



The following Communications have been judged by at least two referees to be “very important papers” and will be published online at [www.angewandte.org](http://www.angewandte.org) soon:

A. Scarso,\* L. Pellizzaro, O. De Lucchi, A. Linden, F. Fabris\*  
**Gas Hosting in Enantiopure Self-Assembled Oximes**

S. Wittrock, T. Becker, H. Kunz\*

**Synthetic Vaccines from Tumor-Associated Glycopeptide Antigens by Immunocompatible Linking to Bovine Serum Albumin through Thioether Formation**

M. Manoli, R. D. L. Johnstone, S. Parsons, M. Murrie, M. Affronte, M. Evangelisti,\* E. K. Brechin\*

**A Ferromagnetic Mixed-Valent Mn Supertetrahedron: Towards Low-Temperature Magnetic Refrigeration with Molecular Clusters**

X. Lou, G. Zhang, I. Herrera, R. Kinach, O. Ornatsky, V. Baranov,\* M. Nitz,\* M. A. Winnik\*

**Polymer-Based Elemental Tags for Sensitive Bioassays**

Y. Sawada, K. Matsumoto, T. Katsuki\*

**Titanium-Catalyzed Asymmetric Epoxidation of Nonactivated Olefins with Hydrogen Peroxide**

Frank H. Westheimer (1912–2007)

## Obituary

E. J. Corey ————— 3610

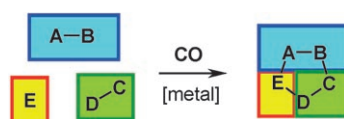
Particles in Water

John Gregory

## Books

reviewed by T. Hofmann,  
F. v. d. Kammer ————— 3611

**An assembly line for heterocycles:** Multicomponent reactions have become a highly valuable tool for the generation of compound libraries particularly of heterocyclic drug candidates. In particular, metal-assisted transformations have enabled the further extension of established methods and development of new strategies. This Highlight presents some innovative examples in which carbon monoxide is used to generate diverse heterocycles.

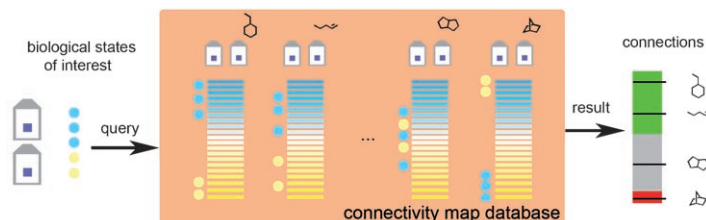


## Highlights

### Heterocyclic Chemistry

M. D. Mihovilovic,\*  
P. Stanetty ————— 3612–3615

Metal-Assisted Multicomponent Reactions Involving Carbon Monoxide—Towards Heterocycle Synthesis



### Small-Molecule Profiles

J.-S. Lee, Y.-T. Chang\* ————— 3616–3618

Understanding the Activity of Small Molecules in the Genomics Context

**Let's look at a map:** Significant improvements in the analysis of gene-expression data and of large data sets have led to the development of the so-called “connectiv-

ity map”, an open database of profiles of small molecules (gene-expression patterns) used to clarify the connections between genes, diseases, and drugs.

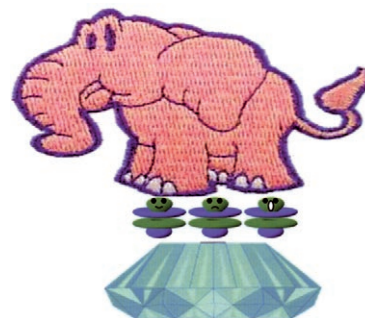
## Reviews

### High-Pressure Chemistry

W. Grochala,\* R. Hoffmann,\* J. Feng,\*  
N. W. Ashcroft\* ————— 3620–3642

The Chemical Imagination at Work  
in Very Tight Places

**Extreme chemistry:** There is a place for chemical intuition and bonding ideas in the structure and electronics of matter under high pressure. When increased density is the only imperative, van der Waals space is first squeezed out. This step is followed by an increase in coordination (often involving multicenter electron-deficient or electron-rich bonding), a differential compression of anions, metallization, Peierls distortions, and a regime of unusual packing and correlation effects.



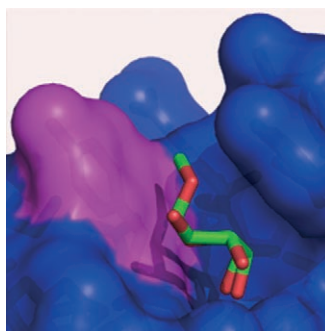
## Communications

### Sugar–Aromatic Complexes

J. Screen, E. C. Stanca-Kaposta,  
D. P. Gamblin, B. Liu, N. A. Macleod,  
L. C. Snoek,\* B. G. Davis,\*  
J. P. Simons\* ————— 3644–3648



IR-Spectral Signatures of Aromatic–Sugar  
Complexes: Probing Carbohydrate–  
Protein Interactions



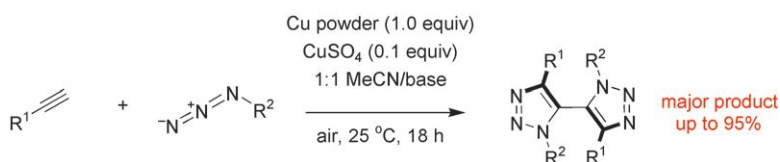
**Key interactions:** Sugar–arene complexes have been created in molecular-beam experiments and observed by IR ion-dip spectroscopy in the gas phase. These complexes are powerful models of the selective recognition seen in protein–sugar complexes, for example between the galactose-specific lectin from *Artocarpus hirsute* and MeGal (see picture).

### Click Chemistry

Y. Angell, K. Burgess\* ————— 3649–3651



Base Dependence in Copper-Catalyzed  
Huisgen Reactions: Efficient Formation of  
Bistriazoles



**Click, click:** Using a carbonate base as an additive in the copper-accelerated click reaction of alkynes and azides led to an oxidative coupling to form predominately 5,5'-bistriazoles (see scheme). The reaction proved more facile for propargylic

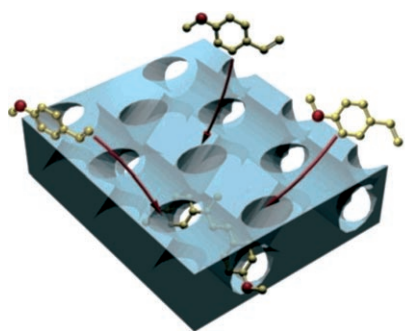
ethers and less hindered substrates. Use of an optically active azide gave separable atropisomeric products and provides a convenient access to optically pure derivatives.

### For the USA and Canada:

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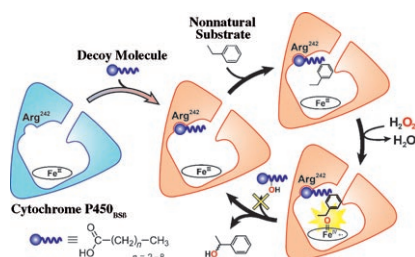


**Pore geometry matters:** Microspectroscopy with polarized light reveals that in the oligomerization of styrene on large ZSM-5 crystals, different pore geometries may lead to distinct reaction products aligned within straight pores of the zeolite. In the body of the crystal, zigzag pore openings provide access for monomer molecules to diffuse inside and react with the dimer, whereas oligomerization at the edges leads toward mainly dimeric products.

### Zeolite Catalysts

M. H. F. Kox, E. Stavitski,  
B. M. Weckhuysen\* — 3652–3655

Nonuniform Catalytic Behavior of Zeolite Crystals as Revealed by In Situ Optical Microspectroscopy



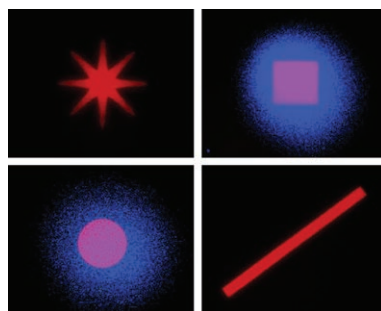
**Trick or treat:** Cytochrome P450<sub>B5B</sub> was transformed into a monooxygenase suitable for practical use by employing a simple substrate trick. The substrate specificity of P450<sub>B5B</sub> was altered drastically by a decoy molecule, while its intrinsic advantage, the use of hydrogen peroxide, was retained. The catalytic activities and the enantioselectivity of the H<sub>2</sub>O<sub>2</sub>–P450<sub>B5B</sub> system are highly dependent on the structure of the decoy molecule.

### Enzyme Catalysis

O. Shoji, T. Fujishiro, H. Nakajima,  
M. Kim, S. Nagano, Y. Shiro,  
Y. Watanabe\* — 3656–3659

Hydrogen Peroxide Dependent Monooxygenations by Tricking the Substrate Recognition of Cytochrome P450<sub>B5B</sub>

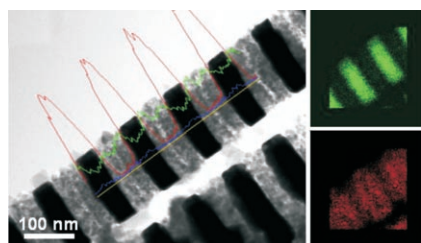
**Ship shape:** At what level of simplicity is emergent behavior possible? An emergent property can arise at the level of a biochemical reaction network. Hemostasis, which regulates blood clotting, can respond to the shape of a patch of surface stimulus. This response can be understood from chemical principles, as demonstrated with numerical simulations and a simplified nonbiological, chemical system.



### Enzyme Models

C. J. Kastrup, F. Shen,  
R. F. Ismagilov\* — 3660–3662

Response to Shape Emerges in a Complex Biochemical Network and Its Simple Chemical Analogue



**Stripped wires:** Multifunctional (magnetic and optical) iron–gold barcode nanowires were electrochemically fabricated using nanoporous templates. Structural analysis by TEM elemental line scan and mapping (see images) clearly revealed the well-separated, bamboo-like nanostructures composed of Fe and Au strips.

### Nanostructures

J. H. Lee, J. H. Wu,\* H. L. Liu, J. U. Cho,  
M. K. Cho, B. H. An, J. H. Min, S. J. Noh,  
Y. K. Kim\* — 3663–3667

Iron–Gold Barcode Nanowires

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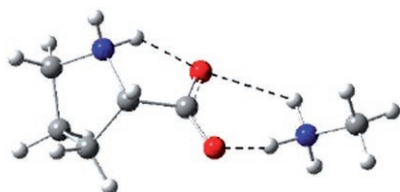


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**Separate charges:** Zwitterionic proline can be effectively stabilized by protonated methylamine in the gas phase (see calculated structure; C gray, H white, N blue, O red), as shown by accurate binding-energy measurements and IR fingerprint spectra. These results imply that organic ions may provide an effective means of stabilizing the zwitterionic structures of amino acids, which are of fundamental importance in biological systems.

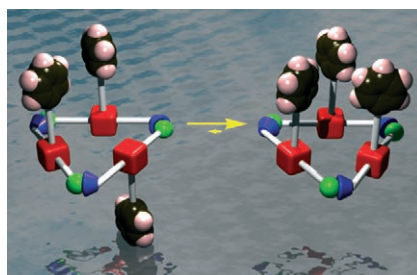
### Zwitterionic Structures

R. Wu, T. B. McMahon\* — 3668–3671

Stabilization of the Zwitterionic Structure of Proline by an Alkylammonium Ion in the Gas Phase



**Ring a ring of benzenes:** An unsubstituted N-confused metalloporphyrin forms both  $C_3$ - and  $C_1$ -symmetric trimers in solution, whereas its monophenyl-substituted analogue forms exclusively  $C_3$ -symmetric trimers (see picture) owing to significant stabilizing noncovalent interactions between the three phenyl groups, as evaluated both experimentally and theoretically.



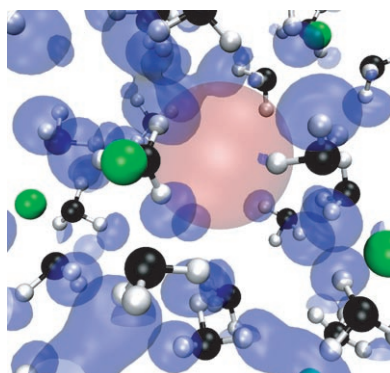
### Benzene Trimers

T. Morimoto, H. Uno, H. Furuta\* — 3672–3675

Benzene Ring Trimer Interactions Modulate Supramolecular Structures



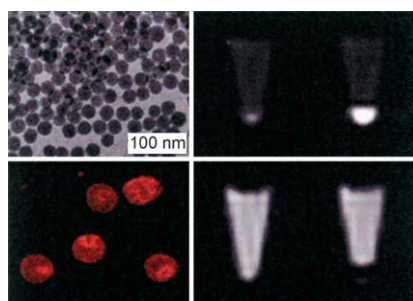
**Where are the solvated electrons** in strongly metallic lithium/ammonia solutions? Ab initio simulations show that they are not preferentially localized close to the void centers (red sphere) but accumulate at the void/solvent interface (blue isosurfaces). Furthermore, void formation itself can be reproduced by simply “stretching” pure ammonia, without any unbound electrons.



### Lithium in Ammonia

A. Chandra,\* D. Marx\* — 3676–3679

Creating Interfaces by Stretching the Solvent Is Key to Metallic Ammonia Solutions



**Lighting things up:** Multifunctional silica nanoparticles containing a luminescent core and a paramagnetic coat are prepared, and their utility as multimodal imaging probes is demonstrated in vitro. Monocyte cells can be labeled with the hybrid nanoparticles with greater than 98% efficiency and do not experience measurable toxicity even at a high loading of 0.123 mg per 5000 cells.

### Imaging Agents

W. J. Rieter, J. S. Kim, K. M. L. Taylor, H. An, W. Lin, T. Tarrant, W. Lin\* — 3680–3682

Hybrid Silica Nanoparticles for Multimodal Imaging



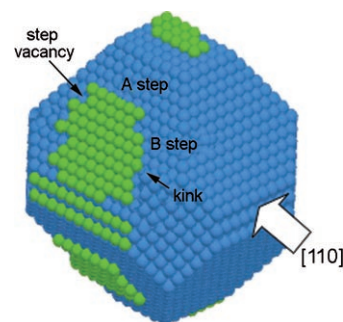


## Electron Microscopy

L. C. Gontard, L.-Y. Chang,  
C. J. D. Hetherington, A. I. Kirkland,  
D. Ozkaya,  
R. E. Dunin-Borkowski\* — **3683–3685**

Aberration-Corrected Imaging of Active  
Sites on Industrial Catalyst Nanoparticles

**Picture perfect:** Information about the local topologies of active sites on commercial nanoparticles can be gained with atomic resolution through spherical-aberration-corrected transmission electron microscopy (TEM). A powder of Pt nanoparticles on carbon black was examined with two advanced TEM techniques based on recent developments in hardware (aberration correction) and computation (exit wavefunction restoration).



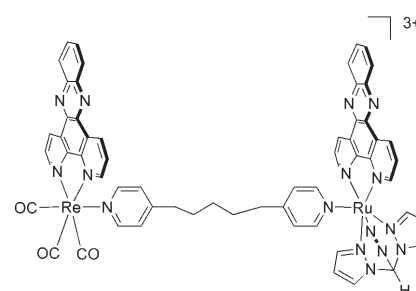
## DNA Intercalators

S. P. Foxon, T. Phillips, M. R. Gill,  
M. Towrie, A. W. Parker, M. Webb,  
J. A. Thomas\* — **3686–3688**



A Multifunctional Light Switch: DNA  
Binding and Cleavage Properties of a  
Heterobimetallic Ruthenium–Rhenium  
Dipyridophenazine Complex

**A DNA Swiss-army knife:** Photo- and biophysical studies on the hetero-dinuclear complex  $[\{Ru(tpm)(dppz)\}(\mu-dpp[5])\{fac-(CO)_3Re(dppz)\}]^{3+}$  (see structure; *tpm* = tris(1-pyrazolyl)methane; *dppz* = dipyrido[3,2-*a*:2',3'-*c*]phenazine; *dpp[5]* = 4,4'-dipyridylpentane) reveal that it is both a DNA light switch and a direct cleaving agent.

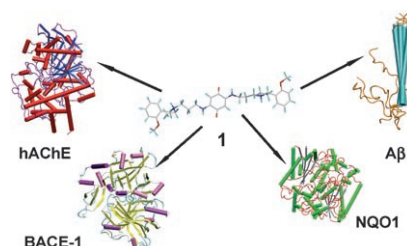


## Neurochemistry

A. Cavalli, M. L. Bolognesi, S. Capsoni,  
V. Andrisano, M. Bartolini, E. Margotti,  
A. Cattaneo, M. Recanatini,  
C. Melchiorre\* — **3689–3692**



A Small Molecule Targeting the  
Multifactorial Nature of Alzheimer's  
Disease



**Sights set on a cure:** Memoquin (**1**) affects several mechanisms relevant to Alzheimer's disease (AD): the formation of reactive oxygen species, the processing and aggregation of amyloid  $\beta$  ( $A\beta$ ) peptides, and acetylcholinesterase activity. In animal models, **1** causes a remarkable decrease in the formation of AD neurodegenerative hallmarks and a significant reversal of behavioral deficits. hAChE, BACE-1, and NQO1 are enzymes targeted by **1**.

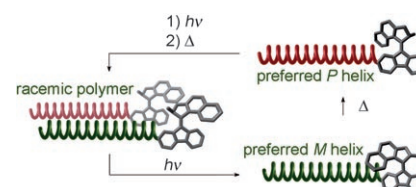
## Macromolecular Helicity

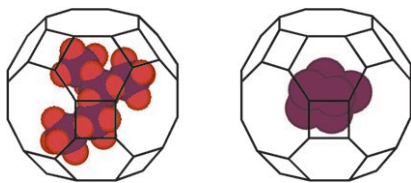
D. Pijper, B. L. Feringa\* — **3693–3696**



Molecular Transmission: Controlling the  
Twist Sense of a Helical Polymer with a  
Single Light-Driven Molecular Motor

**Twisted sisters:** Transmission of chirality from a light-driven rotary molecular motor to the macromolecular level of a polyisocyanate allows fully reversible control of the preferred helical sense of the polymer backbone (see picture).





**Trapped!** The title system (ca. 1 nm diameter; left) catalyzes methanol oxidation with higher turnover rates than clusters on SiO<sub>2</sub> supports. Spatial constraints lead to the preferential oxidation of methanol over larger alcohols. Restricted access to active sites also protects encapsulated Ru clusters (right) against inhibition of ethene hydrogenation by organosulfur compounds.

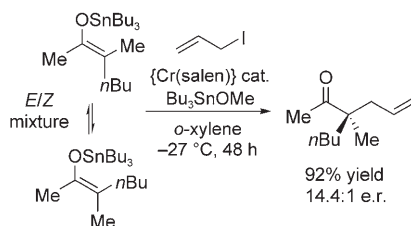
### Encapsulated Catalysts

B.-Z. Zhan, E. Iglesia\* — 3697–3700

RuO<sub>2</sub> Clusters within LTA Zeolite Cages: Consequences of Encapsulation on Catalytic Reactivity and Selectivity



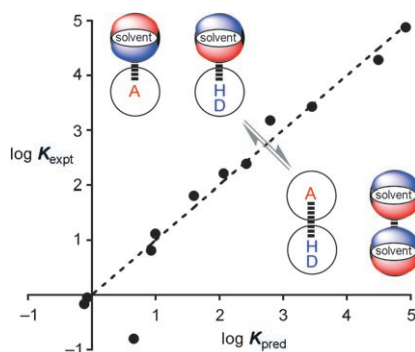
**Straighten out the mixture:** A dynamic mixture of acyclic isomers of tributyltin enolates undergoes a {Cr(salen)}-catalyzed alkylation reaction to generate methyl ketones that contain  $\alpha$ -carbonyl quaternary stereocenters in high yield and enantioselectivity (salen = *N,N'*-bis(salicylidene)ethylenediamine dianion). A mechanism is proposed which involves halide activation by a cationic metal complex.



### Asymmetric Catalysis

A. G. Doyle, E. N. Jacobsen\* — 3701–3705

Enantioselective Alkylation of Acyclic  $\alpha,\alpha$ -Disubstituted Tributyltin Enolates Catalyzed by a {Cr(salen)} Complex

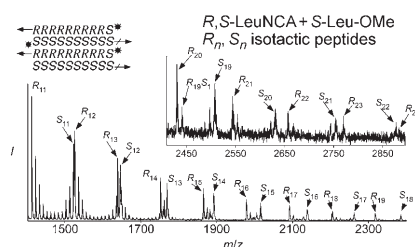


**Environmental issues:** The stability of the 1:1 hydrogen-bonded complex formed between tri-*n*-butylphosphine oxide (A) and perfluoro-*tert*-butyl alcohol (DH) has been measured in a range of solvents. The association constants ( $K_{\text{expt}}$ ) span five orders of magnitude and are predicted ( $K_{\text{pred}}$ ) remarkably accurately by a simple solvent competition model (see picture).

### Molecular Recognition

J. L. Cook, C. A. Hunter,\* C. M. R. Low, A. Perez-Velasco, J. G. Vinter — 3706–3709

Solvent Effects on Hydrogen Bonding



**A pattern on the sheets:** Long oligopeptides of homochiral sequence (see mass spectrum of enantioselectively deuterated compounds) were obtained in the polymerization of racemic *N*-carboxyanhydrides of valine (ValNCA) or leucine (LeuNCA) in aqueous solutions, through self-assembly into racemic antiparallel  $\beta$ -sheet templates.

### Templated Peptide Synthesis

I. Rubinstein, R. Eliash, G. Bolbach, I. Weissbuch, M. Lahav\* — 3710–3713

Racemic  $\beta$  Sheets in Biochirogenesis



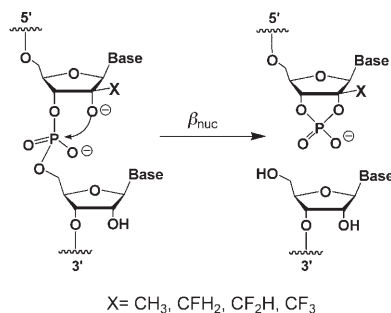


## RNA Cleavage

J.-D. Ye, N.-S. Li, Q. Dai,  
J. A. Piccirilli\* 3714–3717



The Mechanism of RNA Strand Scission:  
An Experimental Measure of the Brønsted  
Coefficient,  $\beta_{\text{nuc}}$



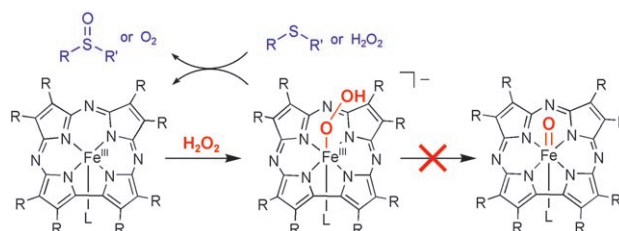
**A closer look at RNA cleavage:** A series of fluorine-bearing nucleotide analogues (X) were incorporated into RNA to perturb the  $\text{pK}_a$  value of the 2'-OH group systematically. These analogues enable the use of physical organic approaches to investigate bonding to the nucleophile during the RNA scission reaction. Measurement of the Brønsted coefficient,  $\beta_{\text{nuc}}$ , provides new insights into the transition-state structure for this classic reaction.

## Ferric Hydroperoxides

W. D. Kerber, B. Ramdhanie,  
D. P. Goldberg\* 3718–3721



$\text{H}_2\text{O}_2$  Oxidations Catalyzed by an Iron(III)  
Corrolazine: Avoiding High-Valent Iron–  
Oxido Species?



**Things aren't always what they seem:** The first iron corrolazine complex is shown to activate  $\text{H}_2\text{O}_2$  towards sulfide oxidation and peroxide dismutation. Although these

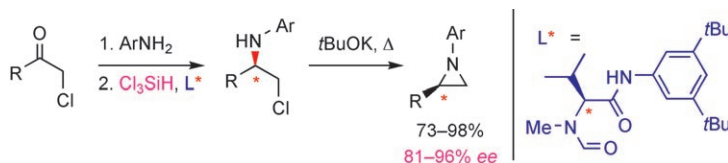
reactions are typical of a high-valent iron-oxido species, the data point to a ferric hydroperoxide species as the active oxidant (see scheme).

## Chiral Aziridines

A. V. Malkov,\* S. Stončius,  
P. Kočovský\* 3722–3724



Enantioselective Synthesis of 1,2-  
Diarylaziridines by the Organocatalytic  
Reductive Amination of  $\alpha$ -Chloroketones



**One-sided triangles:** A simple protocol has been developed for the synthesis of highly useful single enantiomers of 1,2-diarylaziridines. The method involves conversion of the readily available  $\alpha$ -chloroacetophenones into the corre-

sponding imines, followed by an enantioselective reduction with  $\text{Cl}_3\text{SiH}$  catalyzed by the L-valine-derived formamide  $L^*$  and ring closure to give the desired aziridines (see scheme).



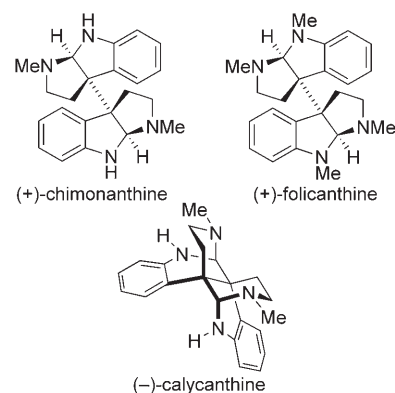
## Natural Products

M. Movassaghi,\*  
M. A. Schmidt 3725–3728

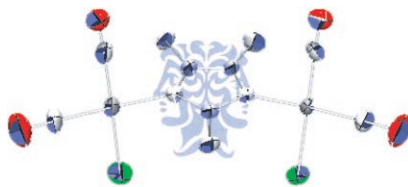


Concise Total Synthesis of (–)-Caly-  
canthine, (+)-Chimonanthine, and  
(+)-Folicanthine

**Conjoined twins:** An efficient and convergent strategy for the synthesis of the dimeric hexahydropyrroloindole alkaloids, (+)-chimonanthine, (+)-folicanthine, and (–)-calycanthine, is described. The simultaneous formation of the vicinal quaternary stereocenters by using a reductive dimerization reaction provides an access to the optically active key intermediate on a gram scale.



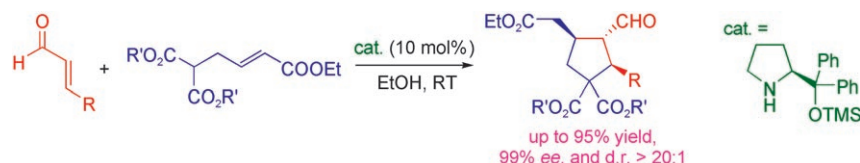
**Double trouble:** A series of complexes of rhodium and iridium has been obtained with a triazolediylidene ligand, which behaves as an extraordinarily versatile Janus-type ligand to form dinuclear complexes. The dinuclear complexes showed improved catalytic properties over their mononuclear counterparts. The picture shows a diiridium complex of 1,2,4-trimethyltriazole-3,5-diylidene (Ir gray, C white, N blue, O red, Cl green).



### N-Heterocyclic Carbenes

E. Mas-Marzá, J. A. Mata,  
E. Peris\* 3729–3731

Triazolediylidenes: A Versatile Class of Ligands for the Preparation of Discrete Molecules of Homo- and Hetero-Binuclear Complexes for Improved Catalytic Applications



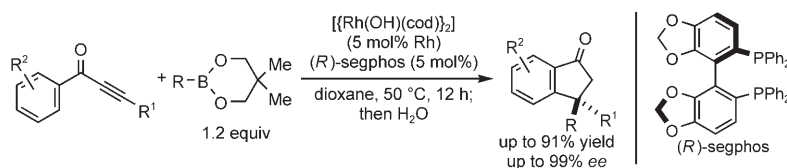
**Do a double take:** A novel highly enantio- and diastereoselective cascade double Michael reaction, in which two C–C bonds and three contiguous stereogenic centers are formed, has been developed. The one-

pot process, which was efficiently catalyzed by the chiral diphenylprolinyl trimethylsilyl (TMS) ether, is a facile approach to synthetically useful chiral cyclopentanes (see scheme).

### Organocatalysis

L. Zu, H. Li, H. Xie, J. Wang, W. Jiang,  
Y. Tang,\* W. Wang\* 3732–3734

Synthesis of Highly Functionalized Chiral Cyclopentanes by Catalytic Enantio- and Diastereoselective Double Michael Addition Reactions



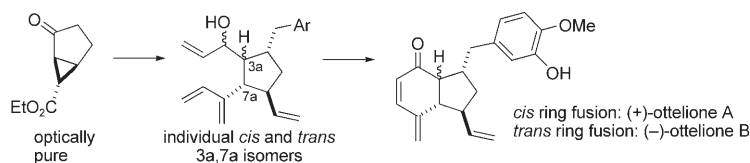
**Rapid access** to enantioenriched indanones that are difficult to obtain by other methods is gained through the highly enantioselective addition of aryl boronates to aryl alkynyl ketones under rhodium catalysis (see scheme). The

resulting 3,3-disubstituted 1-indanones can be converted into a range of potentially useful indan derivatives with high stereoselectivity. R<sup>1</sup> = Si(alkyl)<sub>3</sub>, GeEt<sub>3</sub>; R<sup>2</sup> = F, Me, OMe; cod = 1,5-cyclooctadiene.

### Asymmetric Catalysis

R. Shintani,\* K. Takatsu,  
T. Hayashi\* 3735–3737

Rhodium-Catalyzed Asymmetric Synthesis of 3,3-Disubstituted 1-Indanones



**To A and B:** The extremely powerful antitumor agents ottelione A and B have been synthesized from D-ribose. Key steps in the synthesis involve the use of a cyclopropane unit fused onto a five-

membered ring to control the stereochemistry and later to serve as a precursor to a vinyl group, and a highly regioselective ring-closing metathesis (see scheme).

### Natural Products

D. L. J. Clive,\* D. Liu 3738–3740

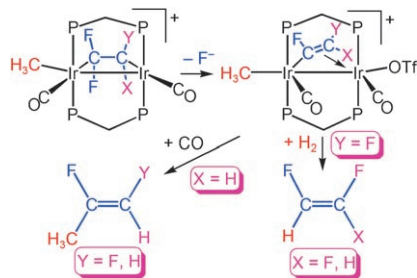
Synthesis of the Otteliones A and B: Use of a Cyclopropyl Group as Both a Steric Shield and a Vinyl Equivalent

## C–F Activation

D. J. Anderson, R. McDonald,  
M. Cowie\* 3741–3744



Carbon–Fluorine Bond Activation in Fluoroolefins: Clear Documentation of Cooperative C–F Bond Activation by Adjacent Metal Centers



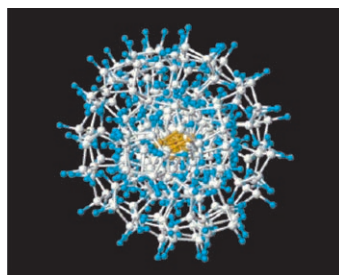
**F<sup>−</sup> off:** Fluoroolefins bridging a pair of iridium centers are susceptible to abstraction of a fluoride ion while those bound to a single metal center are not. The resulting bridging fluorovinyl groups also undergo fluoride abstraction to yield the corresponding vinylidenes, whereas terminal fluorovinyl groups are unreactive. Selective replacement of one fluorine substituent by either hydrogen or a methyl group can be effected (see scheme, Tf = trifluoromethanesulfonyl).

## Polymer Structures

C. Karafilidis, K. Angermund, B. Gabor,  
A. Ruffńska, R. J. Mynott,\*  
G. Breitenbruch, W. Thiel,  
G. Fink\* 3745–3749



Helical Microstructure of Polynorbornene



**Why don't you melt?** The zirconocene-catalyzed polymerization of norbornene proceeds by  $\sigma$ -bond metathesis to give 2-*exo*,7'-*syn* linkages in the main chain and 2-*exo*,2'-*exo*-linked mononorbornyl branches. The resulting helical structure is very compact (see picture, view along helical axis), which hinders conformational changes and explains the insolubility of the polynorbornene and why it cannot be melted.

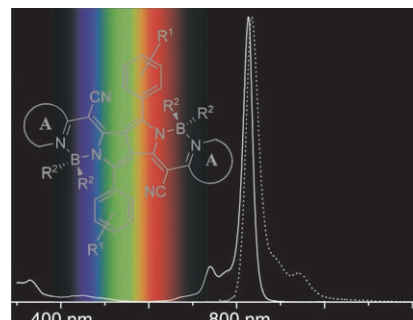
## NIR dyes

G. M. Fischer, A. P. Ehlers, A. Zumbusch,  
E. Daltrozzo\* 3750–3753



Near-Infrared Dyes and Fluorophores Based on Diketopyrrolopyrroles

**NIRly there:** The chromophore of compounds with intramolecular H bonds have been rigidified by substitution of the protons of the N–H...N bridges by BR<sub>2</sub> groups (R = F, Ph). This stiffening and the polymethine character of the chromophores affords dyes with intense fluorescence at room temperature and high NIR absorption, but virtually no absorption in the visible range (see the picture for an example, —: absorption, .....: fluorescence).

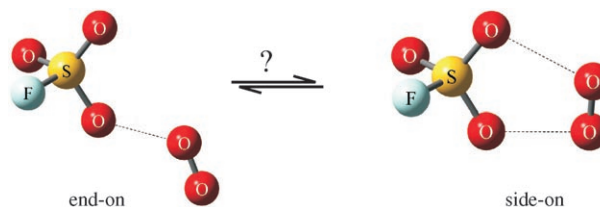


## Fluoroxy Radicals

H. Beckers, P. Garcia, H. Willner,\*  
G. A. Argüello, C. J. Cobos,  
J. S. Francisco\* 3754–3757

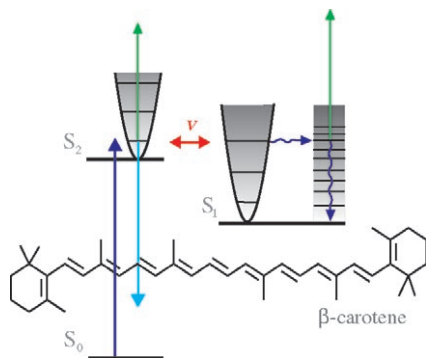


Side-On versus End-On Bonding of O<sub>2</sub> to the FSO<sub>3</sub> Radical: Matrix Isolation and Ab Initio Study of FSO<sub>3</sub>



**Quantum-chemical calculations** predict the existence of two almost equal-energy isomers of the FSO<sub>3</sub> radical with either side-on or end-on bonding of O<sub>2</sub> to FSO<sub>3</sub> (see picture). The structure of this polyoxy

radical isolated in solid noble-gas matrices and formed by the reaction of FSO<sub>3</sub> with molecular oxygen has now been revealed by IR and UV spectroscopic investigations.



**What happens next?** Oscillatory dynamics during internal conversion of  $\beta$ -carotene from the “bright”  $S_2$  to the “dark”  $S_1$  electronic state is observed by femto-second broadband transient absorption and fluorescence. The spectral evolution was described quantitatively by modeling the quantum dynamics in a three-level system, and the vibronic coupling element was determined ( $V \approx 95 \text{ cm}^{-1}$ ).

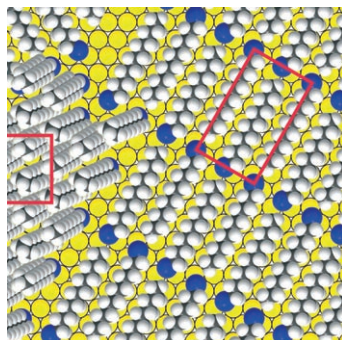
### Femtochemistry

J. L. Pérez Lustres,\* A. L. Dobryakov, A. Holzwarth, M. Veiga — 3758–3761

$S_2 \rightarrow S_1$  Internal Conversion in  $\beta$ -Carotene: Strong Vibronic Coupling from Amplitude Oscillations of Transient Absorption Bands



**Lying low:** Alkanethioacetates do not form the same dense thiolate monolayers on gold that are obtained from alkanethiols. A multitechnique study shows instead the formation of a kinetically stable monolayer with mostly flat-lying thiolate molecules (see model; blue S). This extraordinary structure results from the diminished reactivity of the thioacetate group towards the gold surface.



### Self-Assembled Monolayers

M. G. Badin, A. Bashir, S. Krakert, T. Strunskus, A. Terfort,\* C. Wöll\* — 3762–3764

Kinetically Stable, Flat-Lying Thiolate Monolayers



Supporting information is available on the WWW (see article for access details).



A video clip is available as Supporting Information on the WWW (see article for access details).

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