



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:

S. T. Scroggins, Y. Chi, J. M. J. Fréchet*

Polarity-Directed One-Pot Asymmetric Cascade Reactions Mediated by Two Catalysts in an Aqueous Buffer

D. Loffreda,* F. Delbecq, F. Vigné, P. Sautet

Fast Prediction of Selectivity in Heterogeneous Catalysis from Extended Brønsted–Evans–Polanyi Relations: A Theoretical Insight

M. H. Kox, K. F. Domke, J. P. Day, G. Rago, E. Stavitski, M. Bonn, B. M. Weckhuysen*

Label-Free Chemical Imaging of Catalytic Solids by Coherent Anti-Stokes Raman Scattering and Synchrotron-Based Infrared Microscopy

M. Griesser, D. Neshchadin, K. Dietliker, N. Moszner, R. Liska, G. Gescheidt*

Decisive Reaction Steps at Initial Stages of Photoinitiated Radical Polymerizations

J.-G. Liu, T. Ohta, S. Yamaguchi, T. Ogura, S. Sakamoto, Y. Maeda, Y. Naruta*

Spectroscopic Characterization of a Hydroperoxo–Heme Intermediate of a Synthetic Model: Conversion of a Side-on Peroxy to an End-on Hydroperoxy Complex

A. B. Chaplin, A. S. Weller*

B–H Activation at a Rhodium(II) Center: A Missing Link in the Transition-Metal-Catalyzed Dehydrocoupling of Amine–Boranes

M. Bandini*, A. Eichholzer

Enantioselective Gold-Catalyzed Allylic Alkylation of Indoles with Alcohols: Efficient Route to Functionalized Tetrahydrocarbazoles

G. de Ruiter, E. Tartakovsky, N. Oded, M. E. van der Boom*

Sequential Logic Operations with Surface-Confined Polypyridyl Complexes Having Molecular Random Access Memory Features



“My most exciting discovery to date has been catalytic enantioselective ring-opening reactions.

When I was eighteen I wanted to be a professor of veterinary medicine ...”

This and more about Mark Lautens can be found on page 8602.

Author Profile

Mark Lautens _____ 8602

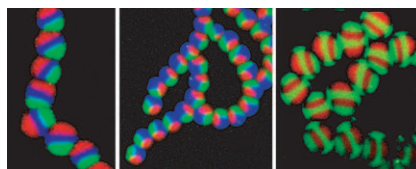
Energy Demand and Climate Change

Franklin Hadley Cocks

Books

reviewed by N. Armaroli _____ 8603

Fibers with a future: An exciting co-electrospinning technique has been developed for the fabrication of multi-component, microstructured, biodegradable materials. These multicompartmental fibers and particles have special relevance to biological imaging, drug delivery, and tissue engineering.



Highlights

Microstructured Materials

M. C. George, P. V. Braun* – 8606–8609

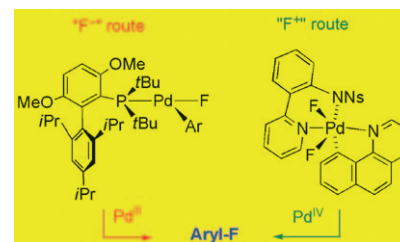
Multicompartmental Materials by Electrohydrodynamic Cojetting

Fluoroarenes

J. M. Brown,*
V. Gouverneur* — 8610–8614

Transition-Metal-Mediated Reactions for C_{sp^2} -F Bond Construction: The State of Play

It's a rate thing: Crucial conceptual advances have allowed for the development of a Pd-mediated pathway to C_{aryl} -F by using either electrophilic (right) or nucleophilic (left) fluorinating reagents. Of significance is the recognition that Pd intermediates reduce the barrier to C-F reductive elimination and the use of an ancillary bulky ligand to force reductive elimination from a 14-electron Pd^{II} complex.

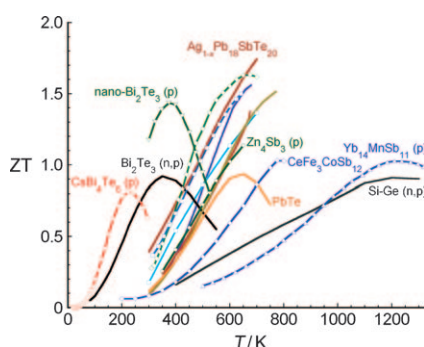


Reviews

Thermoelectrics

J. R. Sootsman, D. Y. Chung,
M. G. Kanatzidis* — 8616–8639

New and Old Concepts in Thermoelectric Materials

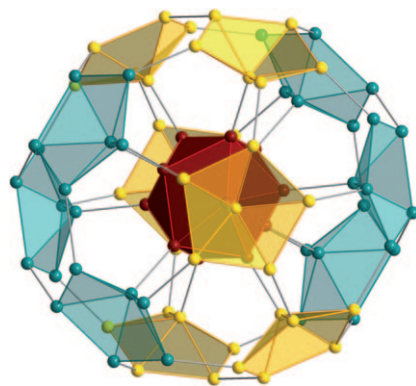


From heat to electricity: At the forefront of research into thermoelectric materials is increasing the thermoelectric figure of merit (ZT), an indicator of the efficiency of electricity generation from heat. For a long time $ZT = 1$ appeared to be an insurmountable barrier however, in the last ten years, this value has been surpassed several times (see diagram).

Boron and Borides

B. Albert,* H. Hillebrecht* — 8640–8668

Boron: Elementary Challenge for Experimenters and Theoreticians



Structural complexity, electron deficiency, unusual bonding situations, and a rich variety of compounds characterize the element boron, for which many unanswered questions on its solid-state chemistry still remain 200 years after its discovery. Theoretical studies on the stability and existence of known and new modifications of the element together with high-pressure and high-temperature experiments have brought new insight (picture: B_{84} unit of β -B).

For the USA and Canada:

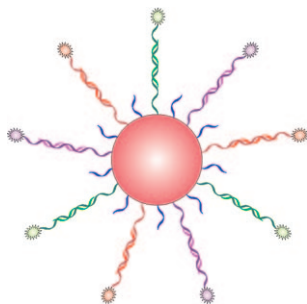
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Meacham Ave., Elmont, NY 11003. Periodicals postage paid at Jamaica, NY 11431. US POSTMASTER: send address changes to *Angewandte Chemie*, Wiley-VCH, 111 River Street, Hoboken, NJ 07030. Annual subscription price for institutions: US\$ 9442/8583 (valid for print and

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

Communications

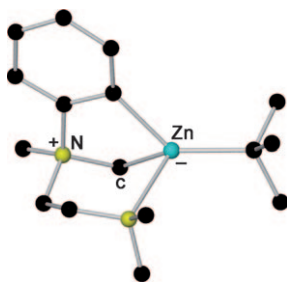
Color vision: Multicolor molecular beacons are constructed from relatively large gold nanoparticles self-assembled with stem-loop probes and helper oligonucleotides (see picture). The nanobeacons can respond differentially to multiple DNA targets, emitting multiple colors.



DNA Analysis

S. Song, Z. Liang, J. Zhang, L. Wang,
G. Li,* C. Fan* ————— **8670–8674**

Gold-Nanoparticle-Based Multicolor
Nanobeacons for Sequence-Specific DNA
Analysis

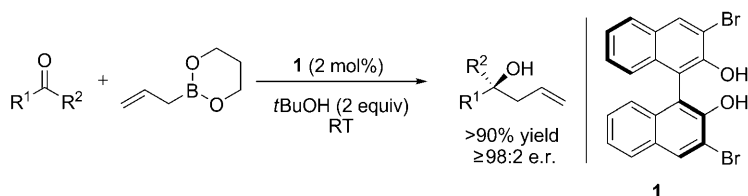


Double role for sodium: Within a synergic sodium TMP-zincate reagent, sodium mediates the direct *ortho*-zincation of chlorobenzene and delivers the TMEDA ligand to nucleophilically attack the benzene ring, thereby generating novel open and cyclic zinc zwitterions (example shown). TMP = 2,2,6,6-tetramethylpiperidine; TMEDA = *N,N,N',N'*-tetramethylethylenediamine.

Zwitterionic Zincation

D. R. Armstrong, L. Balloch, W. Clegg,
S. H. Dale, P. García-Álvarez, E. Hevia,
L. M. Hogg, A. R. Kennedy, R. E. Mulvey,*
C. T. O'Hara ————— **8675–8678**

Synergic Synthesis of Benzannulated
Zincabicyclic Complexes, α -Zincated N
Ylides, through Sodium-TMEDA-
Mediated Zincation of a Haloarene



Allylations

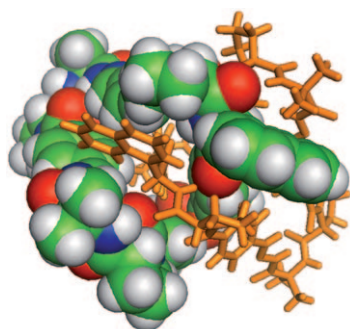
D. S. Barnett, P. N. Moquist,
S. E. Schaus* ————— **8679–8682**

The Mechanism and an Improved
Asymmetric Allylboration of Ketones
Catalyzed by Chiral Biphenols



Giving it a boost: A mechanistic study of the enantioselective asymmetric titled reaction with allyldiisopropoxyborane catalyzed by chiral biphenols revealed a key ligand exchange process which liberates isopropyl alcohol. The addition of *i*PrOH

to the reaction increases the overall rate and enantioselectivity. As a result an improved reaction, employing allyldioxaborinane with **1** and *t*BuOH, resulted in high product yields and enantioselectivities.



A love of self: Narcissistic macrocyclic rings self-assemble into highly ordered, chiral [2]-catenanes displaying high component diastereoselectivity. The picture shows one such structure. One ring is shown in a space-filling representation (C green, O red, N blue, H gray), while the other is shown as orange sticks.

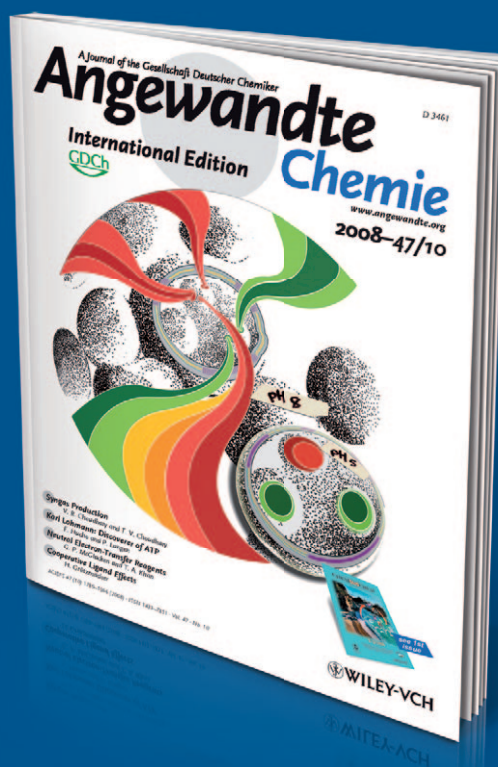
Supramolecular Chemistry

M.-K. Chung, P. S. White, S. J. Lee,
M. R. Gagné* ————— **8683–8686**

Synthesis of Interlocked 56-Membered
Rings by Dynamic Self-Templating



Incredibly inexpensive.



Do chemistry journals really cost so much? Perhaps some do, but certainly not *Angewandte Chemie*! In 2008, an entire institution could subscribe through Wiley InterScience* for 5000 € and get access to 52 issues with over 1600 articles and all associated online search options, and for just 5 % more, the printed issues could be included as well. For full members of the German Chemical Society (GDCh), a personal subscription cost not much more than 300 €, and student GDCh members paid less than 150 €, which is just under 3 € per issue - a price that even compares with high-circulation newsstand publications!

*www.interscience.wiley.com

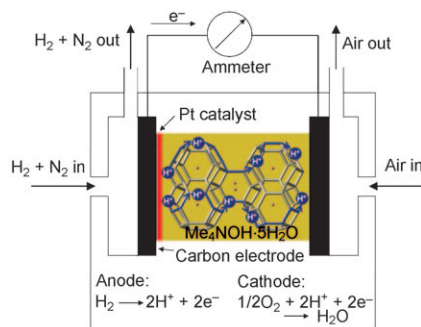


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 **WILEY-VCH**

Icy detector: A simple new concept for the fabrication of amperometric H_2 sensors benefits from an ionic clathrate hydrate (see picture). The physicochemical characteristics of icelike $Me_4NOH \cdot 5H_2O$ lead to good performance of the sensor assembly and fast response and recovery times, even at lower H_2 concentrations.



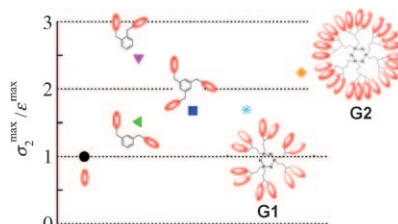
Hydrogen Sensors

J.-H. Cha, W. Lee, H. Lee* — 8687–8690

Hydrogen Gas Sensor Based on Proton-Conducting Clathrate Hydrate



Space invaders: A series of multichromophores (from dimers to dendritic 24-mers) is synthesized, spectroscopically characterized, and theoretically modeled. Enhancement of the two-photon absorption response depends on the number and distribution of chromophoric sub-units (see picture, red) and increases with their proximity. The exciton model indicates purely electrostatic interactions.



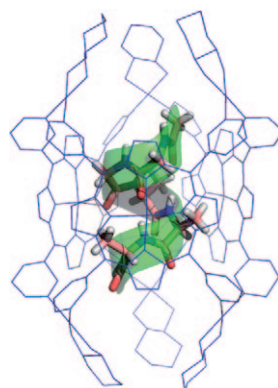
Multichromophores

F. Terenziani, V. Parthasarathy, A. Pla-Quintana, T. Maishal, A.-M. Caminade, J.-P. Majoral, M. Blanchard-Desce* — 8691–8694

Cooperative Two-Photon Absorption Enhancement by Through-Space Interactions in Multichromophoric Compounds



Folding in the confine: Short fragments of tri- to hexapeptides have been encapsulated by a synthetic host in water and folded into their latent helical structures (see crystal structure). Crystallographic analysis of the complexes clearly reveals a mixed conformation of 3_{10} and α helices, thereby illustrating the inherent helical propensity of very short peptide fragments.



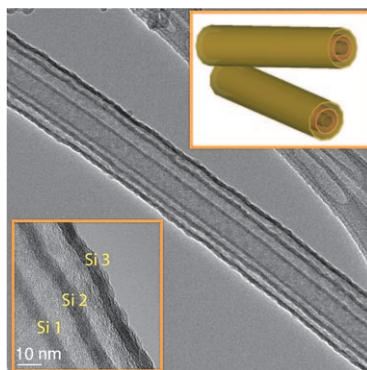
Peptide Folding

Y. Hatakeyama, T. Sawada, M. Kawano, M. Fujita* — 8695–8698

Conformational Preferences of Short Peptide Fragments



Tubular bells and whistles: Building additional functionality into nanotubes by controlled synthesis may enable novel architectures and enhanced (opto)electronic devices. The composition of each wall in novel, hollow, crystalline silicon nanotubes (tube-in-tube and wire-in-tube nanostructures) can be independently defined, and the interwall distance and wall thickness can be controlled (picture: triple-walled nanotube and corresponding TEM images).



Nanotechnology

M. B. Ishai, F. Patolsky* — 8699–8702

Tube-in-Tube and Wire-in-Tube Nano Building Blocks: Towards the Realization of Multifunctional Nanoelectronic Devices

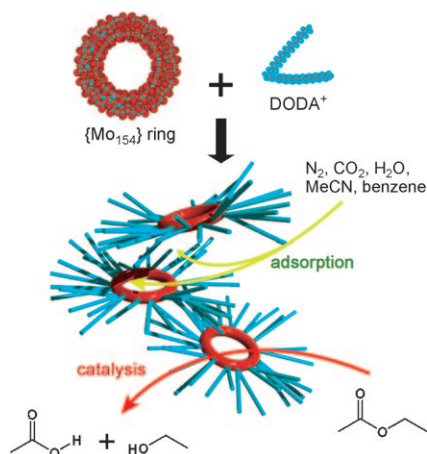


Polyoxometalates

S.-i. Noro,* R. Tsunashima, Y. Kamiya,
K. Uemura, H. Kita, L. Cronin,*
T. Akutagawa,
T. Nakamura* ————— **8703–8706**



Adsorption and Catalytic Properties
of the Inner Nanospace of a Gigantic
Ring-Shaped Polyoxometalate Cluster



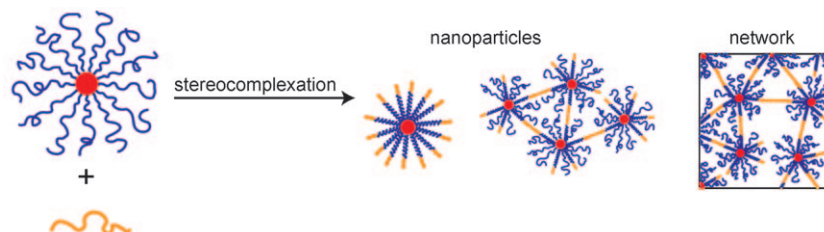
Functional inner space: A gigantic ring-shaped $\{Mo_{154}\}$ polyoxometalate cluster anion can be stabilized by encapsulation in dimethyldioctadecylammonium (DODA) cations. Its inner nanospace enables adsorption of gases and vapors, and it acts as a water-tolerant solid acid catalyst (see scheme).

Polymer-Helix Assemblies

T. K. Goh, J. F. Tan, S. N. Guntari, K. Satoh,
A. Blencowe, M. Kamigaito,*
G. G. Qiao* ————— **8707–8711**



Nano-to-Macroscale Poly(methyl
methacrylate) Stereocomplex Assemblies



We can do so much together: The stereocomplexation of complementary strands of stereoregular poly(methyl methacrylate) (PMMA) to form triple-stranded helices led to a fascinating array of PMMA stereocomplex morphologies. The differ-

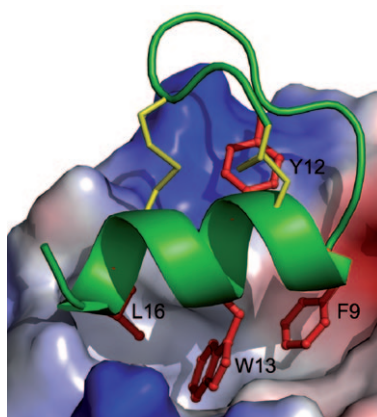
ent polymer-helix assemblies were constructed from a single set of complementary polymers (syndiotactic (blue in the picture) and isotactic PMMA (orange)) by simple tuning of the mixing ratio and concentration.

Protein Structures

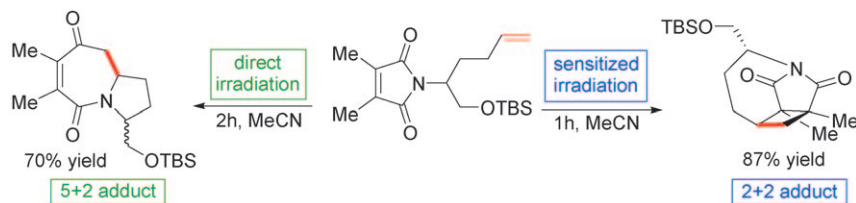
C. Li, M. Pazgier, M. Liu, W.-Y. Lu,
W. Lu* ————— **8712–8715**



Apamin as a Template for Structure-Based
Rational Design of Potent Peptide
Activators of p53



Taking the sting out of tumors: The oncoproteins MDM2 and MDMX negatively regulate the activity and stability of the tumor suppressor protein p53 and are important molecular targets for anti-cancer therapy. Grafting four residues critical for MDM2/MDMX binding to the C-terminal α -helix of apamin (see structure) converts the bee-venom neurotoxin into a novel class of potent p53 activators with potential antitumor activity.



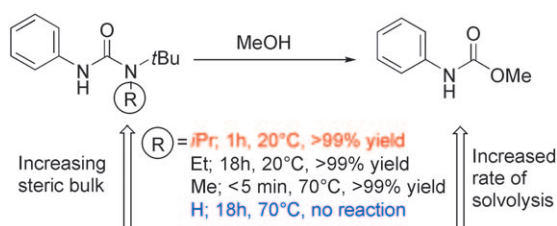
Split personality: Reaction conditions that enable powerful control over the mode of photocycloaddition in maleimides have been developed. Direct irradiation favors

the [5+2] mode whereas sensitized irradiation allows a complete switch to the [2+2] mode (see scheme; TBS = *tert*-butyldimethylsilyl).

Reaction Control

C. Roscini, K. L. Cubbage, M. Berry, A. J. Orr-Ewing,*
K. I. Booker-Milburn* — 8716–8720

Reaction Control in Synthetic Organic Photochemistry: Switching between [5+2] and [2+2] Modes of Cycloaddition



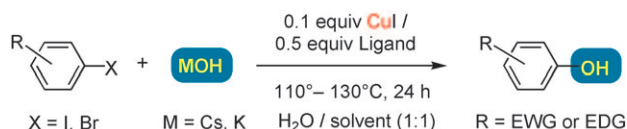
Bigger is better: Sterically hindered dialkyl ureas undergo nucleophilic substitution at dramatically faster rates than their less hindered counterparts (see scheme). Steric decompression upon the formation

of an intermediate isocyanate can explain this counterintuitive behavior. These hindered ureas can be considered as masked reagents that liberate reactive isocyanates in situ.

Solvolysis

M. Hutchby, C. E. Houlden, J. G. Ford, S. N. G. Tyler, M. R. Gagné, G. C. Lloyd-Jones,*
K. I. Booker-Milburn* — 8721–8724

Hindered Ureas as Masked Isocyanates: Facile Carbamoylation of Nucleophiles under Neutral Conditions



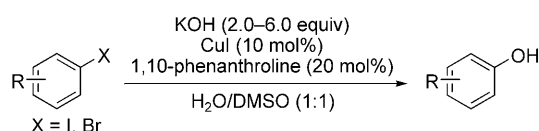
Cheap and cheerful: The direct copper-catalyzed hydroxylation of activated and unactivated aryl iodides or bromides has been achieved using metal hydroxide salts (MOH). Selective hydroxylation in a H₂O/

co-solvent system avoids formation of the related biaryl ether by-product and the low cost of the copper catalytic system makes this method very competitive.

C–O Coupling

A. Tlili, N. Xia, F. Monnier, M. Taillefer* — 8725–8728

A Very Simple Copper-Catalyzed Synthesis of Phenols Employing Hydroxide Salts



A smooth operator: The copper-catalyzed synthesis of phenols from aryl halides was carried out under relatively mild reaction conditions. Alkyl aryl ethers and benzo-

furans could also be prepared smoothly by one-pot domino protocols based on hydroxylation of aryl iodides (see scheme).

Hydroxylation

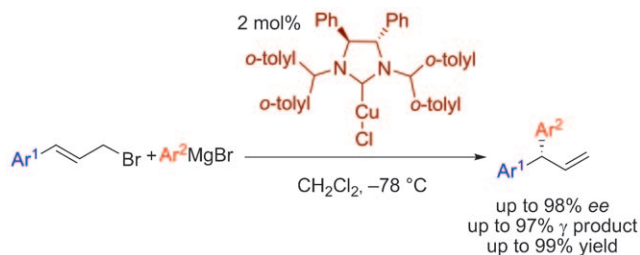
D. Zhao, N. Wu, S. Zhang, P. Xi, X. Su, J. Lan, J. You* — 8729–8732

Synthesis of Phenol, Aromatic Ether, and Benzofuran Derivatives by Copper-Catalyzed Hydroxylation of Aryl Halides



Synthetic Methods

K. B. Selim, Y. Matsumoto, K. Yamada,
K. Tomioka* ————— **8733–8735**



Gamma rules: The title reaction was achieved in a highly regioselective manner using aryl Grignard reagents with monodentate chiral N-heterocyclic carbene–

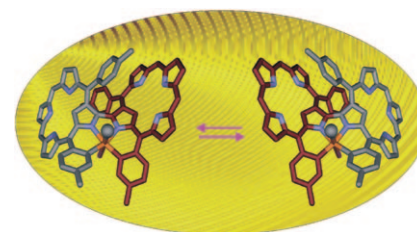
copper(I) catalyst to give diaryl-vinylmethanes with excellent enantiomeric excess in excellent yield (see scheme).

Porphyrinoids

P. J. Chmielewski,* B. Durler, M. Siczek,
L. Szterenberga ————— **8736–8739**

Helical Bis(N-Confused Porphyrins) with Subunits Fused by Double Orthometalation with Platinum: Adaptability of an Apparently Rigid System

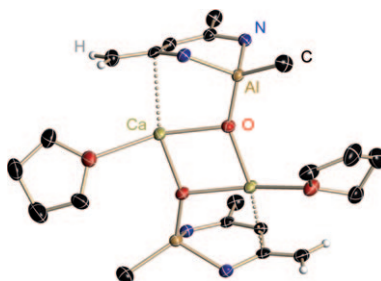
Flexible yet rigid: The flexibility of the porphyrin rings makes racemization or chiral induction still possible with helical metalated bis(N-confused porphyrin) systems, even though the bipyrrole fragment itself is rigid. The porphyrins are peripherally doubly orthometalated with platinum(II), and a series of its oxidative addition products were also obtained (see structures of the iodomethyl adduct: gray and brown C; blue N; orange Pt; purple I).



Alkaline-Earth-Metal Oxides

S. P. Sarish, S. Nembenna, H. W. Roesky,*
H. Ott, A. Pal, D. Stalke, S. Dutta,
S. K. Pati ————— **8740–8742**

Soluble Molecular Dimers of CaO and SrO Stabilized by a Lewis Acid



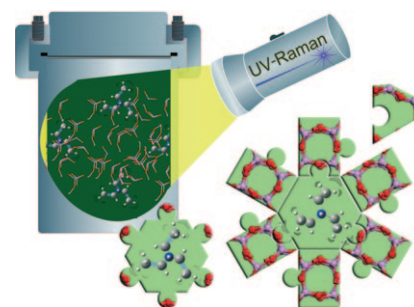
Soluble oxides: The reaction between a β -diketiminato aluminum methyl hydroxide and $[\text{M}\{\text{N}(\text{SiMe}_3)_2\}_2(\text{thf})_2]$ ($\text{M} = \text{Ca}, \text{Sr}$) results in alkaline-earth-metal oxides such as $[\{\text{LAl}(\text{Me})(\mu\text{-O})\text{Ca}(\text{thf})\}_2]$ (see core structure; L is deprotonated β -diketiminato $\text{CH}\{\text{C}(\text{CH}_2)\}(\text{CMe})(2,6\text{-iPr}_2\text{C}_6\text{H}_3\text{N})_2$). These soluble complexes are dimeric in the solid state and contain unprecedented M_2O_2 cores.

Aluminophosphates

F. Fan, Z. Feng, K. Sun, M. Guo, Q. Guo,
Y. Song, W. Li, C. Li* ————— **8743–8747**

In Situ UV Raman Spectroscopic Study on the Synthesis Mechanism of AlPO-5

Piecing together the puzzle: The synthesis mechanism of AlPO-5 was studied using in situ UV Raman spectroscopy in combination with the Fenton reaction. The results reveal molecular-level insights into the role of the template in channel formation as well as the identities of the building blocks in the framework formation.



An exciting exchange: Molecules and surface plasmons may interact through the exchange of photons (see picture) to form new hybrid states in which the photophysical properties of the molecule are altered. This process could form the basis of a new pathway for the modification of the photochemistry and even the chemistry of molecules.

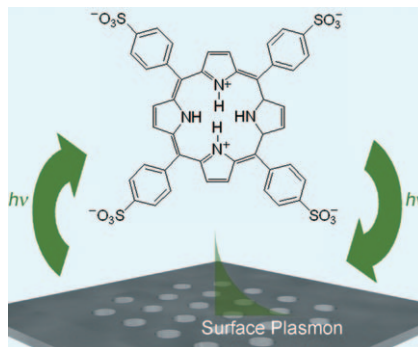
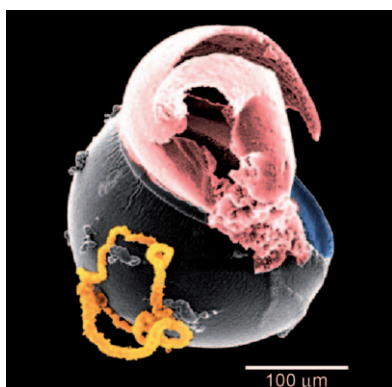


Photo physics

A. Salomon, C. Genet,
T. W. Ebbesen* ————— **8748–8751**

Molecule–Light Complex: Dynamics of
Hybrid Molecule–Surface Plasmon States

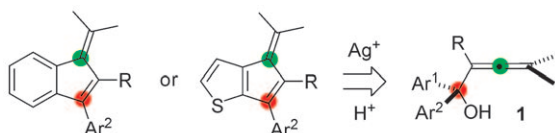


Self propelled: Chemical tubospheres are produced by loading salt solutions into agarose microbeads and exposing them to a solution of sodium silicate. This process creates hollow tubes that are attached to inorganic shells (see picture). The tubes have inner radii down to 3 μm, reach lengths of 0.5 mm, and grow at speeds of up to 50 μm s⁻¹. Tubes that are pinned to bubbles can induce directional bead motion.

Microstructure Growth

R. Makki, M. Al-Humiri, S. Dutta,
O. Steinbock* ————— **8752–8756**

Hollow Microtubes and Shells from
Reactant-Loaded Polymer Beads



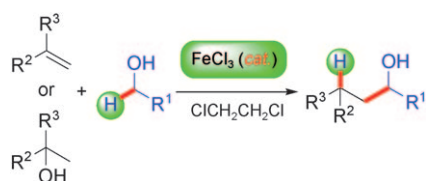
Easy as pie: Expedient access to aryl-substituted benzofulvenes is described. α -Hydroxyallenes **1** that bear two aryl groups at C1 (red circle) are directly transformed into these products at room

temperature by using silver triflate as a catalyst or Brønsted acids. The transformation involves dealkoxylation and subsequent 4 π electrocyclicization (Nazarov reaction).

Nazarov Reactions

P. Cordier, C. Aubert, M. Malacria,*
E. Lacôte,* V. Gandon* — **8757–8760**

Silver and Brønsted Acid Catalyzed
Nazarov-Type Cyclizations To Generate
Benzofulvenes



Synthetic convenience: A wide range of substrates proved to be well tolerated by the novel title transformation. This protocol provides an economical and convenient strategy for the efficient access to structurally diverse secondary alcohols (see scheme).

Iron Catalysis

S.-Y. Zhang, Y.-Q. Tu,* C.-A. Fan,
F.-M. Zhang, L. Shi ————— **8761–8765**

Iron-Catalyzed C(sp³)–C(sp³) Bond
Formation through C(sp³)–H
Functionalization: A Cross-Coupling
Reaction of Alcohols with Alkenes

Natural Products

M. E. Jung,* R. Salehi-Rad — 8766–8769



Total Synthesis of Auripyronone A Using a Tandem Non-Aldol Aldol/Paterson Aldol Process as a Key Step



To aldol or non-aldol: The titled reaction sequence generates the polypropionate **3** from the epoxy alcohol **1** and the ketone **2** as a single diastereomer. Compound **2** was used for an efficient synthesis of auripyronone A using a highly regioselective

hemiketalization of a keto diol and a late-stage spiroketalization onto a stable hemiketal as the final key steps. TBDPS = *tert*-butyldiphenylsilyl, Bz = benzoyl, TES = triethylsilyl.

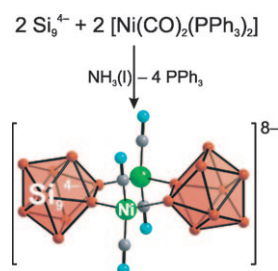


Polyanions

S. Joseph, M. Hamberger, F. Mutzbauer, O. Härtl, M. Meier, N. Korber* — 8770–8772



Chemistry with Bare Silicon Clusters in Solution: A Transition-Metal Complex of a Polysilicide Anion



Pure silicon building blocks in solution are an exciting addition to the repertoire of the materials science of Group 14 elements. The preparation of the complex $[\{\text{Ni}(\text{CO})_2\}_2(\mu\text{-Si}_9)_2]^{8-}$ shows that rational synthetic transformations with the Si_9^{4-} Zintl ion are feasible.

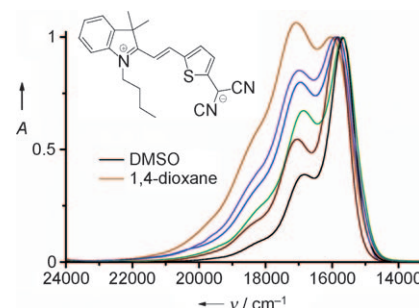
Merocyanines

H. Mustroph,* J. Mistol, B. Senns, D. Keil, M. Findeisen, L. Hennig — 8773–8775



Relationship between the Molecular Structure of Merocyanine Dyes and the Vibrational Fine Structure of Their Electronic Absorption Spectra

Relative intensities of vibronic subbands in the electronic absorption spectra of a new merocyanine dye are influenced by the polarity of the solvent. Increasing solvent polarity leads to an increased π -charge density alternation in the polymethine chain and, consequently, to reduced bond alternation.



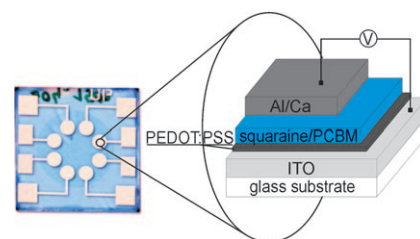
Organic Photovoltaics

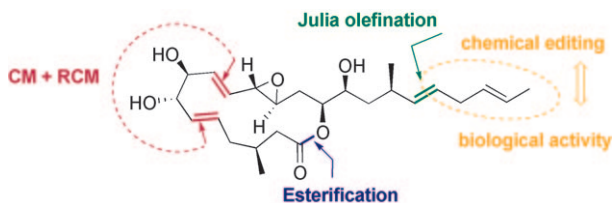
U. Mayerhöffer, K. Deing, K. Größ, H. Braunschweig, K. Meerholz,* F. Würthner* — 8776–8779



Outstanding Short-Circuit Currents in BHJ Solar Cells Based on NIR-Absorbing Acceptor-Substituted Squaraines

Current affairs: Acceptor-substituted squaraines exhibit unprecedented short-circuit current density of up to 12.6 mA cm^{-2} in small-molecule-based solution-processed bulk heterojunction (BHJ) solar cells (see picture). These values are closing the gap to polymer solar cells and together with the NIR absorption of the new cells may lead to wide applications.





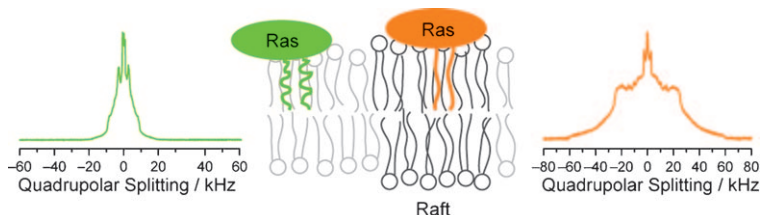
Metathesis on stage: A combined cross-metathesis (CM)/ring-closing metathesis (RCM) approach has led to the stereo-controlled synthesis of iriomoteolide 3a, a smaller but equally cytotoxic congener of amphidinolides. Chemical editing of the

molecule has provided non-natural analogues which have comparable anticancer activity to that of the natural product, thereby allowing the iriomoteolides to be used as probe molecules in chemical biology.

Total Synthesis

R. Cribiú, C. Jäger,
C. Nevado* — 8780–8783

Syntheses and Biological Evaluation of
Iriomoteolide 3a and Analogues



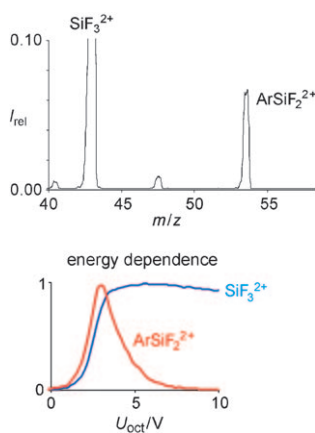
Matching the membrane: The lipid modification of human N-Ras proteins can adapt remarkably to the host membrane by varying its length between 8.7 Å and 15.5 Å, as can be seen in the ^2H NMR spectra of deuterated derivatives (see

picture). In raft-forming biological membranes, Ras is found in the liquid disordered phase as well as in the phase boundary between liquid-crystalline and raft domains.

Membrane Anchoring

A. Vogel, G. Reuther, K. Weise, G. Triola,
J. Nikolaus, K.-T. Tan, C. Nowak,
A. Herrmann, H. Waldmann, R. Winter,
D. Huster* — 8784–8787

The Lipid Modifications of Ras that Sense
Membrane Environments and Induce
Local Enrichment



Noblesse oblige: The SiF_3^{2+} dication, a superelectrophilic reagent, enables the formation of noble gas–silicon compounds in thermal ion–molecule reactions. Bimolecular collisions of mass-selected SiF_3^{2+} dications with argon lead to the dication ArSiF_2^{2+} with a covalent Ar–Si bond as the major product at thermal energies. An unambiguous signal for the NeSiF_2^{2+} dication is also observed, but in much lower yields.

Noble Gas Compounds

J. Roithová,* D. Schröder* — 8788–8790

Silicon Compounds of Neon and Argon



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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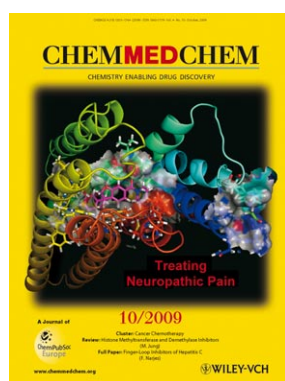
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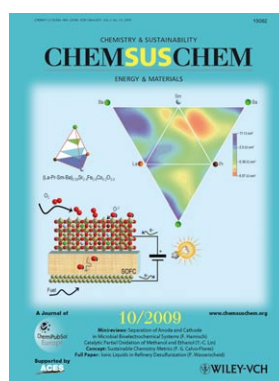
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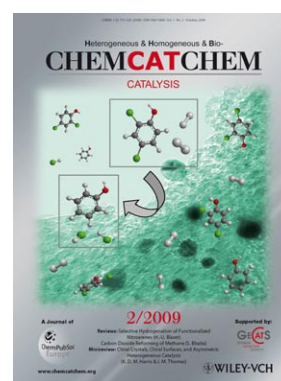
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