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

*A. B. Chaplin, A. S. Weller**
B–H Activation at a Rhodium(I) Center: A Missing Link in the Transition-Metal-Catalyzed Dehydrocoupling of Amine–Boranes

*I. U. Khan, D. Zwanziger, I. Böhme, M. Javed, H. Naseer, S. W. Hyder, A. G. Beck-Sickinger**
Breast Cancer Diagnosis by Neuropeptide Y Analogues: From Synthesis to Clinical Application

Y.-S. Li, F.-Y. Liang, H. Bux, A. Feldhoff, W.-S. Yang, J. Caro**
Metal–Organic Framework Molecular Sieve Membrane: Supported ZIF-7 Layer with High Hydrogen Selectivity by Microwave-Assisted Seeded Growth

S. Yamago, Y. Watanabe, T. Iwamoto*
Synthesis of [8]Cycloparaphenylene from a Square-Shaped Tetranuclear Platinum Complex $[\{Pt(cod)(4,4'\text{-biphenyl})\}_4]$

S. M. Lang, T. M. Bernhardt, R. N. Barnett, U. Landman**
Methane Activation and Catalytic Ethylene Formation on Free Au_2^+

*E. Kang, H. Su Min, J. Lee, M. H. Han, H. J. Ahn, I.-C. Yoon, K. Choi, K. Kim, K. Park, I. C. Kwon**
Nanobubbles from Gas-Generating Polymeric Nanoparticles for Ultrasound Imaging of Living Subjects

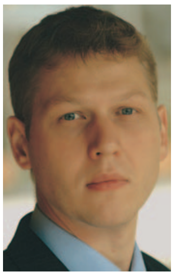
*A. M. Todea, A. Merca, H. Bögge, T. Glaser, J. M. Pigga, M. L. Langston, T. Liu, R. Prozorov, M. Luban, C. Schröder, W. H. Casey, A. Müller**
Porous Capsules $\{(M)M_5\}_{12}Fe^{III}_{30}$ (M = Mo^{VI}, W^{VI}): Sphere-Surface Supramolecular Chemistry with Twenty Ammonium Ions, Related Solution Properties, and Tuning of Magnetic Exchange Interactions

Author Profile

"I chose chemistry as a career because I could make things that did not exist.
If I wasn't a scientist, I would be a writer ..."
This and more about Luis Echegoyen can be found on page 248.

News

G. Ertl



M. Rueping

C. J. Chang

Books

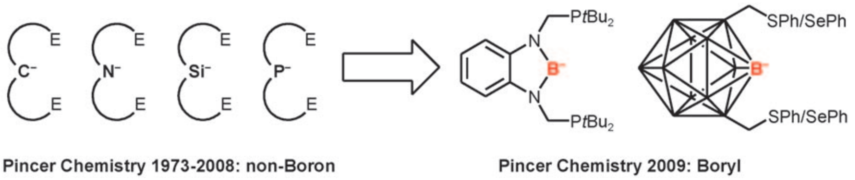
Contents

Highlights

Tridentate Boryl Ligands

J. I. van der Vlugt* ______ **252–255**

Boryl-Based Pincer Systems: New Avenues in Boron Chemistry



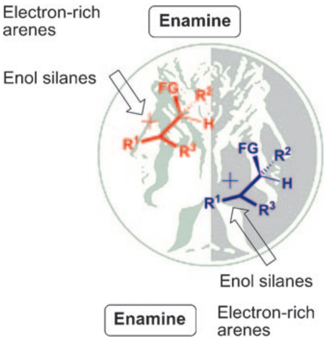
To B or not to B? Incorporation of BH into tridentate ligands has recently allowed the isolation of the first boryl-based pincer complexes. Functionalized diaminoborane and *m*-carbaborane (see picture) form Ir and Pd complexes, respectively. Novel reactivity, including in catalysis, is expected to result from the M−boryl bond and electronic communication with the specific ligand framework.

Carbocations

P. G. Cozzi,* F. Benfatti ______ **256–259**

Stereoselective Reactions with Stabilized Carbocations

Blocking one face: Benzylic carbocations, which are easily generated in situ from alcohols or acetates by Brønsted or Lewis acids, undergo selective facial discrimination in diastereoselective reactions (see scheme; FG = functional group). The A values are responsible for the facial selectivity. Catalytic amounts of various Lewis acids can be employed in discriminating one face of the carbocation.

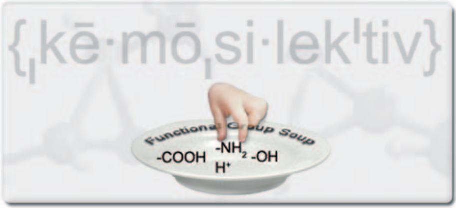


Reviews

Synthesis Design

N. A. Afagh, A. K. Yudin* ______ **262–310**

Chemoselectivity and the Curious Reactivity Preferences of Functional Groups



Spoilt for choice? Chemoselectivity is a central aspect of chemical synthesis, but the endeavor to find the right reaction pathway from the many that are often available can be tedious. A careful analysis of the reaction mechanism can usually help to arrive at the "right pathway".

For the USA and Canada:
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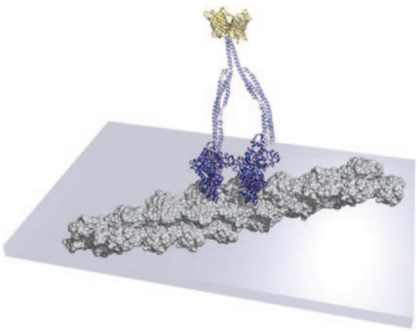
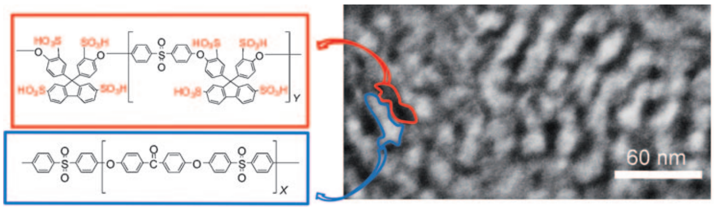
Communications

Nanotechnology



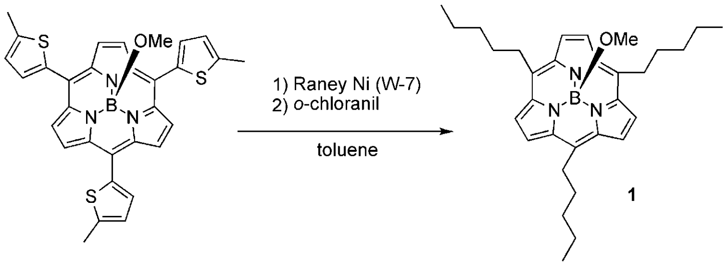
A long life with controlled movement: Engineered protein nanomachines with defined and adjustable functional features offer opportunities for controlling movement on nano- to microscale biohybrid devices. Myosin motors (the picture shows a dimeric motor) have now been constructed with optimized properties with regard to stability and regulation, including switchable processivity and tight control of the velocity of movement.

Fuel-Cell Membranes



Segregation is a key theme of this film: Poly(arylene ether sulfone ketone) multiblock membranes containing highly sulfonated hydrophilic blocks were prepared by postsulfonation. High local concentrations of sulfonic acid groups within the hydrophilic blocks enhanced the phase separation of the two block types (see picture), and the high interconnectivity of the rodlike hydrophilic aggregates led to high proton conductivity even at low humidity.

Porphyrinoids



The desulfurization of meso-trithienylsubporphyrins leads to meso-trialkylsubporphyrins (see scheme). Oxidation at the benzylic position of the meso-alkyl substituent of **1** has been used to prepare a valeryl-substituted subporphyrin that exhibits highly perturbed optical and electrochemical properties compared to other subporphyrins.

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Posters

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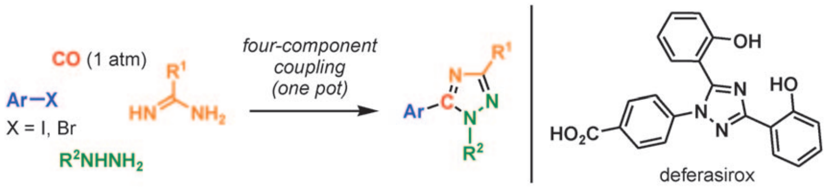

All 4 one: Palladium-catalyzed carbonylation initiates a highly regioselective triazole synthesis. In this four-partner protocol, the aryl halide, amidine, and hydrazine partners are easily varied, allowing the synthesis of fully substituted 1,2,4-triazoles in a modular fashion. This methodology is demonstrated with the synthesis of druglike and/or pharmaceutically relevant molecules such as deferasirox.

Multicomponent Reactions

S. T. Staben,* N. Blaquiere ______ **325 – 328**

Four-Component Synthesis of Fully Substituted 1,2,4-Triazoles

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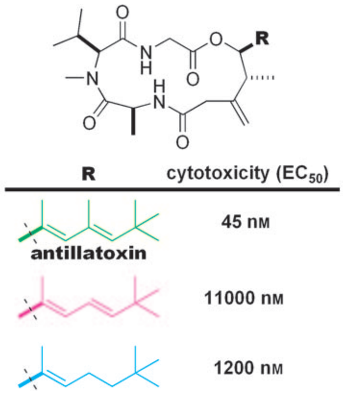

Side-chain plays lead role: Antillatoxin is a cyclic peptide and potent neurotoxin, and the total syntheses and biological evaluations of antillatoxin and its side-chain analogues are reported. A subtle side-chain modification led to dramatic changes in the cytotoxicity; detailed conformational analyses revealed that the shape of the twisted side chain is critical for the biological activity of antillatoxin.

Natural Products

K. Okura, S. Matsuoka, R. Goto, M. Inoue* ______ **329 – 332**

The Twisted Side Chain of Antillatoxin is Important for Potent Toxicity: Total Synthesis and Biological Evaluation of Antillatoxin and Analogues

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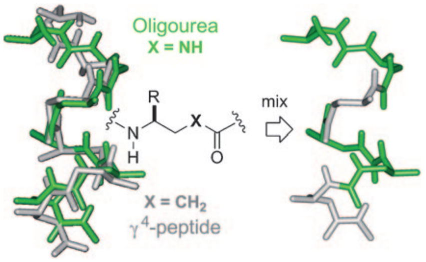
Fraternal twins: Oligoureas and γ-peptides are isosteric, quasi-isostructural helical foldamers endowed with distinct biomolecular recognition properties. Combination of the two backbones to generate urea/amide hybrids (see picture) was found to give more potent yet less cytotoxic antimicrobial helical foldamers.



Helical Foldamers

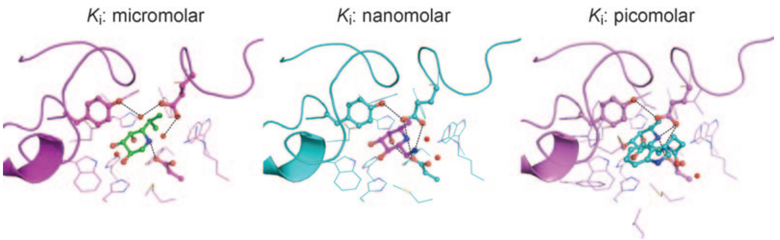
P. Claudon, A. Violette, K. Lamour, M. Decossas, S. Fournel, B. Heurtault, J. Godet, Y. Mély, B. Jamart-Grégoire, M.-C. Averlant-Petit, J.-P. Briand, G. Duportail, H. Monteil, G. Guichard* ______ **333 – 336**

Consequences of Isostructural Main-Chain Modifications for the Design of Antimicrobial Foldamers: Helical Mimics of Host-Defense Peptides Based on a Heterogeneous Amide/Urea Backbone

Enzyme–Inhibitor Complexes

H.-J. Wu, C.-W. Ho, T.-P. Ko, S. D. Popat,
C.-H. Lin,* A. H.-J. Wang* ____ **337–340**

Structural Basis of α-Fucosidase Inhibition by Iminocyclitols with K_i Values in the Micro- to Picomolar Range



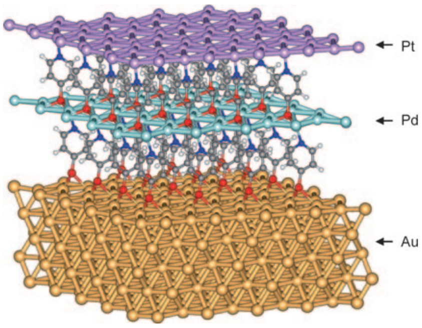
Two loops were found to move inward toward the α-fucosidase active site to produce a closed conformation of complexes with inhibitors with increasing K_i values from the micro- to nanomolar range. Although no further conformational changes in the two loops are observed for inhibitors with sub-nanomolar K_i values, the loops are additionally stabilized by hydrogen bonds and hydrophobic interactions.

Hybrid Composites

F. Eberle, M. Saitner, H.-G. Boyen,*
J. Kucera, A. Gross, A. Romanyuk,
P. Oelhafen, M. D'Olieslaeger,
M. Manolova, D. M. Kolb ____ **341–345**

A Molecular Double Decker: Extending the Limits of Current Metal–Molecule Hybrid Structures

Towards the third dimension: A new type of organic–inorganic hybrid comprises two independent molecular layers separated by a metal monolayer and sandwiched between outer electrodes (see picture). The interlayer reveals metallic properties despite its reduced dimensions and the presence of chemical bonds at both sides. It might be useful as an intermediate electrode to control charge transport through molecule-based devices.

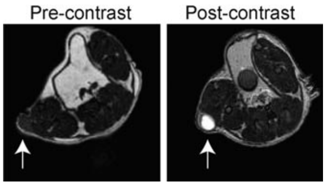


MRI Contrast Agents

Z. Cheng, D. L. J. Thorek,
A. Tsourkas* ____ **346–350**

Gadolinium-Conjugated Dendrimer Nanoclusters as a Tumor-Targeted T_1 Magnetic Resonance Imaging Contrast Agent

Light heavyweight champion: Chemically cross-linked dendrimer nanoclusters (DNCs) have been developed as a platform for preparing targeted magnetic resonance contrast agents. The extremely high gadolinium payload causes the DNCs to show a significant improvement in tumor contrast compared with precontrast images (see picture).

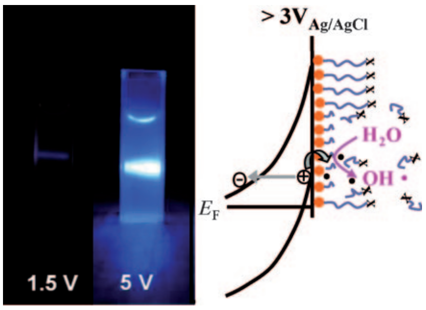


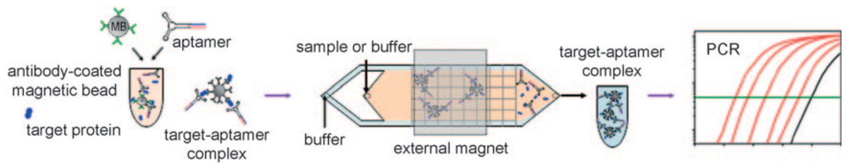
Dark Photocatalysis

Y. Y. Song, P. Roy, I. Paramasivam,
P. Schmuki* ____ **351–354**

Voltage-Induced Payload Release and Wettability Control on TiO_2 and TiO_2 Nanotubes

Please release me: Hydrocarbon monolayers attached to a TiO_2 surface can be cut by electrochemical means, thus allowing active molecules or proteins to be released from the TiO_2 surface (see picture). Requirements are suitable electronic properties of TiO_2 combined with a voltage to cause Schottky barrier breakdown; valence band holes are then sufficiently reactive to generate $OH^{\bullet}$ radicals.





On the MAP: The title technique combines microfluidic magnetic sample preparation with highly specific aptamer-based molecular recognition and quantitative PCR. Using platelet-derived growth factor as a model, a fM detection limit was obtained in fetal bovine serum with a dynamic range that spanned five orders of magnitude.

Microfluidic Biotechnology

A. Csordas, A. E. Gerdon, J. D. Adams, J. Qian, S. S. Oh, Y. Xiao,* H. T. Soh* ______ **355 – 358**

Detection of Proteins in Serum by Micromagnetic Aptamer PCR (MAP) Technology

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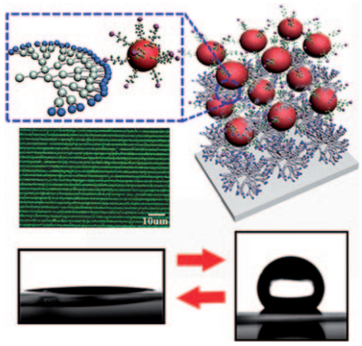

Highly stable photoluminescent (PL) nanocomposite multilayers including quantum dots (see picture) were prepared using a nucleophilic substitution reaction. The assembly of functional nanoparticles in a nonpolar solvent can allow selective deposition and induce a hydrophobic surface with a water contact angle above 115°, which significantly enhances the PL durability of nanocomposite films.

Nanocomposites

B. Lee, Y. Kim, S. Lee, Y. S. Kim, D. Wang, J. Cho* ______ **359 – 363**

Layer-by-Layer Growth of Polymer/Quantum Dot Composite Multilayers by Nucleophilic Substitution in Organic Media

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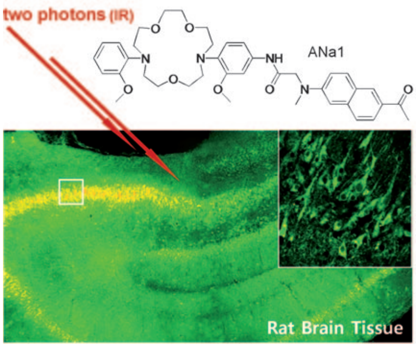
Two photons are better than one: A two-photon probe (ANa1, see picture) shows strong two-photon fluorescence enhancement in response to Na$^+$ and a dissociation constant ($K_d^{\,i}$) of (26 ± 2) mM in the cell. It can be easily loaded into the cells and can selectively detect intracellular free Na$^+$ in live cells and living tissues at a depth of 100–200 μm for a long period of time.



Fluorescent Probes

M. K. Kim, C. S. Lim, J. T. Hong, J. H. Han, H. Y. Jang, H. M. Kim,* B. R. Cho* ______ **364 – 367**

Sodium-Ion-Selective Two-Photon Fluorescent Probe for In Vivo Imaging

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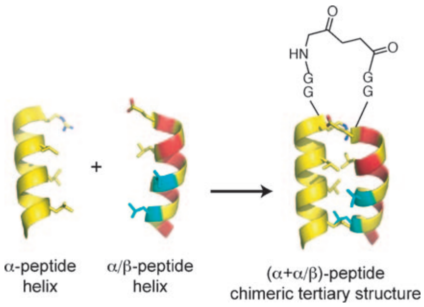


Backbone thioester exchange is used to explore a fundamental type of protein–foldamer packing motif, the association of an α helix and an α/β-peptide foldameric helix, which is analogous to an antiparallel coiled-coil tertiary structure in a pure α-residue backbone. Side-chain packing preferences at this chimeric tertiary interface are comparable to those that determine pairing propensities among antiparallel α helices.

Protein Folding

J. L. Price, E. B. Hadley, J. D. Steinkruger, S. H. Gellman* ______ **368 – 371**

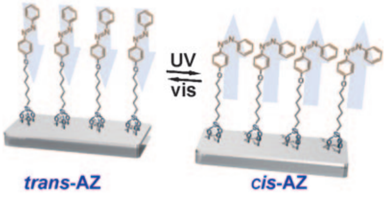
Detection and Analysis of Chimeric Tertiary Structures by Backbone Thioester Exchange: Packing of an α Helix against an α/β-Peptide Helix

Contents

Photocontrolled Superconductivity

A. Ikegami, M. Suda, T. Watanabe, Y. Einaga* ______ **372–374**

Reversible Optical Manipulation of Superconductivity



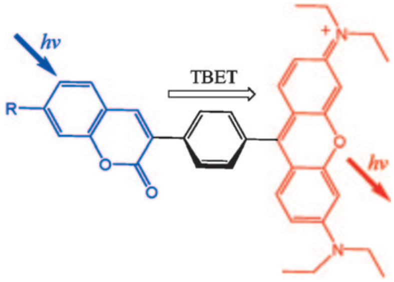
Flicking the switch: Photoirradiation has a dramatic effect on the superconducting properties of a passivated ultrathin Nb film functionalized with a photochromic self-assembled azobenzene (AZ) monolayer (see picture). These effects can be attributed to photoisomerization-induced changes in the amount of charge transfer between the substrate and the photochromic monolayer. The changes are accompanied by alternation of the surface dipoles.

Fluorescent Dyes

W. Lin,* L. Yuan, Z. Cao, Y. Feng, J. Song ______ **375–379**

Through-Bond Energy Transfer Cassettes with Minimal Spectral Overlap between the Donor Emission and Acceptor Absorption: Coumarin–Rhodamine Dyads with Large Pseudo-Stokes Shifts and Emission Shifts

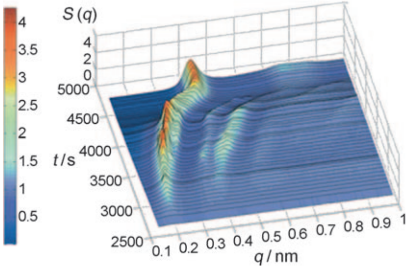
Cassette recording: A new class of coumarin–rhodamine through-bond energy-transfer (TBET) cassettes with minimal spectral overlap between the donor emission and the acceptor absorption (see picture) show large pseudo-Stokes shifts (up to 230 nm) and emission shifts (up to 170 nm). The utility of this TBET platform for TBET-based probe development was demonstrated by a new ratiometric fluorescence pH probe.



Soft Nanoparticle Crystals

W. L. Cheng, M. R. Hartman, D.-M. Smilgies, R. Long, M. J. Campolongo, R. Li, K. Sekar, C.-Y. Hui, D. Luo* ______ **380–384**

Probing in Real Time the Soft Crystallization of DNA-Capped Nanoparticles



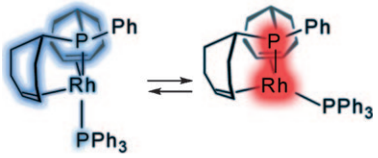
The crystallization events occurring in a drying droplet containing DNA-capped nanoparticles were probed by means of small-angle X-ray scattering (SAXS; the picture shows a series of 1D SAXS patterns recorded over time). Unusual nanoparticle supracrystals form that are both soft and elastic with continuously scalable crystalline states over a wide range of lattice constants.

Electronic Structure

F. F. Puschmann, J. Harmer, D. Stein, H. Rüegger, B. de Bruin,* H. Grützmacher* ______ **385–389**

Electromeric Rhodium Radical Complexes

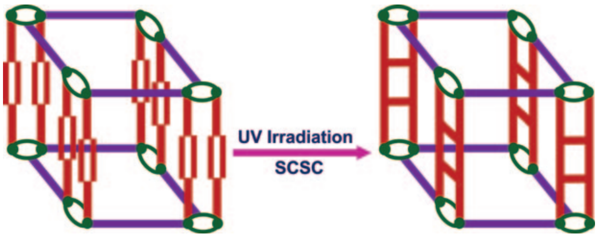
Radical changes: One single P-Rh-P angle determines whether the odd electron in the paramagnetic complex [Rh(trop$_2$PPh)(PPh$_3$)] is delocalized over the whole molecule (see picture, blue) or is localized on the P–Rh unit (red). The two energetically almost degenerate electromers exist in a fast equilibrium, and the "red" complex has the highest spin density at Rh of all low valent Rh complexes observed to date.



Photoreactive MOFs

M. H. Mir, L. L. Koh, G. K. Tan,
J. J. Vittal* ―――――― 390–393

Single-Crystal to Single-Crystal Photochemical Structural Transformations of Interpenetrated 3 D Coordination Polymers by [2+2] Cycloaddition Reactions

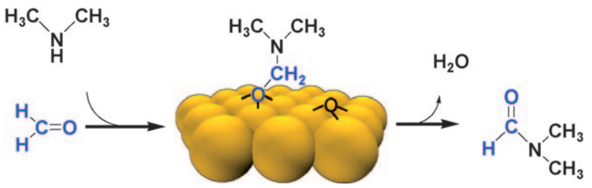


Photosensitive pillars: Perfectly aligned $C{=}C$ bonds in *trans*-1,2-bis(4-pyridyl)-ethene ligand pairs undergo 100% photochemical [2+2] cycloaddition reactions in interpenetrated 3D coordination polymers, accompanied by single-crystal to single-crystal (SCSC) transformation (see scheme).

Gold Catalysis

B. Xu, L. Zhou, R. J. Madix,
C. M. Friend* ―――――― 394–398

Highly Selective Acylation of Dimethylamine Mediated by Oxygen Atoms on Metallic Gold Surfaces



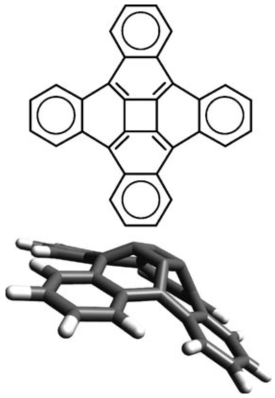
Completely coupled: The acylation of dimethylamine through coupling to formaldehyde occurs with almost 100% selectivity at low coverage of adsorbed O atoms on metallic gold and with a low activation energy. Oxygen creates an active intermediate $(CH_3)_2N_{(a)}$, which attacks the carbonyl carbon of the aldehyde (see picture). A general mechanistic framework for efficient and selective acylation of amines promoted by Au is established.

Small Circulenes

Bharat, R. Bhola, T. Bally, A. Valente,
M. K. Cyrański, Ł. Dobrzycki, S. M. Spain,
P. Rempała, M. R. Chin,
B. T. King* ―――――― 399–402

Quadrannulene: A Nonclassical Fullerene Fragment

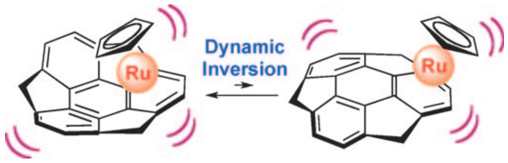
Approaching the curve: A [4]circulene (see picture) has been prepared in five steps. Although the [4]circulene motif has never before been observed, these findings suggest that the square may join the pentagon as a motif for viable fullerenes.



π-Bowl Complexes

T. Amaya, W.-Z. Wang, H. Sakane,
T. Moriuchi, T. Hirao* ―――――― 403–406

A Dynamically Inverting π-Bowl Complex

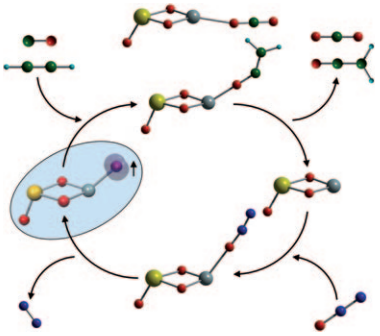


The super bowl: The dynamic inversion behaviour of $[CpRu(\eta^6\text{-sumanene})]PF_6$ (see picture; sumanene $= C_{21}H_{12}$) was investigated in solution by NMR studies. This result, the first elucidation of bowl-to-bowl inversion in π-bowl transition metal complexes, is solvent- and temperature-dependent.

Radical Clusters

M. Nößler, R. Mitrić,
V. Bonačić-Koutecký,* G. E. Johnson,
E. C. Tyo, A. W. Castleman, Jr.* **407 – 410**

Generation of Oxygen Radical Centers in Binary Neutral Metal Oxide Clusters for Catalytic Oxidation Reactions

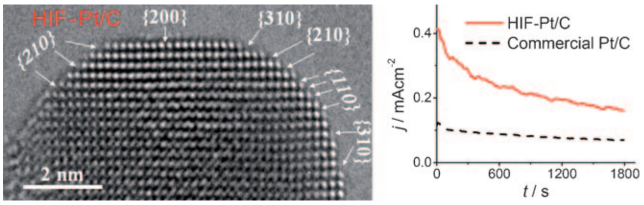


One more, one less: Radical centers are generated in binary neutral zirconium oxide clusters by replacing one Zr atom in single-metal cationic and anionic clusters with radical oxygen centers by an atom that has one less (Sc) or one more electron (Nb). This study reveals that these isoelectronic systems promote similar oxidation reactions and can undergo full catalytic cycles (see picture; Zr yellow, Sc gray, O red, C green, H teal, N blue).

Electrooxidation Catalysts

Z. Y. Zhou, Z. Z. Huang, D. J. Chen,
Q. Wang, N. Tian, S. G. Sun* **411 – 414**

High-Index Faceted Platinum Nanocrystals Supported on Carbon Black as Highly Efficient Catalysts for Ethanol Electrooxidation

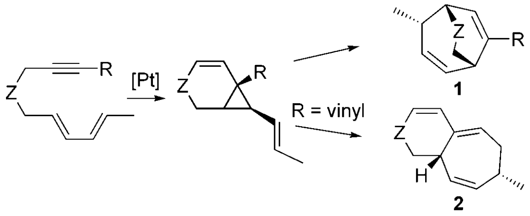


Platinum nanocrystals on carbon black were synthesized by an electrochemical square-wave potential method (HIF-Pt/C, see picture). The nanocrystals have high-index facets and a high density of atomic steps. Thanks to this high density, the catalysts exhibit at least twice the activity and selectivity of commercial Pt/C catalysts for ethanol electrooxidation into CO_2.

Tandem Reactions

S. Y. Kim, Y. Park, Y. K. Chung* **415 – 418**

Sequential Platinum-Catalyzed Cycloisomerization and Cope Rearrangement of Dienynes

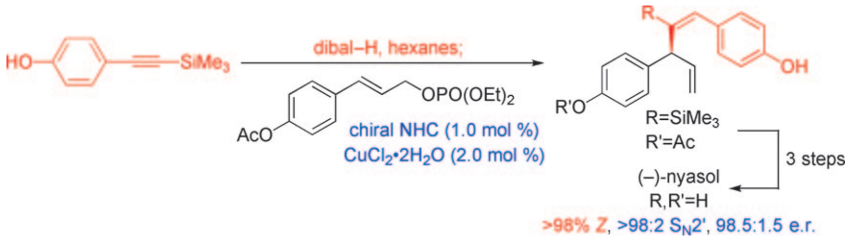


Enyne-enyne-o: Tandem cycloisomerization of dienynes catalyzed by platinum dichloride and subsequent Cope rearrangement affords bicyclic rings (see scheme; Z = NTs, O, CH_2), such as bicyclo[3.2.2]nonadienes **1** or tetrahydrocyclohepta[*c*]pyrans and -pyridines **2**, in high yields.

Enantioselective Synthesis

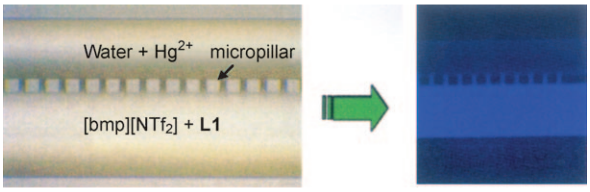
K. Akiyama, F. Gao,
A. H. Hoveyda* **419 – 423**

Stereoisomerically Pure Trisubstituted Vinylaluminum Reagents and their Utility in Copper-Catalyzed Enantioselective Synthesis of 1,4-Dienes Containing *Z* or *E* Alkenes



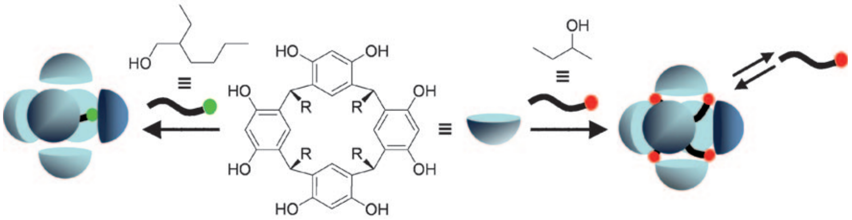
The desired isomer of a chiral 1,4-diene containing an *E* or *Z* double bond can be accessed readily by a regio- and stereoselective hydroalumination of silyl-substituted alkynes and subsequent enantioselective copper-catalyzed allylic alkylation. The utility of the procedure was demonstrated through the synthesis of (−)-nyasol (see scheme). dibal-H = diisobutylaluminum hydride, NHC = N-heterocyclic carbene.

Analytical Microsystems



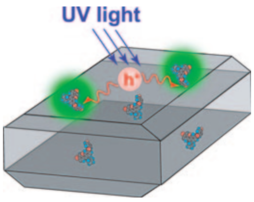
Quick test for quicksilver: A microdevice for liquid–liquid extraction and sensing of mercury ions in water uses an ionic liquid containing a task-specific ionic fluorogenic salt (**L1**) as the extracting liquid phase. The “off–on” sensing response decreases the likelihood of false positives and allows detection of Hg^{2+} in aqueous solution down to 50 ppb.

Molecular Capsules



Knock, knock! Despite the similar high-field ^1H NMR spectra of different alcohols in the presence of hexameric capsules of resorcin[4]arenes, diffusion NMR spectroscopy showed that only some of these alcohols are truly encapsulated in the hexameric capsules, whereas other alcohols are part of the hexameric structure of the capsules (see schematic representation; $R = C_{11}H_{23}$).

Photocatalysis



Proper lighting: Investigation of photocatalytic oxidation by in situ fluorescence imaging with a redox-responsive fluorescent dye (see picture, green) shows that acid treatment of single titanosilicate crystals significantly increases both the adsorption and reaction efficiencies and the heterogeneity of photocatalytic activity among crystals. Furthermore, crystal defects serve as reactive sites during the photocatalytic reaction.

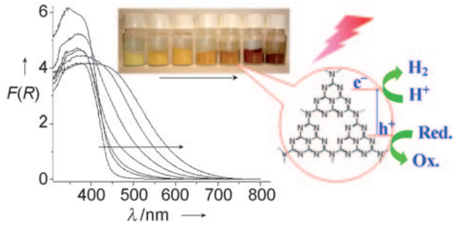
Aerobic Oxidation

Swapping places: The use of carbon to replace transition metals in the selective aerobic oxidation of benzylic alcohols has been demonstrated to be feasible. The HNO_3 additive is activated by the carbon catalyst NSC, which serves as the primary oxidant for alcohol oxidations and is regenerated by O_2. NSC = nanoshell carbon.

Photocatalysis

J. Zhang, X. Chen, K. Takanabe, K. Maeda, K. Domen,* J. D. Epping, X. Fu, M. Antonietti, X. Wang* ______ **441 – 444**

Synthesis of a Carbon Nitride Structure for Visible-Light Catalysis by Copolymerization



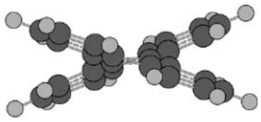
Just like a melon: Carbon nitride photocatalysts (see formula) structurally akin to poly(aminoimino)heptazine (Liebig's melon) can be prepared by direct copolymerization of dicyandiamide with barbituric acid (BA). The picture shows how increasing the amount of BA in the copolymerization mixture (arrows) extends the optical absorption of the products further into the visible region, as is favorable for solar-energy applications.

Liquid Crystals

H. Nagayama, S. K. Varshney, M. Goto, F. Araoka, K. Ishikawa, V. Prasad, H. Takezoe* ______ **445 – 448**

Spontaneous Deracemization of Disc-like Molecules in the Columnar Phase

Chiral discs: Seemingly flat disc-like molecules form a columnar phase and spontaneously segregate into macroscopic chiral domains (deracemization). Quantum chemical calculations suggest that these molecules have a twisted axially chiral core (see picture). Their aggregation into chiral superstructures is evidenced by electronic and vibrational circular dichroism spectra.

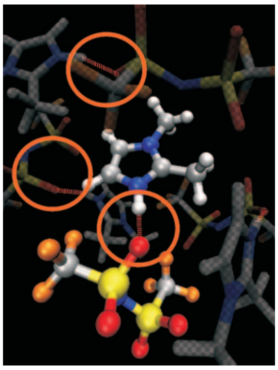


Ionic Liquids

A. Wulf, K. Fumino, R. Ludwig* ______ **449 – 453**

Spectroscopic Evidence for an Enhanced Anion–Cation Interaction from Hydrogen Bonding in Pure Imidazolium Ionic Liquids

Building bridges: Increasing capabilities of hydrogen bond formation in a set of imidazolium ionic liquids allow the direct observation of hydrogen bonding (circled in picture; C gray, H white, F orange, N blue, O red, S yellow) by far-IR spectroscopy. Frequency shifts are assigned to the ability and strength of H-bond formation. Reduced mass effects are negligible and the interactions are local and directional.

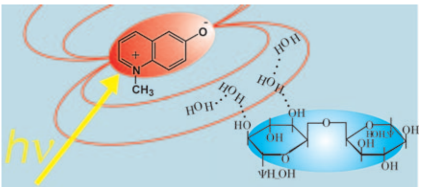


Spectroscopic Methods

M. Sajadi, Y. Ajaj, I. Ioffe, H. Weingärtner,* N. P. Ernsting* ______ **454 – 457**

Terahertz Absorption Spectroscopy of a Liquid Using a Polarity Probe: A Case Study of Trehalose/Water Mixtures

The fluorescence relaxation of *N*-methyl-6-quinolone was used to determine the frequency-dependent permittivity of the molecular environment over a large spectral range (see picture), for example in trehalose–water solution. Local THz-FIR spectroscopy can be realized by covalent attachment of the probe molecule to a supramolecular structure.



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).

Service

Corrigendum

In the Supporting Information for this Communication there was some ambiguity in the way that two ^{1}H NMR spectra were assigned. Upon inspection, the authors noticed that two peaks marked as l and m for Poly(GEMA)$_{RAFT}$ and P2 were wrongly labeled. Therefore, these spectra have been now been correctly assigned and are available online.

The authors apologize for this oversight and emphasize that the changes have no impact on the identity and behavior of the polymers.

Sweet Talking Double Hydrophilic Block Copolymer Vesicles

G. Pasparakis,
C. Alexander* **4847–4850**

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