



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:

T. Mitsudome, K. Mizumoto, T. Mizugaki, K. Jitsukawa, K. Kaneda*
Wacker-Type Oxidation Using a PdCl₂-DMA Catalyst System under Copper-Free Conditions

A. Pal, M. Bérubé, D. G. Hall*
Design, Synthesis, and Screening of a Library of Peptidyl Bisboroxoles as Low Molecular Weight Receptors for Complex Oligosaccharides in Water: Identification of a Receptor for the Tumor Marker TF-Antigen

R. Kaminker, M. Lahav, L. Motiei, M. Vartanian, R. Popovitz-Biro, M. A. Iron, M. E. van der Boom*
Molecular Structure–Function Relations on the Optical Properties and Dimensions of Gold Nanoparticle Assemblies

D.-S. Ahn, S.-Y. Kim, G.-I. Lim, S. Lee,* Y. S. Choi,* S. K. Kim*
Mode-Dependent Fano Resonances in the Predissociation of Diazirine in the S₁ State

P. J. Malinowski, M. Derzsi, Z. Mazej, Z. Jagličić, B. Gawęł, W. Łasocha, W. Grochala*
Anomalously Strong Antiferromagnetism in Silver(II) Sulfate

R. C. Driesener, M. R. Challand, S. E. McGlynn, E. M. Shepard, E. S. Boyd, J. B. Broderick, J. W. Peters, P. L. Roach*
[FeFe]-Hydrogenase Cyanide Ligands Derived from S-Adenosylmethionine-Dependent Cleavage of Tyrosine

E. Kan, H. Xiang, C. Lee, F. Wu, J. Yang, M.-H. Whangbo*
Origin of the Ferroelectricity in Perovskites with s⁰ A-Site Cations: Toward Near-Room-Temperature Multiferroics

A. Ciesielski, S. Lena, S. Masiero, G. P. Spada,* P. Samorì*
Dynamers at the Solid–Liquid Interface: Controlling the Reversible Assembly/Reassembly Process Between Two Highly Ordered Supramolecular Guanine Motifs

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



“The best advice I have ever been given is “Hard work never killed anyone!”
 The worst advice I have ever been given was “Don’t go to Oxford—you’ll flounder.” ...”
 This and more about Simon Aldridge can be found on page 1008.

Author Profile

Simon Aldridge _____ 1008

The Tao of Chemistry and Life

Eugene H. Cordes

Books

reviewed by T. Kolter _____ 1009



All that glitters: Binary catalytic systems of gold(I) complexes and chiral Brønsted acids, which were recently reported by the research groups of Dixon and Gong, represent a significant leap forwards in the area of sequential one-pot reactions.

The special feature of these reactions is the isolobal analogy of the proton and [LAu]⁺ cations, which requires a delicate balance between the reactivities of the acids and the gold complexes.

Highlights

Binary Catalytic Systems

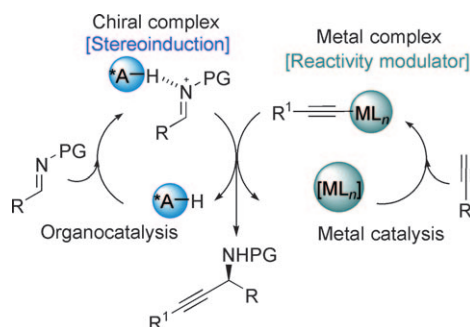
A. S. K. Hashmi,*
 C. Hubbert _____ 1010–1012

Gold and Organocatalysis Combined

Cooperative Catalysis

P. de Armas, D. Tejedor,
F. García-Tellado* — 1013 – 1016

Asymmetric Alkynylation of Imines by Cooperative Hydrogen Bonding and Metal Catalysis



Simple and practical: The asymmetric alkynylation of imines has been revolutionized with two nice examples of a novel model of H-bonding asymmetric metal catalysis with Brønsted acids. The catalytic model comprises two well-differenti-

ated and parallel catalytic cycles: one involving metallic alkynylydes (metal catalysis) and the other one using a chiral Brønsted acid (organocatalysis; see scheme; PG = protecting group).

Minireviews

N₂O Activation

W. B. Tolman* — 1018 – 1024

Binding and Activation of N₂O at Transition-Metal Centers: Recent Mechanistic Insights



No laughing matter: In view of its significance as an atmospheric pollutant and greenhouse gas, an important research goal has been to understand the reactivity of N₂O with metal centers in the gas phase, on solid supports, in enzymes, and as soluble complexes in solution. Recent studies provide provocative mechanistic insights into how N₂O might bind to metal ions and be activated for N–N or N–O bond scission or insertion into M–C or M–H bonds.

Reviews

Clinical Chemistry

J. Durner* — 1026 – 1051

Clinical Chemistry: Challenges for Analytical Chemistry and the Nanosciences from Medicine

From the drawing of blood to diagnosis: Clinical chemistry makes an important contribution to medical diagnostics and the prevention of disease. New insights from the natural sciences and information technologies can reach into medicine by way of clinical chemistry. This Review presents the historic roots of the subject as well as current methods for measurement and analysis. Research projects and future developments are also discussed.



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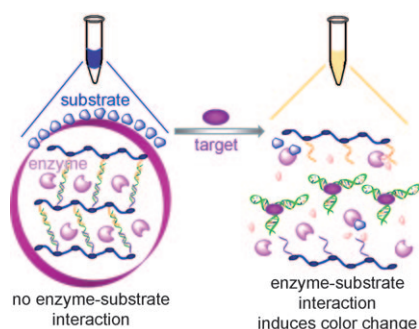
individuals who are personal members of a national chemical society prices are available on request. Postage and handling charges included. All prices are subject to local VAT/sales tax.

Communications

Sensors

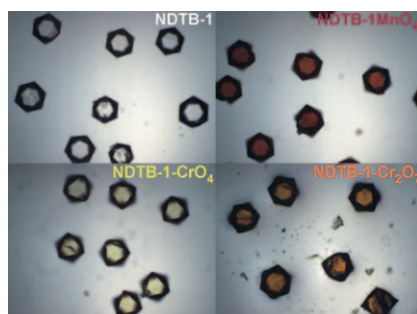
Z. Zhu, C. Wu, H. Liu, Y. Zou, X. Zhang,
H. Kang, C. J. Yang,*
W. Tan* — 1052–1056

An Aptamer Cross-Linked Hydrogel as a
Colorimetric Platform for Visual Detection



Cocaine cracked: The gel-sol transition of an enzyme-caged hydrogel has been efficiently controlled by target binding events, which trigger release of the enzyme to take part in its catalytic role for signal amplification (see picture). As low as 20 ng of cocaine can be visually detected within 10 min without any aid of sophisticated instrumentation.

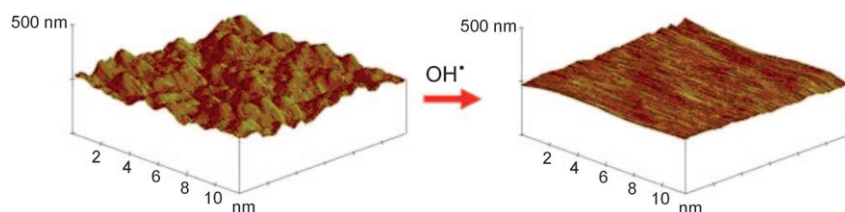
Cleaning up: A cubic thorium borate possesses a porous supertetrahedral cationic framework with extraframework borate anions. These anions are readily exchanged with a variety of environmental contaminants (see picture), especially those from the nuclear industry, including chromate and pertechnetate.



Cationic Framework Materials

S. Wang, E. V. Alekseev,* J. Diwu,
W. H. Casey, B. L. Phillips, W. Depmeier,
T. E. Albrecht-Schmitt* — 1057–1060

NDTB-1: A Supertetrahedral Cationic
Framework That Removes TcO_4^- from
Solution



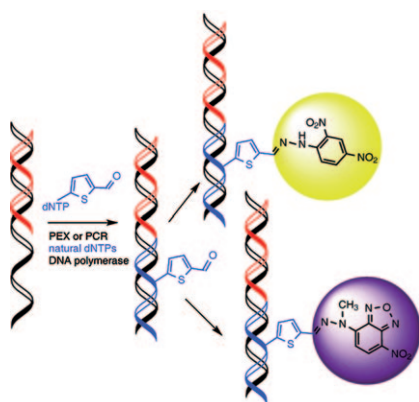
Gold service: The asperities on a polished gold surface are quickly dissolved by the OH^\bullet radicals of Fenton's reagent. The dissolution of Au is rapid at the beginning of the reaction and is negligible when the

asperities have been dissolved. Although the OH^\bullet radicals also oxidize the smooth parts of the Au surface, they do not dissolve them, but form a stable oxide monolayer.

Chemical Polishing

A. M. Nowicka,* U. Hasse, M. Hermes,
F. Scholz* — 1061–1063

Hydroxyl Radicals Attack Metallic Gold



Reactive aldehyde-modified DNA was prepared in two steps by Suzuki cross-coupling of halogenated nucleoside triphosphates (dNTPs) with 4-formylthiophene-2-boronic acid and subsequent polymerase incorporation of the modified nucleotides into DNA (see scheme; PEX = primer extension, PCR = polymerase chain reaction). Formation of hydrazones with arylhydrazines under aqueous conditions was used for DNA staining.

Functionalized DNA

V. Ráindlová, R. Pohl, M. Šanda,
M. Hocek* — 1064–1066

Direct Polymerase Synthesis of Reactive
Aldehyde-Functionalized DNA and Its
Conjugation and Staining with Hydrazines



Frontiers of Chemistry: From Molecules to Systems

A One-Day Symposium

On 21st May 2010 in Paris

at the Maison de la Chimie
(near the Eiffel Tower and Les Invalides)

Speakers



Gerhard Ertl
Nobel Prize 2007



Jean-Marie Lehn
Nobel Prize 1987



Roger Y. Tsien
Nobel Prize 2008



Ada Yonath
Nobel Prize 2009



Luisa De Cola



Alan R. Fersht



Marc Fontecave



Michael Grätzel



Michel Orrit



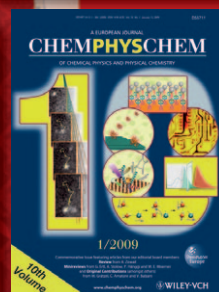
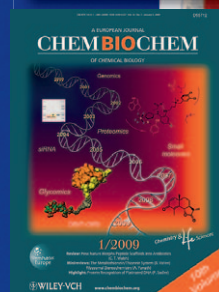
Nicolas Winssinger

Posters

will be displayed also online from 1st April.

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Celebrating 10 Years of



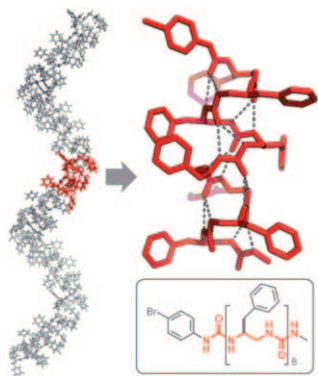
Scientific committee

E. Amouyal, M. Che,
F. C. De Schryver,
A. R. Fersht, P. Göllitz,
J. T. Hynes, J.-M. Lehn

Topics

catalysis, biochemical imaging,
chemical biology, bionanotechnology,
proteomics, spectroscopy, solar cells



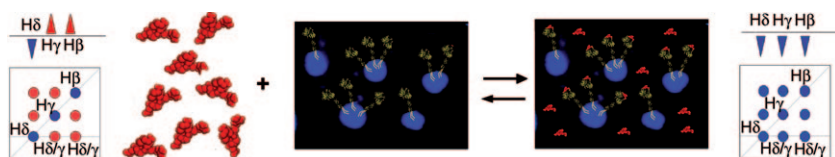


Helical by nature: Urea-based peptidomimetics with proteinogenic side chains are fully helical in the crystalline state (see picture). Four acyclic residues are sufficient to drive complete helix formation with all complementary H-bonding sites being satisfied (up to 14 for a 8-mer). Helices pack head-to-tail to create infinite H-bonded networks with different topologies.

Helical Foldamers

L. Fischer, P. Claudon, N. Pendem, E. Miclet, C. Didierjean, E. Ennifar, G. Guichard* _____ **1067–1070**

The Canonical Helix of Urea Oligomers at Atomic Resolution: Insights Into Folding-Induced Axial Organization



Cell mates: Various ligand–receptor interactions in different human cancer cell lines were probed directly by two-dimensional transferred-NOE spectroscopy (see

picture) to prove recognition specificity and determine an affinity ranking of several ligands.

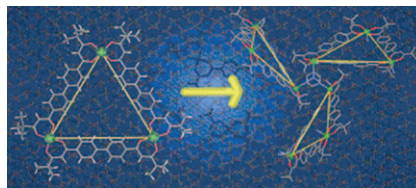
NMR Spectroscopy

S. Mari, C. Invernizzi, A. Spitaleri, L. Alberici, M. Ghitti, C. Bordignon, C. Traversari, G.-P. Rizzardi,* G. Musco* _____ **1071–1074**

2D TR-NOESY Experiments Interrogate and Rank Ligand–Receptor Interactions in Living Human Cancer Cells



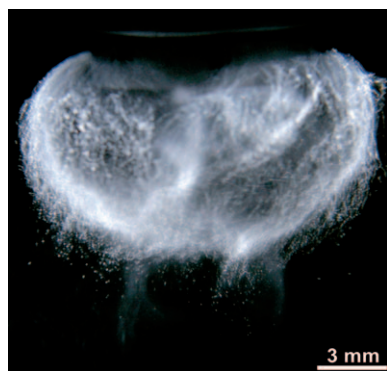
Into the void: A large, cavity-containing trinuclear triangle with accessible binding sites has been synthesized, structurally characterized, and incorporated into a chiral metal–organic framework (see picture). The resulting 3D framework, with a trifunctional amine bridging group, has a void volume of 56% that is accessible to solvent and adsorbs a variety of gases to various degrees.



Self-Assembly

J. K. Clegg, S. S. Iremonger, M. J. Hayter, P. D. Southon, R. B. Macquart, M. B. Duriska, P. Jensen, P. Turner, K. A. Jolliffe, C. J. Kepert, G. V. Meehan, L. F. Lindoy* _____ **1075–1078**

Hierarchical Self-Assembly of a Chiral Metal–Organic Framework Displaying Pronounced Porosity



Bright bubbles! Ultrabright sonoluminescence is observed from concentrated phosphoric acid (see picture). Two independent molecular emission species (excited OH[•] and PO[•]) can be observed from multibubble sonoluminescence; both of them can be used as spectroscopic thermometers to probe the extreme conditions generated during bubble collapse.

Sonoluminescence

H. Xu, N. G. Glumac, K. S. Suslick* _____ **1079–1082**

Temperature Inhomogeneity during Multibubble Sonoluminescence



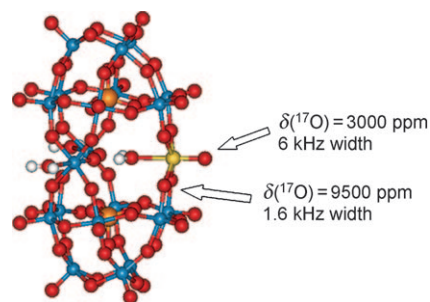
Metal Oxo Complexes

A. Bagno,* R. Bini — 1083–1086



NMR Spectra of Terminal Oxo Gold and Platinum Complexes: Relativistic DFT Predictions

Missing in action: Relativistic density-functional methods provide a reliable framework to predict the features of ^{195}Pt , ^{183}W , and ^{17}O NMR spectra of terminal oxo Pt and Au complexes (see picture for $[\text{P}_2\text{W}_{20}\text{O}_{70}\text{Au}(\text{O})(\text{OH}_2)_3]^{9-}$; Au yellow, W blue, P orange, O red, H white). The complexes have an extremely small HOMO–LUMO gap, and both orbitals are localized on an $\text{M}=\text{O}$ fragment.

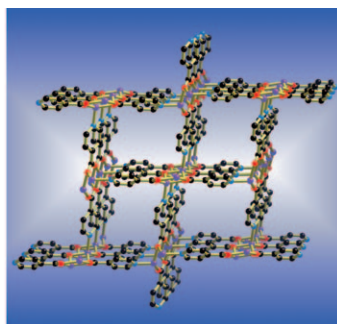
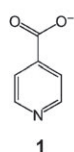


Coordination Polymers

B. F. Abrahams,* M. J. Grannas,
T. A. Hudson, R. Robson* — 1087–1089



A Simple Lithium(I) Salt with a Microporous Structure and Its Gas Sorption Properties



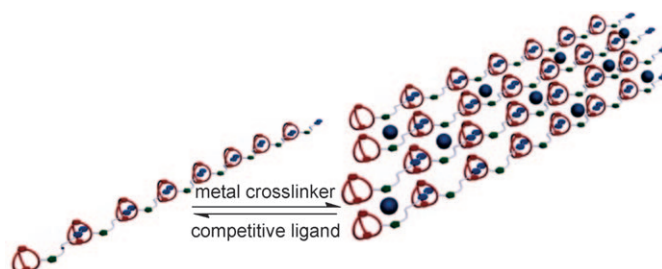
Thoroughly absorbing: The simple combination of lithium ions and isonicotinate anions (**1**) leads to the formation of a lightweight salt (see picture; C black, O red, N blue, Li purple) that is able to reversibly sorb H_2 , N_2 , CO_2 , and CH_4 . The salt exists as a 3D network of micro-channels that are occupied by solvent molecules.

Supramolecular Polymers

F. Wang, J. Zhang, X. Ding, S. Dong,
M. Liu, B. Zheng, S. Li, L. Wu, Y. Yu,
H. W. Gibson, F. Huang* — 1090–1094



Metal Coordination Mediated Reversible Conversion between Linear and Cross-Linked Supramolecular Polymers



The dynamic duo: Different topologies of dynamic supramolecular polymers, such as linear (see picture, left) and cross-linked species (right), can be reversibly

interconverted by external stimuli that utilize host–guest and metal–ligand non-covalent recognition motifs.

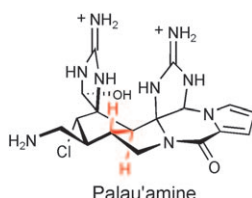


Natural Product Synthesis

I. B. Seiple, S. Su, I. S. Young, C. A. Lewis,
J. Yamaguchi, P. S. Baran* — 1095–1098

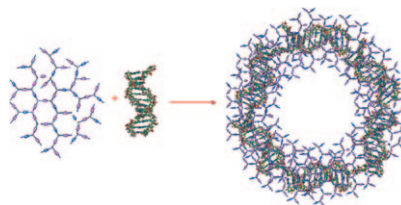


Total Synthesis of Palau'amine



Worth the wait: The long anticipated total synthesis of palau'amine has been accomplished by a route featuring highly chemoselective transformations, cascade reactions, and a remarkable transannular cyclization to secure the unprecedented *trans*-5,5 ring junction (shown in red).

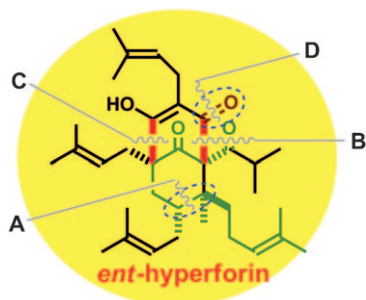
Ring around the rosy: A novel multifunctional disulfide-containing hyperbranched poly(amido amine) that is stimuli-responsive, biocompatible, biodegradable, and photoluminescent can assemble DNA into a well-defined nanoring with strong photoluminescence. The nanoring is stimuli-responsive and the ring wall is biocompatible and controllably bioreducible; it has potential applications in gene and drug delivery, and molecular imaging.



Photoluminescent Nanorings

Y. You,* Z. Yu, M. Cui,
C. Hong* 1099–1102

Preparation of Photoluminescent Nanorings with Controllable Bioreducibility and Stimuli-Responsiveness



Key to success: The first catalytic asymmetric total synthesis of *ent*-hyperforin (see picture) was accomplished by using a Diels–Alder reaction promoted by a chiral cationic iron catalyst (**A**; 96% *ee*, d.r. > 33:1), a diastereoselective Claisen rearrangement (**B**; 12:1 selectivity), an intramolecular aldol reaction (**C**), and a vinylogous Pummerer rearrangement (**D**) as key steps.

Total Synthesis

Y. Shimizu, S.-L. Shi, H. Usuda, M. Kanai,*
M. Shibasaki* 1103–1106

Catalytic Asymmetric Total Synthesis of *ent*-Hyperforin



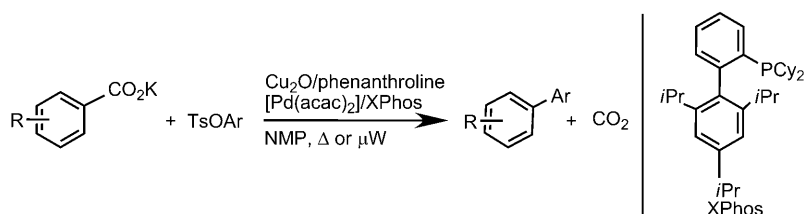
The key to motion: A second-generation molecular rotary motor, which contains a DB24C8 macrocycle ring incorporated into the lower stator half and a dialkyl ammonium ion attached to the upper rotor half, forms a [1]pseudorotaxane in

less polar solvents such as CH_2Cl_2 . In this self-complexing system, acid–base-controlled threading–dethreading movements can be utilized to unlock or lock the molecular motor (see picture).

Molecular Devices

D.-H. Qu, B. L. Feringa* 1107–1110

Controlling Molecular Rotary Motion with a Self-Complexing Lock



A bimetallic copper/palladium catalyst system is disclosed that enables the use of tosylates as carbon electrophiles in decarboxylative coupling reactions. A variety of aromatic carboxylate salts,

regardless of their substitution pattern, have been coupled with these inexpensive and readily available electrophiles to give the corresponding biaryl compounds in good yields (see scheme).

Coupling Reactions

L. J. Gooßen,* N. Rodríguez, P. P. Lange,
C. Linder 1111–1114

Decarboxylative Cross-Coupling of Aryl Tosylates with Aromatic Carboxylate Salts



Copper Catalysis

J. J. Mousseau, J. A. Bull,
A. B. Charette* 1115–1118



Copper-Catalyzed Direct Alkenylation of
N-Iminopyridinium Ylides



A versatile Cu-catalyzed direct C–H alkenylation of N-iminopyridinium ylides, compatible with several different copper sources (including a penny), provides a powerful and inexpensive method for the synthesis of functionalized pyridine

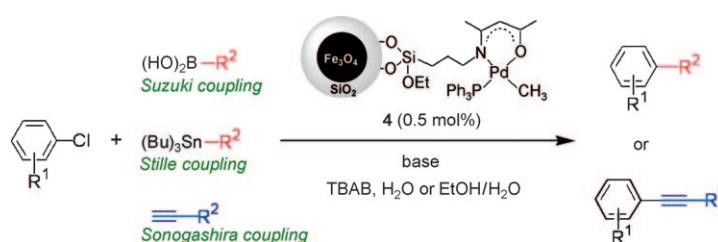
derivatives. Chemoselective functionalization of halide-containing compounds allows the synthesis of alkenyl pyridines containing reactive tethers for further functionalization.

Heterogeneous Catalysis

M.-J. Jin,* D.-H. Lee 1119–1122



A Practical Heterogeneous Catalyst for the
Suzuki, Sonogashira, and Stille Coupling
Reactions of Unreactive Aryl Chlorides

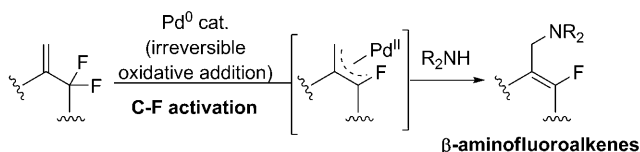


Practical catalyst: A magnetic nanoparticle-supported palladium catalyst was developed for the highly efficient heterogeneous Suzuki, Sonogashira, and Stille

couplings of a wide variety of aryl chlorides. Furthermore, the catalyst could be recycled by facile magnetic separation without any loss of activity.

Organofluorine Compounds

X. Pigeon, M. Bergeron, F. Barabé,
P. Dubé, H. N. Frost,
J.-F. Paquin* 1123–1127



Activation of Allylic C–F bonds:
Palladium-Catalyzed Allylic Amination of
3,3-Difluoropropenes

Lose one, keep one! A wide range of cyclic and acyclic β -aminofluoroalkenes are prepared by the title reaction. The key

fluorinated palladium π -allyl intermediate is generated using a catalytic allylic C–F bond activation.

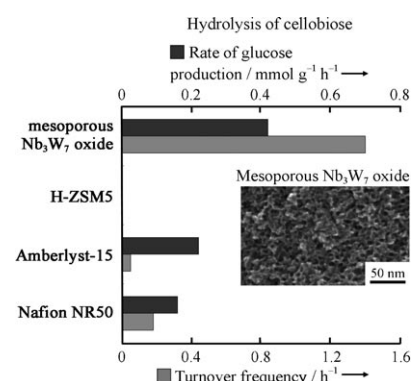
Mesoporous Solid Acid

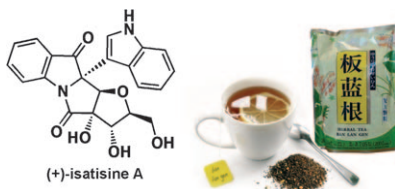
C. Tagusagawa, A. Takagaki, A. Iguchi,
K. Takanabe, J. N. Kondo, K. Ebitani,
S. Hayashi, T. Tatsumi,
K. Domen* 1128–1132



Highly Active Mesoporous Nb–W Oxide
Solid-Acid Catalyst

Pore-ing acid: Mesoporous Nb–W oxides (Nb:W = 3:7) are found to be a recyclable, highly active solid acid. They surpass ion-exchange resins (Nafion NR50 and Amberlyst-15) and zeolites (H-ZSM5 and H-Beta) in Friedel–Crafts alkylation and hydrolysis reactions (see picture). The high activity is due to strong acid sites and a mesoporous structure with a high surface area and easy reactant accessibility.





Isatis indigotica has long been used as a source of therapeutic compounds in traditional Chinese medicine (the roots are the source of Ben Lan Gen herbal tea). In 2007 the leaves of *I. indigotica* yielded isatisine A, an unusual oxidized bis-indole. A concise, enantiospecific total synthesis of isatisine A is reported using a homochiral cyclopropane diester to construct the key tetrahydrofuran ring.

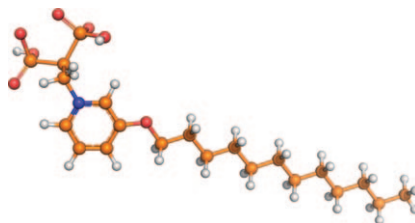
Total Synthesis

A. Karadeolian, M. A. Kerr* 1133–1135

Total Synthesis of (+)-Isatisine A



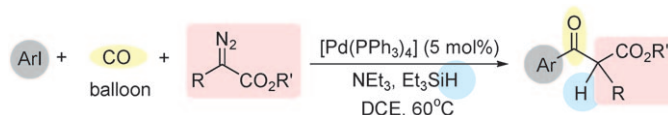
Chain gang: Lipophilic pyridinium bisphosphonates containing long alkyl chains (see picture, P red, N blue, C orange, H gray) stimulate human $\gamma\delta$ T cells expressing the V γ 2V δ 2 T cell receptor. Stimulation with such compounds is more potent than with zoledronate, which is active against breast and prostate cancer. The lipophilic bisphosphonates bind poorly to bone and are thus less likely to cause side effects associated with bisphosphonates in clinical use.



Immunotherapeutic Bisphosphonates

Y. Zhang, R. Cao, F. Yin, F. Y. Lin, H. Wang, K. Krysiak, J. H. No, D. Mukkamala, K. Houlihan, J. K. Li, C. T. Morita,* E. Oldfield* 1136–1138

Lipophilic Pyridinium Bisphosphonates: Potent $\gamma\delta$ T Cell Stimulators



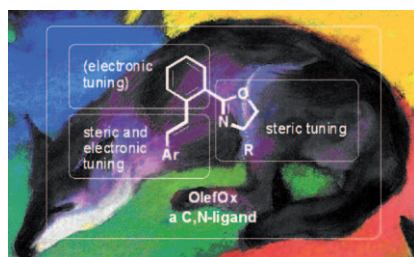
On the move: A palladium-catalyzed reaction of aryl iodides, diazo compounds or *N*-tosylhydrazones, and carbon monoxide affords β -oxo esters or ketones/

enones (see scheme; DCE = 1,2-dichloroethane). The products are delivered with high efficiency through the title sequence.

Synthetic Methods

Z. Zhang, Y. Liu, M. Gong, X. Zhao, Y. Zhang, J. Wang* 1139–1142

Palladium-Catalyzed Carbonylation/Acyl Migratory Insertion Sequence



Foxy ligands: An efficient three-step synthesis allows the new highly modular family of olefin–oxazoline ligands (OlefOx; see picture) to be exploited in asymmetric catalysis. The ease of electronic and steric variation and the successful application in the highly enantioselective rhodium-catalyzed conjugate addition of aryl boronic acids to cyclic enones demonstrate the importance of this new ligand class.

Ligand Design

B. T. Hahn, F. Tewes, R. Fröhlich, F. Glorius* 1143–1146

Olefin–Oxazolines (OlefOx): Highly Modular, Easily Tunable Ligands for Asymmetric Catalysis

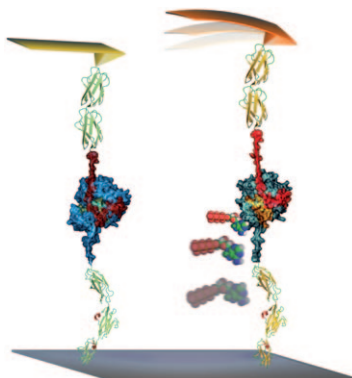


Atomic Force Microscopy

E. M. Puchner, H. E. Gaub* 1147–1150



Exploring the Conformation-Regulated Function of Titin Kinase by Mechanical Pump and Probe Experiments with Single Molecules



Feel the force: A new AFM-based single-molecule pump-and-probe protocol has been used to investigate the function of different force-induced conformations of proteins (see picture). It was found that the autoinhibited enzyme titin kinase reaches its ability to bind ATP after the first two barriers of the complex mechanical activation pathway.

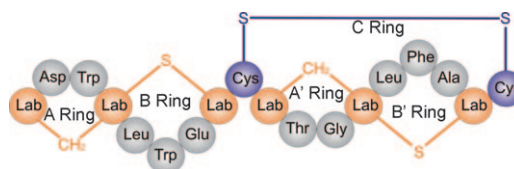


Lantibiotics

K. Meindl, T. Schmiederer, K. Schneider, A. Reicke, D. Butz, S. Keller, H. Gühring, L. Vértessy, J. Wink, H. Hoffmann, M. Brönstrup,* G. M. Sheldrick, R. D. Süßmuth* 1151–1154



Labyrinthopeptins: A New Class of Carbacyclic Lantibiotics



A-maze-ing peptides: The structure of the novel lantibiotic labyrinthopeptin A2 (see picture) contains labionin—an unprecedented carbacyclic, posttranslationally modified amino acid. The identification of the biosynthetic gene cluster of labyrinthopeptins was also successful. Labyrinthopeptin A2 revealed a pronounced in vivo efficacy (attenuation of tactile allodynia) in a mouse model of neuropathic pain.

thopectins was also successful. Labyrinthopeptin A2 revealed a pronounced in vivo efficacy (attenuation of tactile allodynia) in a mouse model of neuropathic pain.

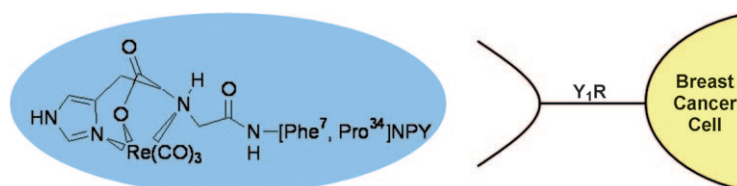


Medicinal Chemistry

I. U. Khan, D. Zwanziger, I. Böhme, M. Javed, H. Naseer, S. W. Hyder, A. G. Beck-Sickingher* 1155–1158

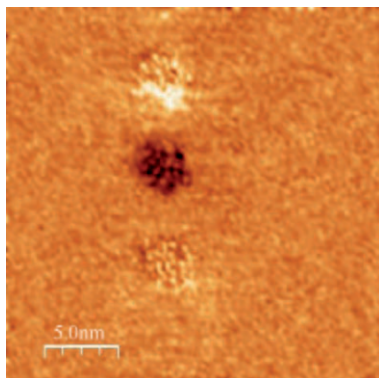


Breast-Cancer Diagnosis by Neuropeptide Y Analogues: From Synthesis to Clinical Application



Selective, sensitive, and specific breast cancer diagnosis: Neuropeptide Y (NPY) and the Y₁R-selective [Phe⁷, Pro³⁴]NPY peptide were labeled with Re/^{99m}Tc and pre-clinically characterized using competition receptor binding and signal transduction assays, and also microscopic,

metabolic-stability, rabbit body-uptake, and protein-binding studies. Selective uptake of the ^{99m}Tc(core)³⁺-(N^αHis-ac)-labeled [Phe⁷, Pro³⁴]NPY analogue in human breast-cancer patients was obtained by whole body scintimammography.



Strung out: Fe^{II} complexes containing pairs of planar terdentate N ligands form regular one-dimensional aggregates on highly oriented pyrolytic graphite, on a length scale of hundreds of nanometers. STM spectroscopy was used to probe the molecular conductance and thus the spin state. The distribution of spin states is random in the single-molecule chain, but local cooperativity sets in when the chain is made up of oligonuclear “beads” (see picture).

Spin Crossover

M. S. Alam, M. Stocker, K. Gieb, P. Müller,* M. Haryono, K. Student, A. Grohmann* _____ **1159–1163**

Spin-State Patterns in Surface-Grafted Beads of Iron(II) Complexes



Supporting information is available on www.angewandte.org (see article for access details).



A video clip is available as Supporting Information on www.angewandte.org (see article for access details).

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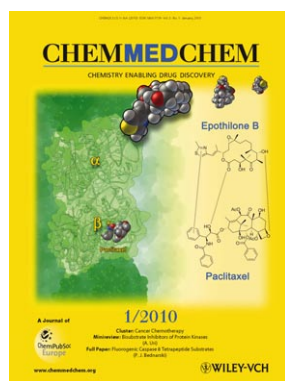
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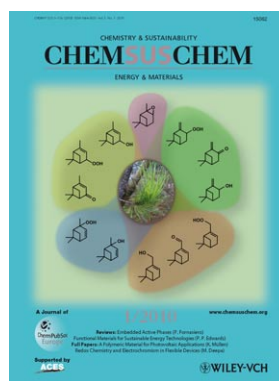
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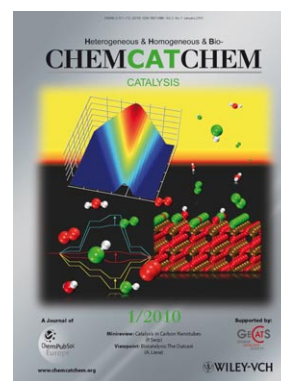
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