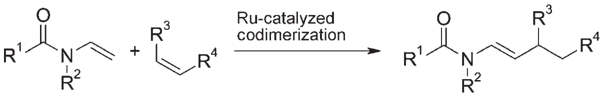
The following Communications have been judged by at least two referees to be “very important papers” and will be published online at www.angewandte.org soon:

Obituary

Books

Highlights

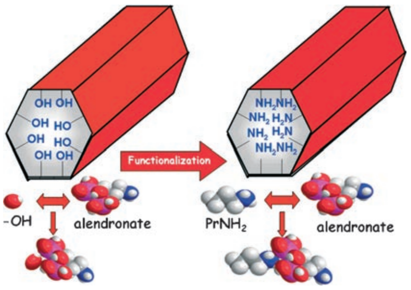
C–C Coupling Reactions



Selective in many aspects: The selective codimerization of enamides with other alkenes to give alkylated enamides has been achieved for the first time in the presence of Ru catalysts. The underlying strategy of utilizing different coordination modes of two olefins at a metal center for selective coupling could open up atom-economic access to functionalized alkenes.

Minireviews

Drug-Delivery Systems



Special delivery: Silica-based ordered mesoporous materials and metal–organic frameworks have been proposed as carriers for the controlled release of drugs. Optimization of the texture properties of mesoporous matrices and changes in the chemical composition of the pore walls lead to better drug-delivery systems as well as stimuli-responsive materials.

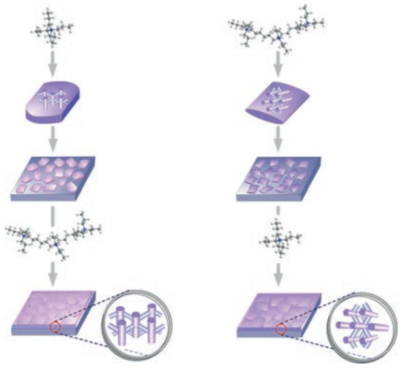
Contents

Reviews

Zeolite Membranes

M. A. Snyder, M. Tsapatsis* **7560 – 7573**

Hierarchical Nanomanufacturing: From Shaped Zeolite Nanoparticles to High-Performance Separation Membranes



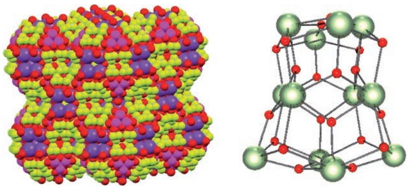
A deeper understanding: New understanding of zeolite growth along with recent advances in molecular engineering of crystal microstructure and morphology, assembly of crystal monolayers, and growth of ordered films with controlled microstructure raise prospects for future success in synthesizing commercially viable zeolite membranes and complex functional materials through hierarchical approaches.

Communications

Uranium Oxo Clusters

G. Nocton, F. Burdet, J. Pécaut, M. Mazzanti* **7574 – 7578**

Self-Assembly of Polyoxo Clusters and Extended Frameworks by Controlled Hydrolysis of Low-Valent Uranium

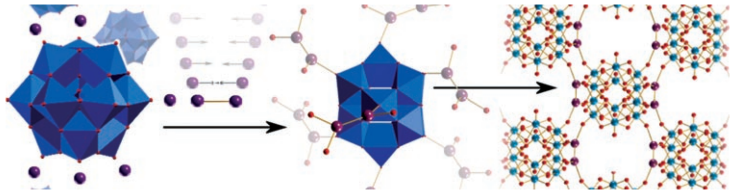


Cores for celebration! Hydrolysis of trivalent uranium compounds with a stoichiometric amount of water leads to mixed-valence (U^{IV}/U^{V}) discrete clusters with the U_6O_8 core and with the unprecedented $U_{12}O_{20}$ core (see picture, right; U green, O red), and to the assembly of U_6O_8 clusters into extended 3D networks of zeolite-like topology with nanosized cavities (left; U pink, K violet, O red, F green).

Functional POM-OFs

C. Streb, C. Ritchie, D.-L. Long, P. Kögerler, L. Cronin* **7579 – 7582**

Modular Assembly of a Functional Polyoxometalate-Based Open Framework Constructed from Unsupported $Ag^{I}\cdots Ag^{I}$ Interactions



No ligand support: Modular assembly of the title system (see picture; Ag purple, W blue, O red) has been achieved using $\{H_3W_{12}O_{40}\}^{5-}$ and $\{Ag_2\}^{2+}$ building blocks to give a material with $[Ag(CH_3CN)_4]^+$ ions in its microporous channels. This framework is constructed by unsupported $Ag^{I}\cdots Ag^{I}$ interactions, and sorption studies show that the framework can reversibly adsorb up to 10 wt% acetonitrile.

For the USA and Canada:
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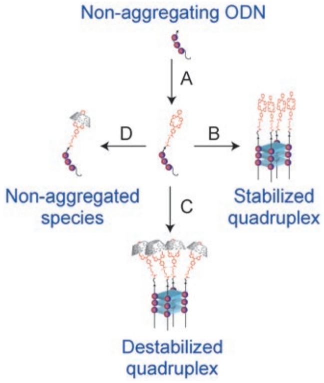
Quadruplex Self-Assembly

J. Jayawickramarajah, D. M. Tagore, L. K. Tsou, A. D. Hamilton* **7583 – 7586**

Allosteric Control of Self-Assembly: Modulating the Formation of Guanine Quadruplexes through Orthogonal Aromatic Interactions

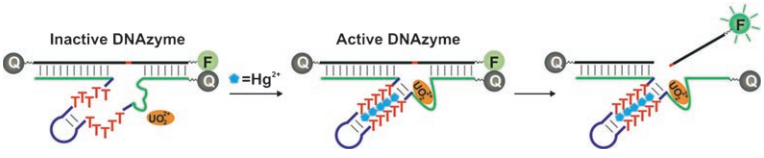


All four one: The covalent attachment (A) of a porphyrin head group onto a guanine-rich oligonucleotide strand can enhance the self-assembly of DNA quadruplexes through porphyrin-based π–π interactions (B). Modulation of these allosteric interactions, by addition of a porphyrin-complexing cyclodextrin derivative, allows for a high degree of control over the formation and disassembly of the guanine quadruplexes (C and D).

Mercury Sensors

J. Liu, Y. Lu* **7587 – 7590**

Rational Design of “Turn-On” Allosteric DNAzyme Catalytic Beacons for Aqueous Mercury Ions with Ultrahigh Sensitivity and Selectivity



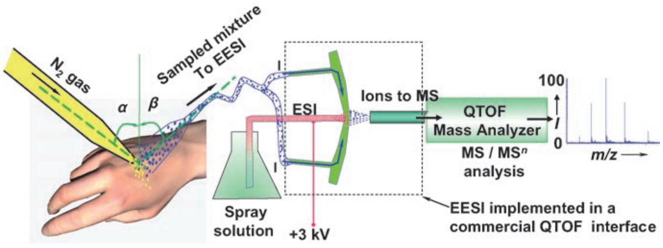
The mercury is rising: A DNAzyme with several thymine residues located close to the catalytic site is rendered inactive. However, thymine–Hg^{2+}–thymine interactions transform the DNAzyme into an active enzyme. When fluorophore/quencher pairs are attached, the DNAzyme can be converted into a highly sensitive and selective mercury sensor with a detection limit of 2.4 nM and a greater than 100 000-fold selectivity over other metal ions.

Analytical Methods

VIP

H. Chen,* S. Yang, A. Wortmann, R. Zenobi* **7591 – 7594**

Neutral Desorption Sampling of Living Objects for Rapid Analysis by Extractive Electrospray Ionization Mass Spectrometry

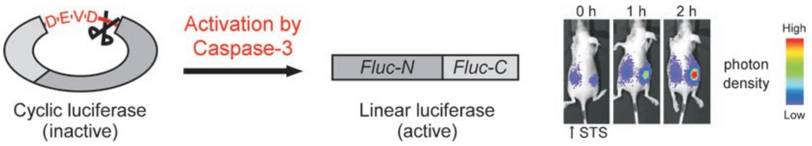


MS comes to life: A novel method to sample surfaces of biological objects uses a neutral gas beam for in vivo EESI mass spectrometric analysis without sample pretreatment (see picture; QTOF = quadropole time-of-flight). The sampling process results in rapid in vivo analyses with reduced ion suppression and without chemical contamination. This strategy can be used in food quality monitoring, homeland security, metabolomics, and clinical diagnosis.

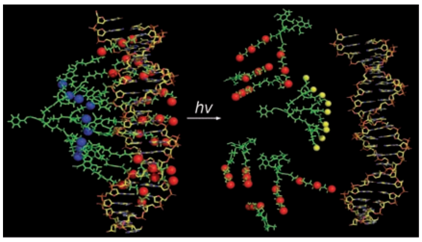
In Vivo Imaging

A. Kanno, Y. Yamanaka, H. Hirano, Y. Umezawa, T. Ozawa* **7595 – 7599**

Cyclic Luciferase for Real-Time Sensing of Caspase-3 Activities in Living Mammals



Illumination for apoptosis: Firefly luciferase connected with a substrate sequence for caspase-3 (DEVD) is cyclized by an intein DnaE. When the cyclic luciferase is expressed in living cells, its activity is greatly decreased because of a steric effect. Activated caspase-3 cleaves the substrate sequence in the cyclic luciferase and the luciferase activity is restored. Quantitative sensing of time-dependent caspase-3 activity in living cells and mice upon extracellular stimuli has been demonstrated (see images).



Catch and release: Multivalent dendrons that have *o*-nitrobenzyl-linked spermine surface groups bind DNA efficiently by multivalent interactions. Cleavage of the *o*-nitrobenzyl groups from the dendron framework, with short UV irradiation, results in a rapid release of DNA, which can be attributed to dendron degradation and charge-switching multivalency (see picture).

Controlled DNA Binding

M. A. Kostiainen,* D. K. Smith,*
O. Ikkala ———— **7600 – 7604**

Optically Triggered Release of DNA from Multivalent Dendrons by Degrading and Charge-Switching Multivalency

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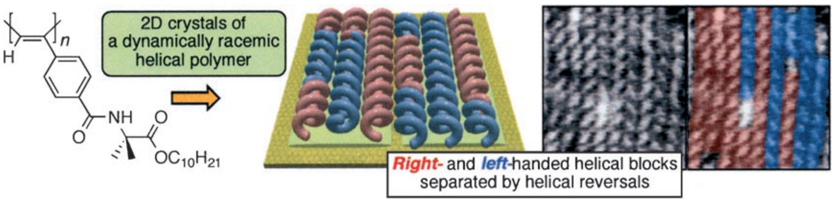


It all makes sense: A dynamically racemic helical polymer crystallizes on graphite upon exposure to an organic solvent vapor, resulting in two-dimensional helix-bundle formation. The enantiomeric right- and left-handed helical blocks separated by helical reversals can be directly visualized by AFM with molecular resolution.

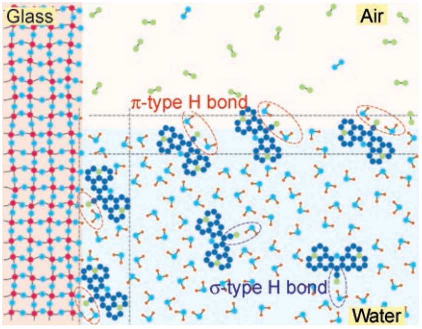
Helical Structures

S.-i. Sakurai,* S. Ohsawa, K. Nagai,
K. Okoshi, J. Kumaki,*
E. Yashima* ———— **7605 – 7608**

Two-Dimensional Helix-Bundle Formation of a Dynamic Helical Poly(phenylacetylene) with Achiral Pendant Groups on Graphite

---

π-Type H-bonds that are not found in bulk liquid phases, where σ-type H-bonds prevail, were detected for the dye rhodamine 800 at air/water and fused silica/water interfaces (see picture) by interface-selective fourth-order nonlinear ($\chi^{(4)}$) Raman spectroscopy in the frequency domain. This new method allows vibrational spectra of organic solutes at liquid interfaces to be obtained in the whole fingerprint region (200–2800 cm^{-1}).

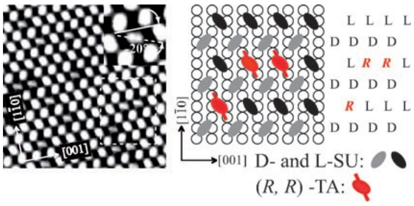


Interfaces

S. Yamaguchi, T. Tahara* ———— **7609 – 7612**

$\chi^{(4)}$ Raman Spectroscopy for Buried Water Interfaces

Site-specific chiral recognition in a two-dimensional heterochiral structure leading to highly enantiospecific substitution by a chiral guest was observed by scanning tunneling microscopy (STM). Thus, in the case of succinic acid (SU) on Cu(110), which forms two chiral motifs denoted D- and L-SU, (*R*,*R*)-tartaric acid ((*R*,*R*)-TA = *R*) only substitutes L sites (see picture; in the STM image *R* appears as "slots" aligned −20° to the [001] direction).



Chiral Recognition

N. Liu, S. Haq, G. R. Darling,
R. Raval* ———— **7613 – 7616**

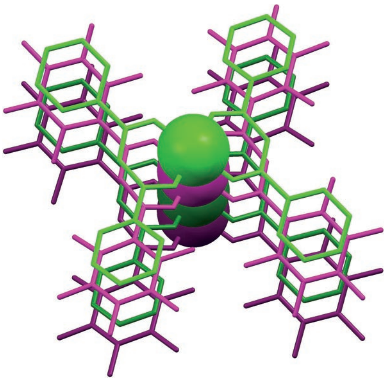
Direct Visualization of Enantiospecific Substitution of Chiral Guest Molecules into Heterochiral Molecular Assemblies at Surfaces

Contents

Metal Arrays

A. Hori,* A. Shinohe, M. Yamasaki,
E. Nishibori, S. Aoyagi,
M. Sakata ______ **7617 – 7620**

1:1 Cross-Assembly of Two β-Diketonate Complexes through Arene–Perfluoroarene Interactions

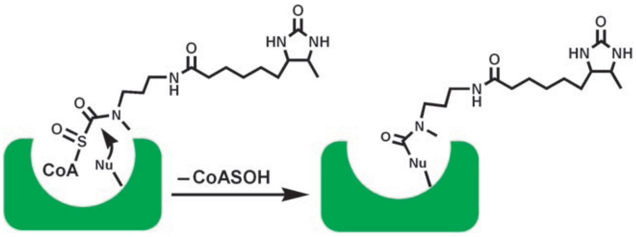


Stack ’em up: Two coordination complexes—one a perfluoroarene-functionalized β-diketonate copper complex (purple, see picture), the other an arene-functionalized metal (M = Cu, Pd, Pt) complex (green)—form alternately layered arrays through arene–perfluoroarene interactions with metal-to-metal distances close to van der Waals contacts to give 1:1 cocrystals as one-dimensional metal wires.

Affinity Probes

Y. Hwang, P. R. Thompson, L. Wang,
L. Jiang, N. L. Kelleher,
P. A. Cole* ______ **7621 – 7624**

A Selective Chemical Probe for Coenzyme A-Requiring Enzymes

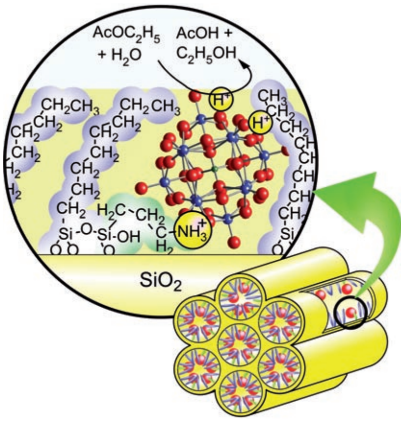


Tagging transferases: A coenzyme A (CoA)-based affinity probe with a sulfoxy-carbamate functionality can selectively identify several acetyltransferases relative to other enzymes and proteins. It leaves behind a desthiobiotin tag that can be used for western blotting and mass spectrometric characterization (see picture; Nu = nucleophile).

Nanostructured Catalysts

K. Inumaru,* T. Ishihara, Y. Kamiya,
T. Okuhara, S. Yamanaka ______ **7625 – 7628**

Water-Tolerant, Highly Active Solid Acid Catalysts Composed of the Keggin-Type Polyoxometalate $H_3PW_{12}O_{40}$ Immobilized in Hydrophobic Nanospaces of Organomodified Mesoporous Silica

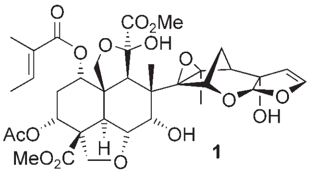


High activity in tight spaces: Polyoxometalate $H_3PW_{12}O_{40}$ surrounded by hydrophobic alkyl groups in the channels of mesoporous silica (see picture) showed exceptionally high catalytic activity for ester hydrolysis in water. The nanostructure based on mesoporous silica allows the aqueous reaction mixture to easily reach the active sites, despite their being surrounded by hydrophobic moieties.

Total Synthesis

G. E. Veitch, E. Beckmann, B. J. Burke, A. Boyer, S. L. Maslen, S. V. Ley* ______ 7629–7632

Synthesis of Azadirachtin: A Long but Successful Journey



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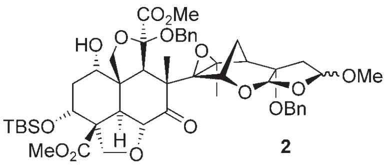


2

22 Years in the making: Azadirachtin (**1**) was synthesized for the first time by a highly convergent approach, utilizing a Claisen rearrangement and a radical cyclization as key steps. End-game strategies relied on intermediate **2**, which could be obtained by synthetic methods as well as by degradation of **1**. Bn = benzyl, TBS = *tert*-butyldimethylsilyl.

Total Synthesis

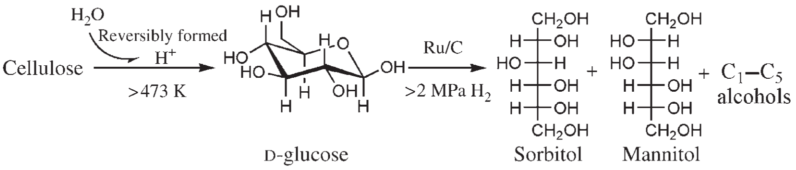
G. E. Veitch, E. Beckmann, B. J. Burke, A. Boyer, C. Ayats, S. V. Ley* 7633–7635

A Relay Route for the Synthesis of Azadirachtin

Sustainable Chemistry

C. Luo, S. Wang, H. Liu* ___ 7636–7639

Cellulose Conversion into Polyols Catalyzed by Reversibly Formed Acids and Supported Ruthenium Clusters in Hot Water



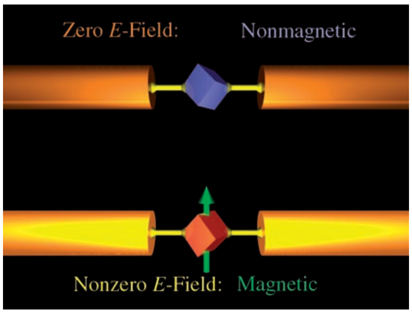
A green approach to efficient conversion of cellulose into hexitols and other lighter alcohols through two steps is reported. In this process, cellulose is hydrolyzed to glucose by acids formed reversibly in situ from hot water, and the glucose is hydrogenated on Ru/C. Understanding of the green aqueous catalytic systems should lead to more efficient conversion of cellulose into fuels and chemicals.

Magnetic Properties

M. Diefenbach,* K. S. Kim* 7640–7643

Towards Molecular Magnetic Switching with an Electric Bias

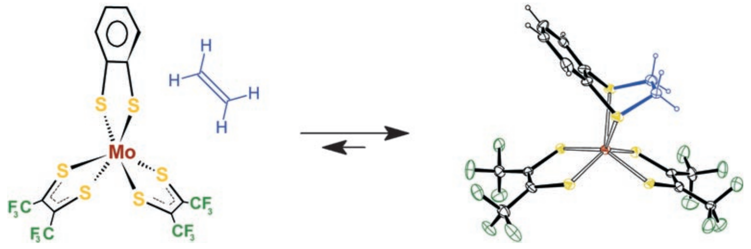
Mesmerizing Molecules: A principle for the direct manipulation of molecular spin states (electron spins) with an electric field is described. This concept is of fundamental interest to both chemistry and physics, and it paves the way for molecular spintronics and quantum computing. Magnetic switching of a molecule with an electric field (see scheme) is demonstrated with quantum-chemical calculations.



Dithiolene Ligands

D. J. Harrison, A. J. Lough, N. Nguyen, U. Fekl* ______ 7644–7647

Push–Pull Molybdenum Trisdithiolenes Allow Rapid Nonconventional Binding of Ethylene at Ligand Sulfur Atoms

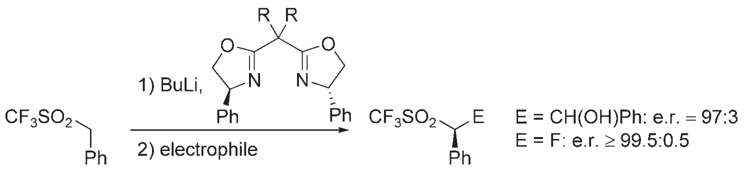


Adding to the mix: Mixed dithiolene complexes of molybdenum, such as [Mo{$S_2C_2(CF_3)_2$}$_2${$S_2(C_6H_4)$}], bind ethylene rapidly, in a nonconventional fashion, at the sulfur atoms (see scheme). In the presence of additional donor ligands, substitution of the metal-bound 2,3-dihydro-1,4-dithiin occurs, providing a new approach to mixed-ligand molybdenum bis- and trisdithiolenes.

Homogeneous Catalysis

S. Nakamura,* N. Hirata, T. Kita, R. Yamada, D. Nakane, N. Shibata, T. Toru* **7648 – 7650**

Highly Enantioselective Reactions of α-Sulfonyl Carbanions of Trifluoromethyl Sulfones

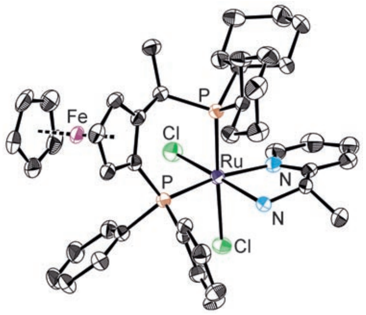


Making a resolution: The catalytic enantioselective reaction of lithiated benzyl trifluoromethyl sulfone with aldehydes delivers products with excellent diastereoselectivity as well as high enantioselectivity. Fluorination of the sulfone with *N*-fluorobenzensulfonimide in the presence of a stoichiometric amount of bis(oxazoline)s resulted in extremely high enantioselectivity (up to 99% *ee*, see scheme).

Asymmetric Catalysis

W. Baratta,* G. Chelucci, E. Herdtweck, S. Magnolia, K. Siega, P. Rigo **7651 – 7654**

Highly Diastereoselective Formation of Ruthenium Complexes for Efficient Catalytic Asymmetric Transfer Hydrogenation

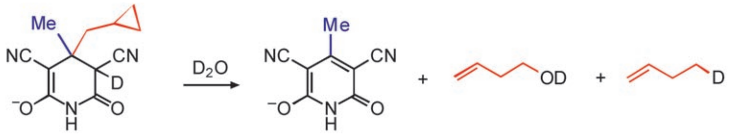


Jumping Josiphos! Ruthenium *cis*-dichloro complexes with matched chiral diphosphane and aminopyridine ligands (see structure) are easily obtained by a one-pot reaction of $[RuCl_2(PPh_3)_3]$ with a Josiphos diphosphane and racemic 1-substituted 1-(pyridin-2-yl)methanamine ligands. These complexes are highly active catalysts for the transfer hydrogenation of ketones, affording turnover frequencies of up to 70000 h^{-1} at 60 °C and *ee* values up to 99%.

Aqueous Radicals

B. Nguyen, K. Chernous, D. Endlar, B. Odell, M. Piacenti, J. M. Brown,* A. S. Dorofeev, A. V. Burasov **7655 – 7658**

Alkyl Radical Generation in Water under Ambient Conditions— A New Look at the Guareschi Reaction of 1897

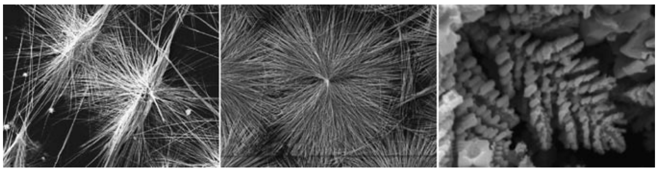


Chemical and biochemical significance of a long-forgotten 19th century observation: The hydrocarbon production from quaternary glutarimides on neutralization in water is the consequence of formation of alkyl radicals, which means that it is possible to generate alkyl radicals under very mild conditions. Oxygen trapping competes with hydrogen abstraction (see scheme).

Nanoarchitectures

X.-F. Shen, X.-P. Yan* **7659 – 7663**

Facile Shape-Controlled Synthesis of Well-Aligned Nanowire Architectures in Binary Aqueous Solution

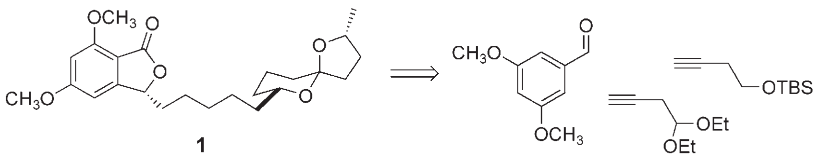


Nanobouquets: Shuttle-like fusiform bundles of L-cysteine–Pb nanowires split and open out to form dandelion-like flowers with well-aligned architectures (see image). The synthetic procedure provides an efficient route to the controllable preparation of well-arranged nanowires and, in a further step, to the formation of hierarchical PbS microstructures.

Natural Product Synthesis

B. M. Trost,* A. H. Weiss — **7664–7666**

An Alkyne Strategy for the Asymmetric Synthesis of Natural Products: Application to (+)-Spirolaxine Methyl Ether

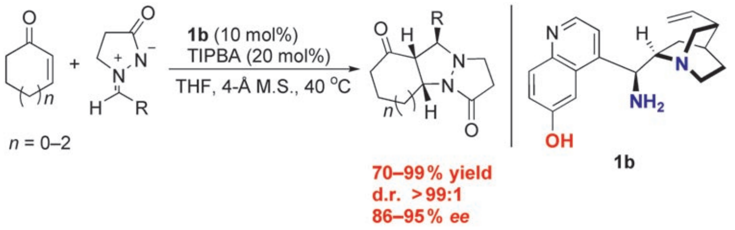


Alkyne of magic: An efficient synthesis of (+)-spirolaxine methyl ether (**1**) was accomplished by using alkynes as versatile synthetic building blocks. Highlights include two ProPhenol-catalyzed asymmetric alkynylations and a Pd-catalyzed spiroketalization (see scheme; TBS = *tert*-butyldiphenylsilyl). The use of alkynes alleviates the chemoselectivity issues of ketones and should be widely applicable to natural product synthesis.

Organocatalysis

W. Chen, W. Du, Y.-Z. Duan, Y. Wu, S.-Y. Yang, Y.-C. Chen* — **7667–7670**

Enantioselective 1,3-Dipolar Cycloaddition of Cyclic Enones Catalyzed by Multifunctional Primary Amines: Beneficial Effects of Hydrogen Bonding



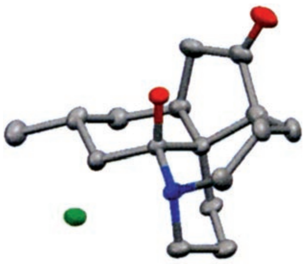
Hydrogen bonding makes a difference: Multifunctional primary amine catalysts derived from cinchona alkaloids are used in the highly enantioselective 1,3-dipolar cycloaddition of cyclic enones and azomethine imines (see scheme; R = aryl, alkyl; TIPBA = 2,4,6-triisopropylbenzenesulfonic acid). The synergistic hydrogen-bonding interaction of catalyst and 1,3-dipole is essential for stereocontrol.

Lycopodium Alkaloids

X. Linghu, J. J. Kennedy-Smith, F. D. Toste* — **7671–7673**

Total Synthesis of (+)-Fawcettimine

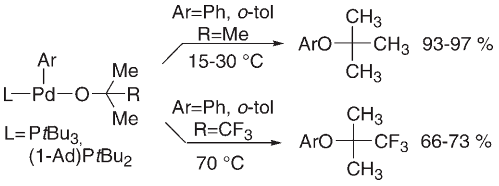
An effective combination: With the first asymmetric total synthesis of fawcettimine (**1**) it has been shown that the use of organocatalytic annulation and gold(I)-catalyzed cyclization reactions provides an effective combination for the synthesis of complex molecules. The absolute configuration of **1** was established through an X-ray structure analysis of its hydrobromide.


(+)-fawcettimine·HBr

C–O Bond Formation

J. P. Stambuli, Z. Weng, C. D. Incarvito, J. F. Hartwig* — **7674–7677**

Reductive Elimination of Ether from T-Shaped, Monomeric Arylpalladium Alkoxides



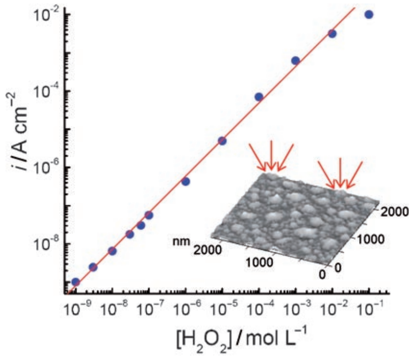
Isolated truth: The synthesis, structures, and reductive elimination chemistry of arylpalladium(II) phenoxide and alkoxide complexes with a single bulky phosphine ligand are reported. These complexes are true intermediates in palladium-catalyzed etherification of aryl halides. They undergo reductive elimination of the alkyl aryl ether directly from the isolated complex (see scheme), and the rates of these reductive eliminations are slower than those from related arylpalladium amido complexes. Ad = adamantyl.

Electrochemical Sensors

A. A. Karyakin,* E. A. Puganova,
I. A. Bolshakov,
E. E. Karyakina ______ **7678 – 7680**

Electrochemical Sensor with Record Performance Characteristics

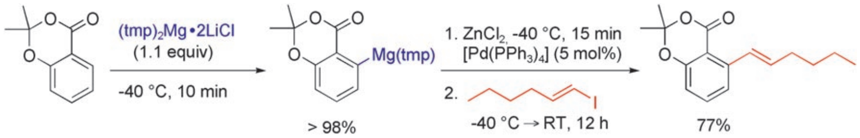
Nanoelectrode sensor arrays were formed by depositing nanostructures of the electrocatalyst Prussian Blue onto an inert carbon support. The sensor thus obtained showed a high sensitivity toward hydrogen peroxide, with a detection limit of 1×10^{-9} mol L^{-1} (i.e., 0.03 ppb), and a broad linear calibration range, which extended over seven orders of magnitude (from 10^{-9} to 10^{-2} M L^{-1} H_2O_2, see graphic).



Synthetic Methods

G. C. Clososki, C. J. Rohbogner,
P. Knochel* ______ **7681 – 7684**

Direct Magnesiation of Polyfunctionalized Arenes and Heteroarenes Using $(tmp)_2Mg\cdot2LiCl$

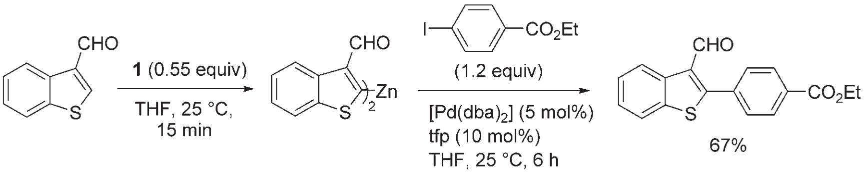


Outstripping lithium bases: A wide range of polyfunctional aryl and heteroaryl magnesium reagents were efficiently prepared in THF by direct magnesiation with $(tmp)_2Mg\cdot2LiCl$, an exceptionally active base. These compounds can serve as intermediates in natural product synthesis. In the example shown a palladium-catalyzed cross-coupling reaction is used to prepare a derivative of salicylic acid.

Synthetic Methods

S. H. Wunderlich,
P. Knochel* ______ **7685 – 7688**

$(tmp)_2Zn\cdot2MgCl_2\cdot2LiCl$: A Chemoselective Base for the Directed Zincation of Sensitive Arenes and Heteroarenes

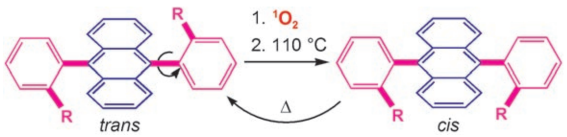


Positive zincing: A wide range of polyfunctional aryl and heteroaryl zinc reagents were efficiently prepared in THF by direct zincation using $(tmp)_2Zn\cdot2MgCl_2\cdot2LiCl$ (**1**), an exceptionally active base. Ester and cyano functions as well as aldehydes and nitro groups are tolerated. dba = dibenzylideneacetone, tmp = 2,2,6,6-tetramethylpiperidide.

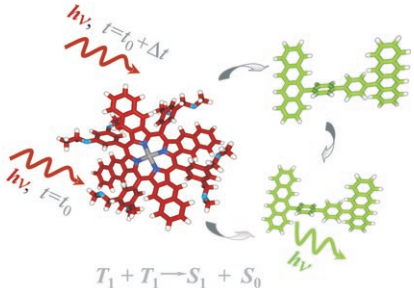
Molecular Devices

D. Zehm, W. Fudickar,
T. Linker* ______ **7689 – 7692**

Molecular Switches Flipped by Oxygen



Singlet oxygen fuels anthracenes and forces an axial rotation of aryl substituents during the oxidation. The molecular switch can be synthesized in only one step from commercially available starting materials. Thermal cleavage of the resulting endoperoxides proceeds quantitatively and affords a simple 180° switch with oxygen as the only waste product. The initial *trans* state is attained by heating, and repetitive cycles are possible.



An emitter/sensitizer couple (see picture; C red/green, H white, N blue, Pd gray) was specially designed for the process of noncoherently excited photon up-conversion. The hypsochromic shift between the energy of the excitation photons and the emitted photons is about 0.7 eV, and an external quantum yield of 0.04 is achieved. As noncoherent excitation source, the near-infrared part of the solar spectrum was used.

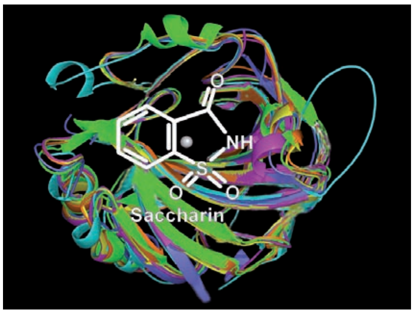
Fluorescence

S. Baluschev,* V. Yakutkin, T. Miteva,*
Y. Avlasevich, S. Chernov, S. Aleshchenkov,
G. Nelles, A. Cheprakov, A. Yasuda,
K. Müllen, G. Wegner ______ **7693 – 7696**

Blue-Green Up-Conversion: Noncoherent Excitation by NIR Light

---

A matter of taste: The sweetner saccharin (a cyclic sulfimide, see picture) is nearly completely absorbed and eliminated through the urine, and is thus exposed to many different proteins in the body. It binds at nanomolar levels to some carbonic anhydrases and this provokes the question of its inert properties. It is known that the plasma level slowly decreases after oral dosing, and CAVI binding could explain its unpleasant metallic aftertaste.



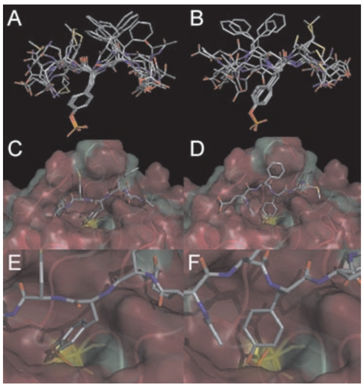
Sweeteners

K. Köhler, A. Hillebrecht,
J. Schulze Wischeler, A. Innocenti,
A. Heine, C. T. Supuran,
G. Klebe* ______ **7697 – 7699**

Saccharin Inhibits Carbonic Anhydrases: Possible Explanation for its Unpleasant Metallic Aftertaste

---

Which substrate will it be? Phosphotyrosine peptide microarrays have allowed the substrate specificity to be mapped for two prototypical protein-tyrosine phosphatases (PTPs): PTP1B and PTPμ. The knowledge gained was used for molecular docking studies (see picture: the docking of a peptide in the binding pocket of PTPμ) and the design of an inhibitor for PTPμ.



Microarrays

M. Köhn, M. Gutierrez-Rodriguez,
P. Jonkheijm, S. Wetzel, R. Wacker,
H. Schroeder, H. Prinz, C. M. Niemeyer,
R. Breinbauer, S. E. Szedlacsek,
H. Waldmann* ______ **7700 – 7703**

A Microarray Strategy for Mapping the Substrate Specificity of Protein Tyrosine Phosphatase

---

With the ferrocene imidazoline palladacycle FIP-Cl as precatalyst, quaternary N-substituted stereocenters can be generated in an asymmetric aza-Claisen rearrangement. Excellent enantioselectivities are obtained even if R and R′ have a similar or identical size (see scheme: e.g. 96% *ee* for CH$_3$/CD$_3$).

Asymmetric Rearrangements

D. F. Fischer, Z.-q. Xin,
R. Peters* ______ **7704 – 7707**

Asymmetric Formation of Allylic Amines with N-Substituted Quaternary Stereocenters by Pd^{II}-Catalyzed Aza-Claisen Rearrangements

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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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