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:

*L. Shi, D. Lundberg, D. G. Musaev, F. M. Menger**
[12]Annulene Gemini Compounds: Structure and Self-Assembly

M. Taoufik, E. Le Roux, J. Thivolle-Cazat, J.-M. Basset**
Discovery of a Catalytic Reaction of Direct Transformation of Ethylene to Propylene, Catalyzed by a Tungsten Hydride Supported on Alumina WH/Al_2O_3: A New Concept of Trifunctional Single-Site Catalyst

*J. K. Jabor, R. Stößer, N. H. Thong, B. Ziemer, M. Meisel**
Unexpected Reactions of $Ag(NCCH_3)_3[(V_2O_3)_2(RPO_3)_4F]$ with H_2 and NO

J. Barluenga, P. Moriel, C. Valdés, F. Aznar*
N-Tosylhydrazones as Reagents for Cross-Coupling Reactions: A Novel Route for the Synthesis of Polysubstituted Olefins

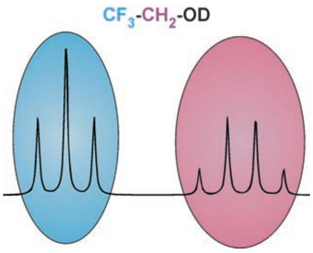
C. Wang, Y. Hou, J. Kim, S. Sun**
A General Strategy for Synthesizing FePt Nanowires and Nanorods

*J. Ge, Y. Hu, Y. Yin**
Highly Tunable Superparamagnetic Colloidal Photonic Crystals

Books

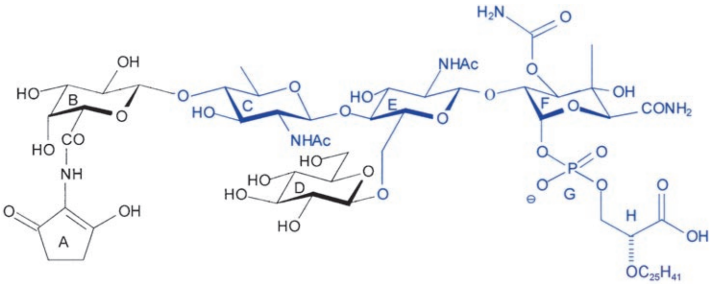
Highlights

Ultralow-Field NMR Spectroscopy



NMR resonances of two nuclei can be obtained in one spectrum at ultralow magnetic fields. At such low fields, chemical shifts become negligible and heteronuclear couplings dominate the NMR spectra. The Highlight gives an overview of "pure *J*-coupling spectroscopy" and (functional) imaging applications at magnetic-field strengths in the range of the Earth's magnetic field.

Antibiotics



The highly active inhibitor of the "forgotten" transglycosylases, moenomycin A, succumbed to total synthesis. The pentasaccharide backbone was constructed employing sulfoxides as glycosyl donors. The recent moenomycin results should establish it as a promising lead structure for new antiinfectives.

Contents

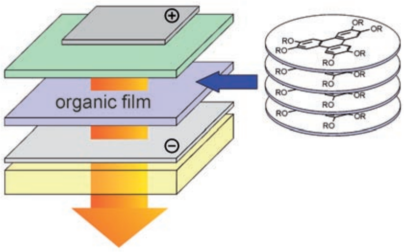
Reviews

Liquid Crystals

S. Laschat,* A. Baro, N. Steinke,
F. Giesselmann,* C. Hägele, G. Scalia,
R. Judele, E. Kapatsina, S. Sauer,
A. Schreivogel, M. Tosoni — **4832 – 4887**

Discotic Liquid Crystals: From Tailor-Made Synthesis to Plastic Electronics

Well stacked? The broad structural diversity and columnar orientation in the liquid-crystalline phase provides access to a variety of potential applications of columnar liquid crystals, such as organic light-emitting diodes (see picture), photovoltaic cells, field-effect transistors, or photoconducting materials in laser printers.

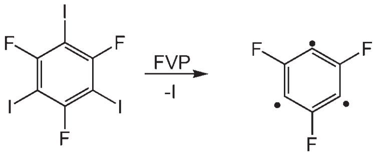


Communications

Dehydroarenes

S. Venkataramani, M. Winkler,
W. Sander* — **4888 – 4893**

Trifluoro-1,3,5-tridehydrobenzene



1 2

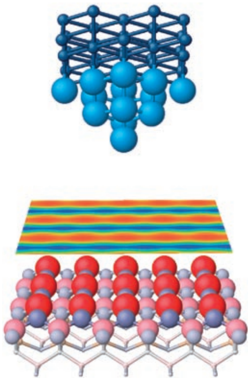
Three stay, three go: Flash vacuum pyrolysis (FVP) of **1** and subsequent trapping of the products in solid argon at 3 K produces trifluoro-1,3,5-tridehydrobenzene (**2**). IR spectroscopy, the photochemical behavior, and quantum chemical calculations confirm that for the first time a derivative of the elusive 1,3,5-tridehydrobenzene could be isolated and spectroscopically characterized.

Surface Chemistry

R. Kováčik, B. Meyer,*
D. Marx — **4894 – 4897**

F Centers versus Dimer Vacancies on ZnO Surfaces: Characterization by STM and STS Calculations

Not what you'd expect: Contrary to what is usually believed for oxide surfaces, density functional calculations show that not *F* centers but missing ZnO dimers are the thermodynamically most favorable type of atomic defect on the nonpolar ZnO surface. Simulated STM data may serve as a fingerprint for experimental identification of the different surface defect types. The picture shows a tunneling tip over ZnO (Zn gray, O red, W blue) with the corresponding STM image in between.



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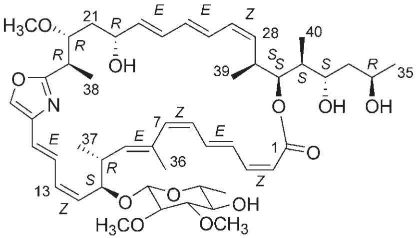
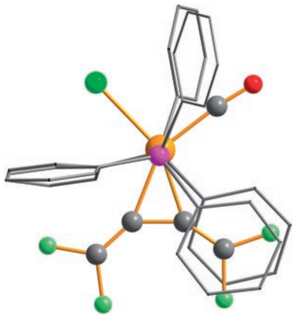


1

The best of both worlds: The configurational assignment of a natural product—chivosazole A (**1**) isolated from myxobacteria—has been achieved for the first time through the combination of structure elucidation and genetic analysis. This was achieved by chemical degradation, analysis of the NMR data in combination with computer-aided conformation analysis, and assignment of the amino acid sequence.

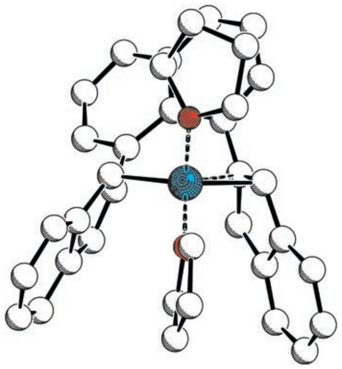
Structure Elucidation

Now tamed: For decades preparative research on 1,1,4,4-tetrafluorobutatriene was not pursued because of the explosive nature of the compound. Two new transition-metal complexes containing a tetrafluorobutatriene ligand (see picture; green F/Cl, gray C, red O, purple P, orange Ir/Rh) have been prepared from the free triene, and both complexes are air-stable.



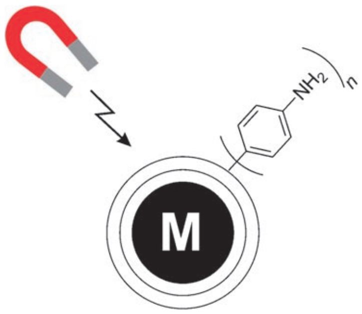
Cumulene Complexes

Zinc has the ansa: Deprotonation of the ligand 1,8-bis(indenyl)naphthalene followed by treatment with $ZnCl_2$ gives an ansa-zincocene. Its crystal structure reveals a σ/(σ,π) coordination of the indenyl units to the central zinc atom (see structure; C white, O red, Zn blue).

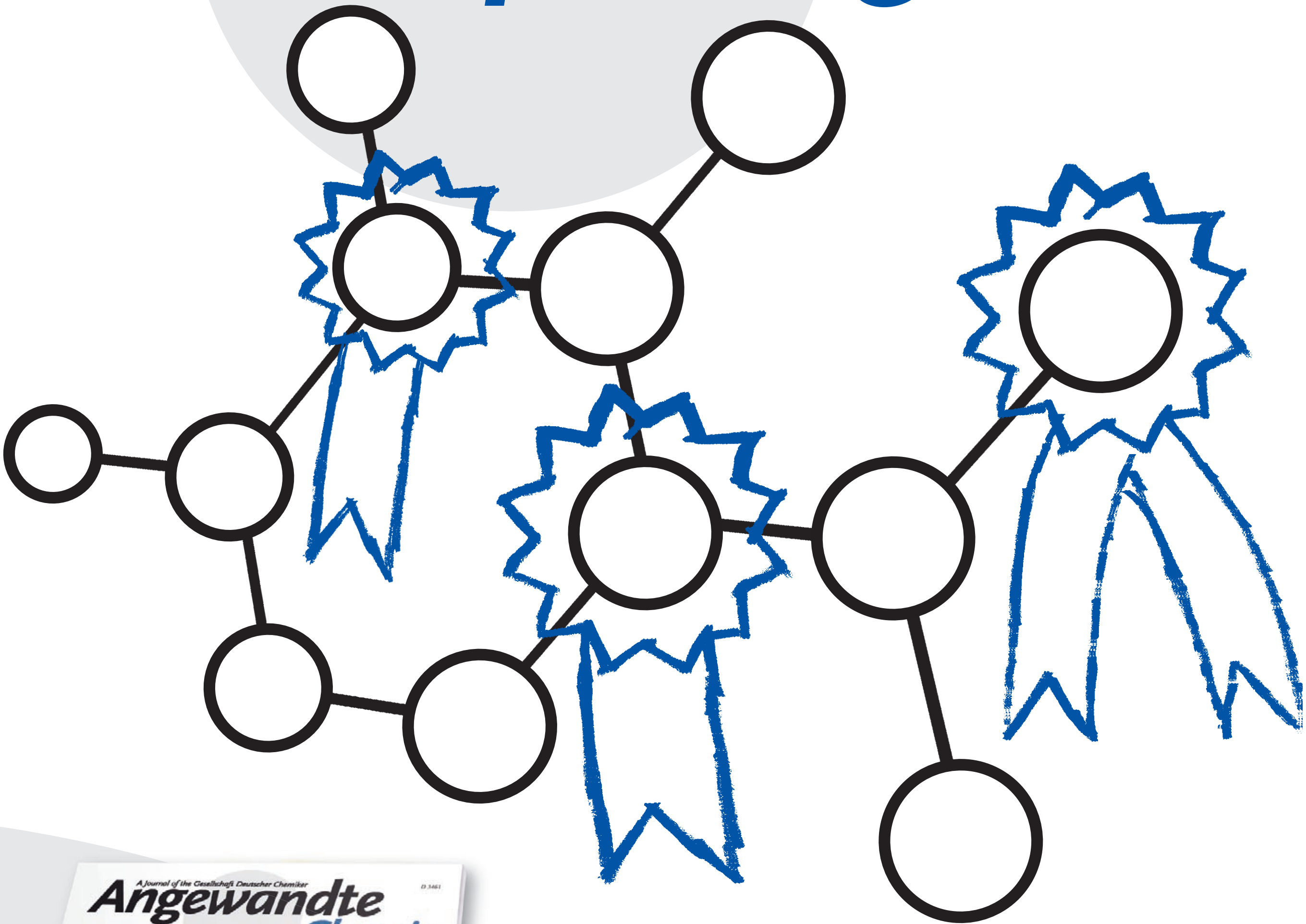


ansa-Metallocenes

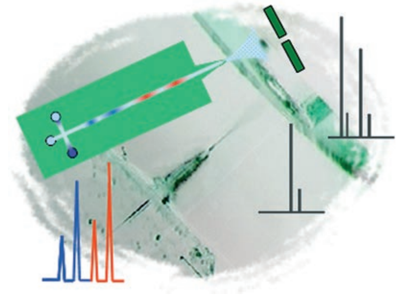
Synthesis in a day! Carbon-coated metal nanoparticles can be covalently functionalized by diazonium chemistry. These colloidal reagents can now serve as a basis to magnetically functionalize molecules during synthesis, enabling their recovery within seconds.



Functional Nanobeads

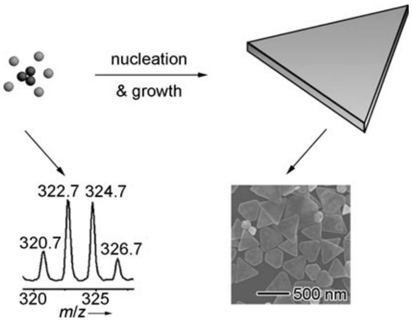


Lab-on-a-Chip



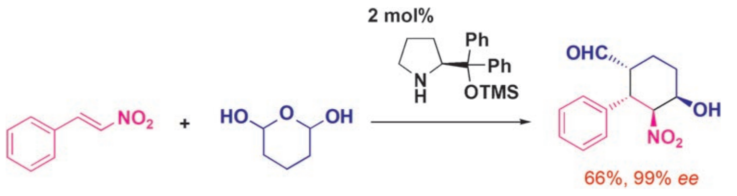
A coupling coup: A novel microfluidic chip with an integrated nanospray emitter enables the first dead-volume-free coupling of glass-chip laboratories with mass spectrometry. An electrophoretic version of the system may be suitable for the separation and analysis of drugs, for example, and has potential for the high-throughput analysis of miniscule amounts of samples.

Silver Nanostructures



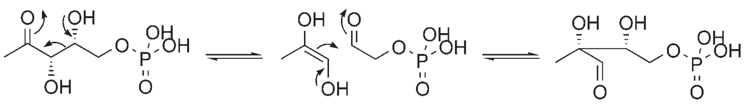
Three's a crowd: Trimeric silver clusters in aqueous $AgNO_3$ solutions have been identified by mass spectrometry (see picture). These clusters are found to dominate nucleation and thus play an important role in the formation of triangular silver nanoplates, which are shown in the SEM image in the lower right part of the picture.

Tandem Reactions



A choice of three: The tandem Michael/Henry reaction of a nitroalkene and pentane-1,5-dial (generated in situ) proceeded efficiently when diphenylprolinol silyl ether was used as an organocatalyst to afford substituted nitrocyclohexanecarbaldehydes with high diastereo- and enantioselectivity (see scheme; TMS: trimethylsilyl). Isomerization under basic or acidic conditions diastereoselectively converts the product into two stereoisomers, without compromising the enantioselectivity.

Enzyme Mechanisms



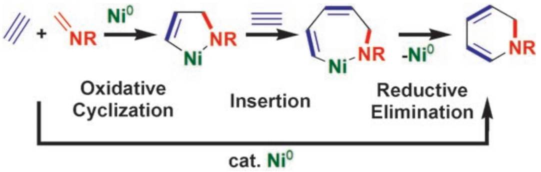
Find the way: 3-[^{2}H]- and 4-[^{2}H]-labeled 1-deoxyxylulose-5-phosphate were synthesized and used to investigate the chemical mechanism of 1-deoxyxylulose-5-phosphate reductoisomerase (DXR) from *E. coli*. The observation of inverse secondary kinetic isotope effects for both labeled substrates indicates that DXR uses a retro-aldol/aldol mechanism in which the recombination reaction is the rate-limiting step (see scheme).

Contents

Nickel-Catalyzed Cycloaddition

S. Ogoshi,* H. Ikeda,
H. Kurosawa **4930–4932**

Formation of an Aza-nickelacycle by Reaction of an Imine and an Alkyne with Nickel(0): Oxidative Cyclization, Insertion, and Reductive Elimination

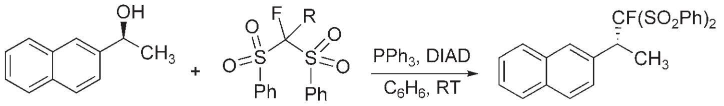


Cyclize, add, reduce: The oxidative cyclization of an imine and an alkyne with nickel(0) followed by the insertion of a second alkyne gives a seven-membered aza-nickelacycle (see scheme). Subsequent reductive elimination yields a 1,2-dihydropyridine. This sequential reaction process is expanded to a one-step nickel-catalyzed [2+2+2] cycloaddition of two alkynes and an imine.

Stereoselective Fluoroalkylation

G. K. S. Prakash,* S. Chacko, S. Alconcel,
T. Stewart, T. Mathew,
G. A. Olah* **4933–4936**

Stereoselective Monofluoromethylation of Primary and Secondary Alcohols by Using a Fluorocarbon Nucleophile in a Mitsunobu Reaction

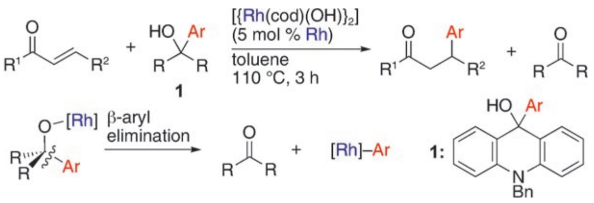


OH is exchanged: An efficient Mitsunobu reaction that uses a fluorinated carbon pronucleophile for the facile synthesis of monofluoromethyl derivatives of alcohols is reported (see scheme; DIAD = diisopropyl azodicarboxylate). This reaction can be performed under mild conditions and is highly feasible for primary, secondary, allylic, benzylic, and alicyclic alcohols. Excellent enantiospecificity is observed for chiral alcohols.

β-Aryl Elimination

T. Nishimura,* T. Katoh,
T. Hayashi* **4937–4939**

Rhodium-Catalyzed Aryl Transfer from Trisubstituted Aryl Methanols to α,β-Unsaturated Carbonyl Compounds



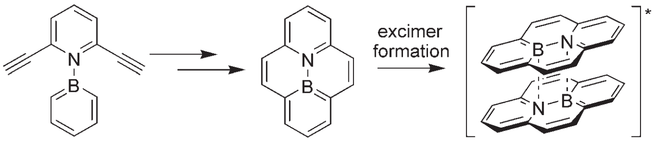
A process of elimination: The rhodium-catalyzed arylation of α,β-unsaturated carbonyl compounds with 9-aryl-10-benzyl-9,10-dihydroacridin-9-ols **1** as arylating reagents proceeds efficiently via β-aryl elimination of the rhodium alkoxide intermediates, to give the 1,4-addition products in high yields (see scheme; Bn = benzyl, cod = cycloocta-1,5-diene).

VIP

Heterocyclic Compounds

M. J. D. Bosdet, W. E. Piers,*
T. S. Sorensen, M. Parvez **4940–4943**

10a-Aza-10b-borapyrenes: Heterocyclic Analogues of Pyrene with Internalized BN Moieties

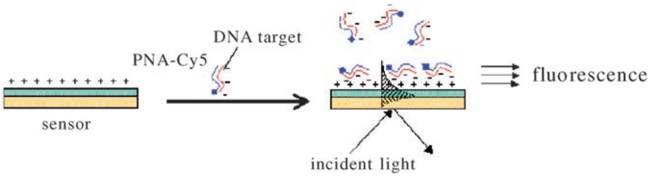


Très BN: The introduction of a B–N dipole into the internal positions of pyrene (see scheme) provides a novel platform for heterocyclic fluorescent species. Comparisons of the photophysical and redox properties between the CC and BN analogues reveal both similarities and differences.

DNA Detection

L.-Q. Chu, R. Förch,
W. Knoll* ———— **4944 – 4947**

Surface-Plasmon-Enhanced Fluorescence Spectroscopy for DNA Detection Using Fluorescently Labeled PNA as "DNA Indicator"

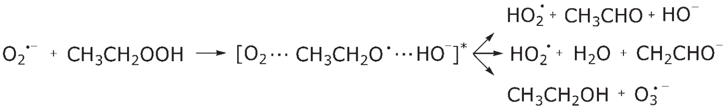


A homogeneous sensor: A new method for the detection of DNA is described. The approach is based on the high surface sensitivity provided by surface-plasmon-enhanced fluorescence spectroscopy and on the different electrostatic properties of peptide nucleic acids (PNAs) and DNA (see scheme). Immobilization of the DNA probes on the sensor surface is not required, which may reduce the complexity and costs of sensor preparation.

Gas-Phase Reactions

S. J. Blanksby,* V. M. Bierbaum,
G. B. Ellison, S. Kato* ———— **4948 – 4950**

Superoxide Does React with Peroxides: Direct Observation of the Haber–Weiss Reaction in the Gas Phase



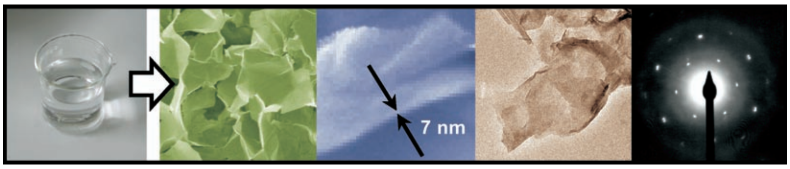
$$O_2^{\bullet-} + CH_3CH_2OOH \rightarrow [O_2\cdots CH_3CH_2O^{\bullet}\cdots HO^-]^* \rightarrow \begin{cases} HO_2^{\bullet} + CH_3CHO + HO^- \\ HO_2^{\bullet} + H_2O + CH_2CHO^- \\ CH_3CH_2OH + O_3^{\bullet-} \end{cases}$$

Radically speaking: Reactions of superoxide with hydrogen peroxide and organic hydroperoxide compounds are observed as efficient processes in the gas phase and yield products—including the ozonide anion—that are consistent with the one-electron reduction of the peroxide species (see scheme).

Nanostructures

Y. Oaki, H. Imai* ———— **4951 – 4955**

One-Pot Synthesis of Manganese Oxide Nanosheets in Aqueous Solution: Chelation-Mediated Parallel Control of Reaction and Morphology

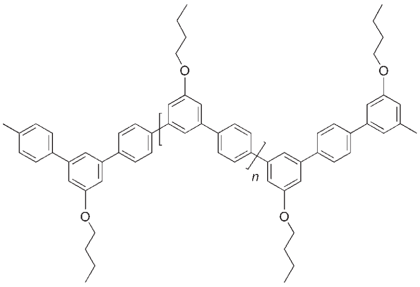


Sheet materials: Manganese oxide nanosheets and their oriented thin films can be prepared by a bottom-up approach in aqueous solution under ambient conditions (left). Chelation between the manganese species and ethylenediaminetetraacetate (EDTA) ligands results in birnessite-type nanosheets (see micrographs and diffraction pattern). The biomimetic pathway allows simultaneous control of synthesis and morphogenesis by the EDTA chelating agent.

Polymer Synthesis

R. Kandre, K. Feldman, H. E. H. Meijer,
P. Smith,* A. D. Schlüter* — **4956 – 4959**

Suzuki Polycondensation Put to Work: A Tough Poly(*meta*-phenylene) with a High Glass-Transition Temperature



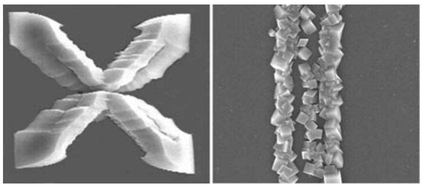
Challenging commercial polycarbonate: Suzuki polycondensation afforded a high-molar-mass poly(*meta*-phenylene) (see structure) which displays not only outstanding toughness, convenient processability, and a high glass-transition temperature, but also considerable resistance against environmental stress-cracking; thus, it rivals high-performance polycarbonate in its fundamental properties.

Contents

Microcrystals

I. S. Chun, J. A. Kwon, H. J. Yoon,
M. N. Bae, J. Hong,
O.-S. Jung* **4960 – 4963**

Interactions of Microcrystals through Free Pyridyl Groups: Microcross and Chain-Arrayed Supercrystals of a Palladium(II) Complex

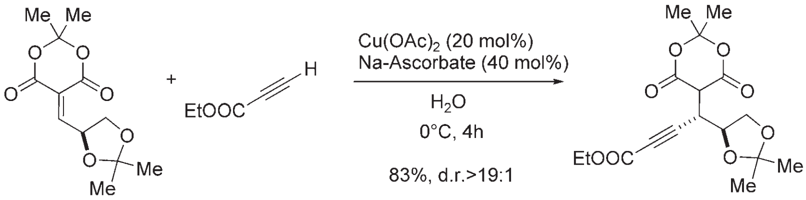


Crosses or chains? The two free pyridyl groups of a dimeric, triethanolamine-triisonicotinate-bridged palladium complex allow the formation of a unique microcross morphology or chain array of microcrystals (see picture), depending on the reaction conditions. These morphologies arise from interactions between microcrystals involving hydrogen bonding of the free pyridyl groups on the microcrystal surface and water molecules.

Conjugate Addition

S. Fujimori, E. M. Carreira* **4964 – 4967**

Cu^I-Catalyzed Conjugate Addition of Ethyl Propiolate



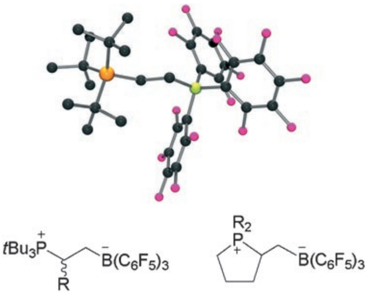
More functionality possible: The conjugate addition of ethyl propiolate to Meldrum's acid derived acceptors is described. These reactions proceed under mild conditions employing substoichiometric amounts of a copper(I) species generated in situ. When chiral acceptors are employed as substrates, the additions are stereoselective and provide the addition adduct with high diastereoselectivity.

Addition Reactions

J. S. J. McCahill, G. C. Welch,
D. W. Stephan* **4968 – 4971**

Reactivity of "Frustrated Lewis Pairs": Three-Component Reactions of Phosphines, a Borane, and Olefins

All or nothing: Sterically frustrated Lewis pairs of phosphines and the borane $B(C_6F_5)_3$ exhibit unprecedented reactivity with olefins to afford both inter- and intramolecular alkanediyl-linked phosphonium borates (see picture; black C, pink F, green B, orange P). These reactions are all the more remarkable given that any pair of these reagents do not react while the combination of all three reagents results in product formation.

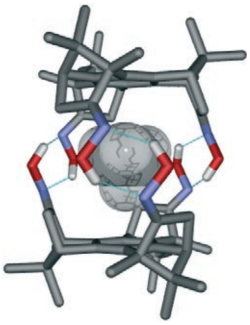


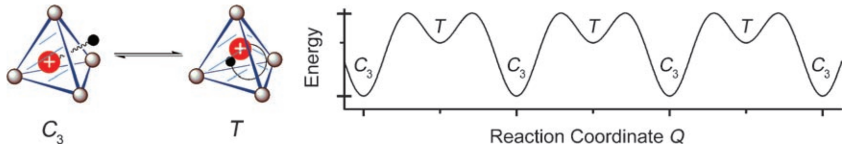
Host–Guest Systems

A. Scarso,* L. Pellizzaro, O. De Lucchi,
A. Linden, F. Fabris* **4972 – 4975**

Gas Hosting in Enantiopure Self-Assembled Oximes

Trapped gas: The enantiopure C_3-symmetric trioxime derived from (+)-*syn*-benzotricamphor forms a dimer in low-polar solvents which encapsulates small molecules, such as Ar, CO, O_2, N_2, and CH_4 (see picture; gray C, blue N, red O, white H). The two benzocyclotrimers are fastened together by six hydrogen bonds between the oxime moieties. Gas binding is fast on the NMR timescale at room temperature, but is slow and complete at lower temperature.





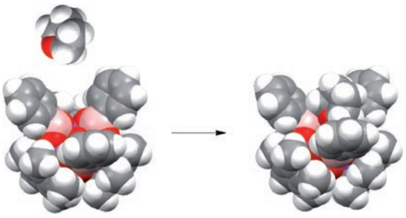
A quick in and out: A supramolecular cluster encapsulates a cation guest with a linear alkyl side chain, which rapidly extends and retracts through apertures in the triangular faces of the tetrahedral host. This motion results in a dynamic second-order Jahn–Teller distortion (see picture).

Jahn–Teller Distortion

B. E. F. Tiedemann,
K. N. Raymond* ——— **4976 – 4978**

Second-Order Jahn–Teller Effect in a Host–Guest Complex

Open and shut cases: Tantalum(V) boronate clusters $[(Cp^*Ta)_3(\mu^2\text{-}\eta^2\text{-}RBO_2)_3(\mu^2\text{-}O)_2(\mu^2\text{-}OH)(\mu^3\text{-}OH)]$ ($Cp^* = \eta^5\text{-}C_5Me_5$; **1**: R = Ph, **2**: R = *i*Bu) with Lewis acidic cavities were prepared. Whereas the cavity of **2** is blocked by the *i*Bu groups, that of **1** is open and can bind Lewis basic guests such as ketones (see picture) by interaction with one boronate and one μ^3-OH ligand.



Molecular Cages

O. Sigouin, C. N. Garon, G. Delaunais,
X. Yin, T. K. Woo, A. Decken,
F.-G. Fontaine* ——— **4979 – 4982**

Synthesis and Characterization of Tantalum(V) Boronate Clusters: Multifunctional Lewis Acid Cages for Binding Guests

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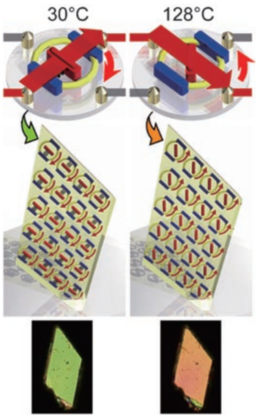

Going 'round the twist: Single crystals of a ferrocenyl-substituted pseudorotaxane have different structures at 30 and 128 °C owing to a thermal crystalline phase transition. This change in crystal structure results from a conformational change that involves rotation of the aromatic ring of the axle component (see scheme), and cooperative unidirectional rotation of the molecules alters the optical anisotropy of the crystal.

Molecular Switches

M. Horie,* T. Sassa, D. Hashizume,
Y. Suzaki, K. Osakada,*
T. Wada ——— **4983 – 4986**

A Crystalline Supramolecular Switch: Controlling the Optical Anisotropy through the Collective Dynamic Motion of Molecules

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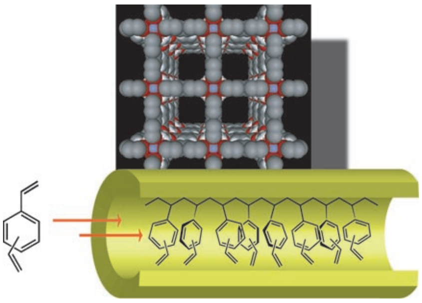


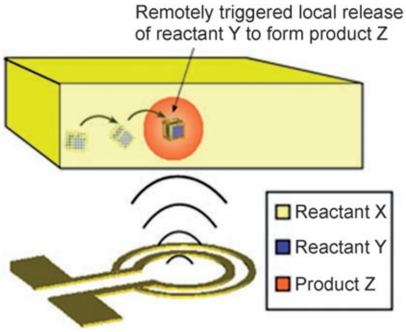
In the groove: Radical polymerization of divinylbenzenes (DVBs) in the one-dimensional nanochannels of porous coordination polymers allows linearly extended topotactic polymerization without cross-linking (see picture). The main factors that influence this polymerization are the channel size and host framework flexibility.

Topotactic Polymerization

T. Uemura, D. Hiramatsu, Y. Kubota,
M. Takata, S. Kitagawa* —— **4987 – 4990**

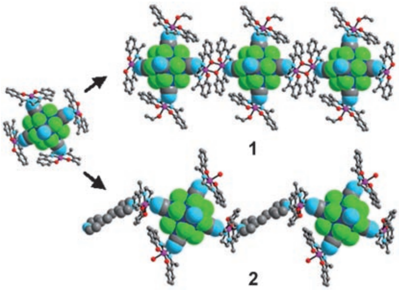
Topotactic Linear Radical Polymerization of Divinylbenzenes in Porous Coordination Polymers

Analytical Chemistry



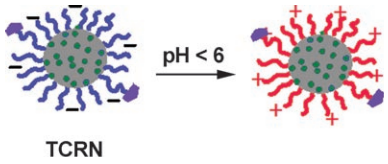
A matter of control: Metallic, self-assembled nanoliter containers have been prepared that can be remotely guided in any chosen spatial trajectory by a magnetic field, as well as remotely triggered by radio-frequency electromagnetic fields to release chemical reactants (see picture). The containers were utilized to facilitate, on demand, spatially localized microfabrication in a capillary and non-invasive chemical delivery to living cells.

Cluster Compounds



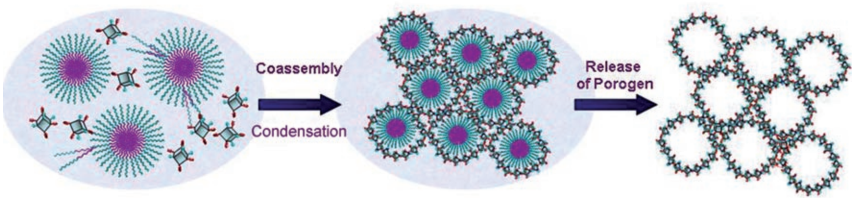
From five to infinity: The pentameric cluster-based supramolecular units $\{[Mn(L)]_4[Nb_6Cl_{12}(CN)_6]\}$ (H_2L = 7-Me-salen) can be extended into two 1D coordination polymers through either the formation of manganese dimers or through ditopic ligands (see picture). In **1** the supramolecular units are linked through phenoxo bridges between the Mn complexes, while in **2** a *trans*-1,2-bis(4-pyridyl)ethylene ligand connects the pentameric assemblies.

Drug Delivery



Reversing the charges: Targeted charge-reversal nanoparticles (TCRNs) comprised of poly(ε-caprolactone)-*block*-polyethyleneimine (PCL-PEI), whose amine groups are converted into amides, are negatively charged at neutral pH but become positively charged at pH < 6 (see picture). TCRNs effectively enter cells, regenerate the PEI layer in lysosomes, and localize in the nucleus for nuclear drug delivery.

Mesoporous Polysilsesquioxanes

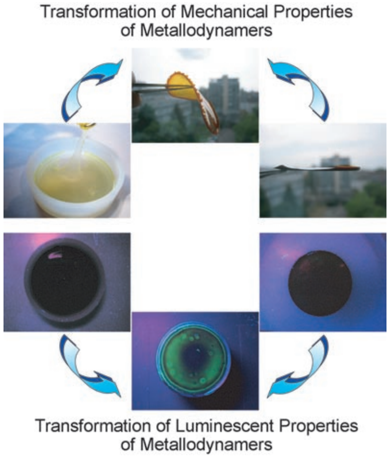


Rounding up the POSSe: Polyhedral oligomeric silsesquioxanes (POSS), one of the smallest hybrid nano building blocks, were weaved into multifunctional hierarchical mesoporous hybrid materials. The mesoporous organic–inorganic hybrid material was synthesized by co-assembly and condensation of the POSS compound around the block copolymer P123 micelles and subsequent release of the porogen (see scheme), resulting in a uniform pore size of 4.40 nm.

Dynamic Polymers

C.-F. Chow, S. Fujii,
J.-M. Lehn* ______ 5007 – 5010

Metallodynamers: Neutral Dynamic Metallosupramolecular Polymers Displaying Transformation of Mechanical and Optical Properties on Constitutional Exchange

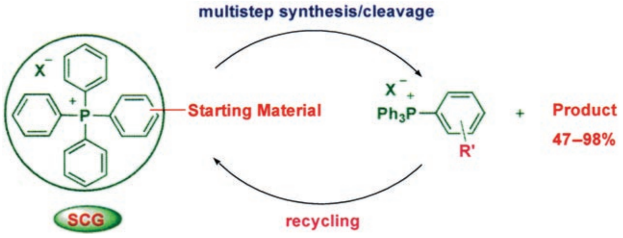


Mix and match: Neutral dynamic metallosupramolecular polymers (metallodynamers) derived from acylhydrazone-based metal coordination centers exchange and reshuffle their components through ligand exchange at the metal coordination site, even in the absence of solvent and catalyst, to modify their constitution. As a result, the materials undergo remarkable changes in both their mechanical and optical properties (see picture).

Synthetic Methods

F. Stazi, D. Marcoux, J.-C. Poupon,
D. Latassa, A. B. Charette* 5011 – 5014

Tetraarylphosphonium Salts as Soluble Supports for the Synthesis of Small Molecules



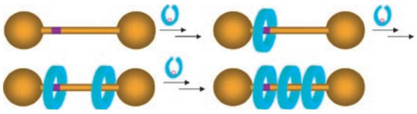
Support groups: Phosphonium salts can be used as solubility-control groups (SCGs) in small-molecule and peptide synthesis (see scheme). The concept, which is a hybrid approach between solution- and solid-phase syntheses, is illustrated by the preparation of 2-substituted piperidine derivatives and a variety of small peptides with high yields and purities. In some cases, recycling of the support is possible.

Rotaxane Synthesis

VIP

A.-M. L. Fuller, D. A. Leigh,*
P. J. Lusby ______ 5015 – 5019

One Template, Multiple Rings: Controlled Iterative Addition of Macrocycles onto a Single Binding Site Rotaxane Thread

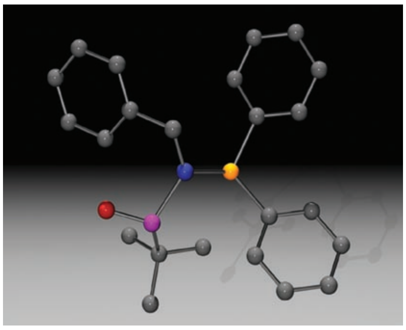


One for all: The iteration of three simple steps—complexation, macrocyclization, and demetalation—enables the synthesis of multi-ring rotaxanes with only one template site (see picture). This efficient and effective strategy enables both the number and the order in which macrocycles are assembled onto a thread to be controlled with unprecedented precision.

P,S Ligands

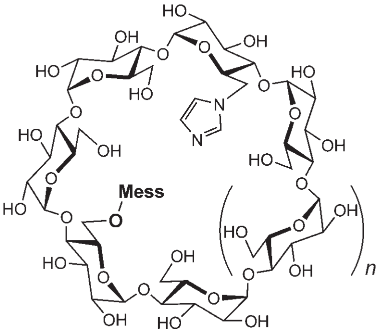
J. Solà, M. Revés, A. Riera,*
X. Verdaguer* ______ 5020 – 5023

N-Phosphino Sulfinamide Ligands: An Efficient Manner To Combine Sulfur Chirality and Phosphorus Coordination Behavior



Four in a row: Four linked heteroatoms (P-N-S-O) constitute the backbone of *N*-phosphino *tert*-butylsulfinamides, a novel class of chiral ligands (see picture, P yellow, N blue, S magenta, O red). Carbon substituents surrounding the central heteroatomic spine transmit the chirality of the sulfur center throughout the ligand. As bidentate P,S ligands in dicobalt–alkyne complexes, they can be used in the asymmetric intermolecular Pauson–Khand reaction.

Cyclodextrins



Remote control: The imidazolyl group in a series of imidazolyl cyclodextrins guides the sulfonylation of a specific hydroxy group in the presence of many others to effect a regioselective 1,22 remote functionalization (see structure; Mess = mesitylenesulfonyl, $n = 0–2$). This process enables the establishment of a novel efficient strategy for the hetero-bifunctionalization and -trifunctionalization of cyclodextrins.

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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Juniorprofessur (W1) für Anorganische Chemie

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