



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 soon:

N. Christinat, R. Scopelliti, K. Severin*

Multicomponent Assembly of Boronic Acid Based Macrocycles and Cages

C. Li, L. Qi

Bio-Inspired Fabrication of 3D Ordered Macroporous Single Crystals of Calcite via a Transient Amorphous Phase

J. Zhuang, H. Wu, Y. Yang, Y. C. Cao*

Controlling Colloidal Superparticle Growth Through Solvophobic Interactions

C. Coleman, D. van der Spoel*

Picosecond Melting of Ice by an Infrared Laser Pulse: A Simulation Study

Foldamers

Stefan Hecht, Ivan Huc

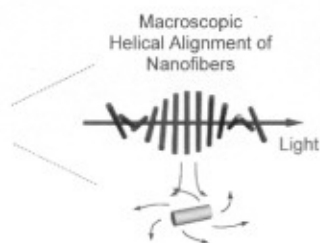
Fullerenes

Fernando Langa, Jean-François Nierengarten

Books

reviewed by S. Gellman _____ 632

reviewed by T. Akasaka _____ 633



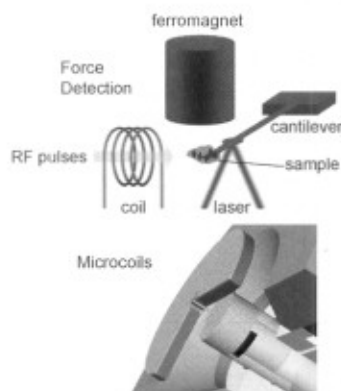
Turn, turn, turn: Achiral supramolecular assemblies are able to align by convective flows or by a vortex flow generated by mechanical stirring in dilute solutions (see picture). The linear dichroism and the linear birefringence created by the alignment result in strong circular dichroism signals.

Highlights

Macroscopic Chirality

G. P. Spada* _____ 636–638

Alignment by the Convective and Vortex Flow of Achiral Self-Assembled Fibers Induces Strong Circular Dichroism Effects



Impressively sensitive: To meet the challenges of miniaturization and limited sample quantity, new approaches for substantially increasing the sensitivity of NMR spectroscopy for gases, liquids, and solids have been developed (for two examples, see picture). These and future developments will assure that NMR spectroscopy remains a major tool for chemistry and other fields for years to come.

NMR Spectroscopy

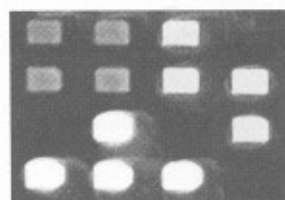
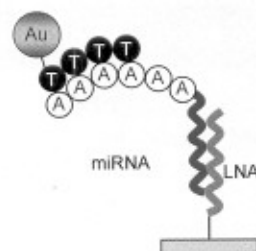
H. W. Spiess* _____ 639–642

NMR Spectroscopy: Pushing the Limits of Sensitivity

Minireviews

A. W. Wark, H. J. Lee,
R. M. Corn* 644-652

Multiplexed Detection Methods for Profiling MicroRNA Expression in Biological Samples



Good things come in small packages: MicroRNA molecules (miRNAs) have recently been discovered to be important regulators of gene expression, and aberrant miRNA expression patterns are increasingly being linked to a variety of

diseases, including cancer. Owing to the special features of miRNAs, the development of quantitative multiplex methods for miRNA profiling is a highly challenging task for bioorganic and analytical chemists. LNA=locked nucleic acid.

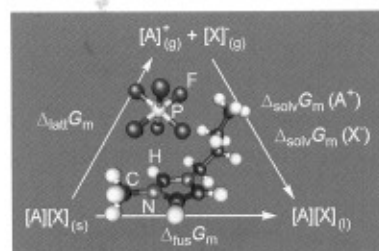
Reviews

Ionic Liquids

H. Weingärtner* _____ 654–670

Understanding Ionic Liquids at the Molecular Level: Facts, Problems, and Controversies

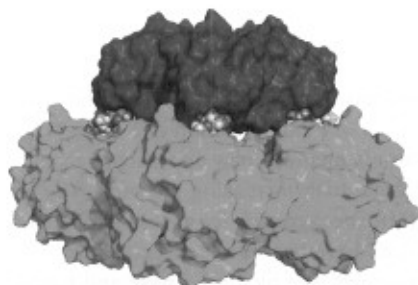
IL-informed? What is actually known about ionic liquids (ILs)? The Review addresses this question from the physicochemical stand point and the molecular basis for the properties of ILs is discussed. The picture shows a cycle for calculating the molar Gibbs energy of fusion, $\Delta_{\text{fus}}G_m$, of an IL from the lattice Gibbs energy, $\Delta_{\text{lat}}G_m$, and the Gibbs energy of solvation, $\Delta_{\text{sol}}G_m$.



Communications

Inhibitors

P. I. Kitov, T. Lipinski, E. Paszkiewicz,
D. Solomon, J. M. Sadowska, G. A. Grant,
G. L. Mulvey, E. N. Kitova, J. S. Klassen,
G. D. Armstrong,
D. R. Bundle* _____ 672-676



A compact heterobifunctional ligand devoid of a linker between binding functionalities induces a supramolecular assembly between two pentameric proteins, Shiga toxin (blue) and serum amyloid P component, a human serum protein (turquoise). The use of an endogenous protein as a template brings about a 10000-fold enhancement of ligand activity.

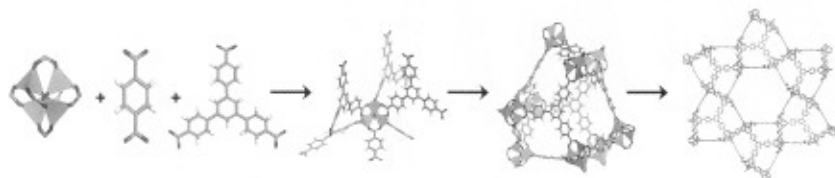


An Entropically Efficient Supramolecular Inhibition Strategy for Shiga Toxins

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electronic / print or electronic delivery); for individuals who are personal members of a national chemical society prices are available on request. Postage and handling charges included. All prices are subject to local VAT/sales tax.



Mixing it up: UCMC-1 (University of Michigan Crystalline Material-1), a mesoporous material with unprecedented levels of microporosity, arises from the coordination copolymerization of a dicarboxylate and a tricarboxylate linker mediated by zinc (see picture; blue tetrahedra Zn₄O clusters, C gray, H white, O red). Exceptionally high surface area derives from the microporous cages lining the 1D mesoporous channels.

Coordination Polymers

K. Koh, A. G. Wong-Fill, A. J. Matzger* 677–680

A Crystalline Mesoporous Coordination Copolymer with High Microporosity



Round the twist: Metalation of [36]octaphyrin 1 provided the Möbius aromatic Pd₂ complex 3 as well as the Hückel antiaromatic Pd₂ complex 2. This method can be applied to other expanded porphyrins and Group 10 metal ions. The aromatic/antiaromatic character was supported by NMR spectroscopy, NICS calculation, and two-photon absorption measurements.

phyrins and Group 10 metal ions. The aromatic/antiaromatic character was supported by NMR spectroscopy, NICS calculation, and two-photon absorption measurements.

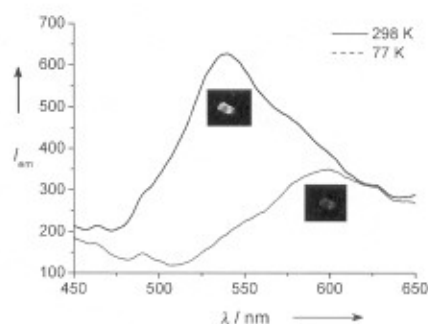
Aromaticity

Y. Tanaka, S. Saito, S. Mori, N. Aratani, H. Shinokubo, N. Shibata, Y. Higuchi, Z. S. Yoon, K. S. Kim, S. B. Noh, J. K. Park, D. Kim,* A. Osuka* 681–684

Metalation of Expanded Porphyrins: A Chemical Trigger Used To Produce Molecular Twisting and Möbius Aromaticity



Temperature-dependent Cu...Cu distances lead to the luminescence thermochromism of copper(I) coordination polymers with cubane-like Cu₄X₄ cluster nodes (X = halogen) according to a study on three copper(I) coordination polymers with a bis-thioether ligand (L). The picture shows solid-state luminescence spectra and photographs of the two-dimensional network polymer [Cu₄I₄L₂]_n at 298 and 77 K.



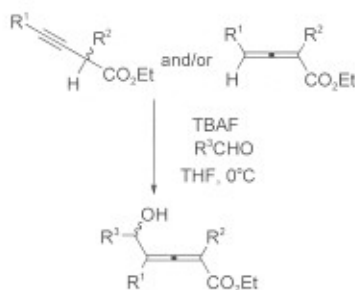
Coordination Polymers

T. H. Kim, Y. W. Shin, J. H. Jung, J. S. Kim, J. Kim* 685–688

Crystal-to-Crystal Transformation between Three Cu^I Coordination Polymers and Structural Evidence for Luminescence Thermochromism



Retroaldol regains control: A counteranion may exert an important effect in promoting thermodynamic control in the TBAF-mediated aldol reaction of propargyl or allenyl esters (or a mixture of both) to produce, under mild conditions, carbinol allenates exclusively (see scheme; TBAF = tetra-*n*-butylammonium fluoride).



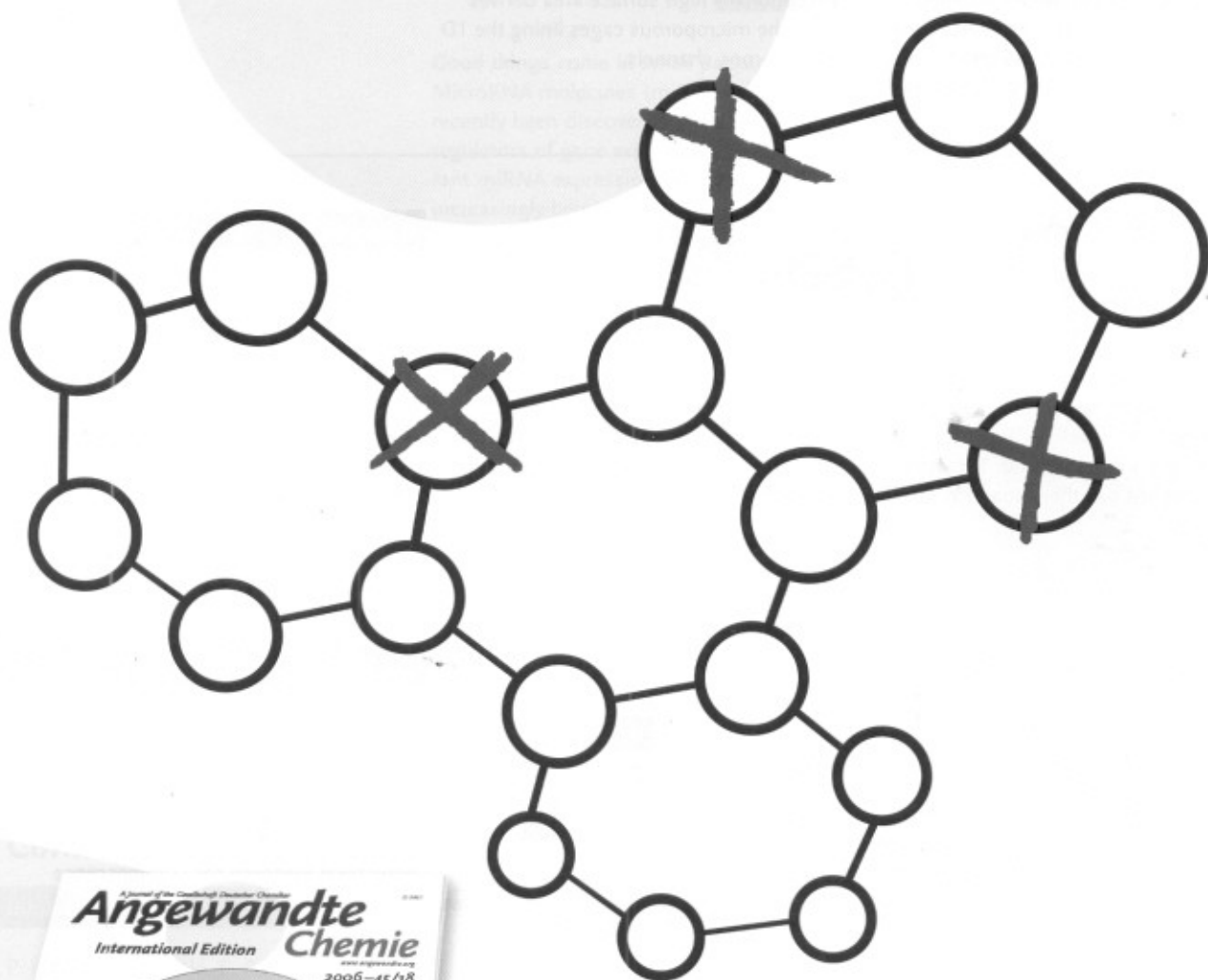
Aldol Reactions

B. Xu, G. B. Hammond* 689–692

Thermodynamically Favored Aldol Reaction of Propargyl or Allenyl Esters: Regioselective Synthesis of Carbinol Allenates



Incredibly selective!



Angewandte Chemie chooses its articles carefully. Most of its Reviews, Highlights, and Essays are written upon invitation, from authors who are among the very best in their fields. Just 30 % of all submitted Communications in 2006 were accepted after peer review - only about 1400 from nearly 5000. Communications that are judged to be exceptionally important within a particular field are featured as Very Important Papers (VIPs).

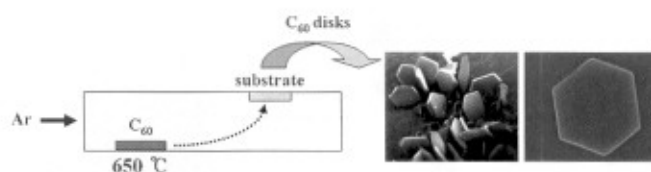
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New jobs for disk jockeys: Single-crystalline C_{60} disks were selectively synthesized on an HOPG substrate by a vapor–solid process with C_{60} powder as the precursor. The photoluminescence intensity of the disks is much greater than that of thin

films and powders. In photoconductivity measurements with a single disk the current was increased by about tenfold. These properties point to intriguing applications.

C_{60} Structures

H. S. Shin, S. M. Yoon, Q. Tang, B. Chon, T. Joo, H. C. Choi* 693–696

Highly Selective Synthesis of C_{60} Disks on Graphite Substrate by a Vapor–Solid Process



Pièces de résistance: The human immunodeficiency virus (HIV) can acquire drug resistance by mutating the key enzyme HIV protease that is being used as a drug target. A strategy has been developed to predict drug resistance that focuses on

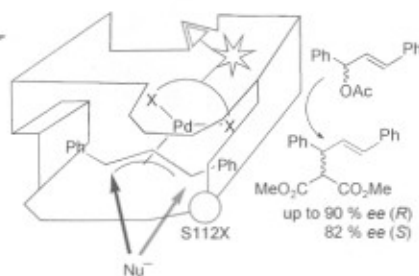
the fact that the virus must retain reasonable efficiency for its catalytic reaction while reducing its affinity to the given drug (γ_M : vitality values for the mutant protein M, K_i : inhibition constant).

Drug Resistance

H. Ishikita, A. Warshel* 697–700

Predicting Drug-Resistant Mutations of HIV Protease

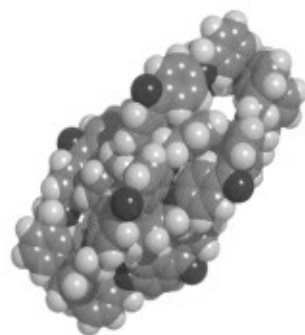
Palladium in the active site: The incorporation of a biotinylated palladium diphosphine within streptavidin yielded an artificial metalloenzyme for the title reaction (see scheme). Chemogenetic optimization of the catalyst by the introduction of a spacer (red star) between biotin (green triangle) and palladium and saturation mutagenesis at position S112X afforded both *R*- and *S*-selective artificial asymmetric allylic alkylases.



Asymmetric Catalysis

J. Pierron, C. Malan, M. Creus, J. Gradinaru, I. Hafner, A. Ivanova, A. Sardo, T. R. Ward* 701–705

Artificial Metalloenzymes for Asymmetric Allylic Alkylation on the Basis of the Biotin–Avidin Technology



Making up a foursome: Self-assembly of 4,4'-(3-pyridinemethoxy)benzophenone (L) and $Pd(NO_3)_2$ resulted in the quantitative formation of a quadruply stranded metallohelicate $[Pd_2(L)_4]$, which undergoes spontaneous dimerization to an unprecedented chiral interlocked metallohelicate $[Pd_2(L)_4]_2$ (see X-ray structure; C black, H white, N blue, O red).

Supramolecular Chemistry

M. Fukuda, R. Sekiya,* R. Kuroda* 706–710

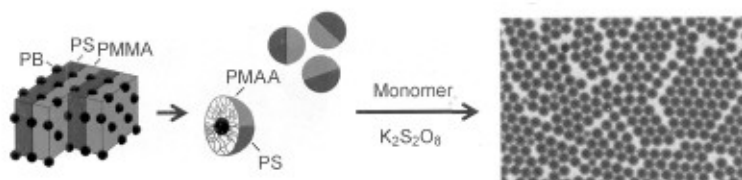
A Quadruply Stranded Metallohelicate and Its Spontaneous Dimerization into an Interlocked Metallohelicate

Emulsion Polymerization

A. Walther, M. Hoffmann,
A. H. E. Müller* 711–714



Emulsion Polymerization Using Janus
Particles as Stabilizers



Two-faced particles: The emulsion polymerization of monomers in the presence of Janus particles, which combine the Pickering effect with amphiphilicity, leads to well-defined and stable latex dispersions (see picture). Only a few Janus

beads are necessary on a latex particle surface to sufficiently stabilize the dispersion against coagulation. PB = polybutadiene, PS = polystyrene, PMMA = poly(methyl methacrylate), PMAA = poly(methacrylic acid).

Iron-Mediated Amination

F. Avenier, E. Gouré, P. Dubourdeaux,
O. Sénèque, J.-L. Oddou, J. Pécaut,
S. Chardon-Noblat, A. Deronzier,
J.-M. Latour* 715–717



Multiple Aromatic Amination Mediated by
a Diiron Complex

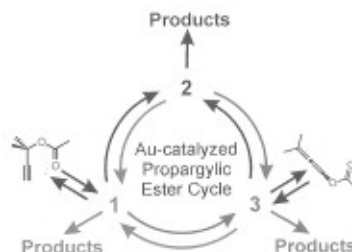


Stop-frame amination: A very efficient amine transfer reaction is mediated by a biomimetic diiron complex, giving both single and double insertions of a tosylamine group into a pendant benzyl group of the ligand (see structure). The mono-insertion product has been shown by X-ray crystallography and other physical methods to be a tosylanilinato diiron(III) complex.

Reaction Mechanisms

A. Correa, N. Marion, L. Fensterbank,
M. Malacria, S. P. Nolan,
L. Cavallo* 718–721

Golden Carousel in Catalysis: The
Cationic Gold/Propargylic Ester Cycle



Round and round it goes: Propargylic esters are versatile substrates for Au-based catalysts. However, under typical conditions the starting Au-coordinated propargylic ester **1** is in rapid equilibrium

with the gold vinylic carbenoid species **2** and with gold allene species **3**. A number of factors dictate which intermediate is lower in energy and which type of products form.

Protein Modifications

J. Guo, J. Wang, J. C. Anderson,
P. G. Schultz* 722–725

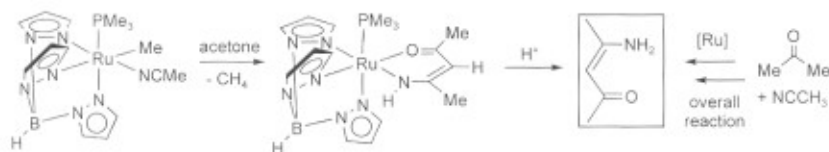


Addition of an α -Hydroxy Acid to the
Genetic Code of Bacteria



Playing tag: The α -hydroxy acid *p*-hydroxy-L-phenyllactic acid has been genetically incorporated into proteins in *E. coli* in response to an amber nonsense codon. The site-specific introduction of

backbone ester mutations was used for the site-specific hydrolysis of an affinity tag, as well as for determination of the energetic contributions of backbone hydrogen bonds to protein stability.



Better H than N: The Ru^{II} fragment $\{\text{TpRu}(\text{PMe}_3)\text{Me}\}$ cleanly activates the sp^3 C–H bonds of functionalized substrates in preference to reactivity with the heterofunctional groups. C–H activation of acetonitrile, acetone, and nitromethane

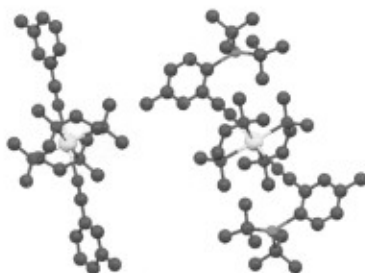
releases CH_4 with subsequent intramolecular C–C or C–N bond-forming reactions. For example, acetone and acetonitrile are converted into free enamine (see scheme).

C–H Activation

N. A. Foley, T. B. Gunnoe,* T. R. Cundari,*
P. D. Boyle, J. L. Petersen — 726–730

Activation of sp^3 Carbon–Hydrogen Bonds by a Ruthenium(II) Complex and Subsequent Metal-Mediated C–C and C–N Bond Formation

Zany zincation: Seemingly simple zincation reactions of aromatic or aliphatic nitriles are in reality highly complicated affairs involving separated ion pairs (see X-ray structure; yellow Na, blue N, green Zn; left cation, right anion) or joined ion pairs. The results also show that the free zincate anions can engage in complicated dismutation processes.

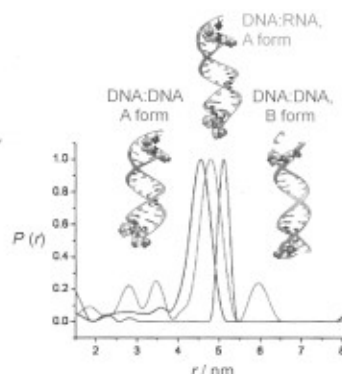


Zincation

W. Clegg, S. H. Dale, E. Hevia, L. M. Hogg,
G. W. Honeyman, R. E. Mulvey,*
C. T. O'Hara, L. Russo — 731–734

Structurally Defined Reactions of Sodium TMP–Zincate with Nitrile Compounds: Synthesis of a Salt-Like Sodium Sodiumzincate and Other Unexpected Ion-Pair Products

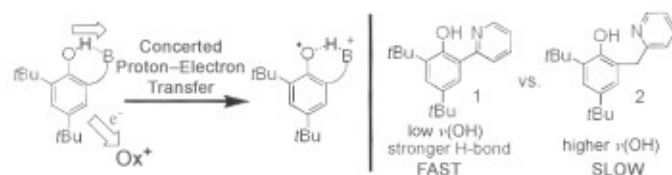
Oh DEER! The DEER technique is capable of detecting even slight conformational changes in site-directed spin-labeled DNA structures, which are extremely difficult to measure by more classical methods. The B–A transition (see picture) in DNA duplexes is induced by trifluoroethanol. DEER can be applied to the study of modifications of DNA induced by various agents.



DNA Structures

G. Sicoli, G. Mathis, O. Delalande,
Y. Boulard, D. Gasparutto,
S. Gambarelli* — 735–737

Double Electron–Electron Resonance (DEER): A Convenient Method To Probe DNA Conformational Changes



Slowed down by a break up: One-electron oxidation of intramolecularly hydrogen-bonded phenol–pyridine compounds **1** and **2** involves proton transfer coupled with electron transfer (see scheme; B = base). At the same driving force, **2** reacts substantially slower than **1**. The

methylene group between the phenol and pyridine groups in **2** breaks the conjugation between the rings, which has a marked effect on the hydrogen bond and is likely the origin of the difference in rate constants.

Concerted Proton–Electron Transfer

T. F. Markle, J. M. Mayer* — 738–740

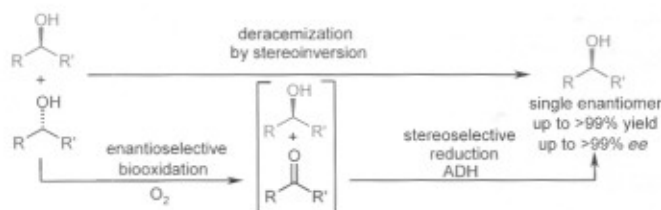
Concerted Proton–Electron Transfer in Pyridylphenols: The Importance of the Hydrogen Bond

Biocatalytic Deracemization

C. V. Voss, C. C. Gruber,
W. Kroutil* 741–745



Deracemization of Secondary Alcohols through a Concurrent Tandem Biocatalytic Oxidation and Reduction



Breaking the mirror: A purified alcohol dehydrogenase (ADH) for stereoselective reduction and whole cells of a microorganism for enantioselective oxidation operated concurrently to effect the ste-

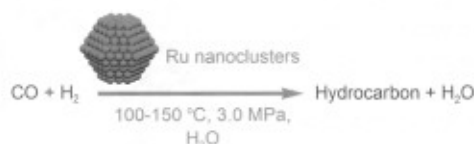
reoinversion of one enantiomer of a racemic secondary alcohol and provide the optically pure alcohol in > 99% yield (see scheme). R, R' = alkyl.

Heterogeneous Catalysis

C.-X. Xiao, Z.-P. Cai, T. Wang, Y. Kou,*
N. Yan* 746–749



Aqueous-Phase Fischer–Tropsch Synthesis with a Ruthenium Nanocluster Catalyst



No need for support: An aqueous-phase Fischer–Tropsch synthesis with a high turnover frequency of 12.9 h^{−1} has been realized over a ruthenium nanocluster catalyst (with particle diameter of 2.0 nm)

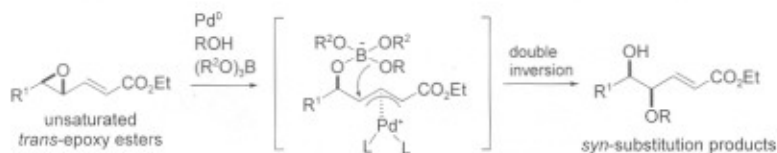
at 150 °C. This catalyst, in the absence of any support, achieves an activity that exceeds those of conventional supported catalysts.

Homogeneous Catalysis

X.-Q. Yu, F. Yoshimura, F. Ito, M. Sasaki,
A. Hirai, K. Tanino,
M. Miyashita* 750–754



Palladium-Catalyzed Stereospecific Substitution of α,β -Unsaturated γ,δ -Epoxy Esters by Alcohols with Double Inversion of Configuration: Synthesis of 4-Alkoxy-5-hydroxy-2-pentenoates



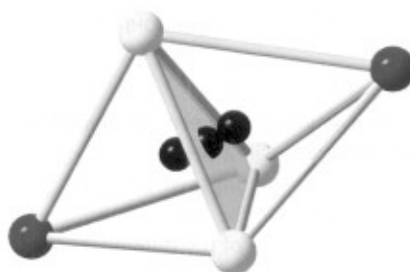
Accessing the inaccessible: Highly functionalized compounds that were previously inaccessible can be readily prepared in a highly stereoselective manner, in high yields, and even in chiral forms by the palladium-catalyzed substitution of

α,β -unsaturated γ,δ -epoxy esters with various alcohols (see scheme). The products formed can serve as versatile synthetic intermediates for further transformations.

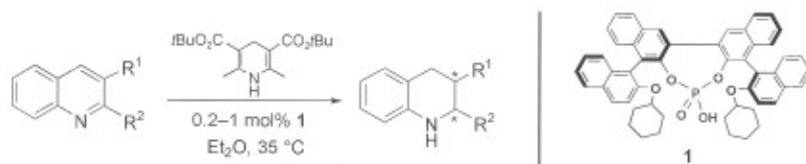
Lithium Ion Conductors

H.-J. Deiseroth,* S.-T. Kong, H. Eckert,
J. Vannahme, C. Reiner, T. Zaiß,
M. Schlosser 755–758

Li₆PS₅X: A Class of Crystalline Li-Rich Solids With an Unusually High Li⁺ Mobility

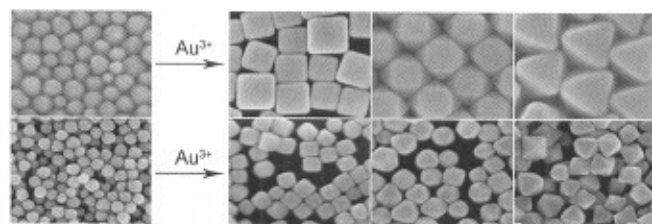


Mobile metal ions: Halide-substituted lithium argyrodites form a new class of Li-rich solids with an unusually high Li mobility. Single-crystal X-ray studies (see picture; Li black, S yellow, I red) at room temperature and MAS-NMR measurements in a wide temperature range provide insights into the Li⁺ ion dynamics.



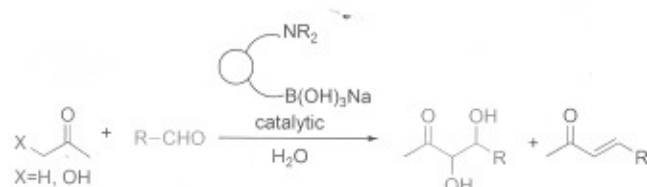
Building a better scaffold: Low loadings (0.2–1 mol%) of new double axially chiral phosphoric acid catalysts **1** based on bisbinol scaffold were used for asymmetric transfer hydrogenation. 2-Aryl- and 2-alkyl-substituted quinolines gave tetrahydro-

quinolines in excellent yields and with up to 98% *ee* and 2,3-disubstituted tetrahydroquinolines were prepared in high diastereo- and enantioselectivities (up to > 20:1 and 92% *ee*).



Shapeshifters: The shape and size of gold nanocrystals were controlled simultaneously through directed surface overgrowth from polyhedral and spherical seeds of different sizes. The resulting cubes, cuboctahedra, and octahedra

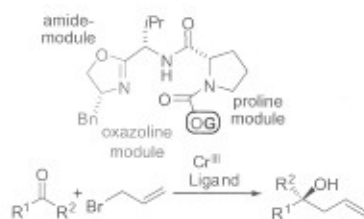
(shown as SEM images for growth from spherical seeds of two sizes) exhibited characteristic optical properties in the visible range, which were analyzed by discrete dipole approximation calculations.



Cooperation is key: *N*-Butyl-1-benzimidazole-2-phenylboronic acid hydroxide complex catalyzes the aldol condensation and aldol addition between hydroxyacetone or acetone, and different aldehydes in water. The catalytic activity results from coop-

erative interactions between the boronate complex and the imidazole function. Aldol condensation gives the unsaturated methyl ketones of acetone, whereas aldol addition predominates with hydroxyacetone.

Is bigger better? By systematically evaluating the size of the substituent *G* in a modular ligand (see picture), a linear free energy relationship was observed relating the product enantiomeric ratio to steric parameters developed by Charton. The reaction studied was the addition of allyl bromide to three different carbonyl substrates.



Asymmetric Catalysis

Q.-S. Guo, D.-M. Du,* J. Xu — 759–762

The Development of Double Axially Chiral Phosphoric Acids and Their Catalytic Transfer Hydrogenation of Quinolines

Gold Nanocrystals

D. Seo, C. I. Yoo, J. C. Park, S. M. Park, S. Ryu,* H. Song* — 763–767

Directed Surface Overgrowth and Morphology Control of Polyhedral Gold Nanocrystals

Aqueous Catalysis

K. Aelvoet, A. S. Batsanov, A. J. Blatch, C. Grosjean, L. G. F. Patrick, C. A. Smethurst, A. Whiting* — 768–770

A Catalytic Aldol Reaction and Condensation through In Situ Boron "Ate" Complex Enolate Generation in Water

Enantioselectivity

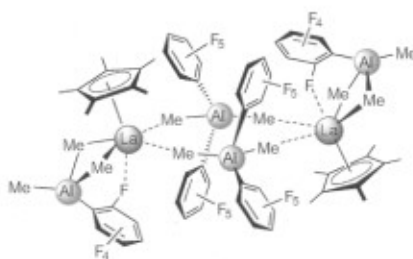
J. J. Miller, M. S. Sigman* — 771–774

Quantitatively Correlating the Effect of Ligand-Substituent Size in Asymmetric Catalysis Using Linear Free Energy Relationships

Polymerization

M. Zimmermann, K. W. Törnroos,
R. Anwander* 775–778

-  Cationic Rare-Earth-Metal Half-Sandwich Complexes for the Living *trans*-1,4-Isoprene Polymerization

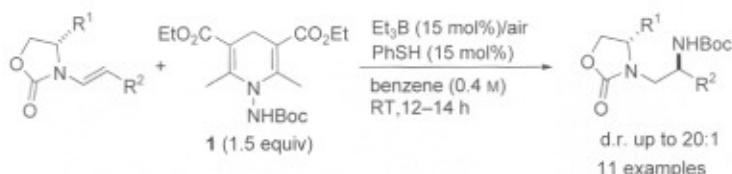


Rare, but well done! Fluorinated borates $[\text{Ph}_3\text{C}][\text{B}(\text{C}_6\text{F}_5)_4]$ and $[\text{PhNMe}_2\text{H}][\text{B}(\text{C}_6\text{F}_5)_4]$ and the borane reagent $\text{B}(\text{C}_6\text{F}_5)_3$ activate rare-earth-metal bis(tetramethylaluminate) complexes in a distinct and efficient manner (an activated catalyst is shown in the picture) for the fabrication of synthetic gutta-percha (99.5% *trans*-1,4-polyisoprene, $M_n = 0.8\text{--}4.4 \times 10^5 \text{ g mol}^{-1}$, $M_n/M_w = 1.18$).

Synthetic Methods

J. Guin, R. Fröhlich, A. Studer* 779–782

-  Thiol-Catalyzed Stereoselective Transfer Hydroamination of Olefins with N-Aminated Dihydropyridines



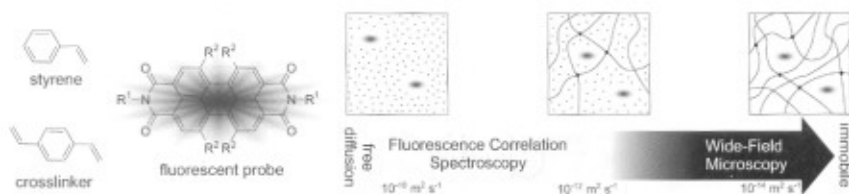
Hantzsch dihydropyridines are popular reducing reagents in organocatalysis. The N-aminated Hantzsch dihydropyridine **1** has now been used as a novel efficient precursor for Boc-protected carbamoyl radicals. The readily prepared reagent was

employed for the radical-transfer hydroamination of various olefins. Stereoselective hydroamination of chiral enecarbamates afforded vicinal diamines in good stereoselectivities. Boc = *tert*-butoxycarbonyl.

VIP Polymerization

D. Wöll, H. Uji-i, T. Schnitzler, J.-I. Hotta,
P. Dedecker, A. Herrmann,
F. C. De Schryver, K. Müllen,
J. Hofkens* 783–787

-  Radical Polymerization Tracked by Single Molecule Spectroscopy



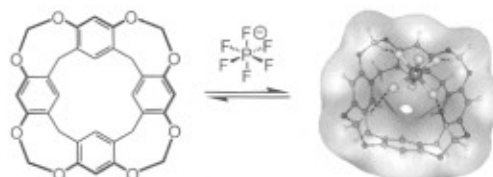
Polymerization—a single molecule view: Molecular motion of free and incorporated single perylene diimide derivatives during radical polymerization of styrene and styrene networks, studied by a combination of fluorescence correlation spec-

troscopy and wide-field microscopy, can be observed over the entire conversion range from free diffusion to immobilization of the dyes. In particular, the presence of evolving heterogeneities could be visualized.

Weak Interactions

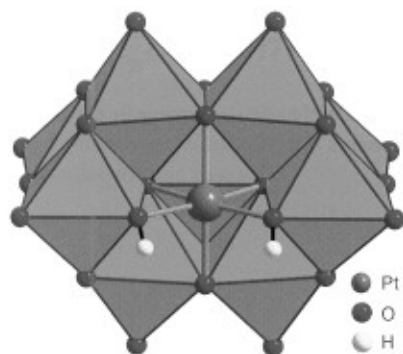
S. S. Zhu, H. Staats, K. Brandhorst,
J. Grunenberg, F. Gruppi, E. Dalcanele,
A. Lützen, K. Rissanen,
C. A. Schalley* 788–792

-  Anion Binding to Resorcinarene-Based Cavities: The Importance of C–H⋯Anion Interactions



Anions rather than cations: While resorcinarenes bind cations through cation– π interactions, methylene-bridged cavities surprisingly bind anions through C–H⋯anion interactions with the acetal

protons at the wider rim (see model of the complex in the scheme). Mass spectrometry, in agreement with theory, provides evidence for anion binding on the concave side of the cavitand bowl.



A new mate for vanadate: The platinum(IV)-containing polyoxovanadate $[\text{H}_2\text{Pt}^{\text{IV}}\text{V}_9\text{O}_{28}]^{5-}$ (**1**) has been synthesized and structurally characterized (see polyhedral representation). The characterization of polyanion **1** in solution by ^{195}Pt NMR spectroscopy is unprecedented in POM chemistry. The facile synthetic procedure with the Pt^{IV} precursor $\text{H}_2\text{Pt}(\text{OH})_6$ appears to be a convenient and general methodology for making a large variety of Pt^{IV} -containing polyoxometalates.

Pt-Containing Polyoxometalates

U. Lee,* H.-C. Joo, K.-M. Park, S. S. Mal, U. Kortz,* B. Keita, L. Nadjro — 793–796

Facile Incorporation of Platinum(IV) into Polyoxometalate Frameworks: Preparation of $[\text{H}_2\text{Pt}^{\text{IV}}\text{V}_9\text{O}_{28}]^{5-}$ and Characterization by ^{195}Pt NMR Spectroscopy



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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