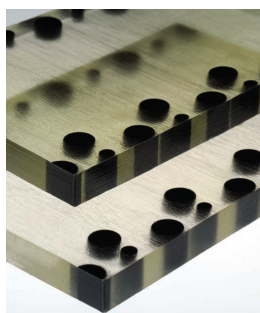


# ADVANCED FUNCTIONAL MATERIALS

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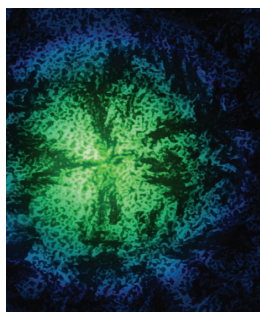
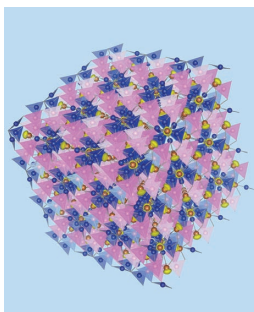


## Surface Engineering

Creating tunable and reversible surfaces is an interesting challenge with no single solution. On page 3641, M. Guttag and M. C. Boyce present a new approach using a composite of stiff particles embedded into a soft matrix which provides a wide range of tunable and locally controllable surface topographies. By applying an external load to the composite, complex surface topographies take shape. (Photo credit: Felice C. Frankel.)

## Thermoelectrics

Tetrahedrites were recently discovered to be high-performance thermoelectrics but the chemical and structural origins of the rattling guests and resultant low thermal conductivity remain elusive. On page 3648, W. La and team establish a clear connection between the local bonding asymmetry and anharmonic rattling modes in  $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$  tetrahedrites, enabled by the chemically active electron lone pairs.

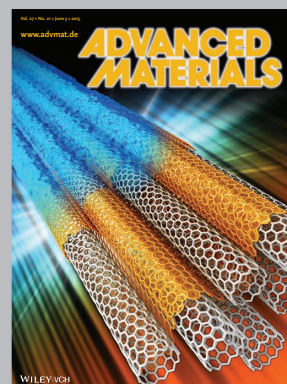
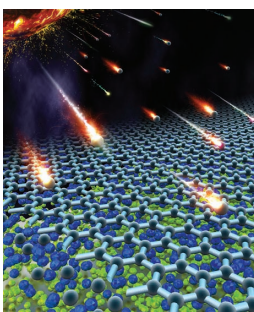


## Organic Electronics

In the study presented by K. Cho, S. Jung, J.-J. Kim, and co-workers on page 3658, partial crosslinking of polymer gate dielectrics (pc-PVP:PMF) allows semiconducting small molecules in solvent to permeate into it. The solvent evaporation during spinning promotes the extraction of TIPS-Pentacene solution onto pc-PVP:PMF surface. The residual solvent in pc-PVP:PMF network evaporates slowly, helping millimeter-sized crystallization of TIPS-Pentacene molecules. Consequently, the OFET devices exhibit high mobilities with maximum value of  $3.40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ .

## Graphene

On page 3666, Q. Yuan, Z. Di, and colleagues take advantage of the dual metal substrate of Ni-coated Cu foils to precisely control layer number of graphene by ion implantation and the layer number of graphene strictly corresponds to the implantation fluence as expected. Besides, the formation mechanism is explored in detail by the experimental analysis and confirmed by theoretical calculations.



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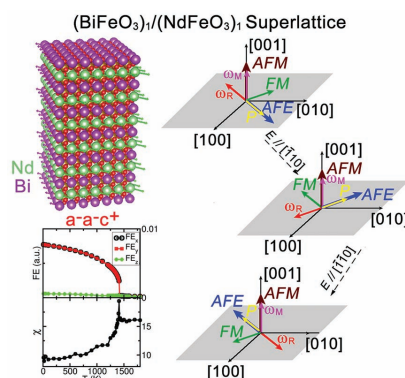
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## FULL PAPERS

The hybrid improper ferroelectricity (HIF) mechanism generates electrical polarization in a superlattice made of two nonpolar materials and is promising for the design of novel multiferroics. A first-principles-based technique is used here to resolve unknown issues about HIF, including the discovery of a path allowing the switching of polarization and magnetization, and how the HIF transition occurs upon cooling.

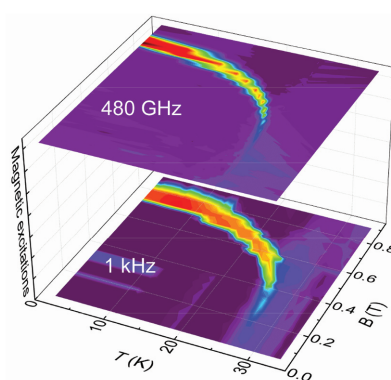


## Multiferroics

B. Xu,\* D. Wang, H. J. Zhao, J. Íñiguez, X. M. Chen, L. Bellaiche\* .....3626–3633

**Hybrid Improper Ferroelectricity in Multiferroic Superlattices: Finite-Temperature Properties and Electric-Field-Driven Switching of Polarization and Magnetization**

Controllable broadband microwave absorption is highly desired for improved electromagnetic shielding and superior signal processing. The absorption of the microwaves in an extremely wide frequency range is demonstrated in layered metamagnets. The effect is controlled by the external magnetic field and should be easily reproduced in artificial metamagnets, that is, magnetic multilayers, allowing a direct tuning of the functional properties.

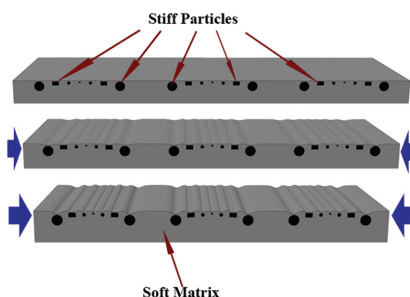


## Microwave Filtering

M. Pregelj,\* O. Zaharko, A. Zorko, M. Gomilšek, O. Sendetskyi, A. Günther, M. Ozerov, S. A. Zvyagin, H. Luetkens, C. Baines, V. Tsurkan, A. Loidl .....3634–3640

**Controllable Broadband Absorption in the Mixed Phase of Metamagnets**

Creating tunable and reversible surfaces is an interesting challenge with no single solution. A new approach using a composite of stiff particles embedded into a soft matrix provides a wide range of tunable and locally controllable surface topographies. By applying an external load to the composite, complex surface topographies take shape.

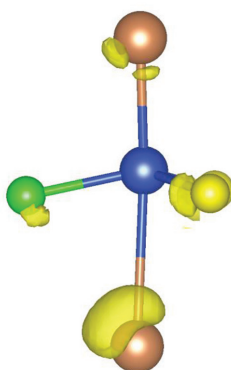


## Surface Engineering

M. Gutttag,\* M. C. Boyce .....3641–3647

**Locally and Dynamically Controllable Surface Topography Through the Use of Particle-Enhanced Soft Composites**

A strong local bonding asymmetry is identified inside a  $\text{Sb}[\text{CuS}_3]\text{Sb}$  atomic cage, in which Cu (blue) forms strong covalent bonding with S (green and yellow) and forms weak covalent bonding with one of the Sb atoms (brown) enabled by the lone-pair electrons. This bonding asymmetry causes the out-of-plane anharmonic rattling which leads to the low thermal conductivity.



## Thermoelectrics

W. Lai,\* Y. Wang, D. T. Morelli, X. Lu .....3648–3657

**From Bonding Asymmetry to Anharmonic Rattling in  $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$  Tetrahedrites: When Lone-Pair Electrons Are Not So Lonely**

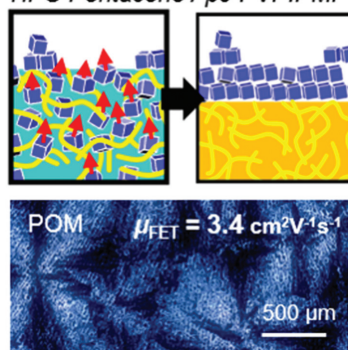
## FULL PAPERS

## Organic Electronics

H. Yoo, H. H. Choi, T. J. Shin, T. Rim,  
K. Cho,\* S. Jung,\* J.-J. Kim\*...3658–3665

**Self-Assembled, Millimeter-Sized  
TIPS-Pentacene Spherulites Grown  
on Partially Crosslinked Polymer Gate  
Dielectric**

**TIPS-Pentacene / pc-PVP:PMF**

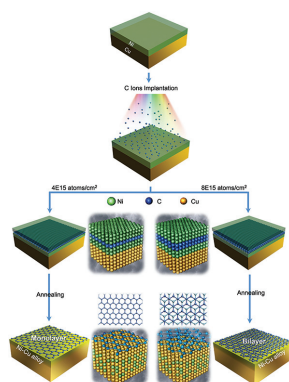


**Partial crosslinking of polymer gate-dielectrics (pc-PVP:PMF)** allows semi-conducting small molecules in solvent to permeate into it. The solvent evaporation during spinning promotes the extraction of TIPS-Pentacene solution onto pc-PVP:PMF surface. The residual solvent in pc-PVP:PMF network evaporates slowly, so it helps millimeter-sized crystallization of TIPS-Pentacene molecules. Consequently, the OFET devices exhibit high mobilities with maximum value of  $3.40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ .

## Graphene

G. Wang, M. Zhang, S. Liu, X. Xie,  
G. Ding, Y. Wang, P. K. Chu, H. Gao,  
W. Ren, Q. Yuan,\* P. Zhang, X. Wang,  
Z. Di\* ..... 3666–3675

**Synthesis of Layer-Tunable Graphene:  
A Combined Kinetic Implantation and  
Thermal Ejection Approach**

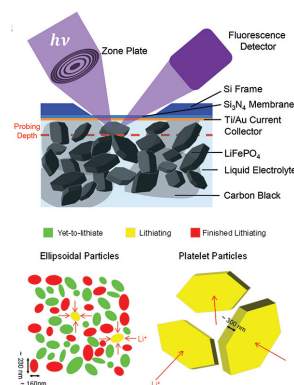


**By taking advantage of the dual metal substrate of Ni-coated Cu foils**, the precise control of layer number of graphene by ion implantation is demonstrated and the layer number of graphene strictly corresponds to the implantation fluence as expected. Besides, the formation mechanism is explored by the experimental analysis in detail and confirmed by the theoretical calculations.

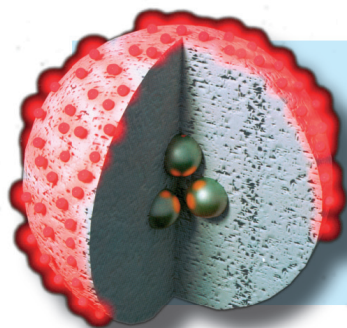
## Fluorescence

Y. Li, J. Nelson Weker, W. E. Gent,  
D. N. Mueller, J. Lim, D. A. Cogswell,  
T. Tyliszczak, W. C. Chueh\* ...3677–3687

**Dichotomy in the Lithiation Pathway of  
Ellipsoidal and Platelet  $\text{LiFePO}_4$  Particles  
Revealed through Nanoscale Operando  
State-of-Charge Imaging**



**A nanoscale liquid imaging platform** is developed to track lithium intercalation in  $\text{LiFePO}_4$  electrodes. The results show that the intercalation pathway strongly depends on the particle morphology and synthesis protocol. Highly faceted ellipsoidal particles intercalate sequentially, with a small number of actively intercalating particles and low electrode utilization. Platelet particles, on the other hand, intercalate simultaneously with a more uniform current distribution.



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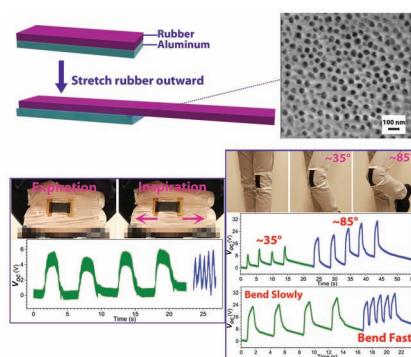
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## FULL PAPERS

A **stretchable-rubber-based triboelectric nanogenerator** is developed, which can not only harvest energy but also serve as self-powered multifunctional sensors. It is composed of a layer of elastic rubber and a layer of aluminum film that acts as the electrode. Electrical outputs are generated by stretching and releasing the rubber. It can be attached to a human body to detect diaphragm breathing and joint motion.



## Biomedical Monitoring

F. Yi, L. Lin, S. Niu, P. K. Yang, Z. Wang, J. Chen, Y. Zhou, Y. Zi, J. Wang, Q. Liao, Y. Zhang,\* Z. L. Wang\* .....3688–3696

## Stretchable-Rubber-Based Triboelectric Nanogenerator and Its Application as Self-Powered Body Motion Sensors

**Human skin-like core/shell material structure** is presented for use in wearable, stretchable electronic systems. Here, an ultralow-modulus elastomer (core) with a thin enclosure (shell) serves to minimize interface stresses and mechanical constraints on natural motions, with ability to strain-isolate the electronics. Demonstration examples exploit emerging commercial classes of stretchable electronic system to wirelessly monitor a subject's motion and body temperature during exercise.

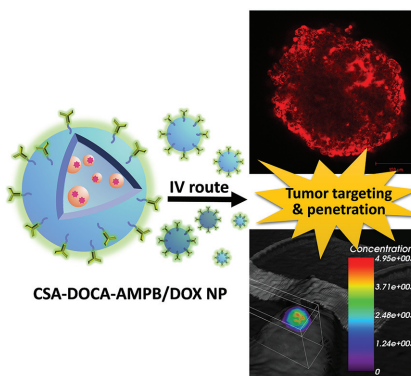


## Epidermal Systems

C. H. Lee, Y. Ma, K.-I. Jang, A. Banks, T. Pan, X. Feng, J. S. Kim, D. Kang, M. S. Raj, B. L. McGrane, B. Morey, X. Wang, R. Ghaffari, Y. Huang,\* J. A. Rogers\* .....3698–3704

## Soft Core/Shell Packages for Stretchable Electronics

(3-Aminomethylphenyl)boronic acid-(AMPB)-functionalized chondroitin sulfate A (CSA)-deoxycholic acid (DOCA)-based nanoparticles (NPs) are prepared for anticancer drug delivery and cancer diagnosis. Doxorubicin (DOX)-loaded CSA-DOCA-AMPB NPs exhibit improved targeting, penetration, and therapeutic efficacies for CD44 receptor-expressed tumors, compared to CSA-DOCA NPs, via CSA-CD44 receptor and boronic-acid-sialic-acid interactions.

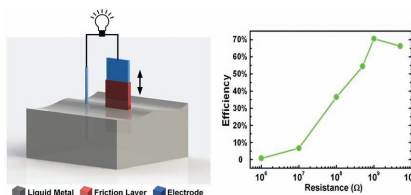


## Drug Delivery

J.-Y. Lee, S.-J. Chung, H.-J. Cho,\* D.-D. Kim\* .....3705–3717

## Phenylboronic Acid-Decorated Chondroitin Sulfate A-Based Theranostic Nanoparticles for Enhanced Tumor Targeting and Penetration

A **liquid-metal-based triboelectric nanogenerator** (LM-TENG) is developed for high power generation through conversion of mechanical energy, which allows total contact between the metal and the dielectric. The LM-TENG exhibits a high output charge density of  $430 \mu\text{C m}^{-2}$ , which is four to five times of that using a solid thin film electrode. An instantaneous energy conversion efficiency as high as 70.6% is demonstrated.



## Energy Harvesting

W. Tang, T. Jiang, F. R. Fan, A. F. Yu, C. Zhang, X. Cao,\* Z. L. Wang\* .....3718–3725

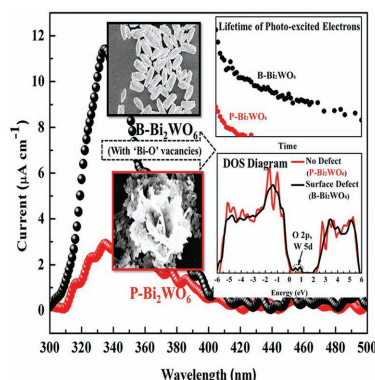
## Liquid-Metal Electrode for High-Performance Triboelectric Nanogenerator at an Instantaneous Energy Conversion Efficiency of 70.6%

## FULL PAPERS

## Photocatalysts

G. Zhang, Z. Y. Hu, M. Sun, Y. Liu,  
L. M. Liu, H. J. Liu,\* C.-P. Huang,  
J. H. Qu, J. H. Li\* ..... 3726–3734

### Formation of $\text{Bi}_2\text{WO}_6$ Bipyramids with Vacancy Pairs for Enhanced Solar-Driven Photoactivity



$\text{Bi}_2\text{WO}_6$  nanobipyramid is successfully fabricated via a facile strategy in which “Bi–O” vacancy pairs are key to increase its solar-light photoactivity.

## Photoresists

A. S. Quick, A. de los Santos Pereira,  
M. Bruns, T. Bückmann,  
C. Rodriguez-Emmenegger,\* M. Wegener,\*  
C. Barner-Kowollik\* ..... 3735–3744

### Rapid Thiol-Yne-Mediated Fabrication and Dual Postfunctionalization of Micro-Resolved 3D Mesostructures

Reactive 3D mesostructures with micrometer feature size are fabricated via dip-in direct laser writing employing the radical thiol-yne coupling reaction. Axial resolution and the polymerization process of the photoresist are investigated. Residual thiols and alkynes are exploited for postmodification reactions, namely thiol-Michael addition and copper-catalyzed azide alkyne cycloaddition, demonstrating successful dual functionalization throughout the structure and on the surface.

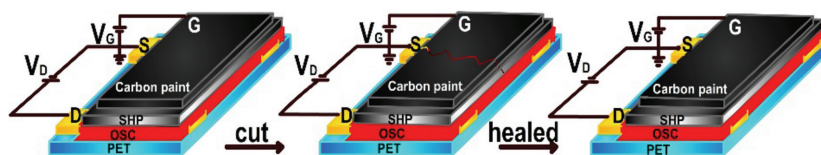


## Organic Electronics

W. Huang, K. Besar, Y. Zhang, S. Yang,  
G. Wiedman, Y. Liu, W. Guo, J. Song,  
K. Hemker, K. Hristova, I. J. Kymissis,  
H. E. Katz\* ..... 3745–3755

### A High-Capacitance Salt-Free Dielectric for Self-Healable, Printable, and Flexible Organic Field Effect Transistors and Chemical Sensor

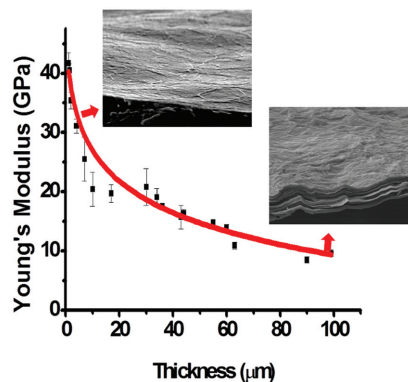
Printable and flexible, low-voltage operating, particularly self-healable electronics is a highly desirable suite of technologies proposed for future intelligent electronic devices such as body monitors and window displays. A key material used in printed logic circuits is reported, the “gate dielectric” insulator, with which all these attributes for the first time are demonstrated.



## Structure–Property Relationship

T. Gong, D. V. Lam, R. Liu, S. Won,  
Y. Hwangbo, S. Kwon, J. Kim,  
K. Sun, J.-H. Kim, S.-M. Lee,  
C. Lee\* ..... 3756–3763

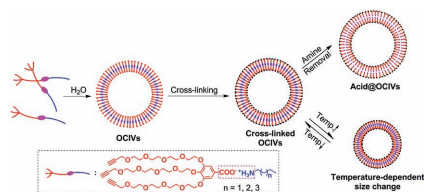
### Thickness Dependence of the Mechanical Properties of Free-Standing Graphene Oxide Papers



The mechanical properties of graphene oxide (GO) papers are shown to be thickness-dependent. This dependence arises from the micro- and macrostructure formed during the fabrication process; microscopy studies reveal that corrugations, void defects, and surface wrinkles all influence the mechanical properties. Establishing this structure–property relationship is expected to enable improved guidelines for the application of GO to mechanical structures.

## FULL PAPERS

A general concept of organic counterion-induced vesicles is established. The introduction of organic counterion not only plays an important role in vesicle construction, but also endows the material with greatly practical values. As examples, a robust luminescence temperature sensor and a highly efficient synergistic catalyst are developed successfully.

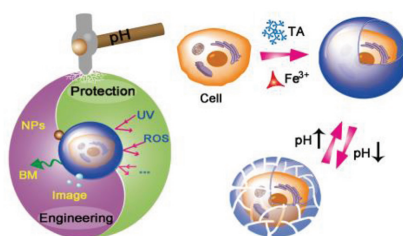


### Synergistic Catalysis

C. Li, S. Zhang,\* J. Pang, Y. Wu, Z. Gu\* .....3764–3774

**Facile Fabrication of Robust Organic Counterion-Induced Vesicles: Reversible Thermal Behavior for Optical Temperature Sensor and Synergistic Catalyst upon Removal of Amine**

Inspired by nature, herein a versatile strategy is reported for encapsulating cells with polyphenolic metal coordination-based shell. The shell provides powerful tools for simultaneously protecting cells and engineering cell surface with nanoparticles, bioactive molecules, and imaging agents. In addition, in contrast to traditional passive shell, the coating here can be removed on-demand with stimuli to evoke the original feature of cells.

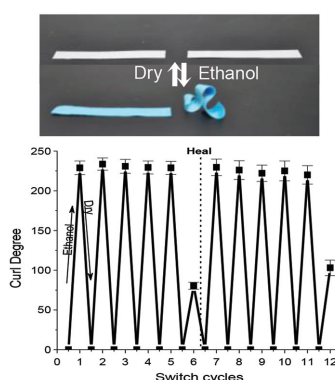


### Cell Encapsulation

W. Li, W. Bing, S. Huang, J. Ren, X. Qu\* .....3775–3784

**Mussel Byssus-Like Reversible Metal-Chelated Supramolecular Complex Used for Dynamic Cellular Surface Engineering and Imaging**

Self-healable adhesive that actuates upon exposure to ethanol is developed by layer-by-layer assembly and paste-to-curl approaches. The degree of curling is easily controlled by controlling exposure to the organic solvent stimulus as well as engineering the thickness and modulus of the inert part of the actuator. The branched poly(ethylenimine)/poly(acrylic acid) film, which is the active component, can easily be self-healed in water, prolonging lifetime of the actuator.



### Actuating Materials

Y. Gu, N. S. Zacharia\* .....3785–3792

**Self-Healing Actuating Adhesive Based on Polyelectrolyte Multilayers**