Summmary of SBU "physical science" faculty currently active at NSLS and/or NSLS-II

Chemistry	Geosciences	Materials Science and Engineering	Physics and Astronomy
1. Soft Matter / Nano-materials Benjamin Chu Robert B. Grubbs Benjamin Hsiao Jonathan G. Rudick 2. Catalysis Chemistry Peter Khalifah Michael G. White 3. Materials Chemistry Nancy Goroff Clare P. Grey Stanislaus Wong 4. Structural Biology Daniel P. Raleigh Orlando D. Schärer 5. Biological Imaging Iwao Ojima Isaac Carrico Liz Boon Nicole S. Sampson Peter J. Tonge	1. Nano-materials/ Environmental John Parise Rich Reeder Martin Schoonen 2. Mineral Physics/Extr Conditions Donald Weidner Robert Liebermann Lars Ehm Baosheng Li Michael Vaughan 3. Planetary Petrology Hanna Nekvasil 4. Imaging/chemical mapping Troy Rasbury	1. Nano-materials/ Environmental/Energy Apps Tadanori Koga Devinder Mahajan Miriam Rafalovich Jonothan Sokolov 2. Thermal spray coatings Sanjay Sampath Herb Hermann Chris Weyant 3. Imaging Michael Dudley Gary Halada	1. Soft Matter / Nano-materials Xu Du 2. Condensed Matter Physics and Materials Science—Correlated Systems Peter Stephens 3. Condensed Matter Physics and Materials Science--Energy Storage Phil Allen, Matt Dawber, Maria Victoria (Marivi) Fernandez-Serra

other statistics

Currently active SBU PS investigators (e.g. No Kirz, Jacobsen etc)

1203 publications in “synchrotron” research

cited ~28,300 times (~ 24/article)

h-index = 75

STEPHENS PW, MIHALY L et al. Supercond K_3C_{60} cit: 616

Zong XH, Kim K, et al. nanofiber membranes cited: 472

STEPHENS et al. CHAINS IN RBC6 cited 421

Pagola S, Stephens PW et al. malaria. Cited: 375

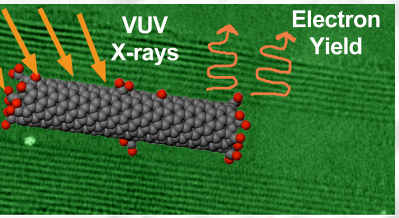
CHAUVET O, et al. ORTHORHOMBIC RBC60, cited: 314

Cleveland CL et al. gold nanocrystals. cited: 283

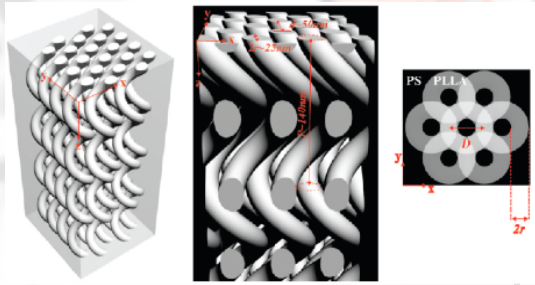
Novel nano/soft functional materials

> Transformational functional materials required for energy/ environmental applications

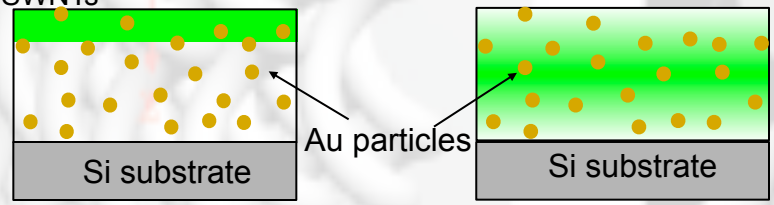
> Materials by design



Unique nano-materials (Wong group) S. Banerjee et al., Chem. Comm., (7), 772-773 (2004) - SWNTs

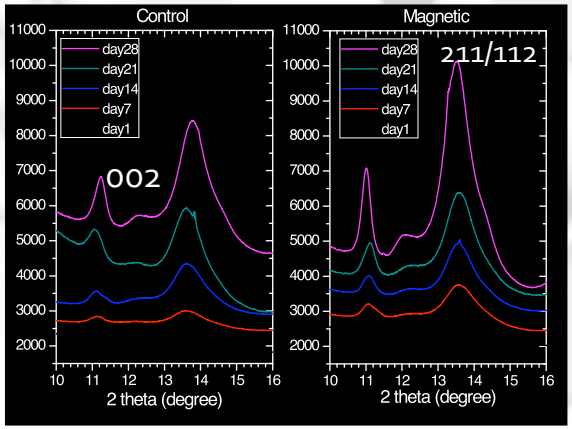


Novel block-copolymers (Hsiao group JACS., 131, 18533 (2009).



"Surface" **"Center"**

Measure "diffusive" motions of the markers which track the local viscosity of the matrix



GiXD spectra of Biominerals; magnetic fields (EMF)enhance biomineralization. DiMasi (BNL) Rafailovich (SBU), Simon (SBU, Dental School)

> Strong track record at NSLS

> Considerable scattering expertise at BNL/SBU alliances

> Already strong SBU--BNL relationships strengthened by soft matter thrust in proposed Center for Advanced Photon Techniques (CAPT) .

> Several suitable beamlines identified and funded at NSLS/NSLS-II

> Future needs at NSLS-II

> Hardware and software to match unique capabilities

> Enhance access of university and industry

> Creation a soft matter network (village) to build capabilities (synthesis, characterization, modeling, application) needed to drive innovation

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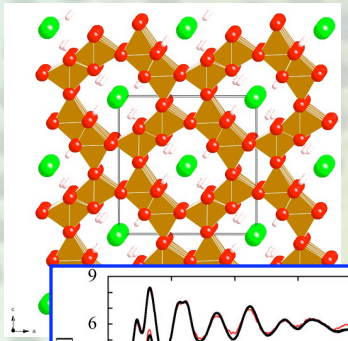
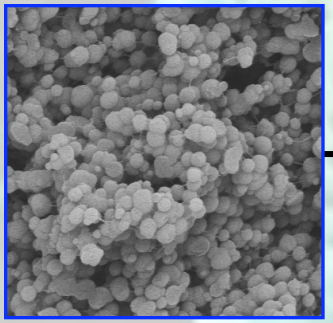
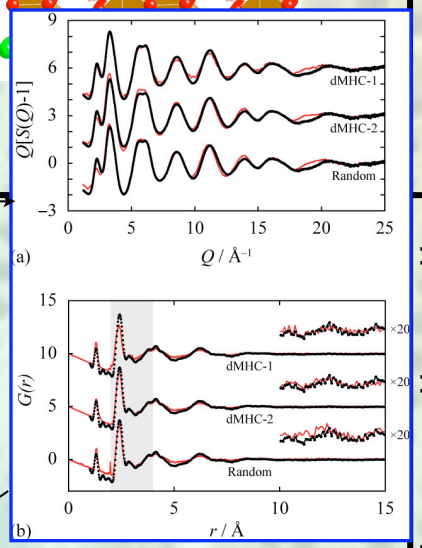
Nano-minerals: key components in the environment

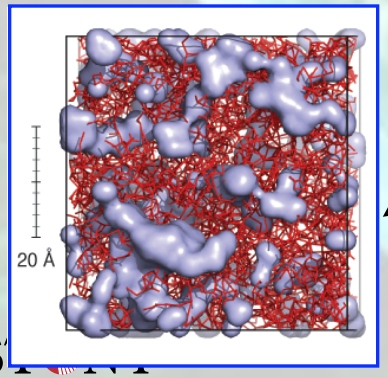
> Important to phase evolution as functions of environmental parameters and biomineralization
  > e.g. Amorphous calcium carbonate

> Materials controlling toxin sequestration and remobilization
  > for example structure evolution of ferrihydrite -> hematite
  > Michel et al (2007) Science
  > Xu et al (2010) Am Mineral.









X-ray Total Scattering from Amorphous Calcium Carbonate Precursors
Reeder, Michel, Phillips, Schmidt, Parise, Goodwin

> High energy beamlines at APS for total scattering

> X17A at NSLS and especially XPD at NSLS-II will provide new opportunities for in operando studies of changes in nano-minerals with time, TP, *f(O_2)*, *f(H_2O)*

> *Chemical Research Center partnership (SBU-Chem/Geo, Temple, Ohio, Penn State)*

> Future - *develop tools for in operando characterization*

> XPD plans for *combined SAXS/WAXS*. Unique insights on nano-mineral on simultaneous length scales

> Crossover between bio-materials and environmental materials (role of organics in in ACC conversion with SAXS/WAXS)

Extreme conditions

> "Action" (tectonics, magnetism etc) at planet interiors

> Push for novel radiation resistant materials

> Emerging interest/expertise in fluid flow/SC CO_2 reactivity, rock interactions, rock mechanics

> Workshops on SC-CO_2 and radiation draw SBU-BNL organizers/interest

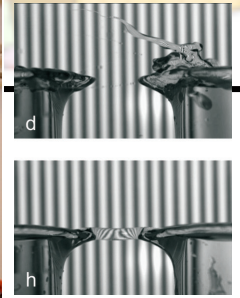
> SBU and BNL diverse interest HV, magnetism, PT

> SBU HP expertise (1 - >10^6 bar) a resource

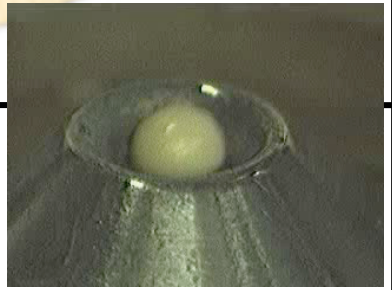
> Diffraction primary method for characterization

> World leading /innovative X-17 HP program

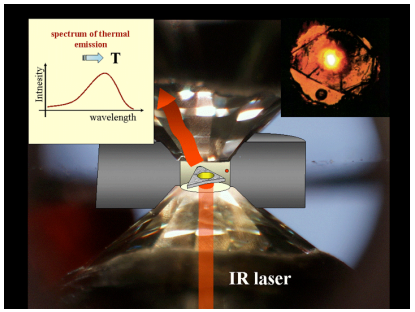
"Paris-Edinburgh" press used at X17-B2



Floating water bridge (30 kV)



Aerodynamically levitated molten droplet



Diamond Anvil cell supports multiple probes

> Future - combinations of technique *in operando*

> "Big iron" HP devices need a base (XPD/4DE)
  > Project beamline XPD; 4DE approved

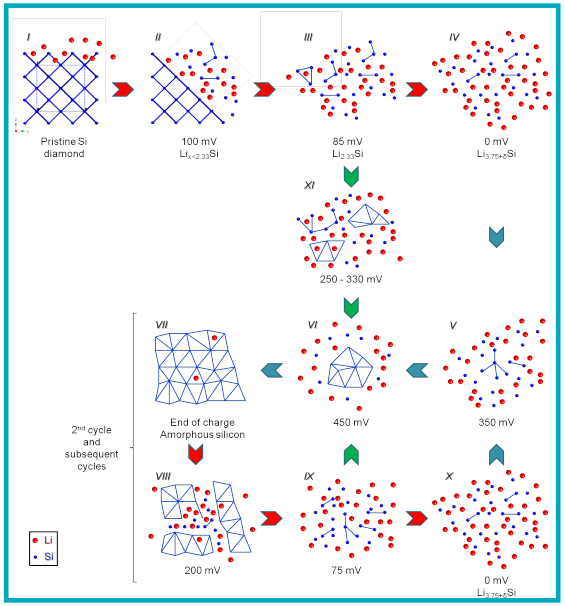
> Diamond anvils relatively easily interfaced

> Cell design-interface software key to day-one operations

>Begin process of further developing "an extreme conditions culture" at NSLS-II

Energy-related applications

> Why devices work and why they fail (Grey group)



Si-anodes: Amorphous phases composed of Li nearby Si-Si clusters and isolated Si environments.

Key, et al., **2010**, submitted to *JACS;*
Key,et al., **2009**, *JACS*, 131, 9239-9249

> Strong industry ties (e.g. topography and batteries)

> EFRC program at SBU

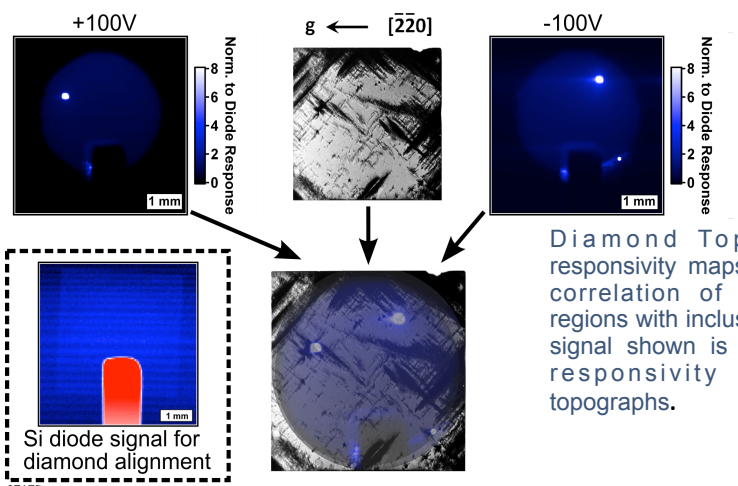
> GE Global work on Na-batteries at X17A

> NYBest involvement (Zhong Zhong)

> Devices inherently heterogeneous

> diffraction key to correlate functionality to meso-structure

> Correlation of functionality in responsivity maps (Dudley group) - Electronic Impact of Inclusions in Diamond



Diamond Topographs and responsivity maps overlaid to show correlation of photoconductive regions with inclusions. Silicon diode signal shown is used to align the responsivity maps with the topographs.

> Future needs - combinations of technique/capability

>Topography: switch between white - monochromatic topography + nanobeam high-res diffraction

> Detector development : large area (10 cm^2), 2 μm spatial resolution, film-like dynamic range

> Space for equipment, cells, *in situ* growth, stressing, annealing, etc. experiments. Long term experiments

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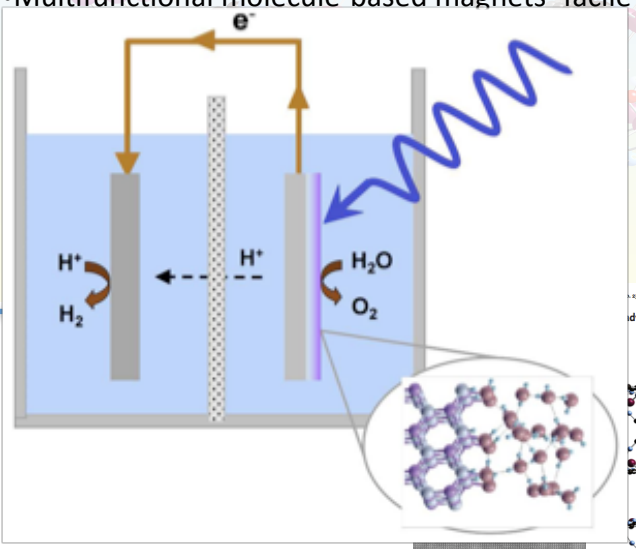
USB Condensed Matter Materials Physics

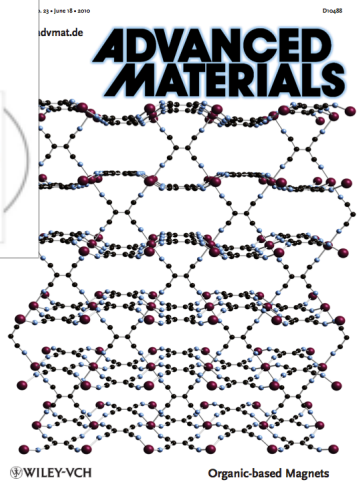
Creating and characterizing new materials with transformational properties and capabilities, based on understanding and control at the atomic scale

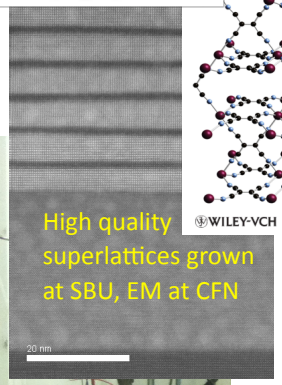
•Semiconductor surfaces can split H_2O.

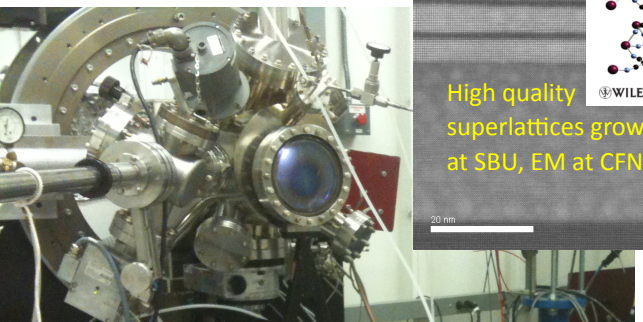
•Physics at interfaces of ferroelectric materials.

•Multifunctional molecule-based magnets -facile "chemical tailoring".









Current work:

New interactions at ferroelectric interfaces,.

Theoretical work on correlation between chemistry of semiconductor surface and ability to catalyze the water oxidation reaction

Advanced structure solution techniques from powder data.

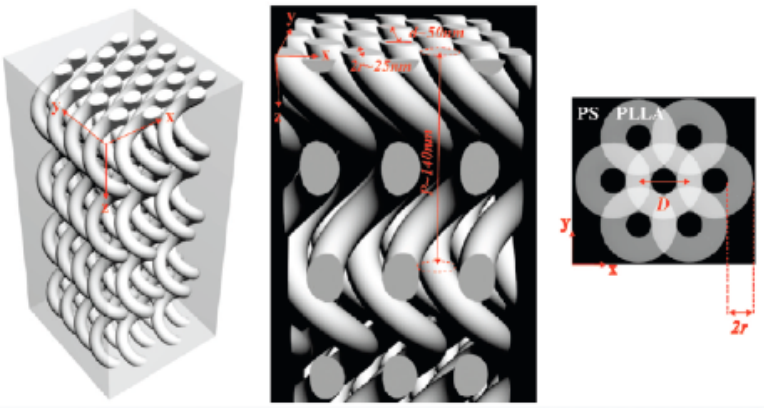
Progress on these and related developments requires:

Surface and interfacial spectroscopies
Time resolved measurements of reaction dynamics
Enhanced ability to perform diffraction and spectroscopic measurements on materials in operating conditions, e.g.,
•high temperature,
•wet environment,
•high gas pressure,
•during vacuum deposition,

Block Copolymers with a Twist

Rong-Ming Ho[1], Yeo-Wan Chiang[1], Edwin L. Thomas[2], Christian Burger[3] and Benjamin S. Hsiao[3]
[1]National Tsing Hua University, [2]Massachusetts Institute of Technology, [3]Stony Brook University

- A new helical phase (H*) from the self-assembly of poly(styrene)-*b*-poly(L-lactide) block copolymers (PS-PLLA BCPs*) was identified. The H* phase consists of hexagonally packed, interdigitated PLLA helical microdomains in a PS matrix; the space group of the phase was determined to be P622.
- A phase diagram of the BCP* was developed to establish the regions in terms of composition and molecular weight, over which the H* phase exists. Also, phase transitions from the H* phase to both the hexagonally packed cylinder phase and gyroid phase were found after long-time annealing, suggesting that the H* phase is a long-lived metastable phase.
- The T_{ODT} values of the PS-PLLA BCPs* always exceed those of the racemic PS-PLA BCPs given equivalent molecular weight and similar composition, indicating that the segregation strength between PS and chiral PLLA blocks is higher than that between PS and racemic PLA blocks. The chiral effect on the molecular interaction is significant and enhances the incompatibility between PS and PLLA blocks in the PS-PLLA BCPs*.



BCPs* comprising chiral entities were designed to fabricate helical architectures (i.e., twisted morphologies) from self-assembly. A new helical phase (H*) with P622 symmetry was discovered in the self-assembly of PS-PLLA BCPs*. Schematic H* phase in which the helical microdomains are packed in a hexagonal lattice with an interdigitated character (left), prospective view (center), and simulated image projected along the z direction of the H* phase (right).

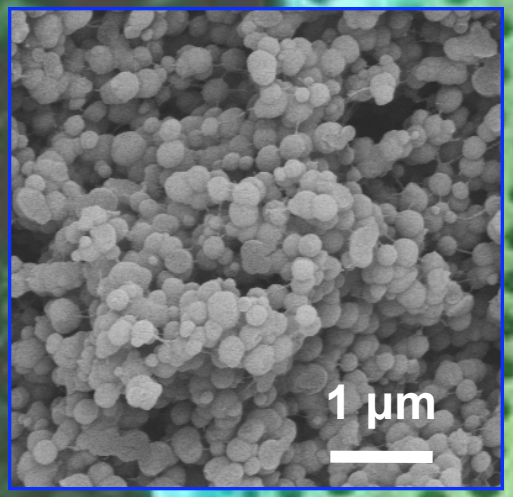
R. M. Ho, Y. W. Chiang, C. K. Chen, H. W. Wang, H. Hasegawa, S. Akasaka, E. L. Thomas, C. Burger, B. S. Hsiao, "Block Copolymers with a Twist," J. AM. CHEM. SOC., 131, 18533 (2009).
Yeo-Wan Chiang, Rong-Ming Ho, Edwin. L. Thomas, Christian Burger and Benjamin S. Hsiao, "A Springlike Behavior of Chiral Block Copolymer with Helical Nanostructure Driven by Crystallization", Advanced Functional Materials, 19(3), 448-459 (2009).

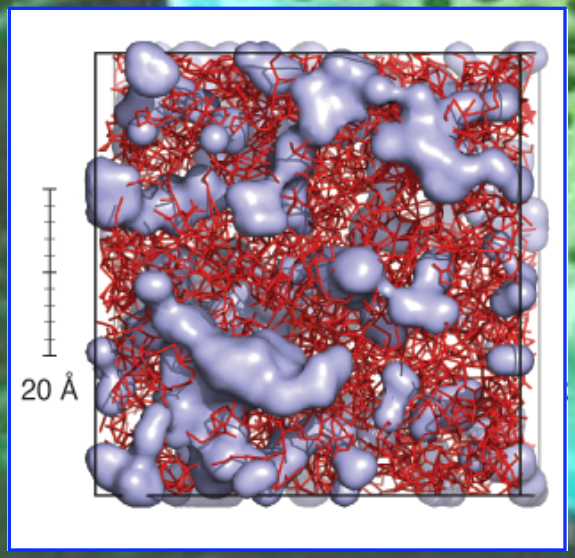
Work performed on beamline X27C.

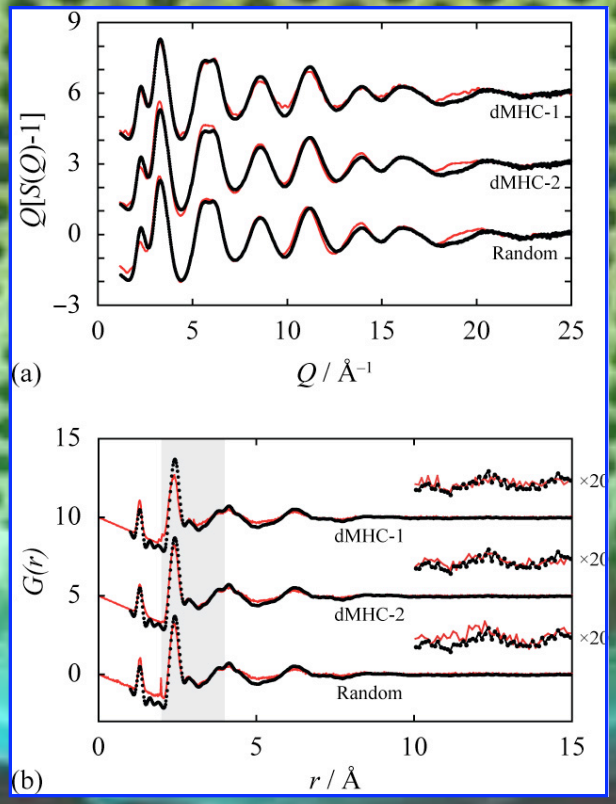
X-ray Total Scattering Studies of Amorphous Calcium Carbonate Precursors

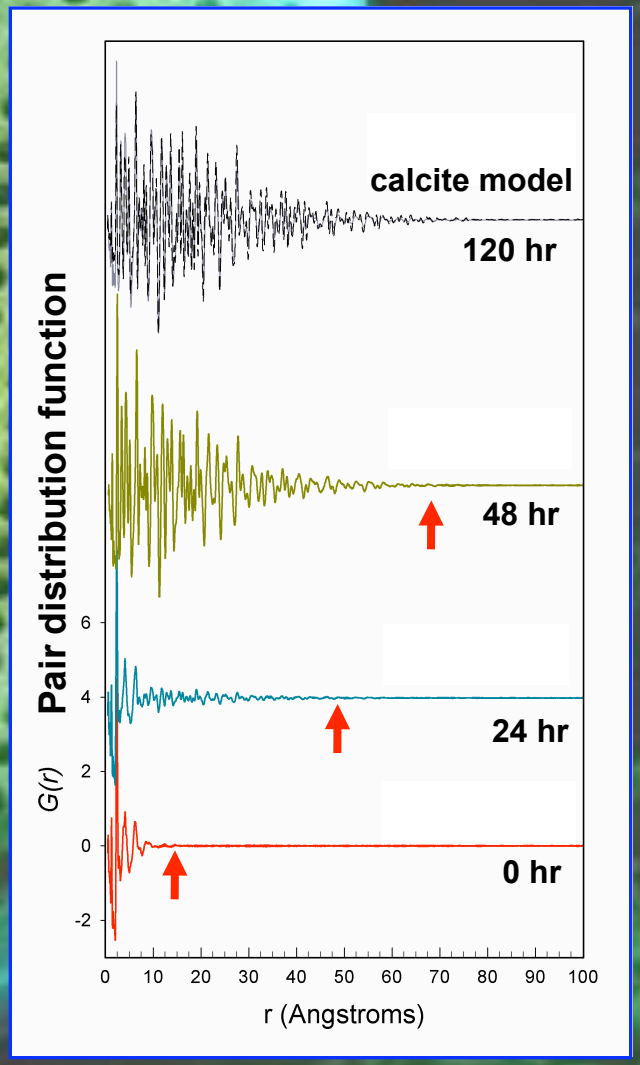
R. J. Reeder, F.M. Michel, B.L. Phillips, M. Schmidt, J.B. Parise, A. Goodwin
Stony Brook University

- Biomineral adapted for functional design
- RMC shows nanoporous structure
- Crystallization pathway modified by dopants

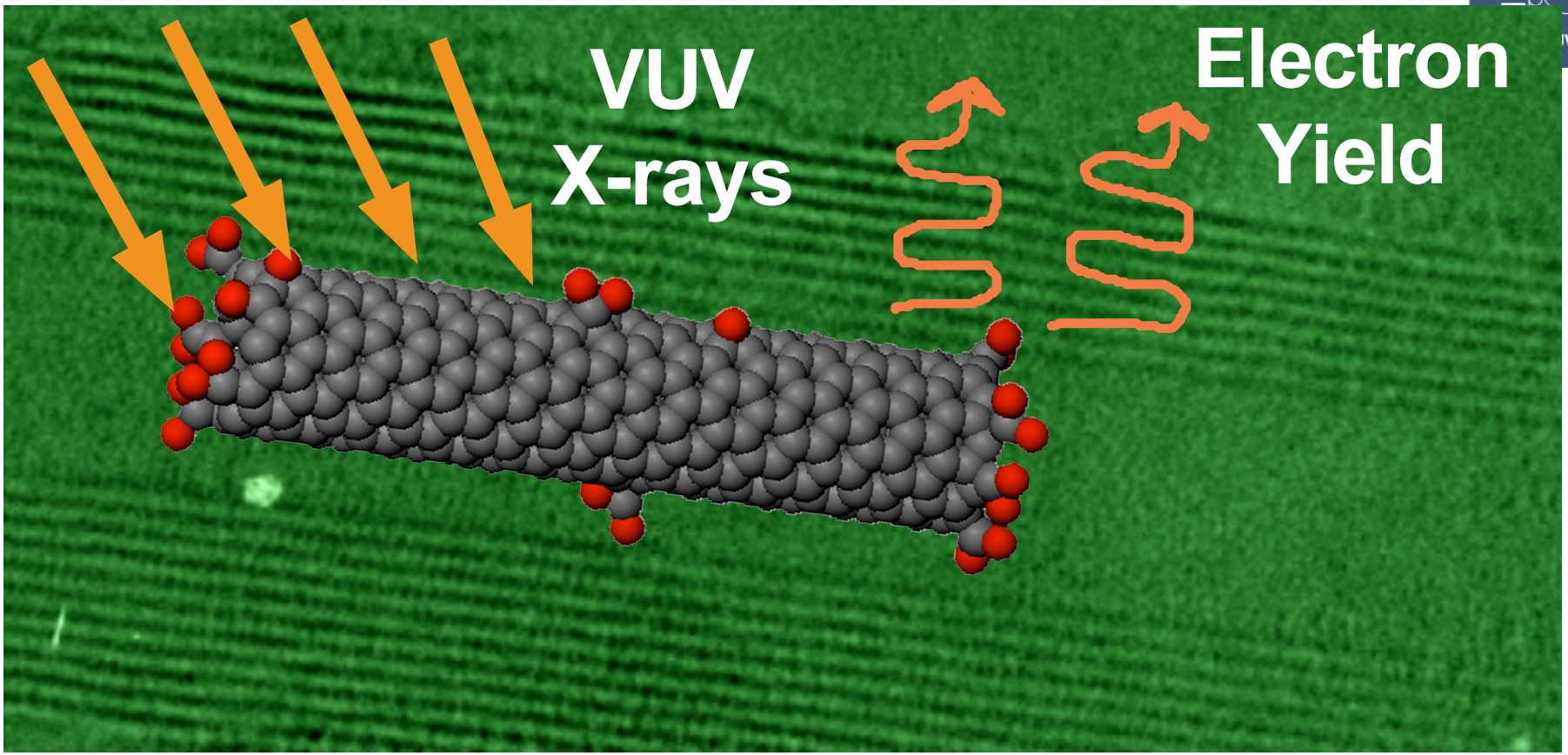








NEXAFS Spectroscopy of Functionalized Carbon Nanotubes



Near-edge X-ray absorption fine structure spectroscopy can be used to track disruption of nanotube electronic structure, alignment, and degree of oxygenation.

References: (a) S. Banerjee et al., *Chem. Commun.*, (7), 772-773 (2004) - SWNTs
(b) S. Banerjee et al., *ChemPhysChem*, v.5, 1416-1422 (2004) - MWNTs
(c) T. Hemraj-Benny et al., *Phys. Chem. Chem. Phys.*, v.7, 1103-1106 (2005) - BN nanotubes
(d) S. Banerjee et al., *J. Phys. Chem. B*, v.109, 8489-8495 (2005) - probing alignment in nanotubes.
(e) T. Hemraj-Benny et al., *Phys. Chem. Chem. Phys.*, v.8, 5038-5044 (2006) – order in arrays.
(f) T-J. Park et al., *J. Phys. Chem. C*, v.112, 10359-10369 (2008) – Fe-containing nanostructures.

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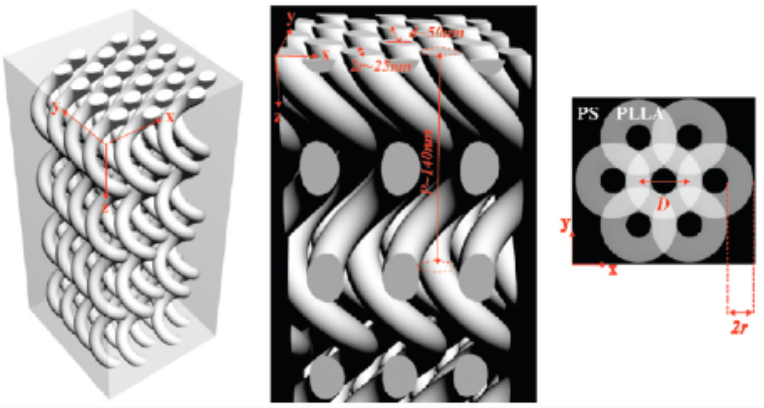

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Yeo-Wan Chiang, Rong-Ming Ho, Edwin. L. Thomas, Christian Burger and Benjamin S. Hsiao, "A Springlike Behavior of Chiral Block Copolymer with Helical Nanostructure Driven by Crystallization", Advanced Functional Materials, 19(3), 448-459 (2009).

Work performed on beamline X27C.

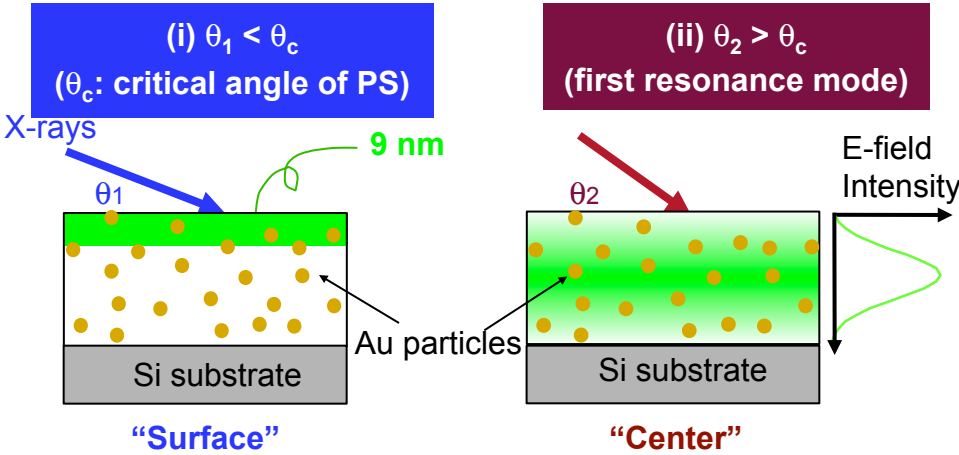
Direct Measurements of Local Properties of Polymers Confined at the Nanoscale

Tad Koga (SBU, Materials Science)

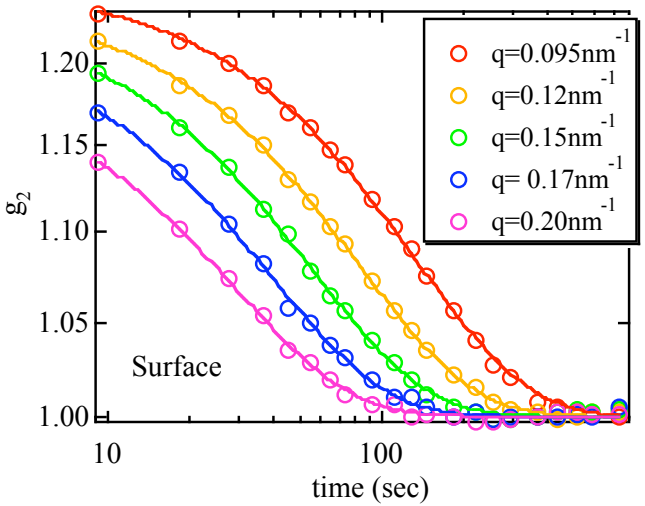
By embedding "dilute" gold nanoparticles in single polystyrene (PS) nanometer films as "markers", we could probe the local viscosity of the free surface at temperatures far above the glass transition temperature (T_g). The technique used was X-ray photon correlation spectroscopy (XPCS) with resonance-enhanced X-ray scattering that allows us to independently measure the marker motions in the regions of interest at the nanometer scale. The results clearly show the viscosity is about 30 % lower than the center of the film. We have found that this reduction is strongly associated with chain entanglements at the free surface rather than the reduction in T_g.

This methodology would open up a new avenue to explore *in-situ* and perturbation-free dynamics at specific surfaces and interfaces of polymer films or membranes to be used for interdisciplinary materials research.

T. Koga, C.Li, M. Endoh, et al., "*Reduced Viscosity of the Free Surface in Entangled Polymer Melt Films*", Physical Review Letters, 104, (2010) 066101-1-4.



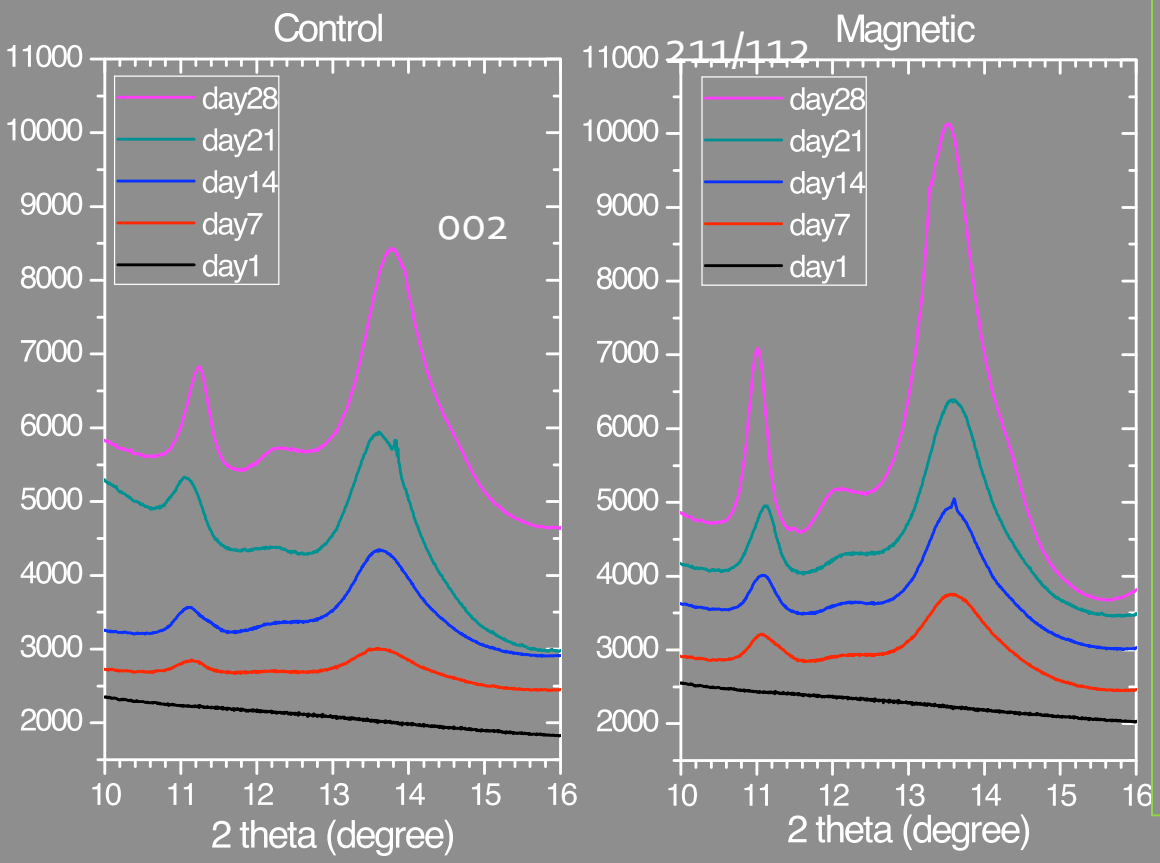
Measure "diffusive" motions of the markers which track the local viscosity of the matrix



The Effects of Static Magnetic Field on Biomineralization of Osteoblasts

Elaine DiMasi (BNL), Xiaolan Ba, Yizhi Meng, Miriam Rafailovich (SBU), Marcia Simon (SBU, Dental School)

GIXD SPECTRA OF BIOMINERALS



- Moderate external magnetic fields (EMF)enhance biomineralization.
- The extra cellular matrix produced in the EMF is more efficient in templating the biomineral deposits.

Sound Velocities of Materials at Extreme Pressures and Temperatures in Conjunction with Synchrotron X-radiation

> Sound velocities measurements provide information about atomic interactions as well as bulk and shear properties at extreme conditions

> Results on geomaterials provide the code to interpret seismic models of Earth's interior in terms of chemical composition and crystal structure (*Li and Liebermann, PNAS, 2009*)

> A unique approach for the study of various time-dependent processes, including phase transformations, deformation and melting

> Precise determination of density for melts/glass at high pressure (*Antao et al., PRL, 2008*)

> Absolute pressure determination on sample and the establishment of pressure scale (*Li et al., JGR, 2005*)

> Large volume press with the capability of probing millimeter sized sample

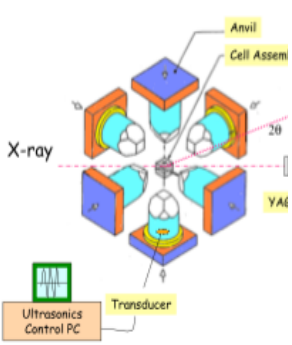
> Simultaneous ultrasonic, X-ray diffraction and X-radiography

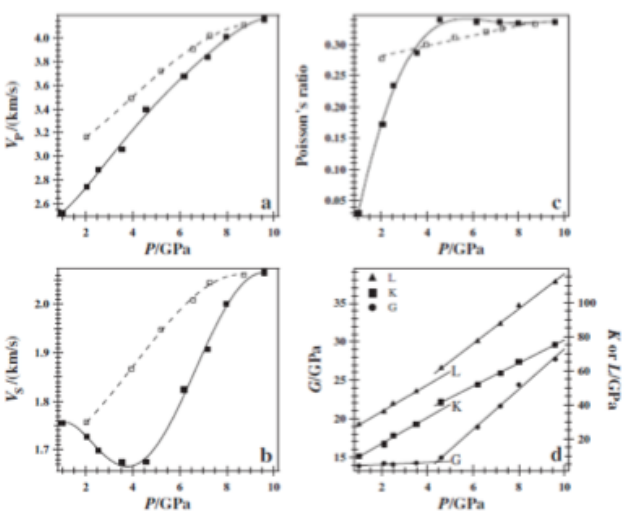
> Compressional and shear wave are measured simultaneously

> In-situ determination of stress with multi-detectors

> Innovative ultrasonic interferometry methods for data acquisition at 1 second level (*Li et al., JAP, 2004*)

> Experiments up to 25 GPa and 1500K at X-17B2 beamline at NSLS.





(Antao et al., PRL, 2008)

Future Development at NSLS II

>Simultaneous PDF and ultrasonic measurements For the determination of melts structure and density

>Coupling of the measurements on mineral-CO2 interactions for sequestration studies

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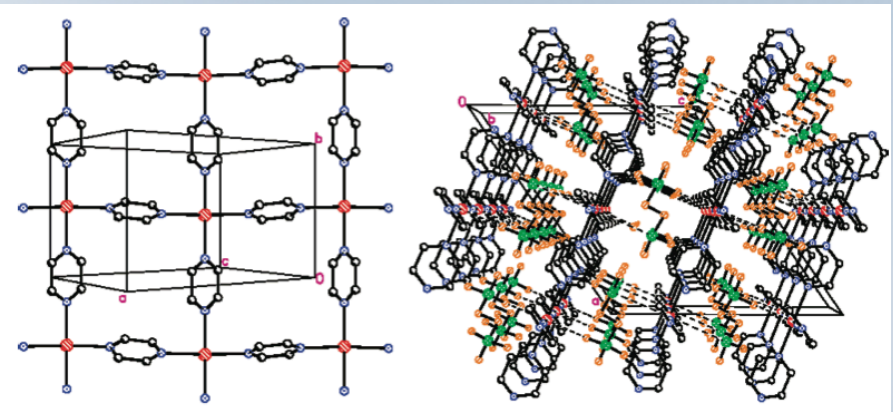
First 2D Silver^{II} Antiferromagnet

J.L. Manson, K. Stone, H.I. Southerland, T. Lancaster, A.J. Steele, M.L. Warter, S.J. Blundell, F.L. Pratt, P.J. Baker, R.D. McDonald, P. Sengupta, J. Singleton, S. Cox, C. Lee, M.-H. Whangbo, C.H. Mielke and P.W. Stephens. Eastern Washington University, Stony Brook University, Oxford University, Rutherford-Appleton Lab., National High Magnetic Field Lab., Los Alamos National Lab., and North Carolina State University.

- High temperature superconductivity is associated with competition between various forms of ordering in layered Cu^{2+} compounds which exhibit antiferromagnetic. However, no other group 1B antiferromagnet has been found.
- $Ag(pyz)_2(S_2O_8)$ (pyz = pyrazine, $C_4H_4N_2$) was first synthesized in 1971, and susceptibility and IR measurements suggested antiferromagnetic interactions between Ag^{2+} ($4d^9$, S=½) ions, but the structure, indeed the precise composition, were not known until our work.
- The crystal structure was solved and refined at the high resolution powder diffractometer at NSLS Beamline X16C, allowing the interpretation of susceptibility and muon spin rotation measurements as a 2D Heisenberg antiferromagnet. The material orders magnetically below T_N = 7.8 K, and the very small value of T_N/J = 0.15 allows the estimate that 2D correlations extend ~1000 lattice spacings at T_N.



The crystal structure of $Ag(pyz)_2(S_2O_8)$, based on 2D square nets of Ag2+ ions resulting from corner-sharing of axially elongated AgN_4O_2 octahedra. Structure determination was complicated by the presence of impurities in samples originally synthesized. Indeed, a method for refining powder diffraction data from samples containing impurity phases was reduced to practice as part of this project: K.H. Stone, S.H. Lapidus, and P.W. Stephens, *J. Appl. Cryst.* **42**, 385-391 (2009). That process guided the synthesis of samples sufficiently pure for the magnetic measurements.

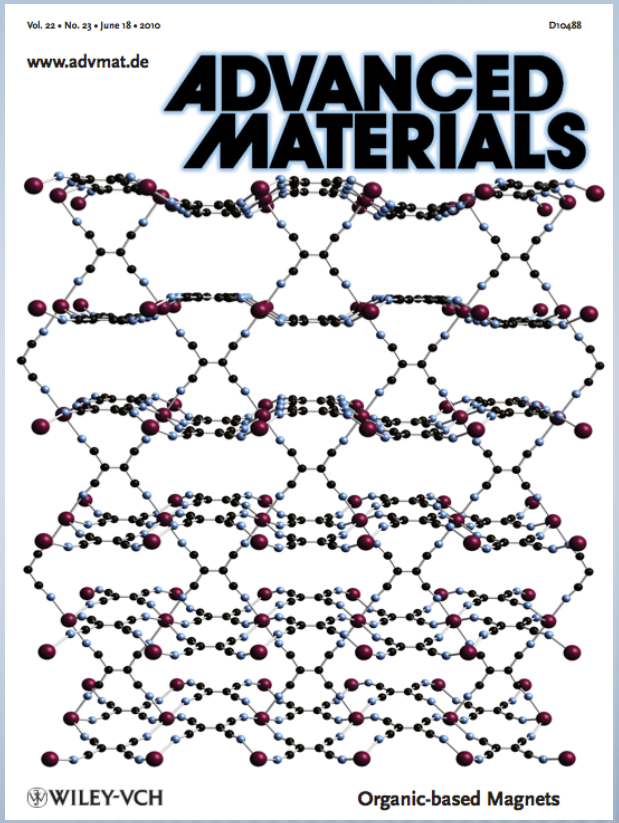
J.L. Manson, K. Stone, H.I. Southerland, T. Lancaster, A.J. Steele, M.L. Warter, S.J. Blundell, F.L. Pratt, P.J. Baker, R.D. McDonald, P. Sengupta, J. Singleton, S. Cox, C. Lee, M.-H. Whangbo, C.H. Mielke and P.W. Stephens, "Characterization of the Antiferromagnetism in $Ag(pyz)_2(S_2O_8)$ (pyz) Pyrazine) with a Two-Dimensional Square Lattice of Ag^{2+} Ions," J. Am. Chem. Soc. ***131****, 4590 (2008).*

Work performed on beamline X16C.

3D *vs.* 2D magnets based on tetracyanoethylene

K.H. Stone, P.W. Stephens, A.C. McConnell, E.Shurdha, K.I. Pokhodnya, and J.S. Miller, Stony Brook University and University of Utah

- Organic-based magnets utilize unpaired electrons residing on organic species to form magnetically ordered materials. They have the potential of combining magnetic properties with other technologically important properties (e.g., electrical or optical) via synthetic methodologies pervasive to organic chemistry.
- The family of metal:TCNE (tetracyanoethylene, $C_2(CN)_4$) magnets have been widely studied, and previous powder diffraction work at X16C has demonstrated a common structural motif of rumpled sheets of M^{II}:μ_4-TCNE.
- Within layer antiferromagnetic interactions between S = 2 (Fe^{++}) or S= 5/2 (Mn^{++}) ions and S = ½ $TCNE^{\bullet-}$ are much stronger than the interlayer interactions in previously known materials, which have ferrimagnetic transition temperatures of ~ 100K. Materials in this family have very different magnetic hysteresis behavior due to the nature of their interlayer magnetic couplings.
- Recently, a new material in this family was discovered with a significantly higher transition temperature of 171 K, which increases to 273 K on application of moderate (15 kbar) pressure. Powder diffraction measurements at X16C elucidated that the higher T_C results from the presence of interlayer bridging $TCNE^{\bullet-}$ which are also magnetically coupled to the Mn^{++} ions, as illustrated here. This points to new opportunities to engineer magnetic properties by appropriate design of the molecular structure.



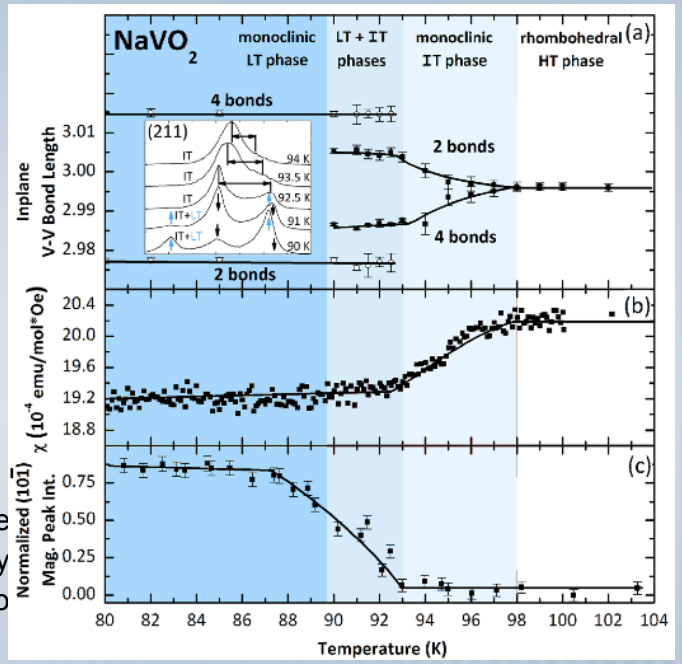
K.H. Stone, P.W. Stephens, A.C. McConnell, E.Shurdha, K.I. Pokhodnya, and J.S. Miller, :$Mn^{II}(TCNE)_{3/2}(I_3)_{1/2}$—A 3D Network-Structured Organic-Based Magnet and Comparison to a 2D Analog," Advanced Materials ***22****, 2514-19 (2010).*

Work performed on beamline X16C.

Successive Orbital Ordering Transitions in $NaVO_2$

T. M. McQueen, P.W. Stephens, Q. Huang, T. Klimczuk, F. Ronning, R.J. Cava, Princeton University, Stony Brook University, NIST Center for Neutron Research, Los Alamos National Lab., and Gdansk University of Technology

- Layered, triangular-lattice transition metal compounds exhibit a wide variety of magnetic and electrical properties as a result of frustrated interactions. At low temperatures, the frustration may be relieved by some small distortion.
- $NaVO_2$ has been proposed as a system that breaks magnetic frustration by orbital ordering, but the nature of observed magnetic transition has been controversial, partly due to poor sample quality arising from rigorous synthetic and handling conditions required to produce this material.
- High resolution powder x-ray diffraction measurements reveal that the material is rhombohedral above 98 K, implying that all in-plane V-V bonds are equal (HT phase).
- From 98 K to 93 K, the material is in a monoclinic intermediate temperature (IT) phase. Susceptibility indicates the development of magnetic correlations, but there is no long range order observed in magnetic neutron diffraction, indicating that they system is still frustrated. The x-ray data shows that the triangular layers distort into a structure creating two long and four short bonds.
- Below 93 K, there is coexistence between the IT phase and a low temperature (LT) phase with reversed distortion – two short and four long bonds. The LT phase breaks the magnetic frustration, and magnetic LRO is observed.



Top: in-plane V-V bond distances, determined from unit cell refinements of high resolution powder diffraction data. Inset shows scans of the rhombohedral (211) reflection, showing a continuous splitting of the peak from the HT to IT phase, and two-phase region of IT and LT forms. Figure also shows magnetic susceptibility and fraction of magnetic phase present.

T. M. McQueen, P.W. Stephens, Q. Huang, T. Klimczuk, F. Ronning, and R.J. Cava, "Successive Orbital Ordering Transitions in $NaVO_2$," Phys. Rev. Lett. ***101****, 166402 (2008)*

Work performed on beamline X16C.

Understanding the (De)lithiation Mechanisms of High Capacity Silicon Electrodes for Lithium-ion Batteries

Baris Key[1], Clare P. Grey[1,2]
[1] Department of Chemistry, Stony Brook University
[2] Chemistry Department, University of Cambridge, UK

- Lithiation and delithiation mechanisms of high capacity negative electrode Si are investigated by using local structure probes such as Solid State Nuclear Magnetic Resonance Spectroscopy and X-ray Pair Distribution Function analysis.
- By characterizing the amorphous phases formed upon lithiation of silicon, the electrochemical processes seen in the discharge and charge cycles were explained in detail for the 1st time. Depending on the state of charge and the local ordering of the lithiated silicides, different mechanisms for (de)lithiation were proposed.
- Understanding these mechanisms is a key step towards building better batteries with these higher capacity electrodes. This will help industry to develop smaller, lighter, safer and cheaper battery packs which will affect mobile electronics (cell phones, laptops etc.), transportation industries (HEVs, EVs) and the grid storage.

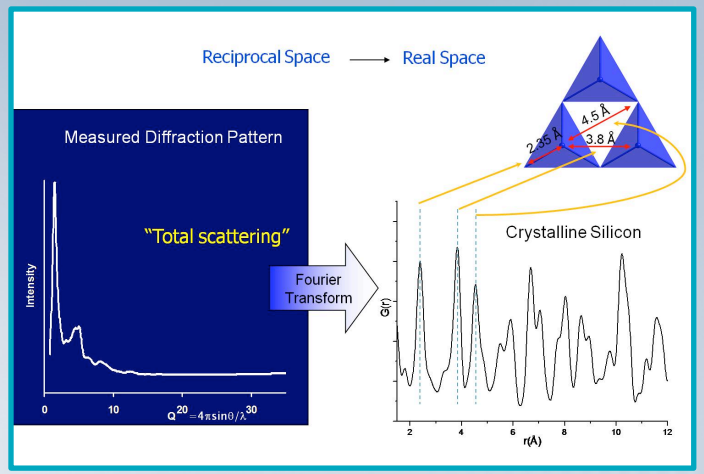


Figure on top: The procedure to obtain PDF. The PDF function contains real space information for all pair-pair correlations in a sample. The PDF data of battery samples combined with solid state NMR data allows characterization of amorphous lithiated silicides.

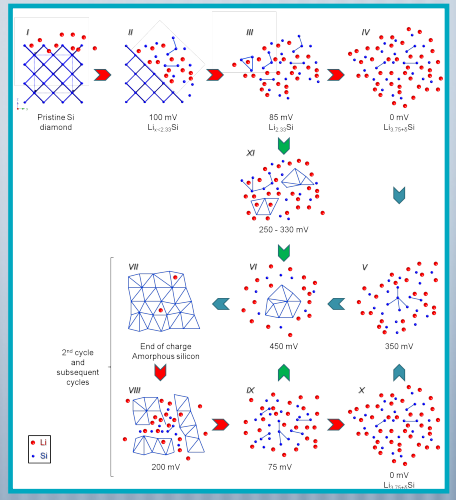


Figure on the side: The complete mechanism(s) for the (de)lithiation of silicon. The characterized amorphous phases are found to be composed of Li nearby Si-Si clusters and Li nearby isolated Si environments.

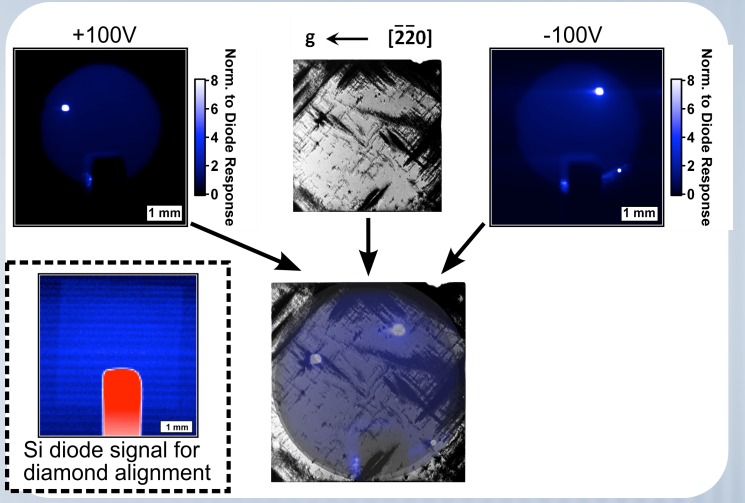
Key, B., Morcrette, M., Tarascon, J. M., and Grey, C. P., **2010**, submitted to *Journal of the American Chemical Society*
Key, B., Bhattacharyya, R., Morcrette, M., Seznec, V., Tarascon, J. M., and Grey, C. P., **2009**, *Journal of the American Chemical Society*, 131, 9239-9249

Work performed on NSLS beamline X7B and APS beamline 11-ID-B

Electronic Impact of Inclusions in Diamond

Erik M. Muller[1], John Smedley[2], Balaji Raghothamachar[1], Mengjia Gaowei[1], Jeffrey W. Keister[3], Ilan Ben-Zvi[4], Michael Dudley[1], and Qiong Wu[4]
[1]Department of Materials Science and Engineering, Stony Brook Universtity; [2] Instrumentation Division; [3] National Synchrotron Light Source II; [4] Collider-Accelerator Division, Brookhaven National Laboratory

- The high mobility, high thermal conductivity, large bandgap and radiation hardness of diamond combine a unique set of properties making devices such as high-flux radiation detectors feasible. However, charge trapping has not been eliminated from fabricated devices therefore resulting in photoconductive gain.
- X-ray topography data are compared with photodiode responsivity maps to identify potential candidates for electron trapping in high purity, single crystal CVD diamond. X-ray topography data indicates that inclusions are a source of electron trapping which affect the performance of diamond X-ray detectors. It was determined that photoconductive gain is only possible with the combination of an injecting contact and charge trapping in the near surface region. Typical photoconductive gain regions are 0.2 mm across; away from these near-surface inclusions the device yields the expected diode responsivity.
- These measurements can provide us with the criteria for choosing diamond crystals for various applications such as diamond amplified photocathode (DAP) in advance.



Topographs and responsivity maps overlaid to show correlation of photoconductive regions with inclusions. Silicon diode signal shown is used to align the responsivity maps with the topographs.

Muller E, Smedley J, Raghothamachar B, Gaowei M, Keister J, Ben-Zvi I, Dudley M, Wu Q., "Electronic Impact of Inclusions in Diamond," in Diamond Electronics and Bioelectronics — Fundamentals to Applications III, edited by P. Bergonzo, J.E. Butler, R.B. Jackman, K.P. Loh, M. Nesladek (Mater. Res. Soc. Symp. Proc. Volume 1203, Warrendale, PA, 2010), 1203-J17-19 .

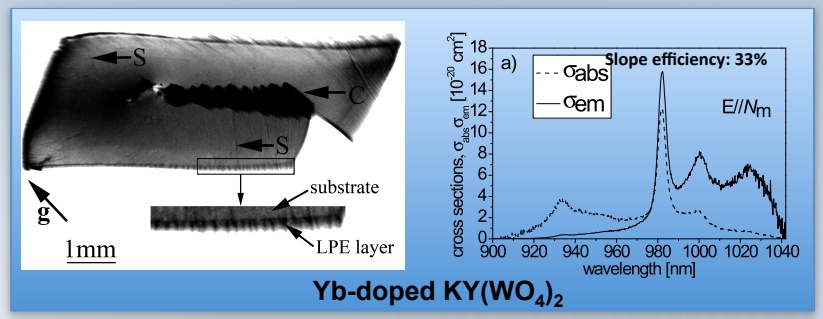
Work performed on beamlines X19C and X6B.

Potassium Rare-Earth Double Tungstates for Thin-Disk Lasers

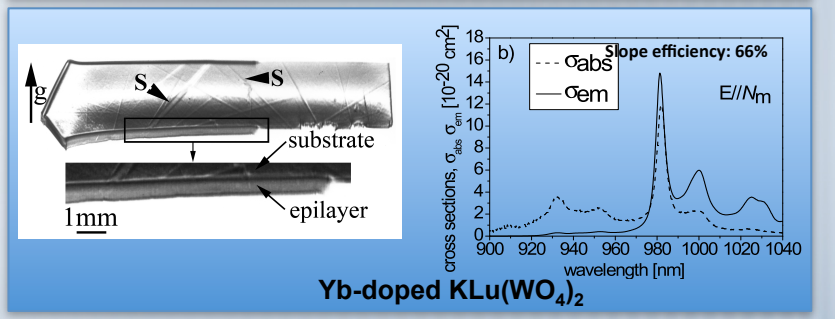
B. Raghothamachar, J. J. Carvajal, M. C. Pujol, X. Mateos, R. Sole, M. Aguilo, F. J. Diaz, M. Dudley
Stony Brook University

- Monoclinic potassium rare earth double tungstates, ($KRE(WO_4)_2$, RE = Y, Lu, Yb; KREW) are well suited as hosts for active lanthanide ion (Ln^{3+}) dopants for diode-pumped solid state lasers, with particular interest in thin-disk laser configurations. Main advantages are the very high values of absorption and emission cross sections of the Ln^{3+} dopants, partly due to the strong anisotropy of these biaxial crystals; and the possibility to dope them with a high concentration of active ions without substantial fluorescence quenching.

- By using synchrotron white beam X-ray topography (SWBXT) at beamline X19C, NSLS, structural defects and strain were mapped in top seeded solution grown (TSSG) bulk $KY(WO_4)_2$ and $KLu(WO_4)_2$ single crystals as well as Yb- and Tm-doped homoepitaxial LPE layers grown on them and correlated with thin-disk laser performance. Tm-doped $KY(WO_4)_2$ epitaxies exhibit stresses that are dominated by effects related to epi-substrate mismatch rather than anisotropy of cyrstal structure. Larger amount of structural stress in Yb-doped $KY(WO_4)_2$ epitaxies leads to lower efficiency in laser operation, while low structural stress in Yb-doped $KLu(WO_4)_2$ Yb:KLuW/KLuW epitaxies leads to much better laser performance.



Yb-doped $KY(WO_4)_2$



Yb-doped $KLu(WO_4)_2$

SWBXT transmission X-topographs of Yb-doped $KY(WO_4)_2$ and Yb-doped $KLu(WO_4)_2$ epitaxies on bulk $KY(WO_4)_2$ and $KLu(WO_4)_2$ and corresponding absorption and emission cross sections of the $^2F_{5/2} \leftrightarrow {}^2F_{7/2}$ transition of Yb^{3+} ion in the epitaxies. Yb-doped $KY(WO_4)_2$ and Yb-doped $KLu(WO_4)_2$. Yb:KYW epitaxial layers is characterized by inhomogeneous stresses while Yb:KLuW epitaxial layers are nearly-strain-free. cw laser operation of Yb:KYW is obtained with a slope efficiency of 33% with respect to the absorbed pump power while a slope efficiency of 66% was achieved for Yb:KLuW layers.

B. Raghothamachar, J. J. Carvajal, M. C. Pujol, X. Mateos, R. Sole, M. Aguilo, F. J. Diaz, M. Dudley, "Synchrotron X-Ray Topography Study of Structural Defects and Strain in Epitaxial Structures of Yb- and Tm-Doped Potassium Rare-Earth Double Tungstates and Their Influence on Laser Performance, J. Electron Mater., 39(6), 823-829 (2010).

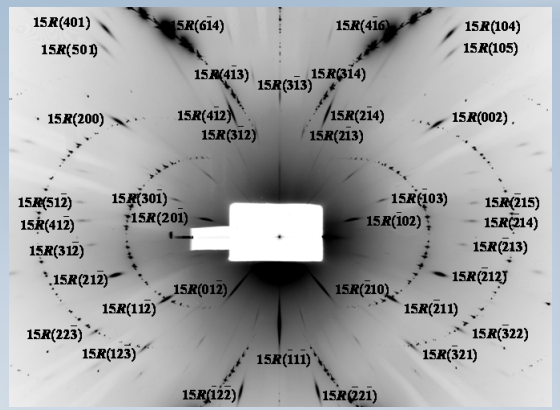
Work performed on beamlines X19C

Single-Crystalline $B_{12}As_2$ on m-plane 15R-SiC

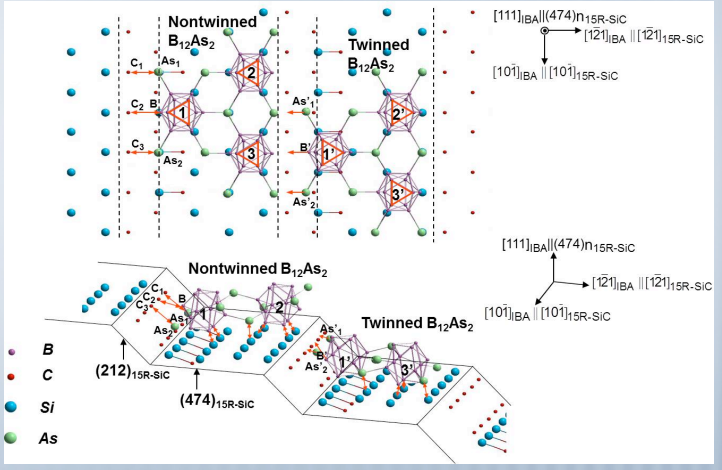
Hui Chen, Guan Wang and Michael Dudley
Stony Brook University

- Single crystal, heteroepitaxial growth of icosahedral $B_{12}As_2$ (IBA, a boride semiconductor) on m-plane 15R-SiC is demonstrated. Previous studies of IBA on other substrates, i.e. (111) Si and (0001) 6H-SiC, produced polycrystalline and twinned epilayers. In contrast, single crystalline and untwinned IBA was achieved on m-plane 15R-SiC. Synchrotron white beam x-ray topography, Raman spectroscopy and high resolution transmission electron microscopy confirm the high quality of the films. High quality growth is shown to be mediated by ordered nucleation of IBA on (474) substrate facets.

- This work demonstrates m-plane 15R-SiC is a good substrate choice to grow high-quality untwinned IBA epilayers for future device applications, such as beta cells, devices capable of producing electrical energy by coupling a radioactive beta emitter to a semiconductor junction.



Laue pattern of a $B_{12}As_2$ film grown on m-plane 15R-SiC showing (353) single growth orientation of the film.



Plan view and 3D perspective view of nontwinned and twinned (353) IBA nucleated on m-plane 15R-SiC surface facets. For the non-twinned IBA, the triangular configuration of B atoms at the bottoms of icosahedra 1, 2 and 3 bond to the similarly oriented triangular configurations of Si atoms exposed on the $(474)_{15R\text{-}SiC}$ terrace. In addition, atoms As_1, B, and As_2 can be well bonded to atoms C_1, C_2, and C_3, respectively, on the neighboring $(212)_{15R\text{-}SiC}$ step riser. In contrast, while the triangular configuration of B atoms at the bottoms of icosahedra 1', 2' and 3' can similarly bond to the $(474)_{15R\text{-}SiC}$ terrace, atoms As'_1, B', As'_2 are not able to reasonably bond to the corresponding C atoms on the neighboring $(212)_{15R\text{-}SiC}$ step riser.

H. Chen, G. Wang, Y. Zhu and M. Dudley et al, "Single-crystalline $B_{12}As_2$ on m-plane (1-100)15R-SiC", Appl. Phys. Lett., 92 (23), 231917 (2008).

Work performed on beamline X19C.

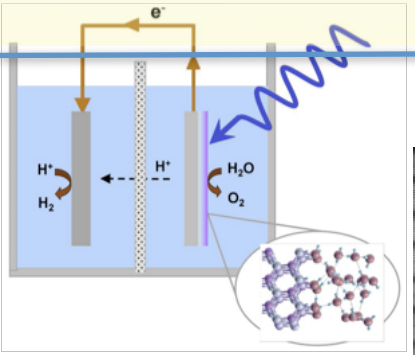
USB Condensed Matter Materials Physics

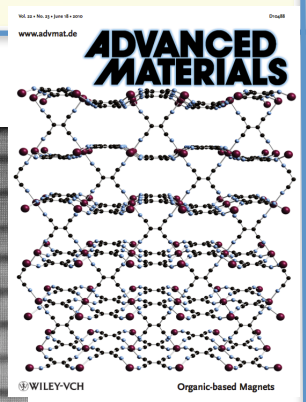
Creating and characterizing new materials with transformational properties and capabilities, based on understanding and control at the atomic scale

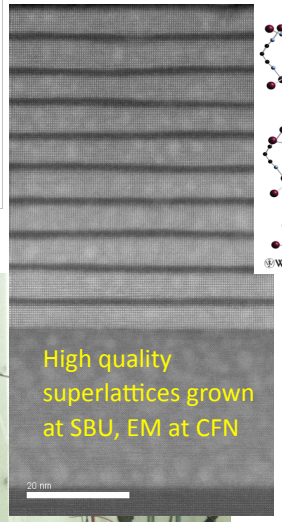
•Semiconductor surfaces can split H_2O using solar photons, but practical materials combining efficiency and durability are needed.

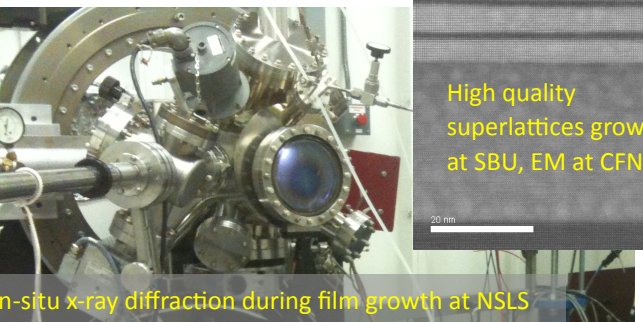
•Physics at the interfaces of ferroelectric materials – Fundamental investigation and harnessing for tailored functionality.

•Multifunctional molecule-based magnets allow tailoring of technologically important properties (e.g., optical, electrical) via synthetic methods pervasive to organic chemistry.









n-situ x-ray diffraction during film growth at NSLS

Current work:

Rapid progress in finding new interactions at ferroelectric interfaces, for example, improper ferroelectricity. These interactions are not yet fully exploited in artificial materials for enhanced ferroelectrics, mulitferroics and piezoelectrics.

How does the chemistry (composition) and physics (polarity) of different semiconductor surfaces correlate with their efficiency to catalyze the water oxidation reaction? We use theoretical and numerical electronic structure methods (density functional theory and beyond).

Advanced diffraction techniques to obtain structural information on samples in available conditions, i.e., not single crystals.

Progress on these and related developments requres:

Surface and interfacial spectroscopies
Time resolved measurements of reaction dynamics
Enhanced ability to perform diffraction and spectroscopic measurements on materials in operating conditions, e.g.,
•high temperature,
•wet environment,
•high gas pressure,
•during vacuum deposition,