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:

*Z. Zhang, Z. Wang, R. Zhang, K. Ding**
Extremely Efficient Titanium Catalyst for the Enantioselective Cyanation of Aldehydes Using Cooperative Catalysis

*Qi Wang, M. Zhang, C. Chen, W. Ma, J. Zhao**
Photocatalytic Aerobic Oxidation of Alcohols on TiO_2: The Acceleration Effect of Brønsted Acids

Ye Fu, Q. Dai, W. Zhang, J. Ren, T. Pan, C. He**
AlkB Domain of Mammalian ABH8 Catalyzes Hydroxylation of 5-Methoxycarbonylmethyluridine at the Wobble Position of tRNA

*S. Rizzato, J. Bergès, S. A. Mason, A. Albinati, J. Kozelka**
Dispersion-Driven Hydrogen Bonding: Theoretically Predicted H–bond between H_2O and Platinum(II) Identified by Neutron Diffraction

*D. R. Dreyer, H. Jia, C. W. Bielawski**
Graphene Oxide: A Convenient Carbocatalyst for Facilitating Oxidation and Hydration Reactions

H. Amouri, J. Moussa, A. K. Renfrew, P. J. Dyson, M. N. Rager, L.-M. Chamoreau*
Metal Complex of Diselenobenzoquinone : Discovery, Structure, and Anticancer Activity

*M. Rauschenberg, S. Bomke, U. Karst, B. J. Ravoo**
Dynamic Peptides as Biomimetic Carbohydrate Receptors

*H. Wu, J. Ge, S. Q. Yao**
Microarray–Assisted High–Throughput Identification of a Cell–Permeable Small Molecule Binder of 14–3–3 Proteins

M. Roth, P. Kindervater, H.-P. Raich, J. Bargon, H. W. Spiess, K. Münnemann**
Continuous ^1H and ^{13}C Signal Enhancement in NMR and MRI Using Parahydrogen and Hollow Fiber Membranes

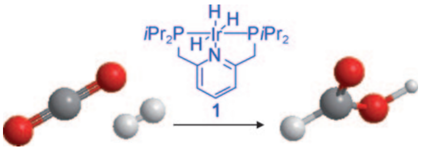
Author Profile

"If I could be anyone for a day, I would be Christopher Columbus at the time he reached America for the first time.
If I were not scientist, I would be a veterinary surgeon …"
This and more about José Barluenga can be found on page 6250.

Books

Highlights

Efficiency is key: Nozaki and co-workers managed to achieve very high turnover numbers for the reduction of carbon dioxide to formic acid using an active Iridium(III)–pincer complex **1.** This Highlight compares Nozaki's work to previously reported state-of-the-art catalysts.

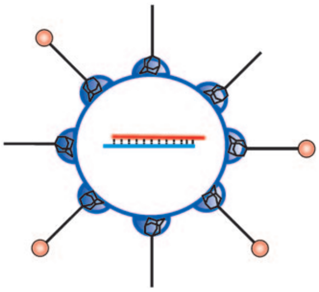


Homogeneous Catalysis

Contents

RNA Interference

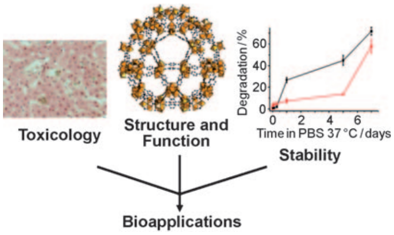
Great hope is placed in the use of RNA interference in the development of novel therapeutics. Recently, small interfering RNAs (siRNAs) were delivered to tumor cells of melanoma patients by nanoparticles consisting of a cyclodextrin polymer, and polyethylene glycol coupled to adamantane and transferrin (see picture). It was demonstrated that the target mRNA in the patients was degraded by a specific RNA interference mechanism.



Minireviews

BioMOFs

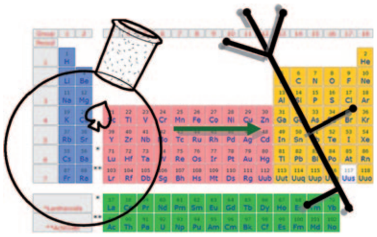
New packaging: A tremendous number of drugs have been developed but not applied further owing to poor bioavailability. To solve this problem, stable non-toxic metal–organic frameworks have been developed as drug-delivery vehicles and even as bioactive species in their own right.



Reviews

Microfluidics

Even flow: Microreactors are a new and convenient tool for liquid–liquid extraction and the optimization of inorganic chemical reactions. Fundamental studies have been carried out by this technique to understand the phenomena of nucleation and growth during chemical processes. Up-to-date data is provided, and the role of microfluidics in the field of inorganic chemistry is discussed.



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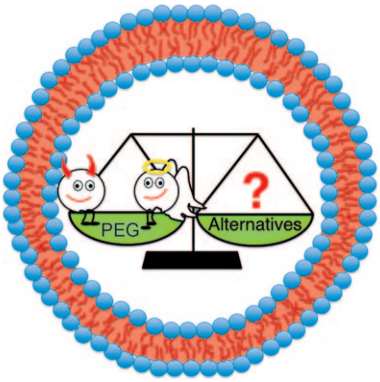
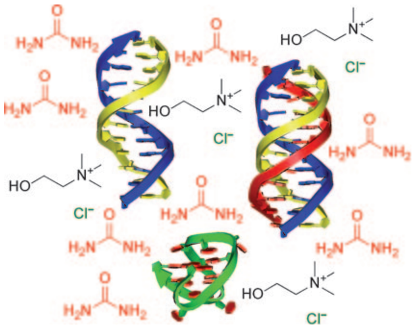


All-round talent PEG? Poly(ethylene glycol) (PEG) surrounds us in our everyday lives and, particularly in drug-delivery applications, counts as a gold standard. This omnipresent polymer is explored from different angles—advantages and disadvantages are evaluated together with alternative polymers.

Drug Delivery

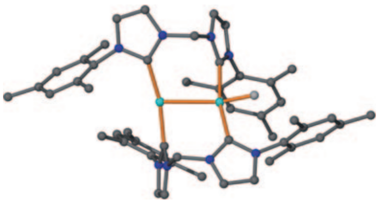
Communications

Hold the water! Deep-eutectic solvents (DESs) are nonvolatile media suitable for a wide range of chemical reactions. The discovery that nucleic acids can form duplex, triplex, and G-quadruplex structures (which in some cases differ from those adopted in aqueous media) in a water-free DES (see picture) opens the enticing possibility that catalytic nucleic acids and enzyme–nucleic acid complexes could also be used in these solvents.



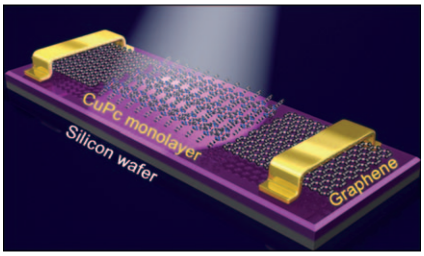
DNA Structures

Half-way house: The first palladium(I) N-heterocyclic carbene complex has been isolated in high yield by the base-assisted reduction of a palladium(II) precursor. The location of the unique terminal hydride (see picture; Pd^I cyan, H white, N blue) was established by neutron crystal structure determination, and the solution fluxional behavior of the complex was explored.



N-Heterocyclic Carbene Complexes

Molecular field-effect transistors with bulk-like carrier mobility (as high as $0.04\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$), high on/off current ratios (over 10^6), and high responsivity are formed by the integration of Langmuir–Blodgett techniques with sophisticated micro/nanofabrication. The transistors are formed from self-assembled uniform monolayers of copper phthalocyanine (CuPc) semiconductors and single-layer graphene as planar contacts.



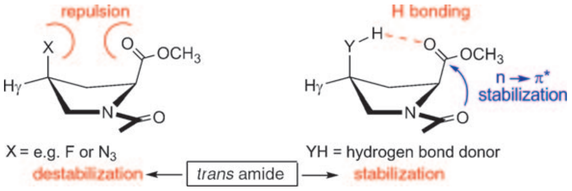
Field-Effect Transistors

Contents

Isomerization

M. Kuemin, Y. A. Nagel, S. Schweizer,
F. W. Monnard, C. Ochsenfeld,*
H. Wennemers* ______ **6324–6327**

Tuning the *cis*/*trans* Conformer Ratio of Xaa–Pro Amide Bonds by Intramolecular Hydrogen Bonds: The Effect on PPII Helix Stability



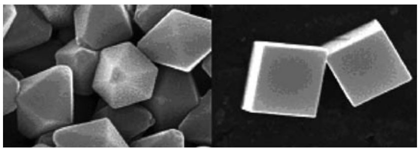
The attraction of H-bonding: Proline derivatives with a C4-*endo* ring pucker and a preference for the *trans* amide conformer are introduced as building blocks to tune the *cis*/*trans* amide conformer ratio in Xaa-Pro bonds within peptides. Noncovalent interactions, such as hydrogen bonding between the substituent at C4 of the proline ring and the amide backbone are key for favoring or disfavoring the *trans* conformer (see picture).

Nanoparticles

J. Z. Yin, Z. N. Yu, F. Gao,* J. J. Wang,
H. Pang, Q. Y. Lu* ______ **6328–6332**

Low-Symmetry Iron Oxide Nanocrystals Bound by High-Index Facets

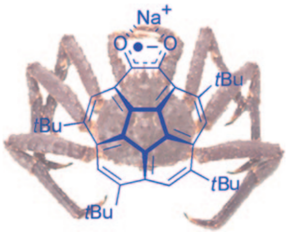
Just the shape of it: Tetrakaidecahedral (see picture, left) and oblique parallelepiped (right) α-Fe_2O_3 polyhedra with exposed high-index facets were obtained in high yield. Magnetic studies show that these two forms of iron oxide have distinct magnetic properties owing to their different shapes and different exposed high-indexed facets.



Bowl-Shaped Radicals

A. Ueda, K. Ogasawara, S. Nishida, T. Ise,
T. Yoshino, S. Nakazawa, K. Sato,
T. Takui,* K. Nakasuji,
Y. Morita* ______ **6333–6337**

A Bowl-Shaped *ortho*-Semiquinone Radical Anion: Quantitative Evaluation of the Dynamic Behavior of Structural and Electronic Features

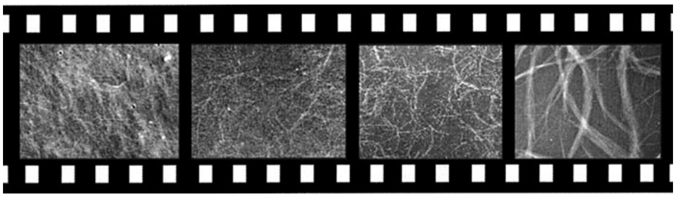


Dish it up: The first bowl-shaped *o*-semiquinone radical with a sodium cation (see picture) has been synthesized, and its 3D spin- and charge-delocalized nature and redox properties have been evaluated. The concave–convex dynamic behavior of the ion pair was also investigated in a quantitative manner using ESR and ENDOR/TRIPLE measurements and sophisticated DFT calculations.

Self-Assembly

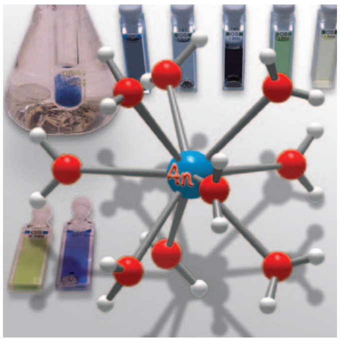
P. Mukhopadhyay,* N. Fujita,* A. Takada,
T. Kishida, M. Shirakawa,
S. Shinkai* ______ **6338–6342**

Regulation of a Real-Time Self-Healing Process in Organogel Tissues by Molecular Adhesives



Say cheeeese! The thixotropic processes that occur in a naphthalenediimide-based organogel can be imaged in real time by TEM and AFM (see picture). The self-assembled 1D fibers disintegrate under mechanical stress and undergo a self-healing process during a resting time to reconstitute the 1D fibers.

Lanthanide-like: X-ray diffraction and density functional theory studies show that $[An(H_2O)_9]^{3+}$ ions of the actinides U^{3+}–Cf^{3+} in crystals of their triflate salts have the same structure and electrostatic bonding as their lanthanide counterparts La^{3+}–Eu^{3+}. Moreover, the nine-coordinate geometry is essentially preserved in solution for the lighter elements (see picture).



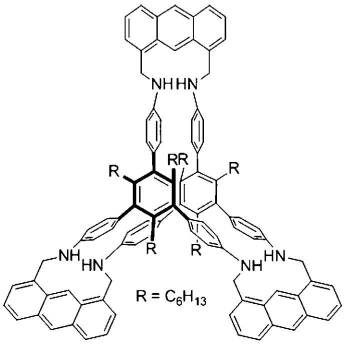
Actinide Chemistry

VIP

C. Apostolidis, B. Schimmelpfennig, N. Magnani, P. Lindqvist-Reis,* O. Walter, R. Sykora, A. Morgenstern, E. Colineau, R. Caciuffo, R. Klenze, R. G. Haire, J. Rebizant, F. Bruchertseifer, T. Fanghänel ______ **6343 – 6347**

$[An(H_2O)_9](CF_3SO_3)_3$ (An = U–Cm, Cf): Exploring Their Stability, Structural Chemistry, and Magnetic Behavior by Experiment and Theory

Carbon capture in an organic cage: A shape-persistent, organic prismatic molecular cage (see structure) was synthesized in one step and high yield from readily accessible starting materials through dynamic covalent chemistry. The resulting cage molecule exhibited high selectivity for the adsorption of CO_2 over N_2 and thus shows promise as a carbon-capture material.

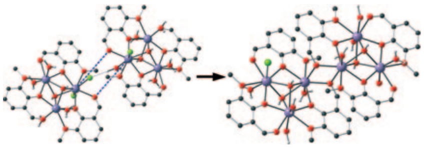


CO_2 Adsorption

Y. Jin, B. A. Voss, R. D. Noble, W. Zhang* ______ **6348 – 6351**

A Shape-Persistent Organic Molecular Cage with High Selectivity for the Adsorption of CO_2 over N_2

Two's company: Coupling two Dy_3 triangles to give Dy_6 (see picture; Dy blue, Cl green, O red) leads to a dramatic enhancement of the barrier height to magnetization reversal. The enhancement has been rationalized by single-crystal magnetometry measurements and ab initio calculations.

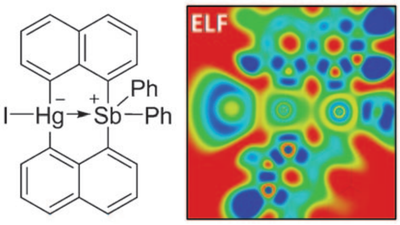


Single-Molecule Magnets

I. J. Hewitt, J. Tang, N. T. Madhu, C. E. Anson, Y. Lan, J. Luzon, M. Etienne, R. Sessoli,* A. K. Powell* ______ **6352 – 6356**

Coupling Dy_3 Triangles Enhances Their Slow Magnetic Relaxation

Fatal attraction: As a result of a unique iodide push–stibonium pull effect, the dinuclear complex (see picture) displays an unprecedented Hg→Sb interaction. The existence of this interaction was examined experimentally and theoretically.



Metal–Metal Interactions

T.-P. Lin, C. R. Wade, L. M. Pérez, F. P. Gabbaï* ______ **6357 – 6360**

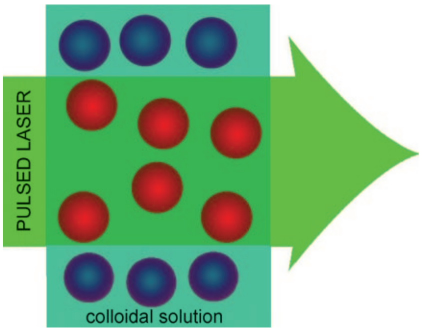
A Mercury→Antimony Interaction

Submicrometer Spheres

H. Wang, A. Pyatenko, K. Kawaguchi, X. Li, Z. Swiatkowska-Warkocka, N. Koshizaki* ________ **6361 – 6364**

Selective Pulsed Heating for the Synthesis of Semiconductor and Metal Submicrometer Spheres

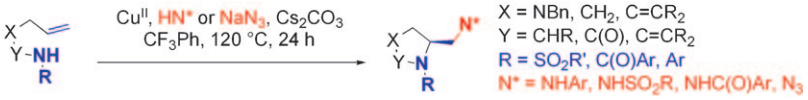
Canned heat: Inhibition of anisotropic crystal growth is critical for synthesizing semiconductor/metal spheres, but it is kinetically difficult to achieve in methods relying on conventional heating. Selective heating of colloidal nanoparticles by pulsed laser irradiation allows size-tailored semiconductor and metal submicrometer spheres to be synthesized.



Copper Catalysis

F. C. Sequeira, B. W. Turnpenny, S. R. Chemler* ________ **6365 – 6368**

Copper-Promoted and Copper-Catalyzed Intermolecular Alkene Diamination

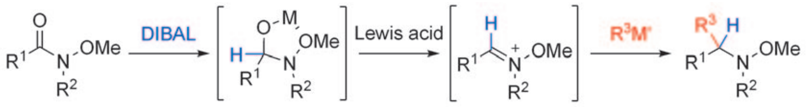


Simply copper: Copper(II)-promoted intra- and intermolecular olefin diaminations bring together two differently functionalized amines into a vicinal relationship. High diastereoselectivity was obtained with chiral alkenes and results of the asymmetric catalysis are promising.

Synthetic Methods

K. Shirokane, Y. Kurosaki, T. Sato,* N. Chida* ________ **6369 – 6372**

A Direct Entry to Substituted *N*-Methoxyamines from *N*-Methoxyamides via *N*-Oxyiminium Ions

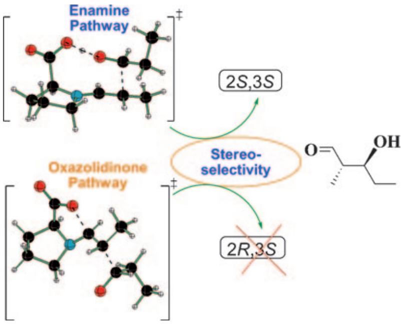


Take the direct path: Sequential nucleophilic addition of *N*-methoxyamides using DIBAL and organometallic reagents provided substituted *N*-methoxyamines in one pot via five-membered chelated intermediates (see scheme, DIBAL = diisobutylaluminum hydride). The reaction allows functionalization of acyclic amides and macrolactams without a preactivation step, which is generally required for inert amide carbonyl groups.

Stereoselectivity

A. K. Sharma, R. B. Sunoj* _ **6373 – 6377**

Enamine versus Oxazolidinone: What Controls Stereoselectivity in Proline-Catalyzed Asymmetric Aldol Reactions?

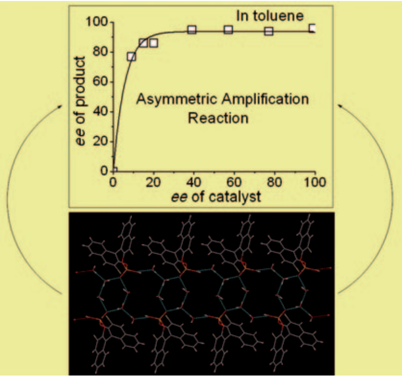


Take the right path: Comparison of the oxazolidinone and enamine pathways in enantioselective aldol reactions by using density functional and ab initio transition states reveals that the oxazolidinone route does not provide the correct stereochemical outcome (see picture), whereas the enamine pathway predicts the correct stereoselectivity.

Asymmetric Amplification

N. Li, X.-H. Chen, S.-M. Zhou, S.-W. Luo, J. Song, L. Ren, L.-Z. Gong* **6378–6381**

Asymmetric Amplification in Phosphoric Acid Catalyzed Reactions

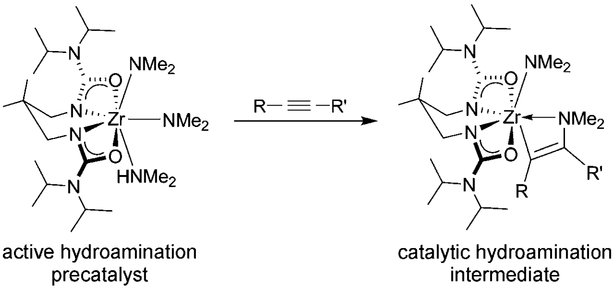


An *ee* amp: Asymmetric amplification in reactions catalyzed by phosphoric acids arose from the formation of less soluble supramolecular structures (see scheme; O red, P yellow, C gray, H white) of the racemic phosphoric acids that participate in hydrogen bonds with the crystalline water molecules. The finding shows that highly enantioselective reactions can be observed by using non-optically pure phosphoric acids.

Catalytic Intermediates

D. C. Leitch, C. S. Turner, L. L. Schafer* **6382–6386**

Isolation of Catalytic Intermediates in Hydroamination Reactions: Insertion of Internal Alkynes into a Zirconium–Amido Bond

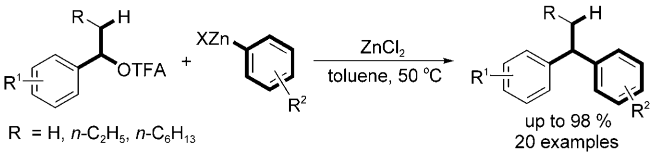


Insert alkyne to continue: Reaction of a bis(ureate) zirconium dimethylamido complex with electron-rich alkynes leads to vinylamine insertion products (see scheme). The insertion of C—C unsaturated bonds into M—N bonds is proposed as a key step in lanthanide chemistry and, more recently, hydroamination reactions catalyzed by Group 4 metals. For the first time, these elusive intermediates have been isolated and characterized on a catalytically active d^0-metal center.

Cross-Coupling

H. Duan, L. Meng, D. Bao, H. Zhang, Y. Li, A. Lei* **6387–6390**

Zinc Chloride Enhanced Arylations of Secondary Benzyl Trifluoroacetates in the Presence of β-Hydrogen Atoms

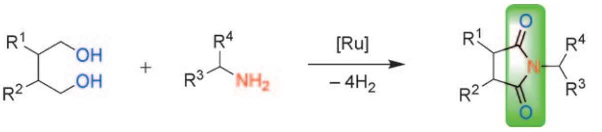


Zinc or swim: Arylation of benzyl trifluoroacetates with arylzinc reagents in the presence of β-hydrogen atoms were realized under mild conditions. Both electron-rich and electron-deficient arene substrates were successfully arylated. This arylation method could offer a very versatile synthetic route to access a series of diversity-oriented diarylalkane motifs. TFA = trifluoroacetyl.

Cyclic Imides

J. Zhang, M. Senthilkumar, S. C. Ghosh, S. H. Hong* **6391–6395**

Synthesis of Cyclic Imides from Simple Diols

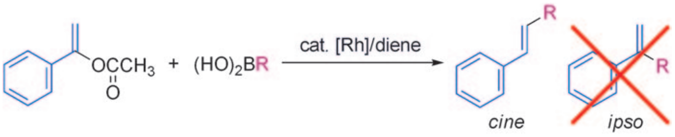


There's something imide so strong: Cyclic imides were synthesized from simple diols with primary amines in a single step using an in-situ-generated ruthenium catalytic system. The atom-economical and operatively simple syntheses of succinimides, phthalimides, and glutarimides, which are important building blocks in natural products and drugs, was also demonstrated.

cine *Substitution*

J.-Y. Yu, R. Shimizu,
R. Kuwano* ______ **6396 – 6399**

Selective *cine* Substitution of 1-Arylethenyl Acetates with Arylboron Reagents, Catalyzed by a Diene/Rhodium Complex

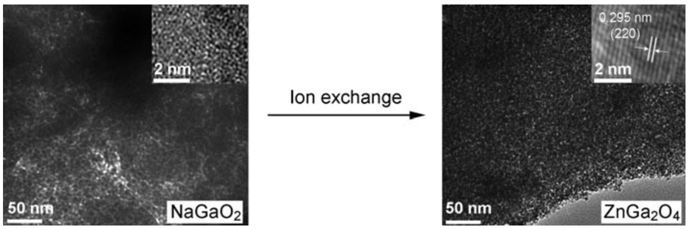


When the crowd says Bo: A carbon–carbon bond is selectively formed at the β position of 1-arylethenyl acetate when the alkenyl substrate is reacted with arylboronic acids in the presence of a cycloocta-1,5-diene/rhodium catalyst. The choice of the ligand is crucial for the unusual *cine* substitution.

Mesoporous Materials

S. C. Yan, S. X. Ouyang, J. Gao, M. Yang, J. Y. Feng, X. X. Fan, L. J. Wan, Z. S. Li,* J. H. Ye, Y. Zhou, Z. G. Zou* **6400 – 6404**

A Room-Temperature Reactive-Template Route to Mesoporous $ZnGa_2O_4$ with Improved Photocatalytic Activity in Reduction of CO_2

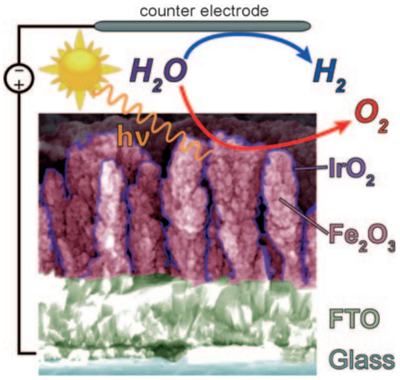


Multimetal mesoporous materials MGa_2O_4 (M = Zn, Ni, Co) can be synthesized by ion-exchange reactions of colloidal mesoporous $NaGaO_2$ (see scheme with TEM images), which acts as both template and precursor, so that a template-removal process can be dispensed with.

Water Splitting

S. D. Tilley, M. Cornuz, K. Sivula,*
M. Grätzel* ______ **6405 – 6408**

Light-Induced Water Splitting with Hematite: Improved Nanostructure and Iridium Oxide Catalysis

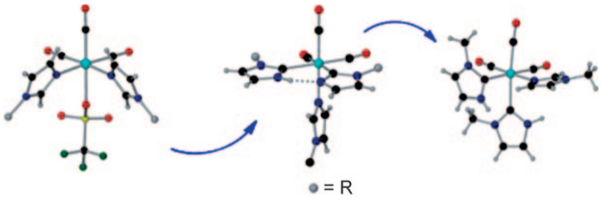


Revved-up rust! Light-induced water splitting over iron oxide (hematite) has been achieved by using a particle-assisted deposition technique and IrO_2-based surface catalysis. Photocurrents in excess of 3 mA cm^{-2} were obtained at +1.23 V versus the reversible hydrogen electrode under AM 1.5 G 100 mW cm^{-2} simulated sunlight. These photocurrents are unmatched by any other oxide-based photoanode. FTO = fluorine-doped tin oxide.

Heterocyclic Carbenes

M. A. Huertos, J. Pérez,* L. Riera,* J. Díaz,
R. López* ______ **6409 – 6412**

From Bis(*N*-Alkylimidazole) to Bis(NH–NHC) in Rhenium Carbonyl Complexes

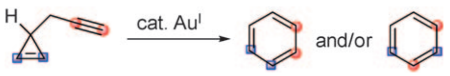


Two birds, one stone: A deprotonation–protonation sequence using only one molar equivalent of base and acid transforms two *N*-alkylimidazole molecules into N-heterocyclic carbene ligands (see scheme; black C, gray H, green F, blue N, red O, cyan Re, yellow S).

Gold Catalysis

C. Li, Y. Zeng, H. Zhang, J. Feng, Y. Zhang, J. Wang* ———— **6413 – 6417**

Gold(I)-Catalyzed Cycloisomerization of Enynes Containing Cyclopropenes

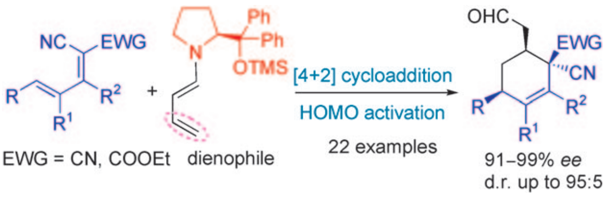


Goldenyne: Gold-catalyzed cycloisomerization reactions of propargyl cyclopropenes afford benzene derivatives in a highly efficient manner. The reaction either proceeds through a mechanism with or without double and triple bond cleavage, depending on the substituents (see scheme).

Asymmetric Synthesis

J.-L. Li, T.-R. Kang, S.-L. Zhou, R. Li, L. Wu, Y.-C. Chen* ———— **6418 – 6420**

Organocatalytic Asymmetric Inverse-Electron-Demand Diels–Alder Reaction of Electron-Deficient Dienes and Crotonaldehyde

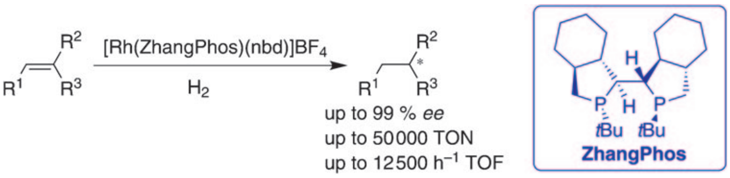


Another pathway for enals: The excellent β,γ-regioselectivity shown in the title reaction under dienamine catalysis efficiently gave highly diastereo- and enantioenriched cyclohexene derivatives with substantial substitution diversity (see scheme; EWG = electron-withdrawing group, TMS = trimethylsilyl).

Ligand Design

X. Zhang, K. Huang, G. Hou, B. Cao, X. Zhang* ———— **6421 – 6424**

Electron-Donating and Rigid P-Stereogenic Bisphospholane Ligands for Highly Enantioselective Rhodium-Catalyzed Asymmetric Hydrogenations



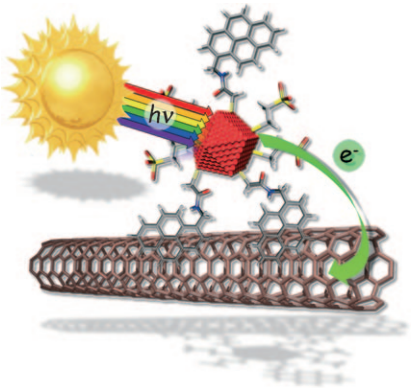
More electron donating, more rigid: A new highly electron-donating P-stereogenic bisphospholane ligand (ZhangPhos) was synthesized in a practical and highly enantioselective manner from a commercially available chiral source. Better or comparable enantioselectivities and reactivities than TangPhos were achieved in rhodium-catalyzed hydrogenation of various functionalized olefins (see scheme; nbd = 3,5-norbornadiene).

Photoactive Devices

C. Schulz-Drost, V. Sgobba, C. Gerhards, S. Leubner, R. M. Krick Calderon, A. Ruland, D. M. Guldi* —— **6425 – 6429**

Innovative Inorganic–Organic Nanohybrid Materials: Coupling Quantum Dots to Carbon Nanotubes

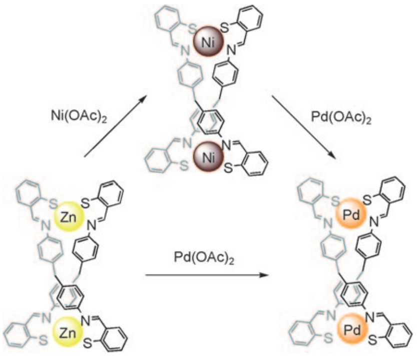
Inorganic meets organic: Covalent bonds (peptide condensation) and noncovalent interactions (π–π stacking) have been employed en route toward versatile donor–acceptor inorganic–organic nanohybrids, QD-pyrene/SWNT. A charge-transfer event within the hybrid transforms the excitonic state of the quantum dot into a charge-transfer state that has a lifetime of several nanoseconds.



Subcomponent Assembly

J. Dömer, J. C. Slootweg, F. Hupka,
K. Lammertsma,
F. E. Hahn* ______________ **6430 – 6433**

Subcomponent Assembly and Transmetalation of Dinuclear Helicates

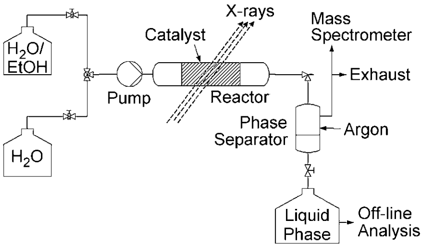


Only available as helicates: Dinuclear zinc and nickel complexes bearing previously inaccessible N,S-N,S ligands have been obtained in subcomponent self-assembly reactions from complexes bearing two 2-thiolatobenzaldehyde ligands and 4,4′-diaminodiphenylmethane. Both complexes react with Pd(OAc)$_2$ by transmetalation to give the dinuclear palladium complex (see scheme).

Reactions in Supercritical Water

S. Rabe, M. Nachtegaal, T. Ulrich,
F. Vogel* ______________ **6434 – 6437**

Towards Understanding the Catalytic Reforming of Biomass in Supercritical Water

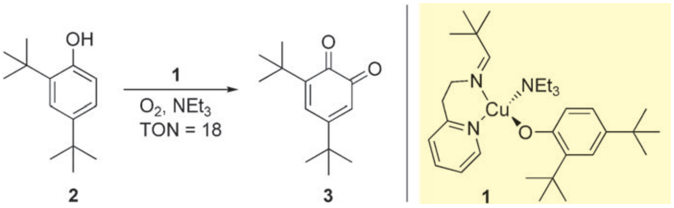


A critical point: The function of a working ruthenium catalyst during the reforming of ethanol in supercritical water was studied in situ using X-ray absorption spectroscopy (see picture). The reforming of ethanol over a supported ruthenium catalyst in sub- and supercritical water is suggested to proceed by the same mechanism as steam reforming at low pressures.

Copper Monooxygenases

M. Rolff, J. Schottenheim, G. Peters,
F. Tuczek* ______________ **6438 – 6442**

The First Catalytic Tyrosinase Model System Based on a Mononuclear Copper(I) Complex: Kinetics and Mechanism



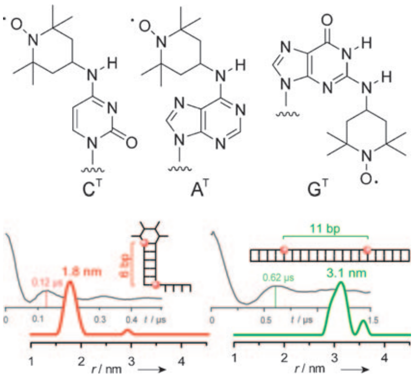
Ready... steady... go! The copper(I) complex **1** not only catalyzes the oxygenation of di-*tert*-butylphenol (DTBP-H) to di-*tert*-butylquinone (DTBQ) in a tyrosinase-like fashion, but also allows the reactive cycle to be studied in a stepwise and controlled manner. This feature opens new insights into the individual stages of the tyrosinase reaction, phenol hydroxylation, and release of the product as quinone. The implications for the enzymatic reaction are discussed.

Spin-Labeled RNA

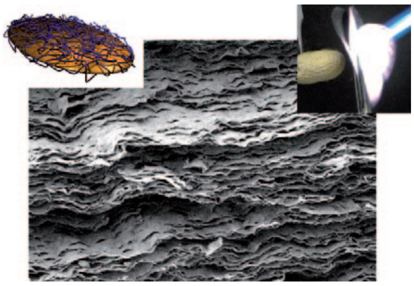
G. Sicoli, F. Wachowius, M. Bennati,*
C. Höbartner* ______________ **6443 – 6447**

Probing Secondary Structures of Spin-Labeled RNA by Pulsed EPR Spectroscopy

Tempo, TEMPO! Spin-labeled RNAs are readily prepared by postsynthetic modification of RNA. This approach provides access to paramagnetic oligonucleotides with position-specific probes in cytidine, adenosine, and guanosine nucleobases (C^T, A^T, G^T, respectively). In this way alternative RNA secondary structures can be investigated by distance measurements using pulsed EPR spectroscopy.



Taking the heat: Hard/soft core/shell colloidal building blocks allow large-scale self-assembly to form nacre-mimetic paper. The strength and stiffness of this material can be tailored by supramolecular ionic bonds. These lightweight biomimetic materials show excellent and tunable mechanical properties and heat and fire-shielding capabilities.



Nacre Paper

A. Walther,* I. Bjurhager, J.-M. Malho, J. Ruokolainen, L. Berglund, O. Ikkala* ———— **6448 – 6453**

Supramolecular Control of Stiffness and Strength in Lightweight High-Performance Nacre-Mimetic Paper with Fire-Shielding Properties

Corrigendum

In this Communication Figure 5 and Figure S5 in the Supporting Information were misinterpreted as a result of an experimental artifact. While we wrote on page 734, that two-photon excited fluorescence of TOPV in the solid state can be unambiguously recorded even on a standard spectrofluorimeter with a Xe lamp as excitation source, a critical reader noticed that a high-intensity laser is still needed to promote a material in the excited state by a two-photon absorption process. In additional experiments we found that the observed emission applying an excitation at 800 nm was a result of the higher order of the diffraction light in the monochromator of the spectrofluorimeter used. Thus, there are also photons available at 400 nm for one-photon excitation, although 800 nm was set as the excitation in the equipment. Diffraction light of a higher order in the monochromator caused the observed emission. This mistake happened as a result of relying on the specifications of the commercial equipment used for the experiments, which suggests that higher-order diffraction light from the monochromator is completely filtered out. This assumption led us to draw an incorrect conclusion because we believed our material has such a strong two-photon absorption that just a Xe lamp could be sufficient to promote the material by a two-photon absorption process. As the content that refers to Figure 5 and Figure S5 is based on a wrong assumption this part of the article is incorrect. A high-intensity laser source is still needed for two-photon excitation, as described elsewhere.[1]

Soon after the publication of our Communication, we noticed a new article by Spano[2] which revealed that some organic dyes that were formerly thought to form J-aggregates based on spectral shifts are actually H-like aggregates. Further investigation suggested that the TOPV molecules may exhibit similar behavior. Although comparison between the molecular and powder absorption (Figure S3 in the Supporting Information) suggests a red-shift of the absorption in the solid state versus in solution, TOPV molecules might form weakly coupled H-aggregate rather than J-aggregate as we had ascribed in our Communication. The small bump at 467 nm in the powder emission spectrum (Figure S3 in the Supporting Information) become apparent in nanoparticles with increasing molecular disorder, and therefore can be assigned to the 0–0 band of H-aggregates.[2,3]

The table of contents entry may also confuse some readers. The meaning of "Low in, high out" used in the Communication refers to the nature of TPA phenomenon. The TOPV molecule emits one higher-energy photon after absorbing two low-energy photons. No quantitative two-photon absorptivity of the crystal is meant in this context.

We thank Dr. Bernd Strehmel, Osterode (Germany) for alerting us to the two-photon deficiencies, and Dr. J. Gierschner, IMDEA-Nanociencia (Spain) for the assignment of J- vs. H-aggregation.

Strong Two-Photon Excited Fluorescence and Stimulated Emission from an Organic Single Crystal of an Oligo(Phenylene Vinylene)

F. Gao, Q. Liao, Z. Z. Xu, Y. H. Yue, Q. Wang, H. L. Zhang*
H. B. Fu* ———— **732–735**

Angew. Chem. Int. Ed. **2010**, *49*

DOI 10.1002/anie.200905428

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[1] B. Strehmel, V. Strehmel in *Advances in Photochemistry*, Vol. *29* (Eds.: D. C. Neckers, W. S. Jenks, T. Wolff), Wiley, Hoboken, **2007**, pp. 111 – 341.

[2] F. C. Spano, *Acc. Chem. Res.* **2010**, *43*, 429.

[3] J. Gierschner, M. Ehni, H.-J. Egelhaaf, B. Milián Medina, D. Beljonne, H. Benmansour, G. C. Bazan, *J. Chem. Phys.* **2005**, *123*, 144914.

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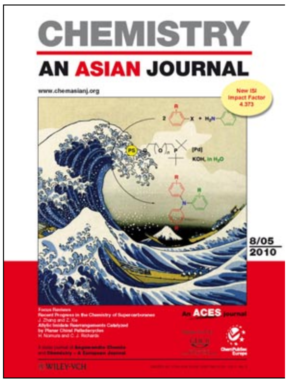

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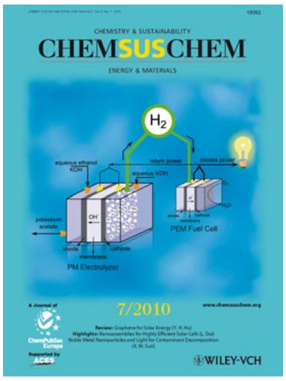

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