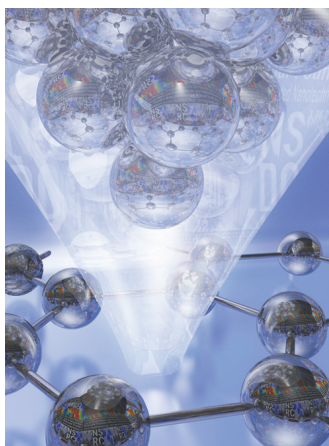


# ADVANCED FUNCTIONAL MATERIALS

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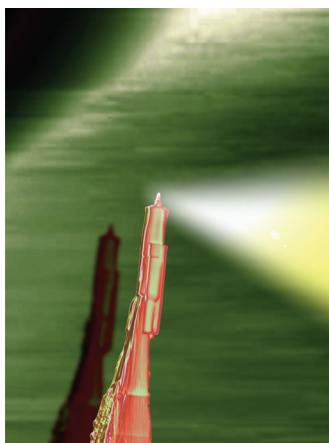
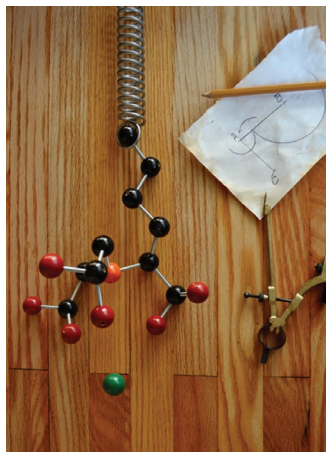


## Scanning Probe Microscopy

An artistic vision of the interaction between the Nanoscale Science Research Centers (NSRCs) and the user community is rendered as a scanning tunneling microscope tip above a surface. NSRCs combine two elements, namely the support of an external user program and an in-house research program, including development of new, commercially unavailable instrumental platforms, operation of unique commercial equipment, and dissemination of this expertise and capabilities to a broad scientific community. This Special Issue, guest-edited by Sergei Kalinin, showcases recent progress in scanning probe microscopy across the NSRCs. Figure courtesy of Stephen Jesse, ORNL.

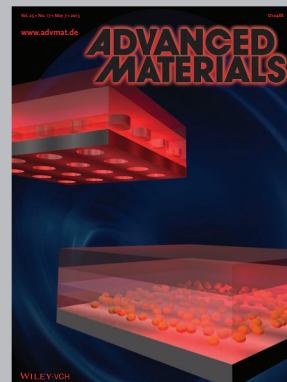
## Dynamic Force Spectroscopy

The photograph on the inside cover illustrates the mechanical measurement of bond energies. This is the essence of dynamic force spectroscopy, which uses the bending of an atomic force microscope cantilever to measure the work required to break a bond. On page 2525, James J. De Yoreo and co-workers show how this technique can be used to probe the binding free energy between biominerals and the proteins that control their growth.



## Synchrotron X-Ray Scanning Tunneling Microscopy

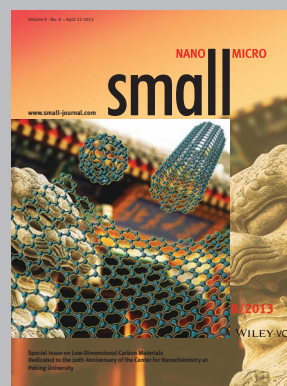
On page 2646 Volker Rose and co-workers present a smart tip for synchrotron X-ray scanning tunneling microscopy that is entirely coated by an insulating oxide, except at the tip apex. The tip is only sensitive to X-ray-excited tunnel currents and not to photoejected electrons caused by classical photoemission, which would degrade the spatial resolution. The bright light in the image, coming in from the right, that hits the tip apex schematically represents the synchrotron beam during an experiment.



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## ESSAY

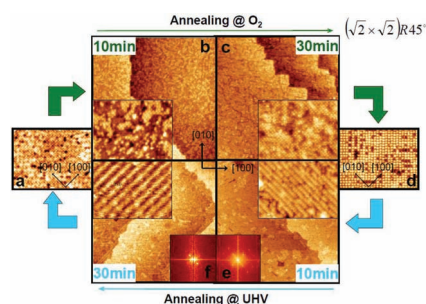
## Scanning Probe Microscopy

S. V. Kalinin\* .....2468–2476

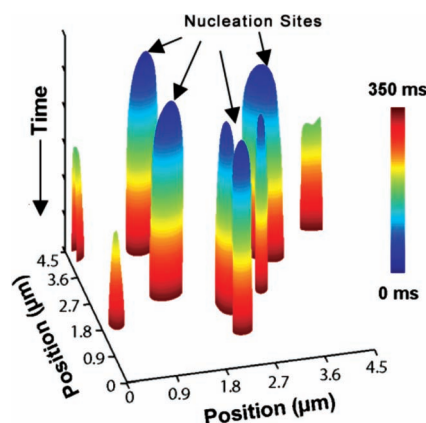
Scanning Probe Microscopy in US Department of Energy Nanoscale Science Research Centers: Status, Perspectives, and Opportunities

## FEATURE ARTICLES

## Strongly Correlated Materials

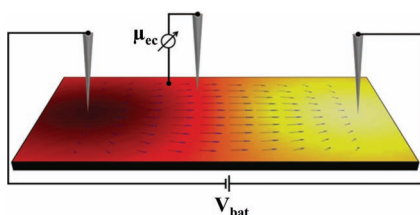
Z. Gai,\* S. V. Kalinin, A.-P. Li, J. Shen,  
A. P. Baddorf .....2477–2489**In Situ Observations and Tuning of  
Physical and Chemical Phenomena  
on the Surfaces of Strongly Correlated  
Oxides****Controllable surface tuning provides the opportunity to study** how structural, electronic, and magnetic properties respond to the broken symmetry and opens avenues for exploration of completely new physical properties. Recent progress in in situ observations and tuning of physical and chemical phenomena on the surfaces of strongly correlated oxide thin films and crystals, including atomic-level structural studies, control, and tuning of the physical properties is reviewed.

## Ferroelectric Materials

R. K. Vasudevan, D. Marincel, S. Jesse,  
Y. Kim, A. Kumar, S. V. Kalinin,  
S. Trolor-McKinstry\* .....2490–2508**Polarization Dynamics in Ferroelectric  
Capacitors: Local Perspective on  
Emergent Collective Behavior and  
Memory Effects****Functional properties of ferroelectric materials** depend both on the residual domain states and on the mobility of domain walls in response to the applied electric and stress fields. Through advanced spectroscopic techniques on model ferroelectric capacitors, it is possible to determine the domain nucleation sites and domain wall velocity as a function of pulse amplitude and widths, in a single experiment.

## FEATURE ARTICLES

**Electrical transport measurement methods with a four-probe scanning tunneling microscope** and recent progress on its applications in nanomaterials with a focus on probing structure-transport relationships at the nanometer-scale are described. A broad range of nanomaterials are covered, including surface supported quasi-1D and 2D electronic systems, semiconducting and metallic nanowires, carbon nanotubes, and graphene. The effects of atomic defects, grain boundaries, interfaces, and electronic interactions are discussed.

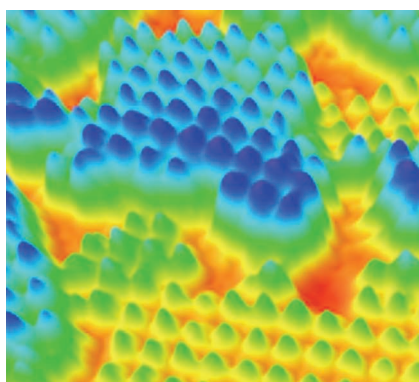


## Scanning Tunneling Microscopy

A.-P. Li,\* K. W. Clark, X.-G. Zhang,  
A. P. Baddorf.....2509–2524

**Electron Transport at the Nanometer-Scale Spatially Revealed by Four-Probe Scanning Tunneling Microscopy**

**Atomic force imaging and spectroscopy provide unique tools for investigating molecular interactions** and dynamics in biomolecular and biomineral systems in situ. Three recent examples are presented that illustrate the use of these methods to gain mechanistic insights into biomolecular controls over mineral formation and the self-assembly of protein matrices, such as the S-layer protein membrane.

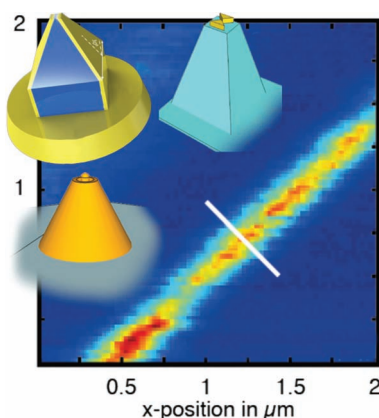


## Biomolecular Assembly

J. J. De Yoreo,\* S. Chung,  
R. W. Friddle.....2525–2538

**In Situ Atomic Force Microscopy as a Tool for Investigating Interactions and Assembly Dynamics in Biomolecular and Biomineral Systems**

**Near-field optical microscopies and spectroscopies** provide potential access to chemical, morphological, physical, and dynamical information at nanometer length scales—information that is difficult to probe by other means. Recent innovations by the nano-optics community and at the Molecular Foundry are reported, which address many of the longstanding “nanospectroscopic imaging” challenges and lay the groundwork for unprecedented nano-optical studies of material properties.

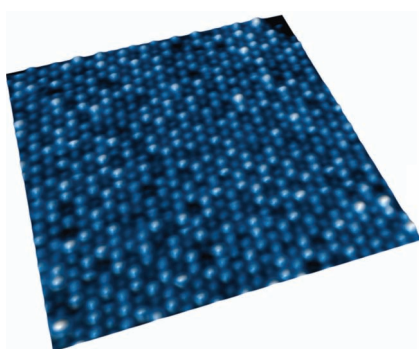


## Nano-Optics

P. J. Schuck,\* A. Weber-Bargioni,\*  
P. D. Ashby, D. F. Ogletree,  
A. Schwartzberg, S. Cabrini\*.....2539–2553

**Life Beyond Diffraction: Opening New Routes to Materials Characterization with Next-Generation Optical Near-Field Approaches**

**Atomic-scale characterization of graphene synthesis on various materials** (SiC, Cu(111), Cu foil, etc.) via scanning tunneling microscopy provides fundamental exploration of growth dynamics, film quality, and the role of defects. The chemical modification of graphene following exposure to atomic hydrogen and molecular assembly are also explored.



## Graphene

E. V. Iski, E. N. Yitamben, L. Gao,  
N. P. Guisinger\*.....2554–2564

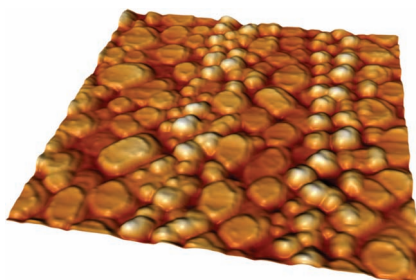
**Graphene at the Atomic-Scale: Synthesis, Characterization, and Modification**

## FEATURE ARTICLES

## Scanning Tunneling Microscopy

T. Y. Chien, J. Chakhalian, J. W. Freeland,  
N. P. Guisinger\* .....2565–2575

### Cross-Sectional Scanning Tunneling Microscopy Applied to Complex Oxide Interfaces



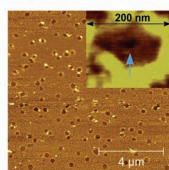
Cross-sectional scanning tunneling microscopy and spectroscopy (XSTM/S) for complex oxides has recently been developed. XSTM/S is an ideal tool to directly probe the electronic properties at interfaces of dissimilar complex oxides. The understanding of the emerging phenomena at complex oxide interfaces could be pushed further with the nanometer-scale electronic information obtained by XSTM/S.

## Nanocomposites

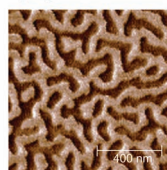
G. A. Montañño,\* P. G. Adams,  
X. Xiao, P. M. Goodwin .....2576–2591

### Scanning Probe Microscopy of Nanocomposite Membranes and Dynamic Organization

AFM of lipid bilayer-templated Au-nanorings



AFM of grafted mixed polymer brushes

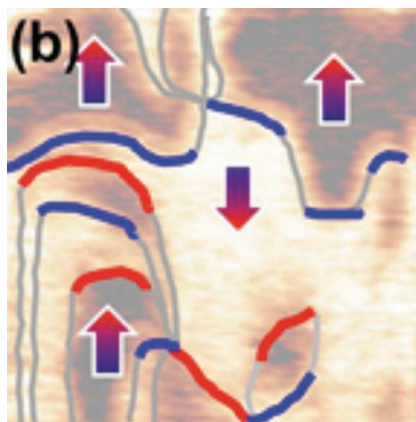


Scanning probe microscopy allows for high-resolution characterization and manipulation of nanocomposite membranes. Several examples are shown and their characterization/manipulation are discussed including lipid/nanoparticle composites and grafted mixed polymer systems.

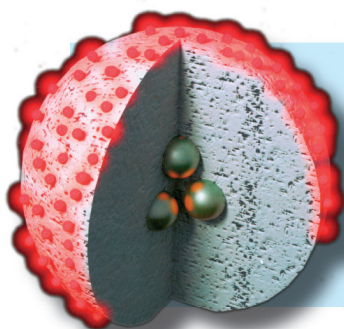
## Ferroelectrics

R. K. Vasudevan, W. Wu,\* J. R. Guest,  
A. P. Baddorf, A. N. Morozovska,  
E. A. Eliseev, N. Balke, V. Nagarajan,\*  
P. Maksymovych,\*  
S. V. Kalinin .....2592–2616

### Domain Wall Conduction and Polarization-Mediated Transport in Ferroelectrics



Nanometer-scale electronic transport in engineered interfaces in ferroelectrics, such as domains and topological defects, has emerged as a topic of broad interest. The use of scanning probe microscopies to access topological defects and directly measure their unique properties is reviewed. It is found that observation of enhanced conduction at domain walls can be attributed to segregation of carriers at charged walls. Furthermore, the potential distribution around a curved or tilted wall can be highly asymmetric, even for nominally uncharged walls due to strain and flexo-electric couplings.



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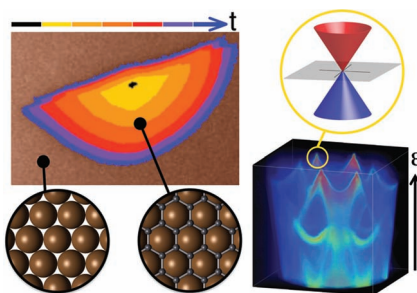
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## FEATURE ARTICLES

**Real-time surface microscopy and in situ spectroscopy** can provide unique insight into graphene and other 2D materials on metal substrates. The power of in situ microscopy in realizing and probing important functionalities in 2D materials is illustrated by reviewing recent progress in understanding scalable graphene growth on metals, processing by selective chemistry at the graphene/metal interface, and important properties such as band structure, work function, etc.

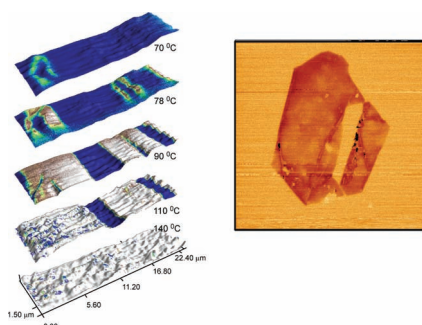


## Graphene

P. Sutter,\* E. Sutter.....2617–2634

## Microscopy of Graphene Growth, Processing, and Properties

**Near-field scanning microwave microscopy** is currently capable of routine imaging of the dielectric constant and conductivity with a spatial resolution below 100 nm. The technique is illustrated with two examples: studies of metal-insulator phase transitions in single-crystalline nanoplatelets of vanadium dioxide and imaging of conductivity inhomogeneities in single- and few-layer graphene grown by chemical vapor deposition.



## Graphene

A. Tselev,\* N. V. Lavrik, A. Kolmakov, S. V. Kalinin .....2635–2645

Scanning Near-Field Microwave Microscopy of VO<sub>2</sub> and Chemical Vapor Deposition Graphene

## FULL PAPERS

**Nanofabricated insulator-coated smart tips** are indispensable for stable tunneling conditions in synchrotron X-ray enhanced scanning tunneling microscopy. An unambiguous and direct way of fingerprinting tunneling to far field transitions of the tip that relies on the simultaneous analysis of the X-ray-induced tip and sample current is presented.

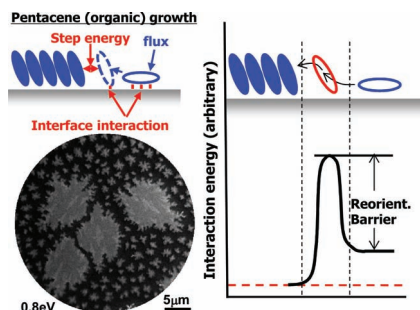


## Characterization Tools

V. Rose,\* K. K. Wang, T. Chien, J. Hiller, D. Rosenmann, J. W. Freeland, C. Preissner, S.-W. Hla .....2646–2652

## Synchrotron X-Ray Scanning Tunneling Microscopy: Fingerprinting Near to Far Field Transitions on Cu(111) Induced by Synchrotron Radiation

**Nucleation and growth processes in organic systems** such as a pentacene film on silicon dioxide are often complicated due to anisotropy in both the molecular shape and the crystal structure. As in the diffusive state the pentacene molecule is in the lying-down configuration, interfacial interaction is different from that at the island's step edge. The strength of interfacial interaction in the diffusion state determines the energy barrier for molecule reorientation, such that the stronger interaction increases the relative stability of diffusing molecules.



## Thin Films

A. Al-Mahboob, Y. Fujikawa, T. Sakurai, J. T. Sadowski\* .....2653–2660

## Real-Time Microscopy of Reorientation Driven Nucleation and Growth in Pentacene Thin Films on Silicon Dioxide