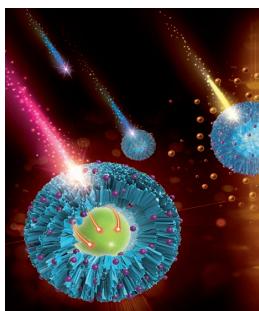


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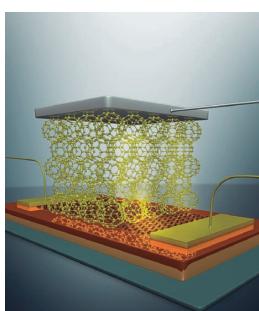
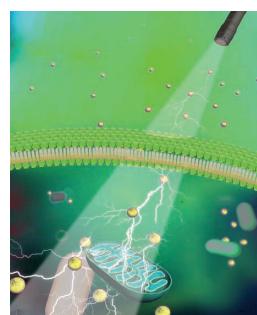


## Photocatalysis

The front cover image demonstrates plasmonic Au nanoparticle decorated, upconverting-core@porous-TiO<sub>2</sub>-shell microspheres that can absorb photons over a wide wavelength range from ultraviolet to near infrared. As reported by D. Ma and team on page 2950, this hybrid structure, rationally designed and realized to greatly benefit from plasmons and upconversion, shows significantly enhanced broadband, photocatalytic activity.

## Cancer Treatment

In situ mitochondria photodynamic therapy is demonstrated to be a simple but effective strategy to inhibit tumor growth. On page 2961, X.-Z. Zhang and co-workers construct a mitochondria-targeted self-delivery system for tumor therapy and imaging. In addition, a dual-stage light irradiation strategy is developed to optimize the antitumor effect both in vitro and in vivo.



## Energy Barriers

Graphene is an ideal candidate for the source electrode in a vertical organic field effect transistor as it has low density of states near the Dirac point and easy gate tunability of the Fermi-level. On page 2972, S. Parui, L. Hueso, and team modulate the energy barrier at a graphene/molecular semiconductor (fullerene) junction by varying the gate electric field, thus opening a promising route toward molecular-semiconductor based devices.



## Smart Hydrogels

Poly(*N*-isopropylacrylamide)-clay nanocomposite hydrogels with a responsive bending property as temperature-controlled manipulators are reported by Z. Liu, L.-Y. Chu, and co-workers on page 2980. The hydrogel manipulators are demonstrated for various applications including encapsulation, capture, and transportation of targeted objects. Such hydrogels provide new materials for designing “smart” soft robots in myriad fields such as manipulators, grippers, and cantilever sensors.

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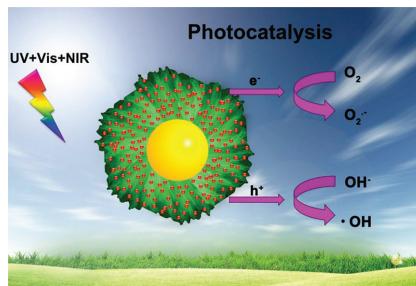
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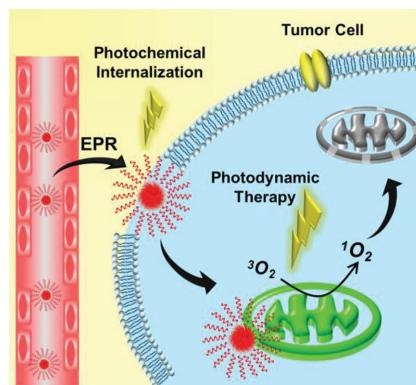
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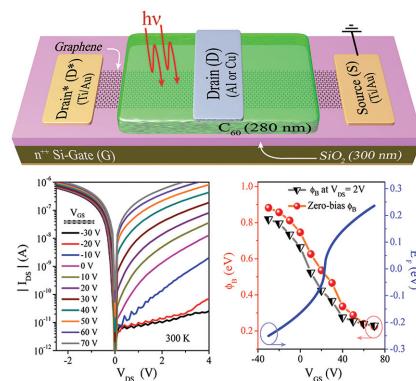
**Au nanoparticle decorated  $\text{NaYF}_4\text{:Yb}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Tm}^{3+}\text{@TiO}_2$  core@porous-shell microspheres** are successfully prepared and exhibit excellent, stable, and broadband photocatalytic activity from UV up to near-infrared by rationally applying plasmon and upconversion concepts into photocatalysis.



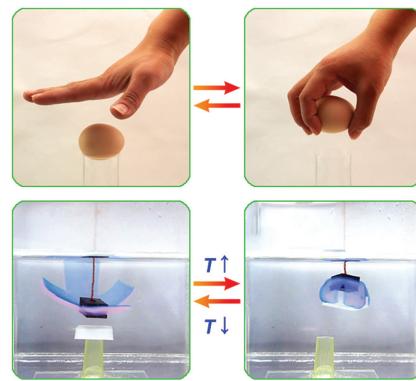
**A mitochondria-targeted self-delivery system** is developed for optical-imaging-guided photodynamic tumor therapy. A dual-stage light irradiation strategy is used to optimize the synergistic effect between photosensitizer and  $(\text{KLAKLAK})_2$ , and significant efficacious tumor inhibition is observed both in vitro and in vivo.



**Graphene is an ideal candidate for the source electrode** in a vertical organic field effect transistor as it has low density of states near the Dirac point and easy gate tunability of the Fermi-level. By varying the gate electric field, the energy-barrier is modulated at a graphene/molecular-semiconductor (fullerene) junction, thus opening a promising route toward molecular-semiconductor based devices.



**Poly(*N*-isopropylacrylamide)-clay nanocomposite hydrogels with responsive bending property** are successfully developed as temperature-controlled manipulators by designing an asymmetrical distribution of nanoclays across the hydrogel thickness. The hydrogels show rapid, reversible, and repeatable thermoresponsive bending characteristics, and are demonstrated as temperature-controlled soft manipulators for applications of encapsulating, grasping, and transporting target objects in aqueous environments.



## Photocatalysis

Z. Xu, M. Quintanilla, F. Vetrone, A. O. Govorov, M. Chaker, D. Ma\* ..... 2950–2960

**Harvesting Lost Photons: Plasmon and Upconversion Enhanced Broadband Photocatalytic Activity in Core@Shell Microspheres Based on Lanthanide-Doped  $\text{NaYF}_4$ ,  $\text{TiO}_2$ , and Au**

## Photodynamic Therapy

K. Han, Q. Lei, S.-B. Wang, J.-J. Hu, W.-X. Qiu, J.-Y. Zhu, W.-N. Yin, X. Luo, X.-Z. Zhang\* ..... 2961–2971

**Dual-Stage-Light-Guided Tumor Inhibition by Mitochondria-Targeted Photodynamic Therapy**

## Energy Barriers

S. Parui,\* L. Pietrobon, D. Ciudad, S. Vélez, X. Sun, F. Casanova, P. Stolar, L. E. Hueso\* ..... 2972–2979

**Gate-Controlled Energy Barrier at a Graphene/Molecular Semiconductor Junction**

## Smart Hydrogels

C. Yao, Z. Liu,\* C. Yang, W. Wang, X.-J. Ju, R. Xie, L.-Y. Chu\* ..... 2980–2991

**Poly(*N*-isopropylacrylamide)-Clay Nanocomposite Hydrogels with Responsive Bending Property as Temperature-Controlled Manipulators**

## FULL PAPERS

## Cationic Nanogels

D. Li, N. Kordalivand, M. F. Fransen, F. Ossendorp, K. Raemdonck, T. Vermonden, W. E. Hennink, C. F. van Nostrum\* ..... 2993–3003

**The model antigen ovalbumin** is chemically conjugated to the cationic nanogels via disulfide bonds. The protein is thereby covalently immobilized in the nanogels in the extracellular environment. Rapid release of conjugated protein occurs once the nanogels are internalized into cells, due to cleavage of the disulfide bonds in the presence of relatively high intracellular levels of glutathione ( $2.5\text{--}10 \times 10^{-3}$  M).

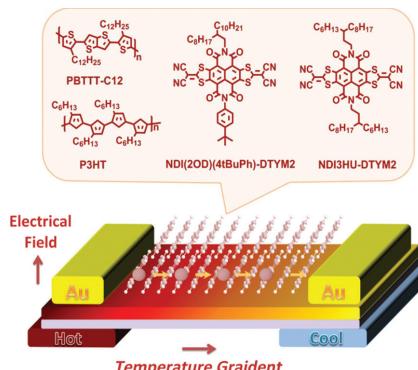


## Reduction-Sensitive Dextran Nanogels Aimed for Intracellular Delivery of Antigens

## Organic Semiconductors

F. Zhang, Y. Zang, D. Huang, C.-a. Di,\* X. K. Gao, H. Sirringhaus, D. B. Zhu\* ..... 3004–3012

## Modulated Thermoelectric Properties of Organic Semiconductors Using Field-Effect Transistors

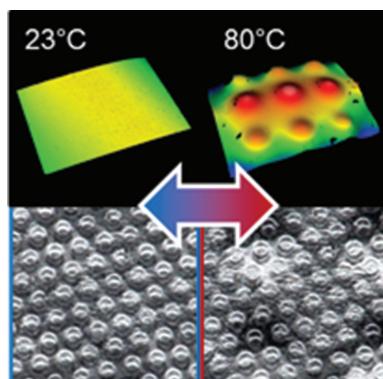


**Investigation of thermoelectric properties of organic semiconductors** is a fundamental issue toward effective application of high-performance organic thermoelectric materials. It is reported on a systematic study of the thermoelectric properties for organic semiconductors via field-effect transistors, which indicates that organic transistors should provide an effective platform to accelerate the screening of promising organic thermoelectric materials.

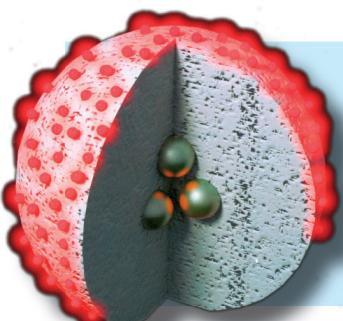
## Shape-Memory

M. Frensemeier, J. S. Kaiser, C. P. Frick, A. S. Schneider, E. Arzt, R. S. Fertig III, E. Kroner\* ..... 3013–3021

## Temperature-Induced Switchable Adhesion using Nickel–Titanium–Polydimethylsiloxane Hybrid Surfaces



**Switchable adhesion** is achieved using a nickel–titanium two-way shape-memory alloy combined with a bioinspired micropatterned dry adhesive silicone rubber. The temperature-induced topography change of the shape memory alloy causes a well-controlled reversible adhesive performance. The switching efficiency is tunable and reaches up to 100%, meanwhile maintaining high reversibility.



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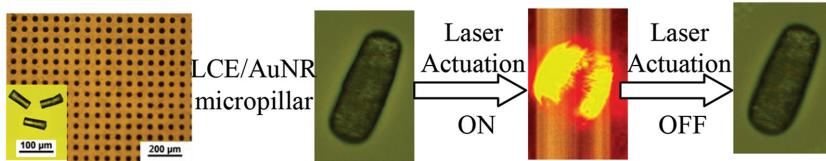
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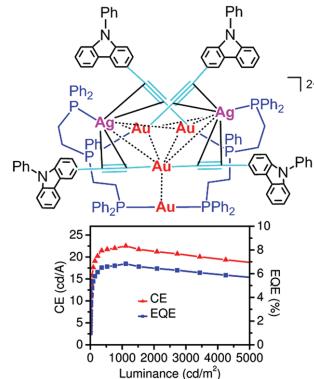
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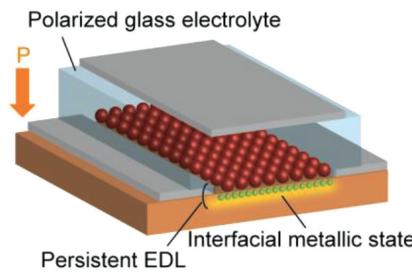
By incorporating of gold nanoparticles (AuNPs) into polyacrylate-based liquid crystal elastomer (LCE), LCE/AuNP micropillars capable of both thermal and photothermal actuation are fabricated and characterized. Upon red laser (635 nm) irradiation, the LCE/AuNR micropillar shows rapid (a few seconds), large amplitude ( $\approx 30\%$  strain), and reversible actuation without perturbing the surrounding environment.



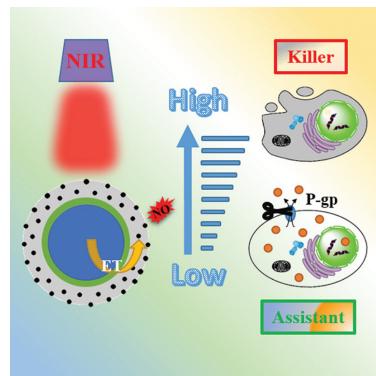
**Cationic  $\text{Au}_4\text{Ag}_2$  aromatic alkynyl cluster complexes** supported by bis(2-diphenylphosphinoethyl)phenylphosphine (dpep) are highly phosphorescent in solid states and films. Solution-processed OLEDs based on  $\text{Au}_4\text{Ag}_2$  complexes exhibit highly efficient electroluminescence.



A **programmable interfacial persistent metallic state** is realized up to near room temperature at the interface between the polarized solid glass–electrolyte and  $\text{SrTiO}_3$ . The electric double layer (EDL) is formed from cations and electrons in the all-solid heterointerface devices. By freezing motion of cations within the EDL, the persistent metallic state is stabilized even after removing the external electric field up to near room temperature.



A new **near-infrared triggered on-demand nitric oxide (NO) delivery nanoplatform** is constructed by incorporating upconversion nanoparticles with light-sensitive NO donors, Roussin's black salt (RBS). By regulating the output power of the laser, the on-demand release of NO is realized and results in multi-functionality of NO for tumor therapy.



## Micropillars

X. Liu, R. Wei, P. T. Hoang, X. Wang,\*  
T. Liu,\* P. Keller\* ..... 3022–3032

## Reversible and Rapid Laser Actuation of Liquid Crystalline Elastomer Micropillars with Inclusion of Gold Nanoparticles

## Electroluminescence

L.-J. Xu, J.-Y. Wang, X.-F. Zhu,  
X.-C. Zeng, Z.-N. Chen\* ..... 3033–3042

Phosphorescent Cationic  $\text{Au}_4\text{Ag}_2$  Alkynyl Cluster Complexes for Efficient Solution-Processed Organic Light-Emitting Diodes

## Charge Carriers

K. Taniguchi,\* T. Fukamichi, K. Itaka,  
H. Takagi ..... 3043–3048

## Programmable Persistent Interfacial Metallic State Induced by Frozen Ions in Inorganic–Glass Solid Electrolyte

## Cancer Treatment

X. Zhang, G. Tian,\* W. Yin, L. Wang,  
X. Zheng, L. Yan, J. Li, H. Su, C. Chen,  
Z. Gu,\* Y. Zhao\* ..... 3049–3056

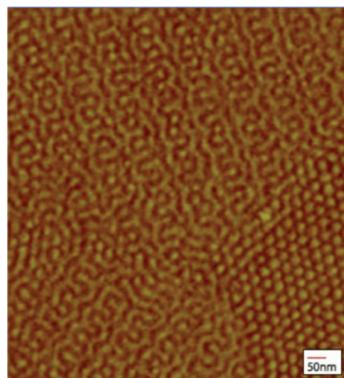
## Controllable Generation of Nitric Oxide by Near-Infrared-Sensitized Upconversion Nanoparticles for Tumor Therapy

## FULL PAPERS

## Block Copolymers

M. A. Chavis, D.-M. Smilgies,  
U. B. Wiesner, C. K. Ober\* ... 3057–3065

**Widely Tunable Morphologies in Block Copolymer Thin Films Through Solvent Vapor Annealing Using Mixtures of Selective Solvents**

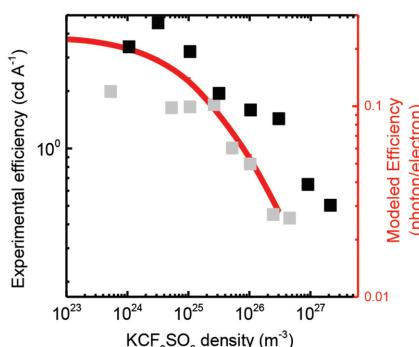


An approach to achieve selective, controlled, morphological ordering from a single, fixed volume fraction, poly(2-hydroxyethyl methacrylate)-block-poly(methyl methacrylate) copolymer is presented. By taking advantage of preferential solvent annealing, it is possible to dial in to a specific block copolymer microstructure that can then be kinetically trapped in the dry film.

## Electrochemical Cells

S. van Reenen, R. A. J. Janssen,  
M. Kemerink\* ..... 3066–3073

**Fundamental Tradeoff between Emission Intensity and Efficiency in Light-Emitting Electrochemical Cells**

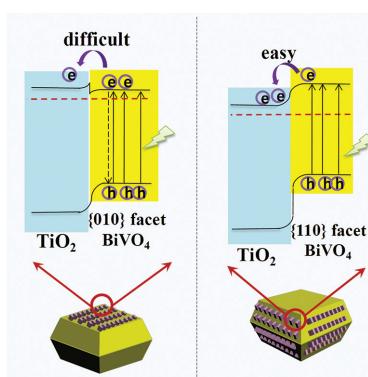


A fundamental tradeoff between emission intensity and efficiency in light-emitting electrochemical cells is reported. The admixed ions in LECs on the one hand improve charge transport by electrochemical doping, but on the other hand reduce the luminescent efficiency by quenching of excitons for  $\text{KCF}_3\text{SO}_3$  densities of  $>10^{25} \text{ m}^{-3}$ .

## Photocatalysis

H. Li, H. Yu, X. Quan,\* S. Chen,  
H. Zhao ..... 3074–3080

**Improved Photocatalytic Performance of Heterojunction by Controlling the Contact Facet: High Electron Transfer Capacity between  $\text{TiO}_2$  and the  $\{110\}$  Facet of  $\text{BiVO}_4$  Caused by Suitable Energy Band Alignment**

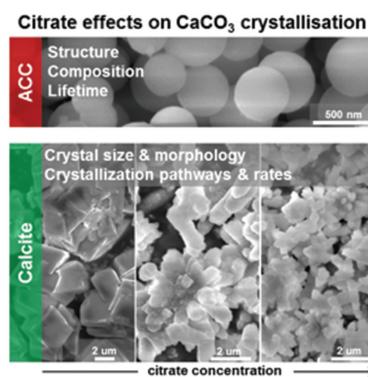


Two kinds of  $\text{BiVO}_4\text{-TiO}_2$  heterojunctions with  $\text{TiO}_2$  grown on the  $\{110\}$  facet and  $\{010\}$  facet of  $\text{BiVO}_4$ , respectively, are prepared. Heterojunction with  $\text{TiO}_2$  grown on the  $\{110\}$  facet of  $\text{BiVO}_4$  possesses higher charge carriers separation efficiency and better photocatalytic performance owing to the lower interfacial energy barrier between conduction band of  $\{110\}$  facet with that of  $\text{TiO}_2$ .

## Calcium Carbonate

D. J. Tobler,\* J. D. Rodriguez-Blanco,  
K. Dideriksen, N. Bovet, K. K. Sand,  
S. L. S. Stipp ..... 3081–3090

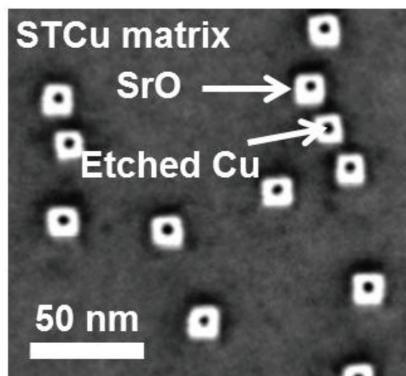
**Citrate Effects on Amorphous Calcium Carbonate (ACC) Structure, Stability, and Crystallization**



The addition of citrate to solutions supersaturated with respect to amorphous calcium carbonate (ACC) leads to more stable ACC with modified structure and composition and direct crystallization to calcite via a spherulitic growth mechanism. At citrate/Ca ratio  $\geq 0.75$ , ACC formation is inhibited and calcite spherulites form directly from solution.

## FULL PAPER

nc-STCu films are grown in vacuum on Nb-doped (001) STO substrate after etching in ammonium hydroxide for 3 h. Metallic copper rods with  $\approx 3$  nm diameter grow in the center of the SrO nanorods in a perovskite matrix, and etching of the Cu by ammonium hydroxide creates uniformly sized nanopores.



## Nanopores

D. H. Kim, X. Y. Sun, N. M. Aimon, J. J. Kim, M. J. Campion, H. L. Tuller, L. Kornblum, F. J. Walker, C. H. Ahn, C. A. Ross\* ..... 3091–3100

A Three Component Self-Assembled Epitaxial Nanocomposite Thin Film

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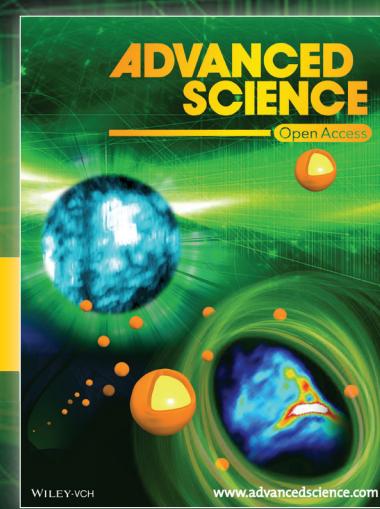
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