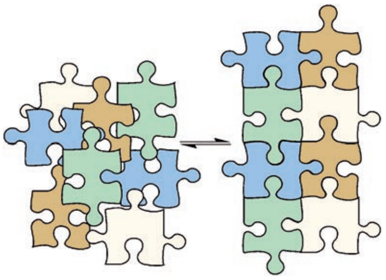
VIP *The following Communications have been judged by at least two referees to be "very important papers" and will be published online at www.angewandte.org soon:*

News

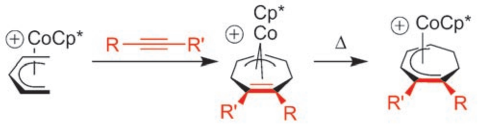
Books

Highlights

Solving the puzzle of how to piece together the building blocks of a multicomponent aggregate should become easier by an improved mastery of reversible orthogonal interactions. In this way the complexity and emergence of artificial multicomponent assemblies may approach those of natural systems.



Reversible Interactions



The magnificent seven: [4+3] cyclizations of unsaturated building blocks to cycloheptane derivatives have been established for some time; now [3+2+2] and more recently [5+2] cyclizations open new routes to cycloheptane derivatives (see scheme: $Cp^* = C_5Me_5$).

Cycloheptane Syntheses

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Contents

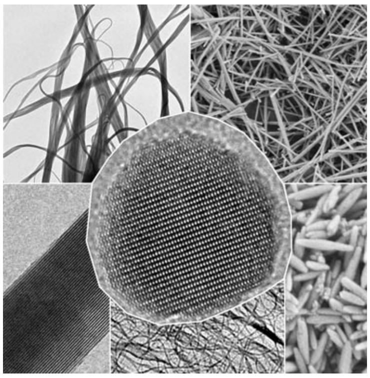
Minireviews

Sol–Gel Processes

N. Pinna,*
M. Niederberger* ________ **5292 – 5304**

Surfactant-Free Nonaqueous Synthesis of Metal Oxide Nanostructures

As few starting materials as possible: Metal oxide nanoparticles and metal-oxide-based organic–inorganic hybrid nanostructures can be prepared in organic solvents under exclusion of water and in the absence of surfactants. Selected chemical formation mechanisms, advantages and limitations of these approaches in comparison to surfactant-directed routes, potential applications and technological aspects are briefly discussed in this Minireview.

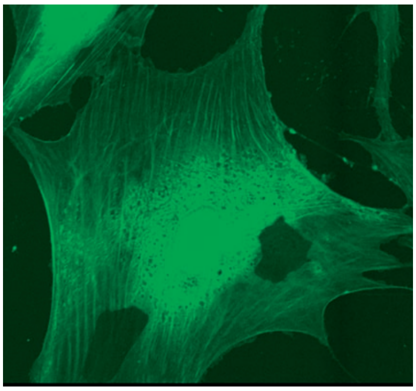


Reviews

Artificial Life

S. Mann* ________ **5306 – 5320**

Life as a Nanoscale Phenomenon



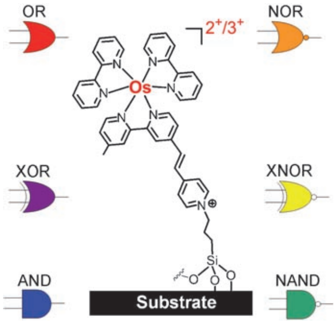
From little to large: Dimensional constraints are critical for the gathering, processing, and transmission of chemical-based information with the consequence that nanoscale miniaturization is fundamental to the emergence and sustainability of cellular life. The deep significance of this relationship is analyzed in this Review.

Communications

Boolean Logic

T. Gupta,
M. E. van der Boom* ________ **5322 – 5326**

Redox-Active Monolayers as a Versatile Platform for Integrating Boolean Logic Gates



A layer of logic: A series of Boolean operations has been demonstrated with redox-active monolayers of osmium and ruthenium complexes on glass substrates. High stability, selective reactivity, and reversible redox-chemistry, coupled with significant optical changes, make these monolayers versatile logic gates. The electron-transfer-based systems are suitable as standalone systems or as operational parts in logic circuits.

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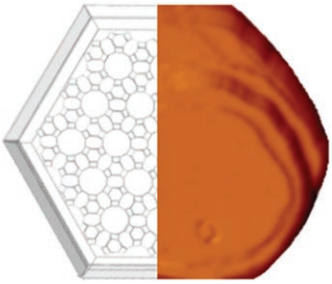
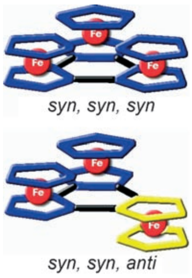
Scratching the surface: Atomic force microscopy has been utilized to study the surface of zeolite L (see AFM image of the hexgonal face of a crystal). Correlations between nanometer high features and the structural units have been determined, with the smallest unit protruding from the surface corresponding to a single cancrinite cage. The mechanism by which growth units attach depends on the crystallographic face.



Zeolite Crystal Growth

R. Brent, M. W. Anderson* **5327 – 5330**

Fundamental Crystal Growth Mechanism in Zeolite L Revealed by Atomic Force Microscopy



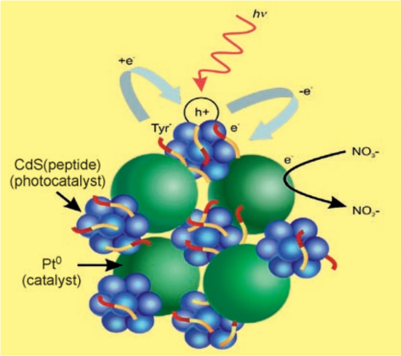
The "fusion" of three ferrocenes into a rigid cyclic array produces the *syn,syn,anti* and *syn,syn,syn* isomers of [(FeCp)$_3$(trindenyl)] (see picture), the cations of which display different mixed-valence properties, namely multiple intervalence-transfer (IT) absorptions appear in the near-IR/IR spectra for the *syn,syn, anti* isomer, while a single band appears in the visible/near-IR spectrum for the *syn, syn,syn* isomer.

Mixed Valency

S. Santi,* L. Orian, A. Donoli, A. Bisello, M. Scapinello, F. Benetollo, P. Ganis, A. Ceccon* **5331 – 5334**

Synthesis of the Prototypical Cyclic Metallocene Triad: Mixed-Valence Properties of [(FeCp)$_3$(trindenyl)] Isomers

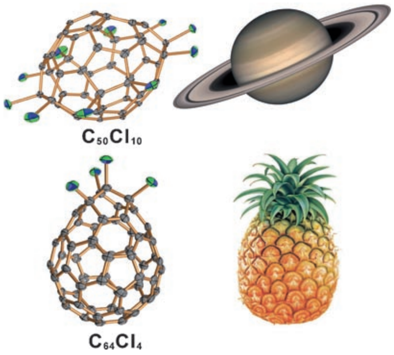
Better than the original: A hybrid nanoparticle system consisting of peptide-coated CdS and Pt nanoparticles acts as an enzyme mimic in the photoactivated reduction of nitrate (see picture). As compared to enzymes, the biotemplated nanoparticles exhibit enhanced activity, are optically activated, and can be used at elevated temperatures. Moreover, electron mediators and exogenous sacrificial electron donors are not needed.



Bio-Nanotechnology

J. M. Slocik, A. O. Govorov, R. R. Naik* **5335 – 5339**

Photoactivated Biotemplated Nanoparticles as an Enzyme Mimic



Planet pineapple: The structures of two isolated pentagon rule (IPR)-violating chlorofullerenes (C$_{50}$Cl$_{10}$ and C$_{64}$Cl$_4$; see picture) are determined by X-ray crystallography and their stabilities rationalized by relief of strain at the active pentagon-fusion carbon atoms whilst maintaining the aromaticity of their carbon skeletons.

IPR-Violating Fullerenes

X. Han, S. J. Zhou, Y. Z. Tan, X. Wu, F. Gao, Z. J. Liao, R.-B. Huang, Y.-Q. Feng, X. Lu, S. Y. Xie,* L.-S. Zheng **5340 – 5343**

Crystal Structures of Saturn-Like C$_{50}$Cl$_{10}$ and Pineapple-Shaped C$_{64}$Cl$_4$: Geometric Implications of Double- and Triple-Pentagon-Fused Chlorofullerenes

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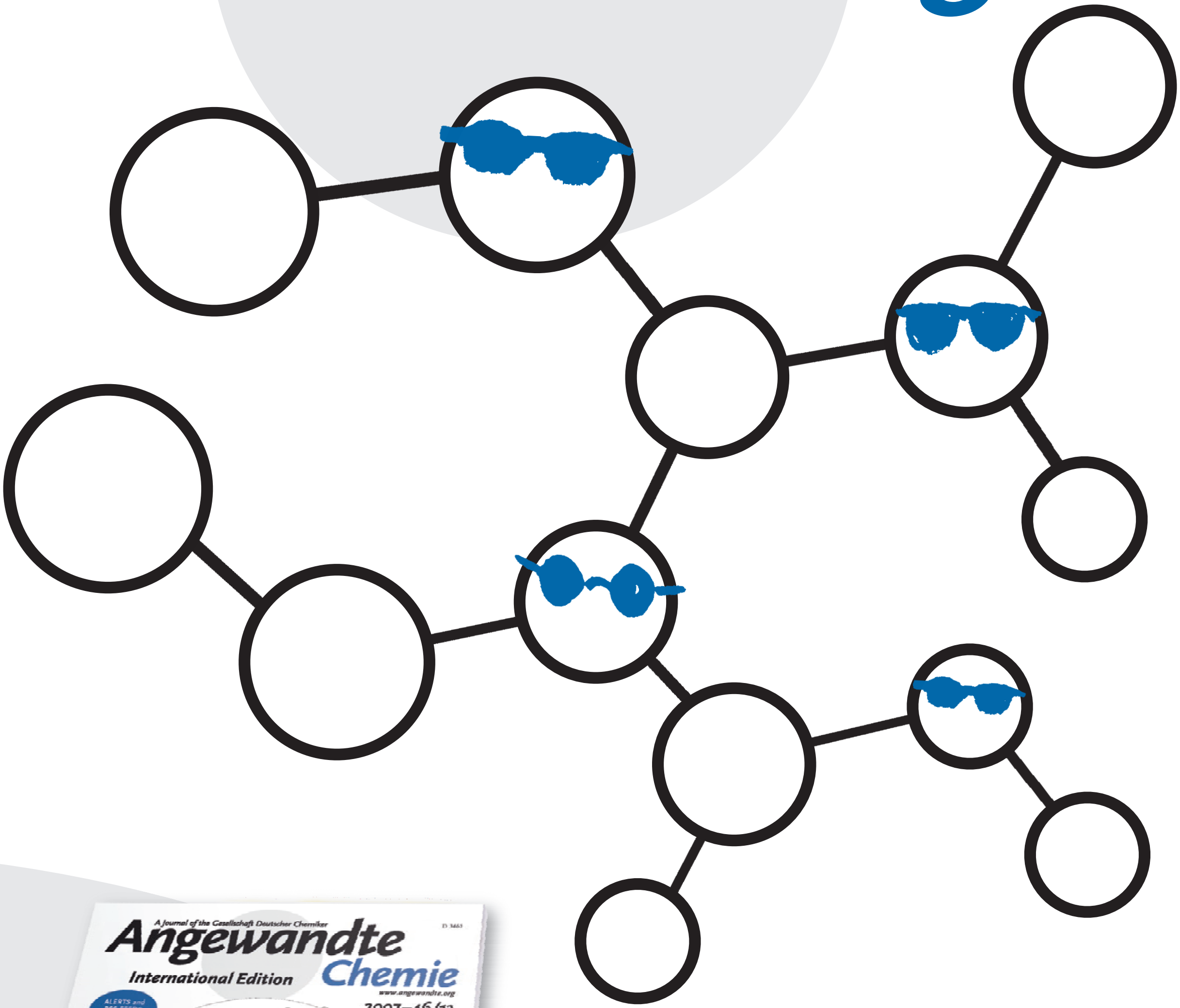
Incredibly incognito!



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2007–46/13

1st April Issue

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Controlling Magnetic Properties

O. Sato et al.

Natural Product (Bio)synthesis

H. B. Bode and R. Müller

C-H and C-C Bond Activation

ACIEFS 46 (13) 2123–2332 (2007) · ISSN 1433–7851 · Vol. 46 · No. 13

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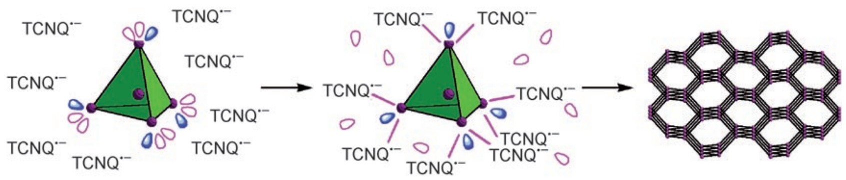
Self-Assembly

Y. L. Bai, J. Tao,* R.-B. Huang,
L.-S. Zheng ______ 5344 – 5347

The Designed Assembly of Augmented Diamond Networks From Predetermined Pentanuclear Tetrahedral Units

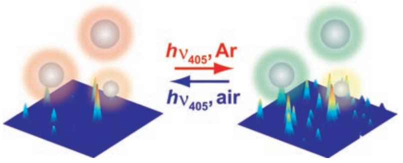


Step by step: Pentanuclear clusters (see picture) are synthesized as building units for the construction of three-dimensional, diamondlike networks. The ESI mass spectra of these clusters reveal that the pentanuclear units maintain their structural integrity before condensing into frameworks, thereby allowing a successful step-by-step assembly.

Fluorescence Spectroscopy

T. Tachikawa, T. Ishigaki, J.-G. Li,
M. Fujitsuka, T. Majima* __ 5348 – 5352

Defect-Mediated Photoluminescence Dynamics of Eu^{3+}-Doped TiO_2 Nanocrystals Revealed at the Single-Particle or Single-Aggregate Level

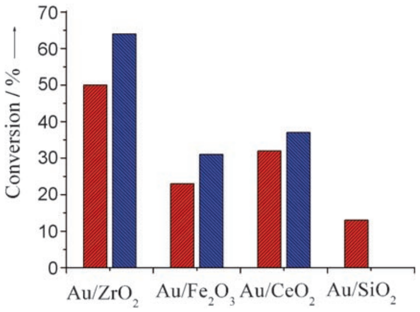


Single-particle fluorescence spectroscopy was used to investigate defect-mediated photoluminescence (PL) dynamics of undoped and Eu^{3+}-doped TiO_2 nanoparticles, and revealed that free excitons in the photoirradiated TiO_2 host can excite both interior and surface Eu^{3+} ions, while trapped excitons at the surface only excite the latter. The picture shows PL images of TiO_2:Eu^{3+} nanoparticles during laser excitation in air and argon atmospheres.

Nanostructured Catalysts

X. Chen, H.-Y. Zhu,* J.-C. Zhao,
Z.-F. Zheng, X.-P. Gao* ___ 5353 – 5356

Visible-Light-Driven Oxidation of Organic Contaminants in Air with Gold Nanoparticle Catalysts on Oxide Supports

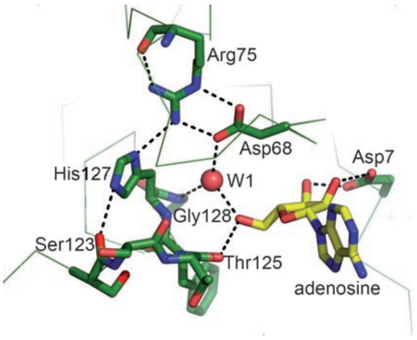


Staying cool in the sun: When illuminated with visible light, gold nanoparticles dispersed on various metal oxide supports show significant activity in the oxidation of formaldehyde and methanol in air at room temperature. HCHO conversion under illumination of blue light (see picture, blue bars) and under red light (red bars) are compared.

Enzyme Catalysis

H. Deng, C. H. Botting,
J. T. G. Hamilton, R. J. M. Russell,
D. O'Hagan* ______ 5357 – 5361

S-Adenosyl-L-methionine:Hydroxide Adenosyltransferase: A SAM Enzyme



Not so DUF: A DUF62 enzyme from the archaeon *Pyrococcus horikoshii* OT3 converts *S*-adenosyl-L-methionine (SAM) into adenosine through the nucleophilic attack of a hydroxide ion derived from water (see picture of the active site). The highly conserved nature of Asp68, Arg75, and His127 throughout the DUF62 protein superfamily suggests the wide-spread distribution of this novel catalytic activity in microorganisms.
DUF = domain of unknown function.

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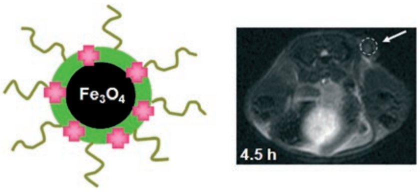
Multifunctional Nanoparticles

M. K. Yu, Y. Y. Jeong, J. Park, S. Park, J. W. Kim, J. J. Min, K. Kim, S. Jon* **5362 – 5365**

Drug-Loaded Superparamagnetic Iron Oxide Nanoparticles for Combined Cancer Imaging and Therapy In Vivo

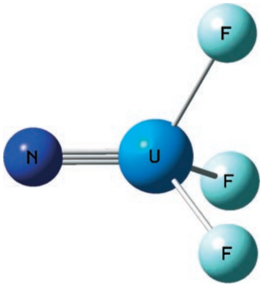
Multitasking rust ball: The questions of where a tumor is located, whether drugs are properly delivered to the tumor, and what the therapeutic response of the tumor is, could be answered by using simple but smart, drug-loaded superparamagnetic iron oxide nanoparticles that can accumulate in tumor with high efficiency thanks to the presence of rationally engineered polymer coating layers.



Multiple Bonds

L. Andrews,* X. Wang, R. Lindh, B. O. Roos, C. J. Marsden **5366 – 5370**

Simple N≡UF$_3$ and P≡UF$_3$ Molecules with Triple Bonds to Uranium



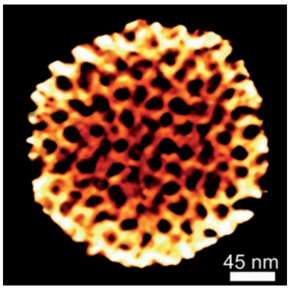
UN-beatable? Laser-ablated uranium atoms activate NF$_3$ and PF$_3$ to form the N≡UF$_3$ and P≡UF$_3$ molecules containing novel terminal nitride and phosphide functional groups. These molecules are identified from matrix infrared spectra and theoretical methods. The N≡UF$_3$ molecule contains the strongest triple bond to uranium in a ternary compound.

Mesoporous Metal

Y. Yamauchi,* A. Sugiyama, R. Morimoto, A. Takai, K. Kuroda* **5371 – 5373**

Mesoporous Platinum with Giant Mesocages Templated from Lyotropic Liquid Crystals Consisting of Diblock Copolymers

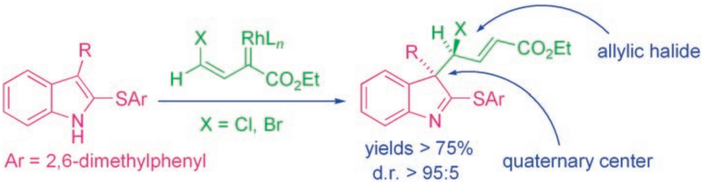
Giant metal cages: Mesoporous Pt particles, with mesocages connected closely in three dimensions (see picture), are prepared by an electrodeposition process through soft templating from lyotropic liquid crystals of diblock copolymers. The size of the mesocages is the largest (about 15 nm) reported in mesoporous metals. The method can be extended to other metals and mesostructures, and the mesopores can be controlled over a range of pore sizes.



Synthetic Methods

V. Boyarskikh, A. Nyong, J. D. Rainier* **5374 – 5377**

Highly Diastereoselective Sulfonium Ylide Rearrangements to Quaternary Substituted Indolines



Quat. centers: Sulfonium ylide intermediates generated from halogenated vinyl diazoacetates and 2-thioindoles in the presence of a Rh^{II} complex undergo highly diastereoselective [3,3] rearrangements to indolines having an asymmetric quaternary carbon center with an adjacent halogenated stereocenter (see scheme).

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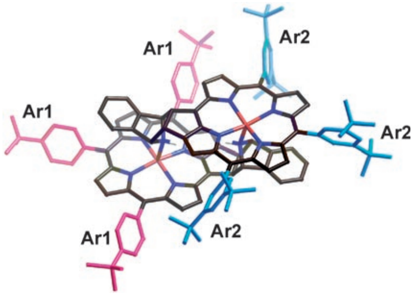
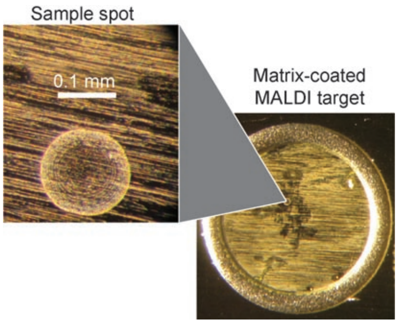
Positive discrimination: Palladium-catalyzed [3+2] annulation of meso-bromoporphyrin with norbornene derivatives efficiently provides novel chiral meso, β-fused porphyrins with a rigid structure (as shown). A chiral benzoazanorbornene-fused zinc porphyrin assembles selectively to form the slipped cofacial dimer in a heterochiral association, which allows construction of a heterodimer from two porphyrin components through chiral discrimination.

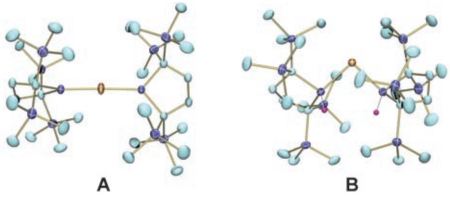


Self-Assembly

Getting a look-in: An optimized MALDI-MS procedure has been developed to detect endogenous primary metabolites directly in the cell extract. A detection limit corresponding to metabolites from less than a single cell has been attained, opening the door to single-cell metabolomics by mass spectrometry.

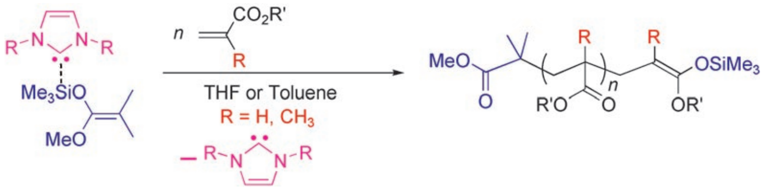


Analytical Methods



Hydrogens make a difference: The first 14-electron bis(silylene)palladium (**A**) and 12-electron bis(silyl)palladium complexes (**B**) are synthesized from a stable dialkylsilylene (see picture; gold: Pd; dark blue: Si; light blue: C; magenta: H). The remarkable difference between the L-M-L angles of these two complexes is rationalized by applying the Walsh diagram.

Silylene Complexes



Neutral but versatile: N-heterocyclic carbenes are very efficient catalysts for the group-transfer polymerization of acrylates and methacrylates in both polar and apolar media. These organocatalysts were also used to design *all*-(meth)acrylic di- and triblock copolymers.

Organocatalyzed Polymerization

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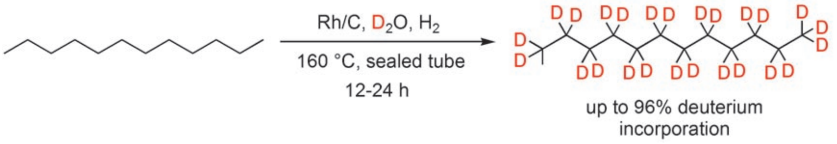
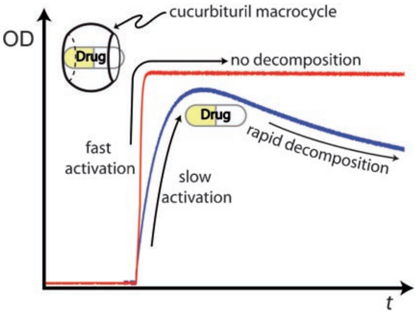
C–H Activation



Easy access: In the presence of Rh/C in D_2O under H_2 at 160 °C the H/D exchange reaction of unfunctionalized alkanes can easily occur (see scheme). Inexpensive reagents and mild reaction conditions are used, and fully deuterated products can be obtained after a simple work up procedure.

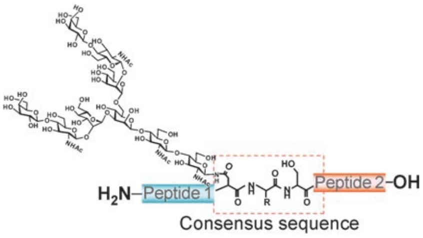
Supramolecular Chemistry



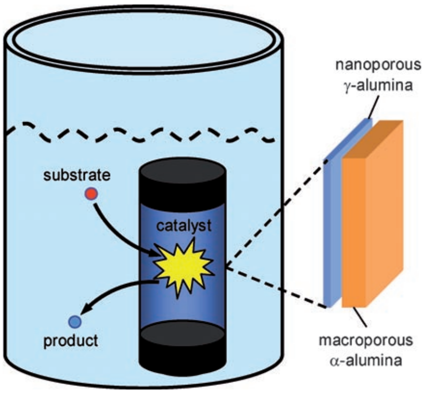
Controlling the drug supply: UV and NMR spectroscopic studies show that drugs such as omeprazole and lansoprazole, popular drugs for the treatment of gastric acid related diseases, are stable for several weeks by using cucurbit[7]uril as a macrocyclic host (see scheme). The drugs are also more rapidly converted into their active forms by an acid-promoted reaction catalyzed by a supramolecular pK_a shift.

Glycopeptide Ligation

Serine in site: A native chemical ligation concept at the serine site is described (see picture). This method was achieved by the CNBr conversion reaction from methylcysteine, which could be obtained by selective S methylation. This new method allowed a consensus sequence that is generally incorporated in N-linked oligosaccharides to be used as a ligation site in the course of glycopeptide synthesis.



Applied Catalysis



A cat with nine lives: Catalyst separation and recycling is the bane of homogeneous catalysis. This problem is addressed by a two-layered ceramic membrane cylinder that allows the diffusion of reactants and products in and out, but keeps the catalyst trapped. The concept is demonstrated for the enantioselective transfer hydrogenation of acetophenone to (S)-phenylethyl alcohol with large molecular catalysts anchored on Fréchet-type dendrimers.

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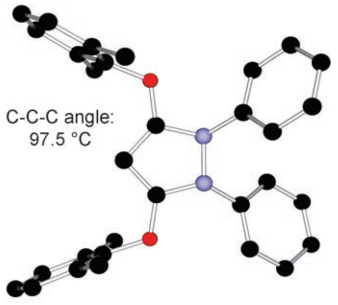


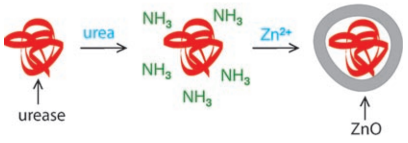
Push–push substitution patterns render allenes so flexible that the usually linear C=C=C fragment can be accommodated in a five-membered ring composed of only nitrogen and carbon atoms (see structure; C black, N blue, O red). These cyclic allenes appear to be strong donor η^1 ligands for alkali and transition metals.

Bent Allenes

V. Lavallo, C. A. Dyker,* B. Donnadieu, G. Bertrand* ______ **5411 – 5414**

Synthesis and Ligand Properties of Stable Five-Membered-Ring Allenes Containing Only Second-Row Elements

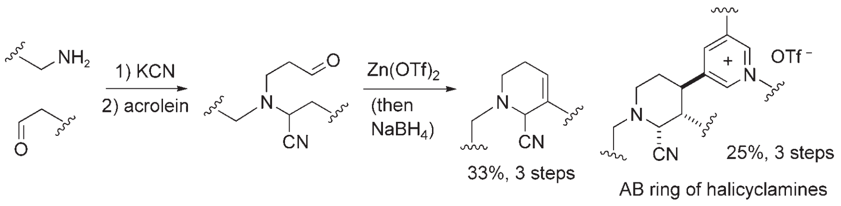
Reactor core: Hybrid core–shell urease–ZnO nanostructures have been synthesized (see picture). The crystallization takes place under mild biological conditions, thus ensuring its easy implementation in other room-temperature syntheses of oxide semiconductor materials.



Bio-Nanoreactors

R. de la Rica, H. Matsui* ___ **5415 – 5417**

Urease as a Nanoreactor for Growing Crystalline ZnO Nanoshells at Room Temperature

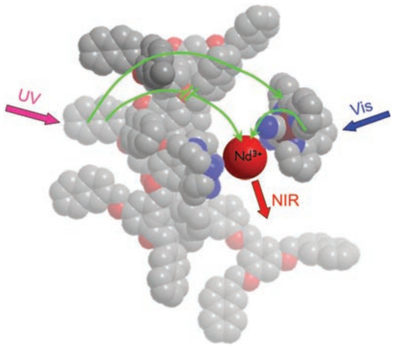


Without the isolation of intermediates, 5,6-dihydropyridinium salts and their nitrile derivatives were prepared as shown in the scheme. Cyclization in the presence of an aminopentadienal led to the formation of the AB ring system of the halicyclamines, or, under alternative conditions, the fused AB ring system of the manzamines. Tf = trifluoromethanesulfonyl.

Natural Products

J.-C. Wypych, T. M. Nguyen, P. Nuhant, M. Bénéchie, C. Marazano* **5418 – 5421**

Further Insight from Model Experiments into a Possible Scenario Concerning the Origin of Manzamine Alkaloids



A luminescence mediator: An unusual lanthanide complex, exploiting a dendrimer and a ruthenium(II) complex as ligands, collects ultraviolet and visible light and exhibits luminescence in the near infrared region (see picture, N Blue, Ru brown, O red, C gray). In the absence of the ruthenium component the dendrimer is unable to transfer energy to the Nd^{III} ion, even though it is directly coordinated to it.

Supramolecular Chemistry

C. Giansante, P. Ceroni,* V. Balzani, F. Vögtle ______ **5422 – 5425**

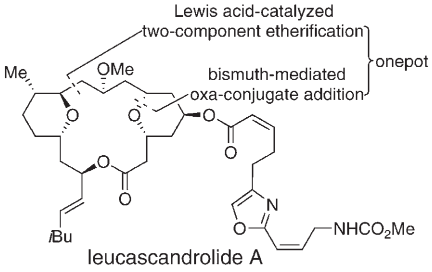
Self-Assembly of a Light-Harvesting Antenna Formed by a Dendrimer, a Ru^{II} Complex, and a Nd^{III} Ion

Natural Product Synthesis

P. A. Evans,* W. J. Andrews **5426 – 5429**

A Sequential Two-Component Etherification/Oxa-Conjugate Addition Reaction: Asymmetric Synthesis of (+)-Leucascandrolide A Macrolactone

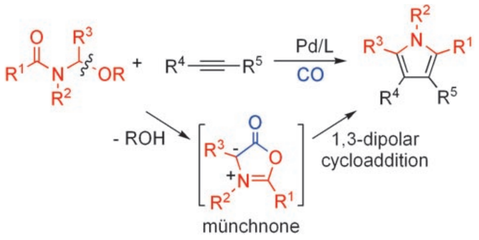
A smooth operator: The asymmetric synthesis of the macrolactone of (+)-leucascandrolide A (see structure) has been accomplished through a convergent route (longest linear sequence of 14 steps) in 20% overall yield. The assembly of the 1,5-bis(tetrahydropyran) core in a single operation provides the most concise synthetic approach developed to date.



Cycloaddition

Y. Lu, B. A. Arndtsen* ______ **5430 – 5433**

Palladium Catalyzed Synthesis of Münchnones from α-Amidoethers: A Mild Route to Pyrroles

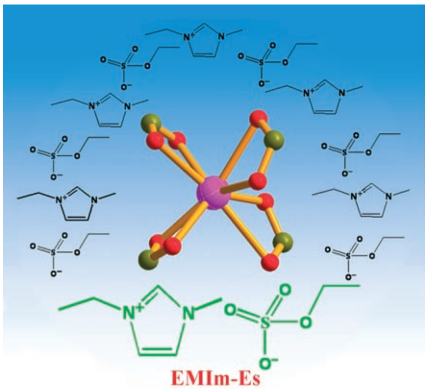


Having the munchies: A mild palladium-catalyzed method to activate the carbon–oxygen bond of α-amidoethers has been developed and applied to carbonylation chemistry (see scheme). A münchnone intermediate is generated in situ and undergoes a 1,3-dipolar cycloaddition with alkynes to give diversely substituted pyrroles.

Metal–Organic Frameworks

J. Zhang, S. Chen, X. Bu* __ **5434 – 5437**

Multiple Functions of Ionic Liquids in the Synthesis of Three-Dimensional Low-Connectivity Homochiral and Achiral Frameworks



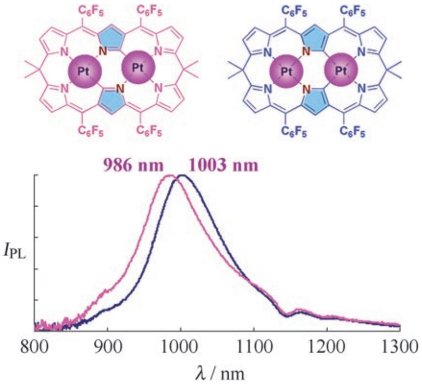
One foot in the door: In the self-assembly of a homochiral or achiral low-connectivity metal–dicarboxylate framework, an ionic liquid (shown: EMIm-Es) undergoes either complete entrapment, partial (cation-only), or no entrapment. The structure type depends on the ionic liquid and whether a structure-directing agent is used.

Calixphyrins

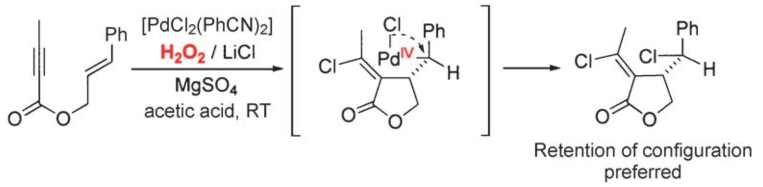
D.-H. Won, M. Toganoh, Y. Terada, S. Fukatsu, H. Uno, H. Furuta* ______ **5438 – 5441**

Near-Infrared Emission from Bis-Pt^{II} Complexes of Doubly N-Confused Calix[6]phyrins(1.1.1.1.1.1)

Light from confusion: A set of doubly N-confused calix[6]phyrins bearing two coordination sites (NNNC,NNNC or NNNN,NNCC) were synthesized. Their bis-Pt^{II} complexes exhibit near-infrared emission around 1000 nm, which could make these compounds ideal for biological applications.

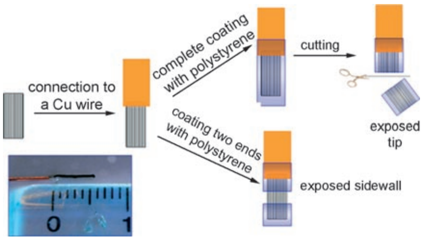


Oxidative Cyclization



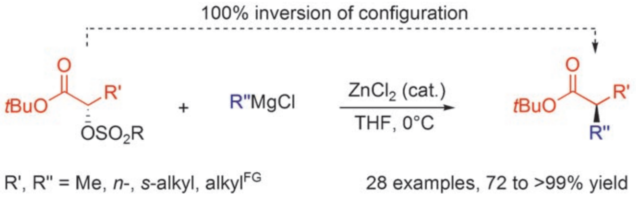
Gently does it: The Pd-catalyzed oxidative cyclization of enynes with hydrogen peroxide, a simple, inexpensive, and environmental benign oxidant, is proposed to proceed through a catalytic Pd^{II}/Pd^{IV} cycle in which the key C–Cl bond-forming step is the formation of a Pd^{IV} intermediate (see scheme). This is followed by a direct reductive elimination to generate C–Cl bonds with retention of the configuration of the $C(sp^3)$ center.

Carbon Nanotubes



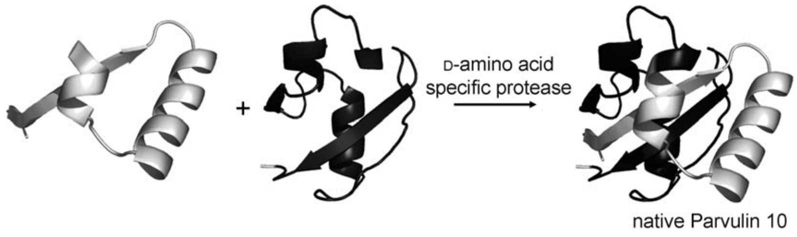
More than tips: The electrochemical properties of the tips and sidewalls of carbon nanotubes can be compared when specific regions are selectively masked with a nonconducting polymer coating (e.g. polystyrene) such that the electrolyte has access to one region only (see scheme). Surprisingly electrochemistry at carbon nanotube electrodes is not always facilitated by the nanotube tip.

Synthetic Methods



Zinc chloride does the trick and efficiently catalyzes the enantiospecific cross-coupling of α-hydroxy ester triflates with Grignard reagents under mild conditions. Enantiopure α-hydroxy esters are directly available from the chiral pool or by diazotization of α-amino acids. Substantial variations in both reacting partners are tolerated making this methodology an attractive alternative to enolate alkylation featuring a reversal of polarity.

Protein Synthesis by D-Peptidases



Side reactions side-stepped: A D-amino acid specific protease was used for the first time in the chemoenzymatic synthesis of a native protein, the peptidyl prolyl *cis*/*trans* isomerase parvulin 10 from *E. coli* (see scheme). Side reactions that complicate synthesis with L-amino acid specific enzymes are avoided in this approach.

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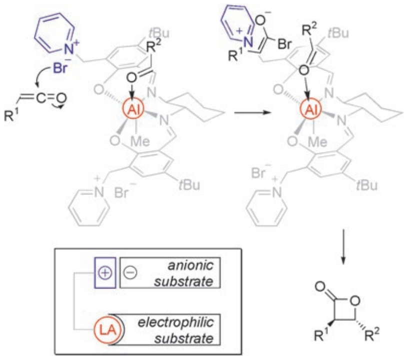
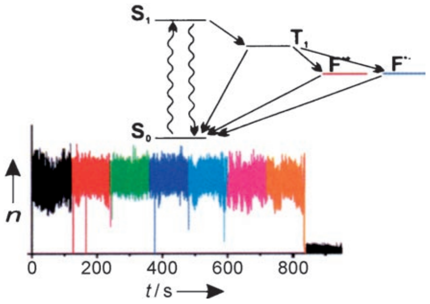
Dual-Activation Catalysis

Opposites attract. A novel concept for dual-activation catalysis takes advantage of the cooperative action of aprotic contact ion pair catalysis and Lewis acid catalysis. This strategy was used for the first *trans*-selective catalytic asymmetric [2+2] cyclocondensation of acyl halides with aliphatic aldehydes, furnishing *trans*-configured 3,4-disubstituted β-lactones with high stereoselectivity.



Fluorescence Spectroscopy



On the ROXS: Photobleaching represents one of the main limitations of modern fluorescence microscopy. A reducing and oxidizing system (ROXS) has been introduced that recovers triplet states as well as charge-separated states through electron-transfer reactions (see picture). Thus the blinking and photobleaching of fluorophores is strikingly reduced.

Supporting information is available on www.angewandte.org (see article for access details).

A video clip is available as Supporting Information on www.angewandte.org (see article for access details).

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Corrigendum

Synergistic Effects of B/N Doping on the Visible-Light Photocatalytic Activity of Mesoporous TiO_2

G. Liu, Y. Zhao, C. Sun, F. Li, G. Q. Lu,* H.-M. Cheng* ———— **4516–4520**

Angew. Chem. Int. Ed. **2008**, *47*

DOI 10.1002/anie.200705633

The authors of this Communication wish to cite additional papers relevant to their work. Reports by Fornasiero et al.[23a] and Sun et al.,[23c] both dealing with B,N-codoped TiO_2, were inadvertently omitted from the references cited in this Communication. In those reports, nitrogen doping was realized not by thermal treatment route in the presence of an ammonia atmosphere, but by a wet-chemistry route, and no B—N bond was formed. The complete reference [23] is given below.

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[23] a) V. Gombac, L. Rogatis, A. Gasparotto, G. Vicario, T. Montini, D. Barreca, G. Balducci, P. Fornasiero, E. Tondello, M. Graziani, *Chem. Phys.* **2007**, *339*, 111; b) S. In, A. Oriov, R. Berg, F. Garcia, S. Pedrosa-Jimenez, M. S. Tikhov, D. S. Wright, R. M. Lambert, *J. Am. Chem. Soc.* **2007**, *129*, 13790; c) Q. Ling, J. Sun, Q. Zhu, *Appl. Surf. Sci.* **2008**, *254*, 3236.

Corrigendum

Stereoselective Synthesis of β-D-Mannopyranosides with Reactive Mannopyranosyl Donors Possessing a Neighboring Electron-Withdrawing Group—a Second Corrigendum

A. A.-H. Abdel-Rahman, S. Jonke, E. S. H. El Ashry, R. R. Schmidt ———— **2972–2974**

Angew. Chem. Int. Ed. **2002**, *41*

DOI 10.1002/1521-3773-(20020816)41:16<2972::AID-ANIE2972>3.0.CO;2-4

In the first Corrigendum published from Schmidt et al.,[1] it was revealed that the data published in footnote [25] of the original Communication[2] was that of an unidentified byproduct, and not that of alcohol **3**. It is now clear that the data in footnote [28] is also incorrect, and does not pertain to the glycoside **10β**. The incorrect assignment resulted from the same type of error as for **3**. The editorial office thanks Dr. Prasanna Jayalath and Dr. Ming Li from the laboratory of Professor David Crich for the preparation of authentic samples of **3** and **10β**[3] and for bringing this matter to our attention.

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[1] A. A.-H. Abdel-Rahman, S. Jonke, E. S. H. El Ashry, R. R. Schmidt, *Angew. Chem.* **2004**, *116*, 4489; *Angew. Chem. Int. Ed.* **2004**, *43*, 4389.

[2] A. A.-H. Abdel-Rahman, S. Jonke, E. S. H. El Ashry, R. R. Schmidt, *Angew. Chem.* **2002**, *114*, 3100–3103; *Angew. Chem. Int. Ed.* **2002**, *41*, 2972–2974.

[3] Disaccharide **10β** was obtained by an unambiguous route involving classical neighboring-group-directed ß-glucosylation, selective removal of the participating ester, oxidation, and selective reduction of the uloside to the ß-mannoside.

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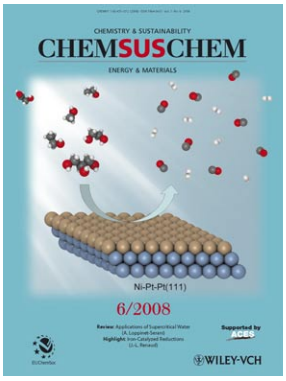

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