VIP *The following Communications have been judged by at least two referees to be "very important papers" and will be published online at www.angewandte.org soon:*

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

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

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

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

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

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

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

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

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

*Q. Liu, G. Li, J. He, J. Liu, P. Li, A. Lei**
Palladium-Catalyzed Aerobic Oxidation and Carbonylation of Arylboronate Esters under Mild Conditions

A. C. Filippou, O. Chernov, K. W. Stumpf, G. Schnakenburg*
Metal–Silicon Triple Bonds: The Molybdenum Silylidyne Complex $[Cp(CO)_2Mo{=}SiR]$

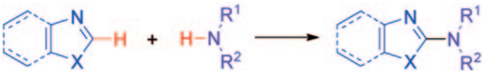
Author Profile

"The biggest problem that chemists face is increasing the signal to noise ratio.
My favorite piece of research is our preparation of dialane in solid hydrogen …"
This and more about Lester Andrews can be found on page 2278.

Books

Highlights

C–H Functionalization



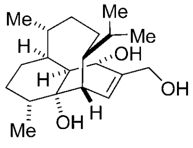
Three amigos: Direct oxidative amination of the C2-position of azoles enables the synthesis of important classes of heterocycle in an efficient and atom-economic manner (see scheme). Recent research has uncovered three distinct approaches to this transformation.

Contents

Total Synthesis

J.-Y. Lu, D. G. Hall* ________ **2286 – 2288**

Fragmentation Enables Complexity in the First Total Synthesis of Vinigrol



Vinigrol

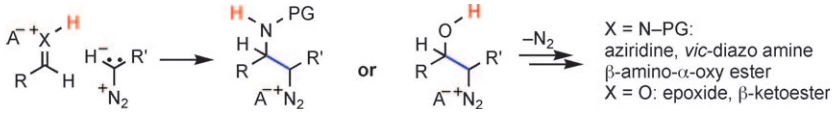
Vinigrol at last: A unique tricyclic framework and eight contiguous stereogenic centers have made vinigrol a challenging but highly prized target for the synthetic community. The first total synthesis of vinigrol was recently completed by Baran and co-workers using an intramolecular Diels–Alder reaction, a Grob fragmentation, and a series of creative endgame functionalization reactions.

Minireviews

Brønsted Acid Catalysis

J. N. Johnston,* H. Muchalski, T. L. Troyer ________ **2290 – 2298**

To Protonate or Alkylate? Stereoselective Brønsted Acid Catalysis of C–C Bond Formation Using Diazoalkanes



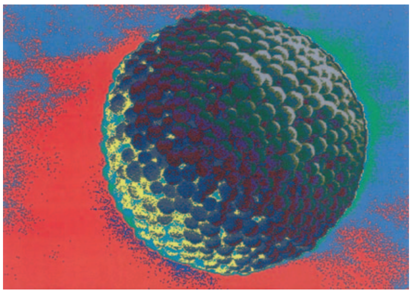
Brønsted acids, both achiral and chiral, have been used to promote the formation of carbon–carbon and carbon–heteroatom bonds with a growing number of diazoalkane derivatives (see scheme). These transformations are remarkable owing to their ability to skirt competitive diazo protonation, a reaction that has long been used to prepare esters efficiently and cleanly from carboxylic acids.

Reviews

Liquid Chromatography

K. K. Unger,* R. Ditz, E. Machtejevas, R. Skudas ________ **2300 – 2312**

Liquid Chromatography—Its Development and Key Role in Life Science Applications



For large and small molecules: Liquid chromatography is one of the most versatile technologies in the life sciences. It can be used for femtomol level analytics as well as for the ton-scale purification of drugs, and it offers a solution to abundance problems in the investigation and purification of biological systems. The picture shows a porous microparticle for liquid chromatography made by controlled agglomeration of nonporous nanoparticles.

For the USA and Canada:
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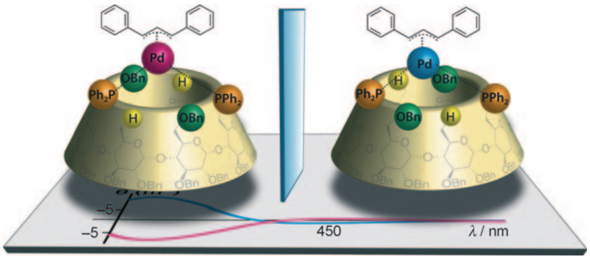
Communications

Inherent Chirality

S. Guieu, E. Zaborova, Y. Blériot, G. Poli, A. Jutand, D. Madec, G. Prestat, M. Sollogoub* ______ **2314 – 2318**

Can Hetero-Polysubstituted Cyclodextrins be Considered as Inherently Chiral Concave Molecules?

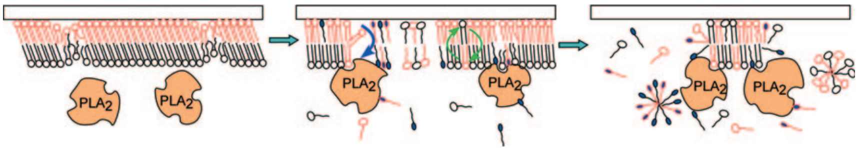


Learning curve: Regioselective twofold deprotection of benzylated cyclodextrins with diisobutylaluminum hydride affords products that can behave as enantiomers. For example, they can act as ligands for enantioselective Pd^0-catalyzed reactions and their complexes display opposite circular dichroism signals (see picture). They can thus be seen as being inherently chiral cycle surrogates.

Lipids

Y. Tong, N. Li, H. Liu, A. Ge, M. Osawa, S. Ye* ______ **2319 – 2323**

Mechanistic Studies by Sum-Frequency Generation Spectroscopy: Hydrolysis of a Supported Phospholipid Bilayer by Phospholipase A$_2$



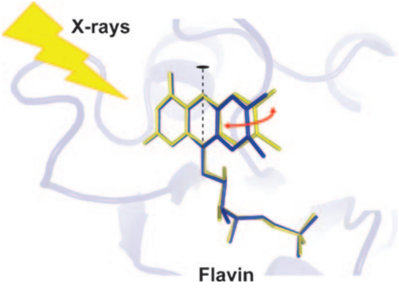
Sum mothers do 'ave 'em: Sum-frequency generation spectroscopy was used to investigate the hydrolysis mechanism of a dipalmitoylphosphatidylcholine bilayer by phospholipase A$_2$ (PLA$_2$). This study describes the structural changes and the hydrolysis mechanism in each leaflet (red and black) of a supported lipid bilayer at a molecular level.

Conformation Analysis

Å. K. Røhr, H.-P. Hersleth, K. K. Andersson* ______ **2324 – 2327**

Tracking Flavin Conformations in Protein Crystal Structures with Raman Spectroscopy and QM/MM Calculations

Damaged goods? Detailed knowledge of the cofactor conformation is essential for the functional analysis of flavoenzyme crystal structures. However, photoelectrons generated by X-rays during crystal-data collection can reduce the flavin cofactor and thus change its geometry (see picture). Monitoring of the flavin vibrational modes by Raman spectroscopy during X-ray crystal-data collection provided important information on the actual flavin state.

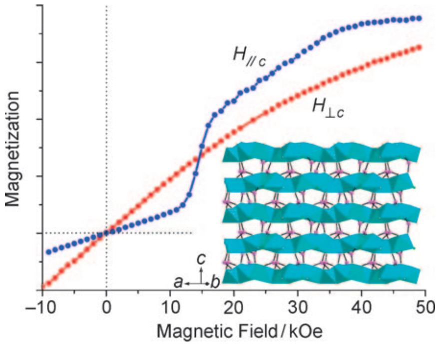


Magnetic Anisotropy

H. Xing, W. Yang, T. Su, Y. Li, J. Xu, T. Nakano,* J. Yu,* R. Xu ____ **2328 – 2331**

Ionothermal Synthesis of Extra-Large-Pore Open-Framework Nickel Phosphite $5\,H_3O\cdot[Ni_8(HPO_3)_9Cl_3]\cdot 1.5\,H_2O$: Magnetic Anisotropy of the Antiferromagnetism

Microporous magnets: The structure (inset) of the title compound features two-dimensional nickel–oxygen/chloride 18-ring layers, which are stacked along the *c* axis and pillared by HPO$_3$ units. Magnetic anisotropy measurements (see picture) on aligned single crystals indicate that the magnetic easy axis is perpendicular to the layer plane.



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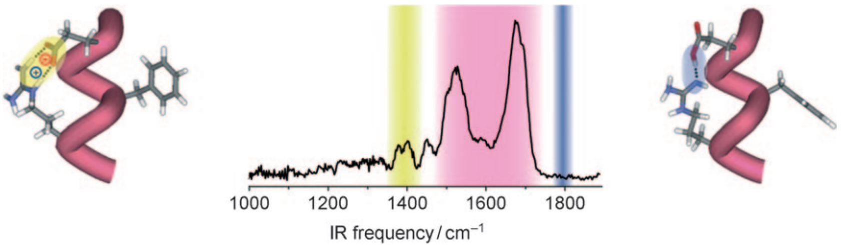
Neutral Zwitterions

A. M. Rijs,* G. Ohanessian,* J. Oomens, G. Meijer, G. von Helden, I. Compagnon* ——— **2332 – 2335**

Internal Proton Transfer Leading to Stable Zwitterionic Structures in a Neutral Isolated Peptide

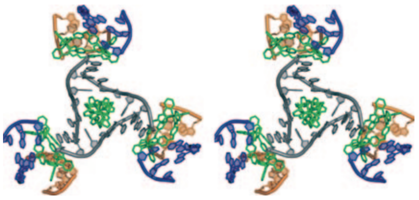


No solution for the zwitterion: An acid- and base-containing pentapeptide was designed to explore the possibility of zwitterion formation in the gas phase, in the absence of a net charge. Internal proton transfer between peptide side chains in vacuo gave a zwitterion (highlighted in yellow; the canonical form is highlighted in blue), which was identified by gas-phase IR spectroscopy.

DNA Networks

D. R. Boer, J. M. C. A. Kerckhoffs, Y. Parajo, M. Pascu, I. Usón, P. Lincoln, M. J. Hannon,* M. Coll* ——— **2336 – 2339**

Self-Assembly of Functionalizable Two-Component 3D DNA Arrays through the Induced Formation of DNA Three-Way-Junction Branch Points by Supramolecular Cylinders

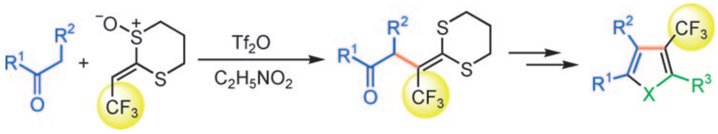


Bringing all the pieces together: By arranging the blunt-end duplex DNA strands of "continuous" three-way junctions around themselves through noncovalent interactions and inducing the formation of local "discontinuous" three-way junctions, supramolecular cylinders promote the self-assembly of 3D DNA networks with tunable physical and chemical properties (see the arrangement of the junctions in the portion of the lattice structure shown).

Synthetic Methods

T. Kobatake, S. Yoshida, H. Yorimitsu,* K. Oshima* ——— **2340 – 2343**

Reaction of 2-(2,2,2-Trifluoroethylidene)-1,3-dithiane 1-Oxide with Ketones under Pummerer Conditions and Its Application to the Synthesis of 3-Trifluoromethyl-Substituted Five-Membered Heteroarenes

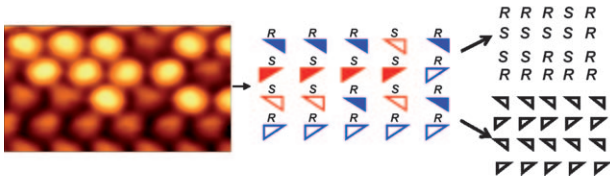


Easy as pie: With the aid of triflic anhydride, the title reaction resulted in nucleophilic attack of the carbonyl oxygen atom onto the activated cationic sulfur center and subsequent [3,3]-sigmatropic rearrangement (see scheme). The products are precursors of the difficult-to-synthesize five-membered 3-trifluoromethyl heteroarene compounds.

Chirality

M. Forster, M. S. Dyer, M. Persson, R. Raval* ——— **2344 – 2348**

2D Random Organization of Racemic Amino Acid Monolayers Driven by Nanoscale Adsorption Footprints: Proline on Cu(110)



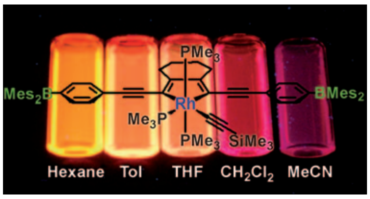
Amino acids find their feet: Scanning tunneling microscopy of racemic (*R*,*S*)-proline on Cu(110) reveals rows of random chiral amino acid sequences, showing that organization is not governed by molecular chirality. Instead, the system is dictated by a strict heterochiral adsorption-footprint template, in which each adsorption position can be occupied by either enantiomer (see picture), resulting in a random solid solution in 2D.

Photophysics

A. Steffen, M. G. Tay, A. S. Batsanov, J. A. K. Howard, A. Beeby,* K. Q. Vuong, X.-Z. Sun, M. W. George,* T. B. Marder* ______ **2349 – 2353**

2,5-Bis(*p*-R-arylethynyl)rhodacyclopentadienes Show Intense Fluorescence: Denying the Presence of a Heavy Atom

Heavy-metal light show: Photophysical studies show unprecedented high fluorescence quantum yields (Φ_f up to 69%, $\tau_f \approx 1$–3 ns) and unexpectedly slow intersystem crossing for a series of rhodium complexes. This new class of compounds challenges our understanding of the behavior of excited electronic states and the role of the heavy atom in intersystem crossing processes. Mes = mesityl, THF = tetrahydrofuran, Tol = toluene.

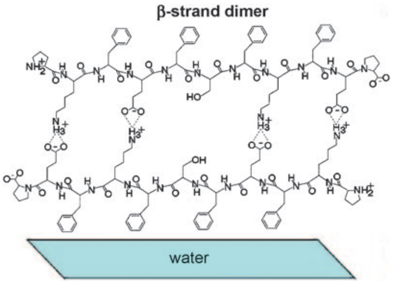


Peptide Films

A. Birman, K. Kjaer, Y. Prior, I. Nevo,* L. Leiserowitz* ______ **2354 – 2357**

Laser-Induced Alignment of Self-Assembled Films of an Oligopeptide β Sheet on the Water Surface

A pulsed IR laser beam was used to align a peptide at the air–water interface. This peptide was designed to form a cyclic β-strand dimer through Glu–Lys interactions in solution, which, when spread onto water, yielded a self-assembled β-sheet bilayer (see picture) following solvent evaporation. During this process illumination with linearly polarized laser light induced formation of an aligned crystalline film, whereas circular polarization did not.

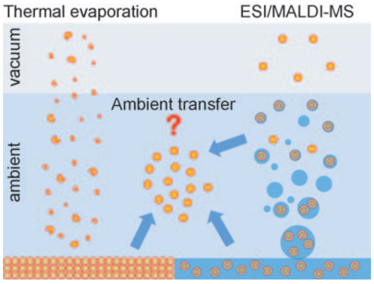


Spectroscopy

K. Chingin, V. Frankevich, R. M. Balabin, K. Barylyuk, H. Chen, R. Wang, R. Zenobi* ______ **2358 – 2361**

Direct Access to Isolated Biomolecules under Ambient Conditions

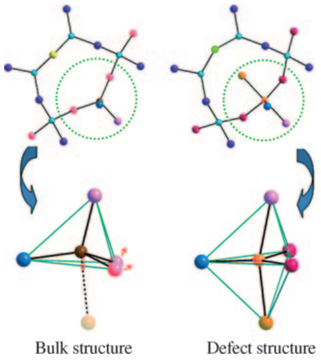
In the transfer of nonvolatiles into the gas phase, thermal evaporation is extremely inefficient because of the rapid chemical degradation (left). Generally, nonvolatiles can be brought into the gas phase by soft-ionization methods such as ESI or MALDI for investigation by MS (right). Now a method has been developed to access isolated biomolecules in the gas phase under ambient conditions (center).



Oxide Ion Conductors

M. R. Li, X. Kuang, S. Y. Chong, Z. Xu, C. I. Thomas, H. J. Niu, J. B. Claridge,* M. J. Rosseinsky* ______ **2362 – 2366**

Interstitial Oxide Ion Order and Conductivity in $La_{1.64}Ca_{0.36}Ga_3O_{7.32}$ Melilite

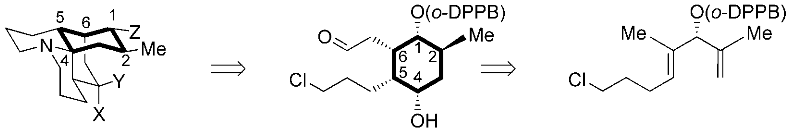


The spatial ordering of interstitial oxide charge carriers in $La_{1.64}Ca_{0.36}Ga_3O_{7.32}$ melilite reduces the ion conductivity. The complex ordering pattern is produced by the accommodation of local structural relaxation around the interstitial oxides, which extends beyond the simple formation of GaO_5 trigonal bipyramids from GaO_4 tetrahedra (see picture; brown, orange, and teal are Ga, all others O) in disordered structures at low interstitial concentrations.

Natural Product Synthesis

K. M. Laemmerhold,
B. Breit* ______ 2367 – 2370

Total Synthesis of (+)-Clavolonine, (−)-Deacetylfawcettiine, and (+)-Acetylfawcettiine



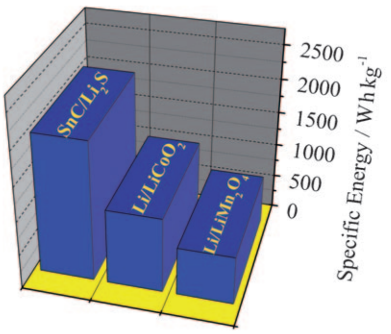
(+)-clavolonine (**1**): X,Y = O, Z = OH
(−)-deacetylfawcettiine (**2**): X = H, Y = OH, Z = OH
(+)-acetylfawcettiine (**3**): X = H, Y = OAc, Z = OAc

Three lycopodium alkaloids have been prepared by using a new strategy for constructing the central cyclohexane core (see scheme; *o*-DPPB = *ortho*-diphenylphosphanyl benzoyl). Thus stereoselective synthesis relying on substrate control was implemented; the key step involved a sequence of a (*o*-DPPB)-directed hydroformylation/carbonylene reaction and (*o*-DPPB)-directed hydroformylation.

Lithium Ion Batteries

J. Hassoun, B. Scrosati* ______ 2371 – 2374

A High-Performance Polymer Tin Sulfur Lithium Ion Battery

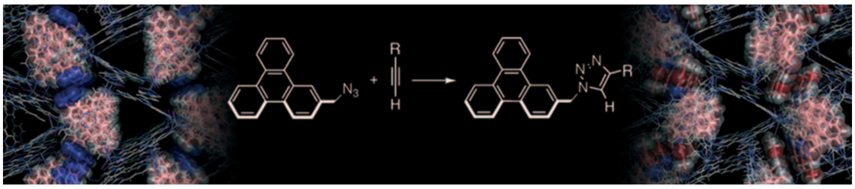


Above and beyond: A tin sulfur lithium ion battery has an energy density value of the order of 1000 Wh kg^{-1}, which is five times higher than those of conventional batteries that containing intercalated materials (see picture). Replacement of the conventional liquid electrolyte with a polymer membrane brings improvements in the fabrication and safety of the battery.

Porous Networks

T. Kawamichi, Y. Inokuma, M. Kawano,
M. Fujita* ______ 2375 – 2377

Regioselecitive Huisgen Cycloaddition within Porous Coordination Networks

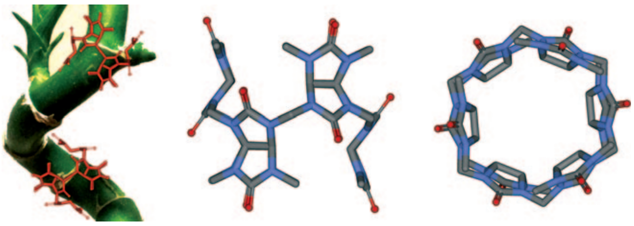


Pore it on: Huisgen 1,3-dipolar cycloaddition reactions of 2-(azidomethyl)triphenylene and alkynes (see scheme) took place within the pores of a porous coordination network in a single-crystal-to-single-crystal fashion. Columnar π stacks and nanosized pores in the network complex enforce a particular orientation of the reactants such that the 1,4-substituted isomer of the 1,2,3-triazole product is selectively produced.

Anion Receptors

J. Svec, M. Necas,
V. Sindelar* ______ 2378 – 2381

Bambus[6]uril

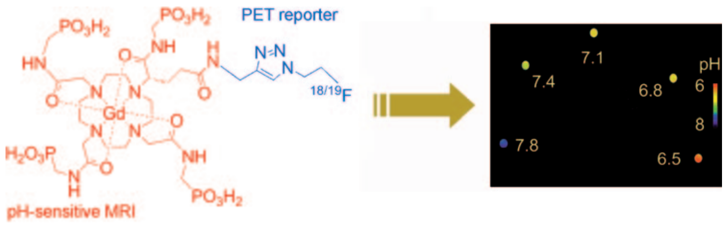


Be my guest! A macrocyclic hexamer, the structure of which is reminiscent of part of the bamboo-plant stem (see picture), was directly prepared by the condensation of a glycoluril derivative and formaldehyde. The macrocycle bound halide anions with high affinity and selectivity.

Imaging Agents

L. Frullano, C. Catana, T. Benner,
A. D. Sherry, P. Caravan* ___ **2382 – 2384**

Bimodal MR–PET Agent for Quantitative pH Imaging



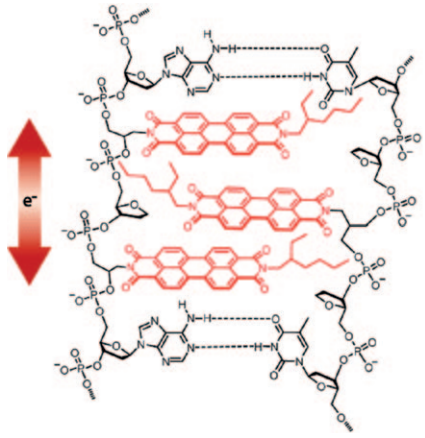
Two unknowns: The relaxivity of activatable magnetic resonance (MR) contrast agents depends on environmental factors, such as the pH value or enzymatic activity, but the MR signal depends on relaxivity and agent concentration. Simultaneous positron emission tomography (PET) and MR imaging (MRI) with the bimodal, pH-responsive MR–PET agent shown enabled the direct determination of concentration and T_1, and hence the pH value.

Electron Conduits

T. M. Wilson, T. A. Zeidan
M. Hariharan, F. D. Lewis,*
M. R. Wasielewski* ___ **2385 – 2388**

Electron Hopping among Cofacially Stacked Perylenediimides Assembled by Using DNA Hairpins

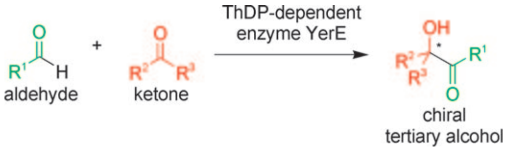
Charge on through: Perylenediimide chromophores incorporated into DNA hairpins serve as base-pair surrogates and form a zipperlike intercalated structure (see picture). Electron hopping was observed within the chromophore stacks upon one-electron reduction.



Enzyme Catalysis

P. Lehwald, M. Richter, C. Röhr, H.-w. Liu,
M. Müller* ___ **2389 – 2392**

Enantioselective Intermolecular Aldehyde–Ketone Cross-Coupling through an Enzymatic Carboligation Reaction

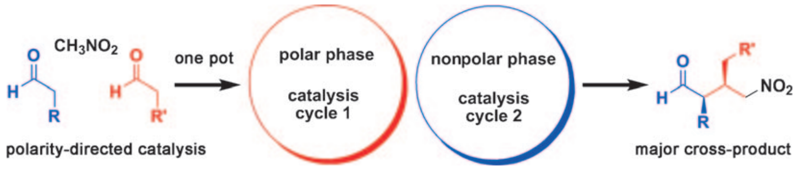


Happy new YerE: The first example of the title reaction is presented using a ThDP-dependent enzyme catalyst. The substrate tolerance of the enzyme is very broad and includes cyclic and open-chain ketones, as well as diketones and α- and β-ketoesters as acceptor substrates. The absolute configurations of two enzymatic products were determined by single-crystal structure analysis.

Organocatalysis

S. T. Scroggins, Y. Chi,
J. M. J. Fréchet* ___ **2393 – 2396**

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

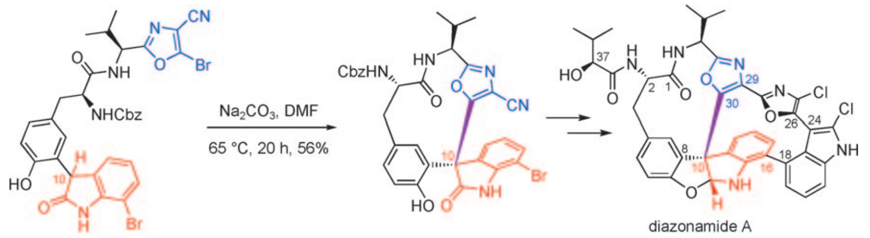


A crossing between phases: Polarity makes a difference in distinguishing substrates of otherwise nearly identical chemical reactivities. A one-pot cascade reaction involving nitromethane and two aliphatic aldehydes with similar reactivities has been developed (see scheme). The use of a biphasic reaction medium with two different organic catalysts results in the controlled incorporation of both aldehyde substrates into a single major cross-product.

Natural Products

C.-K. Mai, M. F. Sammons,
T. Sammakia* ______ **2397 – 2400**

A Concise Formal Synthesis of Diazonamide A by the Stereoselective Construction of the C10 Quaternary Center

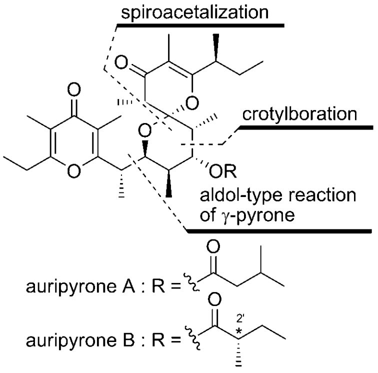


Protecting groups are overrated! A formal total synthesis of diazonamide A is described. The key step in this synthesis is an intramolecular S_NAr reaction between an oxindole and a bromooxazole. Interestingly, this reaction proceeds best using the mild base Na_2CO_3, and with no protecting groups on the oxindole nitrogen atom or phenol groups of the cyclization precursor.

Natural Product Synthesis

I. Hayakawa, T. Takemura, E. Fukasawa,
Y. Ebihara, N. Sato, T. Nakamura,
K. Suenaga, H. Kigoshi* ______ **2401 – 2405**

Total Synthesis of Auripyrones A and B and Determination of the Absolute Configuration of Auripyrone B



The winner takes it al-dol: The total synthesis of auripyrones A and B was achieved using a diastereoselective aldol-type reaction with 2,6-diethyl-3,5-dimethyl-4-pyrone as a key step. The stereostructure and absolute configuration of auripyrone B has also been determined.

O–O Activation

I. Garcia-Bosch, A. Company, J. R. Frisch,
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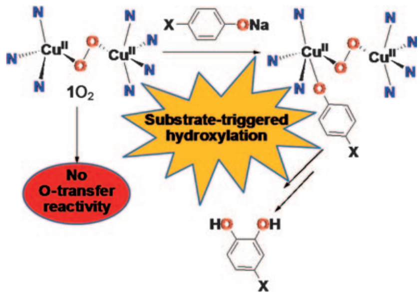
M. Costas* ______ **2406 – 2409**

O_2 Activation and Selective Phenolate *ortho* Hydroxylation by an Unsymmetric Dicopper μ-η^1:η^1-Peroxido Complex

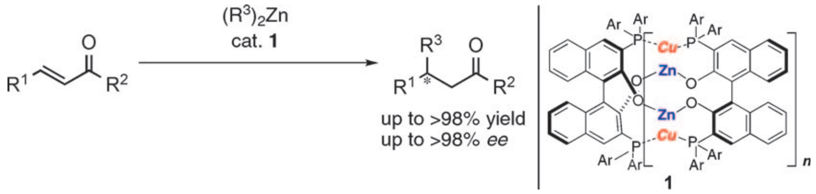


Unusual reactivity: A novel unsymmetric dicopper complex gives rise to the unsymmetric species **1**-O_2 having a μ-η^1:η^1-O_2 binding mode and reactivity patterns not previously observed for symmetric analogues. It is unreactive in oxygen atom transfer reactions, but it can selectively bind phenolate and mediate its *ortho* hydroxylation, thereby demonstrating a conceptually different tyrosinase model with exquisite selectivity.

Multinuclear Catalyst

K. Endo,* M. Ogawa,
T. Shibata* ______ **2410 – 2413**

Multinuclear Catalyst for Copper-Catalyzed Asymmetric Conjugate Addition of Organozinc Reagents



A whole lot o' metal: An efficient copper-catalyzed asymmetric conjugate addition was achieved using a binol-derived ligand. The catalytic system has a turnover number of 2000, and the excellent catalytic performance could be attributed to the generation of a multinuclear complex such as **1**. binol = 2,2′-dihydroxy-1,1′-binaphthyl.

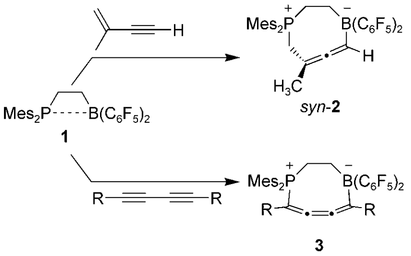
Contents

Frustrated Lewis Pairs

C. M. Mömming, G. Kehr, B. Wibbeling, R. Fröhlich, B. Schirmer, S. Grimme, G. Erker* ________ 2414 – 2417

Formation of Cyclic Allenes and Cumulenes by Cooperative Addition of Frustrated Lewis Pairs to Conjugated Enynes and Diynes

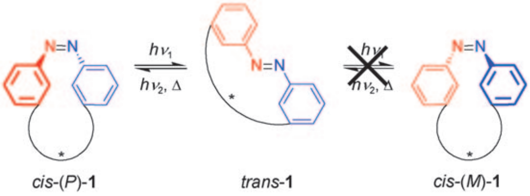
Eight, not six: The intramolecular frustrated Lewis pair **1** reacts with conjugated enynes and diynes by 1,4-addition to yield the corresponding eight-membered cyclic allenes **2** and cumulenes **3**, respectively. The thermodynamically more favorable 1,2-addition (formation of six-membered-ring products) is not observed, which indicates that the observed addition is a kinetically controlled reaction.



Chirality Switch

G. Haberhauer,* C. Kallweit 2418 – 2421

A Bridged Azobenzene Derivative as a Reversible, Light-Induced Chirality Switch

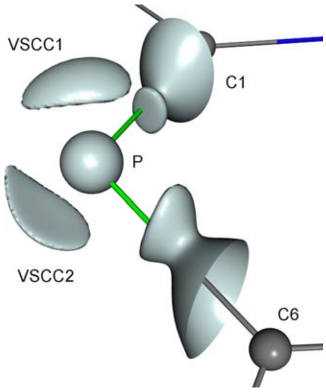


A new trick for an old dog: The azobenzene derivative **1** was used to demonstrate the light-induced spatially directed (unidirectional) *trans*→*cis* isomerization for the first time. This important effect expands the range of applications for azobenzene derivatives.

Phosphorus Lone Pairs

J. Henn, K. Meindl, A. Oechsner, G. Schwab, T. Koritsanszky, D. Stalke* ________ 2422 – 2426

Charge Density Distribution in a Metallaphosphane



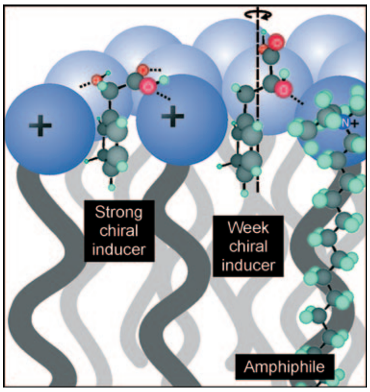
Two lone pairs: The metallaphosphane [Me$_2$Al(μ-Py)$_2$P] contains a divalent P^{III} atom. Charge density investigations reveal the presence of two lone pairs at the central phosphorus atom. This result explains why the phosphorus atom in PPy$_2$(H) can be employed in a μ-bridging fashion to two {W(CO)$_5$} moieties in the unsupported dinuclear complex [{(OC)$_5$W}$_2$(μ-P)Py$_2$(H)], and thus behaves as a four-electron donor.

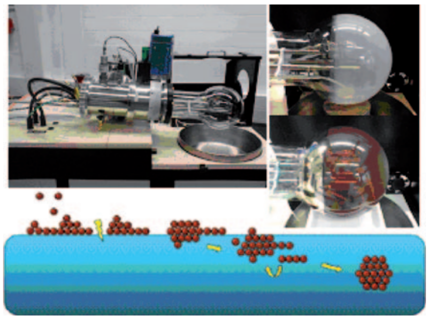
Chiral Induction

U. C. Dawin, H. Dilger, E. Roduner, R. Scheuermann, A. Stoykov, F. Giesselmann* ________ 2427 – 2430

Chiral Induction in Lyotropic Liquid Crystals: Insights into the Role of Dopant Location and Dopant Dynamics

Avoided-level-crossing muon spin resonance was applied to chiral dopants added to a nematic lyotropic liquid crystal to obtain information on the chiral induction in soft matter. The dopant solubilization at the surface of the amphiphile micelles (see model) is clarified. Reduced dopant dynamics is correlated with a strong chiral induction.





Easy peasy: Physical vapor deposition by substrate evaporation into ionic liquids is a simple, clean, and facile way to prepare metal and metal–metal oxide colloids that are stable for long periods of time. The use of ionic liquids means that the liquid does not need to be frozen, as is otherwise the case, nor are additional stabilizers necessary.

Nanoparticle Synthesis

K. Richter, A. Birkner, A.-V. Mudring* ______ **2431 – 2435**

Stabilizer-Free Metal Nanoparticles and Metal–Metal Oxide Nanocomposites with Long-Term Stability Prepared by Physical Vapor Deposition into Ionic Liquids

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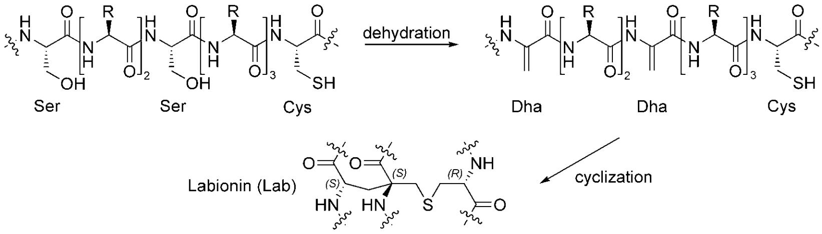

A way through the labyrinth: LabKC is an enzyme encoded in the biosynthesis gene cluster (*lab* gene cluster) of the labyrinthopeptin producer *Actinomadura namibiensis*. Some of its genes are homologous with those in other actinomycetes strains, for example, the model organism *Streptomyces coelicolor*. The functional assignment of LabKC as a kinase–cyclase suggests the formation of a new post-translational modification by a consecutive double Michael addition (see scheme; Dha = α,β-dehydroalanine).

Enzymatic C–C Bond Formation

W. M. Müller, T. Schmiederer, P. Ensle, R. D. Süssmuth* ______ **2436 – 2440**

In Vitro Biosynthesis of the Prepeptide of Type-III Lantibiotic Labyrinthopeptin A2 Including Formation of a C–C Bond as a Post-Translational Modification

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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

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

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

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