 1971; Vol. IV/4, p 267 (review). (b) Doering, W. v. E.; Guyton, C. A. *J. Am. Chem. Soc.* **1978**, *100*, 3229. (c) Lingnau, J.; Stickler, M.; Meyerhoff, G. *Eur. Polym. J.* **1980**, *16*, 785.

(4) Examples of thermally induced intramolecular [2 + 2] cycloadditions of molecules having two multiple bonds separated by three atoms to form cyclobutanes or cyclobutenes are rare. Reported to date have been the exclusive head-to-head mode to yield bicyclo[3.2.0]heptane skeletons, cf.: (a) Oppolzer, W.; Loosli, H.-R. *Helv. Chim. Acta* **1974**, *57*, 2605. (b) Klemm, L. H.; Hwang, Y. N.; McGuire, T. M. *J. Org. Chem.* **1976**, *41*, 3813. (c) Doering, W. v. E., personal communication cited in: Dewar, M. J. S.; Wade, L. E. *J. Am. Chem. Soc.* **1977**, *99*, 4417. (d) Shea, K. J.; Wise, S. *Tetrahedron Lett.* **1978**, 2283. (e) Shea, K. J.; Wise, S.; Burke, L. D.; Davis, P. D.; Gilman, J. W.; Greely, A. C. *J. Am. Chem. Soc.* **1982**, *104*, 5708. The exclusive head-to-tail mode to yield bicyclo[3.1.1]heptane skeletons, cf.: (f) ref 4a. (g) Ramamurthy, V.; Liu, R. S. H. *J. Org. Chem.* **1974**, *39*, 3435. as well as mixed modes, cf.: (h) Meinwald, J.; Kapecki, J. A. *J. Am. Chem. Soc.* **1972**, *94*, 6235. (i) Nelsen, S. F.; Gillespie, J. P. *Ibid.* **1972**, *94*, 6238.

0002-7863/83/1505-6712$01.50/0