

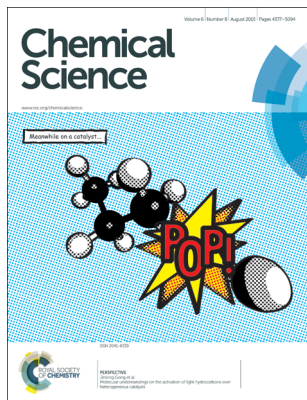
# Chemical Science

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## IN THIS ISSUE

ISSN 2041-6539 CODEN CSHCBM 6(8) 4377–5094 (2015)



### Cover

See Jinlong Gong *et al.*, pp. 4403–4425.  
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### Inside cover

See Jean-François Nierengarten, Dirk M. Guldi, Enrique Ortí, Nazario Martín *et al.*, pp. 4426–4432.  
Image reproduced by permission of Nazario Martín from *Chem. Sci.*, 2015, 6, 4426.

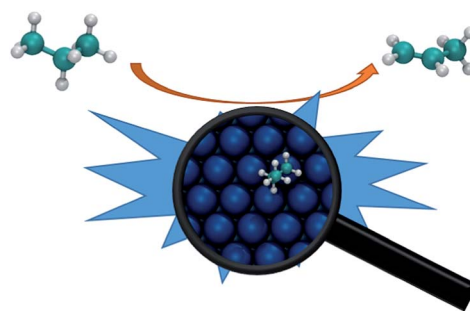
## PERSPECTIVE

4403

### Molecular understandings on the activation of light hydrocarbons over heterogeneous catalysts

Zhi-Jian Zhao, Cheng-chau Chiu and Jinlong Gong\*

This review describes recent progress on mechanistic understanding of heterogeneous catalytic dehydrogenation reactions of light alkanes.



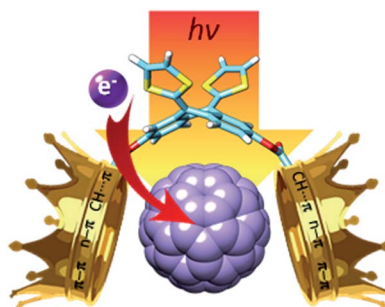
## EDGE ARTICLES

4426

### Unveiling the nature of supramolecular crown ether–C<sub>60</sub> interactions

L. Moreira, J. Calbo, R. M. Krick Calderon, J. Santos, B. M. Illescas, J. Aragón, J.-F. Nierengarten,\* D. M. Guldi,\* E. Ortí\* and N. Martín\*

Preparation of exTTF-(crown ether)<sub>2</sub> receptors, which host C<sub>60</sub>, to understand the nature of the fullerene–crown ether interaction. A combination of experimental and *in silico* studies suggest that it results from the interplay of donor–acceptor,  $\pi$ – $\pi$ ,  $n$ – $\pi$  and CH $\cdots$  $\pi$  interactions.



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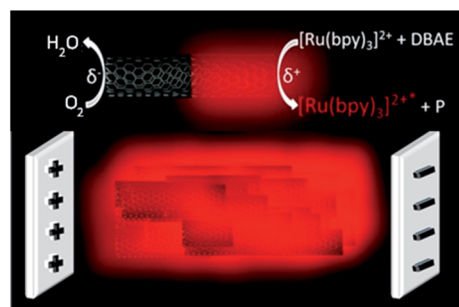


4433

**3D electrogenerated chemiluminescence: from surface-confined reactions to bulk emission**

Milica Sentic, Stéphane Arbault, Laurent Bouffier, Dragan Manojlovic, Alexander Kuhn\* and Neso Sojic\*

Electrogenerated chemiluminescence is extended to the 3D by generating light at the level of millions of micro-emitters addressed remotely by bipolar electrochemistry.

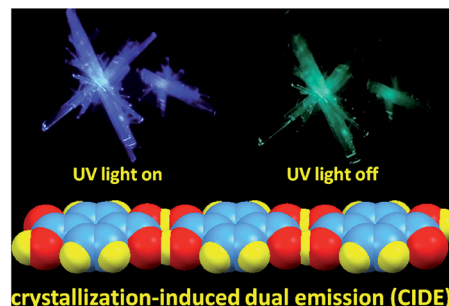


4438

**Crystallization-induced dual emission from metal- and heavy atom-free aromatic acids and esters**

Yongyang Gong, Lifang Zhao, Qian Peng, Di Fan, Wang Zhang Yuan,\* Yongming Zhang\* and Ben Zhong Tang

Crystallization-induced dual emission (fluorescence and phosphorescence) is observed in a group of pure organic aromatic acids and esters.

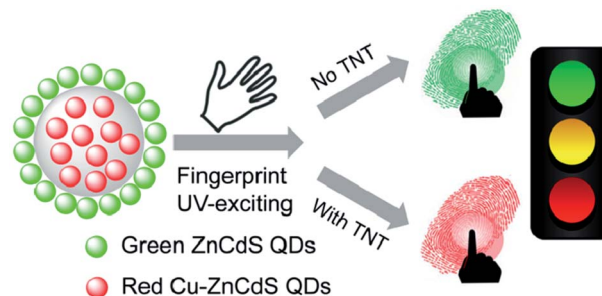


4445

**Dual-emitting quantum dot nanohybrid for imaging of latent fingerprints: simultaneous identification of individuals and traffic light-type visualization of TNT**

Peng Wu, Chaoying Xu, Xiandeng Hou, Jing-Juan Xu\* and Hong-Yuan Chen\*

A nanohybrid was employed for fingerprint imaging that was capable of simultaneous identification of individuals and TNT visualization in a "traffic-light" manner.

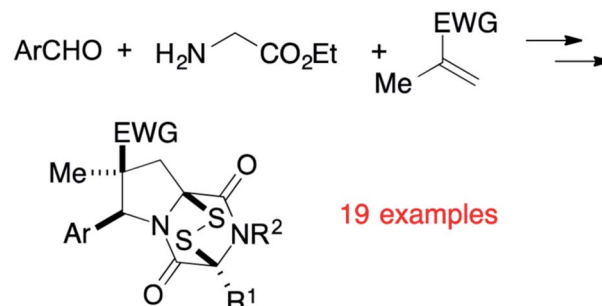


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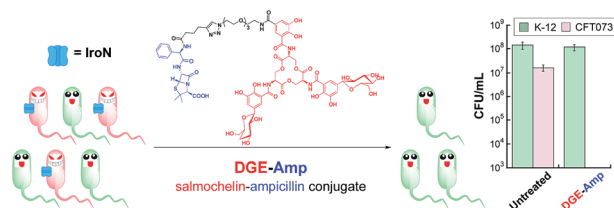
**Tricyclic analogues of epidithiodioxopiperazine alkaloids with promising *in vitro* and *in vivo* antitumor activity**

Marcus Baumann, André P. Dieskau, Brad M. Loertscher, Mary C. Walton, Sangkil Nam, Jun Xie, David Horne\* and Larry E. Overman\*

A short synthesis of 1,4-dioxohexahydro-6*H*-3,8*a*-epidithiopyrrolo[1,2-*a*]pyrazines will enable future mechanistic and translational studies of these structurally novel and promising clinical antitumor candidates.



4458



### Targeting virulence: salmochelin modification tunes the antibacterial activity spectrum of $\beta$ -lactams for pathogen-selective killing of *Escherichia coli*

Phoom Chairatana, Tengfei Zheng and Elizabeth M. Nolan\*

New antibiotics are required to treat bacterial infections and counteract the emergence of antibiotic resistance.

4472

### Object Image

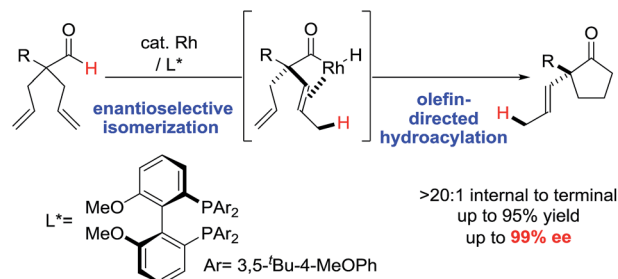


### Small molecular logic systems can draw the outlines of objects via edge visualization

Jue Ling, Gaowa Naren, Jessica Kelly, David B. Fox and A. Prasanna de Silva

Like a child with a crayon, logical molecules produce outline drawings from a template.

4479

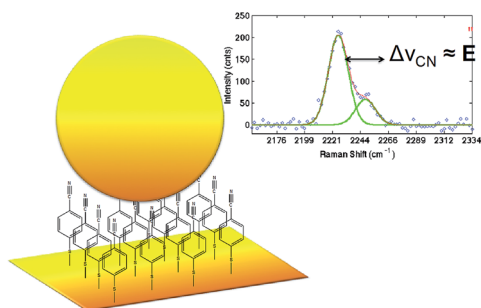


### Rh-catalyzed desymmetrization of $\alpha$ -quaternary centers by isomerization-hydroacylation

Jung-Woo Park, Kevin G. M. Kou, Daniel K. Kim and Vy M. Dong\*

A Rh-catalyzed desymmetrization of  $\alpha, \alpha$ -bis(allyl)aldehydes occurs by enantioselective isomerization followed by olefin-directed hydroacylation.

4484



### Alkyl-nitrile adlayers as probes of plasmonically induced electric fields

Daniel T. Kwasnieski, Hao Wang and Zachary D. Schultz\*

Adsorbed mercaptoalkylnitriles show Stark shifts on plasmonic surfaces that provide a direct measurement of the induced electric field.



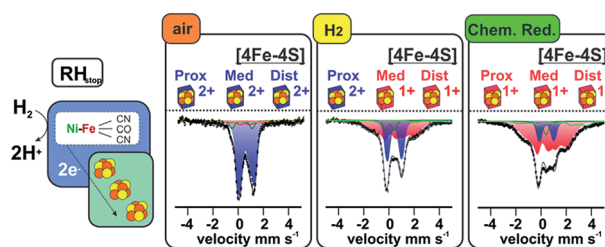


4495

## Cofactor composition and function of a H<sub>2</sub>-sensing regulatory hydrogenase as revealed by Mössbauer and EPR spectroscopy

Federico Roncaroli, Eckhard Bill,\* Bärbel Friedrich, Oliver Lenz, Wolfgang Lubitz\* and Maria-Eirini Pandelia\*

A regulatory hydrogenase is characterised by Mössbauer, EPR and FTIR yielding insight into structure and function of this dihydrogen sensor.

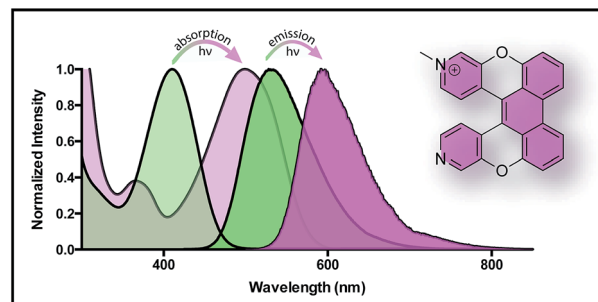


4508

## Synthesis and properties of lysosome-specific photoactivatable probes for live-cell imaging

Mai N. Tran, Robert-André F. Rarig and David M. Chenoweth\*

We describe the synthesis and application of a new class of large Stokes shift lysosome-specific photoactivatable probes for live-cell imaging.

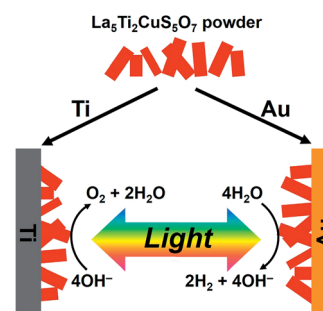


4513

## Photoanodic and photocathodic behaviour of La<sub>5</sub>Ti<sub>2</sub>CuS<sub>5</sub>O<sub>7</sub> electrodes in the water splitting reaction

Guijun Ma, Yohichi Suzuki, Rupashree Balia Singh, Aki Iwanaga, Yosuke Moriya, Tsutomu Minegishi, Jingyuan Liu, Takashi Hisatomi, Hiroshi Nishiyama, Masao Katayama, Kazuhiko Seki, Akihiro Furube, Taro Yamada and Kazunari Domen\*

La<sub>5</sub>Ti<sub>2</sub>CuS<sub>5</sub>O<sub>7</sub> embedded into the surface of Au and Ti substrates shows a photocurrent attributable to HER and OER.

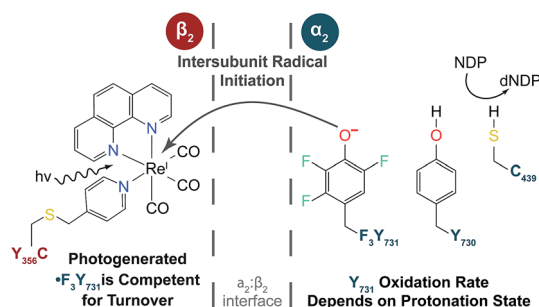


4519

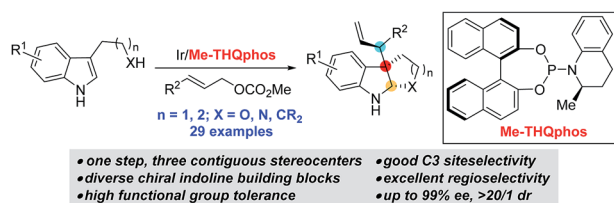
## Direct interfacial Y<sub>731</sub> oxidation in α<sub>2</sub> by a photoβ<sub>2</sub> subunit of *E. coli* class Ia ribonucleotide reductase

David Y. Song, Arturo A. Pizano, Patrick G. Holder, JoAnne Stubbe\* and Daniel G. Nocera\*

Proton-coupled electron transfer (PCET) is a fundamental mechanism important in a wide range of biological processes including the universal reaction catalysed by ribonucleotide reductases (RNRs) in making *de novo*, the building blocks required for DNA replication and repair.



4525

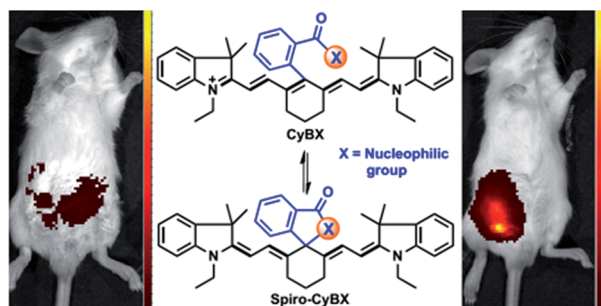


### Ligand-enabled Ir-catalyzed intermolecular diastereoselective and enantioselective allylic alkylation of 3-substituted indoles

Xiao Zhang, Wen-Bo Liu, Hang-Fei Tu and Shu-Li You\*

A ligand-enabled Ir-catalyzed diastereoselective and enantioselective allylic alkylation of 3-substituted indoles is reported, providing indoline products containing three contiguous stereocenters in one step with high site-, regio-, diastereo- and enantioselectivities from a wide range of readily available starting materials.

4530

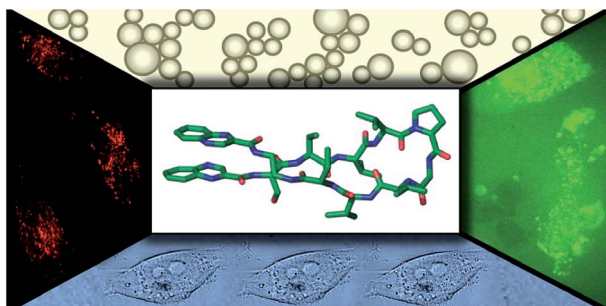


### A simple and effective "capping" approach to readily tune the fluorescence of near-infrared cyanines

Longwei He, Weiyang Lin,\* Qiuyan Xu, Mingguang Ren, Haipeng Wei and Jian-Yong Wang

A simple and effective capping approach was introduced to readily tune the fluorescence of NIR cyanines.

4537

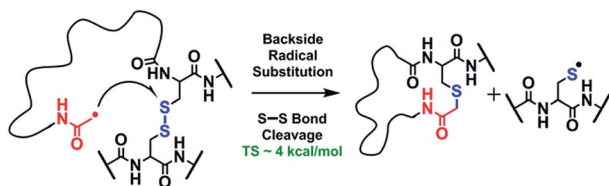


### A new quinoxaline-containing peptide induces apoptosis in cancer cells by autophagy modulation

Rubí Zamudio-Vázquez, Saška Ivanova, Miguel Moreno, María Isabel Hernandez-Alvarez, Ernest Giralt, Axel Bidon-Chanal, Antonio Zorzano,\* Fernando Albericio\* and Judit Tulla-Puche\*

The most cytotoxic compound from a library of quinoxaline-containing peptides is endocytosed into HeLa cells, accumulates in acidic compartments, and blocks autophagy by altering lysosomal function, leading to apoptosis activation.

4550



### Mechanisms and energetics of free radical initiated disulfide bond cleavage in model peptides and insulin by mass spectrometry

Chang Ho Sohn, Jinshan Gao, Daniel A. Thomas, Tae-Young Kim, William A. Goddard III and J. L. Beauchamp\*

Direct radical substitution at sulfur initiates disulfide bond cleavage by hydrogen-deficient radicals in peptides and proteins.

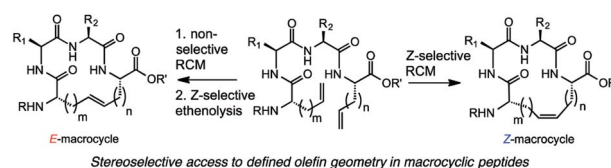


4561

### Stereoselective synthesis of macrocyclic peptides via a dual olefin metathesis and ethenolysis approach

Shane L. Mangold and Robert H. Grubbs\*

A metathesis strategy for controlling olefin geometry within macrocyclic peptides has been achieved using catalyst-directed RCM and ethenolysis.

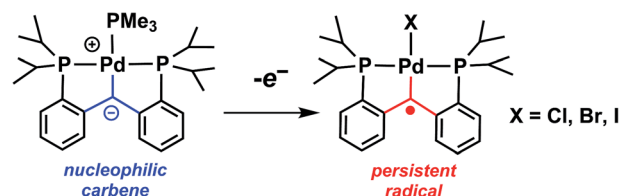


4570

### Palladium carbene complexes as persistent radicals

C. C. Comanescu, M. Vyushkova and V. M. Iluc\*

A series of palladium(II) persistent radical carbene complexes,  $[PC(sp^2)P]PdX$  ( $X = Cl, Br, I$ ), was synthesized from the nucleophilic carbene  $[PC(sp^2)P]PdPMe_3$ .

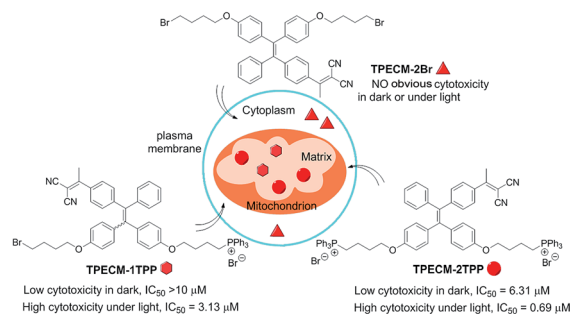


4580

### Image-guided combination chemotherapy and photodynamic therapy using a mitochondria-targeted molecular probe with aggregation-induced emission characteristics

Chong-Jing Zhang, Qinglian Hu, Guangxue Feng, Ruoyu Zhang, Youyong Yuan, Xianmao Lu and Bin Liu\*

Mitochondria-targeted AIE photosensitizers show multifunctions of targeted and image-guided combination chemotherapy and photodynamic therapy.

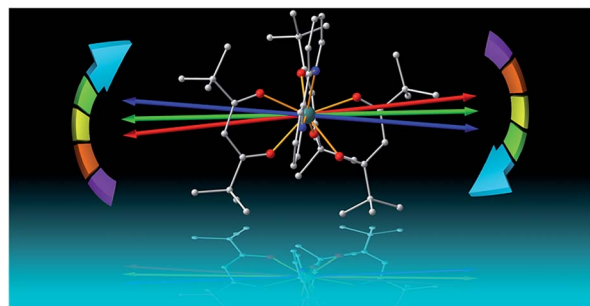


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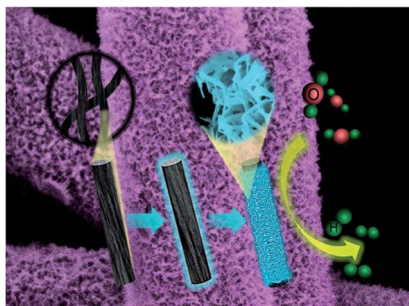
### Does the thermal evolution of molecular structures critically affect the magnetic anisotropy?

Kang Qian, José J. Baldoví, Shang-Da Jiang,\* Alejandro Gaita-Ariño,\* Yi-Quan Zhang, Jacob Overgaard, Bing-Wu Wang, Eugenio Coronado\* and Song Gao\*

In the absence of a critical phase transition, one can safely use the crystal structure information determined at liquid nitrogen temperature in magnetic anisotropy research.



4594

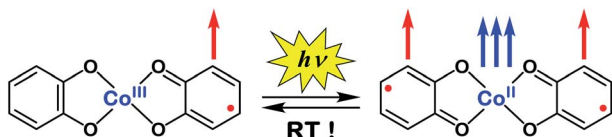


### Cobalt diselenide nanobelts grafted on carbon fiber felt: an efficient and robust 3D cathode for hydrogen production

Ya-Rong Zheng, Min-Rui Gao, Zi-You Yu, Qiang Gao, Huai-Ling Gao and Shu-Hong Yu\*

An easily scaled-up 3D CoSe<sub>2</sub>/CFF hierarchical electrode has been developed as a highly active and stable hydrogen evolution reaction cathode.

4599



### Bidirectional photoswitching of magnetic properties at room temperature: ligand-driven light-induced valence tautomerism

Alexander Witt, Frank W. Heinemann and Marat M. Khusniyarov\*

A unique molecular switch – a cobalt dioxolene complex featuring photoisomerizable ligands – allows unprecedented control of magnetic properties with light at room temperature.

4610

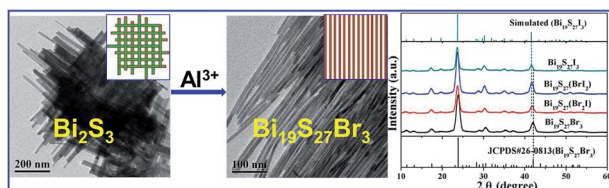


### Oxalyl amide assisted palladium-catalyzed synthesis of pyrrolidones via carbonylation of $\gamma$ -C(sp<sup>3</sup>)-H bonds of aliphatic amine substrates

Chao Wang, Li Zhang, Changpeng Chen, Jian Han, Yingming Yao\* and Yingsheng Zhao\*

The first Pd-catalyzed regioselective  $\gamma$ -carbonylation of oxalyl amide protected aliphatic amines with carbon monoxide leading to synthesis of pyrrolidones has been developed.

4615



### Shape and composition control of Bi<sub>19</sub>S<sub>27</sub>(Br<sub>3-x</sub>I<sub>x</sub>) alloyed nanowires: the role of metal ions

Yihui Wu, Huanhuan Pan, Xin Zhou, Mingrun Li, Bin Zhou, Chi Yang, Wen-Hua Zhang,\* Jiansheng Jie\* and Can Li\*

Highly uniform single-crystalline Bi<sub>19</sub>S<sub>27</sub>(Br<sub>3-x</sub>I<sub>x</sub>) alloyed nanowires (NWs) (0 ≤ x ≤ 3) were achieved for the first time. The NWs show a composition-independent band gap and have great application potential in optoelectronic devices.

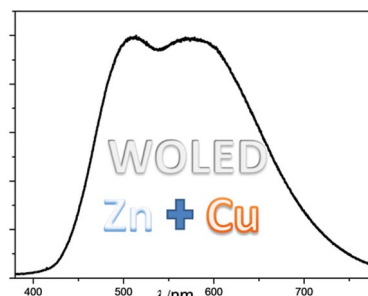


4623

### Luminescent zinc(II) and copper(I) complexes for high-performance solution-processed monochromatic and white organic light-emitting devices

G. Cheng, G. K.-M. So, W.-P. To, Y. Chen, C.-C. Kwok, C. Ma, X. Guan, X. Chang, W.-M. Kwok and C.-M. Che\*

High performance orange (EQE up to 15.64%) and white (EQE up to 6.88%) solution processed OLEDs fabricated solely with emitters of non-platinum group metals were reported. The white device has CIE coordinates of (0.42, 0.44) and CRI of 81.

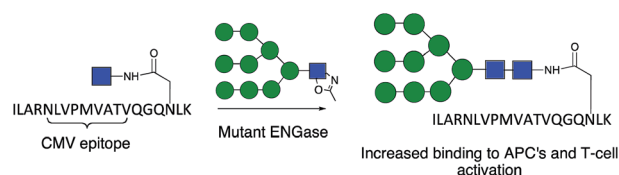


4636

### Convergent chemo-enzymatic synthesis of mannosylated glycopeptides; targeting of putative vaccine candidates to antigen presenting cells

Julie D. McIntosh, Margaret A. Brimble,\* Anna E. S. Brooks, P. Rod Dunbar,\* Renata Kowalczyk, Yusuke Tomabechi and Antony J. Fairbanks\*

Convergent chemo-enzymatic synthesis of mannosylated glycopeptides enhances uptake by human antigen presenting cells whilst preserving the immunogenicity of peptide epitopes.

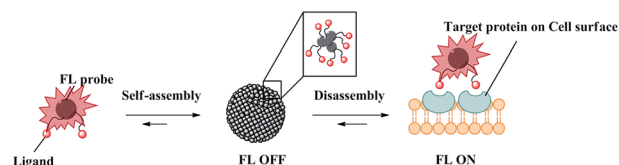


4643

### Near-infrared fluorescence activation probes based on disassembly-induced emission cyanine dye

Tai-Cheng Hou, Ying-Yi Wu, Po-Yi Chiang and Kui-Thong Tan\*

In the presence of target analyte, bright fluorescence in the near-IR region is emitted through the recognition-induced disassembly of the probe aggregate.

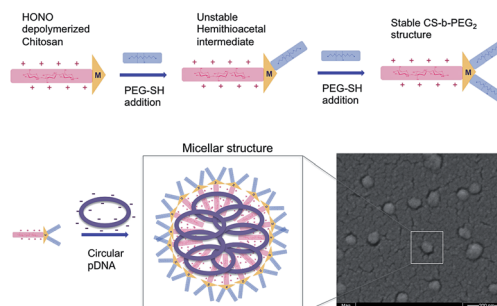


4650

### Regioselective thioacetylation of chitosan end-groups for nanoparticle gene delivery systems

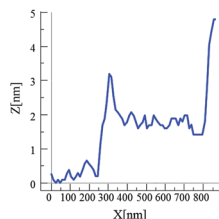
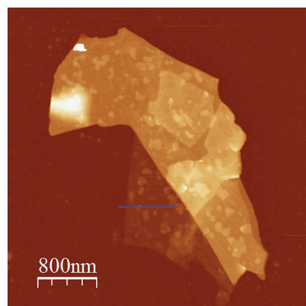
V. D. Pickenhahn, V. Darras, F. Dziopa, K. Biniecki, G. De Crescenzo, M. Lavertu\* and M. D. Buschmann\*

We present a novel, aqueous thiol-based conjugation strategy that constitutes an alternative to the oxime-click pathway for generating a reactive end-group on chitosan (CS), which could also be applicable to other polymers.





4665

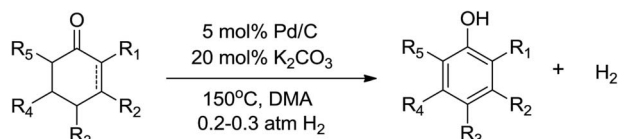


### Graphene related magnetic materials: micromechanical exfoliation of 2D layered magnets based on bimetallic anilate complexes with inserted $[\text{Fe}^{\text{III}}(\text{acac}_2\text{-trien})]^+$ and $[\text{Fe}^{\text{III}}(\text{sal}_2\text{-trien})]^+$ molecules

Alexandre Abhervé, Samuel Mañas-Valero, Miguel Clemente-León\* and Eugenio Coronado\*

The Scotch tape method has been used for the exfoliation of layered coordination compounds formed by a 2D bimetallic anilate-based anionic network and  $\text{Fe}(\text{III})$  cationic complexes placed between or within the layers.

4674

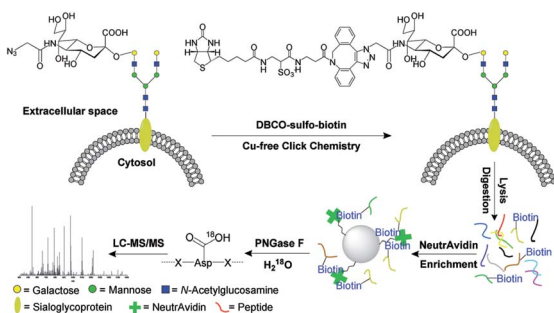


### Reaction-activated palladium catalyst for dehydrogenation of substituted cyclohexanones to phenols and $\text{H}_2$ without oxidants and hydrogen acceptors

Jingwu Zhang, Qiangqiang Jiang, Dejun Yang, Xiaomei Zhao, Yanli Dong and Renhua Liu\*

A combination of  $\text{Pd/C}$  and  $\text{H}_2$  is found to dehydrogenate a wide range of substituted cyclohexanones and 2-cyclohexenones to their corresponding phenols with high isolated yields, with  $\text{H}_2$  as the only byproduct.

4681

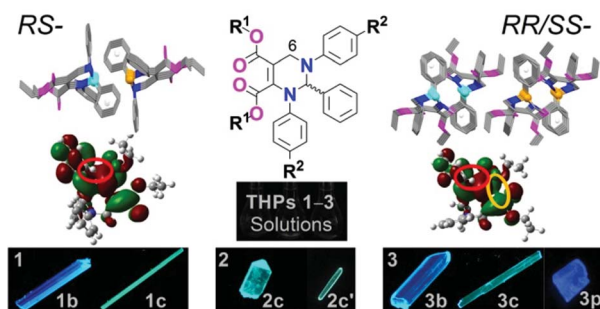


### Systematic and site-specific analysis of *N*-sialoglycosylated proteins on the cell surface by integrating click chemistry and MS-based proteomics

Weixuan Chen, Johanna M. Smeekens and Ronghu Wu\*

A method integrating metabolic labeling, copper-free click chemistry and MS-based proteomics is effective to globally and site-specifically analyze surface *N*-sialoglycoproteins.

4690



### Insight into the strong aggregation-induced emission of low-conjugated racemic C6-unsubstituted tetrahydropyrimidines through crystal-structure–property relationship of polymorphs

Qihua Zhu, Yilin Zhang, Han Nie, Zujin Zhao, Shuwen Liu,\* Kam Sing Wong\* and Ben Zhong Tang\*

Racemic low-conjugated non-emissive THPs 1–3 can form highly emissive *RS*- and *RR/SS*-packing polymorphs with mixed through-bond and through-space conjugation.

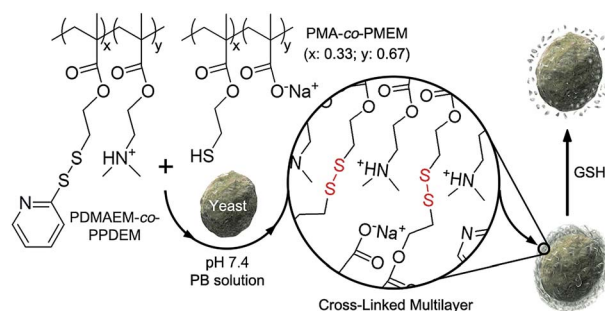


4698

### Cytocompatible *in situ* cross-linking of degradable LbL films based on thiol–exchange reaction

Sung Ho Yang,\* Jinsu Choi, L. Palanikumar, Eun Seong Choi, Juno Lee, Juan Kim, Insung S. Choi\* and Ja-Hyoung Ryu\*

A highly cytocompatible LbL process was developed, based on the thiol–exchange reaction, for forming *in situ* cross-linked and degradable films under physiologically mild conditions.

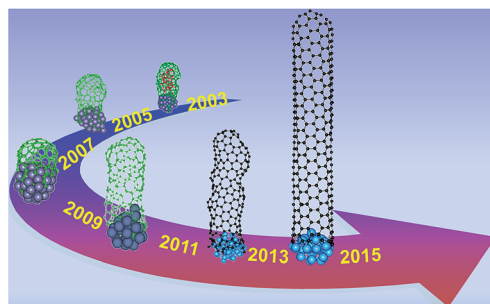


4704

### Atomistic simulation of the growth of defect-free carbon nanotubes

Ziwei Xu, Tianying Yan\* and Feng Ding\*

The atomistic simulation of defect-free SWCNT growth is realized for the first time after 12 years of continuous effort.

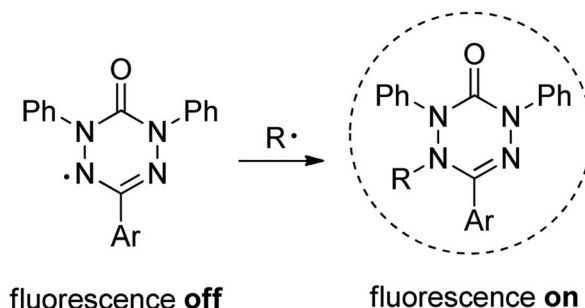


4712

### Profluorescent verdazyl radicals – synthesis and characterization

David Matuschek, Steffen Eusterwiemann, Linda Stegemann, Carsten Doerenkamp, Birgit Wibbeling, Constantin G. Daniliuc, Nikos L. Doltsinis,\* Cristian A. Strassert,\* Hellmut Eckert\* and Armido Studer\*

The synthesis and characterization of various 6-oxo-verdazyl radicals and their diamagnetic styryl radical trapping products are presented.

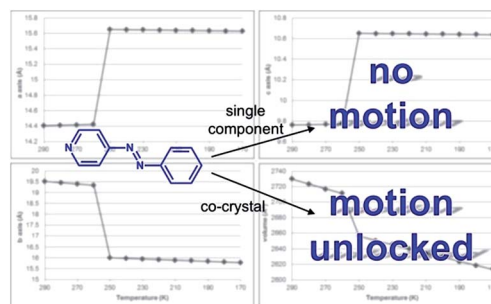


4717

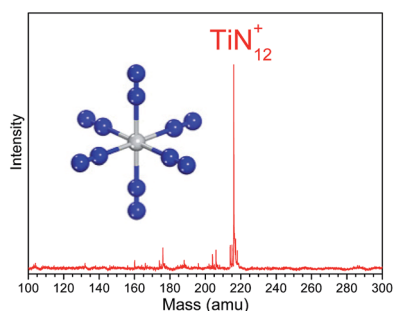
### Achieving dynamic behaviour and thermal expansion in the organic solid state *via* co-crystallization

Kristin M. Hutchins, Ryan H. Groeneman, Eric W. Reinheimer, Dale C. Swenson and Leonard R. MacGillivray\*

Molecular motion of an azo functional group is 'unlocked' *via* co-crystallizations.



4723

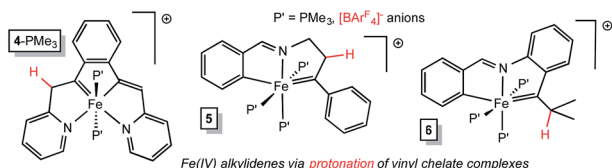


### Experimental observation of $\text{TiN}_{12}^+$ cluster and theoretical investigation of its stable and metastable isomers

Ke-Wei Ding, Xiao-Wei Li, Hong-Guang Xu, Tao-Qi Li,\*  
Zhong-Xue Ge,\* Qian Wang\* and Wei-Jun Zheng\*

In the  $O_h$  symmetric structure of  $\text{TiN}_{12}^+$ , the interaction between Ti and  $\text{N}_2$  weakens the N–N bond significantly.

4730

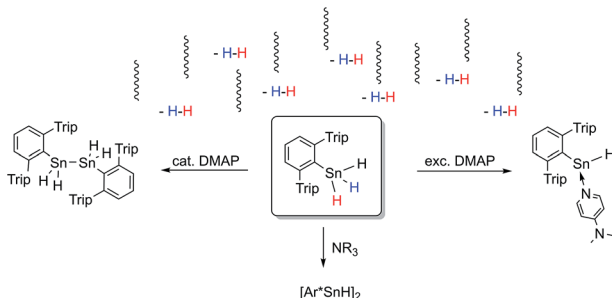


### Fe(IV) alkylidenes via protonation of Fe(II) vinyl chelates and a comparative Mössbauer spectroscopic study

Brian M. Lindley, Ala'aeddeen Swidan, Emil B. Lobkovsky,  
Peter T. Wolczanski,\* Mario Adelhardt, Jörg Sutter  
and Karsten Meyer

Fe(IV) alkylidenes are produced via protonation of Fe(II) vinyl chelate complexes.

4737

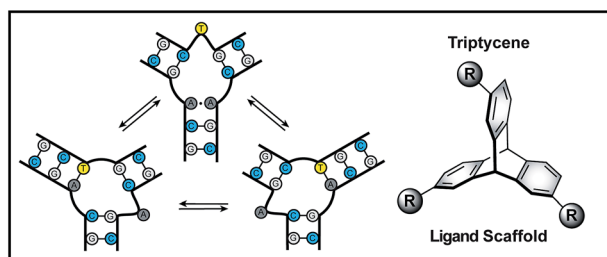


### A nitrogen-base catalyzed generation of organotin(II) hydride from an organotin trihydride under reductive dihydrogen elimination

Christian P. Sindlinger, Andreas Stasch, Holger F. Bettinger  
and Lars Wesemann\*

Amine bases are shown to induce reductive elimination of dihydrogen from terphenyltin trihydride.

4752



### Triptycene-based small molecules modulate (CAG)·(CTG) repeat junctions

Stephanie A. Barros and David M. Chenoweth\*

A triptycene-based scaffold is used to develop a new class of ligands for modulating the structure of junction forming trinucleotide repeat expansion sequences.

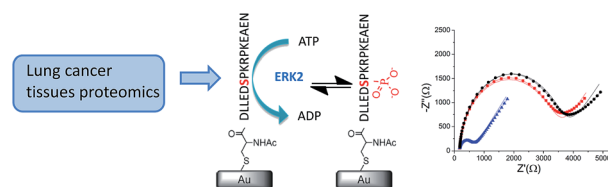


4756

### Integrating proteomics with electrochemistry for identifying kinase biomarkers

Einav Amit, Rofeamor Obena, Yi-Ting Wang, Roman Zhuravel, Aaron James F. Reyes, Shir Elbaz, Dvir Rotem, Danny Porath, Assaf Friedler,\* Yu-Ju Chen\* and Shlomo Yitzchaik\*

We present an integrated approach for highly sensitive identification and validation of substrate-specific kinases as cancer biomarkers.

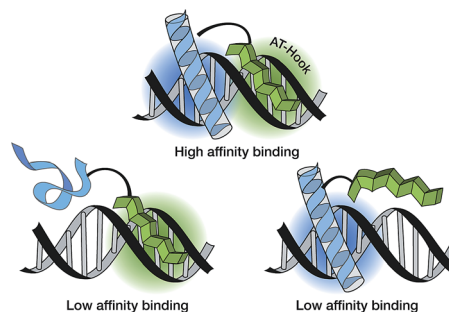


4767

### The AT-Hook motif as a versatile minor groove anchor for promoting DNA binding of transcription factor fragments

Jéssica Rodríguez, Jesús Mosquera, Jose R. Couceiro, M. Eugenio Vázquez\* and José L. Mascareñas\*

We report the development of chimeric DNA binding peptides comprising a DNA binding fragment of natural transcription factors (the basic region of a bZIP protein or a monomeric zinc finger module) and an AT-Hook peptide motif.

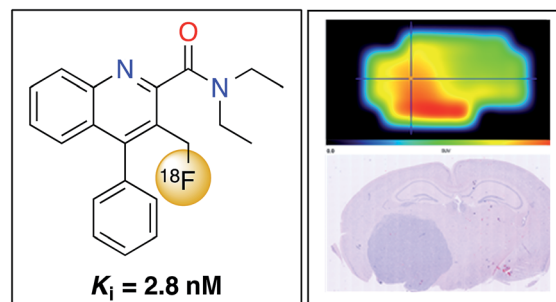


4772

### A novel $^{18}\text{F}$ -labelled high affinity agent for PET imaging of the translocator protein

Adele Blair, Filip Zmuda, Gaurav Malviya, Adriana A. S. Tavares, Gilles D. Tamagnan, Anthony J. Chalmers, Deborah Dewar, Sally L. Pimlott and Andrew Sutherland\*

A novel  $^{18}\text{F}$ -labelled quinoline-2-carboxamide has been characterised as a novel PET imaging agent for the translocator protein.

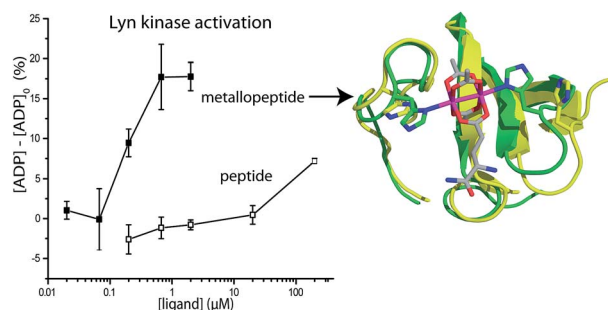


4778

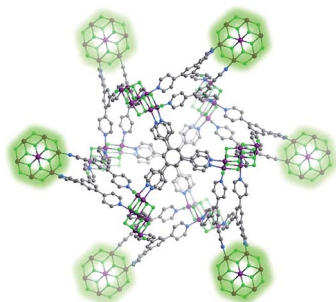
### Potent and selective inhibition of SH3 domains with dirhodium metalloinhibitors

Farrukh Vohidov, Sarah E. Knudsen, Paul G. Leonard, Jun Ohata, Michael J. Wheadon, Brian V. Popp, John E. Ladbury and Zachary T. Ball\*

Specific, designed histidine–rhodium interactions allow a metallopeptide to bind Lyn kinase with nanomolar affinity and to activate kinase activity.



4784

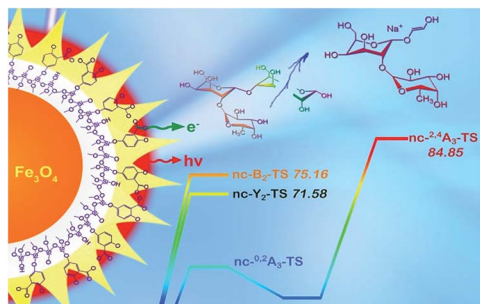


### Double-walled pyr topology networks from a novel fluoride-bridged heptanuclear metal cluster

Kai-Jie Chen, John J. Perry IV, Hayley S. Scott, Qing-Yuan Yang and Michael J. Zaworotko\*

Two isostructural networks with double-walled pyr topology comprised of novel fluoride-bridged heptanuclear metal clusters and 3-connected ligands have been synthesized and characterized by X-ray diffraction, thermogravimetric analysis, and gas sorption experiments.

4790

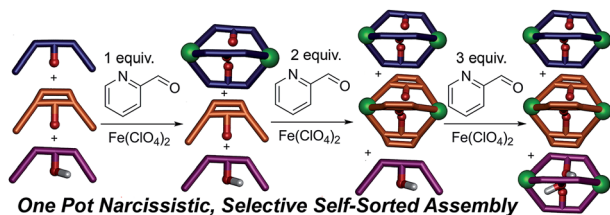


### UV-activated multilayer nanomatrix provides one-step tunable carbohydrate structural characterization in MALDI-MS

Rofeamor P. Obena, Mei-Chun Tseng, Indah Primadona, Jun Hsiao, I-Che Li, Rey Y. Capangpangan, Hsiu-Fong Lu, Wan-Sheung Li, Ito Chao, Chun-Cheng Lin and Yu-Ju Chen\*

Our work highlights DHB@MNP-induced pseudo-MS/MS for oligosaccharide characterization, with some insights on this nanoparticle-mediated energy transfer dynamics.

4801

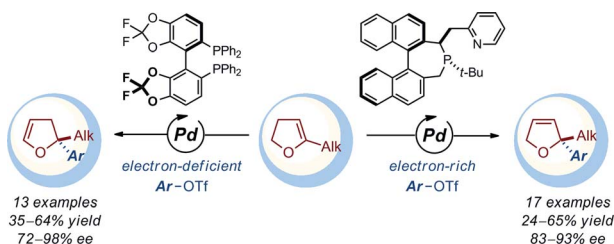


### High fidelity sorting of remarkably similar components via metal-mediated assembly

Lauren R. Holloway, Michael C. Young, Gregory J. O. Beran and Richard J. Hooley\*

Subtle differences in coordination angle and rigidity lead to narcissistic self-sorting between highly similar individual components upon metal-mediated assembly.

4807



### Access to enantioenriched 2,3- and 2,5-dihydrofurans with a fully substituted C2 stereocenter by Pd-catalyzed asymmetric intermolecular Heck reaction

Gustavo M. Borrajo-Calleja, Vincent Bizet, Thomas Bürgi and Clément Mazet\*

A palladium catalyzed intermolecular asymmetric Heck reaction provides access to valuable 2,3- and 2,5-dihydrofurans with a fully substituted C2 stereocenter with high levels of regio- and enantiocontrol.



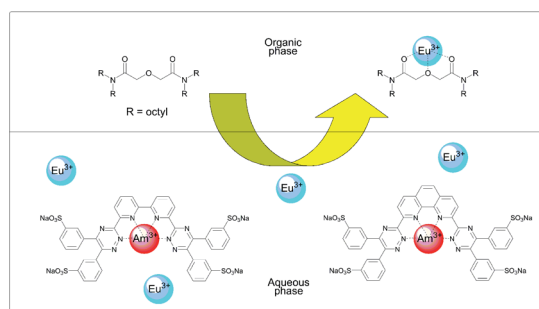


4812

## Hydrophilic sulfonated bis-1,2,4-triazine ligands are highly effective reagents for separating actinides(III) from lanthanides(III) via selective formation of aqueous actinide complexes

Frank W. Lewis,\* Laurence M. Harwood,\*  
Michael J. Hudson, Andreas Geist, Valery N. Kozhevnikov,  
Petr Distler and Jan John

Tetrasulfonated bis-1,2,4-triazine ligands can selectively complex and separate actinides from lanthanides in aqueous nitric acid with very high selectivities.

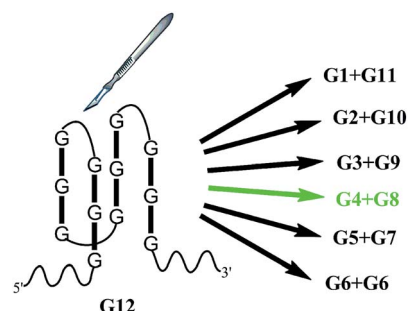


4822

## How to split a G-quadruplex for DNA detection: new insight into the formation of DNA split G-quadruplex

Jinbo Zhu, Libing Zhang, Shaojun Dong  
and Erkan Wang\*

A magic "law of 4 : 8" to split the G-quadruplex for DNA detection has been found.

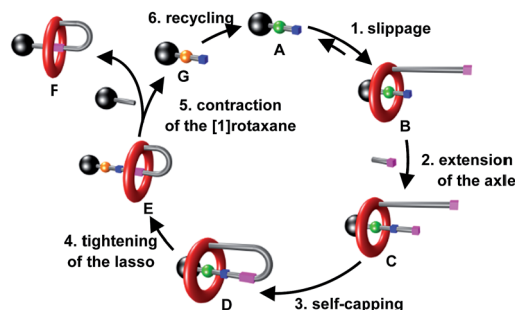


4828

## Synthesis of triazolium-based mono- and tris-branched [1]rotaxanes using a molecular transporter of dibenzo-24-crown-8

P. Waelès, C. Clavel, K. Fournel-Marotte and F. Coutrot\*

A general synthesis to mono- and multi-branched [1]rotaxanes that are devoid of any efficient template is reported.

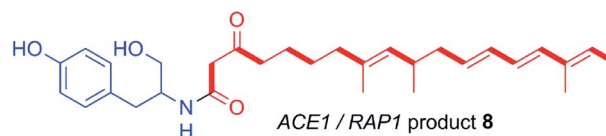


4837

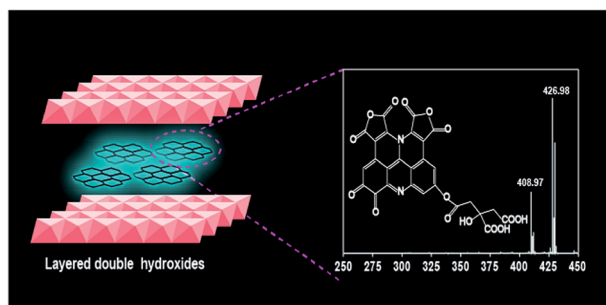
# Heterologous expression of the avirulence gene *ACE1* from the fungal rice pathogen *Magnaporthe oryzae*

Z. Song, W. Bakeer, J. W. Marshall, A. A. Yakasai,  
R. M. Khalid, J. Collemare, E. Skellam, D. Tharreau,  
M.-H. Lebrun, C. M. Lazarus, A. M. Bailey, T. J. Simpson  
and R. J. Cox\*

Heterologous expression of key components of the *Magnaporthe grisea* *ACE1* gene cluster produces a potential precursor of cryptic avirulence signalling compounds that induce resistance to *M. grisea* in rice.



4846

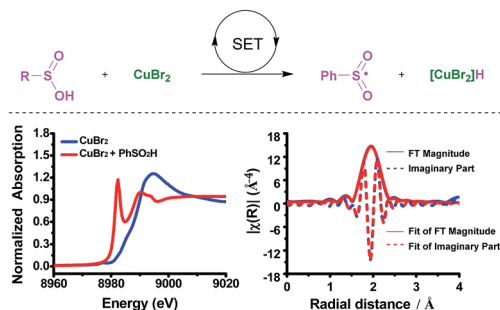


### Structure observation of graphene quantum dots by single-layered formation in layered confinement space

Liqing Song, Jingjing Shi, Jun Lu and Chao Lu\*

We observe the structure of single-layered graphene quantum dots prepared in the 2D confined space of layered double hydroxides.

4851

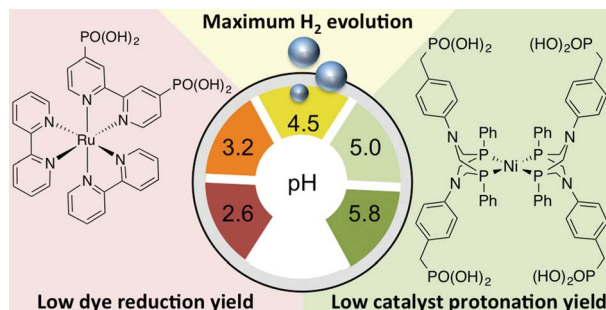


### Operando X-ray absorption and EPR evidence for a single electron redox process in copper catalysis

Qingquan Lu, Jian Zhang, Pan Peng, Guanghui Zhang, Zhiliang Huang, Hong Yi, Jeffrey T. Miller and Aiwen Lei\*

A single electron redox process between Cu(II) and a sulfinic acid, and characterization of the formed Cu(I) are clearly shown using *operando* X-ray absorption and EPR evidence.

4855

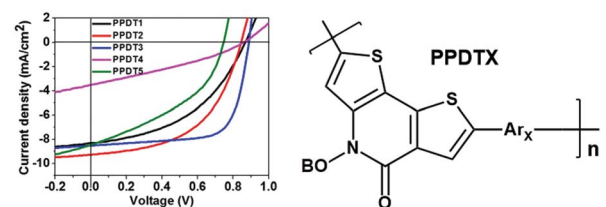


### Unravelling the pH-dependence of a molecular photocatalytic system for hydrogen production

Anna Reynal,\* Ernest Pastor, Manuela A. Gross, Shababa Selim, Erwin Reisner\* and James R. Durrant\*

The electron-donating ability of the sacrificial agent and the protonation of the catalyst determine the optimum pH for hydrogen production.

4860



### Wide bandgap OPV polymers based on pyridinedithiophene unit with efficiency >5%

Alexander M. Schneider, Luyao Lu, Eric F. Manley, Tianyue Zheng, Valerii Sharapov, Tao Xu, Tobin J. Marks, Lin X. Chen\* and Luping Yu\*

We report the properties of a new series of wide band gap photovoltaic polymers based on the *N*-alkyl 2-pyridone dithiophene (PDT) unit.

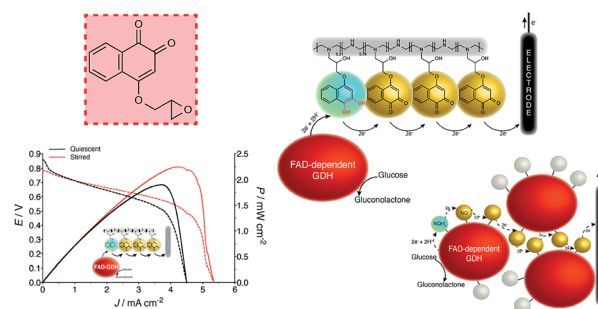


4867

## Rational design of quinones for high power density biofuel cells

Ross D. Milton, David P. Hickey, Sofiene Abdellaoui, Koun Lim, Fei Wu, Boxuan Tan and Shelley D. Minteer\*

Rationally designing quinones to label GDH and create a redox hydrogel that delivers high OCP, current and power densities.

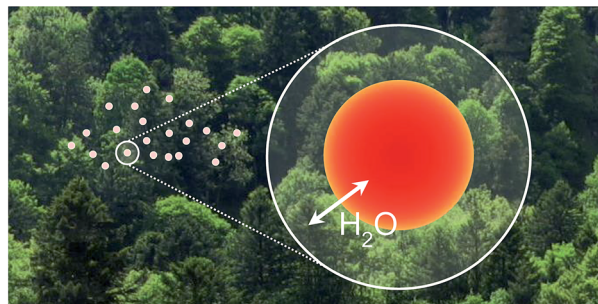


4876

## Water diffusion in atmospherically relevant $\alpha$ -pinene secondary organic material

Hannah C. Price,\* Johan Mattsson, Yue Zhang, Allan K. Bertram, James F. Davies, James W. Grayson, Scot T. Martin, Daniel O'Sullivan, Jonathan P. Reid, Andrew M. J. Rickards and Benjamin J. Murray\*

We report the first direct measurements of water diffusion coefficients in secondary organic aerosol.

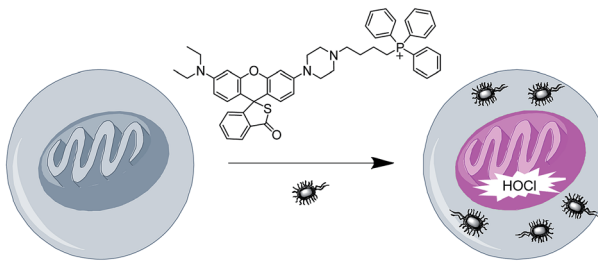


4884

## HOCl can appear in the mitochondria of macrophages during bacterial infection as revealed by a sensitive mitochondrial-targeting fluorescent probe

Jin Zhou, Lihong Li, Wen Shi,\* Xinghui Gao, Xiaohua Li and Huimin Ma\*

HOCl can appear in the mitochondria of macrophages during bacterial infection as revealed by a new sensitive mitochondrial-targeting fluorescent probe.

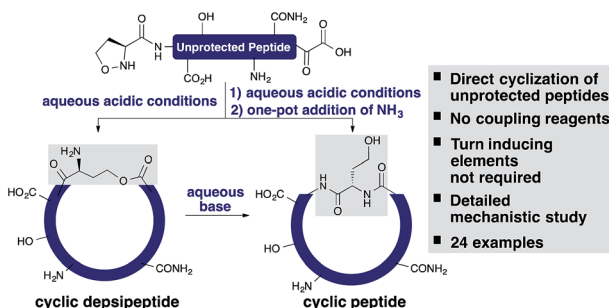


4889

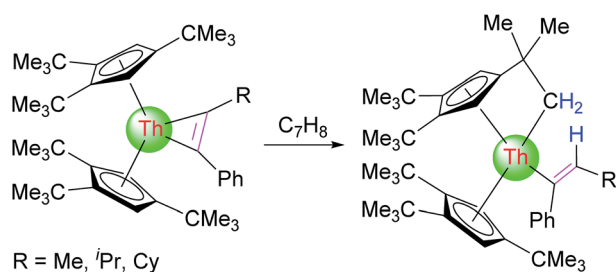
## Spontaneous head-to-tail cyclization of unprotected linear peptides with the KAHA ligation

Florian Rohrbacher, Gildas Deniau, Anatol Luther and Jeffrey W. Bode\*

The  $\alpha$ -ketoacid-hydroxylamine (KAHA) ligation enables the direct cyclization of unprotected peptides upon cleavage, without coupling reagents or purification of precursors. We report the synthesis of a library of 24 cyclic peptides and a detailed mechanistic study.



4897

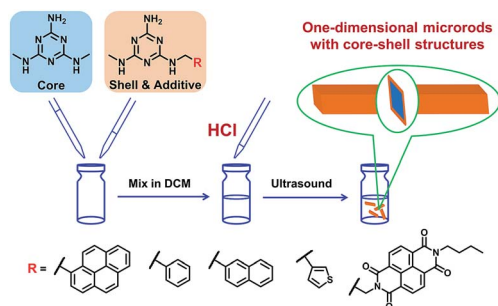


### C–H bond activation induced by thorium metallacyclopentadiene complexes: a combined experimental and computational study

Bo Fang, Lei Zhang, Guohua Hou, Guofu Zi,<sup>\*</sup> De-Cai Fang<sup>\*</sup> and Marc D. Walter<sup>\*</sup>

Thorium metallacyclopentadienes derived from phenyl(alkyl) acetylenes are very reactive complexes that undergo selective intramolecular C–H bond activation.

4907

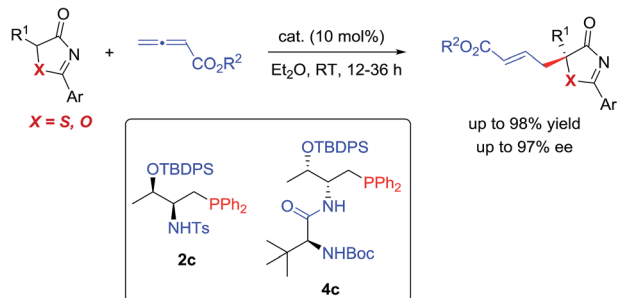


### Self-assembling 1D core/shell microrods by the introduction of additives: a one-pot and shell-tunable method

Jun Xu, Hongde Yu, Liulin Yang, Guanglu Wu, Zhiqiang Wang, Dong Wang and Xi Zhang<sup>\*</sup>

A one-pot method for the fabrication of 1D core/shell microrods with tunable shell compositions by the introduction of additives.

4912

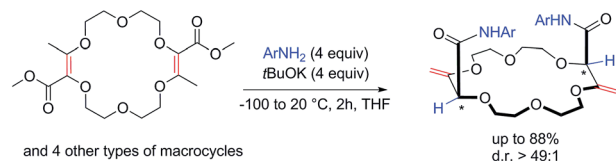


### Highly enantioselective construction of tertiary thioethers and alcohols via phosphine-catalyzed asymmetric $\gamma$ -addition reactions of 5H-thiazol-4-ones and 5H-oxazol-4-ones: scope and mechanistic understandings

Tianli Wang, Zhaoyuan Yu, Ding Long Hoon, Kuo-Wei Huang, Yu Lan<sup>\*</sup> and Yixin Lu<sup>\*</sup>

A new method for facile access to enantioenriched tertiary thioethers/alcohols.

4923



### Remote stereoselective deconjugation of $\alpha,\beta$ -unsaturated esters by simple amidation reactions

Mahesh Vishe, Radim Hrdina, Amalia I. Poblador-Bahamonde, Céline Besnard, Laure Guénée, Thomas Bürgi and Jérôme Lacour<sup>\*</sup>

The amidation of macrocyclic conjugated esters affords in one-pot single (chiral)  $\beta,\gamma$ -unsaturated diastereomers via effective remote stereocontrol.

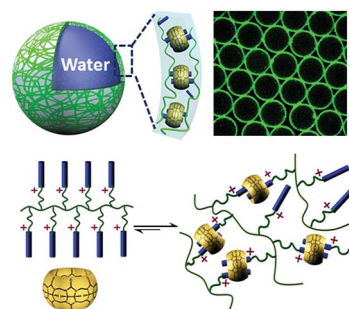


4929

### Supramolecular hydrogel microcapsules *via* cucurbit[8]uril host–guest interactions with triggered and UV-controlled molecular permeability

Ziyi Yu, Jing Zhang, Roger J. Coulston, Richard M. Parker, Frank Biedermann, Xin Liu, Oren A. Scherman\* and Chris Abell\*

Host–guest assembly at the interface of microfluidic droplets offers a versatile strategy to construct supramolecular hydrogel microcapsules with “smart” cargo release.

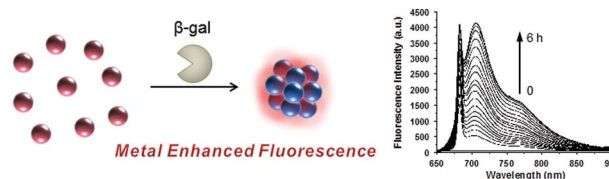


4934

### An enzyme-responsive metal-enhanced near-infrared fluorescence sensor based on functionalized gold nanoparticles

Zhanghua Zeng, Shin Mizukami, Katsumasa Fujita and Kazuya Kikuchi\*

An enzyme-responsive NIR nanosystem based on MEF was fabricated by surface functionalization of gold nanoparticles. Sensors based on this strategy are promising for enzyme detection in early diagnostic imaging and *in vivo* applications.

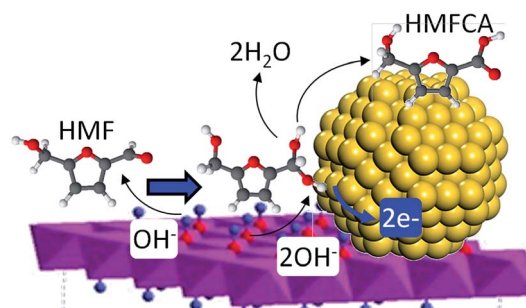


4940

### Solid base catalysed 5-HMF oxidation to 2,5-FDCA over Au/hydrotalcites: fact or fiction?

Leandro Ardemani, Giannantonio Cibir, Andrew J. Dent, Mark A. Isaacs, Georgios Kyriakou, Adam F. Lee,\* Christopher M. A. Parlett, Stephen A. Parry and Karen Wilson\*

Synergistic effects between alkali-free hydrotalcites and gold nanoparticles afford efficient heterogeneous catalysts for the cascade oxidation of 5-HMF to 2,5-FDCA.

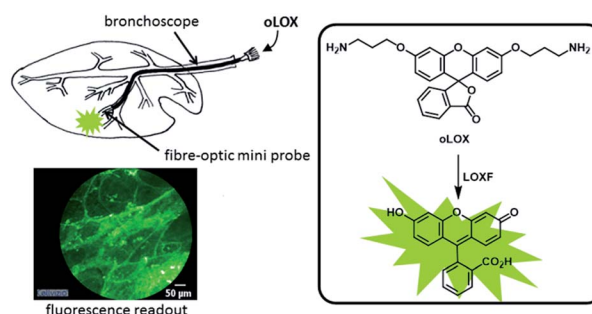


4946

### Optical molecular imaging of lysyl oxidase activity – detection of active fibrogenesis in human lung tissue

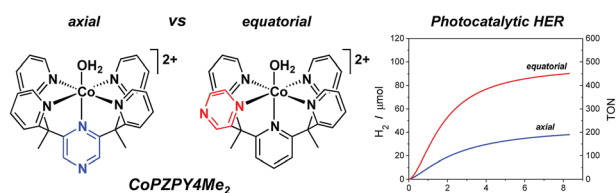
Tashfeen Aslam, Amy Miele, Sunay V. Chankeshwara, Alicia Megia-Fernandez, Chesney Michels, Ahsan R. Akram, Neil McDonald, Nik Hirani, Chris Haslett, Mark Bradley\* and Kevin Dhaliwal\*

A fluorogenic probe provides real-time measurement of lysyl oxidase activity in *ex vivo* asinine and human lung tissue.





4954

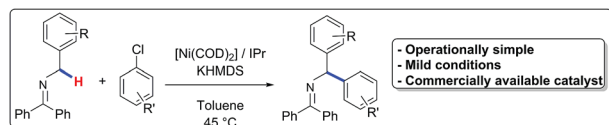


### Bioinspired design of redox-active ligands for multielectron catalysis: effects of positioning pyrazine reservoirs on cobalt for electro- and photocatalytic generation of hydrogen from water

Jonah W. Jurss, Rony S. Khnayzer, Julien A. Panetier, Karim A. El Roz, Eva M. Nichols, Martin Head-Gordon,<sup>\*</sup> Jeffrey R. Long,<sup>\*</sup> Felix N. Castellano<sup>\*</sup> and Christopher J. Chang<sup>\*</sup>

We report the effects of installing redox-active pyrazines at distinct positions in a series of isostructural Co catalysts.

4973

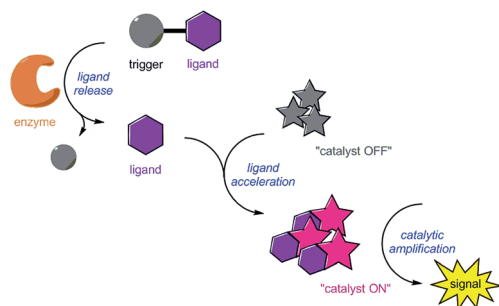


### Synthesis of (diarylmethyl)amines using Ni-catalyzed arylation of C(sp<sup>3</sup>)-H bonds

José A. Fernández-Salas, Enrico Marelli and Steven P. Nolan<sup>\*</sup>

The first nickel catalyzed deprotonative cross coupling between C(sp<sup>3</sup>)-H bonds and aryl chlorides is reported, allowing the challenging arylation of benzylamines in the absence of directing group or stoichiometric metal activation.

4978

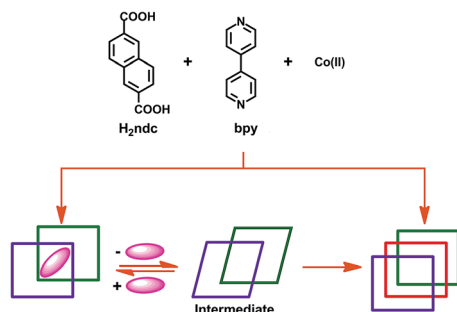


### Signal transduction and amplification through enzyme-triggered ligand release and accelerated catalysis

Sean Goggins, Barrie J. Marsh, Anneke T. Lubben and Christopher G. Frost<sup>\*</sup>

An enzyme-triggered catalytic signal amplification cascade is described through the design of a novel enzyme substrate that selectively activates an organometallic transfer hydrogenation catalyst once triggered.

4986



### Isolation of a structural intermediate during switching of degree of interpenetration in a metal-organic framework

Himanshu Aggarwal, Raj Kumar Das, Prashant M. Bhatt and Leonard J. Barbour<sup>\*</sup>

A structural intermediate has been isolated for the first time during switching of interpenetration from twofold to threefold in the MOF [Co<sub>2</sub>(ndc)<sub>2</sub>(bpy)].

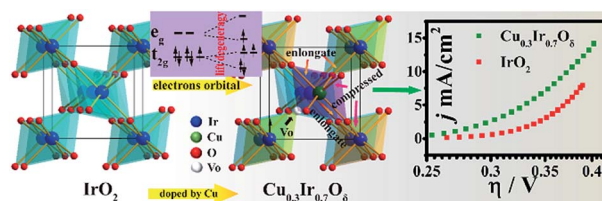


4993

## An efficiently tuned d-orbital occupation of IrO<sub>2</sub> by doping with Cu for enhancing the oxygen evolution reaction activity

Wei Sun, Ya Song, Xue-Qing Gong,\* Li-mei Cao and Ji Yang\*

Tuning Ir d-orbital occupation *via* doping Cu into the IrO<sub>2</sub> lattice to prepare a highly efficient oxygen evolution reaction catalyst, Cu<sub>0.3</sub>Ir<sub>0.7</sub>O<sub>6</sub>.

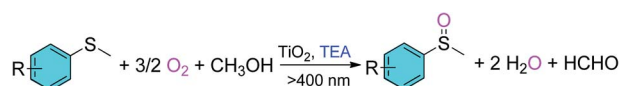


5000

## Tertiary amine mediated aerobic oxidation of sulfides into sulfoxides by visible-light photoredox catalysis on TiO<sub>2</sub>

Xianjun Lang, Wei Hao, Wan Ru Leow, Shuzhou Li,\* Jincai Zhao and Xiaodong Chen\*

The selective aerobic oxidation of sulfides into sulfoxides on TiO<sub>2</sub> under visible-light irradiation was accomplished through synergistic catalysis with triethylamine.

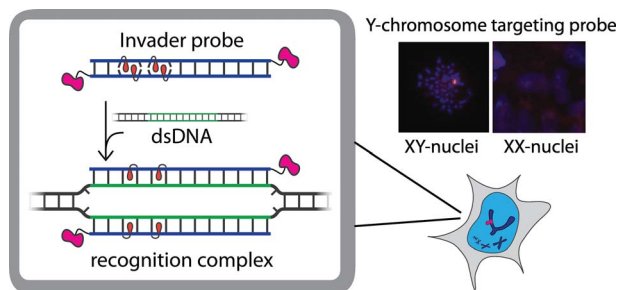


5006

## Invader probes: harnessing the energy of intercalation to facilitate recognition of chromosomal DNA for diagnostic applications

Dale C. Guenther, Grace H. Anderson, Saswata Karmakar, Brooke A. Anderson, Bradley A. Didion, Wei Guo, John P. Verstegen and Patrick J. Hrdlicka\*

Optimized Invader probes enable efficient ( $C_{50} < 1 \mu\text{M}$ ), fast ( $t_{50} < 3 \text{ h}$ ), kinetically stable ( $>24 \text{ h}$ ), and single nucleotide specific recognition of DNA targets.

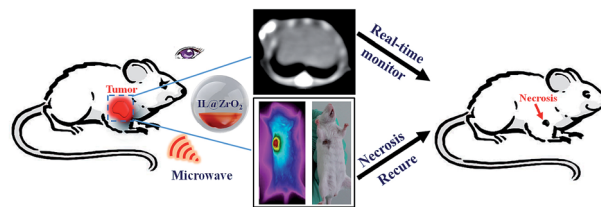


5016

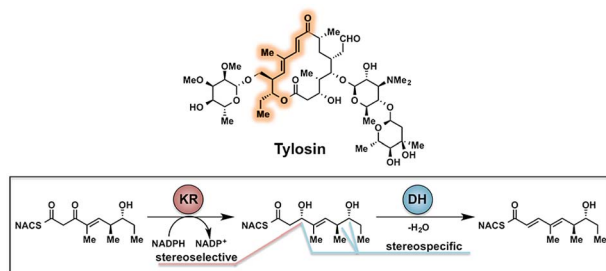
## A smart all-in-one theranostic platform for CT imaging guided tumor microwave thermotherapy based on IL@ZrO<sub>2</sub> nanoparticles

Haitang Shi, Meng Niu, Longfei Tan, Tianlong Liu, Haibo Shao, Changhui Fu, Xiangling Ren, Tengchuang Ma, Jun Ren, Linlin Li, Huiyu Liu, Ke Xu,\* Jianxin Wang, Fangqiong Tang and Xianwei Meng\*

This paper develops a simple multifunctional theranostic platform using an IL@ZrO<sub>2</sub> nanostructure for CT imaging guided tumor microwave thermotherapy.



5027

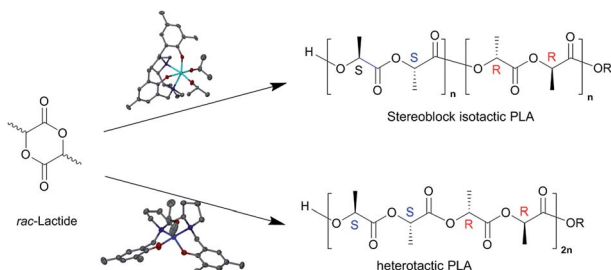


### Tylosin polyketide synthase module 3: stereospecificity, stereoselectivity and steady-state kinetic analysis of $\beta$ -processing domains *via* diffusible, synthetic substrates

William D. Fiers, Greg J. Dodge, Yang Li, Janet L. Smith, Robert A. Fecik\* and Courtney C. Aldrich\*

Natural and modified substrates coupled with LC-MS/MS analysis of products revealed the stereospecificity and stereoselectivity of a polyketide didomain.

5034

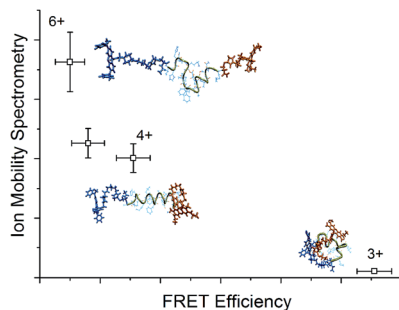


### Metal influence on the iso- and hetero-selectivity of complexes of bipyrrrolidine derived salan ligands for the polymerisation of *rac*-lactide

Matthew D. Jones,\* Lauren Brady, Paul McKeown, Antoine Buchard, Pascal M. Schäfer, Lynne H. Thomas, Mary F. Mahon, Timothy J. Woodman and John P. Lowe

A series complexes based on 2,2'-bipyrrrolidine based salan ligands have been prepared and either isotactic or heterotactic PLA have been prepared.

5040

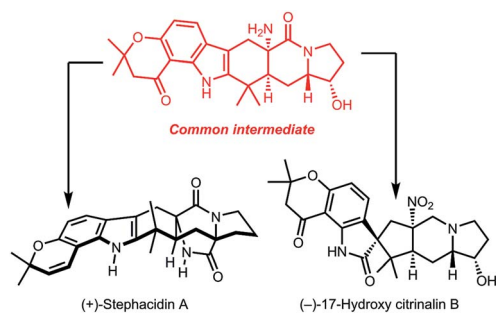


### Conformational changes in amyloid-beta (12–28) alloforms studied using action-FRET, IMS and molecular dynamics simulations

Steven Daly, Alexander Kulesza, Frederic Poussigue, Anne-Laure Simon, Chang Min Choi, Geoffrey Knight, Fabien Chirot, Luke MacAleese, Rodolphe Antoine and Philippe Dugourd\*

The gas phase conformations of two amyloid beta mutants are studied by multiple techniques to elucidate the origin of the different aggregation behaviour.

5048



### Unified approach to prenylated indole alkaloids: total syntheses of (–)-17-hydroxy-citrinalin B, (+)-stephacidin A, and (+)-notoamide I

Eduardo V. Mercado-Marin and Richmond Sarpong\*

The first strategy that provides reverse-prenylated indole alkaloids that bear a characteristic bicyclo[2.2.2]diazaoctane as well as those that lack this structural motif is reported.

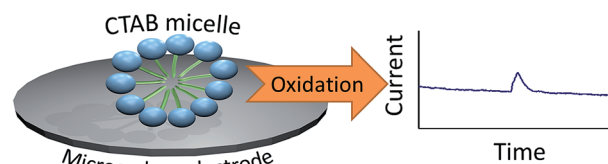


5053

### Electrochemical detection of single micelles through 'nano-impacts'

H. S. Toh and R. G. Compton\*

CTAB (cetyltrimethylammonium bromide) micelles are detected directly via the novel electrochemical method of 'nano-impacts' through oxidation of its bromide content.

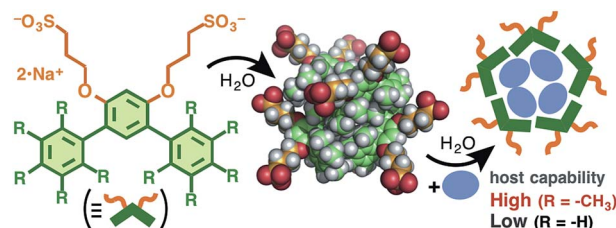


5059

### Well-defined aqueous nanoassemblies from amphiphilic meta-terphenyls and their guest incorporation

Yusuke Okazawa, Kei Kondo, Munetaka Akita and Michito Yoshizawa\*

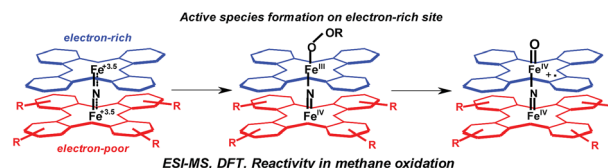
Spherical molecular assemblies with diameters of  $\sim 2$  nm were quantitatively formed in water from new amphiphilic meta-terphenyls and the nanoassembly with methyl groups provides superior host capability for fluorescent dyes.



5063

### Site-selective formation of an iron(IV)–oxo species at the more electron-rich iron atom of heteroleptic $\mu$ -nitrido diiron phthalocyanines

Ümit Işci, Abayomi S. Faponle, Pavel Afanasiev, Florian Albrieux, Valérie Briois, Vefa Ahsen, Fabienne Dumoulin\*, Alexander B. Sorokin\* and Sam P. de Visser\*



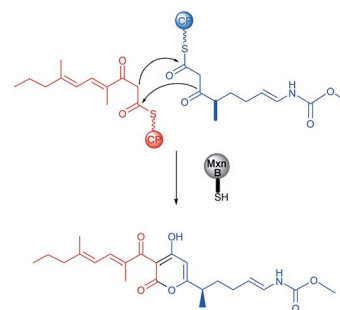
A combination of MS and computation on  $\mu$ -nitrido bridged diiron complexes reveals  $\text{H}_2\text{O}_2$  binding to the complex and generates an oxidant capable of oxidizing methane.

5076

### *In vitro* reconstitution of $\alpha$ -pyrone ring formation in myxopyronin biosynthesis

H. Sucipto, J. H. Sahner, E. Prusov, S. C. Wenzel, R. W. Hartmann, J. Koehnke\* and R. Müller\*

$\alpha$ -Pyrone rings exist in many polyketide synthase (PKS) derived natural products. We report the first *in vitro* reconstitution of  $\alpha$ -pyrone ring formation by a type I PKS using chemically synthesized substrates.



## 5086

## Hyukin Kwon, Wei Jiang and Eric T. Kool\*

## 5087

Chun Y. Chow, Hélène Bolvin, Victoria E. Campbell, Régis Guillot, Jeff W. Kampf, Wolfgang Wernsdorfer, Frédéric Gendron, Jochen Autschbach, Vincent L. Pecoraro\* and Talal Mallah\*

## 5088

## Qiao Song, Fei Li, Zhiqiang Wang and Xi Zhang\*

## 5090

Dali Wang, Chunlai Tu, Yue Su, Chuan Zhang, Udo Greiser, Xinyuan Zhu,\* Deyue Yan and Wenxin Wang\*