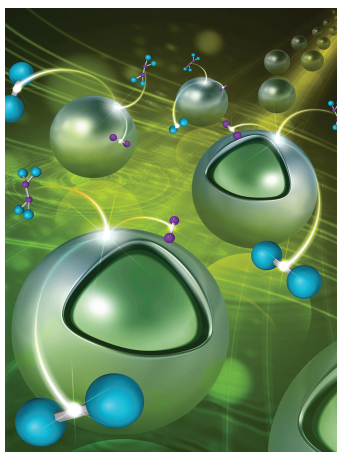


# ADVANCED FUNCTIONAL MATERIALS

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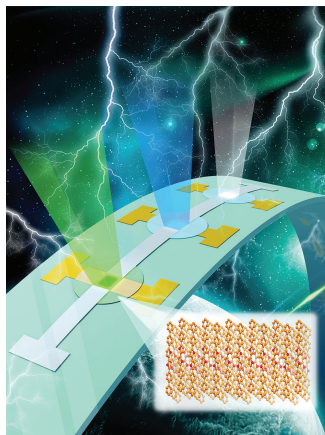
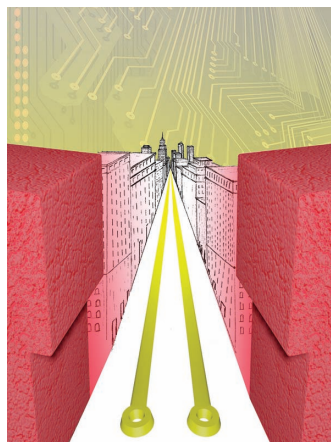


## Nanocatalysts

On page 7073 Y. Zhang and co-workers report the design and synthesis of novel non-precious  $\text{Cu@Fe}_3\text{Ni}_3$  core@shell nanocatalysts, exhibiting 100%  $\text{H}_2$  selectivity and high activity for the complete decomposition of hydrous hydrazine under ambient conditions, which would certainly promote the effective application of hydrous hydrazine as a promising hydrogen storage material.

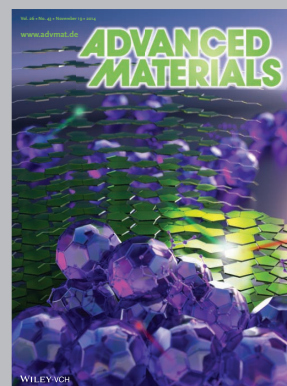
## Organic Crystals

Organic single-crystal-based organic light-emitting devices (OLEDs) with bright and homogeneous luminescence are realized by J. Feng, H.-B. Sun, and colleagues on page 7085 by employing a template stripping technique. Both surface and edge emission are realized by using two kinds of crystals. Highly polarized EL and white emission are also observed from the crystal-based OLEDs with high flexibility and mechanical robustness. This method can be adopted to fabricate the crystal-based optoelectronic devices, providing an avenue to promote their broad applications.



## Self-Assembly

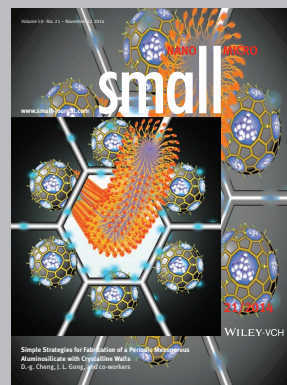
Beyond Manhattan type profiles in block copolymer nanostructures. On page 7078 C. He and M. P. Stoykovich present an approach, analogous to that in bilayer photoresist films, to achieve tunable interface shapes and sidewall profiles in block copolymers self-assembled in thin films. Such interfacial control provides the unique opportunity to design block copolymer nanostructures specially tuned for pattern transfer into functional materials. Cover design by Chunlin He and Kathryn Morrissey.



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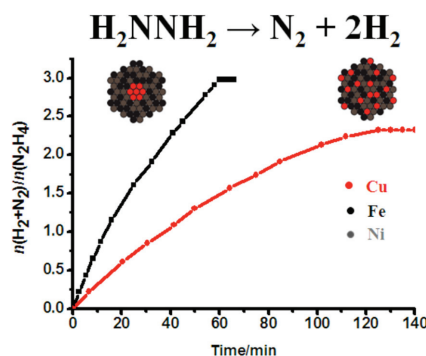
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# FULL PAPERS

## Nanocatalysts

J. Wang, Y. Li, Y. Zhang\* ..... 7073–7077

### Precious-Metal-Free Nanocatalysts for Highly Efficient Hydrogen Production from Hydrous Hydrazine

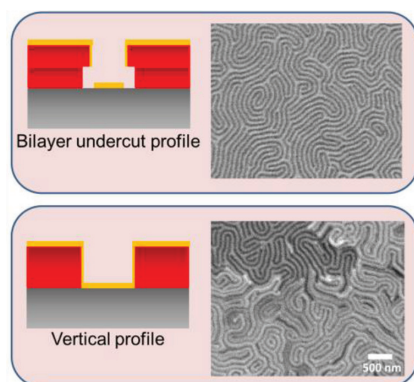


Hydrous hydrazine has great potential as a suitable liquid-hydrogen storage material, provided that a low-cost and high-performance dehydrogenation catalyst is developed. The core@shell nanocatalyst Cu@Fe<sub>3</sub>Ni<sub>5</sub> enables hydrazine decomposition such that the H<sub>2</sub>-producing reaction is completely favored over the ammonia-producing reaction, making hydrous hydrazine a promising hydrogen storage material.

## Self-Assembly

C. He, M. P. Stoykovich\* ..... 7078–7084

### Profile Control in Block Copolymer Nanostructures using Bilayer Thin Films for Enhanced Pattern Transfer Processes



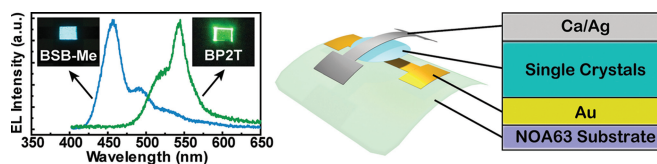
Bilayer thin films of lamellar-forming poly(styrene-*block*-methyl methacrylate) are self-assembled and thermally cross-linked on wafer substrates in a layer-by-layer process. This layer-by-layer approach can provide novel control over the interface profile of block copolymer nanostructures and enables an undercut sidewall profile that enhances metal lift-off processes for pattern transfer.

## Organic Crystals

R. Ding, J. Feng,\* X.-L. Zhang, W. Zhou, H.-H. Fang, Y.-F. Liu, Q.-D. Chen, H.-Y. Wang, H.-B. Sun\* ..... 7085–7092

### Fabrication and Characterization of Organic Single Crystal-Based Light-Emitting Devices with Improved Contact Between the Metallic Electrodes and Crystal

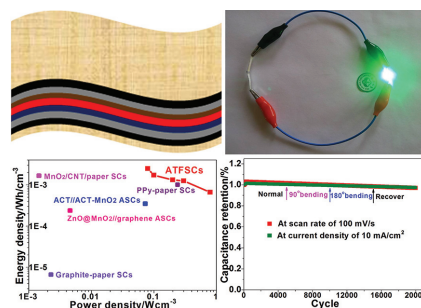
Organic single-crystal-based LEDs with bright and homogeneous luminescence are realized due to an improved contact between the crystals and both top and bottom electrodes by employing the template stripping technique. Both surface and edge emission are realized by using two kinds of crystals. Highly polarized EL and white emission are also observed from the crystal-based OLEDs. Moreover, the crystal-based OLEDs exhibit high flexibility and mechanical robustness.



## Supercapacitors

J.-X. Feng, S.-H. Ye, A.-L. Wang, X.-F. Lu, Y.-X. Tong, G.-R. Li\* ..... 7093–7101

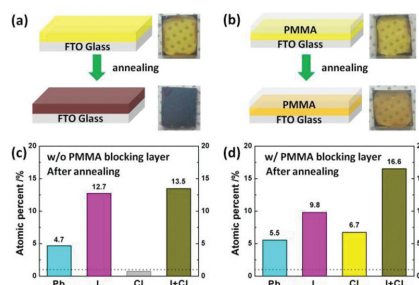
### Flexible Cellulose Paper-based Asymmetrical Thin Film Supercapacitors with High-Performance for Electrochemical Energy Storage



Cellulose paper (CP)-based asymmetrical thin film supercapacitors are assembled by using sandwich-structured graphite/Ni/Co<sub>2</sub>NiO<sub>4</sub>-CP as positive electrode and graphite/Ni/AC-CP as negative electrode. The assembled devices exhibit large C<sub>sp</sub> (7.6 F/cm<sup>3</sup> at 5 mV/s), high volumetric energy density (2.48 mWh/cm<sup>3</sup>, 80 Wh/kg), high volumetric power density (0.79 W/cm<sup>3</sup>, 25.6 kW/kg), and excellent cycle stability (less 4% C<sub>sp</sub> loss after 20 000 cycles).

## FULL PAPERS

To understand the formation mechanism of  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  perovskite, two testing structures, perovskite (precursor mixture)/FTO and PMMA (polymethyl methacrylate)/perovskite (precursor mixture)/FTO, are designed. The different annealing results of these two structures suggest that the formation of  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  perovskite is likely driven by the release of gaseous  $\text{CH}_3\text{NH}_3\text{Cl}$  through an intermediate organolead mixed halide phase.

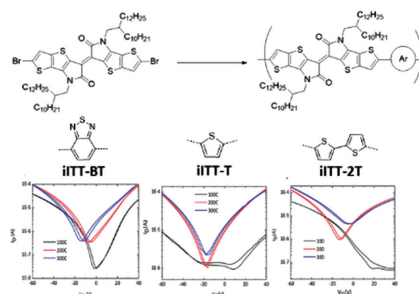


## Photovoltaics

H. Yu, F. Wang, F. Xie, W. Li, J. Chen, N. Zhao\* ..... 7102–7108

### The Role of Chlorine in the Formation Process of " $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ " Perovskite

A series of new isoindigo semiconducting polymers with thieno[3,2-*b*]thiophene as the donor moiety are synthesized. By fusing an extra aromatic ring to the isoindigo core, absorption profiles are shifted into the near-IR with optical band gaps as narrow as 1.05 eV. Organic top-gate bottom-contact transistor devices are fabricated demonstrating good ambipolar mobilities.

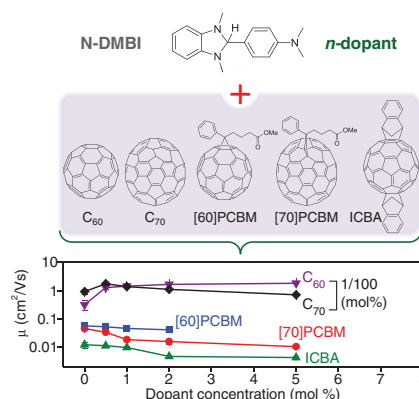


## Conjugated Polymers

I. Meager,\* M. Nikolka, B. C. Schroeder, C. B. Nielsen, M. Planells, H. Bronstein, J. W. Rumer, D. I. James, R. S. Ashraf, Aditya Sadhanala, P. Hayoz, J.-C. Flores, H. Sirringhaus, I. McCulloch... 7109–7115

### Thieno[3,2-*b*]thiophene Flanked Isoindigo Polymers for High Performance Ambipolar OFET Applications

Doping of organic semiconductors is a key technology for tuning their charge transport properties. Here a systematic comparison of n-type doping of solution-processed un/substituted fullerenes with regard to optical, electrical, and morphological properties, is presented. In case of  $\text{C}_{60}$  and  $\text{C}_{70}$ , n-doping is found to both increase the electron mobility and improve the bias stress stability of the transistors.

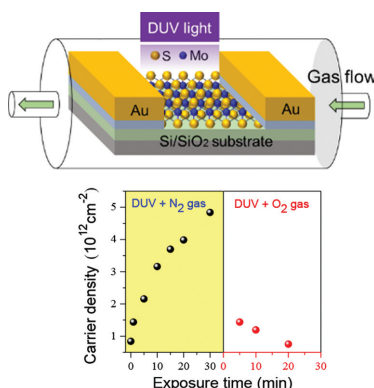


## Molecular Doping

S. Rossbauer, C. Müller, T. D. Anthopoulos\* ..... 7116–7124

### Comparative Study of the N-Type Doping Efficiency in Solution-processed Fullerenes and Fullerene Derivatives

The charge-carrier density of single-, bi-, and few-layer  $\text{MoS}_2$  nanosheets can be finely and reversibly tuned with  $\text{N}_2$  and  $\text{O}_2$  gas in the presence of deep-ultraviolet (DUV) light. The exposure to  $\text{N}_2$  gas in the presence of DUV light significantly improves the drain-to-source current, carrier density, and charge-carrier mobility for  $\text{MoS}_2$  nanosheets.



## Charge Transport

A. K. Singh, S. Andleeb, J. Singh, H. T. Dung, Y. Seo, J. Eom\* ..... 7125–7132

### Ultraviolet-Light-Induced Reversible and Stable Carrier Modulation in $\text{MoS}_2$ Field-Effect Transistors

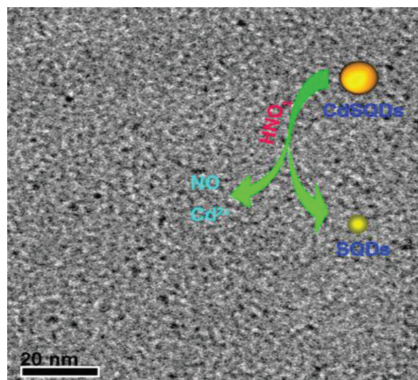


## FULL PAPERS

## Quantum Dots

S. X. Li,\* D. J. Chen, F. Y. Zheng,  
H. F. Zhou, S. X. Jiang,  
Y. J. Wu ..... 7133–7138

### Water-Soluble and Lowly Toxic Sulphur Quantum Dots

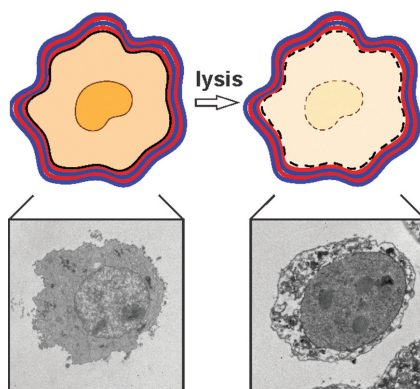


**Sulphur quantum dots** are first synthesized by a ox-red reaction at the water–oil interface. The sulphur quantum dots show good water solubility, excellent stability, and lowly toxicity. Sulphur quantum dots sensitized TiO<sub>2</sub> photocatalyze the generation of H<sub>2</sub> from a water/methanol mixture upon irradiation with UV. A Fe<sup>3+</sup> ions fluorescence sensor demonstrates good linearity and high selectiveness.

## Immune Therapy

L. Lybaert, E. De Vlieghere, R. De Rycke,  
N. Vanparijs, O. De Wever, S. De Koker,  
B. G. De Geest\* ..... 7139–7150

### Bio-Hybrid Tumor Cell-Templated Capsules: A Generic Formulation Strategy for Tumor Associated Antigens in View of Immune Therapy

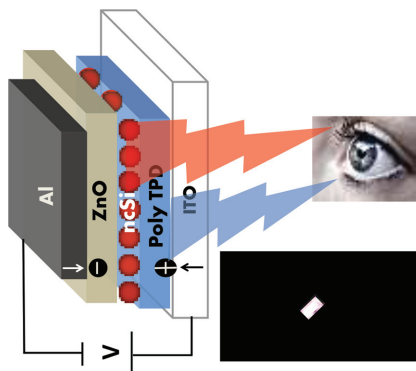


**Melanoma cells** are coated via layer-by-layer deposition of poly(vinylpyrrolidone) and tannic acid followed by lysis through hypo-osmotic treatment. The coating is optimized to preserve the cellular integrity and optimal retention of the cellular proteins within the cell-templated capsules. In addition, a proof-of-concept study is performed to demonstrate the encapsulation of heat shock proteins as potential immune-activators.

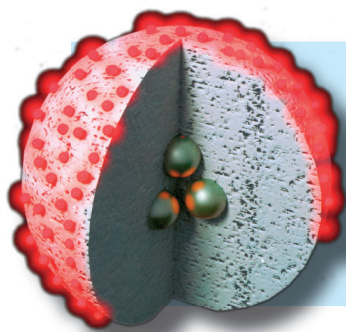
## Hybrid Electronics

B. Ghosh, Y. Masuda, Y. Wakayama,  
Y. Imanaka, J.-i. Inoue, K. Hashi,  
K. Deguchi, H. Yamada, Y. Sakka,  
S. Ohki, T. Shimizu,  
N. Shirahata\* ..... 7151–7160

### Hybrid White Light Emitting Diode Based on Silicon Nanocrystals



**A novel design of white light emitting diodes** emerges to meet the growing global demand for resource sustainability while preserving health and environment. The proposed WLED contains a hybrid bi-layer of silicon nanocrystals (ncSi) and luminescent polymer in its device active region, and works at sufficiently low turn-on voltage (3.5 V) by taking advantages of the large Stokes shift inherent to ncSi.



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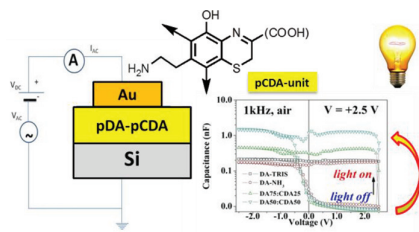
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## FULL PAPERS

**Incorporation of cysteinyl-dopamine(CDA)-derived units into polydopamine (pDA)** leads to an innovative bioinspired material combining the photosensitizing properties of red human hair pigments with the electrical properties of melanins. The resulting mixed-type biopolymers exhibit a markedly enhanced response to UV light and open the doorway to unprecedented bio-inspired metal-insulator-semiconductor photo-capacitor hybrid devices with tunable optical response for light sensing applications.

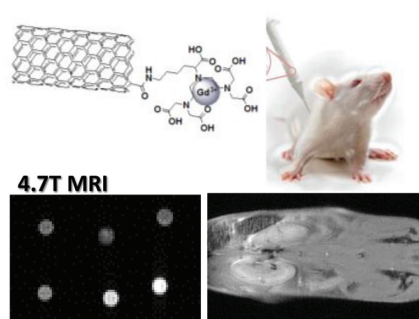


### Light Sensing

M. Ambrico,\* N. F. Della Vecchia, P. F. Ambrico, A. Cardone, S. R. Cicco, T. Ligonzo, R. Avolio, A. Napolitano, M. d'Ischia\* .....7161–7172

### A Photoresponsive Red-Hair-Inspired Polydopamine-Based Copolymer for Hybrid Photocapacitive Sensors

**Oxidized multi-walled carbon nanotubes** are provided with positive MRI contrast properties by covalent functionalization with the chelating ligand DTPA followed by chelation to  $Gd^{3+}$ . Despite the intrinsic  $T_2$  contrast of oxidized CNTs, the covalent anchoring of paramagnetic gadolinium ions on CNT sidewall allows high dispersibility and efficient positive MR contrast both in vitro in labeled cells and in vivo after intravenous injection of CNTs in mice.

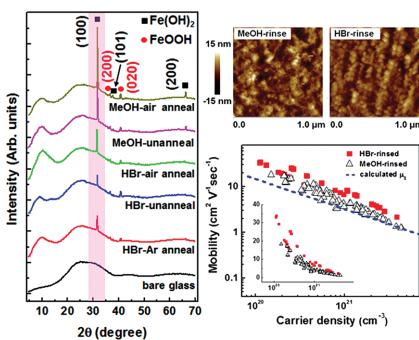


### Nanomedicine

I. Marangon, C. Ménard-Moyon, J. Kolosnjaj-Tabi, M. L. Béoutis, L. Lartigue, D. Alloeyau, E. Pach, B. Ballesteros, G. Autret, T. Ninjbadgar, D. F. Brougham, A. Bianco,\* .....7173–7186

### Covalent Functionalization of Multi-walled Carbon Nanotubes with a Gadolinium Chelate for Efficient $T_1$ -Weighted Magnetic Resonance Imaging

**Heavily-doped poly(3,4-ethylenedioxythiophene)(PEDOT) films with carrier density higher than  $\sim 10^{20} \text{ cm}^{-3}$**  are produced using oxidative chemical-vapor-deposition. The mechanisms for the conductivity decrease and carrier transport behavior in heavily-doped PEDOT, which are important to organic optoelectronic device applications that show a strong effect of air-exposure and low temperature annealing on the device stability and performance, are suggested.

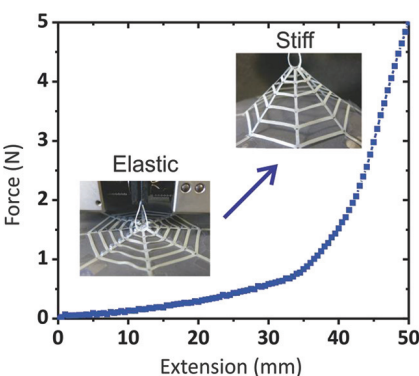


### Conducting Polymers

S. Lee, D. C. Paine, K. K. Gleason\* .....7187–7196

### Heavily Doped poly(3,4-ethylenedioxythiophene) Thin Films with High Carrier Mobility Deposited Using Oxidative CVD: Conductivity Stability and Carrier Transport

**Adaptive Young's moduli of composites** are demonstrated by combining different materials and controlling their geometries. As applied strain increases, the critical component that dictates the mechanical strength of the composite shifts from an elastic material to a stiff material, resulting in discrete steps in increasing moduli. The characteristics of the steps can be engineered to accommodate specific requirements of applications.



### Adaptive Materials

J.-H. So, A. S. Tayi, F. Güder, G. M. Whitesides\* .....7197–7204

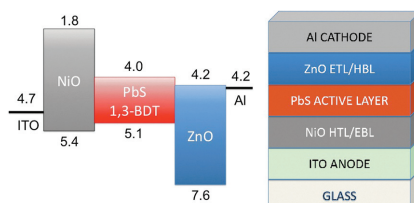
### Stepped Moduli in Layered Composites

## FULL PAPERS

## Photodetectors

J. R. Manders, T.-H. Lai, Y. An,  
W. Xu, J. Lee, D. Y. Kim, G. Bosman,\*  
F. So\* ..... 7205–7210

**Low-Noise Multispectral Photodetectors  
Made from All Solution-Processed  
Inorganic Semiconductors**

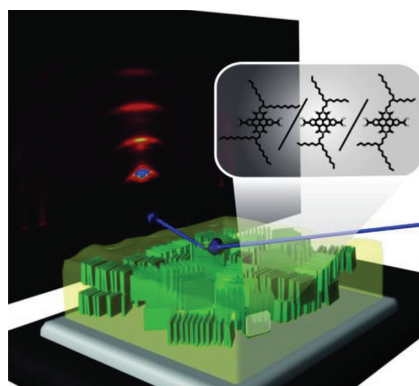


**Air-stable, all-solution processed all-inorganic multispectral photodetectors** using PbS quantum dots as the photo-active layer, colloidal ZnO nanoparticles as the electron transport/hole blocking layer, and solution-derived NiO as the hole transport/electron blocking layer are reported. The resulting devices have very low dark current density with a noise equivalent power on the order of tens of picowatts across the detection spectra. The performance parameters are comparable to commercially available Si, Ge, and InGaAs photodetectors and the device has a long unencapsulated storage lifetime in air.

## Organic Electronics

E. Gann, X. Gao, C.-a. Di,  
C. R. McNeill\* ..... 7211–7220

**Phase Transitions and Anisotropic  
Thermal Expansion in High Mobility  
Core-expanded Naphthalene Diimide  
Thin Film Transistors**



**In situ grazing incidence wide angle X-ray scattering** monitors the crystallinity and phase behavior of thin film organic systems during heating. In the case of solution-processed core expanded naphthalene diimide small molecules, the branching location of alkyl side chains tunes in-plane thermal expansion coefficients which in turn correlate with mobility of the quenched film.