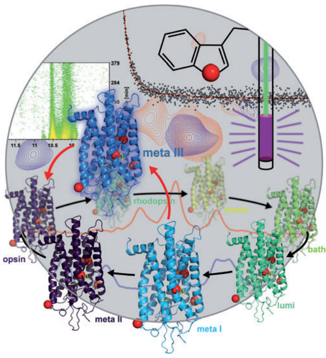
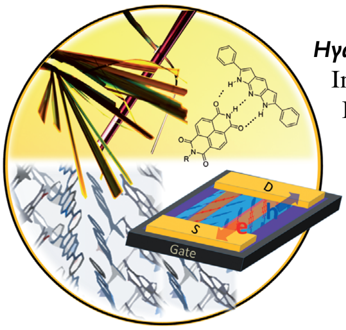
Two bowl-shaped molecules ...

... of complementary electronic character, a donor truxene derivative and an acceptor hemifullerene, have been found to form heteromolecular dimers in solution with binding constants on the order of $10^3\,\text{M}^{-1}$, as described by E. Ortí, D. M. Guldi, L. T. Scott, N. Martín, et al. in their Communication on page 2170 ff. This is the first time that a fullerene fragment has been found to mimic the behavior of C_{60} as an electron acceptor.

Rhodopsin Dynamics

H. Schwalbe et al. describe in their Communication on page 2078 ff. the use of time-resolved liquid-state NMR spectroscopy to study the kinetics of the light-activation process of the mammalian visual dim-light photoreceptor rhodopsin.





Hydrogen Bonds

In their Communication on page 2138 ff., H. T. Black and D. F. Perepichka show that the solid-state arrangement of donor/acceptor components in crystalline p/n heterojunctions is controlled by complementary hydrogen bonding.

Synthetic Methods

In their Communication on page 2212 ff., M. Pohmakotr et al. describe a stereoselective cascade process that involves radical cyclization and *ipso*-1,4-aryl migration for the synthesis of 3,3-difluoro-2-propanoylbicyclo[3.3.0]octanes.

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Service

Author Profile

*"My biggest motivation is curiosity.
The most exciting thing about my research is not knowing what comes next. …"*
This and more about Alexander Eychmüller can be found on page 2028.

News


H. Zhang


Y. Xie

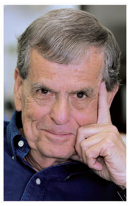

K. Ding


X. Feng


B. Han


W.-H. Fang


C. Amatore


A. Ciechanover


R. R. Schrock


C. Corminboeuf

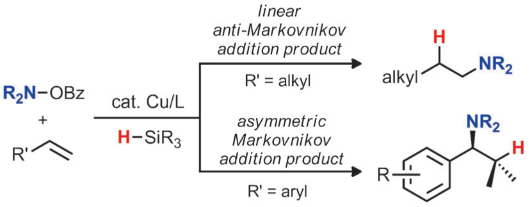

J. Waser


E. Reisner

Books

Highlights

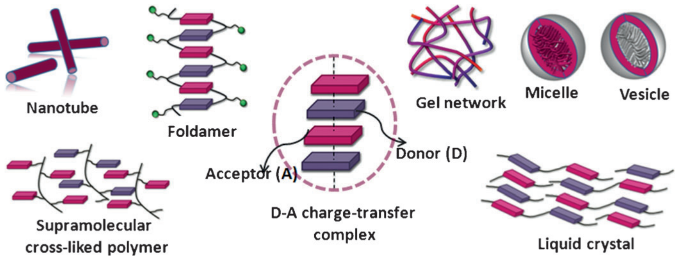
Hydroamination



Adding value: The catalytic hydroamination of alkenes with amines holds promise to rapidly deliver value-added chiral amines from simple and accessible starting materials. The use of mild, Cu-catalyzed electrophilic amination strategies for the regioselective preparation of both linear and chiral branched amines is highlighted.

Minireviews

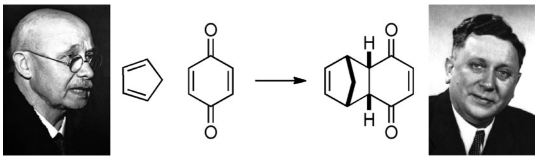
Noncovalent Interactions



Riot of color: Alternate stacking of aromatic donor and acceptor building blocks by complementary and directional charge-transfer interactions produce versatile supramolecularly assembled materials including micelles, vesicles, nanotubes, fibrillar gels, folded polymers, cross-linked networks, and liquid-crystalline phases.

Reviews

Diels–Alder Reactions of Quinones



Master of six-membered rings: The reaction of a quinone (dienophile) with a diene was described by Diels and Alder in 1928. This Review gives an overview of the application of this reaction as a pivotal step in the total synthesis of natural products and other complex molecules.

For the USA and Canada:
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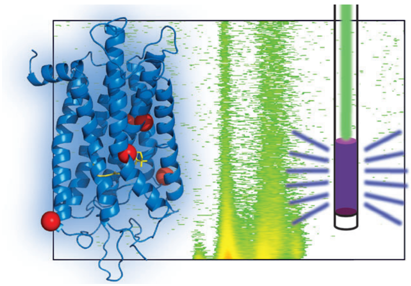
Communications

Rhodopsin Dynamics

J. Stehle, R. Silvers, K. Werner, D. Chatterjee, S. Gande, F. Scholz, A. Dutta, J. Wachtveitl, J. Klein-Seetharaman, H. Schwalbe* ______ **2078 – 2084**

Characterization of the Simultaneous Decay Kinetics of Metarhodopsin States II and III in Rhodopsin by Solution-State NMR Spectroscopy

Let there be light! Time-resolved liquid-state NMR spectroscopy has been used to study the kinetics of the light-activation process of the mammalian visual dim-light photoreceptor rhodopsin. The data reveal two parallel processes occurring subsequent to rhodopsin activation. The metarhodopsin states II and III emerge in parallel in a relative ratio of approximately 3:1.



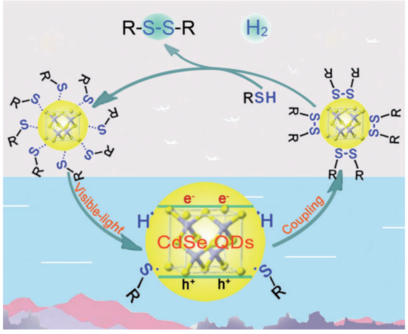
Frontispiece

Photocatalysis

X.-B. Li, Z.-J. Li, Y.-J. Gao, Q.-Y. Meng, S. Yu, R. G. Weiss, C.-H. Tung, L.-Z. Wu* ______ **2085 – 2089**

Mechanistic Insights into the Interface-Directed Transformation of Thiols into Disulfides and Molecular Hydrogen by Visible-Light Irradiation of Quantum Dots

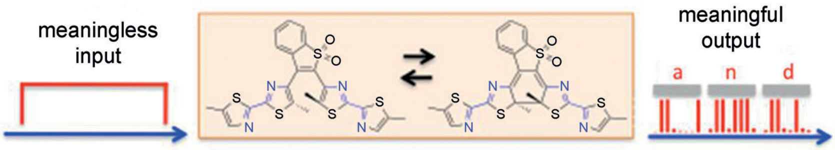
On the dot: A clean and efficient catalytic method for the preparation of disulfides from a variety of thiols in the absence of sacrificial reagents or external oxidants is described. Irradiation of CdSe quantum dots (QDs) with visible light results in good to excellent yields of the disulfides and equivalent amounts of H_2. Mechanistic studies provide evidence for the formation of QD-bound RS$^•$ and H$^•$ radicals as reaction intermediates.



Photochromism

Y. Wu, Y. Xie, Q. Zhang, H. Tian, W. Zhu,* A. D. Q. Li* ______ **2090 – 2094**

Quantitative Photoswitching in Bis(dithiazole)ethene Enables Modulation of Light for Encoding Optical Signals



Molecular photoswitches: A bis(dithiazole)ethane-based compound can be photoswitched between its ring-open and ring-closed states quantitatively with excellent fatigue resistance and high thermal stability. The quantitative reversibility allows the photoswitching light to control other light travelling through the medium, a phenomenon of transferring information encoded in one light ray to others.


Angewandte Chemie
International Edition
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Medication Detection by a Combinatorial Fluorescent Molecular Sensor
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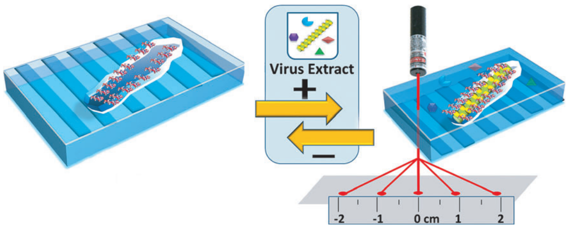
Biosensors

W. Bai, D. A. Spivak* ______ **2095 – 2098**

A Double-Imprinted Diffraction-Grating Sensor Based on a Virus-Responsive Super-Aptamer Hydrogel Derived from an Impure Extract

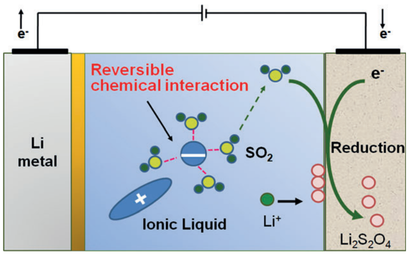


Eye see it: A strategy for the naked-eye detection of target compounds by using shrink-responsive hydrogels is presented. A "double imprinting" method was used, in which a virus-bioimprinted hydrogel is further micromolded into a diffraction-grating sensor by using imprint-lithography techniques. The bioimprinting was performed with an impure extract, which eliminates the need for a pure template.

Lithium Batteries

H. B. Xing,* C. Liao, Q. W. Yang,
G. M. Veith, B. K. Guo, X. G. Sun,*
Q. L. Ren, Y. S. Hu, S. Dai* **2099 – 2103**

Ambient Lithium–SO_2 Batteries with Ionic Liquids as Electrolytes

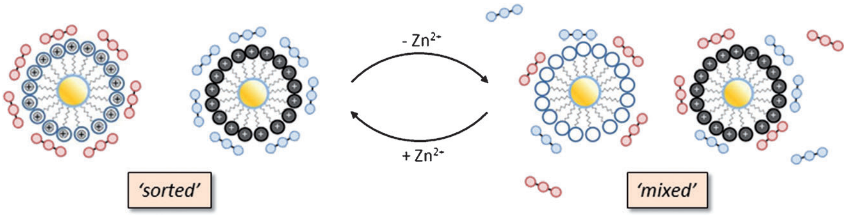


A novel ambient Li-SO_2 battery was developed through the introduction of ionic liquid (IL) electrolytes with tailored basicities to capture SO_2 by a reversible chemical absorption (see picture). By tuning the interactions of ILs with SO_2, a high energy density and good discharge performance with operating voltages above 2.8 V were obtained.

Self-Sorting

C. Pezzato, P. Scrimin,
L. J. Prins* ______ **2104 – 2109**

Zn^{2+}-Regulated Self-Sorting and Mixing of Phosphates and Carboxylates on the Surface of Functionalized Gold Nanoparticles

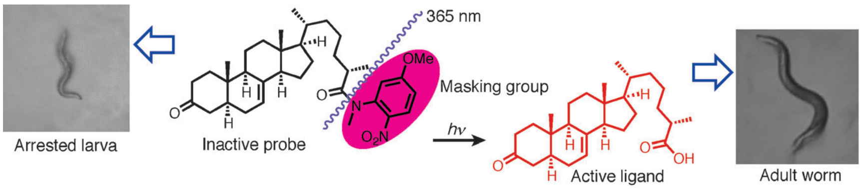


Order versus chaos: A mixture of phosphate- and carboxylate-containing molecules spontaneously separated into the two components on the surface of two different nanoparticles in the presence of Zn^{2+}. The self-sorting process could be reversed by the addition of a scavenger ligand for the Zn^{2+} ions (see picture).

Optogenetics

J. C. Judkins, P. Mahanti, J. B. Hoffman,
I. Yim, A. Antebi,
F. C. Schroeder* ______ **2110 – 2113**

A Photocleavable Masked Nuclear-Receptor Ligand Enables Temporal Control of *C. elegans* Development

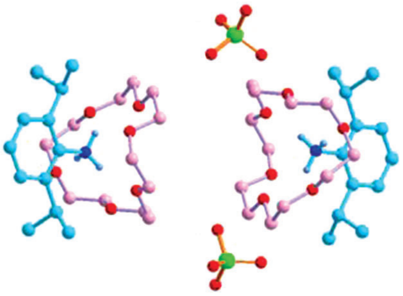


Life goes by in a flash: Photocleavable amides of recently identified *C. elegans* steroids that bind to a vitamin D receptor homologue enable precise temporal control of worm development and lifespan. The synthetic amide-masked receptor ligands are biocompatible and are retained in the worm in their inactive forms for many days.

Molecular Ferroelectrics

Y. Zhang, H.-Y. Ye, D.-W. Fu, R.-G. Xiong* ______ 2114 – 2118

An Order–Disorder Ferroelectric Host–Guest Inclusion Compound

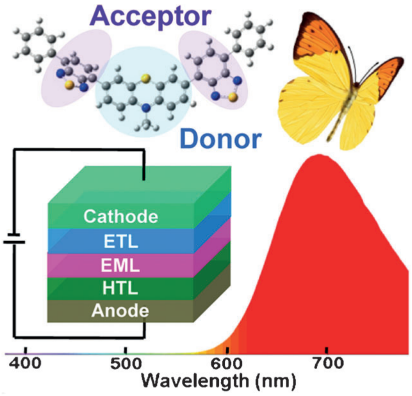


The host–guest inclusion compound [2,6-diisopropylanilinium([18]crown-6)](ClO_4) (see picture, C blue/pink, N dark blue, O red, Cl green) exhibits a paraelectric–ferroelectric phase transition at 132 K. The origin of the ferroelectricity is attributed to order–disorder transitions of the host [18]crown-6 and ClO_4^- ion rather than the rotation of the guest cation, in contrast to other rotational host–guest inclusion ferroelectrics.

Optoelectronic Materials

L. Yao, S. Zhang, R. Wang, W. Li, F. Shen, B. Yang,* Y. Ma* ______ 2119 – 2123

Highly Efficient Near-Infrared Organic Light-Emitting Diode Based on a Butterfly-Shaped Donor–Acceptor Chromophore with Strong Solid-State Fluorescence and a Large Proportion of Radiative Excitons

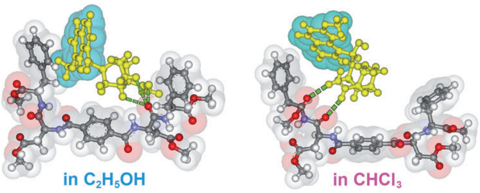


Exceeding the limit: An organic light-emitting diode (OLED) based on a butterfly-shaped near-infrared donor–acceptor chromophore (see structure) with efficient solid-state and aggregation-induced emission behavior showed a high external quantum efficiency of 1.54%. Its radiative exciton ratio of 48% greatly surpassed the limit of 25% in conventional fluorescent OLEDs.

Chiral Gels

G. Qing, X. Shan, W. Chen, Z. Lv, P. Xiong, T. Sun* ______ 2124 – 2129

Solvent-Driven Chiral-Interaction Reversion for Organogel Formation

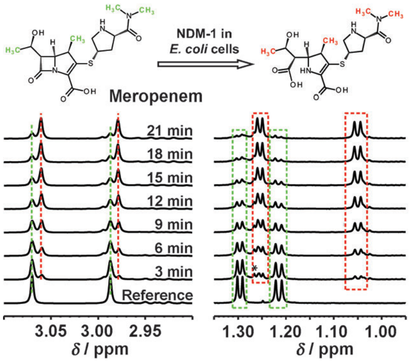


Get smart: A system for gel formation in which the solvent not only provides a medium but also determines the nature of the stereoselective interaction between a gelator and a chiral guest provides important insight into the assembly of chiral gelators. The solvent-driven preferential complexation of one or another chiral guest (see picture) enables the design of organogels with new "smart" features for various applications.

In Vivo NMR Spectroscopy

J. Ma, S. McLeod, K. MacCormack, S. Sriram, N. Gao, A. L. Breeze,* J. Hu* ______ 2130 – 2133

Real-Time Monitoring of New Delhi Metallo-β-Lactamase Activity in Living Bacterial Cells by ^1H NMR Spectroscopy

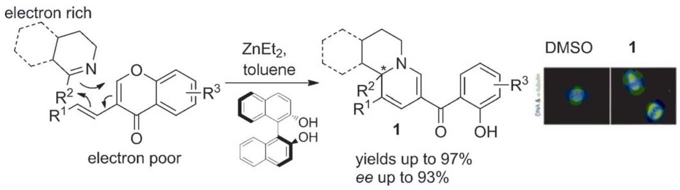


Spying on superbugs: The hydrolysis of the antibiotic meropenem by *Escherichia coli* cells carrying the gene for New Delhi Metallo-β-Lactamase (NDM-1), which confers antibiotic resistance, was monitored by NMR spectroscopy in real time. IC_{50} values for NDM-1 inhibitors were assessed in vivo. Spermine, a porin inhibitor, was shown to reduce the rate of meropenem degradation in living cells, although in vitro, the inhibitory effect of spermine is minimal.

Asymmetric Synthesis

V. Eschenbrenner-Lux, P. Küchler,
S. Ziegler, K. Kumar,*
H. Waldmann* ________ 2134 – 2137

An Enantioselective Inverse-Electron-Demand Imino Diels–Alder Reaction



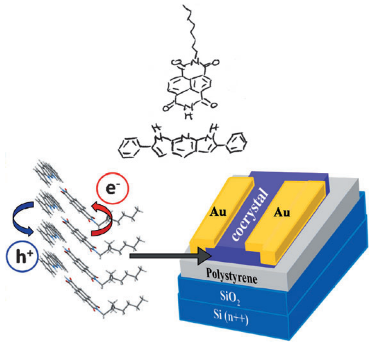
Inverse demand! The first enantioselective inverse electron-demand imino Diels–Alder reaction involving electron-poor dienes has been developed and used to generate a natural product inspired compound collection which revealed a highly potent modulator of mitosis.

Organic Semiconductors

H. T. Black,
D. F. Perepichka* ________ 2138 – 2142

Crystal Engineering of Dual Channel p/n Organic Semiconductors by Complementary Hydrogen Bonding

Inside Back Cover

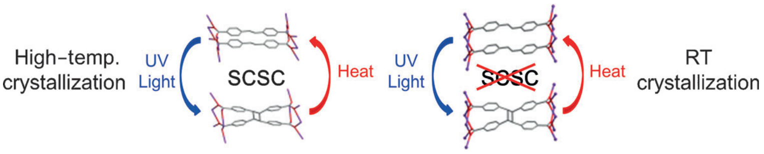


A complementary pair: Complementary hydrogen bonding has been used to control the solid-state arrangement of donor/acceptor components in crystalline p/n heterojunctions (see picture). The self-assembly process by hydrogen bonding modulates the charge-transfer interactions between the donor and acceptor, makes possible precise control over the heterojunction structure, and leads to a combination of charge-transport properties of the individual components.

Reversible C–C Bond Formation

G. K. Kole, T. Kojima, M. Kawano,*
J. J. Vittal* ________ 2143 – 2146

Reversible Single-Crystal-to-Single-Crystal Photochemical Formation and Thermal Cleavage of a Cyclobutane Ring

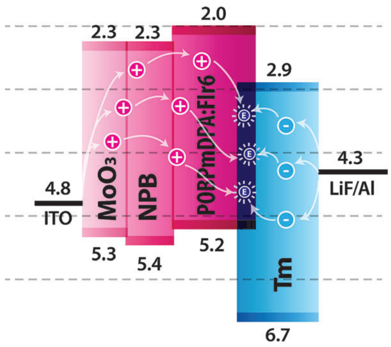


You can be recycled, Mr. Bond! The influence of crystal packing on the retention of single-crystal nature during cyclobutane formation and cleavage was investigated. The polymorph formed at high temperature retains its single-crystal (SC) nature during reversible photochemical formation and thermal cleavage in a SCSC manner, whereas the polymorph obtained at room temperature does not.

Blue Phosphorescent Organic LEDs

C. Fan, L. Zhu, T. Liu, B. Jiang, D. Ma,*
J. Qin, C. Yang* ________ 2147 – 2151

Using an Organic Molecule with Low Triplet Energy as a Host in a Highly Efficient Blue Electrophosphorescent Device

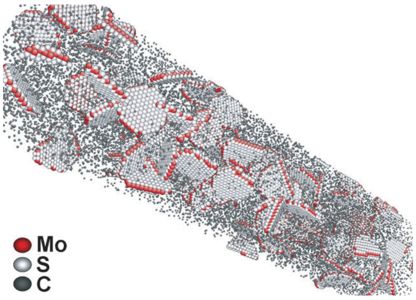


A host material with low triplet energy (2.71 eV) was designed by integrating diarylamine and diphenylphosphine oxide into a biphenyl skeleton. Highly efficient organic LEDs that are based on the blue phosphor FIr6, with a maximum current efficiency of 37 cd A^{-1} and power efficiency of 40 Lm W^{-1}, were achieved by using this host material.

Energy Storage

C. Zhu, X. Mu, P. A. van Aken, Y. Yu,*
J. Maier* **2152–2156**

Single-Layered Ultrasmall Nanoplates of MoS_2 Embedded in Carbon Nanofibers with Excellent Electrochemical Performance for Lithium and Sodium Storage

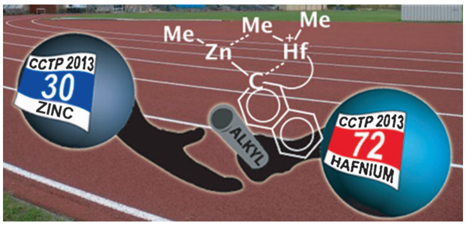


Single-layered ultrasmall nanoplates (thickness ca. 0.4 nm and lateral dimension ca. 4.0 nm) of MoS_2 embedded in thin carbon nanofibers were prepared by electrospinning followed by heat treatment. The high rate performance and cycling stability for both lithium and sodium storage is remarkable and also fundamentally exciting as the conversion reactions are expected to become reversible for such isolated nanodots.

Heterobimetallic Complexes

L. Rocchigiani, V. Busico, A. Pastore,
G. Talarico,* A. Macchioni* **2157–2161**

Unusual Hafnium–Pyridylamido/ER_n Heterobimetallic Adducts ($ER_n = ZnR_2$ or AlR_3)

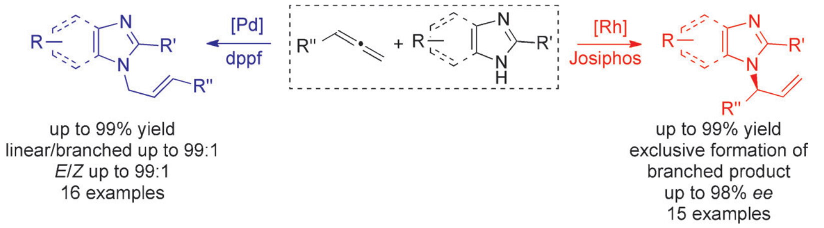


Pass the alkyl: NMR spectroscopy and DFT calculations provide compelling evidence for the formation of heterobimetallic $[\{N^-,N\}HfMe(\mu\text{-}R)(\mu\text{-}C_{aryl})ER_{n-1}][B(C_6F_5)_4]$ adducts when industrially relevant Symyx/Dow Hf^{IV}–pyridylamido catalysts for (coordinative chain transfer) olefin polymerization interact with ER_n (E = Al or Zn, R = alkyl group).

Asymmetric Catalysis

K. Xu, N. Thieme, B. Breit* **2162–2165**

Atom-Economic, Regiodivergent, and Stereoselective Coupling of Imidazole Derivatives with Terminal Allenes

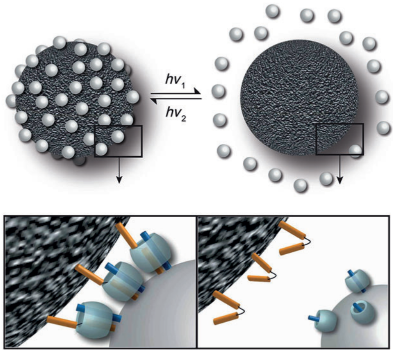


Taking control: New Rh- and Pd-catalyzed regiodivergent and stereoselective intermolecular coupling reactions of imidazole derivatives with monosubstituted allenes are reported. Using a Rh^I/Josiphos system, perfect regioselectivities and high enantiomeric excess were obtained, while a Pd^{II}/dppf system gave the linear products with high regioselectivities and high *E*/*Z* selectivities. dppf = 1,1′-bis(diphenylphosphino)ferrocene, Josiphos = (*R*)-1-[(S_P)-2-(dicyclohexylphosphino)ferrocenyl]ethyldialkylphosphine.

Supramolecular Self-Assembly

Y. Lan, Y. Wu, A. Karas,
O. A. Scherman* **2166–2169**

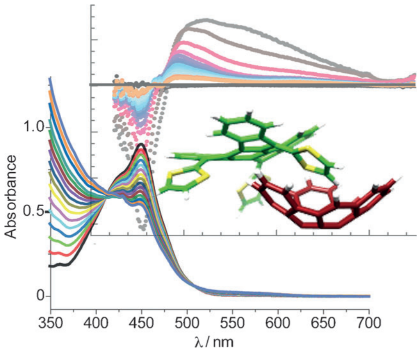
Photoresponsive Hybrid Raspberry-Like Colloids Based on Cucurbit[8]uril Host–Guest Interactions



Reversible assembly of colloids: Hybrid raspberry-like colloids prepared by employing cucurbit[8]uril as a supramolecular linker to assemble functional polymeric nanoparticles onto silica microspheres can be disassembled and reassembled in an efficient and controllable manner (see picture). This supramolecular self-assembly method makes possible the preparation of colloids with sophisticated structures and properties.

Fullerene Fragments

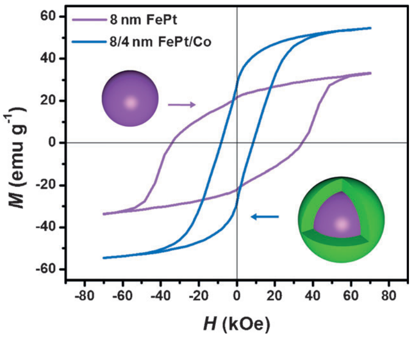
Front Cover



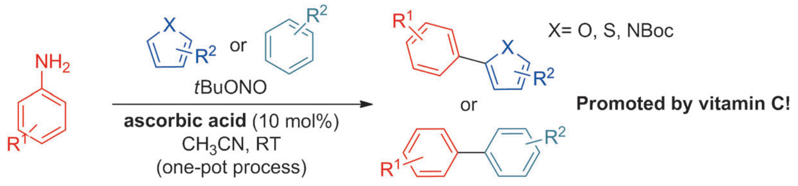
Bowl meets bowl: The association of a fullerene fragment, hemifullerene $C_{30}H_{12}$, with an electron-donating bowl-shaped tetrathiafulvalene derivative (truxTTF) has been investigated. Electron transfer from truxTTF to $C_{30}H_{12}$ to form the fully charge-separated species takes place upon irradiation of the associate with light, constituting the first example in which a fullerene fragment mimics the electron-accepting behavior of the fullerenes within a supramolecular complex.

Intermetallic Phases

Exchange-coupled nanoparticles: A general protocol is reported for the synthesis of exchange-coupled nanoparticles composed of hard magnetic face-centered tetragonal FePt as the core and soft magnetic Co (or Ni, Fe_2C) as the shell. These core/shell nanoparticles show shell-thickness-dependent magnetic properties (see picture; H = applied field, M = magnetization).

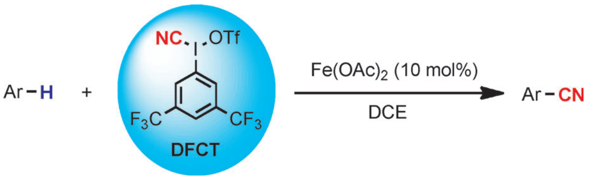


C–H Arylation



Vitamin Coupling: A catalytic amount of ascorbic acid acted as a radical initiator in a metal-free direct C–H arylation of arenes and heteroarenes with anilines (see scheme; Boc = *tert*-butoxycarbonyl). The reaction requires no heating or irradiation, is operationally simple, and constitutes a greener approach to arylation.

Synthetic Methods



Direct facile cyanation today! A direct cyanation of aryl C–H bonds with 3,5-di(trifluoromethyl)phenyl(cyano)iodonium triflate (DFCT) under Fe^{II} catalysis was successfully applied to a wide range of aromatic substrates, including polycyclic structures and heteroaromatic compounds (see scheme). The reaction proceeded efficiently under mild conditions.

Synthetic Biology

Z. Xiang, V. K. Lacey, H. Ren, J. Xu,
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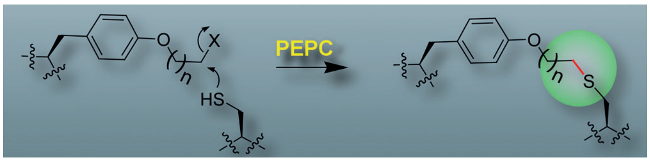
L. Wang* ———— 2190 – 2193

Proximity-Enabled Protein Crosslinking through Genetically Encoding Haloalkane Unnatural Amino Acids

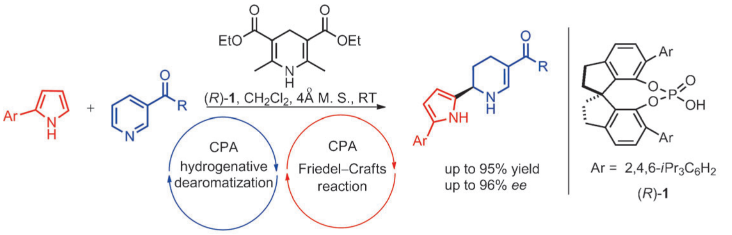


Building bridges: By expanding the genetic code, a series of haloalkane unnatural amino acids (Uaas) were incorporated into proteins at specific sites. These Uaas only form a covalent thioether bond with cysteine when the two residues are in proximity. This strategy of proximity-enabled protein crosslinking (PEPC) provides diversity and flexibility in covalent bond formation for protein research and protein engineering.

Asymmetric Catalysis

S.-G. Wang, S.-L. You* ———— 2194 – 2197

Hydrogenative Dearomatization of Pyridine and an Asymmetric Aza-Friedel–Crafts Alkylation Sequence

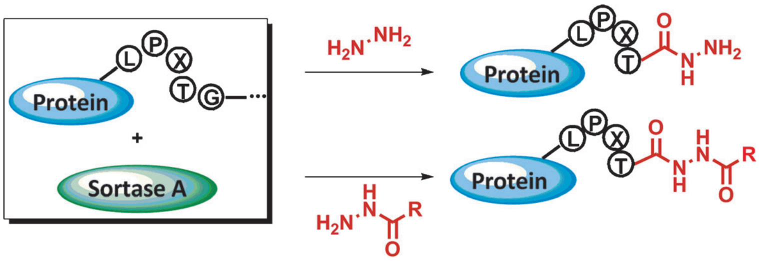


The highly efficient synthesis of enantiomerically enriched substituted piperidines in good to excellent yields and enantioselectivity has been realized by using the title reaction, which is catalyzed by the chiral phosphoric acid (CPA) (*R*)-**1**.

Protein Modifications

Y.-M. Li, Y.-T. Li, M. Pan, X.-Q. Kong,
Y.-C. Huang, Z.-Y. Hong,
L. Liu* ———— 2198 – 2202

Irreversible Site-Specific Hydrazinolysis of Proteins by Use of Sortase

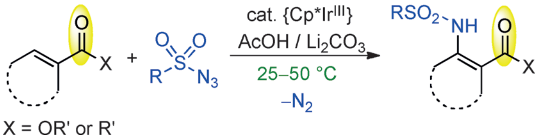


Takes all sort(ase)s: Sortase A mediated hydrazinolysis reactions of proteins with hydrazine or its derivatives were developed as an efficient process for the production of recombinant protein hydrazides. This reaction provides an alternative approach for the semichemical synthesis as well as C-terminal modification of proteins.

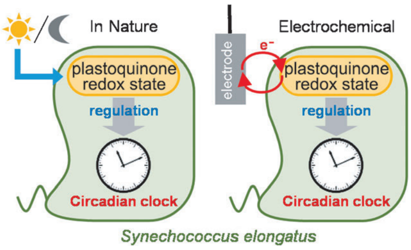
C–H Amidation

J. Kim, S. Chang* ———— 2203 – 2207

Iridium-Catalyzed Direct C–H Amidation with Weakly Coordinating Carbonyl Directing Groups under Mild Conditions

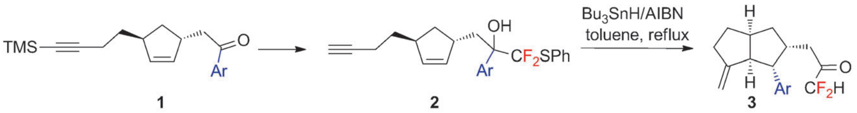


Mild and direct: The iridium-catalyzed direct C–H amidation of weakly coordinating substrates, particularly of those with ester and ketone groups, under very mild conditions has been developed. With the aid of a combined use of acetic acid and lithium carbonate as additives, the reaction proceeded with high efficiency. Cp* = pentamethylcyclopentadienyl.



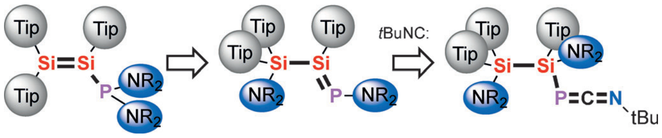
Let's do the time warp again: Electrochemically controlled extracellular electron transfer can regulate the circadian clock of the cyanobacterium *Synechococcus elongatus* under constant light intensity through periodic modification of the intracellular redox state. This method could potentially allow for direct or indirect regulation of the biological clock across a range of species.

Circadian Rhythm



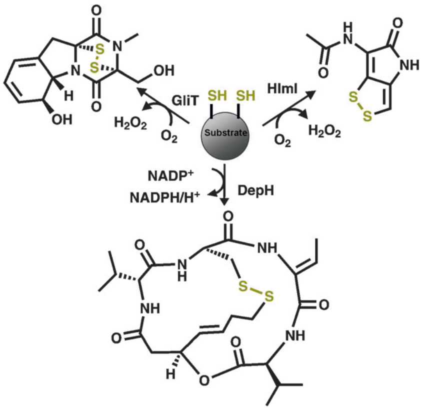
Rad cascades: The title reaction of the compounds **2**, which are readily obtained by fluoride-catalyzed nucleophilic addition of PhSCF$_2$TMS to the chiral ketocyclopentenes **1**, was found to provide novel access to the asymmetric synthesis of the 3,3-difluoro-2-propanoylbicyclo[3.3.0]octanes **3**. AIBN = 2,2′-azobis(2-methylpropionitrile), TBAF = tetra-*n*-butylammonium fluoride, THF = tetrahydrofuran, TMS = trimethylsilyl.

Synthetic Methods



By-product free conversion of double bonds between heavier elements occurs by the sequential shifting of amino substituents (NR$_2$; see picture, blue) from phosphorus to the adjacent silicon atoms. The spontaneous isomerization of the plausibly transient phosphinodisilenes (Tip = 2,4,6-*i*Pr$_3$C$_6$H$_2$) affords phosphasilenes, one of which is transformed upon addition of an isonitrile into a 1-aza-3-phosphaallene.

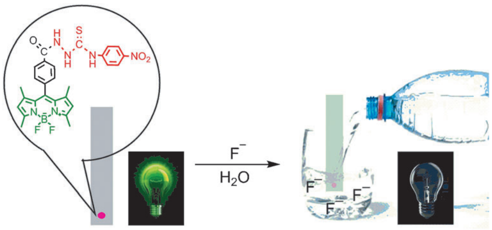
Heavier Double Bonds



Whiff of sulfur: Disulfide bonds are essential for the bioactivity of natural products such as gliotoxin, a virulence factor of the human pathogen *A. fumigatus*, the antibiotic holomycin (*S. clavuligerus*), and the anticancer drug romidepsin (*C. violaceum*). Structural and biochemical analyses of the thiol oxidases GliT, HlmI, and DepH identify a common reaction mechanism but with different substrate tolerance.

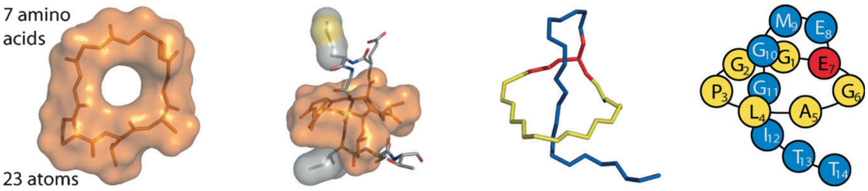
Thiol Oxidation

Fluoride Sensing



Dip and detect: BODIPY dyes with strong hydrogen-bond-donating amidothiourea receptors can detect fluoride in organic–aqueous media. The probe embedded in a nitrocellulose test strip enables a quantitative detection of fluoride in pure aqueous medium by using a simple lateral-flow fluorescence reader.

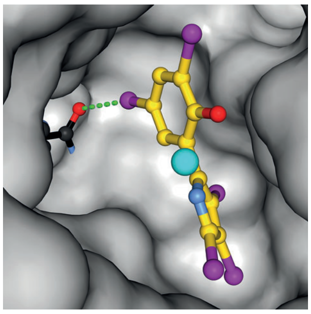
Lasso Peptides



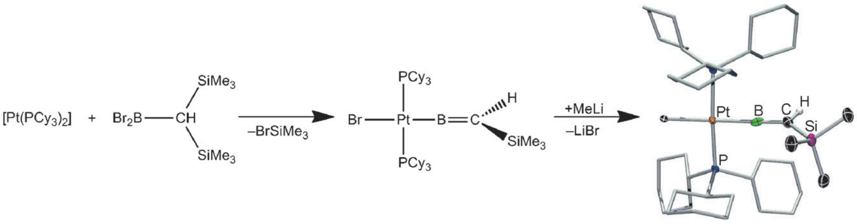
Lowering the bar: Xanthomonins I–III are the first examples of lasso peptides that feature macrolactam rings with seven residues. The structures of xanthomonins I and II (see picture) are described, together with a thorough mutational analysis that revealed the smallest amino acid residue that can still act as a plug to maintain a thermally stable lasso fold in this new class of lasso peptides.

Herbicides

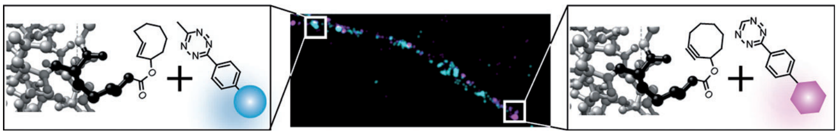
Halogenated natural alkaloids: Nowadays, many organisms develop resistances against known antiinfectives or herbicides. Therefore, new chemical entities as well as new targets are in the focus of current research. Pseudilins, highly halogenated marine natural products, inhibit the third enzyme of the non-mevalonate pathway (IspD) by an allosteric mode of action by using halogen bonding and metal ion coordination. Pseudilins thus show herbicidal and antiplasmodial activity.



Alkylideneboryl Complexes



The oxidative addition of an alkyldibromoborane to a zerovalent platinum complex with subsequent elimination of the bromosilane at room temperature leads to the first terminally coordinated transition metal alkylideneboryl complex. The *trans* bromide ligand can be exchanged for a methyl group through salt metathesis (see scheme). DFT calculations revealed that like imino- and oxoboryl ligands, the alkylideneboryl ligand is an excellent σ-donor and displays a strong *trans* influence.



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Angewandte Corrigendum

The authors of this Communication wish to cite an additional contribution. For this reason, reference [18] must be added on page 13606, right column, at the end of the second paragraph: "Direct transformation of readily available alkanes into valuable complex alkyl ethers by transition-metal-catalyzed C(sp3)–H functionalization of unactivated methylenes is arguably a highly efficient and atom-economic method toward these compounds.[18]"

An Efficient Palladium-Catalyzed C–H Alkoxylation of Unactivated Methylene and Methyl Groups with Cyclic Hypervalent Iodine (I^{3+}) Oxidants

G. Shan, X. Yang, Y. Zong,
Y. Rao* ———— **13606–13610**

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[18] During the revision our manuscript, an alkyl ether synthesis (alkoxylation of the β-C(sp^3)–H bonds) through sp^3 (methylene) C–H activation was reported by Shi's group with a new pyridine-based bidentate directing group and $PhI(OAc)_2$ as the oxidant. γ-Alkoxylation of C(sp^2)–H bonds was also performed in this work: F. Chen, S. Zhao, F. Hu, K. Chen, Q. Zhang, S. Zhang, B. Shi, *Chem. Sci.* **2013**, *4*, 4187..