



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](http://www.angewandte.org) soon:

S. G. Srivatsan, N. J. Greco, Y. Tor\*

**Highly Emissive Fluorescent Nucleoside Signals the Activity of Toxic Ribosome-Inactivating Proteins**

M. Mascal\*, E. B. Nikitin

**Direct, High-Yield Conversion of Cellulose into Biofuel**

P. García-Álvarez, D. V. Graham, E. Hevia, A. R. Kennedy, J. Klett, R. E. Mulvey\*, C. T. O'Hara, S. Weatherstone

**Unmasking Representative Structures of TMP-Active Hauser and Turbo Hauser Bases**

T. Robert, J. Velder, H.-G. Schmalz\*

**Enantioselective Copper-Catalyzed 1,4-Addition of Grignard Reagents to Cyclohexenone Using Taddol-Derived Phosphine–Phosphite Ligands and 2-Methyltetrahydrofuran as a Solvent**

A. D. Shaller, W. Wang, H. Gan, A. D. Q. Li\*

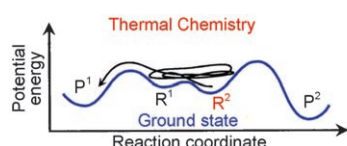
**Tunable Molecular Assembly Codes Direct Reaction Pathways**

Supramolecular Catalysis

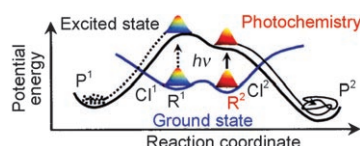
Piet W. N. M. van Leeuwen

## Books

reviewed by A. Lützen — 6320



**Trust is good, control is better:** In thermal processes in which the interconversion of reactant conformers  $R^{1/2}$  is faster than the reaction itself, selectivity can be tailored by modulating the energy barriers of competing reaction paths. Ultrafast pho-



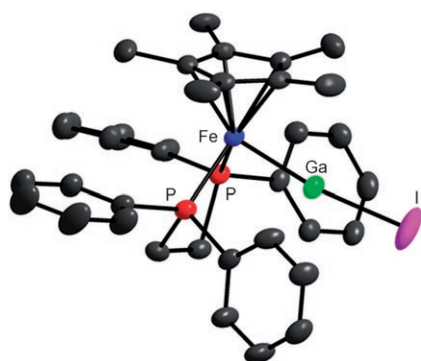
toexcitation promotes separate conformers to different regions of the potential energy surface, where the wave packet may reach different conical intersections  $Cl^{1/2}$ , giving rise to product selectivity ( $P^{1/2}$ ).

## Highlights

### Reaction Selectivity

M. Olivucci\*, F. Santoro\* — 6322–6325

Chemical Selectivity through Control of Excited-State Dynamics



**EX-traordinary alternative:** The first structurally characterized complex,  $[Cp^*Fe-(dppe)(Gal)]^+$  ( $Cp^* = C_5Me_5$ ,  $dppe = 1,2$ -bis(diphenylphosphanyl)ethane), in which a diatomic molecule (EX) from elements of Group 13 (E) and 17 (X) is coordinated to a transition-metal fragment, is the most recent contribution to the long-lasting debate about the analogies between CO and EX. The picture shows the molecular structure of the complex cation with a terminal Gal group.

### Coordination Chemistry

H.-J. Himmel\*, G. Linti\* — 6326–6328

Gal as Ligand in Transition-Metal Complexes—An Alternative to CO or  $N_2$ ?

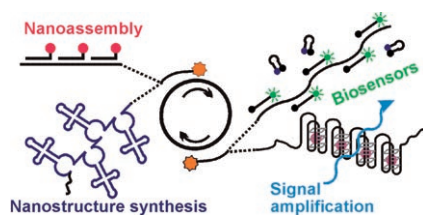
## Minireviews

### Nanobiotechnology

W. Zhao, M. M. Ali, M. A. Brook,\*  
Y. Li\* ————— 6330–6337



Rolling Circle Amplification: Applications in Nanotechnology and Biodetection with Functional Nucleic Acids



**A fruitful marriage:** Rolling circle amplification (RCA), an enzymatic reaction in which long single-stranded DNA molecules are produced, is a useful technique for the generation of novel nanostructures and large-scale DNA templates for periodic nanoassemblies (see picture). In combination with functional nucleic acids, RCA becomes a biodetection method with applications in genomics, proteomics, diagnostics, biosensing, and drug discovery.

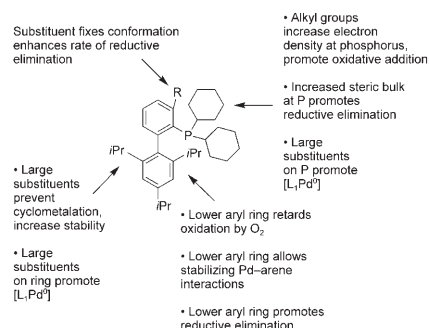
## Reviews

### Palladium Catalysis

D. S. Surry, S. L. Buchwald\* 6338–6361

Biaryl Phosphane Ligands in Palladium-Catalyzed Amination

Since the first description of dialkylbiaryl phosphane ligands in 1998, numerous uses in the synthesis of pharmaceuticals, functional materials, natural products, and heterocycles have been found. The applications of ligands of this class in palladium-catalyzed amination reactions are reviewed.



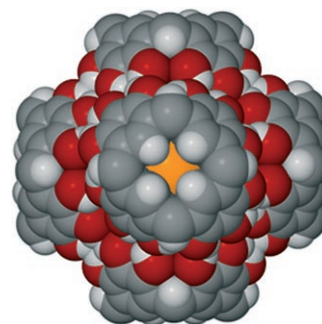
## Communications

### Self-Assembly

K. Swaminathan Iyer, M. Norret,  
S. J. Dalgarno, J. L. Atwood,  
C. L. Raston\* ————— 6362–6366

Loading Molecular Hydrogen Cargo within Viruslike Nanocontainers

**Space capsules:** Self-organized molecular capsules based on six C-alkylpyrogallol-[4]arene cavitand type molecules, held together by hydrogen bonds, can be disassembled by shearing in liquids on a rapidly rotating disk. Rapid uptake of hydrogen in the thin film on the disk is possible, which can be entrapped in the molecular capsules as they reassemble to the parent state (hexamer) in bulk solution.



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electronic / print or electronic delivery); for 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.



**Rapid resolution:** A new catalyst system for the oxidative kinetic resolution of secondary alcohols leads to dramatic rate increases. This system allows the use of a diamine to provide access to either en-

antiomer of a range of alcohols with good selectivity factors (see scheme). This method has been applied to the formal total synthesis of (–)-amurensinine.

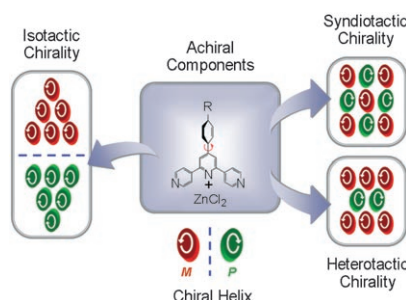
### Kinetic Resolution

D. C. Ebner, R. M. Trend, C. Genet,  
M. J. McGrath, P. O'Brien,\*  
B. M. Stoltz\* ————— 6367 – 6370

Palladium-Catalyzed Enantioselective  
Oxidation of Chiral Secondary Alcohols:  
Access to Both Enantiomeric Series



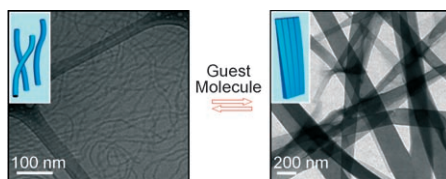
**Making a resolution:** A unique example of chiral/racemic polymorphs, which are obtained from achiral components (see picture), illustrates the evolution in chirality from partial to complete spontaneous resolution, and the phenomenon of supramolecular isotactic, syndiotactic, and heterotactic chirality.



### Supramolecular Chirality

X.-Z. Li, M. Li, Z. Li, J.-Z. Hou,  
X.-C. Huang, D. Li\* ————— 6371 – 6374

Concomitant and Controllable Chiral/  
Racemic Polymorphs: From Achirality to  
Isotactic, Syndiotactic, and Heterotactic  
Chirality



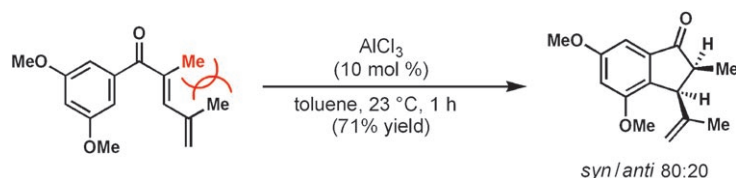
**Sticking together:** Diblock molecules containing a Y-shaped aromatic segment with a linear poly(ethylene oxide) head and branched alkyl chains at the tail have been synthesized. Cylindrical fibers are formed by the aggregation of these mole-

cules in aqueous solution (see picture). Intercalation of an aromatic guest induces lateral stacking of the aggregates to form flat ribbons and a dramatic change from a gel to a fluid solution.

### Functional Nanofibers

E. Lee, J.-K. Kim, M. Lee\* — 6375 – 6378

Lateral Association of Cylindrical  
Nanofibers into Flat Ribbons Triggered by  
“Molecular Glue”



**1,3-'s a crowd:** The facility of the AlCl<sub>3</sub>-catalyzed Nazarov cyclization of electron-rich aryl dienones was found to be dependent upon the substitution of the diene portion (see scheme). For α,γ-substituted systems, pronounced 1,3-

allylic strain in the reactant is alleviated in the transition state for electrocyclization, leading to enhanced reactivity. DFT calculations support this analysis and have proven to be predictive of reactivity.

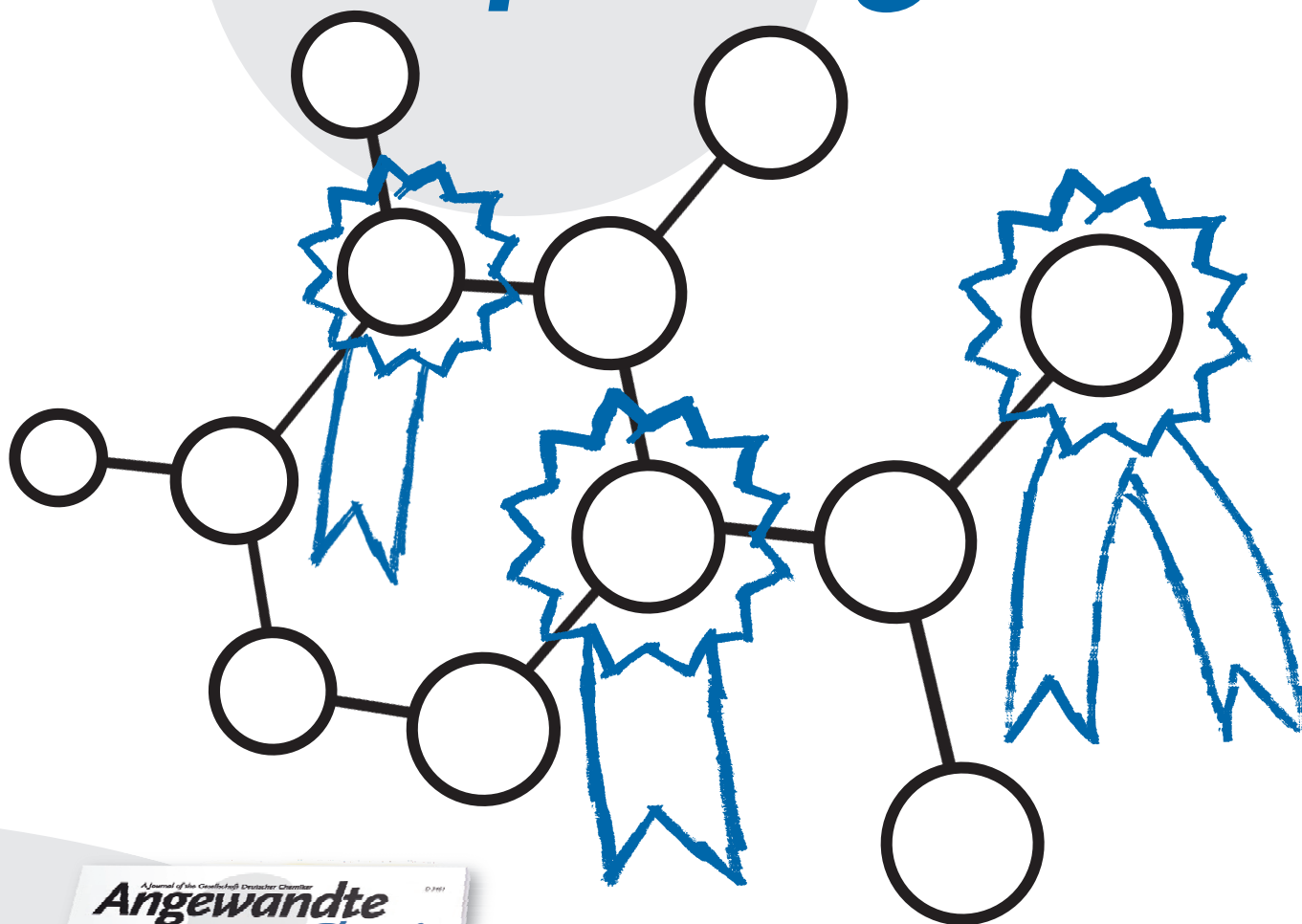
### Cyclizations

A. P. Marcus, A. S. Lee, R. L. Davis,  
D. J. Tantillo,\* R. Sarpong\* 6379 – 6383

Pronounced Steric Effects of Substituents  
in the Nazarov Cyclization of Aryl Dienyl  
Ketones



# Incredibly prestigious!



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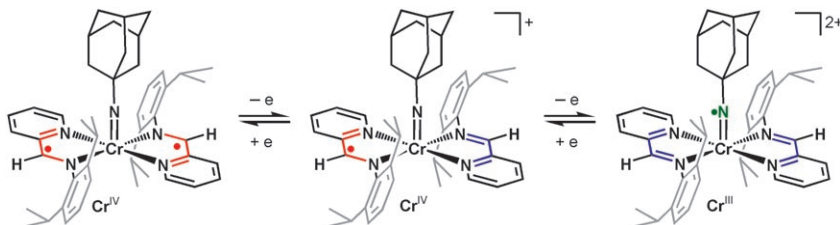
Angewandte Chemie is simply the best! With an Impact Factor of 10.232 (2006), Angewandte Chemie is considerably ahead of comparable journals. Such a high value is predominantly a reflection of the high quality of our Communications. The Reviews in Angewandte Chemie are unquestionably among the most important articles in their fields, however their contribution to the Impact Factor is much less significant than people tend to assert.

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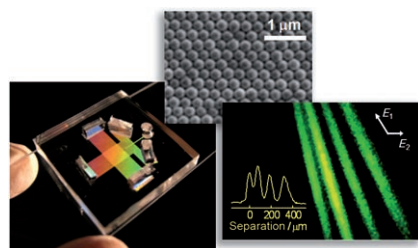
**Radically different oxidation:** An investigation of the electronic interplay between ligand radical(s) and a high-valent metal center in the three-member electron-transfer series shown in the picture

reveals that, upon oxidation and removal of both ligand radicals, the chromium center becomes reduced from Cr<sup>IV</sup> to Cr<sup>III</sup> with concomitant formation of an imidyl radical (NR)<sup>•</sup>.

### Ligand Radicals

C. C. Lu,\* S. DeBeer George,  
T. Weyhermüller, E. Bill, E. Bothe,  
K. Wieghardt\* 6384–6387

An Electron-Transfer Series of High-Valent Chromium Complexes with Redox Non-Innocent, Non-Heme Ligands



**Fleeting separation:** A general microfluidic approach has been developed for the simple and fast patterning of large-scale crack-free colloidal nanoarrays in complex microdevices. By using this technique, a 2D microfractionator for high-throughput continuous-flow separation of 2–50 kbp DNA fragments into different deflection angles with high resolution was devised.

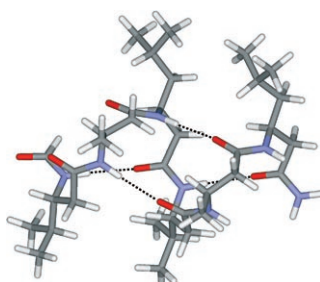
### Microfluidics

Y. Zeng, M. He,  
D. J. Harrison\* 6388–6391

Microfluidic Self-Patterning of Large-Scale Crystalline Nanoarrays for High-Throughput Continuous DNA Fractionation



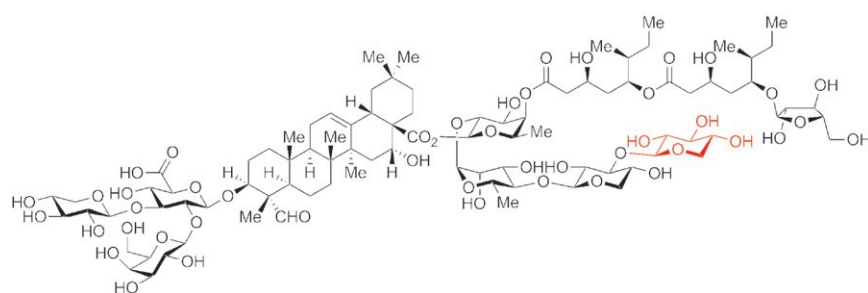
**Chiroptical tool:** Good agreement between the experimental and the quantum-chemically simulated Raman optical activity (ROA) spectrum of a  $\beta$  peptide adopting the (M)-3<sub>14</sub>-helical conformation (see model peptide; blue N, red O, gray C) demonstrates that ROA spectroscopy has the potential to provide complete structures in solution, including their absolute helicity.



### Peptide Structures

J. Kapitán, F. Zhu, L. Hecht, J. Gardiner,  
D. Seebach,\* L. D. Barron\* 6392–6394

Solution Structures of  $\beta$  Peptides from Raman Optical Activity



**A helping hand:** The adjuvant QS-21-xylose (see structure; the xylose residue is shown in red) was obtained in pure form by chemical synthesis. When combined with its synthetic apiose isomer to furnish synthetic QS-21 (sQS-21), the saponin

combination was shown to aid the production of antibodies in mice when injected with a melanoma vaccine. The preparation of sQS-21 provides access to pure, functional saponin adjuvants of defined composition.

### Natural Products

K. Deng, M. M. Adams, P. Damani,  
P. O. Livingston, G. Ragupathi,\*  
D. Y. Gin\* 6395–6398

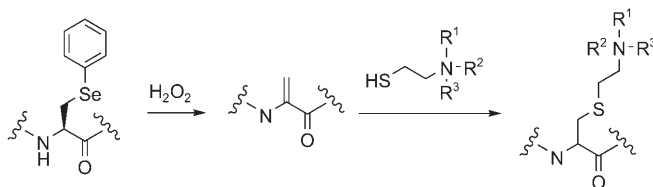
Synthesis of QS-21-Xylose: Establishment of the Immunopotentiating Activity of Synthetic QS-21 Adjuvant with a Melanoma Vaccine





## Protein Modification

J. Guo, J. Wang, J. S. Lee,  
F. G. Schultz\* — 6399 – 6401



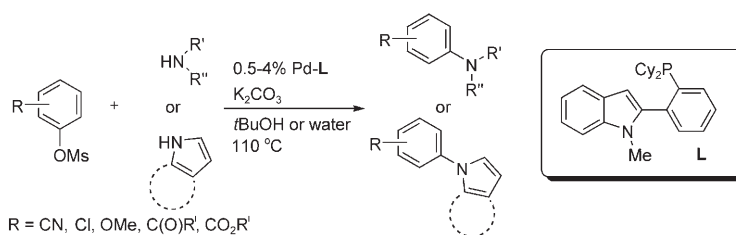
Site-Specific Incorporation of Methyl- and Acetyl-Lysine Analogues into Recombinant Proteins

**Location, location, location:** A new chemoselective method to site- and degree-specifically install lysine analogues into recombinant proteins provides facile access to near-homogeneously methyl-

ated and acetylated histone analogues (see scheme;  $R^1$ ,  $R^2$ ,  $R^3$  = H, Me, acetyl), which can be used to study the impact of histone modification on chromatin structure and function.

## Homogeneous Catalysis

C. M. So, Z. Zhou, C. P. Lau,  
F. Y. Kwong\* — 6402 – 6406



Palladium-Catalyzed Amination of Aryl Mesylates

**Aminated adventures:** The new phosphine ligand **L** in combination with a  $\text{Pd}(\text{OAc})_2$  precursor provides the first palladium-catalyzed amination of unacti-

vated aryl mesylates. This catalyst system can be applied in an aqueous reaction medium without detrimental effect.

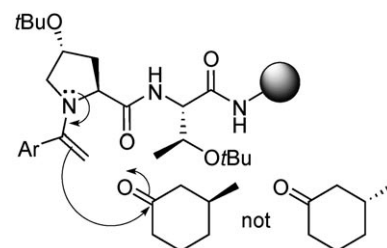
## Organocatalysis

R. D. Carpenter, J. C. Fetting, K. S. Lam,  
M. J. Kurth\* — 6407 – 6410



Asymmetric Catalysis: Resin-Bound Hydroxypropylthreonine Derivatives in Enamine-Mediated Reactions

**Control of stereochemistry** is achieved using two TentaGel-bound di-*tert*-butoxy-protected hydroxypropyl-threonine catalysts (see picture, sphere represents TentaGel). These catalysts mediate asymmetric tandem enamine/Michael reactions with high enantioselectivity and complete diastereoselectivity; the choice of catalyst depends on the desired absolute configuration.

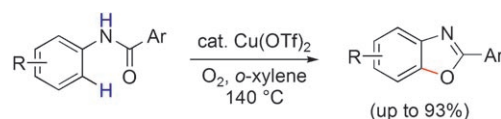


## Homogeneous Catalysis

S. Ueda, H. Nagasawa\* — 6411 – 6413

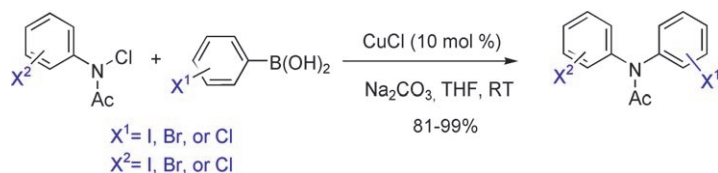


Synthesis of 2-Arylbenzoxazoles by Copper-Catalyzed Intramolecular Oxidative C–O Coupling of Benzanilides



**No need for additives:** A wide variety of functionalized 2-arylbenzoxazoles can be prepared with high functional-group tolerance and regioselectivity by a copper-catalyzed intramolecular oxidative C–O

coupling of benzanilides. The catalytic cycle is completed by the regeneration of the copper catalyst using molecular oxygen as a terminal oxidant without the need for additives.



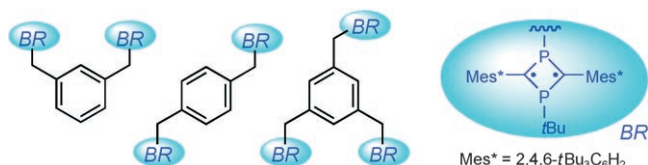
**N-Cl is no competition:** Aryl halides were tolerated in an efficient ligandless CuCl-catalyzed electrophilic amination reaction of arylboronic acids with *N*-chloroamides

(see scheme; Ac = acetoxy). This coupling proceeded smoothly at ambient temperature, and products were obtained with good to excellent yields.

## Cross-Coupling

C. He, C. Chen, J. Cheng, C. Liu, W. Liu, Q. Li, A. Lei\* 6414–6417

Aryl Halide Tolerated Electrophilic Amination of Arylboronic Acids with *N*-Chloroamides Catalyzed by CuCl at Room Temperature



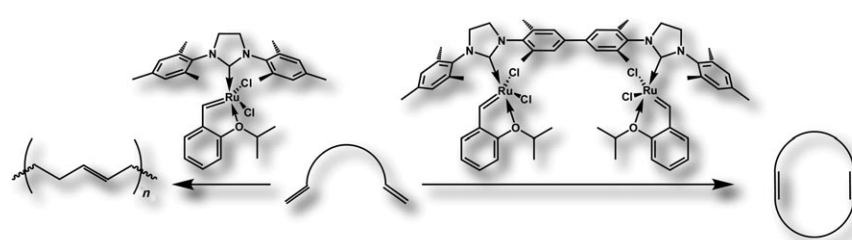
**Biradical chromophore bridge:** Di- and trimethylenephényl moieties facilitate a through-space interaction between sterically demanding biradical subunits (see picture), enabling the design of new

electronic materials. The solid-state structure of a bis(biradical) derivative illustrates the proximal positioning of two biradical subunits, allowing for direct interaction between them.

## Biradicals

S. Ito,\* J. Miura, N. Morita, M. Yoshifuji,\* A. J. Arduengo, III 6418–6421

Poly(Biradicals): Oligomers of 1,3-Diphosphacyclobutane-2,4-diyl Units



**Two ring or not to ring:** Novel diruthenium olefin metathesis catalysts show a tendency to avoid oligomerization and favor cyclic dimerization when the distances

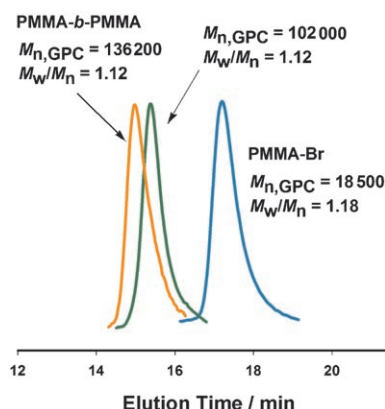
between the ruthenium centers and between the diene extremities match (see scheme).

## Ring-Closing Metathesis

E. Tzur, A. Ben-Asuly, C. E. Diesendruck, I. Goldberg, N. G. Lemcoff\* 6422–6425

Homodinuclear Ruthenium Catalysts for Dimer Ring-Closing Metathesis

**Living polymers:** Iron(III) complexes with phosphorus-containing ligands are efficient catalysts for atom transfer radical polymerization in the absence of any radical initiator or reducing agent. The polymers produced have good molecular weights and narrow polydispersities as determined by gel permeation chromatography (see scheme). The controlled nature of the  $\text{FeBr}_3/\text{dppp}$  system was confirmed by a chain-extension experiment.  $M_{n,\text{GPC}}$  in  $[\text{g mol}^{-1}]$ .



## Polymerization

Z. Xue, N. T. B. Linh, S. K. Noh,\* W. S. Lyoo 6426–6429

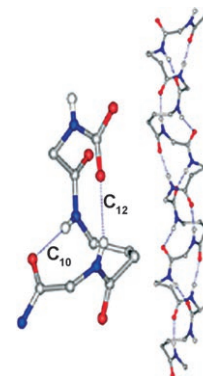
Phosphorus-Containing Ligands for Iron(III)-Catalyzed Atom Transfer Radical Polymerization

## Helical Structures

P. G. Vasudev, S. Chatterjee, K. Ananda, N. Shamala,\* P. Balaram\* — **6430–6432**

Hybrid  $\alpha\gamma$  Polypeptides: Structural Characterization of a  $C_{12}/C_{10}$  Helix with Alternating Hydrogen-Bond Polarity

**A different twist:** A mixed  $C_{12}/C_{10}$  helix with alternating hydrogen-bond directionality has been observed in the crystal structure of the hybrid  $\alpha\gamma$  peptide Boc-Leu-Gpn-Leu-Aib-OMe (see structure; Boc: *tert*-butoxycarbonyl; Leu: leucine; Gpn: 1-(aminomethyl)cyclohexanecarboxylic acid (gabapentin); Aib: aminoisobutyric acid). The structure may be generalized to a regular mixed helix.



## Spin-Crossover Nanocrystals

I. Boldog, A. B. Gaspar,\* V. Martínez, P. Pardo-Ibañez, V. Ksenofontov, A. Bhattacharjee, P. Gülich, J. A. Real\* — **6433–6437**

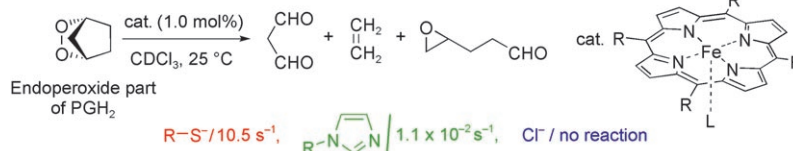


**Nanocrystals of the three-dimensional, spin-crossover, porous coordination framework**  $[\text{Fe}(\text{pz})\text{Pt}(\text{CN})_4] \cdot n\text{H}_2\text{O}$  (pz = pyrazine;  $n \leq 2.5$ ) have been synthesized from water-in-oil microemulsions. The surfactant-free nanocrystals

readily desorb water and the resulting anhydrous compounds exhibit thermally induced electronic bistability accompanied by a pronounced color change (see picture; HS = high spin, LS = low spin) close to room temperature.

## Enzyme Mechanisms

T. Yamane, K. Makino, N. Umezawa, N. Kato, T. Higuchi\* — **6438–6440**

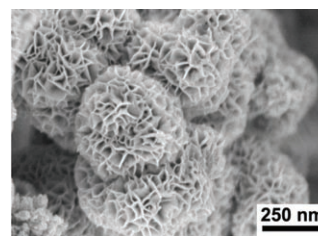
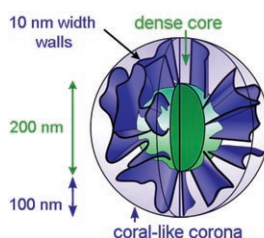


**A coordinated effort:** The isomerization mechanism of prostaglandin  $\text{H}_2$  ( $\text{PGH}_2$ ), which is catalytically isomerized to prostacyclin or thromboxane  $\text{A}_2$  by cytochrome P450s, was investigated using a heme-thiolate complex and an endoperoxide.

Isomerization of endoperoxides proceeded very rapidly with this complex, whereas imidazole- or chloride-ligated heme had slight or no catalytic activity (see scheme).

## Core–Corona Particles

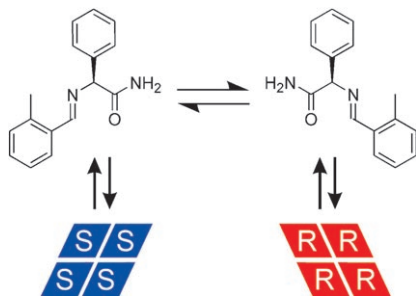
D. Portehault, S. Cassaignon,\* N. Nassif, E. Baudrin, J.-P. Jolivet — **6441–6444**



**No template and no organic medium** are needed for the low-temperature, one-pot aqueous route to layered manganese oxides with a hierarchical core–corona

architecture (see picture). Particle formation occurs by a two-step precipitation process with two different kinetics.



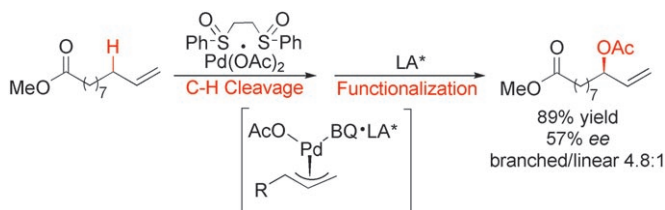


**Shaken and stirred:** The emergence of single chirality in the solid state during grinding of a slurry has been kinetically studied for a phenylglycine amide (see scheme). The insight obtained into the underlying process of attrition-enhanced Ostwald ripening enables the definition of suitable conditions to increase the deracemization rate drastically.

## Homochirality

W. L. Noorduin, H. Meekes, W. J. P. van Enckevort, A. Millemaggi, M. Leeman, B. Kaptein,\* R. M. Kellogg,\* E. Vlieg\* — 6445–6447

Complete Deracemization by Attrition-Enhanced Ostwald Ripening Elucidated



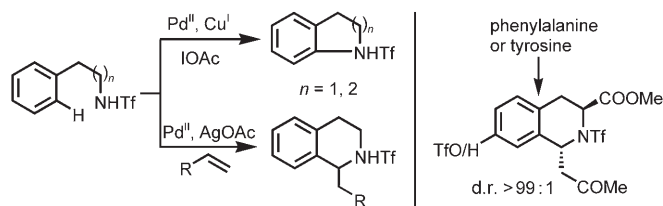
**Skip the chiral ligand:** A chiral Lewis acid strategy for effecting asymmetric induction in oxidative systems not amenable to strongly binding chiral ligands is disclosed and its mechanism investigated.

The highest levels of enantioselectivity for allylic C–H oxidation of terminal olefins is reported (see scheme; BQ = 1,4-benzoquinone, LA\* = chiral Lewis acid)

## C–H Oxidation

D. J. Covell, M. C. White\* — 6448–6451

A Chiral Lewis Acid Strategy for Enantioselective Allylic C–H Oxidation



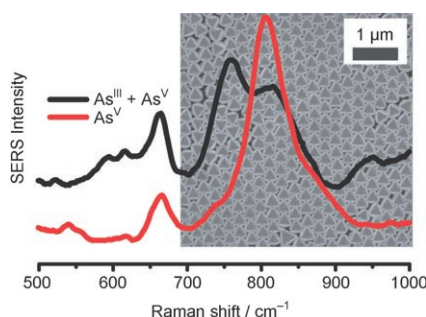
**Hand in hand:** A versatile C–H activation route for the synthesis of indolines, tetrahydroquinolines, and tetrahydroisoquinolines from simple aryethylamines relies on a one-pot iodination and amination

reaction (see scheme, Tf = trifluoromethanesulfonyl). The natural amino acids phenylalanine, tyrosine, and tryptophan can be converted into various heterocycles by using this technology.

## Heterocycle Synthesis

J.-J. Li, T.-S. Mei, J.-Q. Yu\* — 6452–6455

Synthesis of Indolines and Tetrahydroisoquinolines from Arylethylamines by PdII-Catalyzed C–H Activation Reactions



## Getting to the bottom of groundwater:

The development of a reliable, portable, and simple-to-use device for detecting arsenic in groundwater is urgently needed in developing nations such as Bangladesh, where contaminated groundwater is at the root of a public health crisis. Toward this end, a highly sensitive platform utilizing surface-enhanced Raman spectroscopy (SERS, see picture) is used to quantitatively detect arsenate in water down to 1 ppb.

## Arsenic Detection

M. Mulvihill, A. Tao, K. Benjauthrit, J. Arnold, P. Yang\* — 6456–6460

Surface-Enhanced Raman Spectroscopy for Trace Arsenic Detection in Contaminated Water



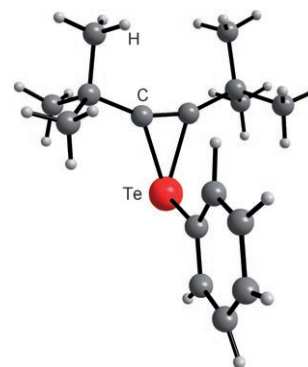
## Selenium and Tellurium

H. Poleschner,\* K. Seppelt – 6461 – 6464



Selenirenium and Tellurirenium Ions

**Canted chalcogens:** Relatively stable selenirenium salts ( $(R^1_2C_2SeR)^+X^-$  ( $R^1 = \text{tert-alkyl}$ ) are prepared by reaction of  $RSe^+X^-$  with acetylenes; the bond angle at selenium is only  $38^\circ$ . Highly unstable tellurirenium salts ( $(R^1_2C_2TePh)^+X^-$  from  $PhTe^+X^-$  and di-*tert*-alkylacetylenes have a bond angle at tellurium of  $34^\circ$  (see picture). These three-membered-ring cations were experimentally and theoretically investigated.

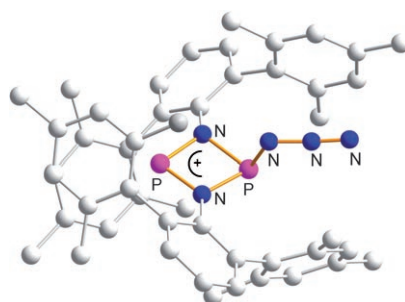


## P/N Cations

D. Michalik, A. Schulz,\* A. Villinger, N. Weding – 6465 – 6468



A Cationic Cyclic Phosphorus(III) Azide



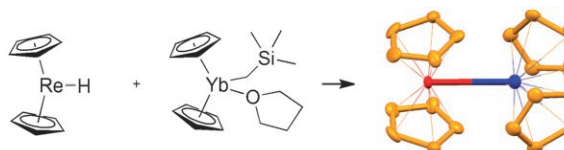
**Azide abounds:** Novel cyclic phosphorus–nitrogen cations have been synthesized by chloride abstraction from kinetically stabilized 1-chloro-*cyclo*-1,3-diphosphadiazanes.  $GaCl_3$ -assisted  $Cl/N_3$  exchange in 1-chloro-*cyclo*-1,3-diphosphadiazanium salts leads to 1-azido-*cyclo*-1,3-diphosphadiazanium-2,4-diazanium- $\mu$ -azido-hexachloridodigallate, a salt with an azide group in the cation and in the anion.

## Bimetallic Complexes

M. V. Butovskii, O. L. Tok, F. R. Wagner, R. Kempe\* – 6469 – 6472



Bismetallocenes: Lanthanoid–Transition-Metal Bonds through Alkane Elimination



**The reaction of monohydride complexes** of the late transition metals with lanthanoid alkyl derivatives affords heterobimetallic compounds with direct metal–metal

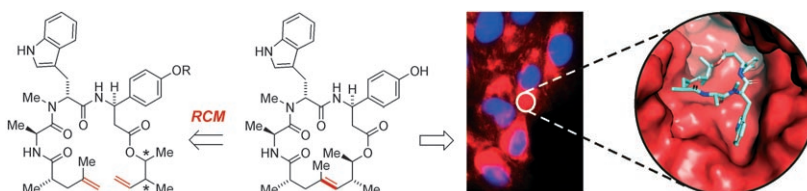
bonds. These covalent bonds are strongly polar and can be considered as donor–acceptor bonds.

## Drug Design

H. Waldmann,\* T.-S. Hu, S. Renner, S. Menninger, R. Tannert, T. Oda, H.-D. Arndt\* – 6473 – 6477

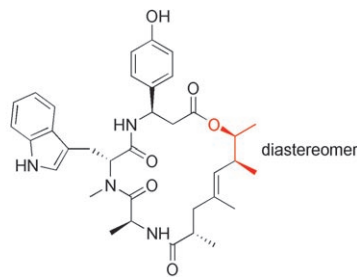
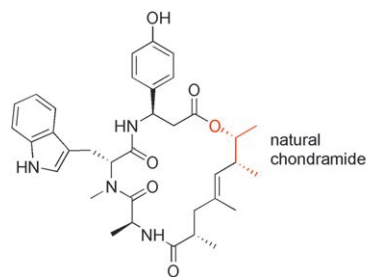


Total Synthesis of Chondramide C and Its Binding Mode to F-Actin



**Actin glue:** An *E*-selective ring-closing metathesis as the key step allowed the solid-phase-based total synthesis of the F-actin stabilizer chondramide C as well as the establishment of its hitherto unknown

stereochemistry. A strong influence of the polyketide configuration was revealed in cellular assays. Docking studies on the F-actin filament structure led to a detailed model of the binding site.



**Two solutions for one problem:** Of the four isomers of chondramide C synthesized, the two shown in the scheme

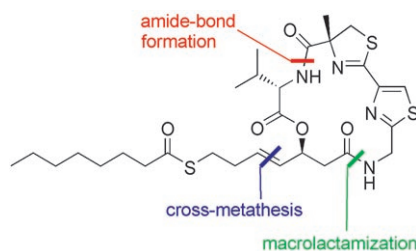
exhibit nearly the same conformation of the peptide segment and consequently unfold equal biological activities.

## Total Synthesis

U. Eggert, R. Diestel, F. Sasse, R. Jansen, B. Kunze, M. Kalesse\* — 6478–6482

Chondramide C: Synthesis, Configurational Assignment, and Structure–Activity Relationship Studies

**A modular synthesis** of the marine natural product largazole and related synthetic analogues is described. Largazole was prepared in 19% overall yield through a synthetic route with a longest linear sequence of nine steps. Activity tests showed the necessity of the thiobutenyl moiety for antiproliferative activity.

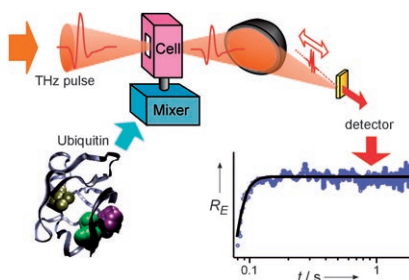


## Natural Products Synthesis

T. Seiser, F. Kamena, N. Cramer\* — 6483–6485

Synthesis and Biological Activity of Largazole and Derivatives

**Take a look around:** Kinetic terahertz absorption (KITA) spectroscopy has been used to monitor the attenuation and phase shift of a terahertz electric field transmitted through a protein during refolding. Comparison with data from fluorescence, circular dichroism, and small angle X-ray scattering shows that KITA monitors rearrangements of the hydration water–protein interactions during early collapse and secondary structure formation.



## Terahertz Spectroscopy

S. J. Kim, B. Born, M. Havenith, M. Gruebele\* — 6486–6489

Real-Time Detection of Protein–Water Dynamics upon Protein Folding by Terahertz Absorption Spectroscopy



Supporting information is available on [www.angewandte.org](http://www.angewandte.org) (see article for access details).



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

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