

# ADVANCED FUNCTIONAL MATERIALS

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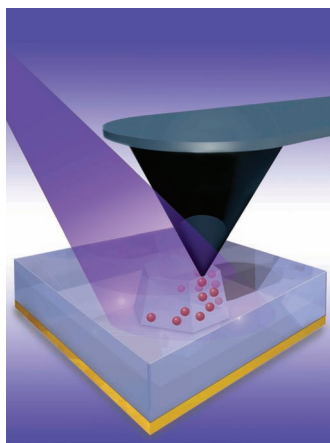


## Immunosensors

The electrogenerated chemiluminescence (ECL) of a novel phenyleneethynylene derivative (4,4'-(2,5-dimethoxy-1,4-phenylene)bis(ethyne-2,1-diyl) dibenzoic acid) that has carboxylic acid groups at the para positions in aqueous solution is studied and its first immunosensing application is reported. On page 3899, Jinghua Yu and co-workers report that the nanotubular mesoporous Pt-Ag alloy nanoparticles are used in the ECL immunosensor as a signal amplification carrier to obtain ultrasensitive ECL analysis.

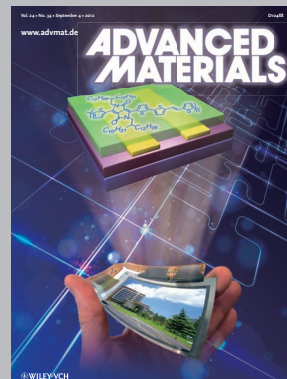
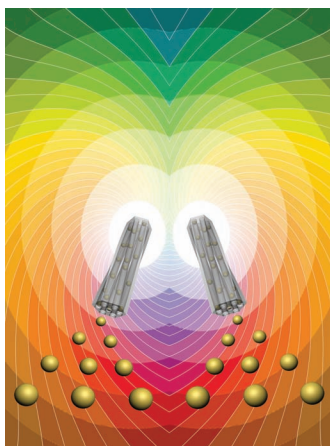
## Chiral Nanoparticles

The chiroptical properties of pure inorganic material are reported by Shunai Che and co-workers on page 3784. The properties are investigated by dispersing achiral metal nanoparticles into chiral mesoporous silica, due to collectively asymmetric plasmon interactions of metal nanoparticles arranged along a helical pore orientation. Such chiral materials will have novel applications for biosensing, chiral recognition, and detection, based on the importance of surface plasmon resonance (SPR) in biological structures.



## Photoinduced Conductivity

Domain walls (DWs) in highly insulating, millimeter-thick lithium niobate ( $\text{LiNbO}_3$ ) single crystals show a pronounced electric conductivity when being illuminated by super-bandgap light. As reported by Lukas M. Eng and co-workers on page 3936, this conductivity can be tuned by engineering the tilting angle of the DWs with respect to the polar axis. The results are obtained using conductive atomic force microscopy and macroscopic transport measurements at various temperatures.



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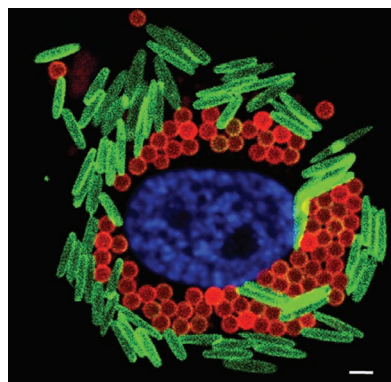
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## FULL PAPERS

## Particulate Drug Delivery

P. Kolhar, S. Mitragotri\* .....3759–3764

## Polymer Microparticles Exhibit Size and Shape Dependent Accumulation around the Nucleus after Endocytosis



The intracellular accumulation and subsequent spatial segregation of spherical and rod-shaped microparticles are studied. Both spherical and rod-shaped particles exhibit perinuclear accumulation upon cellular internalization; however binary mixtures of particles spatially segregate in the cytoplasm based on their shape and size. Larger particles exhibit preferential accumulation closer to the nucleus.

## Self-Assembly

N. Hendler, B. Belgorodsky, E. D. Mentovich, S. Richter,\* L. Fadeev,\* M. Gozin\* .....3765–3776

Multiple Self-Assembly Functional Structures Based on Versatile Binding Sites of  $\beta$ -Lactoglobulin

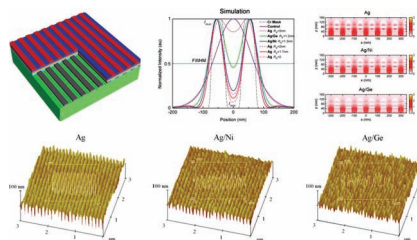
A synthetic methodology for construction of novel multifunctional materials in a bottom-up fashion is demonstrated by combination of highly ordered self-assembly nanostructures with chemical versatility of the orderly doped  $\beta$ -lactoglobulin protein-based fibrils. Well-defined complexes of  $\beta$ -lactoglobulin with retinoic acid, protoporphyrin IX or carboxyfullerene ligands are converted into ligand-doped fibrils with designed physical properties.



## Lithography

H. Liu, B. Wang, L. Ke, J. Deng, C. C. Choy, M. S. Zhang, L. Shen, S. A. Maier,\* J. H. Teng\* .....3777–3783

## High Contrast Superlens Lithography Engineered by Loss Reduction

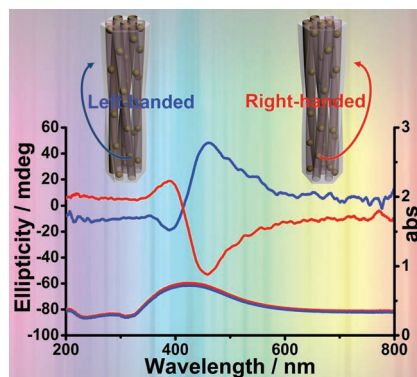


Sub-diffraction-limited resolution and high contrast thin flat silver superlenses (Ag, Ag/Ni and Ag/Ge) are engineered through refining interfacial roughness to reduce scattering loss. A smooth interface is a key factor for realizing high performance superlens lithography while the seed layer effect is detrimental. This report verifies that a superlens is a potential tool for nano-photolithography.

## Chiral Nanoparticles

J. Xie, Y. Duan, S. Che\* .....3784–3792

## Chirality of Metal Nanoparticles in Chiral Mesoporous Silica

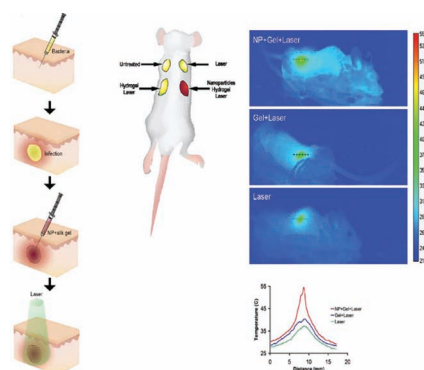


Chiroptical properties of pure inorganic material are investigated in detail by dispersing achiral Ag nanoparticles into chiral mesoporous silica, due to collectively asymmetric plasmon interactions of Ag nanoparticles arranged along helical pore orientation. Such chiral material will develop novel applications for biosensing, chiral recognition and detection based on the importance of surface plasmon resonance (SPR) in biological structures.



## FULL PAPERS

A single 10-minute laser treatment of a subcutaneous infection in mice preserves the general tissue architecture, while achieving a bactericidal effect, and even results in complete eradication in some cases. The unique materials platform presented can provide the basis for an alternative treatment of focal infections.

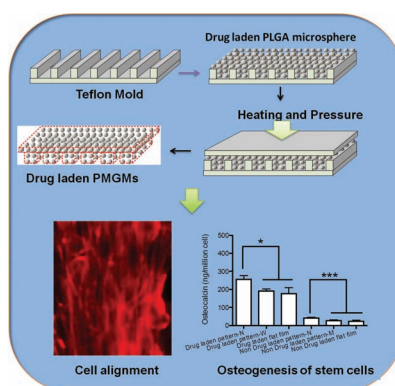


## Biomedical Applications

N. Kojic, E. M. Pritchard, H. Tao, M. A. Brenckle, J. P. Mondia, B. Panilaitis, F. Omenetto,\* D. L. Kaplan\* .....3793–3798

## Focal Infection Treatment using Laser-Mediated Heating of Injectable Silk Hydrogels with Gold Nanoparticles

Newly developed cell differentiated factor laden, polymer microsphere-based grooved micropatterns (PMGMs) generated by Teflon microfluidic chips that combine topographical and chemical/biological cues promote better osteogenic commitment of adipose-derived stem cells when compared with an individual stimulation alone. The results indicate the great potential of PMGMs for stem cell research and bone regenerative therapies.

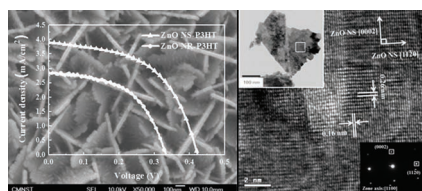


## Micropatterning

X. T. Shi,\* S. Chen, J. H. Zhou, H. J. Yu, L. Li, H. K. Wu\* .....3799–3807

## Directing Osteogenesis of Stem Cells with Drug-Laden, Polymer-Microsphere-Based Micropatterns Generated by Teflon Microfluidic Chips

The ZnO nanosheet (NS) framework, which is constructed on nanoneedles/indium tin oxide substrate via a room-temperature chemical bath deposition, exhibits a slightly larger absorption edge and a faster electron transport rate compared to a ZnO nanorod (NR) array. Superior interface properties are demonstrated in the ZnO NS framework/poly(3-hexylthiophene) (P3HT) hybrid, resulting in an enhancement of the solar cell efficiency compared to ZnO NR-P3HT hybrid.

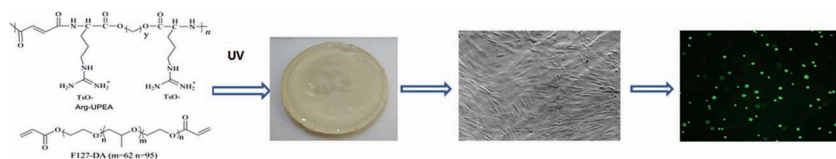


## Solar Cells

Y.-H. Sung, W.-P. Liao, D.-W. Chen, C.-T. Wu, G.-J. Chang, J.-J. Wu\* .....3808–3814

## Room-Temperature Tailoring of Vertical ZnO Nanoarchitecture Morphology for Efficient Hybrid Polymer Solar Cells

Unsaturated arginine poly(ester amide)s are prepared to improve the biocompatibility of Pluronic hydrogels. This is confirmed using fibroblast cell attachment and endothelial cell encapsulation tests.



## Hydrogels

J. Wu, D. Wu, M. A. Mutschler, C.-C. Chu\* .....3815–3823

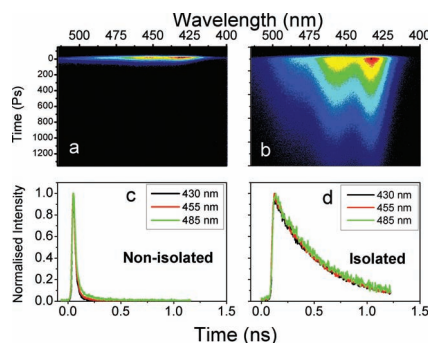
## Cationic Hybrid Hydrogels from Amino-Acid-Based Poly(ester amide): Fabrication, Characterization, and Biological Properties

## FULL PAPERS

## Conjugated Polymers

H. A. Al-Attar,\*  
A. P. Monkman .....3824–3832

**Room-Temperature Phosphorescence From Films of Isolated Water-Soluble Conjugated Polymers in Hydrogen-Bonded Matrices**

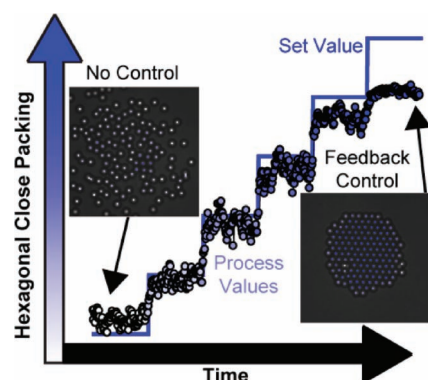


A new method to fabricate solid films with densely packed near perfectly isolated, non-interacting luminescent polymer chains that yield very high photoluminescence quantum yield (PLQY) and, more remarkably, room temperature phosphorescence is reported. The method uses water-soluble conjugated polymers (WSCP) and a polymeric surfactant such as poly(vinyl alcohol) (PVA) and poly(vinyl pyrrolidone) (PVP).

## Self-Assembly

J. J. Juárez, M. A. Bevan\* .....3833–3839

**Feedback Controlled Colloidal Self-Assembly**

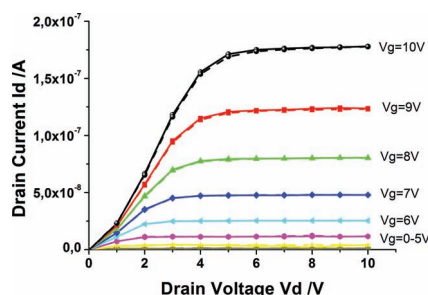


A conceptually new approach that employs feedback control to quantitatively and reversibly guide the dynamic evolution of a colloidal self-assembly process is demonstrated. The key to this approach is the use of free energy landscape models to inform feedback control laws that close the loop between real-time sensing of order parameters and electric field mediated actuation.

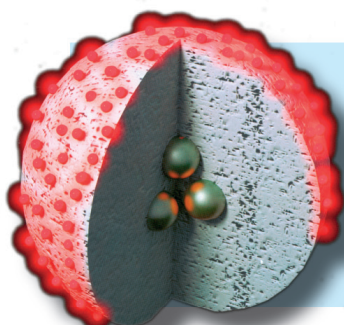
## Organic Electronics

I. Tsydel,\* M. Kucinska, T. Marszalek, R. Rybakiewicz, A. Nosal, J. Jung, M. Gazicki-Lipman, C. Pitsalidis, C. Gravalidis, S. Logothetidis, M. Zagorska, J. Ulanski .....3840–3844

**High-Mobility and Low Turn-On Voltage n-Channel OTFTs Based on a Solution-Processable Derivative of Naphthalene Bisimide**



Application of zone-casting technique leads to air-operating n-channel field-effect transistors (FETs), which have electron mobility that is improved by three orders of magnitude compared to transistors in which the same active layer, a naphthalene bisimide derivative, is spin-coated.



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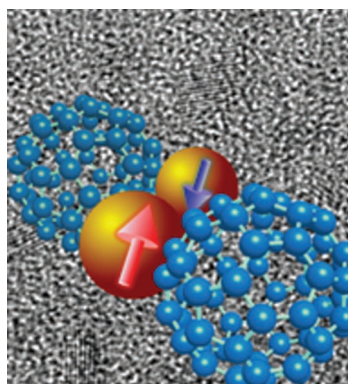
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## FULL PAPERS

**Co nanoparticles in a C<sub>60</sub> matrix are studied** and a novel magnetoelectric coupling appears in the material. A large magnetic switching effect is realized, where an external electric field controls the magnetization alignment of the Co nanoparticles and an external magnetic field controls the charging states of the nanoparticles.

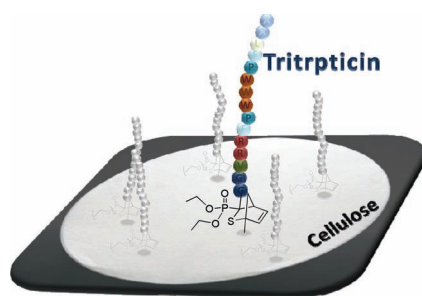


### Magnetic Materials

Y. Sakai, E. Tamura,\* S. Toyokawa,  
E. Shikoh, V. K. Lazarov, A. Hirohata,  
T. Shinjo, Y. Suzuki,  
M. Shiraishi\* .....3845–3852

### Observation of Magnetic-Switching and Multiferroic-Like Behavior of Co Nanoparticles in a C<sub>60</sub> Matrix

**Peptide-cellulose hybrid materials are generated via mild hetero Diels-Alder (HDA) cycloaddition** to graft thioamide functional oligopeptides onto solid cellulose substrates bearing highly reactive cyclopentadienyl moieties. Oligopeptides (including antimicrobial tritrypticin) are functionalized employing strongly electron deficient thiocarbonyl thio compounds resulting in biomacromolecules bearing a thioamide endgroup. In-depth investigations in solution and on the solid peptide functional substrate were performed confirming a highly efficient grafting process.

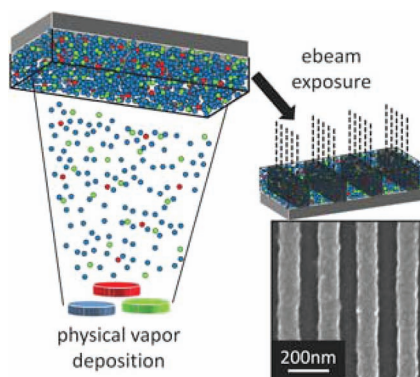


### Hybrid Materials

T. Tischer, A. S. Goldmann, K. Linkert,  
V. Trouillet, H. G. Börner,\*  
C. Barner-Kowollik\* .....3853–3864

### Modular Ligation of Thioamide Functional Peptides onto Solid Cellulose Substrates

**The homogeneous molecular material distribution of a multicomponent molecular glass electron beam resist** is systematically investigated for solvent-based spin-coated and solvent-free physical vapor deposited thin films. The applied screening methods and combinatorial optimizations allow the investigation of dissolution behavior, resist sensitivity, and overall lithographic performance demonstrating the advantages of applying physical vapor deposition for film preparation.

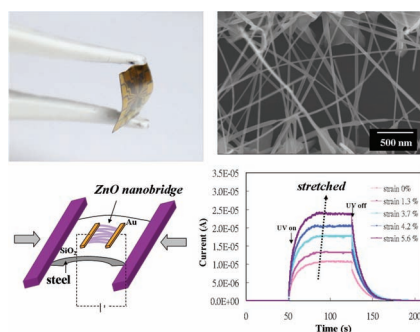


### Thin Films

T. Kolb, C. Neuber, M. Krysak,  
C. K. Ober, H.-W. Schmidt\* .....3865–3873

### Multicomponent Physical Vapor Deposited Films with Homogeneous Molecular Material Distribution Featuring Improved Resist Sensitivity

**A high-performance flexible nanowire (NW)-array photosensor with a large area** is developed by directly growing horizontal single-crystalline Al-doped ZnO NW arrays across Au microelectrodes patterned on flexible SiO<sub>2</sub>/steel substrates. The device shows excellent photosensing properties with an ultra-high UV/visible rejection ratio, photoresponsivity, and sensitivity. Tensile strain further enhances the photoresponsivity, which is due to a piezo-photonic effect.



### Photosensors

R.-C. Wang,\* H.-Y. Lin, C.-H. Wang,  
C.-P. Liu\* .....3875–3881

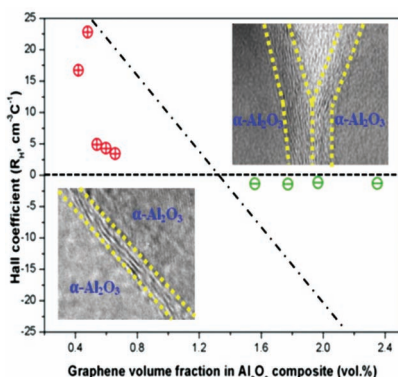
### Fabrication of a Large-Area Al-Doped ZnO Nanowire Array Photosensor with Enhanced Photoresponse by Straining

## FULL PAPERS

## Graphene

Y. Fan, W. Jiang,  
A. Kawasaki\*.....3882–3889

### Highly Conductive Few-Layer Graphene/ $\text{Al}_2\text{O}_3$ Nanocomposites with Tunable Charge Carrier Type



Few-layer graphene/ $\text{Al}_2\text{O}_3$  nanocomposites with good electrical conductivity are prepared by using a titration method followed by spark plasma sintering. It is found that the charge carrier type changes from p-type to n-type as graphene content becomes higher. This conversion is deduced to be related to the doping effect induced by  $\text{Al}_2\text{O}_3$  matrix and is thickness-dependent with respect to few-layer graphene.

## Surface Modification

T. Jalkanen, E. Mäkilä, Y.-I. Suzuki,  
T. Urata, K. Fukami, T. Sakka,  
J. Salonen,\* Y. H. Ogata.....3890–3898

### Studies on Chemical Modification of Porous Silicon-Based Graded-Index Optical Microcavities for Improved Stability Under Alkaline Conditions

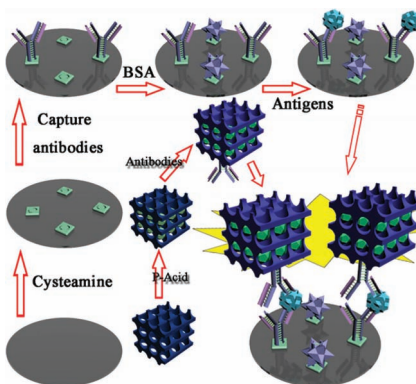


Chemical modification of graded-index microcavities based on porous silicon is studied with the aim of producing a functionalized surface with improved stability under alkaline conditions. Thermal carbonization treatments are employed for obtaining microcavities with hydrophilic and hydrophobic surface terminations, respectively. Addition of undecylenic acid to the hydrocarbonized surface is demonstrated as a viable method for obtaining functionalized surface with greatly improved stability.

## Nanotubes

M. Yan, L. Ge, W. Q. Gao, J. Yu,\*  
X. R. Song, S. G. Ge, Z. Y. Jia,  
C. C. Chu.....3899–3906

### Electrogenerated Chemiluminescence from a Phenyleneethynylene Derivative and its Ultrasensitive Immunosensing Application Using a Nanotubular Mesoporous Pt–Ag Alloy for Signal Amplification

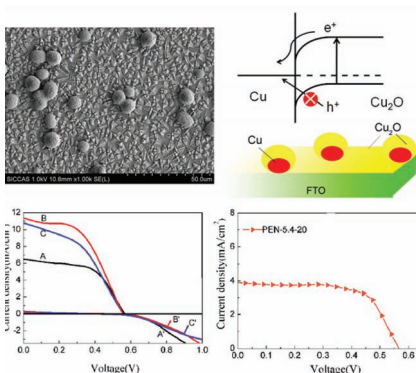


Electrogenerated chemiluminescence (ECL) of novel phenyleneethynylene derivatives with carboxylic acid groups at the para positions in aqueous solutions is studied and the first immunosensing applications are shown. This ECL immunosensor exhibits high sensitivity, good reproducibility, rapid response, and long-term stability.

## Solar Cells

F. Shao, J. Sun,\* L. Gao, J. Luo, Y. Liu,  
S. Yang .....3907–3913

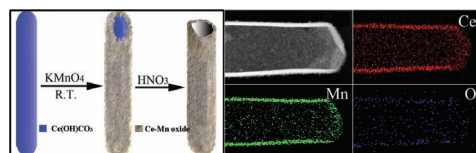
### High Efficiency Semiconductor-Liquid Junction Solar Cells based on $\text{Cu}/\text{Cu}_2\text{O}$



The abundance and nontoxicity of  $\text{Cu}_2\text{O}$ , as well as the good solar spectral absorption, allows for the possibility of constructing efficient solar cells with  $\text{Cu}_2\text{O}$ .  $\text{Cu}/\text{Cu}_2\text{O}$  is prepared by a low-cost and easily scalable solution processing technique; the solar cells based on the obtained  $\text{Cu}/\text{Cu}_2\text{O}$  have a best conversion efficiency of 3.13% under simulated AM1.5G. The efficiency of the flexible cell based on  $\text{Cu}/\text{Cu}_2\text{O}$  films is 1.44%.

## FULL PAPER

**Ce–Mn oxide nanotubes are fabricated by an interfacial reaction** between  $\text{Ce}(\text{OH})\text{CO}_3$  templates and  $\text{KMnO}_4$  aqueous solution, followed by selective wash with  $\text{HNO}_3$ . The resulting nanotubes exhibit higher catalytic activity towards CO oxidation and excellent adsorption capacity of Congo red. This work represents a demonstration of an interfacial oxidation–reduction reaction-engaged process capable of generating binary oxide hollow nanostructures with different shapes and compositions.

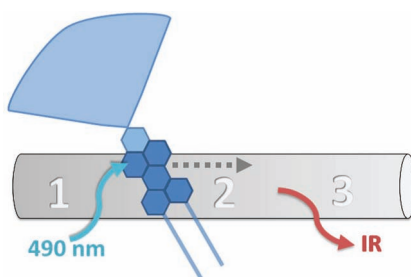


## Catalytic Nanotubes

G. Chen, F. Rosei, D. Ma\* .....3914–3920

**Interfacial Reaction-Directed Synthesis of Ce–Mn Binary Oxide Nanotubes and Their Applications in CO Oxidation and Water Treatment**

**A novel perylene-based functional surfactant that suspends nanotubes independent of pH** in aqueous solution and forms energy transfer complexes with the nanotube is presented. Upon excitation of the adsorbant an emission from the nanotube is observed, while the adsorbant emission is completely quenched. This energy transfer between the perylene core and the nanotube in biocompatible environments is demonstrated using photoluminescence and is observed for all chiralities.

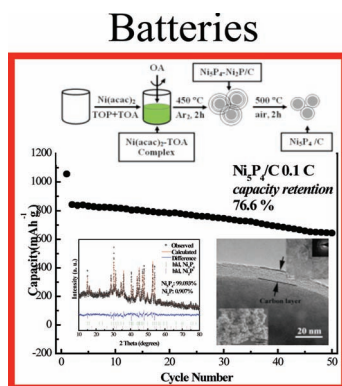


## Photoluminescence

F. Ernst,\* T. Heek, A. Setaro, R. Haag, S. Reich .....3921–3926

**Energy Transfer in Nanotube-Perylene Complexes**

**Monophase nickel phosphide/carbon ( $\text{Ni}_5\text{P}_4/\text{C}$ ) composites** are controllably synthesized via a two-step reaction: a wet-chemistry reaction and a solid-state reaction. Highly crystalline  $\text{Ni}_5\text{P}_4/\text{C}$  spheres with uniform morphology are obtained. The composites exhibit a high specific capacity and rate performance, as well as good cyclic stability, making them one of the most promising anode materials for lithium-ion batteries.

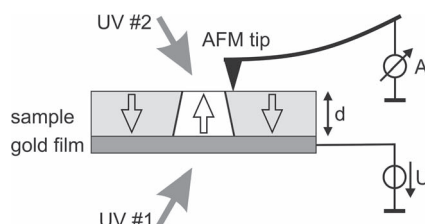


## Batteries

Y. Lu, J.-P. Tu,\* Q.-Q. Xiong, J.-Y. Xiang, Y.-J. Mai, J. Zhang, Y.-Q. Qiao, X.-L. Wang,\* C.-D. Gu, S. X. Mao .....3927–3935

**Controllable Synthesis of a Monophase Nickel Phosphide/Carbon ( $\text{Ni}_5\text{P}_4/\text{C}$ ) Composite Electrode via Wet-Chemistry and a Solid-State Reaction for the Anode in Lithium Secondary Batteries**

**Domain walls (DWs) in highly insulating, millimeter thick lithium niobate ( $\text{LiNbO}_3$ ) single crystals** show a pronounced electric conductivity when being illuminated by super-bandgap light. This conductivity can be tuned by engineering the tilting angle of the DWs with respect to the polar axis. The results are obtained using conductive atomic force microscopy and macroscopic transport measurements at various temperatures.



## Ferroelectrics

M. Schröder, A. Haußmann, A. Thiessen, E. Soergel, T. Woike, L. M. Eng\* .....3936–3944

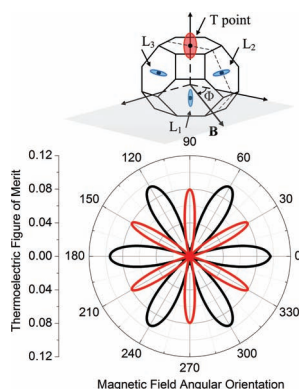
**Conducting Domain Walls in Lithium Niobate Single Crystals**

## FULL PAPERS

### Thermoelectric Materials

A. Popescu,  
L. M. Woods\* .....3945–3949

#### Valleytronics, Carrier Filtering and Thermoelectricity in Bismuth: Magnetic Field Polarization Effects

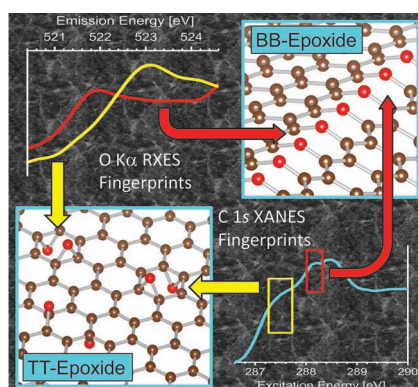


**Transport control in bismuth by tuning its charge** and thermal properties via the orientation of an applied external magnetic field is proposed. It is demonstrated that the direction dependent polarization effects of the magnetic field can remove the Dirac valley degeneracy in bismuth electronic structure suggesting that valleytronics is a possible way to control thermoelectricity.

### Graphene

A. Hunt, D. A. Dikin, E. Z. Kurmaev,  
T. D. Boyko, P. Bazylewski,  
G. S. Chang, A. Moewes\* .....3950–3957

#### Epoxide Speciation and Functional Group Distribution in Graphene Oxide Paper-Like Materials

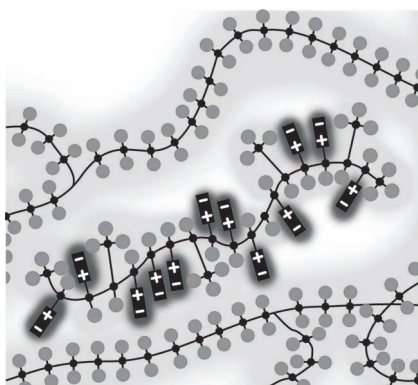


**When epoxides align, they can unzip the graphene lattice.** This process is an aging effect that may be mitigated using intercalation of dodecylamine. The randomly placed epoxides and the aligned epoxides are easily differentiated using either C 1s X-ray absorption near-edge structure spectroscopy (XANES) or O Kα resonant X-ray emission spectroscopy (RXES) techniques.

### Actuators

S. Risse,\* B. Kussmaul, H. Krüger,  
G. Kofod.....3958–3962

#### Synergistic Improvement of Actuation Properties with Compatibilized High Permittivity Filler



**Electric field-driven actuator materials are improved by an increase in permittivity** and by a reduction in stiffness. A synergistic enhancement method is presented that is based on a macromolecular plasticizing filler molecule with a combination of both high dipole moment and compatibilizer moieties, synthesized to simultaneously ensure improvement of electromechanical properties and compatibility with the host matrix.