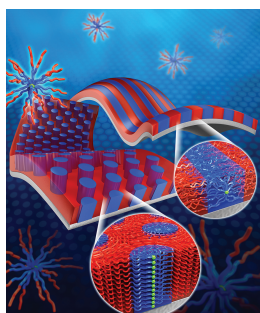


# ADVANCED FUNCTIONAL MATERIALS

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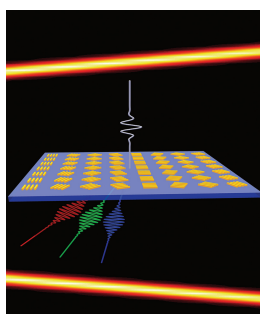
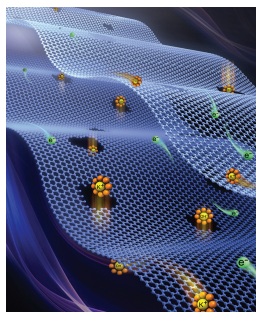


## Oriented Nanodomains

On page 5414 J.K. Kim and co-workers use star-shaped 18 arm poly(methyl methacrylate)-block-polystyrene copolymers to demonstrate that cylindrical and lamellar nanodomains can be vertically aligned on flexible substrates without the use of an additional layer to neutralize the preferential affinity between the substrate and each block. This method will contribute toward commercialization of direct self-assembly for next-generation integrated circuit and data storage media.

## Carbon Nanomeshes

The controlled synthesis of thin-sheet hexagonal carbon nanomeshes by chemical vapor deposition, is reported by Z. B. Lei, J. S. Qiu and co-workers on page 5420. The carbon nanomeshes are rich with large, in-plane mesopores, with controlled pore size, density, and geometry. They are created by the etching of carbon atoms with in situ generated  $\text{Fe}_2\text{O}_3$  and they exhibit dramatically accelerated ion kinetics with potential application as electrodes for supercapacitors.

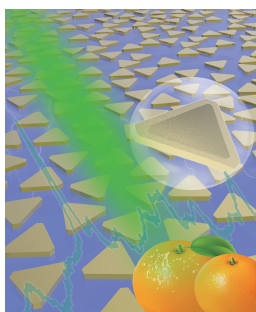


## Beam Deflectors

On page 5428, J. Li, S. Chen, J. Tian and co-workers theoretically and experimentally demonstrate a high performance broadband circularly polarized beam deflector based on multi-rod metasurfaces. The high efficiency and broadband characteristics of the proposed metasurfaces arise from plasmonic hybridization. The polarization degree of the incident light can also be determined using the multirod metasurfaces.

## Core/Shell Nanostructures

An etching-free epitaxial growth method is developed to convert Ag nanostructures of various morphologies into their Ag@Au core/shell counterparts. Anisotropic ones such as nanoplates exhibit excellent plasmonic properties and chemical stability, as shown on page 5435 by C. Gao, Y. Yin and co-workers. As a result, the core/shell structures display high activity and reliability in many plasmon-based applications, such as chemical sensing by surface-enhanced Raman spectroscopy.



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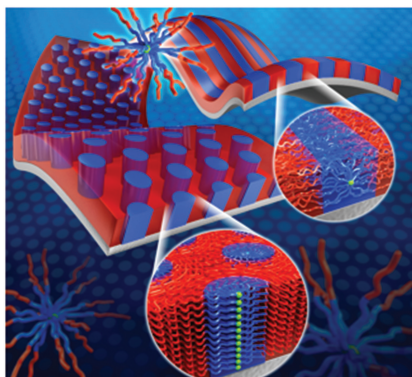
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## FULL PAPERS

**Vertically oriented lamellar and cylindrical nanodomains** are developed by employing 18 arm star-shaped (PMMA-*b*-PS)<sub>18</sub> through a self-neutralization concept. Without a solvent annealing or neutralization process of the substrate, vertically oriented lamellar and cylindrical nanodomains are successfully achieved on various substrates, ranging from semiconductors (Si, SiO<sub>x</sub>) to metal (Au), polymer-brushed substrates, and flexible polymer substrates.

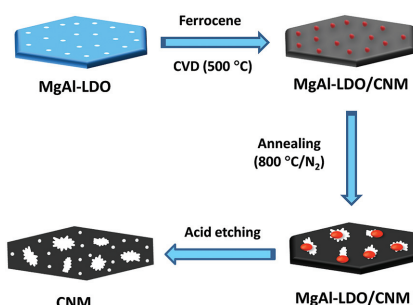


## Oriented Nanodomains

S. Jang, K. Lee, H. C. Moon, J. Kwak, J. Park, G. Jeon, W. B. Lee, J. K. Kim\* .....5414–5419

**Vertical Orientation of Nanodomains on Versatile Substrates through Self-Neutralization Induced by Star-Shaped Block Copolymers**

**Growth of thin-sheet carbon nanomesh (CNM) on MgAl-layered double oxides**, with controllable in-plane pore structure, is reported. The large mesopores with pore size varying from 10 to 50 nm offer additional ion channels that greatly promote ion kinetics across the 2D CNM plane, leading to an improved capacitive performance with much shortened relaxation time constant.

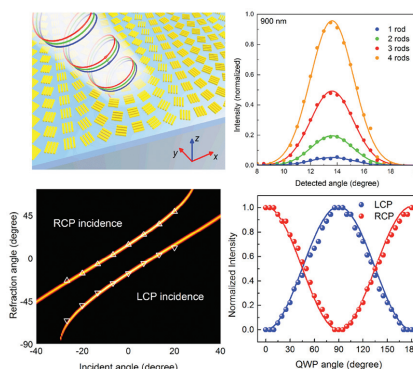


## Carbon Nanomeshes

H. J. Wang, L. Zhi, K. Q. Liu, L. Q. Dang, Z. H. Liu, Z. B. Lei,\* C. Yu, J. S. Qiu\* .....5420–5427

**Thin-Sheet Carbon Nanomesh with an Excellent Electrocapacitive Performance**

**A high-performance broadband circularly polarized beam deflector** is demonstrated based on multirod metasurfaces. The distinct high efficiency and broadband characteristics of the metasurfaces are theoretically and experimentally shown to result from the plasmonic hybridization. Determination of the polarization degree of the incident light based on the Poincaré sphere is shown to be easy.

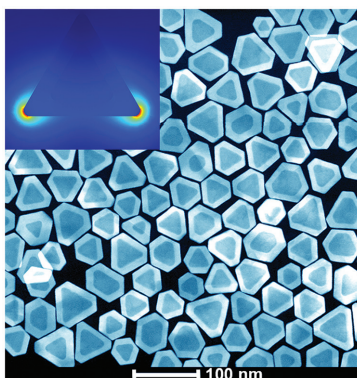


## Beam Deflectors

Z. Liu, Z. Li, Z. Liu, J. Li, H. Cheng, P. Yu, W. Liu, C. Tang, C. Gu, J. Li,\* S. Chen,\* J. Tian\* .....5428–5434

**High-Performance Broadband Circularly Polarized Beam Deflector by Mirror Effect of Multinorod Metasurfaces**

**Ag@Au core/shell nanostructures** are synthesized by epitaxial growth of Au on Ag nanostructures, with the anisotropic ones such as nanoplates showing Ag-like plasmonic properties and high stability. Sulfite plays a critical role in preventing galvanic replacement and oxidative etching of the Ag nanostructures. The resulting nanocrystals may find broad use in many plasmon-based applications for superior activities and durability.



## Core/Shell Nanostructures

H. Liu, T. Liu, L. Zhang, L. Han, C. Gao,\* Y. Yin\* .....5435–5443

**Etching-Free Epitaxial Growth of Gold on Silver Nanostructures for High Chemical Stability and Plasmonic Activity**

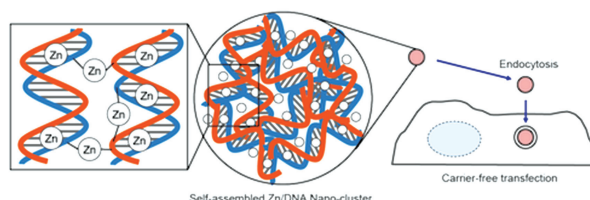
## FULL PAPERS

## Gene Delivery

K. S. Lim, D. Y. Lee, G. M. Valencia,  
Y.-W. Won,\* D. A. Bull\* ..... 5445–5451

**Nano-Self-Assembly of Nucleic Acids  
Capable of Transfection without  
a Gene Carrier**

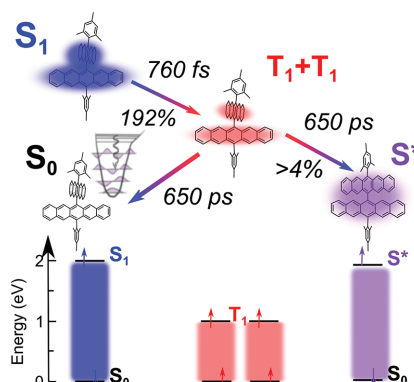
$\text{Zn}^{2+}$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  are able to assemble nucleic acids into a nanocluster that is capable of spontaneous internalization into cells.  $\text{Zn}^{2+}$  shows superior gene transfection efficiency among the three divalent metal ions tested. Various endocytosis mechanisms are involved in the cellular uptake of the Zn/DNA cluster. This study represents a new platform for carrier-free gene delivery.



## Pentacene Dimers

S. Lukman, A. J. Musser, K. Chen,  
S. Athanopoulos, C. K. Yong,  
Z. Zeng, Q. Ye, C. Chi, J. M. Hodgkiss,  
J. Wu,\* R. H. Friend,  
N. C. Greenham\* ..... 5452–5461

**Tuneable Singlet Exciton Fission  
and Triplet–Triplet Annihilation in an  
Orthogonal Pentacene Dimer**

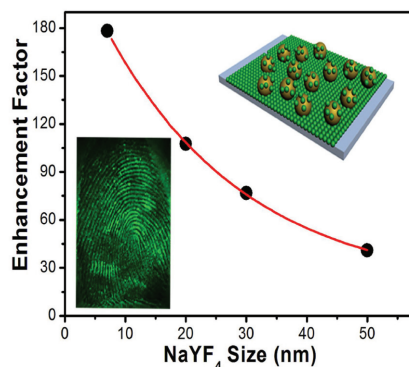


An orthogonal covalently bonded pentacene dimer (DP-Mes) shows nearly quantitative singlet exciton fission on a sub-picosecond timescale that is within the nuclear relaxation time scale. By applying geometrical constraints, the fate of triplet–triplet annihilation is altered. These properties closely depend on molecular geometry and suggest a role for nuclear relaxation in controlling singlet exciton fission and triplet–triplet annihilation.

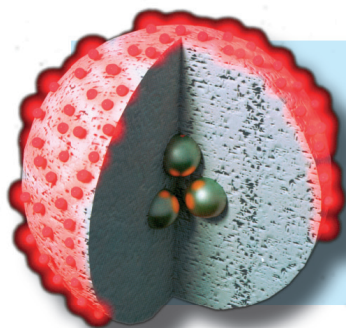
## Surface Plasmons

X. Chen, W. Xu,\* L. H. Zhang, X. Bai,  
S. B. Cui, D. L. Zhou, Z. Yin, H. W. Song,\*  
D.-H. Kim ..... 5462–5471

**Large Upconversion Enhancement  
in the “Islands” Au–Ag Alloy/ $\text{NaYF}_4$ :  
 $\text{Yb}^{3+}$ ,  $\text{Tm}^{3+}$ / $\text{Er}^{3+}$  Composite Films, and  
Fingerprint Identification**



An “islands” Au–Ag/ $\text{NaYF}_4$ :  $\text{Yb}^{3+}$ ,  $\text{Tm}^{3+}$  composite film is prepared by an organic self-assembly and removal template method. The Au–Ag alloy has the stable and repeatable properties and exhibits super-broad surface plasmon resonance band extending to 1100. It obtained  $\approx 180$ -fold enhancement of upconversion luminescence for fingerprint identification.



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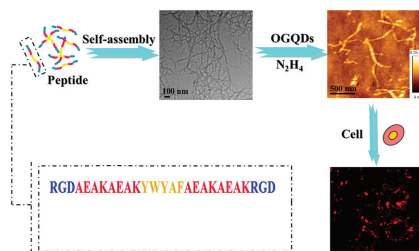
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## FULL PAPERS

**Peptide nanofibers decorated with graphene quantum dots for simultaneous targeting and imaging of tumor cells** are successfully fabricated based on non-covalent interactions. The nanohybrids possess the properties of strong fluorescence, good biocompatibility, and show the capability of labeling live tumor cells specially and efficiently.

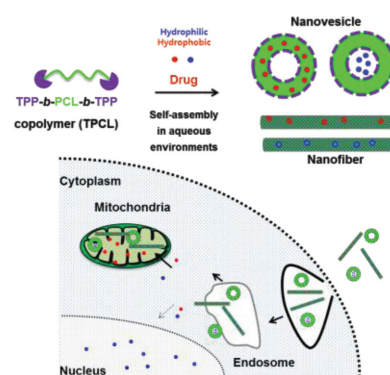


## Cellular Imaging

Z. Su,\* H. Shen, H. Wang, J. Wang, J. Li, G. U. Nienhaus, L. Shang,\* G. Wei\* .....5472–5478

**Motif-Designed Peptide Nanofibers Decorated with Graphene Quantum Dots for Simultaneous Targeting and Imaging of Tumor Cells**

**Amphiphilic triphenylphosphonium (TPP)-poly( $\epsilon$ -caprolactone)(PCL)-TPP (TPP-PCL-TPP, TPCL) polymers** are synthesized and self-assembled to form nanovesicles and nanofibers. TPCL nanoparticles (NPs) themselves exhibit cancer-killing properties and can carry either hydrophilic drugs or hydrophobic drugs. Anticancer drug-loaded TPCL NPs show synergistically strong anticancer effects via a combination of the loaded chemical drugs and TPCL-based NPs.

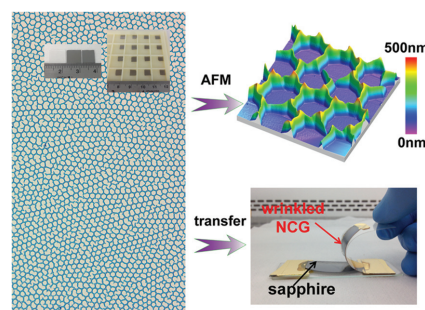


## Nanomedicine

D. Y. Cho, H. Cho, K. Kwon, M. Yu, E. Lee, K. M. Huh, D. H. Lee, H. C. Kang\* .....5479–5491

**Triphenylphosphonium-Conjugated Poly( $\epsilon$ -caprolactone)-Based Self-Assembled Nanostructures as Nanosized Drugs and Drug Delivery Carriers for Mitochondria-Targeting Synergistic Anticancer Drug Delivery**

**A unique hexagonal wrinkled pattern is generated on nanocrystalline graphitic films** grown on polished *c*-plane sapphire substrate via a simple delamination–buckling process. The dimensional features of the wrinkles can be manipulated by adjusting the thickness of deposited films. In addition, a high-fidelity dry transfer method for transfer of wrinkled films onto arbitrary substrates is developed for further applications.

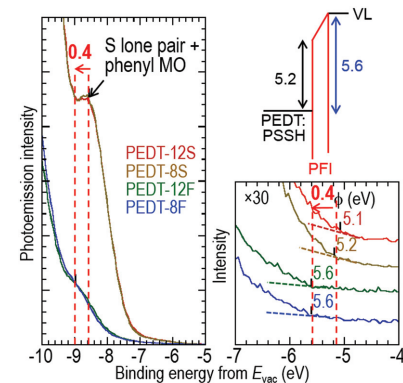


## Surface Wrinkles

Y. Liu, Kenry, Y. Guo, S. Sonam, S. K. Hong, M. H. Nai, C. T. Nai, L. Gao, J. Chen, B. J. Cho, C. T. Lim, W. Guo,\* K. P. Loh\* .....5492–5503

**Large-Area, Periodic, Hexagonal Wrinkles on Nanocrystalline Graphitic Film**

**The ultrahigh workfunctions of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonic acid) (PEDT:PSSH):perfluorinated ionomer (PFI) blends** result from a dipolar surface layer associated with surface segregation of PFI chains. Despite their higher workfunctions, their hole contacts to organic semiconductors with ionization potentials deeper than 5.2 eV remain nonohmic. Work function matching through the use of an insulating dipolar layer is not efficient for providing ohmic contacts.



## Organic Semiconductors

D. Belaineh, J.-K. Tan, R.-Q. Png,\* P.-F. Dee, Y.-M. Lee, B.-N. N. Thi, N.-S. Ridzuan, P. K. H. Ho.....5504–5511

**Perfluorinated Ionomer-Modified Hole-Injection Layers: Ultrahigh-Workfunction but Nonohmic Contacts**

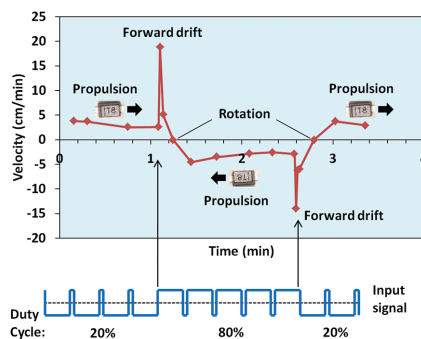


# FULL PAPERS

## Self-Propelling Microcircuits

R. Sharma, O. D. Velev\* ..... 5512–5519

### Remote Steering of Self-Propelling Microcircuits by Modulated Electric Field

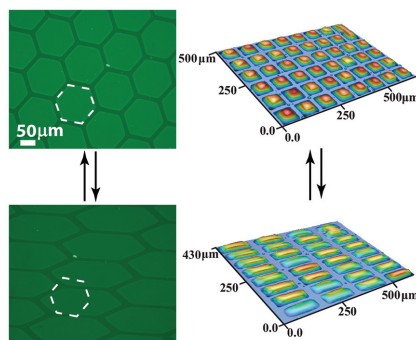


The direction of self-propulsion of diodes floating on a water surface, powered by external alternating (AC) electric fields, can be controlled on-demand by electronically modifying the symmetry of the applied AC waveform. Changes in the duty cycle of the signal inducing short-lasting DC fields can be used to remotely reverse its direction of motion or make it move sideways.

## Droplet Arrays

J. J. Bowen, J. M. Taylor, C. P. Jurich, S. A. Morin\* ..... 5520–5528

### Stretchable Chemical Patterns for the Assembly and Manipulation of Arrays of Microdroplets with Lensing and Micromixing Functionality

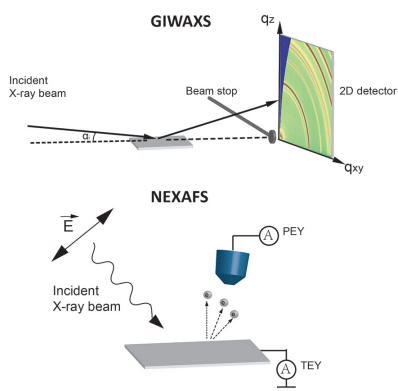


Chemically patterned silicone films drive the assembly of microdroplets into ordered arrays. These microdroplets can be “stretched”, a capability that was used to drive mixing in the droplets and to change the lensing characteristics of the array. Synthesizing “stretchable” chemical patterns of different geometries and functional groups provides a method to explore soft, mechanoresponsive surfaces with rapidly reversible properties unavailable in hard systems.

## Perovskite Films

W. Huang, F. Huang, E. Gann, Y.-B. Cheng,\* C. R. McNeill\*... 5529–5536

### Probing Molecular and Crystalline Orientation in Solution-Processed Perovskite Solar Cells

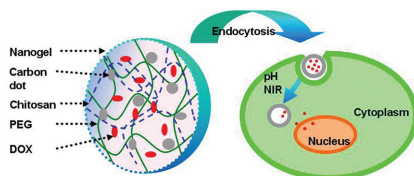


Using a combination of grazing-incidence wide-angle X-ray scattering (GIWAXS) and near-edge X-ray absorption fine structure (NEXAFS) spectroscopy, the orientational alignment of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  crystallites and  $\text{CH}_3\text{NH}_3^+$  cations are separately probed. The orientation of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  crystallites is sensitive to film thickness, solvent evaporation rate, and the underlying  $\text{TiO}_2$  morphology. However,  $\text{CH}_3\text{NH}_3^+$  cations exhibit a random molecular orientation that is independent of the  $\text{TiO}_2$  architecture and the perovskite film thickness.

## Hybrid Nanogels

H. Wang, J. Di, Y. Sun, J. Fu, Z. Wei, H. Matsui, A. del C. Alonso, S. Zhou\* ..... 5537–5547

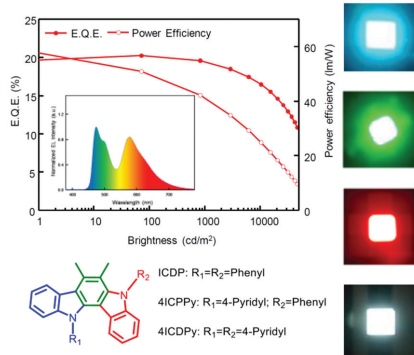
### Biocompatible PEG-Chitosan@Carbon Dots Hybrid Nanogels for Two-Photon Fluorescence Imaging, Near-Infrared Light/pH Dual-Responsive Drug Carrier, and Synergistic Therapy



One-pot, surfactant-free synthesis of bio-compatible and multifunctional hybrid nanogels composed of poly(ethylene glycol), chitosan, and carbon dots is reported. Such hybrid nanogels demonstrate great promise for in vivo medical diagnosis and synergistic therapy because they can realize simultaneous pH and near-infrared light dual-responsive drug release and two-photon fluorescence cellular imaging.

## FULL PAPERS

**Three new indolocarbazole-based host materials**, ICDP, 4ICPPy, and 4ICDPy, are synthesized and used as hosts for various color phosphorescent organic light-emitting diodes. The blue, green, and red devices using 4ICDPy as the host all exhibit very high device efficiencies. In addition, a two-color white device shows an external quantum efficiency of 20.3% and power efficiency of  $50.9 \text{ lm W}^{-1}$  with excellent color stability.

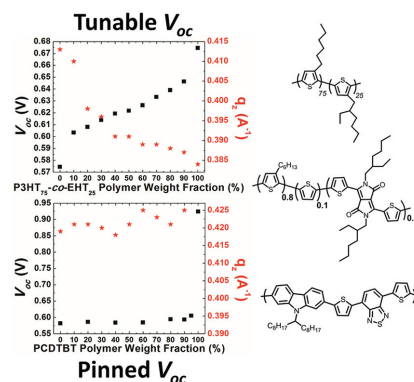


## Indolocarbazoles

C.-C. Lai, M.-J. Huang, H.-H. Chou,  
 C.-Y. Liao, P. Rajamalli, C.-H.  
 Cheng\* .....5548–5556

*m*-Indolocarbazole Derivative as a  
 Universal Host Material for RGB and  
 White Phosphorescent OLEDs

**The first direct structural study of ternary-blend bulk heterojunction active layers** in solar cells that demonstrate tunable open-circuit voltage ( $V_{oc}$ ) is performed. Physical mixing of the polymer donors, quantified by the square of the square-root difference in surface energies, leads to ensemble-average electronic character that results in composition-dependent  $V_{oc}$  when these blends are incorporated in solar cells.



## Organic Solar Cells

P. P. Khlyabich, A. E. Rudenko,  
 B. C. Thompson, Y.-L. Loo\* .....5557–5563

Structural Origins for Tunable Open-  
 Circuit Voltage in Ternary-Blend  
 Organic Solar Cells

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