

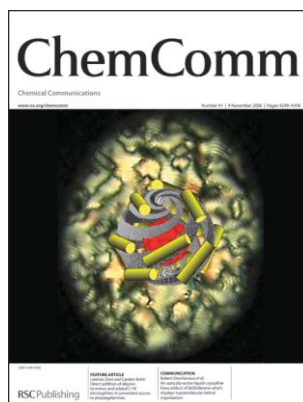
## IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (41) 4249–4348 (2006)



### Cover

See Jagadese J. Vittal *et al.*, page 4276.  
The cover picture depicts the formation of metastable orthorhombic AgInS<sub>2</sub> nanocrystals by decomposing [(Ph<sub>3</sub>P)<sub>2</sub>AgIn(SCOPh)<sub>4</sub>] in dodecanethiol and oleic acid at 125–200 °C. Image reproduced by permission of Lu Tian, Hendry Izaac Elim, Wei Ji and Jagadese J. Vittal, *Chem. Commun.*, 2006, 4276.



### Inside cover

See Robert Deschenaux *et al.*, page 4282.  
A chiral nematic phase for a liquid crystal substituted [60]fullerene separating from the liquid. Superimposed above is the proposed molecular 'boojom' structure of the fullerene by J. W. Goodby. Image reproduced by permission of Stéphane Campidelli, Torsten Brandmüller, Andreas Hirsch, Isabel M. Saez, John W. Goodby and Robert Deschenaux, *Chem. Commun.*, 2006, 4282.

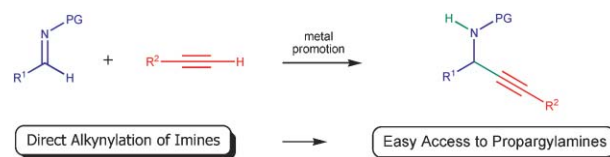
## FEATURE ARTICLE

4263

### Direct addition of alkynes to imines and related C=N electrophiles: A convenient access to propargylamines

Lorenzo Zani and Carsten Bolm\*

Advances in the preparation of propargylic amines *via* the direct addition of alkynes to imines and related carbon–nitrogen electrophiles in the presence of metal catalysts or promoters are summarised.



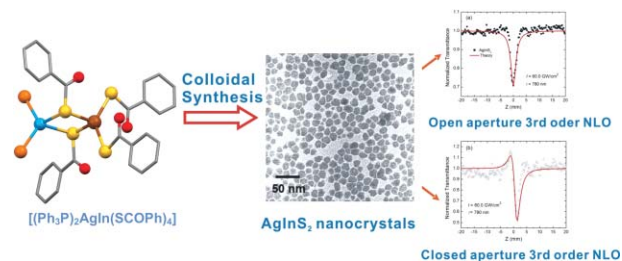
## COMMUNICATIONS

4276

### One-pot synthesis and third-order nonlinear optical properties of AgInS<sub>2</sub> nanocrystals

Lu Tian, Hendry Izaac Elim, Wei Ji\* and Jagadese J. Vittal\*

Colloidal nanocrystals of metastable orthorhombic AgInS<sub>2</sub> obtained by decomposing [(Ph<sub>3</sub>P)<sub>2</sub>AgIn(SCOPh)<sub>4</sub>] in dodecanethiol and oleic acid at 125–200 °C, show significant third order non-linear optical properties.



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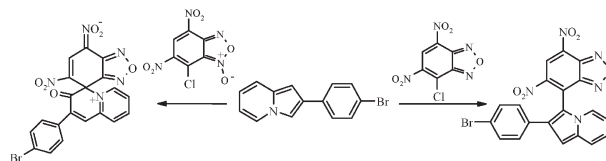
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4279

**Ring opening and ring closure in an indolizine structure activated through  $S_NAr$  coupling with superelectrophilic 4,6-dinitrobenzofuroxan, an unusual intramolecular oxygen transfer from a N-oxide functionality**

Sergey Kurbatov, Artem Tatarov, Vladimir Minkin, Régis Goumont and François Terrier\*

Coupling of superelectrophilic 4,6-dinitrobenzofuroxan with a  $\pi$ -excessive indolizine structure affords a strongly dipolar substitution product which undergoes a facile but unusual rearrangement induced by an intramolecular oxygen atom transfer from the N-oxide functionality of the DNBF moiety.

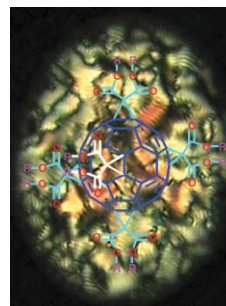


4282

**An optically-active liquid-crystalline hexa-adduct of [60]fullerene which displays supramolecular helical organization**

Stéphane Campidelli, Torsten Brandmüller, Andreas Hirsch,\* Isabel M. Saez, John W. Goodby\* and Robert Deschenaux\*

Polyaddition of mesogenic moieties to  $C_{60}$  were found to yield chiral supermolecular nanoparticles which exhibit iridescent helical chiral nematic phases.

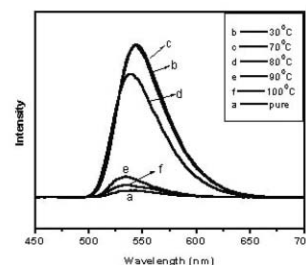
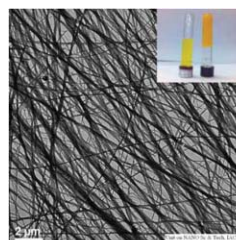


4285

**A two component thermoreversible hydrogel of riboflavin and melamine: Enhancement of photoluminescence in the gel form**

Swarup Manna, Abhijit Saha and Arun K. Nandi\*

A new thermoreversible hydrogel of riboflavin and melamine supramolecular complex ( $\geq 0.02\%$ , w/v) shows enhanced photoluminescence properties through H-bonding.

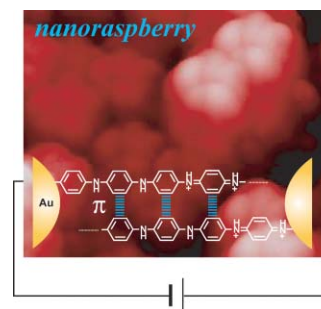


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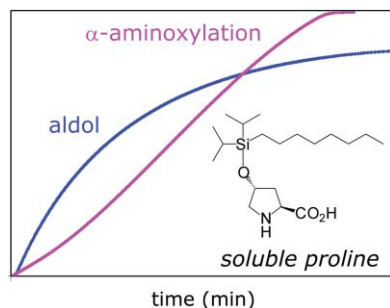
**One-step preparation of positively-charged gold nanoraspberry**

Hiroshi Shiigi,\* Yojiro Yamamoto, Naonobu Yoshi, Hidenobu Nakao and Tsutomu Nagaoka

Self-assembling particles were prepared by a one-step synthesis of a raspberry-like aggregate based on three-dimensional particle–aniline oligomer–particle repeated sequences. Their properties could help the realization of nanoscale electronics and molecular devices.



4291

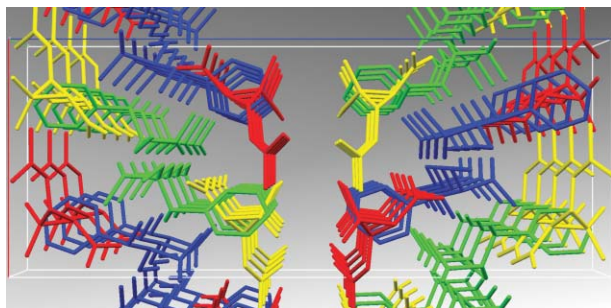


### A mechanistic rationalization of unusual kinetic behavior in proline-mediated C–O and C–N bond-forming reactions

Suju P. Mathew, Martin Klussmann, Hiroshi Iwamura, David H. Wells, Jr., Alan Armstrong and Donna G. Blackmond\*

Kinetic evidence supports the role of the product in the catalytic cycle of proline-mediated  $\alpha$ -aminoxylation and  $\alpha$ -amination, providing a model for the evolution of efficiency in catalysis.

4294

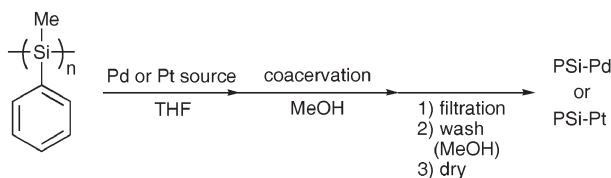


### $p_1, n_1$ Salts: self-assembled supramolecular structures sequestering racemates. Diastereomeric separation and enantiomeric enrichment of *trans*-chrysanthemic acid

Goffredo Rosini,\* Claudia Ayoub, Valerio Borzatta, Andrea Mazzanti, Emanuela Marotta and Paolo Righi

$p_1, n_1$  Salts can be exploited as a racemate sequestrant. An application to technical mixtures of chrysanthemic acids (ChA) allowed the separation of *trans*- and *cis*-ChA.

4297

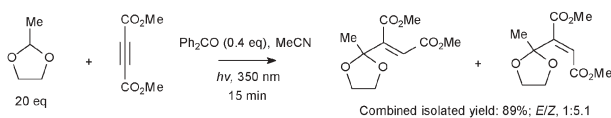


### Polysilane-supported Pd and Pt nanoparticles as efficient catalysts for organic synthesis

Hidekazu Oyamada, Ryo Akiyama, Hiroyuki Hagio, Takeshi Naito and Shū Kobayashi\*

Polysilane-supported Pd and Pt catalysts have been prepared for the first time, and used successfully in hydrogenation, Suzuki and Sonogashira reactions, and hydrosilylation respectively: the reactions proceeded in high yields, and the catalysts could be recovered quantitatively by simple filtration and reused.

4300



### The rapid photomediated alkenylation of 2-alkyl-1,3-dioxolanes with alkynes: a stereoelectronically assisted chain reaction

Niall W. A. Geraghty\* and Andrea Lally

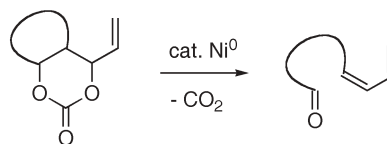
The photomediated addition of 2-alkyl-1,3-dioxolanes to alkynes is remarkably rapid for stereoelectronic reasons. This C–C bond forming reaction demonstrates the potential of photochemically generated radicals in organic synthesis.

4303

### Nickel catalyzed Grob fragmentation: $\omega$ -dienyl aldehydes synthesis

Masahiko Mori, Masanari Kimura, Yushi Takahashi and Yoshinao Tamaru\*

Nickel(0) catalyzes decarboxylative fragmentation reaction of cyclic carbonates to  $\omega$ -dienyl aldehydes *via* novel  $\beta$ -carbon elimination of  $\pi$ -allylnickel intermediates.

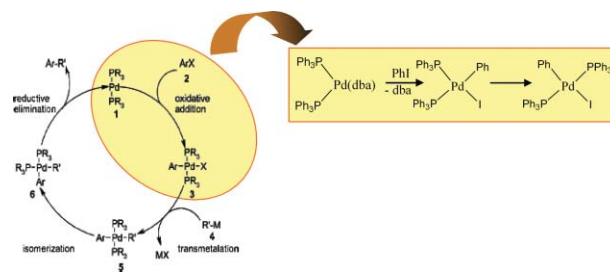


4306

### *In situ* investigation of the oxidative addition in homogeneous Pd catalysts by synchronised time resolved UV-Vis/EXAFS

Gemma Guilera,\* Mark A. Newton, Charlene Polli, Sakura Pascarelli, Meritxell Guinó and King Kuog (Mimi) Hii\*

Simultaneous structuro-kinetic information obtained *in situ* *via* time resolved stopped-flow/UV-Vis spectroscopy/dispersive EXAFS (EDE) experiments elucidated a two-step process for the addition of iodobenzene to [(Ph<sub>3</sub>P)<sub>2</sub>Pd(dba)].

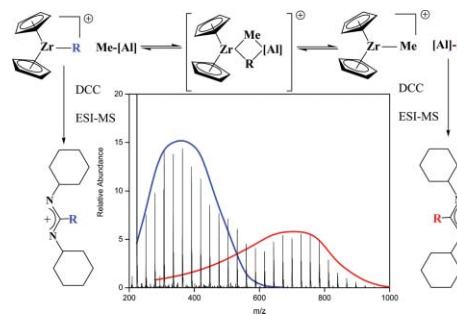


4309

### Chain transfer to aluminium in MAO-activated metallocene-catalyzed polymerization reactions

Esther Quintanilla, Fabio di Lena and Peter Chen\*

Electrospray ionization mass spectrometry (ESI-MS) of MAO-activated, Cp<sub>2</sub>ZrCl<sub>2</sub>-catalyzed polymerizations, quenched with carbodiimides, identifies and quantifies alkylzirconocenium species and higher alkyl aluminiums.

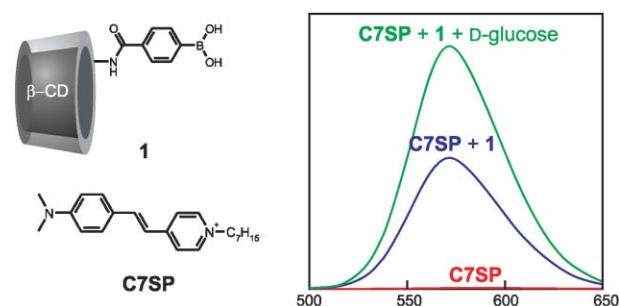


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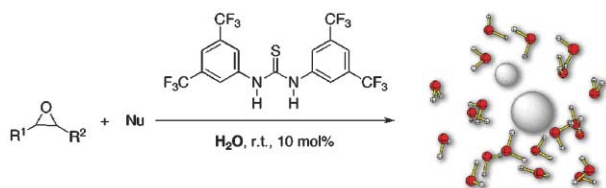
### Pseudorotaxane-type fluorescent receptor exhibiting unique response to saccharides

Akiyo Yamauchi, Yoshiko Sakashita, Kazuaki Hirose, Takashi Hayashita and Iwao Suzuki\*

Pseudorotaxane-type complexes of phenylboronic acid-modified  $\beta$ -cyclodextrin with 1-heptyl-4-(4'-dimethylaminostyryl)pyridinium were found to be fluorescent saccharide receptors, the fluorescence of which was enhanced by monosaccharides with D-glucose selectivity.



4315

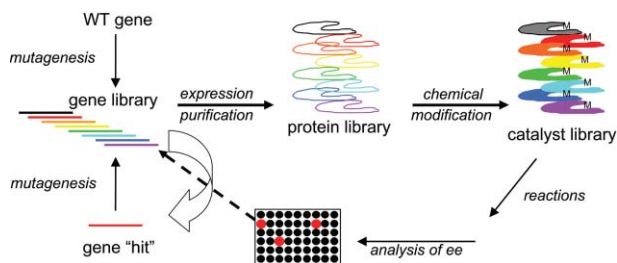


### Hydrophobic amplification of noncovalent organocatalysis

Christian M. Kleiner and Peter R. Schreiner\*

The effects of hydrogen-bonding organocatalysts and water for the acceleration of epoxide openings with a variety of nucleophiles are additive and lead to excellent yields of the catalyzed reactions in water.

4318

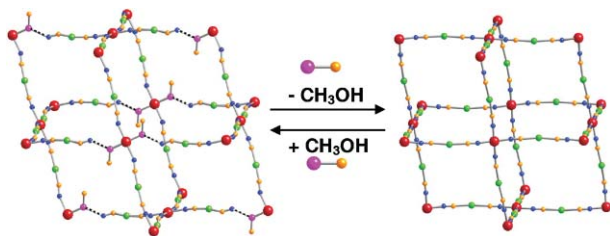


### Directed evolution of hybrid enzymes: Evolving enantioselectivity of an achiral Rh-complex anchored to a protein

Manfred T. Reetz,\* Jérôme J.-P. Peyralans, Andrea Maichele, Yu Fu and Matthias Maywald

The concept of utilizing the methods of directed evolution for tuning the enantioselectivity of synthetic achiral metal–ligand centers anchored to proteins has been implemented experimentally for the first time.

4321



### Coordination polymers undergoing spin crossover and reversible ligand exchange in the solid

Ana Galet, M. Carmen Muñoz and José Antonio Real\*

A crystalline-state ligand exchange reaction involving substitution-active iron(II) coordination sites able to selectively recognise guest  $\text{CH}_3\text{OH}$  molecules induces the reversible transformation of a non porous 2-D coordination polymer into a triple interpenetrated 3-D microporous framework.

4324



### Photo-deposition and film formation of benzenethiol monolayer protected clusters in halogenated media

Michael Busby\* and Franco Scandola\*

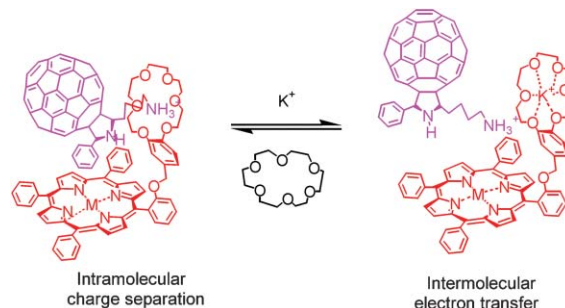
2.1 nm benzenethiol monolayer protected gold clusters undergo solvent dependant photoinduced deposition with potential applications in the field of nanoparticle film formation.

4327

### Electron transfer switching in supramolecular porphyrin–fullerene conjugates held by alkylammonium cation-crown ether binding

Atula S. D. Sandanayaka, Yasuyuki Araki, Osamu Ito, Raghu Chitta, Suresh Gadde and Francis D'Souza

Reversible switching between intra- to intermolecular electron transfer paths has been accomplished by adding and extracting potassium ions to the supramolecular porphyrin–fullerene conjugates.

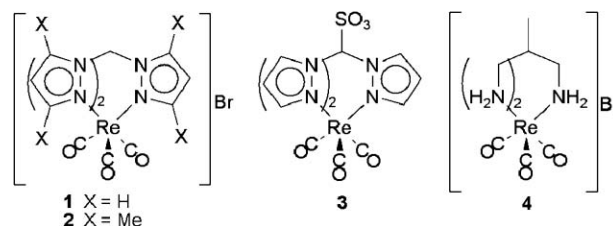


4330

### Aqueous preparation and physiological stability studies of Re(CO)<sub>3</sub>(tripodal) compounds

Richard S. Herrick,\* Tim J. Brunker,\* Caroline Maus, Kerianne Crandall, Anil Cetin and Christopher J. Ziegler

The preparation of compounds 1–4 was achieved under aqueous conditions to test their possible use as bifunctional chelating agents for <sup>99m</sup>Tc diagnostic imaging. The crystal structures of 2–4 are reported.

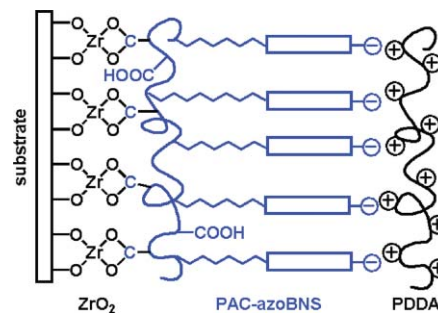


4332

### A facile room temperature layer-by-layer deposition process for the fabrication of ultrathin films with noncentrosymmetrically oriented azobenzene chromophores

En-Hua Kang, Pengcheng Jin, Yanqiang Yang, Junqi Sun\* and Jiacong Shen

Using a combination of layer-by-layer assembly of polyelectrolytes and the surface sol–gel process, layered ultrathin films with noncentrosymmetrically oriented azobenzene chromophores were fabricated.

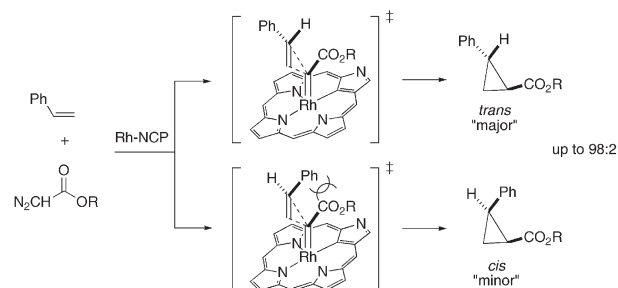


4335

### Rhodium N-confused porphyrin-catalyzed alkene cyclopropanation

Tepei Niino, Motoki Toganoh, Bruno Andrioletti and Hiroyuki Furuta\*

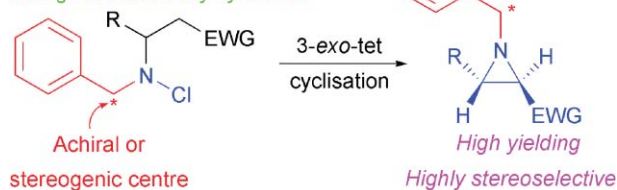
Alkene cyclopropanation reactions catalyzed by rhodium N-confused porphyrins (Rh-NCP) were examined, and excellent regioselectivity (*trans* : *cis* = 98 : 2) was observed in high yield, displaying the crucial role of *confusion* in catalytic reactions.



4338



Cheap reagents  
Straightforward easy synthesis

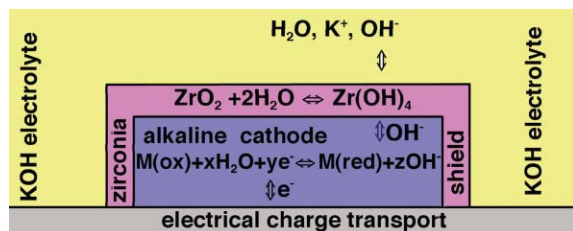


### Stereoselective synthesis of *N*-alkylaziridines from *N*-chloroamines

Sean P. Bew,\* D. L. Hughes, Nicholas J. Palmer, Vladimir Savic, Katy M. Soapi and Martin A. Wilson

In an efficient three-step process that utilises *N*-chloroamines as key intermediates, the synthesis of racemic and chiral non-racemic *N*-alkylaziridines has been accomplished.

4341

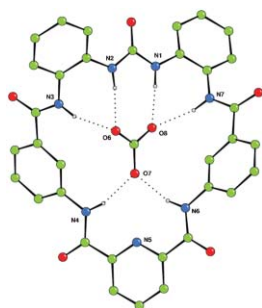


### Cathodic chemistry of high performance Zr coated alkaline materials

Stuart Licht,\* Xingwen Yu and Dong Zheng

A low level zirconia modification significantly stabilizes energetic cathodes under consideration to improve the energy storage capacity of alkaline batteries.

4344



### Anion-binding modes in a macrocyclic amidourea

Simon J. Brooks, Philip A. Gale\* and Mark E. Light

A new macrocyclic amidourea has been shown to have a high selectivity for carboxylates in DMSO-*d*<sub>6</sub>-water mixtures. The crystal structure of the carbonate complex of the macrocycle has been elucidated showing the anion bound by six hydrogen bonds in the solid state.



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