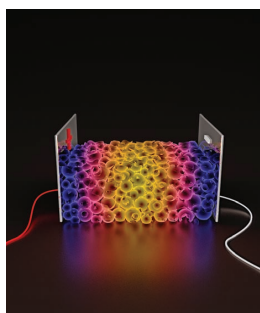


# ADVANCED FUNCTIONAL MATERIALS

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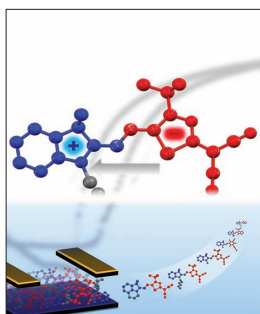
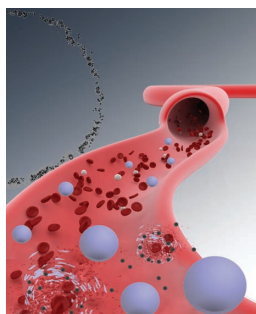


## Graphene Aerogels

On page 28, a fundamental study of the direct resistive heating of emulsion-templated graphene aerogels demonstrates reliable, fast, energy-efficient and uniform temperature control of the graphene framework. R. Menzel, M. S. P. Shaffer, and team's findings lay a framework that may be exploited in a wide range of novel applications for nanocarbon aerogels, including flow-through local gas heaters, temperature-sensitive catalytic reactions, and thermally regenerable filter and storage systems.

## Hybrid Materials

Enabling safe and efficient drug delivery across the blood-brain barrier (BBB) remains to be challenging. On page 36 of this issue, T. Lammers, P. Koczera, F. Kiessling, and colleagues show that polymer-based microbubbles, containing iron oxide nanoparticles within their shell, can be employed to mediate and monitor BBB permeation. Such theranostic constructs and concepts are considered to be useful for tailoring the treatment of central nervous system disorders.

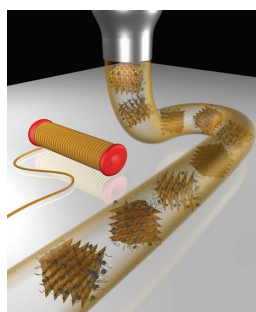


## Organic Electronics

F. Würthner and team investigate the suitability of twenty dipolar merocyanine dyes as active layer in organic thin-film transistors on page 44. While the device performance varies significantly up to  $0.18 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , it is shown that a good performance of some dyes occurs when they exhibit a high ground state dipole moment and well-defined sterics, resulting in a beneficial brick-work-type packing within the thin film.

## Graphene Oxide

The use of large graphene oxide (GO) sheets as a filler enables the continuous fabrication of elastomeric composite fibers with remarkable stiffness and no loss of stretchability compared to the parent elastomeric polymer. J. M. Razal, G. G. Wallace, and co-workers report the production of the fibers on page 94, and provide insights into how the GO sheet size influences the fiber mechanical properties.



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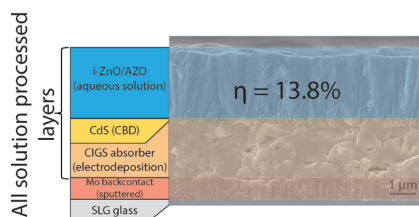
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## FEATURE ARTICLE

A  $\text{Cu(In,Ga)Se}_2$  thin film solar cell with efficiency of 13.8% is demonstrated where all constituent layers (except the metal back contact) are processed from aqueous solutions of metal salts using industrially scalable processes.



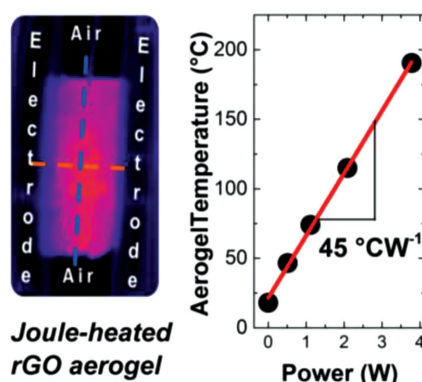
## Photovoltaic Devices

Y. E. Romanyuk,\* H. Hagendorfer, P. Stücheli, P. Fuchs, A. R. Uhl, C. M. Sutter-Fella, M. Werner, S. Haass, J. Stükelberger, C. Broussillou, P.-P. Grand, V. Bermudez, A. N. Tiwari ..... 12–27

**All Solution-Processed Chalcogenide Solar Cells – from Single Functional Layers Towards a 13.8% Efficient CIGS Device**

## FULL PAPERS

A fundamental study of the direct resistive heating of graphene aerogels demonstrates efficient, uniform, fast, and repeatable Joule heating. The outlined principles pave a route towards new applications of three-dimensional nano-carbon networks as viable light-weight, high-surface-area local gas heaters and temperature-controlled catalyst and adsorber supports.

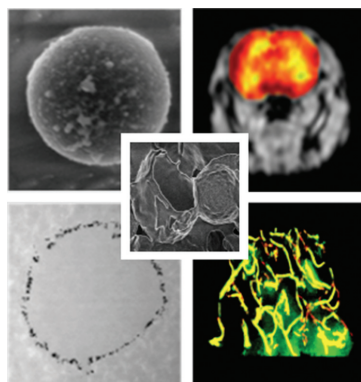


## Graphene Aerogels

R. Menzel,\* S. Barg, M. Miranda, D. B. Anthony, S. M. Bawaked, M. Mokhtar, S. A. Al-Thabaiti, S. N. Basahel, E. Saiz, M. S. P. Shaffer\* ..... 28–35

**Joule Heating Characteristics of Emulsion-Templated Graphene Aerogels**

Polymer-based microbubbles containing ultrasmall superparamagnetic iron oxide (USPIO) nanoparticles within their shell can be used to simultaneously mediate and monitor blood-brain barrier (BBB) permeation and to enable efficient macromolecular (model) drug delivery into the central nervous system.



## Hybrid Materials

T. Lammers,\* P. Koczera,\* S. Fokong, F. Gremse, J. Ehling, M. Vogt, A. Pich, G. Storm, M. van Zandvoort, F. Kiessling\* ..... 36–43

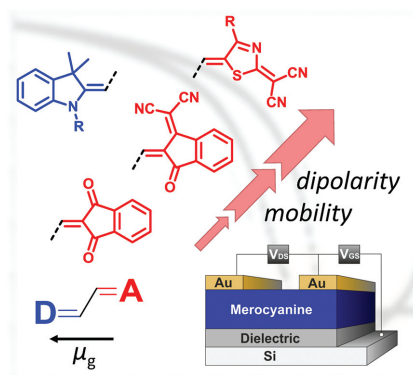
**Theranostic USPIO-Loaded Microbubbles for Mediating and Monitoring Blood-Brain Barrier Permeation**

# FULL PAPERS

## Organic Electronics

A. Liess, L. Huang, A. Arjona-Esteban, A. Lv, M. Gsänger, V. Stepanenko, M. Stolte, F. Würthner\* ..... 44–57

### Organic Thin Film Transistors Based on Highly Dipolar Donor–Acceptor Polymethine Dyes

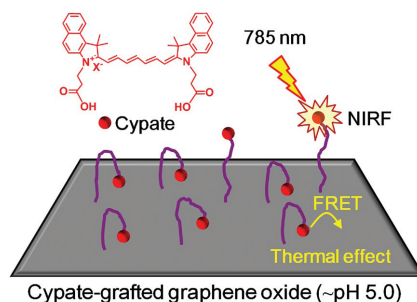


Twenty dipolar donor–acceptor-substituted polymethine dyes (merocyanines) are investigated as an active layer in organic thin film transistors. It is shown that the diverse performance of the devices can be attributed to either low or high dimensional ordering in the crystal structure which in the latter case leads to mobilities as high as  $0.18 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ .

## Photothermal Therapy

M. Guo, J. Huang, Y. B. Deng, H. Shen, Y. F. Ma, M. X. Zhang, A. J. Zhu, Y. L. Li, H. Hui, Y. Y. Wang, X. L. Yang, Z. J. Zhang,\* H. B. Chen\* ..... 59–67

### pH-Responsive Cyanine-Grafted Graphene Oxide for Fluorescence Resonance Energy Transfer-Enhanced Photothermal Therapy

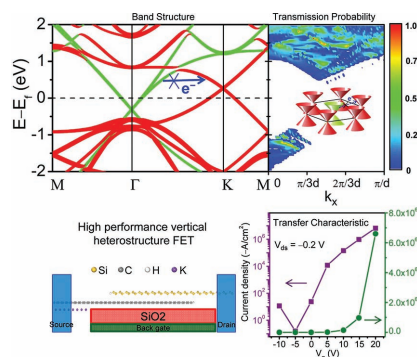


A smart cyanine-grafted GO nanoconjugate (GO-Cypate) with pH-dependent FRET effect, which triggers pH-responsive photothermal effect and thus results in enhanced PTT efficacy with tumor ablation upon NIR irradiation after a single-dose intravenous injection, is reported.

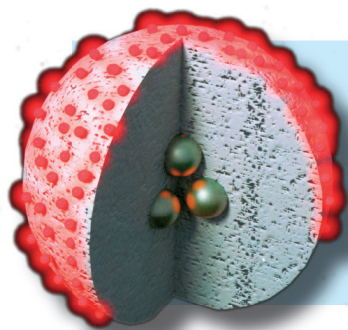
## Dirac Materials

Y. Wang, Z. Ni, Q. Liu, R. Quhe, J. Zheng, M. Ye, D. Yu, J. Shi, J. Yang, J. Li, J. Lu\* ..... 68–77

### All-Metallic Vertical Transistors Based on Stacked Dirac Materials



Electron transport from one Dirac material to the other near  $E_f$  is forbidden by momentum mismatch if the two Dirac cones of different layers are well separated. All-metallic field effect transistor can be designed out of Dirac materials with a large transport gap and a high on/off current ratio of over  $10^4$  based on ab initio quantum transport simulations.



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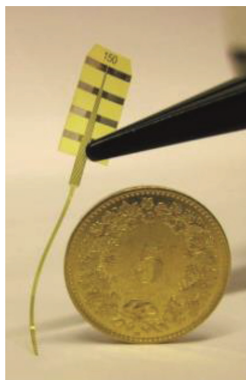
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## FULL PAPERS

**Incorporating glassy carbon electrodes into flexible neural probes** allows for implants to be used as chemical sensors as well as electrical sensors and stimulators. The devices are microfabricated on carrier wafers out of polyimide, platinum, titanium, and glassy carbon. The process demonstrated here is generally applicable for the integration of various high temperature materials into flexible devices.

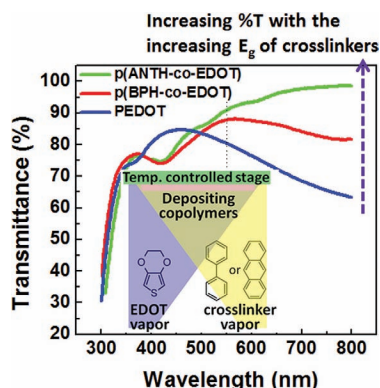


## Biomedical Implants

J. J. VanDersarl,\* A. Mercanzini,  
P. Renaud\* ..... 78–84

**Integration of 2D and 3D Thin Film Glassy Carbon Electrode Arrays for Electrochemical Dopamine Sensing in Flexible Neuroelectronic Implants**

**Band gap-tuned PEDOT copolymers** are successfully demonstrated using oCVD with the incorporation of cross-linking monomers of anthracene or biphenyl. oCVD offers single-step synthesis, deposition and doping of copolymers. oCVD copolymers show significantly improved visible-regime transparency compared to homopolymer PEDOT, which is promising for many organic optoelectronic devices that require high optical transmittance with electrical conductivity.

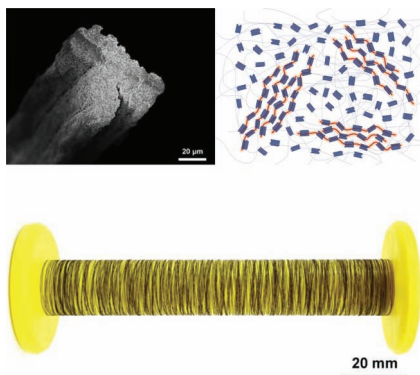


## Conducting Polymers

S. Lee, K. K. Gleason\* ..... 85–93

**Enhanced Optical Property with Tunable Band Gap of Cross-linked PEDOT Copolymers via Oxidative Chemical Vapor Deposition**

**A straightforward approach to prepare liquid crystalline dispersions** of polyurethane/graphene oxide (PU/GO) formulations has enabled the fabrication of high performance elastomeric composite fibers. The PU/GO fibers containing large GO sheets display remarkable reinforcement in modulus and yield stress (low strain properties) without compromising the extensibility and stretchability (high strain properties) inherent to PU. These fibers are produced using a very simple, continuous, and highly scalable fiber wet-spinning process.

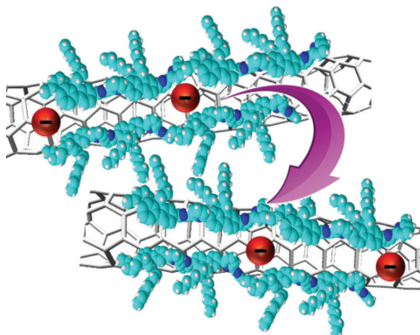


## Graphene Oxide

M. Z. Seyedin, J. M. Razal,\* P. C. Innis,  
R. Jalili, G. G. Wallace\* ..... 94–104

**Achieving Outstanding Mechanical Performance in Reinforced Elastomeric Composite Fibers Using Large Sheets of Graphene Oxide**

**The temperature-dependent electrical transport in polymer-sorted semiconducting carbon nanotube networks** is elucidated in this work. The source-drain current and mobility in polymer-sorted carbon nanotube network-based field-effect transistors decrease with decreasing temperature attributable to fluctuation-induced tunneling. The barrier for charge transport inversely correlates with carbon nanotube network thickness, for the probability to sample conductive pathways with fewer inter-tube junctions increases with increasing thickness.



## Carbon Nanotube Networks

J. Gao, Y.-L. Loo\* ..... 105–110

**Temperature-Dependent Electrical Transport in Polymer-Sorted Semiconducting Carbon Nanotube Networks**

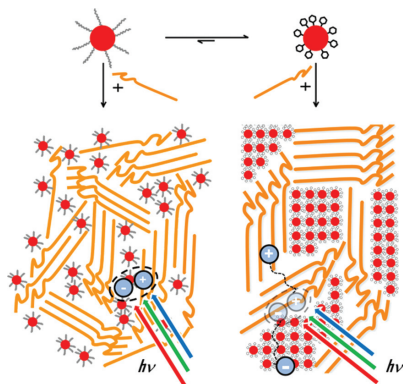


## FULL PAPERS

## Hybrid Nanocomposites

C. Giansante,\* R. Mastria,  
G. Lerario, L. Moretti, I. Kriegel,  
F. Scotognella, G. Lanzani, S. Carallo,  
M. Esposito, M. Biasiucci, A. Rizzo,  
G. Gigli..... 111–119

### Molecular-Level Switching of Polymer/ Nanocrystal Non-Covalent Interactions and Application in Hybrid Solar Cells

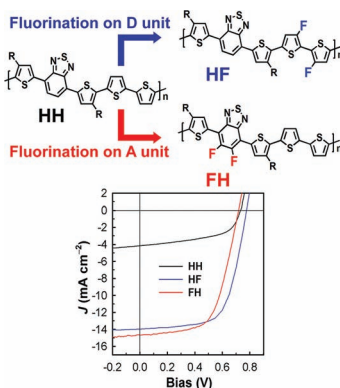


**Hybrid nanocomposites with switched morphology** are obtained by mediating non-covalent interactions between conjugated polymers and semiconductor nanocrystals via the pending moiety of organic ligands at the nanocrystal surface. Morphology switching deeply impacts the optoelectronic properties of polythiophene/PbS nanocrystal composites, as demonstrated by achieving unprecedented photovoltaic device performances for this blend material. Photo-induced processes at the hybrid interface are investigated and discussed.

## Photovoltaic Devices

J. W. Jo, S. Bae, F. Liu, T. P. Russell,  
W. H. Jo\*..... 120–125

### Comparison of Two D–A Type Polymers with Each Being Fluorinated on D and A Unit for High Performance Solar Cells

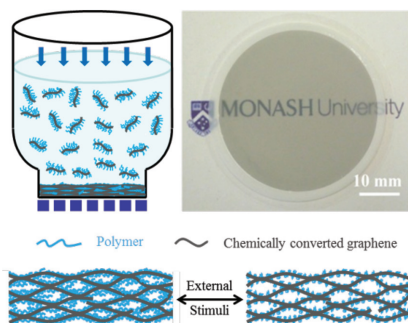


**Two types of fluorinated D–A polymers with each being fluorinated on D and A unit** are designed and synthesized. The D–A polymer with fluorinated D unit exhibits well-developed fibril network, low bimolecular recombination and high hole mobility, leading to a high PCE of 7.10%, which is higher than the PCE (6.41%) of the polymer with fluorinated A unit.

## Smart Membranes

Y. Wang, S. Chen, L. Qiu, K. Wang,  
H. Wang, G. P. Simon, D. Li\*... 126–133

### Graphene-Directed Supramolecular Assembly of Multifunctional Polymer Hydrogel Membranes

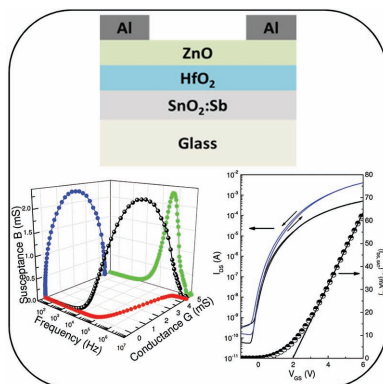


**A versatile approach to the fabrication of multifunctional polymer hydrogel membranes** is demonstrated by exploiting the unique micro-corrugated 2D configuration of chemically converted graphene, its self-assembly behavior and rich supramolecular interactions. A range of water soluble polymers can be readily processed into such membranes with tunable nanostructures to suit a variety of potential applications.

## Semiconductors

M. Esro, G. Vourlias, C. Somerton,  
W. I. Milne, G. Adamopoulos\*... 134–141

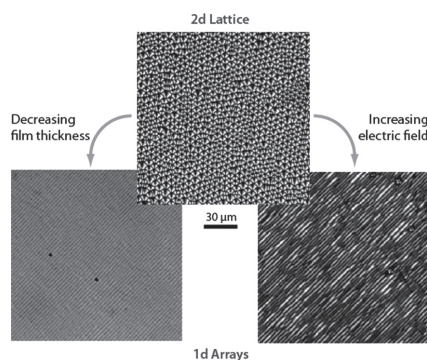
### High-Mobility ZnO Thin Film Transistors Based on Solution-processed Hafnium Oxide Gate Dielectrics



**Solution-processed metal oxide thin film transistors (TFTs)** employing sequential spray coated antimony-doped tin oxide (SnO<sub>2</sub>:Sb) gate electrodes, hafnium oxide (HfO<sub>2</sub>) gate dielectrics and zinc oxide (ZnO) semiconducting channels are demonstrated. The transistors show excellent characteristics in terms of high transparency, hysteresis-free operation, low operation voltage, high electron mobility, and On/Off current modulation ratio.

## FULL PAPERS

**Rapid formation of periodic defect patterns** can be induced and controlled in smectic liquid crystal films by applying electric fields and varying the film thickness at the smectic-nematic phase transition. The pattern type (1d/2d) and period persist in the smectic phase in a non-equilibrium state, stabilized by large topological barriers, even after switching the field off.

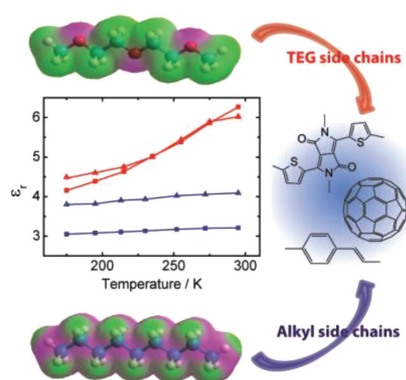


## Liquid Crystals

I. Gryn, E. Lacaze, R. Bartolino, B. Zappone\* ..... 142–149

### Controlling the Self-Assembly of Periodic Defect Patterns in Smectic Liquid Crystal Films with Electric Fields

A **synthetic strategy** is presented for the dielectric constant enhancement of organic semiconductors. It is demonstrated that fullerene derivatives, DPP- and PPV-based polymers show a marked increase of the relative dielectric constant when being functionalized with TEG side chains. Density functional theory calculations attribute such enhancements to the rapid reorientations of ethylene glycol dipoles on the side chains.

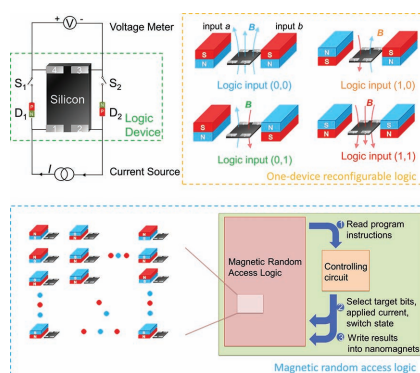


## Charge Transport

S. Torabi, F. Jahani, I. Van Severen, C. Kanimozhi, S. Patil, R. W. A. Havenith, R. C. Chiechi, L. Lutsen, D. J. M. Vanderzande, T. J. Cleij, J. C. Hummelen, L. J. A. Koster\* ..... 150–157

### Strategy for Enhancing the Dielectric Constant of Organic Semiconductors Without Sacrificing Charge Carrier Mobility and Solubility

**Magnetoelectronics logic** shows promise for surpassing the development limits of CMOS logic and arouses profound attentions. Here, silicon-based current-controlled reconfigurable magnetoresistance logic is experimentally demonstrated. This logic device performs four basic Boolean logic (AND, OR, NAND, and NOR) in one device. It has the advantages of reconfiguration, ultralow consumption, instant-on performance, and would bridge the processor-memory gap.



## Silicon-Based Devices

Z. C. Luo, X. Z. Zhang,\* C. Y. Xiong, J. J. Chen ..... 158–166

### Silicon-Based Current-Controlled Reconfigurable Magnetoresistance Logic Combined with Non-Volatile Memory