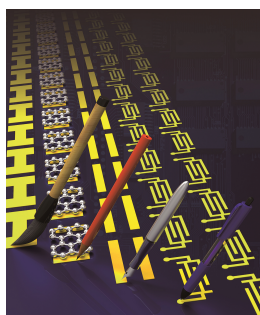


# ADVANCED FUNCTIONAL MATERIALS

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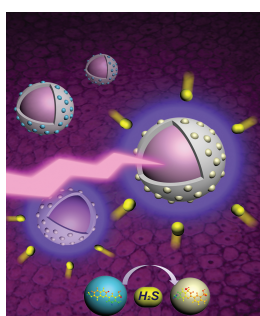
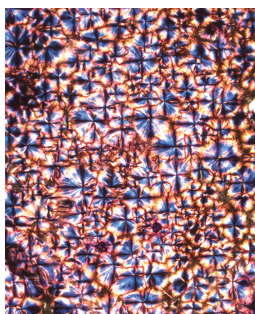


## Pen-Based Writing

Due to the emerging demand for methods of fabricating electronics in a cost-effective, easily accessible way that allows for rapid prototyping way, novel techniques to design and manufacture electronics are required. On page 165, F. Li, F. Xu, and co-workers review the recent innovations allowing ubiquitously available pens to be employed to directly write electronics, as inspired by the daily act of hand writing.

## Disease Diagnostics

With worldwide epidemic outbreaks occurring on a regular basis, there is urgent need to improve and establish new strategies toward rapid, simple, and affordable detection of target analytes. Birefringence generated from versatile lipid based liquid crystal cubic phase leads to a unique optical signal that can be used to detect analytes of medical interest, including bacteria, viruses, and parasites, as shown by R. Mezzenga and co-workers on page 181.

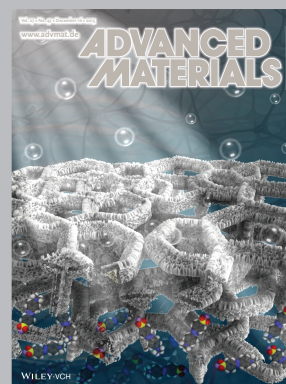
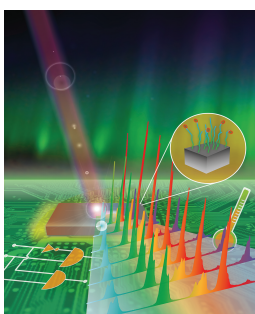


## Sensors

Hydrogen sulfide is regarded as one of the most significant gas signaling molecules for a variety of physiological and pathological processes. On page 191, L. Yuan, Y.-T. Chang, and co-workers design a H<sub>2</sub>S sensing nano-platform based on dye assembled upconversion nanoparticles. These powerful sensors demonstrate highly selective and rapid responses to H<sub>2</sub>S in vitro and in cells, for a variety of applications in environmental and bioimaging fields

## Luminescent Thermometers

The thermal gradients generated at submicrometer scale are the limiting factor of device integration in micro- and nanoelectronics. Silicon functionalized surfaces combine the benefits of electronic circuits with those of molecular probes. On page 200, L. D. Carlos, A. Millán, and co-workers describe the first self-assembled Tb<sup>3+</sup>/Eu<sup>3+</sup>-doped-polymer-based monolayer that acts as a ratiometric thermometer, displaying bi-stability that can be used as an optically active molecular demultiplexer.



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## FEATURE ARTICLE

Inspired by daily hand-writing, ubiquitously available pens have recently been employed to directly write electronics. Increasing demand for cost-effective and easily accessible fabrication of electronics that also allows for rapid prototyping calls for novel techniques to design and manufacture electronics. State-of-art advances in pen-based writing electronics and emerging applications are summarized.



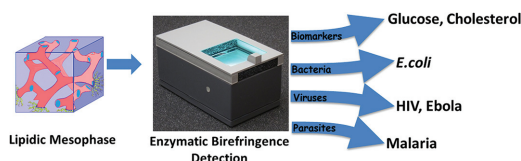
## Pen-Based Writing

Z. Li, H. Liu, C. Ouyang, W. Hong Wee, X. Cui, T. Jian Lu, B. Pingguan-Murphy, F. Li,\* F. Xu\* ..... 165–180

## Recent Advances in Pen-Based Writing Electronics and their Emerging Applications

## FULL PAPERS

Lipidic cubic mesophases are proposed as a general tool to detect analytes of biomedical and epidemic significance by exploiting in meso enzymatic reactions. Due to nanoconfinement effects, the converted substrates crystallize, generating a detectable birefringence. By exploiting this optical signal, it is shown that enzymatic assays can be designed for simple, real-time detection of analytes comprising biomarkers, bacteria, viruses, and parasites, illuminating new pathways toward scalable detection technologies.

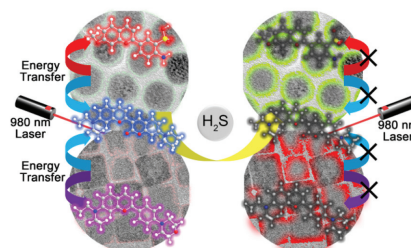


## Disease Diagnostics

J. J. Vallooran, S. Handschin, S. M. Pillai, B. N. Vetter, S. Rusch, H.-P. Beck, R. Mezzenga\* ..... 181–190

## Lipidic Cubic Phases as a Versatile Platform for the Rapid Detection of Biomarkers, Viruses, Bacteria, and Parasites

The request of powerful sensor probes for environmental and biomaging fields has attracted intensive and enthusiastic research during the past decades. Hereby a new dye-assembled  $\text{H}_2\text{S}$  sensing platform—dye-assembled upconversion nanoparticles (UCNPs)—is designed. These probes with emission spectra ranging from visible to near-infrared (NIR) are applicable for  $\text{H}_2\text{S}$  detection in cells and mice serum.

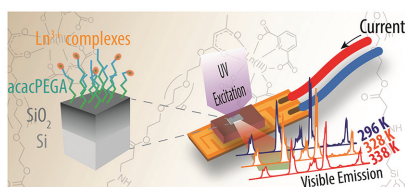


## Sensors

J. Peng, C. L. Teoh, X. Zeng, A. Samanta, L. Wang, W. Xu, D. Su, L. Yuan,\* X. Liu, Y.-T. Chang\* ..... 191–199

## Development of a Highly Selective, Sensitive, and Fast Response Upconversion Luminescent Platform for Hydrogen Sulfide Detection

The first self-assembled  $\text{Tb}^{3+}/\text{Eu}^{3+}$ -doped based polymer monolayer producing a ratiometric temperature readout is fabricated on Si surfaces. The functionalized surface presents reversible bistability that can be used as an optically active molecular demultiplexer with a thermal sensitivity up to  $1.43 \text{ K}^{-1}$  at 323 K, a cycle–recycle reliability of 98.6%, and a temperature uncertainty of less than 0.3 K.



## Luminescent Thermometers

M. Rodrigues, R. Piñol, G. Antorrena, C. D. S. Brites, N. J. O. Silva, J. L. Murillo, R. Cases, I. Díez, F. Palacio, N. Torras, J. A. Plaza, L. Pérez-García, L. D. Carlos,\* A. Millán\* ..... 200–209

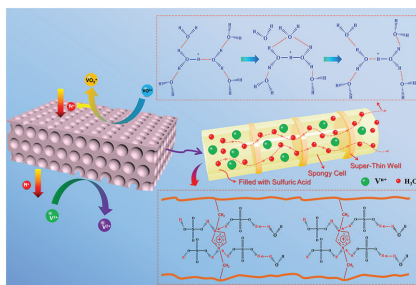
## Implementing Thermometry on Silicon Surfaces Functionalized by Lanthanide-Doped Self-Assembled Polymer Monolayers

FULL PAPERS

Batteries

Y. Zhao, M. Li, Z. Yuan, X. Li,\* H. Zhang,\*  
I. F. J. Vankelecom..... 210–218

**Advanced Charged Sponge-Like Membrane with Ultrahigh Stability and Selectivity for Vanadium Flow Batteries**

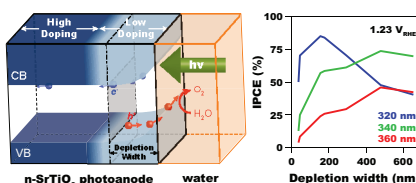


A charged sponge-like membrane with a pore-wall covered imidazolium-based crosslinked structure is designed and fabricated. The prepared membranes exhibit excellent selectivity and stability for vanadium flow battery application.

Water Splitting

M. Liu,\* J. L. Lyons, D. Yan,  
M. S. Hybertsen ..... 219–225

**Semiconductor-Based Photoelectrochemical Water Splitting at the Limit of Very Wide Depletion Region**

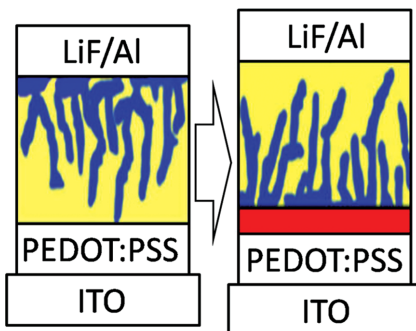


Water photolysis over semiconducting metal oxide is studied at the limit of very wide depletion region for *n*-SrTiO<sub>3</sub> photoanodes. Through a graded doping architecture, the depletion region is widened to an unprecedented depth of 500+ nm, which optimizes the trade-off between photon collection and minority carrier delivery, leading to an incident photon-to-current efficiency over 70% for its weak indirect band gap absorption.

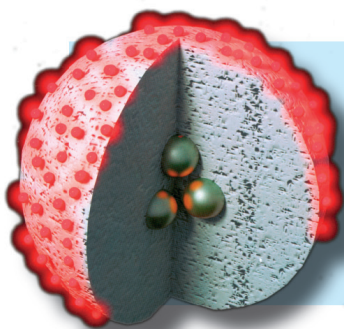
Polymer Solar Cells

B. Meng, Z. Y. Wang, W. Ma,\* Z. Y. Xie,  
J. Liu,\* L. X. Wang ..... 226–232

**A Cross-Linkable Donor Polymer as the Underlying Layer to Tune the Active Layer Morphology of Polymer Solar Cells**



A cross-linkable donor polymer is developed and used as the underlying layer to improve the vertical composition distribution of donor:acceptor in the active layer of polymer solar cells (PSCs). With the improvement, the regular PSC device based on PTB7:PC<sub>71</sub>BM active layer exhibits power conversion efficiency increase from 7.41% to 8.55%.



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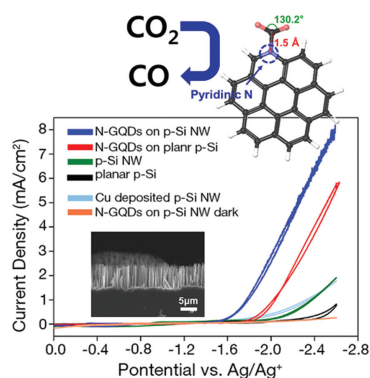
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**Highly selective CO<sub>2</sub> to CO photo-conversion** catalyzed by N-doped graphene quantum sheets (N-GQs) on a p-type Si nanowire is demonstrated. The photocatalytic system produces CO with the chemical selectivity of more than 95%, which is comparable with that of the noble metals such as Au and Re. The developed system provides a new means for utilizing CO<sub>2</sub> as a usable chemical feedstock.

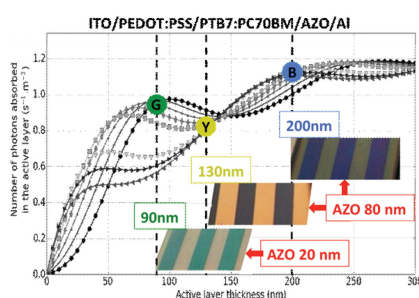


### Quantum Sheets

K. D. Yang, Y. Ha, U. Sim, J. An, C. W. Lee, K. Jin, Y. Kim, J. Park, J. S. Hong, J. H. Lee, H.-E. Lee, H.-Y. Jeong, H. Kim,\* K. T. Nam\* .....233–242

### Graphene Quantum Sheet Catalyzed Silicon Photocathode for Selective CO<sub>2</sub> Conversion to CO

**A facile Al-doped ZnO nanocrystal synthesis for solution-processed electron extraction layer** and playing as well as optical spacer in regular polymer solar cells is presented. A high power conversion efficiency together with fine-tuning of the reflected cell color is obtained. Thick layers of this annealing-free conductive film (up to 80 nm) together with thick layers of active blends (up to 200 nm) strongly improve the compatibility of polymer solar cells with large-scale robust solution processing.

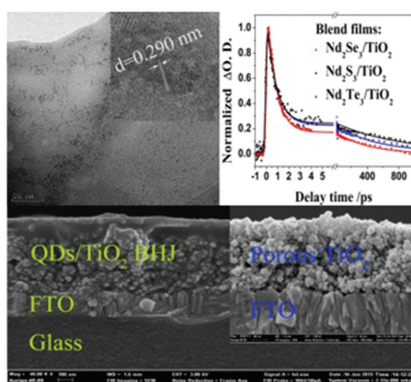


### Organic Solar Cells

M. Gaceur, S. B. Dkhil, D. Duché, F. Bencheikh, J.-J. Simon, L. Escoubas, M. Mansour, A. Guerrero, G. Garcia-Belmonte, X. Liu, M. Fahlman, W. Dachraoui, A. K. Diallo, C. Videlot-Ackermann, O. Margeat,\* J. Ackermann\* .....243–253

### Ligand-Free Synthesis of Aluminum-Doped Zinc Oxide Nanocrystals and their Use as Optical Spacers in Color-Tuned Highly Efficient Organic Solar Cells

**Lead/cadmium-free and optimized neodymium chalcogenide quantum dots (QDs)** have been synthesized by varying the ratio of neodymium to chalcogenide and their promising applications to solar cells have been demonstrated in terms of optical properties, energy level alignment, and charge transfer dynamics, etc. Nd<sub>2</sub>Se<sub>3</sub> QDs exhibit a faster charge transfer rate and a conversion efficiency of 3.19% has been achieved.

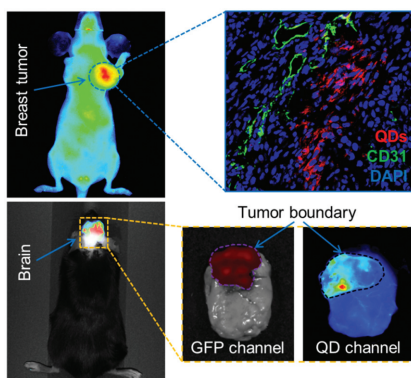


### Quantum Dots

Q. Li,\* X. Jin, Y. Yang, H. Wang, H. Xu, Y. Cheng, T. Wei, Y. Qin, X. Luo, W. Sun,\* S. Luo\* .....254–266

### Nd<sub>2</sub>(S, Se, Te)<sub>3</sub> Colloidal Quantum Dots: Synthesis, Energy Level Alignment, Charge Transfer Dynamics, and Their Applications to Solar Cells

**Highly luminescent cadmium-free CuInSe<sub>2</sub>/ZnS core/shell quantum dots** are demonstrated as a tumor-targeted imaging probe. This probe exhibits excellent photostability, long circulation time, and minimal toxicity. Intense intratumoral signals are detected using three luminescence imaging modalities: near-infrared, time-gated, and two-photon. The probe clearly denotes tumor boundaries and diffusely infiltrating tumor cells in a glioblastoma mouse model.



### Tumor Imaging

X. Liu, G. B. Braun, H. Zhong, D. J. Hall, W. Han, M. Qin, C. Zhao, M. Wang, Z.-G. She, C. Cao, M. J. Sailor, W. B. Stallcup, E. Ruoslahti, K. N. Sugahara\* .....267–276

### Tumor-Targeted Multimodal Optical Imaging with Versatile Cadmium-Free Quantum Dots

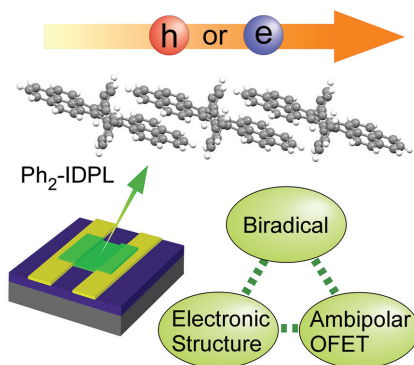


FULL PAPERS

Organic Electronics

H. Koike,\* M. Chikamatsu, R. Azumi, J. Tsutsumi, K. Ogawa, W. Yamane, T. Nishiuchi, T. Kubo, T. Hasegawa, K. Kanai..... 277–283

Stable Delocalized Singlet Biradical Hydrocarbon for Organic Field-Effect Transistors

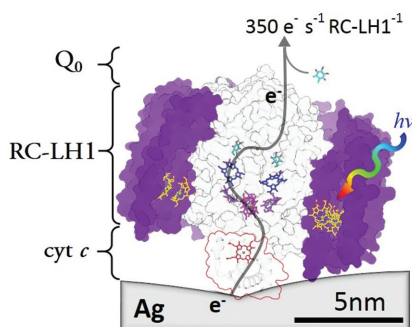


A singlet biradical nature enables high mobility comparable to that of amorphous Si in organic field-effect transistors (OFETs). This paper focuses on the electronic structure and electrical properties of OFETs of a delocalized singlet biradical hydrocarbon, the diphenyl derivative of *s*-indacenodiphenalene (Ph<sub>2</sub>-IDPL). Ph<sub>2</sub>-IDPL holds significant promise as a new semiconducting material for high-performance organic devices.

Bioelectronics

V. M. Friebe, J. D. Delgado, D. J. K. Swainsbury, J. M. Gruber, A. Chanaewa, R. van Grondelle, E. von Hauff, D. Millo, M. R. Jones, R. N. Frese\* ..... 285–292

Plasmon-Enhanced Photocurrent of Photosynthetic Pigment Proteins on Nanoporous Silver

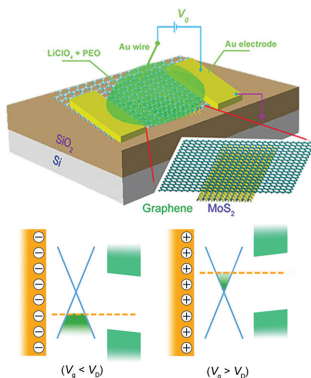


A novel biohybrid photocathode consisting of photosynthetic complexes is revealed on a nanostructured silver electrode. This photocathode nets record photocurrents up to 416  $\mu\text{A cm}^{-2}$  in part thanks to a 2.5-fold plasmon-enhanced light harvesting efficiency.

Excitons

Y. Li, C.-Y. Xu,\* J.-K. Qin, W. Feng, J.-Y. Wang, S. Zhang, L.-P. Ma, J. Cao, P. A. Hu, W. Ren, L. Zhen\* ..... 293–302

Tuning the Excitonic States in MoS<sub>2</sub>/Graphene van der Waals Heterostructures via Electrochemical Gating

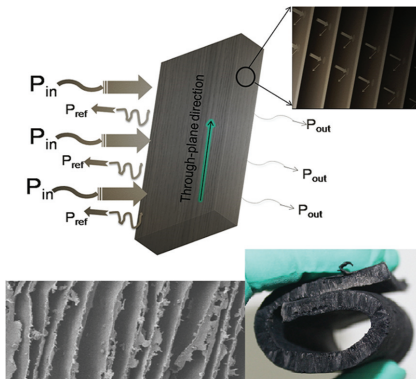


Using monolayer MoS<sub>2</sub>/graphene heterostructures to construct electric double-layer devices with an ion-gels dielectric, the possibility of tuning the photoluminescence properties through electrochemical gating is realized. This tunability can be achieved by either modulating the carrier densities of the counterparts or the band alignment at the interface, and the dominant factor determining the PL tuning can be determined.

Carbon Nanotubes

Z. Zeng, H. Jin,\* M. Chen, W. Li, L. Zhou, Z. Zhang\* ..... 303–310

Lightweight and Anisotropic Porous MWCNT/WPU Composites for Ultrahigh Performance Electromagnetic Interference Shielding



Lightweight, flexible and anisotropic porous composites are assembled by a simple freeze-drying method. The composites contain wide range of mass ratios of multiwalled carbon nanotube in waterborne polyurethane matrix, and show giant electromagnetic interference shielding effectiveness, with the relevant specific shielding effectiveness greater than other shielding materials ever reported.