

°C and identical with the sample isolated in the methylation studies.

1-Amino-1-methyl-2-phenylperhydroazepinium Iodide (10). A solution of 1-amino-2-phenylperhydroazepine (3, 7.24 g) in acetonitrile (20 mL) was heated at reflux with methyl iodide (5 mL) for 17 h under nitrogen. Removal of the solvent gave a solid which was recrystallized from absolute ethanol: yield 3.55 g; mp 165–167 °C; IR (Nujol) 3.07, 3.18 (NH), 14.16 (C₆H₅) μm; NMR (CDCl₃-Me₂SO-*d*₆) δ 1.50–2.50 (m, 8 H, 3,4,5,6-CH₂), 3.07, 3.29 (2 s, 3 H, CH₃), 3.67–4.48 (m, 2 H, 7-CH₂), 4.85–5.33, (m, 1 H, 2-CH), 5.45–5.87 (br singlets, 2 H, NH₂), 7.32–7.90 (m, 5 H, C₆H₅). The singlets at δ 5.45 and 5.87 underwent slow exchange with D₂O.

Studies on the Acetylation of 1-Amino-1-methyl-2-phenylperhydroazepinium Iodide (10). (A) The iodide 10 was not affected by heating with acetic anhydride in acetonitrile for 6 h.

(B) The iodide 10 (2.66 g) was heated under nitrogen with ethyl acetate (0.71 g) and potassium *tert*-butoxide (0.90 g) in *tert*-butyl alcohol (50 mL) at 85–90 °C for 42 h. The resulting reaction mixture was filtered, and the filtrate upon removal of the solvent gave a yellow oil which distilled at 117–120 °C (0.5 mm); yield 0.34 g. The product, on the basis of its IR and NMR spectra, appeared to be a mixture of 1-methyl-1-(6-phenyl-5-hexenyl)-hydrazine (12) and 1-methyl-3-phenylperhydro-1,2-diazocine (13) in a ratio of 1:1.4: IR (neat) 3.05 (weak), 3.27, 3.41, 3.51, 3.58, 5.93 (weak), 6.23, 6.68, 6.79, 6.96, 7.40, 8.35, 9.35, 9.81, 9.97, 10.36, 11.45, 12.02, 13.10, 13.45, 13.85, 14.27 μm; NMR (CDCl₃) δ 1.17–2.00 (m, 10.3 H, CH₂, NH, NH₂, CH₂N, CH₂C=C), 2.00–3.00 (m, 10.3 H, CH₂, NH, NH₂, CH₂N, CH₂C=C), 2.40 (s, 3 H, NCH₃), 4.02 (t, 0.7 H, CHC₆H₅, *J* = 5 Hz), 6.13 (dt, 0.5 H, olefinic, *J* = 15, 6 Hz), 6.44 (d, 0.5 H, olefinic, *J* = 15 Hz), 7.00–7.62 (m, 5 H, aromatic).

The absorptions in the IR fingerprint region (8.35, 9.35, 9.81, 9.97, 11.45, 12.02, 13.10, 13.85, and 14.27 μm) were identical with those found for an authentic sample of 13.⁴ The triplet in the NMR spectrum at δ 4.02 (*J* = 5 Hz) for the benzylic proton of 13 occurs at the same point as that reported in the spectrum of an authentic sample.⁴

Registry No. 1, 3466-82-8; 2, 77153-75-4; 3, 77153-76-5; 4, 77153-77-6; 6, 77153-78-7; (E)-7, 77153-79-8; 10, 77153-80-1; 12, 77172-41-9; 13, 49868-87-3.

Ring Opening of Cyclopropyl Ketones by Trimethylsilyl Iodide

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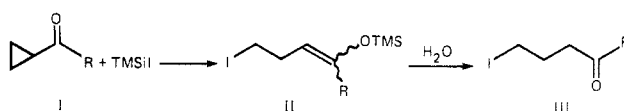
Recent advances in the generation of three-membered rings have led to the steadily increasing usage of cyclopropyl derivatives as reagents for organic synthesis.¹ In this respect, the nucleophilic ring opening of electron-deficient cyclopropane derivatives has received considerable attention and has been recently reviewed.² Similarly, the acid-catalyzed ring opening of cyclopropylcarbinyl alcohols has also been successfully employed for the synthesis (often stereoselective) of a variety of useful olefin derivatives.³

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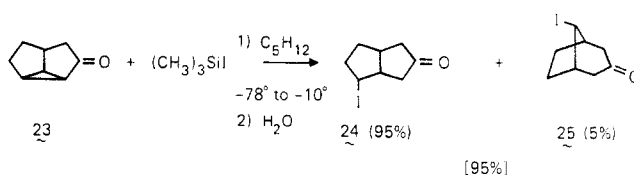
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Scheme I



Scheme II



Unlike the ring opening of cyclopropyl ketones and esters by nucleophiles which usually requires diactivation, a number of electrophilically initiated ring openings of monofunctional cyclopropyl derivatives have been reported.⁴ These have traditionally employed acidic reagents and often require reaction conditions which are not compatible with sensitive functionality. In addition, the regioselectivity of the ring opening, particularly in polycyclic systems, is somewhat unpredictable and may be a sensitive function of reaction conditions, structure, substituents, etc.^{4b} Nevertheless, in spite of the many uncertainties, the electrophilic ring opening of cyclopropyl carbonyl derivatives has been successfully utilized in a number of natural product syntheses.⁵

Trimethylsilyl iodide (Me₃SiI) is a highly electrophilic reagent of considerable synthetic importance.⁶ The reactivity of this reagent with α,β-unsaturated enones^{6f} suggested that cyclopropyl ketones could be similarly transformed to generate either γ-iodo ketones or the corresponding iodotrimethylsilyl enol ethers (see Scheme I), depending on the reaction conditions. We describe here in some detail the general utility of this reagent for the former purpose.

The ring opening which proceeds rapidly under very mild conditions ultimately generates the corresponding γ-iodo ketones upon hydrolytic workup (Table I).^{7,8} In-

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
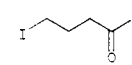



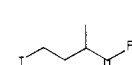



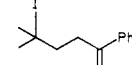



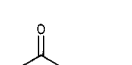







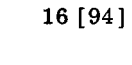

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(7) Consistent spectral data were obtained for all compounds. The iodo ketones were reduced by using tri-*n*-butyltin hydride for comparison with authentic samples. Satisfactory analytical and/or high-resolution mass spectroscopic data were obtained for new compounds or suitable derivatives.

(8) Under these reactions conditions, simple cyclopropylcarboxylic acid esters were unreactive. Reactivity was, however, observed for diactivated esters such as 1,1-dicarbethoxycyclopropane and 6,6-dimethyl-5,7-dioxaspiro[2.5]octane-4,8-dione, which yielded the expected iodo esters in good yield after 6–48 h at 25 °C.

(9) Under the stated conditions, less than 5% of 4-iodocyclohexanone was produced (NMR analysis). When the Me₃SiI was added at higher temperatures (i.e., 25 °C), the ring opening became slightly less regioselective.

Table I

reactant	conditions ^a	product [total yield, %]	product	conditions ^a	product [total yield, %] ^a
 1a, R = Me b, R = Ph	-20 °C, 0.5 h; 25 °C, 2 h	 12a [85] ^b b [88]	 7	-20 °C, 0.5 h	 18 [95] ^a
 2	-10 °C, 1 h; 25 °C, 1 h	 13 [98]	 8	0 °C, 1 h; 25 °C, 2 h	 19 [90]
 3	25 °C, 2 h	 14 [98]	 9	-20 °C, 0.5 h	 20 [89]
 4	-5 °C, 1 h; 25 °C, 1 h	 15 [98]	 10	-10 °C, 1 h; 25 °C, 2 h	 21a (90) ^d
 5	-10 °C, 1 h; 25 °C, 2 h	 16 [94]	 11	-10 °C, 1 h; 25 °C, 2 h	 22a (54)
 6	-10 °C, 1 h; 25 °C, 2 h	 17 [96]			 22b (46)
					[95]
					[90]

^a 1.2 equiv of Me₃SiI, CCl₄. ^b Absolute yield. ^c The predominate isomer was tentatively assigned the trans configuration on the basis of its NMR spectrum. ^d Relative yields. ^e Individual yields given in percent in parentheses.

terestingly, the regioselectivity of this transformation is often quite high¹⁰ and for rigid polycyclic derivatives (e.g., 7–9 and 23) involves cleavage of the cyclopropane bond which most effectively overlaps the π orbitals of the carbonyl group in a manner reminiscent of that observed for the cleavage of related systems in dissolving-metal reductions.¹¹ This selectivity is particularly significant for 7 and 9, since both compounds result in rather nonregioselective product formation with other electrophilic reagents.^{4,6,12} In this respect, tricyclo[3.3.0.0^{2,8}]octan-3-one (23) serves as a well studied model for comparison between Me₃SiI and other electrophilic reagents. Accordingly, the reaction of 23 with Me₃SiI results in the regioselective formation of 2-iodobicyclo[3.3.0]octan-7-one (~95%; Scheme II). This selectivity compares quite favorably with

that observed for the lithium–ammonia reduction of 23 and is considerably better than that reported for other electrophilic procedures.^{4,11d,13,14}

In summary, it appears that trimethylsilyl iodide, under exceedingly mild reaction conditions, can be employed to generate synthetically useful γ -iodo ketones from the corresponding cyclopropyl derivatives. Under these conditions, the regioselectivity of bond cleavage in rigid systems is often very high and is predicted by bond overlap considerations in a manner similar to that described for dissolving-metal reductions.

Further investigations in this and related areas are proceeding.

Experimental Section

All solvents were routinely dried and distilled before use. ¹H NMR spectra were recorded on a Varian HA-100 using tetramethylsilane as an internal standard. Infrared spectra were taken on a Perkin-Elmer 297 instrument. The mass spectra were re-

(10) It is interesting that 3 generates the tertiary iodide 14 in relatively good yield. This suggests the development of positive charge on the β -carbons of the cyclopropane ring which can become a determining factor in conformationally unbiased systems. Further mechanistic investigations are proceeding.

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(12) The tricyclic ketone 9 reacted rapidly with 30% hydrogen bromide in acetic acid (25 °C) to yield a mixture of fused ring and spirocyclic bromo ketones as determined by IR and NMR analyses. This mixture was debrominated with tri-*n*-butyltin hydride to yield a 58/42 mixture of spiro[4.5]decan-2-one and 2-decalone: Miller, R. D.; McKean, D. R., unpublished results.

(13) The reaction of 23 with pyridinium hydrochloride in refluxing acetonitrile for extended periods (2–3 days) results in a 4:1 mixture of 2-chlorobicyclo[3.3.0]octan-7-one and 8-chlorobicyclo[3.2.1]octan-3-one: Miller, R. D.; McKean, D. R., unpublished results.

(14) In a manner similar to that reported for acetyl methane-sulfonate,⁴ the use of trimethylsilyl iodide in the presence of a suitably sterically hindered base with a nonhydrolytic workup leads to the regioselective generation of iodotrimethylsilyl enol ethers. The production of materials of this type and their subsequent reactivity will be described in a subsequent publication.

corded on an AEI-MS-30. GLC analyses were accomplished with a Hewlett-Packard 5750 machine using a glass column ($1/4 \times 6$ ft) packed with 10% SE-30 on GAS-CHROM Q. The γ -iodo ketones were fully characterized by their spectral data. Subsequently, they were converted into the corresponding γ -keto sulfides by reaction with lithium thiophenoxide in THF for high-resolution mass spectroscopic analysis. In some cases, the γ -iodo ketones were also reduced to the corresponding ketones by using tri-*n*-butyltin hydride for comparison with authentic samples. This technique was also employed when isomeric mixtures of iodo ketones were obtained from the reaction of trimethylsilyl iodide with certain cyclopropyl ketones to facilitate analysis. These results were compared with those obtained by direct spectroscopic analysis of the γ -iodo ketone mixtures.

Materials. Methylcyclopropyl ketone and phenylcyclopropyl ketone, purchased from Aldrich Chemical Co., were dried over 4-Å molecular sieves and used without further purification. 1-Methylcyclopropyl phenyl ketone (2) was generated from the corresponding carboxylic acid by treatment with phenyllithium in ether. In a similar manner, 3 was produced from the carboxylic acid which was in turn available by the basic hydrolysis of the corresponding nitrile.¹⁵ 3-Cyclopropylcyclohex-2-en-1-one (4) was formed by treatment of commercially available 3-ethoxycyclohex-2-en-1-one with cyclopropyllithium followed by hydrolysis with 5% hydrochloric acid in THF (25 °C). The bicyclic ketone 5 was generated by treatment of the respective acid with methylolithium in ether. The acid was produced by basic hydrolysis of the corresponding ethyl ester resulting from the decomposition of ethyl diazoacetate catalyzed by Cu_2I_2 in cyclohexene. The cyclopropyl ketones 6-8, 10, and 11 were produced from the corresponding enones by the method described by Vorbrüggen and co-workers.¹⁶ Tricyclo[3.3.0.0^{2,8}]octan-3-one (23) was prepared as described by Monti and co-workers.^{11d} In a similar fashion, the tricyclic ketone 9 was prepared from 3-(1-cyclohexenyl)propionic acid.

General Procedure for the Ring Opening of Cyclopropyl Ketones with Iodotrimethylsilane. Ring opening of 2 is representative. To a solution of 160 mg (1 mmol) of 2 in 3 mL of carbon tetrachloride at -10 °C was added 0.155 mL (1.1 mmol) of iodotrimethylsilane. The solution was then stirred 1 h at -10 °C and 1 h at 25 °C. The solution was diluted with 50 mL of ether and washed with 25 mL of saturated Na_2SO_3 solution. The organic phase was removed and dried (MgSO_4). Solvent removal yielded 282 mg (98%) of the iodo ketone 13. The following compounds were also obtained. 12a: $^1\text{H NMR } \delta$ (CCl_4) 3.12 (t, $J = 7$ Hz, 2 H), 2.49 (t, $J = 7$ Hz, 2 H), 1.82-2.17 (m with superimposed s, 5 H); IR (neat) 3000, 2980, 1715, 1425, 1370, 1220, 1180 and 790 cm^{-1} .⁴ⁱ 12b: $^1\text{H NMR } \delta$ (CCl_4) 7.76 (m, 2 H), 7.25 (m, 3 H), 3.20 (t, $J = 7$ Hz, 2 H), 2.97 (t, $J = 7$ Hz, 2 H), 2.12 (m, 2 H); IR (neat) 3060, 2960, 1685, 1600, 1580, 1450, 1225, 1215, 990, 750, 740, and 690 cm^{-1} .⁴ⁱ 13: $^1\text{H NMR } \delta$ (CCl_4) 7.83 (m, 2 H), 7.28 (m, 3 H), 3.6 (m, 1 H), 3.1 (m, 2 H), 1.6-2.5 (m, 2 H), 1.15 (d, $J = 6$ Hz, 6 H); IR (neat) 3080, 3060, 3020, 2960, 1680, 1595, 1580, 1450, 1255, 1220, 970, 705 cm^{-1} . 14: $^1\text{H NMR } \delta$ (CCl_4) 7.80 (m, 2 H), 7.32 (m, 3 H), 3.09 (m, 2 H), 1.88 (m with superimposed s, 8 H); IR (neat) 2975, 2930, 1715, 1460, 1390, 1100 cm^{-1} . 15: $^1\text{H NMR } \delta$ (CCl_4) 5.66 (br s, 1 H), 3.12 (t, $J = 7$ Hz, 2 H), 1.77-2.45 (m, 10 H); IR (neat) 3015, 2940, 1670, 1625, 1430, 1330, 1255, 1195, 890 cm^{-1} . 16: $^1\text{H NMR } \delta$ (CCl_4) 3.95 (ddd, $J = 11, 11, 5$ Hz, 1 H), 1.0-2.7 (m with s (2.05) superimposed, 14 H); IR (neat) 2940, 2860, 1715, 1450, 1360, 1170, 1145, 655 cm^{-1} . 17: $^1\text{H NMR } \delta$ (CCl_4) 3.09 (m, 2 H), 1.1-2.82 (m, 13 H); IR (neat) 2940, 2860, 1700, 1455, 1180 cm^{-1} . 18: $^1\text{H NMR } \delta$ (CCl_4) 3.2 (d, $J = 6$ Hz, 2 H), 1.4-2.6 (m, 7 H); IR (neat) 2960, 2900, 1740, 1400, 1180, 1160 cm^{-1} . 19: $^1\text{H NMR } \delta$ (CDCl_3) 3.2 (d, $J = 4.5$ Hz, 2 H), 1.2-2.7 (m, 9 H); IR (neat) 2940, 2860, 1710, 1450, 1430, 1295, 1225, 1175 cm^{-1} .⁴ⁱ 20: $^1\text{H NMR } \delta$ (CCl_4) δ 4.43 (m, 1 H), 1.22-2.3 (m with s (2.17) superimposed, 14 H); IR (neat) 2940, 2860, 1740, 1450, 1405, 1170 cm^{-1} .

Analysis of Iodo Ketone Mixtures Produced upon Ring Opening of Cyclopropyl Ketones. Iodo ketone mixtures pro-

duced from reaction of cyclopropyl ketones with Me_3SiI were analyzed by reduction of the mixtures with tributyltin hydride. The ketones obtained from reduction were then compared spectrally and chromatographically with authentic samples. Reduction of a mixture of 22a and 22b is representative. To a refluxing solution of 0.31 g (1.07 mmol) of tri-*n*-butyltin hydride in 5 mL of cyclohexane was added dropwise a solution containing 0.246 g (0.98 mmol) of a mixture of 22a and 22b and a few milligrams of azobis(isobutyronitrile) in 3 mL of cyclohexane. The solution then heated to reflux for 4 h. The solution was then diluted with 75 mL of ether, and the resulting organic phase was washed with 25 mL of 5% potassium fluoride solution, water, and saturated sodium chloride solution and then dried (MgSO_4). The residue obtained after solvent removal was chromatographed on 15 g of neutral alumina (activity II) with 5% ether in hexane to yield 120 mg of product. The ketone mixture was then compared spectrally and chromatographically with authentic material and shown to consist of 54% 3-methylcycloheptanone and 46% cyclooctane.

Registry No. 1a, 765-43-5; 1b, 3481-02-5; 2, 26921-44-8; 3, 5685-43-8; 4, 34194-40-6; 5, 13332-18-8; 6, 5743-85-1; 7, 4160-49-0; 8, 5771-58-4; 9, 13705-50-5; 10, 2862-90-0; 11, 16335-43-6; 12a, 3695-29-2; 12b, 65488-05-3; 13, 77070-49-6; 14, 77070-50-9; 15, 77070-51-0; trans-16, 19093-22-2; 17, 77070-52-1; 18, 71987-94-5; 19, 72003-75-9; 20, 77070-53-2; trans-21a, 71988-01-7; 21b, 77070-54-3; 22a, 71987-95-6; 22b, 77070-55-4; 23, 20826-85-1; 24, 77070-56-5; 25, 77070-57-6; trimethylsilyl iodide, 16029-98-4.

Palladium-Catalyzed Reactions: Stereoselective Synthesis of Substituted Cyclopropanes Related to Chrysanthemic Acid. A Simple Route to *cis*-Chrysanthemonitrile

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Considerable interest continues to be shown in functionalized cyclopropanes¹ because of their insecticidal properties. The cyclopropanecarboxylic acid with a *cis* vinylic side chain (as in several synthetic pyrethroids)² is a more potent insecticide than the corresponding *trans* compound. We report here a convenient and general approach for the synthesis of substituted functionalized cyclopropanes (e.g., 5 and 6, Scheme I) in which $\text{C}_1\text{-C}_3$ bond formation occurs via an intramolecular $\text{S}_{\text{N}}\text{V}$ (via 4) mechanism and its application to the preparation of *cis*-chrysanthemonitrile (3).⁴

The acyclic precursors 1 are available via organopalladium intermediates.⁵ The reaction of the *cis* mon-

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