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:*

*M. Alcarazo, T. Stork, A. Anoop, W. Thiel, A. Fürstner**
Steering the Surprisingly Modular π-Acceptor Properties of N-Heterocyclic Carbenes: Implications for Gold Catalysis

*D. Bojer, A. Venugopal, B. Neumann, H.-G. Stammler, N. W. Mitzel**
Lewis Base Induced Reductions in Organolanthanide Chemistry

S.-H. Kim, Su Y. Lee, S.-M. Yang**
Janus Microspheres for Highly Flexible and Impregnable Water-Repelling Interfaces

*L. Frullano, C. Catana, T. Benner, A. D. Sherry, P. Caravan**
A Bimodal MR-PET Agent for Quantitative pH Imaging

*K. Schober, E. Hartmann, H. Zhang, R. M. Gschwind**
^{1}H DOSY Spectra of Highly Enantioselective Ligands: A Fast and Simple NMR-Specroscopy Method to Optimize Catalytic Reaction Conditions

*A. M. Scott, A. B. Ricks, M. T. Colvin, M. R. Wasielewski**
Comparing Spin-Selective Charge Transport through Donor-Bridge–Acceptor Molecules having Different Oligomeric Aromatic Bridges

*D. Figgen, A. Koers, P. Schwerdtfeger**
NWHClI: A Small and Compact Chiral Molecule with Large Parity Violation Effects in the Vibrational Spectrum

S. Pal, Z. Deng, B. Ding, H. Yan, Y. Liu**
DNA-Origami-Directed Self-Assembly of Discrete Silver Nanoparticle Architectures

A. M. Nowicka, U. Hasse, G. Sievers, M. Donten, Z. Stojek, S. Fletcher, F. Scholz**
Selective Knock–Out of Active Sites on a Gold Surface

C. Costentin, M. Robert, J. Savéant, C. Tard
Inserting a Hydrogen Bond Relay between Proton Exchanging Sites in Proton–Coupled Electron Transfers

Author Profile

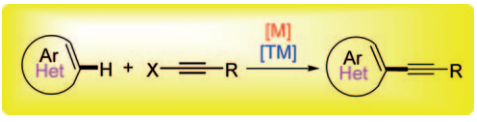
"My favorite piece of research is to simulate and elucidate processes of biological regulation through structurally specified models.
The part of my job which I enjoy the most is that ideas can be proven by experiments …"
This and more about Horst Kunz can be found on page 2092.

Horst Kunz ____ **2092**

Books

Synthesis of Solid Catalysts — Krijn P. de Jong — *reviewed by M. Behrens* ____ **2095**

Highlights



Getting straight to the point: Recent developments in the direct functionalization of unreactive C–H bonds have established the title reaction (see scheme). Various main-group (M) and transition metals (TM) serve as efficient promoters or catalysts for this transformation.

C–H Activation

A. S. Dudnik, V. Gevorgyan* **2096 – 2098**

Formal Inverse Sonogashira Reaction: Direct Alkynylation of Arenes and Heterocycles with Alkynyl Halides

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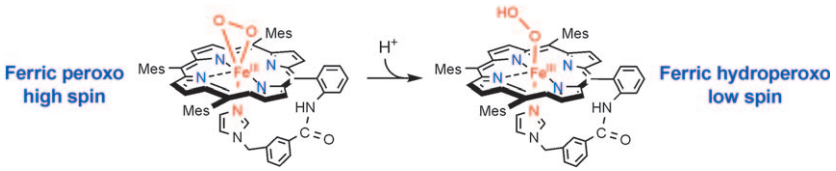
Contents

Bioinorganic Chemistry

S. P. de Visser, J. S. Valentine, W. Nam* ——— **2099 – 2101**

A Biomimetic Ferric Hydroperoxo Porphyrin Intermediate



OOH, my! The protonation of a side-on high-spin ferric peroxo species yields the corresponding end-on low-spin ferric hydroperoxo intermediate (see picture), which is a precursor of Compound I and has been frequently proposed as a reactive species in heme enzymes. This ferric hydroperoxo complex can be used to study reactivities of similar species in substrate oxygenation and hydroperoxide O–O bond cleavage reactions.

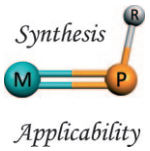
Minireviews

Phosphinidenes

H. Aktaş, J. C. Slootweg, K. Lammertsma* ——— **2102 – 2113**

Nucleophilic Phosphinidene Complexes: Access and Applicability

Make it and break it! An overview of the different methodologies to synthesize nucleophilic phosphinidene complexes is presented. The emerging applicability of this class of phosphorus reagents as phosphinidene transfer agents that also posses the established reactivity of the isoelectronic metal-complexed carbenes makes them viable synthetic targets for a broad spectrum of chemical conversions.

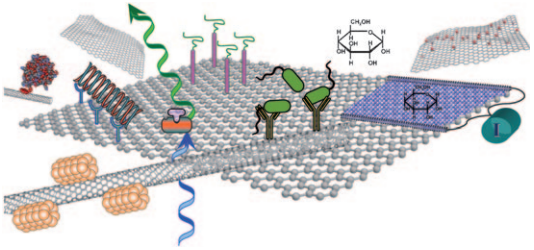


Reviews

Biosensors and Carbon Nanomaterials

W. Yang, K. R. Ratinac, S. P. Ringer, P. Thordarson, J. J. Gooding, F. Braet* ——— **2114 – 2138**

Carbon Nanomaterials in Biosensors: Should You Use Nanotubes or Graphene?



Coiled or unrolled? Many research groups are working to incorporate carbon nanotubes or graphene into prototype biosensors (see picture). This Review critically explores the latest advances in electrochemical, electrical, and optical biosensors that use carbon nanotubes and graphene, and compares the biosensing performance of these unique allotropes of carbon.

For the USA and Canada:
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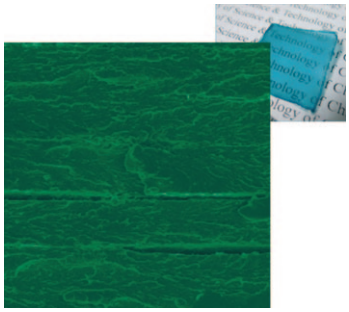
Communications



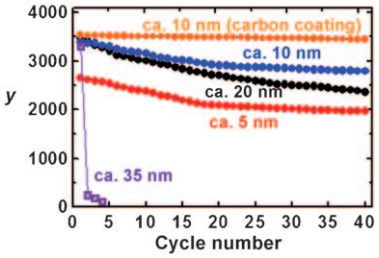
Like mother nature: Biologically inspired organic–inorganic hybrid films comprising alternating layers of chitosan and micro- and nanoplatelets of layered double hydroxides mimic the unique layered microstructures of seashell nacre (see picture) and display high tensile strength. Moreover, the hybrid films can be endowed with the optical properties of the platelets.

Hybrid Composites

H. B. Yao, H. Y. Fang, Z. H. Tan, L. H. Wu, S. H. Yu* ——— **2140 – 2145**

Biologically Inspired, Strong, Transparent, and Functional Layered Organic–Inorganic Hybrid Films

Well-dispersed Si nanocrystals with sizes of approximately 5, 10, and 20 nm were prepared in reverse micelles at high pressure and 380 °C and investigated as anode materials for lithium batteries. The 10 nm sized nanocrystals show a first charge capacity y of 3380 mAh g^{-1} and the highest capacity retention of 81 % after 40 cycles, which can be increased to 96 % by carbon coating (see picture).

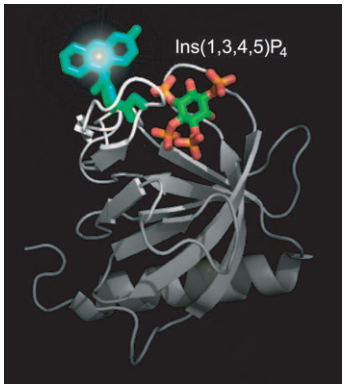


Lithium Batteries

H. Kim, M. Seo, M.-H. Park, J. Cho* ——— **2146 – 2149**

A Critical Size of Silicon Nano-Anodes for Lithium Rechargeable Batteries

Don't shoot the messenger! An optical sensor for D-*myo*-inositol 1,3,4,5-tetrakisphosphate ($Ins(1,3,4,5)P_4$), an intracellular second messenger, is constructed by introducing a fluorophore at a unique cysteine residue within a mutant of the pleckstrin homology (PH) domain of the general receptor for phosphoinositides 1 (GRP1). The biosensor visualizes the $Ins(1,3,4,5)P_4$ dynamics by agonist stimulation in single live cells.



Biosensors

R. Sakaguchi, K. Tainaka, N. Shimada, S. Nakano, M. Inoue, S. Kiyonaka, Y. Mori, T. Morii* ——— **2150 – 2153**

An In Vivo Fluorescent Sensor Reveals Intracellular $Ins(1,3,4,5)P_4$ Dynamics in Single Cells

Flexisense: Films of graphene oxide and reduced graphene oxide are printed onto a flexible plastic surface (see picture), using inkjet techniques, which are used to detect chemically aggressive vapors such as NO_2 and Cl_2. Vapors in the 100 ppm–500 ppb concentration range can be detected in an air sample without the aid of a vapor concentrator.

Graphene Sensors

V. Dua, S. P. Surwade, S. Ammu, S. R. Agnihotra, S. Jain, K. E. Roberts, S. Park, R. S. Ruoff,* S. K. Manohar* ——— **2154 – 2157**

All-Organic Vapor Sensor Using Inkjet-Printed Reduced Graphene Oxide

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Frontiers of Chemistry:
From Molecules to Systems
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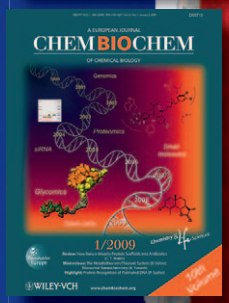

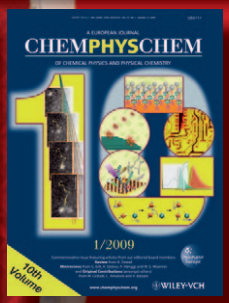

On 21st May 2010 in Paris
at the Maison de la Chimie
(near the Eiffel Tower and Les Invalides)

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Nobel Prize 2007


Jean-Marie Lehn
Nobel Prize 1987


Roger Y. Tsien
Nobel Prize 2008


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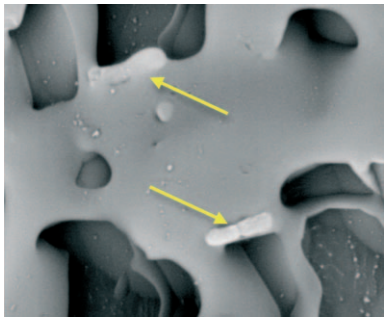
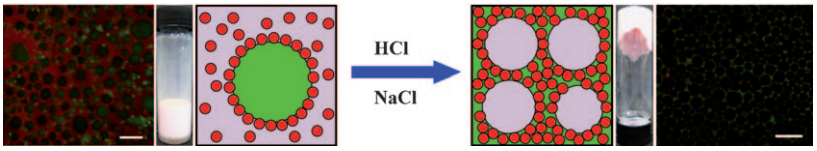


Taking fish out of water: Freeze-drying can be used to incorporate bacteria in a deep-eutectic solvent with outstanding preservation of integrity and viability. (Intact *E. coli* are marked by arrows in the image.) These findings open interesting perspectives for the use of whole microorganisms in biocatalytic processes carried out in nonaqueous solvents.

Bacteria in Nonaqueous Solution

M. C. Gutiérrez,* M. L. Ferrer, L. Yuste, F. Rojo, F. del Monte* —— **2158 – 2162**

Bacteria Incorporation in Deep-eutectic Solvents through Freeze-Drying

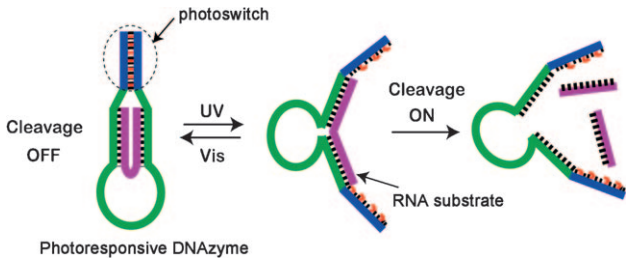


Two triggers: Phase inversion of a particle-stabilized oil–water system from an ordinary oil-in-water emulsion (see picture, left; green: oil, pink: water) to a water-in-oil high-internal-phase emulsion (right) at a fixed oil/water ratio (27:73 vol%) can be simply driven by either a change of pH value or salt concentration in a single system. Scale bars: 30 μm.

Emulsions

G. Sun, Z. Li, T. Ngai* —— **2163 – 2166**

Inversion of Particle-Stabilized Emulsions to Form High-Internal-Phase Emulsions



Photons as fuel: A photoresponsive DNA enzyme was constructed that can work at the single-molecule level. Complete ON–OFF photoswitching of RNA digestion was realized by photoregulating the topological structure of a DNAzyme/RNA complex. The key components of the photoswitch are azobenzene units.

DNAzymes

M. G. Zhou, X. G. Liang,* T. Mochizuki, H. Asanuma* —— **2167 – 2170**

A Light-Driven DNA Nanomachine for the Efficient Photoswitching of RNA Digestion

Eraserhead: A reversible nanoporous antireflection coating can be fabricated by assembly of layered double hydroxide nanoparticles with polyanions using an electrostatic layer-by-layer method followed by calcination. Antireflection properties of the coating can be switched between the porous and non-porous state by alternating calcination and rehydration processes.

Nanotechnology

J. Han, Y. Dou, M. Wei,* D. G. Evans, X. Duan —— **2171 – 2174**

Erasable Nanoporous Antireflection Coatings Based on the Reconstruction Effect of Layered Double Hydroxides

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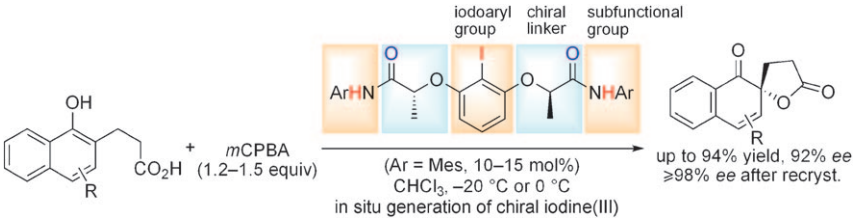
Hypervalent Compounds

M. Uyanik, T. Yasui, K. Ishihara* ——— 2175 – 2177

Enantioselective Kita Oxidative Spirolactonization Catalyzed by In Situ Generated Chiral Hypervalent Iodine(III) Species



The iodines(III) have it: The rational design of a conformationally flexible C_2-symmetric iodosylarene catalyst has been used for the enantioselective Kita oxidative spirolactonization. The reaction occurs through secondary n–σ* or hydrogen-bonding interactions between the chiral catalyst and the substrate. Mes = mesityl (2,4,6-trimethylphenyl).

Guanidines

M.-H. Larraufie, C. Ollivier, L. Fensterbank,* M. Malacria,* E. Lacôte* ——— 2178 – 2181

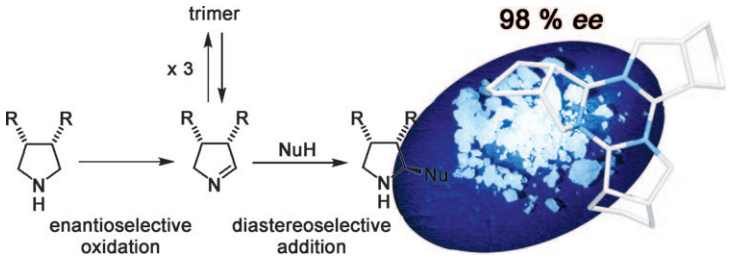
Radical Synthesis of Guanidines from *N*-Acyl Cyanamides

Center stage: Additions of nitrogen-centered radicals to cyanamide compounds provided the first radical synthesis of aromatic polycyclic guanidine derivatives (see scheme). Modular assembly of the substrates allows for a rapid increase of the molecular complexity of scaffolds, which have potential applications for medicinal chemistry.

Chiral Imines

V. Köhler, K. R. Bailey, A. Znabet, J. Raftery, M. Helliwell, N. J. Turner* — 2182 – 2184

Enantioselective Biocatalytic Oxidative Desymmetrization of Substituted Pyrrolidines

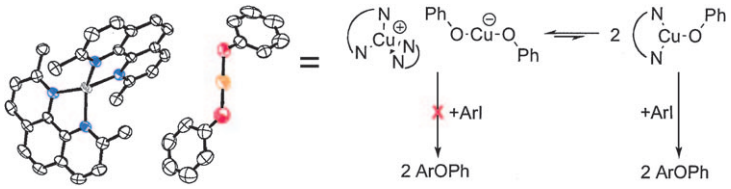


Made up out of air: The highly enantioselective oxidation of 3,4-substituted *meso*-pyrrolidines to Δ^1-pyrrolines is reported. The reaction is catalyzed by monoamine oxidase from *Aspergillus niger* (MAO-N D5) and utilizes molecular oxygen from air as the stoichiometric oxidant. The corresponding Δ^1-pyrrolines serve as useful building blocks for the synthesis of L-proline analogues and α-amino nitriles of high enantiomeric purity.

Cross-Coupling

J. W. Tye, Z. Weng, R. Giri, J. F. Hartwig* ——— 2185 – 2189

Copper(I) Phenoxide Complexes in the Etherification of Aryl Halides



No copping out! Copper(I) phenoxide complexes containing chelating ligands (see picture), proposed intermediates in copper-catalyzed etherification of aryl halides, have been synthesized and fully characterized. The kinetic and chemical competence of the isolated complexes are demonstrated for the synthesis of aryl phenyl ethers, and experiments provide evidence against mechanistic pathways involving the formation of either free or caged radicals.

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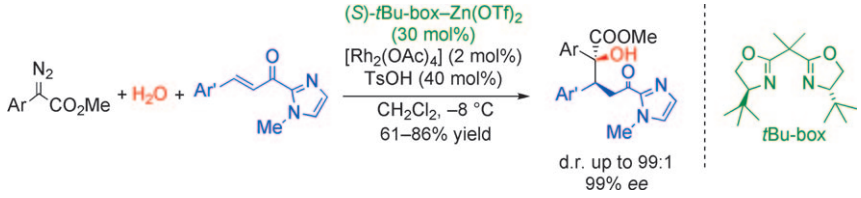
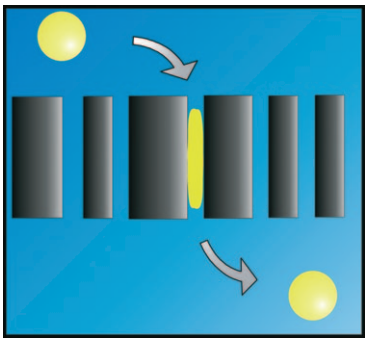


Together they make a difference: An aryl diazoacetate, H_2O, and an α,β-unsaturated 2-acyl imidazole give γ-hydroxyketones with a quaternary carbon stereocenter with excellent selectivity only if all three catalyst components shown in the scheme are present. The Michael addition step did not occur when a similar reagent mixture was treated with the $[Rh_2(OAc)_4]$ catalyst alone. OTf = triflate, Ts = *p*-tosyl.

Multicomponent Reactions

X.-Y. Guan, L.-P. Yang, W. Hu* ——— 2190 – 2192

Cooperative Catalysis in Multicomponent Reactions: Highly Enantioselective Synthesis of γ-Hydroxyketones with a Quaternary Carbon Stereocenter



Tight fit: Hydrogel microparticles can conform to and pass through pores up to 10 times smaller than the particle diameter (see picture). Network flexibility of this extent is unprecedented in the context of pore passage. Microgels even show such compressibility and prepenetration at size ratios and pressure differentials comparable to physiological renal filtration conditions, making them of potential interest in drug-delivery applications.

Ultrasoft Colloids

G. R. Hendrickson, L. A. Lyon* ——— 2193 – 2197

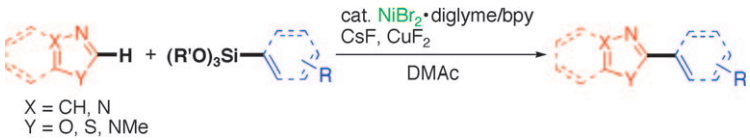
Microgel Translocation through Pores under Confinement

NHC me, now you don't: A tridentate bis(aryloxide)NHC-chelating ligand readily coordinates to Zr^{IV} and undergoes an unprecedented THF-promoted benzyl migration from the zirconium metal center to the $C_{carbene}$ atom of the NHC. NHC = N-heterocyclic carbene.

Carbenes

C. Romain, K. Miqueu, J.-M. Sotiropoulos, S. Bellemin-Laponnaz,* S. Dagorne* ——— 2198 – 2201

Non-Innocent Behavior of a Tridentate NHC Chelating Ligand Coordinated onto a Zirconium(IV) Center



Breaking the Si-lence: The direct cross-coupling of heteroarenes with aryl- or alkenylsilanes proceeds efficiently, in the presence of a nickel catalyst, to create a wide range of aryl– and alkenyl–heteroaryl bonds efficiently (see scheme; DMAc = *N*,*N*-dimethylacetamide, bpy = 2,2′-bipyridine). This catalysis represents a new avenue for using organosilanes in the catalytic C–H functionalization chemistry.

C–H Functionalization

H. Hachiya, K. Hirano, T. Satoh, M. Miura* ——— 2202 – 2205

Nickel-Catalyzed Direct C–H Arylation and Alkenylation of Heteroarenes with Organosilicon Reagents

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[5+2] Cycloaddition

F. Inagaki, K. Sugikubo, Y. Miyashita, C. Mukai* _____ **2206 – 2210**

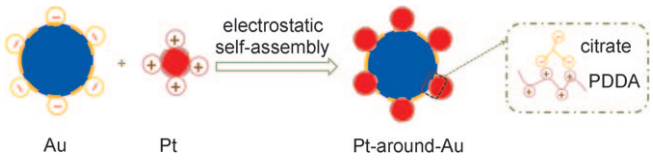
Rhodium(I)-Catalyzed Intramolecular [5+2] Cycloaddition Reactions of Alkynes and Allenylcyclopropanes: Construction of Bicyclo[5.4.0]undecatrienes and Bicyclo[5.5.0]dodecatrienes

Come on allene: The $[\{RhCl(CO)_2\}_2]$- or $[\{RhCl(CO)dppp\}_2]$-catalyzed intramolecular [5+2] cycloaddition of alkyne-allenylcyclopropanes under mild conditions affords the bicyclo[5.4.0]undecatriene or the larger bicyclo[5.5.0]dodecatriene frameworks (see scheme; dppp = 1,3-bis(diphenylphosphino)propane).

Nanoparticle Catalysts

S. Zhang, Y. Shao, G. Yin,* Y. Lin* _____ **2211 – 2214**

Electrostatic Self-Assembly of a Pt-around-Au Nanocomposite with High Activity towards Formic Acid Oxidation

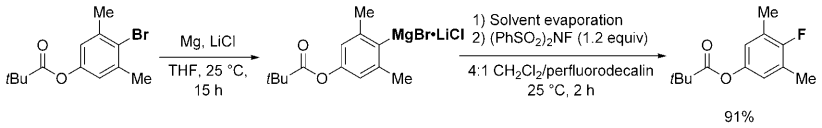


Opposites attract: A Pt-around-Au nanocomposite is synthesized by electrostatic self-assembly (see picture; PDDA = poly(diallyldimethylammonium chloride)). This catalyst shows significantly improved activity towards formic acid oxidation versus pure Pt catalysts. The possible reason is the efficient spillover of HCOO from Au to the surrounding Pt nanoparticles, where HCOO is further oxidized to CO_2.

Electrophilic Fluorination

S. Yamada, A. Gavryushin, P. Knochel* _____ **2215 – 2218**

Convenient Electrophilic Fluorination of Functionalized Aryl and Heteroaryl Magnesium Reagents



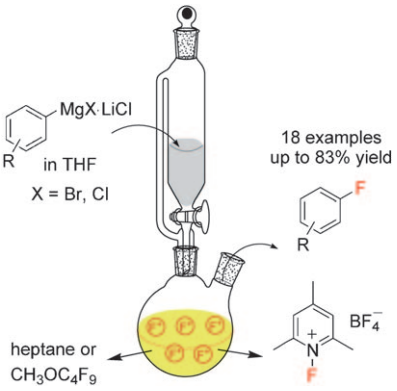
Give me an "F": Electrophilic fluorination of various aromatic and heteroaromatic Grignard reagents is smoothly performed with $(PhSO_2)_2NF$ as fluorinating agent in a 4:1 mixture of CH_2Cl_2/perfluorodecalin (see scheme). This solvent system allows minimization of most side reactions.

Selective Fluorination

P. Anbarasan, H. Neumann, M. Beller* _____ **2219 – 2222**

Efficient Synthesis of Aryl Fluorides

Creating C–F bonds: A novel electrophilic fluorination of aryl and heteroaryl Grignard reagents has been discovered and was used for the efficient synthesis of various aryl fluoride derivatives (see picture; THF = tetrahydrofuran).



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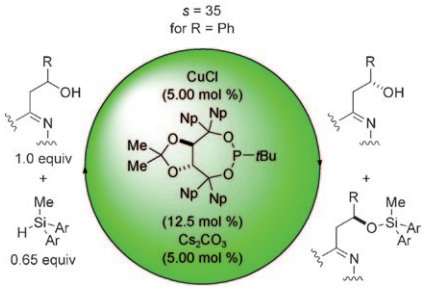
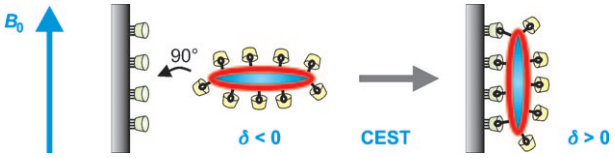
Silicon alley: Si–H and H–OR are enantioselectively coupled in the presence of a chiral Cu–H complex. In this way, the kinetic resolution of racemic mixtures of alcohols is accomplished through asymmetric protection with standard silanes (see scheme; R = aryl or alkyl, Ar = 3,5-xylyl, Np = 2-naphthyl; *s* = selectivity factor).



Kinetic Resolution

A. Weickgenannt, M. Mewald, T. W. T. Muesmann, M. Oestreich* ——— 2223 – 2226

Catalytic Asymmetric Si–O Coupling of Simple Achiral Silanes and Chiral Donor-Functionalized Alcohols

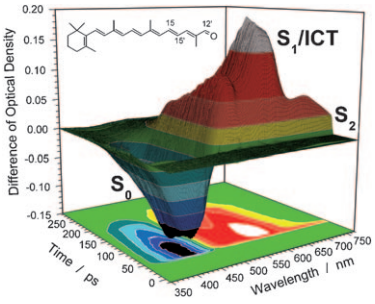


Searching for orientation: The alignment of aspherical paramagnetic liposomes in a magnetic field makes them attractive contrast agents for magnetic resonance (MR) imaging. The orientation of aspherical liposomes in solution, and the orientation change upon binding to a target surface was determined by using a simple MR technique (see picture).

Imaging Agents

D. Burdinski,* J. A. Pikkemaat, M. Emrullahoglu, F. Costantini, W. Verboom, S. Langereis, H. Grüll, J. Huskens ——— 2227 – 2229

Targeted LipoCEST Contrast Agents for Magnetic Resonance Imaging: Alignment of Aspherical Liposomes on a Capillary Surface



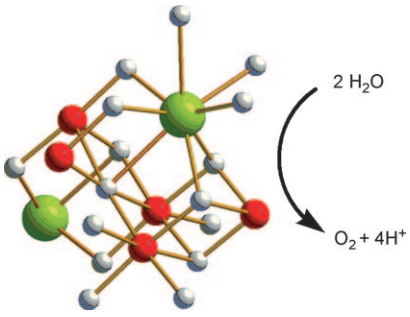
Superquick espionage: Attaching an aldehyde function to a simple carotenoid generates a powerful laser spectroscopy probe for elucidating solvation dynamics and polarity in ionic liquids with ultrafast pump–supercontinuum-probe broadband absorption spectroscopy.

Femtochemistry in Ionic Liquids

K. Oum,* P. W. Lohse, F. Ehlers, M. Scholz, M. Kopczynski, T. Lenzer* ——— 2230 – 2232

12′-Apo-β-caroten-12′-al: An Ultrafast "Spy" Molecule for Probing Local Interactions in Ionic Liquids

Biomimetic and efficient: Mixed calcium manganese(III) oxides (see structure; Ca green, Mn red, O white) with elemental compositions and structures mimicking the active site of photosystem II were found to be highly active catalysts for the oxidation of water to molecular oxygen. As for PS II, the presence of Ca^{2+} greatly enhances the catalyst performance in comparison to the related manganese-only system Mn_2O_3.



Water Oxidation

M. M. Najafpour, T. Ehrenberg, M. Wiechen, P. Kurz* ——— 2233 – 2237

Calcium Manganese(III) Oxides ($CaMn_2O_4 \cdot x H_2O$) as Biomimetic Oxygen-Evolving Catalysts

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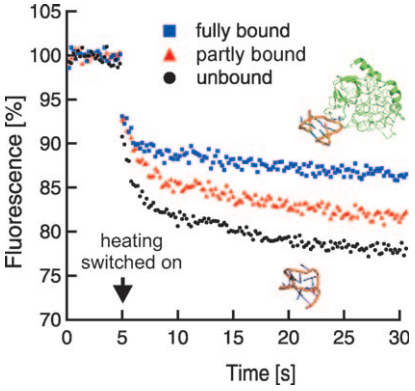
Contents

Bioanalytical Chemistry

P. Baaske,* C. J. Wienken, P. Reineck, S. Duhr, D. Braun ——— **2238 – 2241**

Optical Thermophoresis for Quantifying the Buffer Dependence of Aptamer Binding

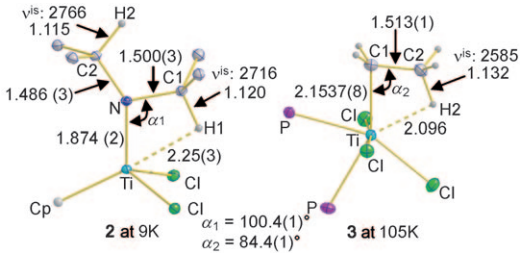


Some like it hot: A robust and fast method for characterizing aptamers relies on the distinct thermophoretic movements of molecules in microscopic temperature gradients (see diagram). The binding properties of proteins and even small molecules can be measured within seconds, and less than 1 μL of sample is required. Notably, the technique works well in complex liquids such as human serum.

Agostic Interactions

W. Scherer,* D. J. Wolstenholme, V. Herz, G. Eickerling,* A. Brück, P. Benndorf, P. W. Roesky* ——— **2242 – 2246**

On the Nature of Agostic Interactions in Transition-Metal Amido Complexes



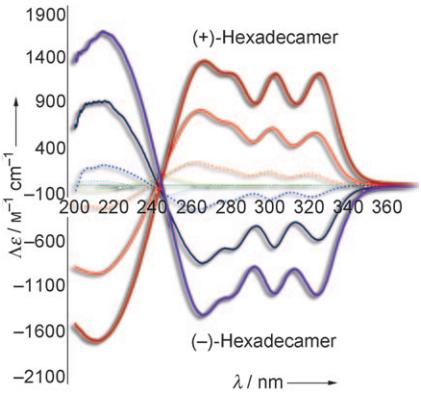
Not the same: β-H elimination in alkyl species containing a β-agostic interactions (see picture, right) is facilitated by negative hyperconjugative delocalization of the M–C bonding electron pair over the alkyl backbone. This process is significantly hindered by the π character of the M–N bond in corresponding amido complexes with agostic interactions (left). Hence, both types of agostic interactions should be discriminated from one another.

Chiral Oligomers

P. Rivera-Fuentes, J. L. Alonso-Gómez, A. G. Petrovic, F. Santoro, N. Harada, N. Berova, F. Diederich* ——— **2247 – 2250**

Amplification of Chirality in Monodisperse, Enantiopure Alleno-Acetylenic Oligomers

Enantiomerically pure alleno-acetylenic oligomers of defined lengths were synthesized by the palladium-mediated oxidative homocoupling of optically pure 1,3-diethynylallenes. The large amplification of their chiroptical properties strongly suggests the formation of helical secondary structures. This assignment is supported by time-dependent quantum chemical calculations.



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

In this Communication the first two authors are listed in reverse order. The correct order is: Yanling Dong, Hui Zheng, Xin Wang, Wenjian Weng, Gaorong Han, Ning Ma, Piyi Du.

Super High Threshold Percolative Ferroelectric/Ferrimagnetic Composite Ceramics with Outstanding Permittivity and Initial Permeability

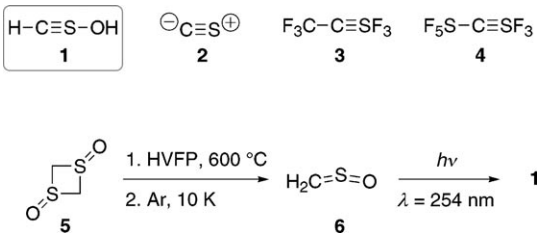
H. Zheng, Y. Dong, X. Wang, W. Weng, G. Han, N. Ma,* P. Du* — **8927–8930**

Angew. Chem. Int. Ed. **2009**, *48*

DOI 10.1002/anie.200904269

Corrigendum

Seppelt's compound **3** should be $F_3C{-}C{\equiv}SF_3$ and not $F_3S{-}C{\equiv}SF_3$. The corrected Scheme 1 is shown here. The authors apologize for this oversight.



A Formal Carbon–Sulfur Triple Bond: H–C≡S–O–H

P. R. Schreiner,* H. P. Reisenauer, J. Romanski, G. Mloston* — **8133–8136**

Angew. Chem. Int. Ed. **2009**, *48*

DOI 10.1002/anie.200903969

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