the polyfunctional vinyl iodide **14** gave the desired triene ester **20** in 79% yield with complete retention of all alkene stereochemistries. Deprotection of **20** with HF gave the free alcohol **21** [mp 100–102 °C, $[\alpha]_D$ +102.1° (*c* 1.0, CH_2Cl_2), 92%] without any isomerization of the triene.[13] Hydrolysis of **21** gave 94% of the hydroxy acid **22**;[14] acetylation of **22** followed by hydrolysis of the resulting mixed anhydride gave the acetoxy acid **23** (99%), presenting the left half of the target in suitably protected form.

To construct a chiral synthon corresponding to C-3 through C-8 of the right-half amino diene, we employed the readily available anhydrogalactoside **24**[15] as a chiral source (Scheme II). The stereogenic center at C-6 was generated by trans-diaxial epoxide opening of **24** using 10 equiv of Me_2Mg,[16] to yield 95% of the diol **25**. This was converted to deoxy acetonide **26** in 64% overall yield by the following sequence: (1) selective silylation of equatorial hydroxyl group, (2) conversion of the axial hydroxyl group to the imidazole thiocarbamate followed by radical deoxygenation,[17] (3) *n*-Bu_4NF desilylation, and (4) condensation with acetone/$FeCl_3$.[18] Hydrogenolytic debenzylation of **26** and then Swern oxidation followed by buffered $KMnO_4$ and diazomethane gave ester **27** [mp 47–49 °C, $[\alpha]_D$ −25.4° (*c* 1.6, CH_2Cl_2)] in 70% yield over four steps from **26**.

With the six carbons of **27** corresponding to neooxazolomycin C-3 through C-8 as marked, we used the ester group of **27** as the electrophile in a cyclocondensation[19] with the dianion of amidomalonate **28**. Formation of the dianion of **28** and reverse addition at −78 °C to **27** in THF gave a mixture of **29**α and **29**β (1:1.4 ratio) in 82% yield based on recovered ester (49%).[20] After chromatographic separation, the desired lactam **29**α was rearranged to the thioacetal ester **30** in nearly quantitative yield. This was converted by O-silylation at C-7, saponification, Fujisawa reduction[21] to the carbinol **31**,[22] and silylation of the new hydroxyl group to the fully elaborated thioacetal **32**, mp 118–120 °C, in 59% overall yield from **30**.

Hydrolysis of **32** to the aldehyde **33** (99%), mp 138–139 °C, paved the way for completion of the right half. Treatment of **33** with $CHI_3/CrCl_2$ gave a 70% yield of the (*E*)-vinyl iodide **34** (mp 134–136 °C).[23] Quantitative desilylation[24] gave the triol **35**, which was condensed with our FMOC-amino propenylstannane reagent **36** (1.1 equiv)[25] under Stille conditions,[12] to afford cleanly the (*E*,*E*)-dienylamide **37** (mp 95–98 °C, 84%). Double O-acetylation gave 96% of the diacetate **38**.

Our synthesis culminated in the reaction of the protected acid **23**, *N*,*N*-bis(2-oxo-3-oxazolidinyl)phosphorodiamidic chloride (1.1 equiv), and Et_3N (2.2 equiv) to give the activated anhydride,[26] to which was added a CH_2Cl_2 solution of the free amine prepared by DBU (2 equiv) deprotection[25] of the FMOC diacetate **38** (1 equiv). Reaction for 1 h produced neooxazolomycin triacetate (**39**) in 60% yield. The spectroscopic properties of our synthetic triacetate were in full agreement with those reported for naturally derived **39**.[1a] Finally, careful hydrolysis of **39** with LiOH (10 equiv) followed by acidification gave a 67% yield of pure neooxazolomycin (**1**), identical with an authentic sample by 300-MHz ^{1}H NMR, IR, TLC (silica gel and reverse phase) in several solvent systems, HPLC, and FAB mass spectrometric comparisons.[27]

Supplementary Material Available: Spectral data and physical properties for compounds **4–17**, **19–23**, **25–27**, and **29–39** (10 pages). Ordering information is given on any current masthead page.

(12) Stille, J. K.; Groh, B. L. *J. Am. Chem. Soc.* **1987**, *109*, 813.
(13) Deprotection using other conventional reagents (Bu_4NF, Py·HF, CsF, KF) failed.
(14) Hydrolysis of ester **21** requires assistance from the β-hydroxyl group via hydrogen bonding; silyl-protected ester **20** could not be hydrolyzed.
(15) Buchanan, J. G.; Fletcher, R. *J. Chem. Soc.* **1965**, 6316.
(16) Parker, K. A.; Babine, R. E. *Tetrahedron Lett.* **1982**, *23*, 1763. No product from Payne rearrangement was observed. Treatment of the TBS ether of **24** with excess Me_2CuLi gave an 8:1 mixture of the undesired regioisomer. Reaction of **24** with excess Me_2CuLi gave a 1:1 mixture of regioisomers.
(17) For this modified Barton deoxygenation, see: Rasmussen, J. R.; Slinger, C. J.; Kordish, R. J.; Newman-Evans, D. D. *J. Org. Chem.* **1981**, *46*, 4843.
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(20) This is contrary to the stereochemical outcome in the chiral acyclic model ester (*S*)-MOM lactate, suggesting that the use of a more conformationally flexible acyclic ester synthon could preferentially give the desired α-isomer **29**α (see ref 19); however, direct formation of the acyclic dithioacetal ester from **27** failed.
(21) Fujisawa, T.; Mori, T.; Sato, T. *Chem. Lett.* **1983**, 835.
(22) To confirm the structure, intermediate **31** was converted to the known triacetate which had been prepared from **39** by Uemura et al.; see ref 1a.
(23) Takai, K.; Nitta, K.; Utimoto, K. *J. Am. Chem. Soc.* **1986**, *108*, 7408.
(24) Deprotection at this stage was necessary, because removal of TBS groups from the synthetic disilyl ether of **37** or trisilyl ether of **1** under all conventional methods proceeded poorly.
(25) The vinylstannane **36** was prepared from propargylamine by the sequence following: (1) FMOCCl/Py/CH_2Cl_2; (2) Bu_3SnH/AIBN (Kende, A. S.; DeVita, R. J. *Tetrahedron Lett.* **1990**, *31*, 307).
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Allenyl Chloromethyl Sulfones: New Dienophile–Diene Synthons. A Simple Iterative Ring-Growing Procedure[1]

Eric Block* and David Putman

Department of Chemistry
State University of New York at Albany
Albany, New York 12222

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We report the preparation and applications of new organosulfur reagents, allenyl chloromethyl sulfones (**1**), $RR'C{=}C{=}CHSO_2CH_2Cl$, functioning as potent dienophiles whose Diels–Alder adducts give 1,3-dienes with base, thus allowing two-step "cyclohomologation" of dienes. Examples of this class of reagents include the parent compound, chloromethyl 1,2-propadienyl sulfone (**2**), $H_2C{=}C{=}CHSO_2CH_2Cl$, chloromethyl tetradeca-1,2-dienyl sulfone (**3**), $CH_3(CH_2)_{10}CH{=}C{=}CHSO_2CH_2Cl$, and chloromethyl 3-methylbuta-1,2-dienyl sulfone (**4**), $Me_2C{=}C{=}CHSO_2CH_2Cl$. Reagents **1** were developed in the course of seeking new applications of the Ramberg–Bäcklund reaction in which the necessary reaction components, sulfonyl group and α-halogen, are already present in the same reagent.[2] We describe herein the use of **1** in a novel iterative ring-growing procedure for construction of linear fused carbocycles.

The choice of **1** was suggested by the known high reactivity of sulfonylallenes as dienophiles due to their low LUMO,[3] the anticipated susceptibility of the allylic sulfone Diels–Alder adduct toward base-induced elimination, and a simple projected synthesis of **1** via coupling of chloromethylsulfenyl chloride, $ClCH_2SCl$,[4] with propargylic alcohols ($RR'C(OH)C{\equiv}CH$) giving *S*-chloromethyl propargyl sulfenates, $ClCH_2SOCRR'C{\equiv}CH$ (**5**), [2,3]-sigmatropic rearrangement[5] of **5** to chloromethyl 1,2-alkadienyl sulfoxides, $RR'C{=}C{=}CHS(O)CH_2Cl$ (**6**), and oxidation

(1) Presented at the 199th National Meeting of the American Chemical Society, April 24, 1990, Boston, MA.
(2) (a) Block, E.; Aslam, M.; Eswarakrishnan, V.; Gebreyes, K.; Hutchinson, J.; Iyer, R.; Laffitte, J.-A.; Wall, A. *J. Am. Chem. Soc.* **1986**, *108*, 4568. (b) Block, E.; Aslam, M. *Org. Synth.* **1987**, *65*, 90.
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(5) Braverman, S.; Stabinsky, Y. *Isr. J. Chem.* **1967**, *5*, 125.

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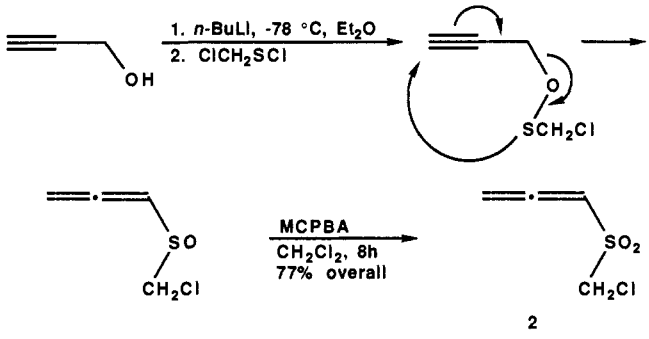
Scheme I



of **6** to **1**. A variety of propargylic alcohols are available through addition of $HC{\equiv}CMgBr$[6] to carbonyl compounds, $RR'C{=}O$.

The success of our procedure depended in part on our discovery that Douglass's synthesis[4] of $ClCH_2SCl$ from the solid chlorination product of dimethyl disulfide, CH_3SCl_3, can be substantially improved if the latter is prepared and decomposed as a 10% solution in CH_2Cl_2. Under these conditions, pure $ClCH_2SCl$ can be conveniently and safely prepared on a large scale in almost quantitative yield. Formation of the sulfenate ester is best conducted by allowing an ethereal solution of 1 equiv of the lithium salt of the propargylic alcohol to react at −78 °C with $ClCH_2SCl$ and then repeatedly filtering the mixture as it warms to remove LiCl (which otherwise catalyzes decomposition). Concentration affords chloromethyl 1,2-propadienyl sulfoxide (**7**), $H_2C{=}C{=}CHS(O)CH_2Cl$ (from propargyl alcohol), chloromethyl tetradeca-1,2-dienyl sulfoxide (**8**), $CH_3(CH_2)_{10}CH{=}C{=}CHS(O)CH_2Cl$ (from dodecanal/$HC{\equiv}CMgBr$ adduct), or chloromethyl 3-methylbuta-1,2-dienyl sulfoxide (**9**), $Me_2C{=}C{=}CHS(O)CH_2Cl$ (from commercially available 2-methyl-3-butyn-2-ol), respectively. Oxidation (MCPBA) of these allenyl sulfoxides affords **2** (a colorless, odorless solid, mp 39 °C) (Scheme 1), **3** (a colorless solid, mp 37 °C), and **4** (a colorless oil) in 47%, 69%, and 61% overall yields, respectively.[7,8]

A 2:1 mixture of 1,2-bis(methylene)cyclohexane[2b] and **2** was warmed to 60 °C for 3 h and the product was diluted with THF, treated with 1 equiv of KO-*t*-Bu at 0 °C, and worked up to directly afford triene **11** in 57% overall yield via Ramberg–Bäcklund[2] rearrangement of chloromethyl sulfone **10**.[9] Repetition of the process twice gave tetraene **12** and then pentaene **13**, each in 85% overall yield (Scheme II). In these reactions, reagent **2** is functioning as a synthetic equivalent of 1,2,3-butatriene. Table I summarizes the results of application of the above procedure to various Diels–Alder adducts obtained from reaction of **2**–**4** with cyclopentadiene, furan, furfural diethyl acetal, 1,3-cyclohexadiene,

Scheme II

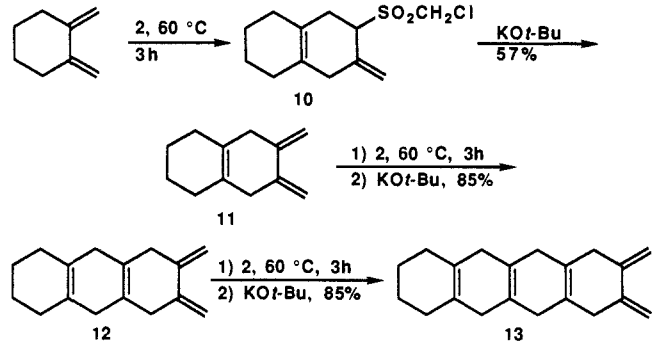
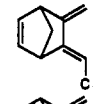
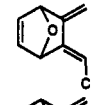


Table I. Synthesis of 1,3-Dienes Using Chloromethyl 1,2-Propadienyl Sulfone (**2**), Chloromethyl 1,2-Tetradecadienyl Sulfone (**3**), and Chloromethyl 3-Methyl-1,2-butadienyl Sulfone (**4**)

Entry	Diene	Dienophile	Diels-Alder Conditions	Product (Yield)[a]
1		2	25 °C, 5 min	 (85%)[b]
2		2	60 °C, 3 h	 (85%)[c]
3		2	60 °C, 2 h	 (68%)
4		2	60 °C, 2 h	 (60%)[b]
5		2	60 °C, 3 h	 (57%)[d]
6	 11	2	60 °C, 3 h	 (85%) 12
7	 12	2	60 °C, 3 h	 (85%) 13
8		2	80 °C, 5 h	 (57%)
9		3	80 °C, 30 min	 (60%) $C_{11}H_{23}$-*n*
10		3	80 °C, 3 h	 (49%) $C_{11}H_{23}$-*n*
11		4	25 °C, 5 min	 (85%)

[a] All new compounds have been fully characterized spectroscopically. [b] Alder, K.; Hartung, S.; Netz, O. *Chem. Ber.* **1957**, *90*, 1. Bowe, M. A. P.; Miller, R. G. J.; Rose, J. B.; Wood, D. G. M. *J. Chem. Soc.* **1960**, 1541. [c] Roth, W. R.; Humbert, H.; Wegner, G.; Erker, G.; Exner, H.-D. *Chem. Ber.* **1975**, *108*, 1655. [d] Bailey, W. J.; Hudson, R. H.; Liau, C.-W. *J. Am. Chem. Soc.* **1958**, *80*, 4358.

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(7) Preparation of **2**: A solution of MeSSMe (30 mL) in CH_2Cl_2 (300 mL) is treated at −78 °C with Cl_2 with a 9-mm glass tube until Cl_2 is in excess (yellow-green slurry). The flask (drying tube) is warmed to 25 °C, and HCl evolves. Concentration in vacuo (100 mm) affords pure $ClCH_2SCl$ (bp 50–55 °C/115 mm; 75 g, 97% yield), a pungent yellow liquid, ^1H NMR δ 5.1 (s). Propargyl alcohol (9 mL; 0.16 mol) in Et_2O (500 mL) is treated at −78 °C with *n*-BuLi (66 mL, 0.16 mol), and then $ClCH_2SCl$ (19 g, 0.16 mol) is added dropwise. After 30 min, the solution is allowed to warm to 25 °C as it is repeatedly filtered through a frit covered by a thin layer of silica gel to remove the LiCl. Concentration gives **7** as a yellow oil or colorless needles, mp 49–49.5 °C, from ether/hexane: ^1H NMR δ 6.00 (t, 1 H, J = 6 Hz), 5.3 (d, 2 H, J = 6 Hz), 4.4 (s, 2 H); IR 3000 (m), 1925 (m), 1060 (s) cm^{-1}. This is dissolved in CH_2Cl_2 (250 mL) and oxidized overnight at 25 °C with MCPBA (50–60%; 47 g, 0.16 mol). The solution is washed ($NaHSO_3$ (3×); $NaHCO_3$ (3×)), dried ($MgSO_4$), and concentrated and the product recrystallized from Et_2O/hexane, to give **2**, a colorless solid, mp 39–39.5 °C (11.7 g, 47%): ^1H NMR δ 6.26 (t, 1 H, J = 6.3 Hz), 5.62 (d, 2 H, J = 6.3 Hz), 4.53 (s, 2 H); ^{13}C NMR δ 212.25 (=C=), 95.17 (=CH), 84.34 ($=CH_2$), 58.26 (CH_2); IR 3000 (m), 1965 (m), 1328 (s), 1247 (m), 1148 (s), 1120 (s), 872 (m) cm^{-1}. Compound **3**, obtained in 69% yield after chromatography (silica gel, 5:1 CH_2Cl_2/hexane), was a colorless solid, mp 37–37.5 °C: ^1H NMR δ 6.15 (m, 1 H), 5.99 (dd, 1 H, J = 6 Hz), 4.48 (s, 2 H), 2.23 (m, 2 H), 1.47 (m, 2 H), 1.24 (br s, 16 H), 0.86 (t, 3 H, J = 7 Hz); ^{13}C NMR δ 208.9 (=C=), 101.3 (=CH), 95 (=CH), 57.9 (CH_2), 31.8 (CH_2), 29.5 (CH_2), 29.4 (CH_2), 29.3 (CH_2), 29.2 (CH_2), 28.9 (CH_2), 28.4 (CH_2), 27.6 (CH_2), 22.6 (CH_2), 14.0 (CH_3); IR 2924 (vs), 2853 (vs), 1954 (m), 1465 (m), 1330 (s), 1147 (s) cm^{-1}. Compound **4**, obtained in 61% yield after chromatography, was a colorless oil: ^1H NMR δ 5.98 (m, 1 H), 4.46 (s, 2 H), 1.89 (d, 6 H, J = 3.3 Hz); ^{13}C NMR δ 207.5 (=C=), 107.3 (=C), 92.6 (=CH), 57.6 (CH_2), 19.4 (CH_3); IR 3015 (m), 2950 (m), 1960 (m), 1320 (vs), 1115 (vs) cm^{-1}.

(8) All new compounds show correct elemental analysis and/or MS molecular ions.

(9) The lower yield of **11** compared to **12** and **13** is due to formation of significant quantities of a minor Diels–Alder adduct in addition to major adduct **10**. This minor adduct, which we have been unable to isolate in pure form, is apparently decomposed by base, giving unidentified products, which are removed during workup of **11**. The regioselectivity of the Diels–Alder reactions with **2** is higher with **11** and **12**.

1,2-bis(methylene)cyclohexane, and 2,3-dimethyl-1,3-butadiene.

Homologues of 1,2-bis(methylene)cyclohexane related to **11**, prepared by lengthier procedures, have been employed in syntheses of pentacene.[10] *cis*-1,4-Dichloro-2-butene has also been employed as a dienophile–diene synthon but requires "severe and carefully controlled reaction conditions [typically several days at 190–200 °C], was somewhat erratic", and gave only moderate yields.[11] Furthermore, *cis*-1,4-dichloro-2-butene does not react with either furan or 1,3-cyclohexadiene.[12] We shall report elsewhere on the application of reagents **1** in the synthesis of substituted polyacenes.

Acknowledgment. We thank David Block and Ruben Tommasi for experimental assistance and gratefully acknowledge support from the donors of the Petroleum Research Fund, administered by the American Chemical Society, the Herman Frasch Foundation, and the National Science Foundation.

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Design, Preparation, and Electron Spin Resonance Detection of a Ground-State Undecet ($S = 5$) Hydrocarbon

Ikuo Fujita, Yoshio Teki, Takeji Takui, Takamasa Kinoshita, and Koichi Itoh*

Department of Chemistry, Faculty of Science
Osaka City University, Sugimoto 3-3-138, Sumiyoshi-ku
Osaka 558, Japan

Fumio Miko and Yasuhiko Sawaki

Department of Applied Chemistry, Faculty of Engineering
Nagoya University, Nagoya 464, Japan

Hiizu Iwamura,*,† Akira Izuoka,‡ and Tadashi Sugawara‡

Institute for Molecular Science, Okazaki 444, Japan

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Synthetic organomagnetic materials such as organic ferromagnets[1] are a recent topic attracting both academic and industrial interest. As part of our program for obtaining purely organic ferromagnets,[2-4] we have synthesized and detected an aromatic hydrocarbon **1** which has an undecet electronic ground state with 10 parallel spins ($S = 5$). This is the highest spin multiplicity known to date for organic molecules. This novel aromatic hydrocarbon has been designed by utilizing topological symmetry of its π electron network.[2,3] The behavior of many spins in such hydrocarbons as well as in other organic high-spin molecules reported by several authors[5] is of key importance for the theory of organic magnetism.[2a,3,6]

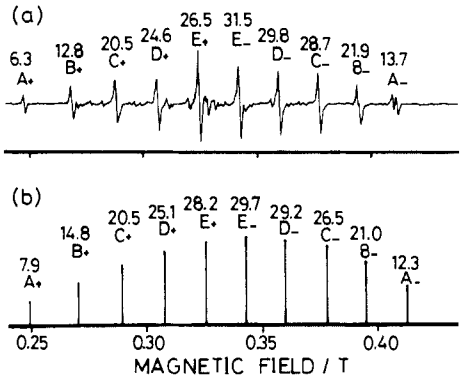
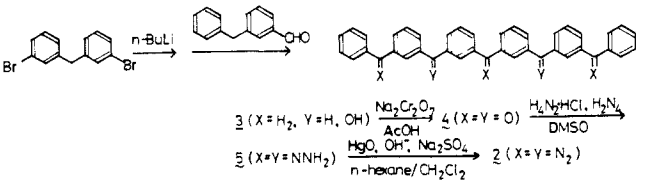
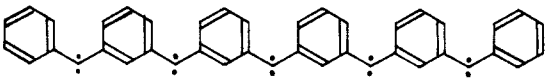


Figure 1. ESR spectra observed after photolysis at 77 K with the magnetic field along the *b* axis of the 1,3-dibenzoylbenzene crystal (*Pbca* space group). (a) Observed at 50 K. The microwave frequency is 9438.9 MHz. (b) Theoretical stick spectrum obtained by the exact diagonalization of the spin Hamiltonian at $\nu = 9438.9$ MHz. The figures above each line represent the relative signal intensities.

Scheme I





1

Hydrocarbon **1** was generated at 77 K by photolysis of the pentakis(diazo) precursor **2** which was diluted in a single crystal of 1,3-dibenzoylbenzene (*Pbca* space group). The photolysis was carried out with an XBO 500-W high-pressure mercury lamp using a quartz rod which guided the light into an X-band TE_{102} cavity of a Bruker ESP300 spectrometer equipped with an Oxford ESR910 variable temperature controller. The mixed crystals were grown in the dark by slowly cooling a benzene-d_6 solution containing 1,3-dibenzoylbenzene and 0.0027 mol fraction of **2**.

The pentakis(diazo) compound **2** was prepared as in Scheme I. Bis(3-bromophenyl)methane[7] was lithiated and allowed to react with excess 3-benzylbenzaldehyde[8] to give **3**. Oxidation of **3** with $Na_2Cr_2O_7$ gave **4**, mp 249–253 °C in 58% yield based on the dibromide: IR 1660 cm^{-1}.[9] Pentahydrazone **5**, mp 100–103 °C,

† Present address: Department of Chemistry, Faculty of Science, The University of Tokyo, Hongo, Bunkyo, Tokyo 113, Japan.
‡ Present address: Department of Pure and Applied Sciences, College of Arts and Sciences, The University of Tokyo, Komaba, Megro, Tokyo 153, Japan.

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