Argonne NATIONAL LABORATORY

... for a brighter future

Chemistry Division Overview for Postdoc Pre-Lunch

Al Wagner

March 23, 2007

U.S. Department of Energy

UChicago Argonne LLC

Office of Science
U.S. DEPARTMENT OF ENERGY

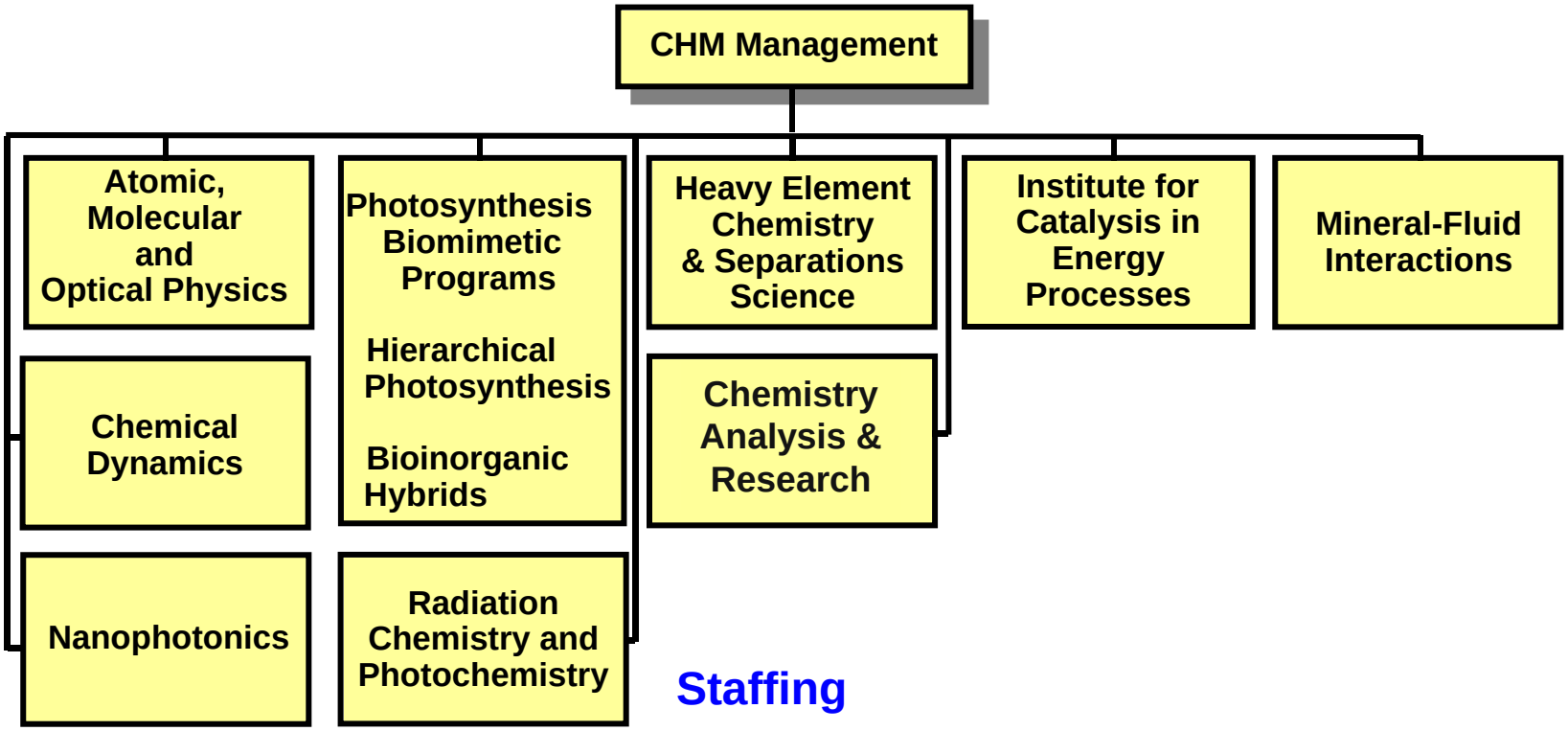
A U.S. Department of Energy laboratory managed by UChicago Argonne, LLC

Outline

1. **Overview of the division:**
   - group structure and staffing

2. **Overview of each group**
   - scientific activity
   - major accomplishments in the past 5 years
   - major new directions
   - major collaborations at Argonne, UC, Northwestern,…

Chemistry Division Overview: Group Structure & Staffing



Staffing

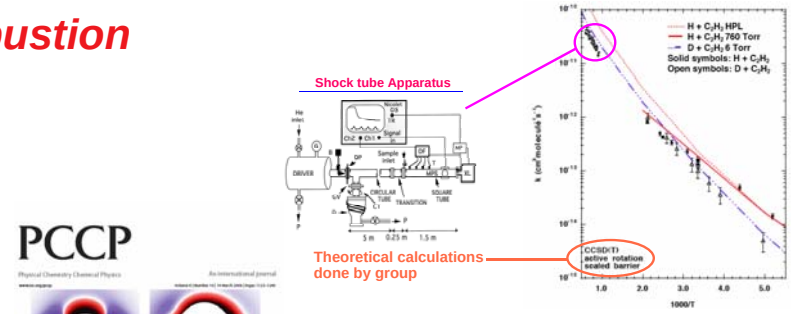
- 44 FTE scientific staff
- 9 FTE support staff:
  ESH, Accounting, Personnel,Building, Secretarial, Computing, Glassblowing
- ~20 FTE temporary staff (postdocs, STAs)

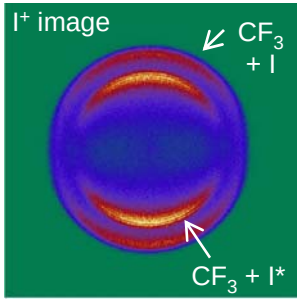
Chemistry Division Overview: Group Activities

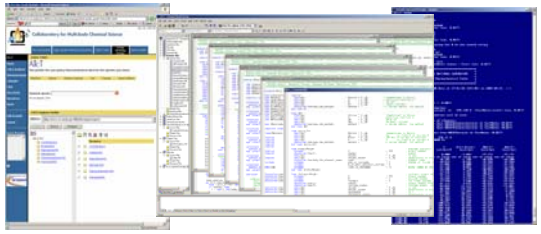
Chemical Dynamics Program

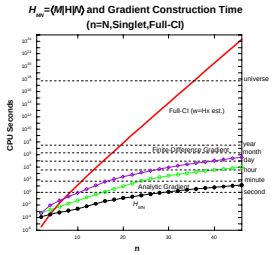
Understanding the fundamental chemistry of combustion

- **vertically integrated effort in experimental and theoretical combustion**
- **from electronic structure to rate constants and mechanism reduction**
- **thermochemistry, chemical dynamics, and chemical kinetics**
- **critical mass sufficient to address complex issues**
- **quality staff attracts new funding**
  - MICS - SciDAC - Computational Nanophotonics









Chemistry Division Overview: Group Activities

Chemical Dynamics Program

- **major accomplishments in the past 5 years**
  - **reaction paths without transition states**
  - **thermodynamics advances: ATcT, corrected heat of formation of OH**
  - **revolutionary non-linear way of doing electronic structure calculations**

- **major new directions for the next 5 years**
  - **alternative fuels**
  - **two senior hires from BNL**
  - **connections to applied programs in ES** ***(LDRD strategic initiative idea)***

- **major collaborations at Argonne, UC, Northwestern,…**
  - **collaborations with MCS**
  - **past collaborations with ES on X-ray transparent engine**
  - **collaborations with Northwestern, the CRF at Sandia Livermore, and many universities around the world**

Chemistry Division Overview: Group Activities

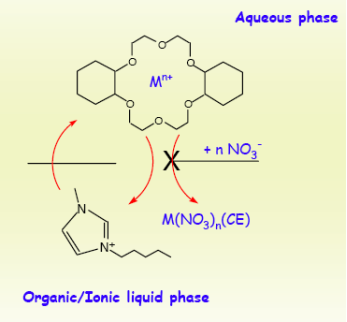
Heavy Elements Chemistry and Separations Science Program:

Atomic and molecular-scale chemistry of heavy elements and fission products

- **discovering new actinide bonding patterns**

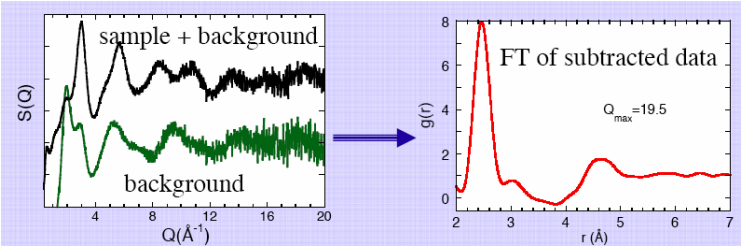
QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

- **understanding third phases in separations**



- **balance of pathways in ionic liquid l-l separations**

- **Actinide facility for APS (e.g., HEXS for Cm^{+3}/1M-$HClO_4$)**



Chemistry Division Overview: Group Activities

Heavy Elements Chemistry and Separations Science Program

- **major accomplishments in the past 5 years**
  - **discovery of actinyl nanospheres**
  - **description of energetics of third phase formation**
  - **discovery that ionic liquids are not "green" in liquid/liquid separations**

- **major new directions for the next 5 years**
  - **actinide biochemistry** →
  - **synthesis hire, hire in complementary gas-phase studies, theory hire?**
  - **basic/applied studies in advanced nuclear energy and waste disposal**
    - **interfacial properties in liquid/liquid separations**
    - **aggregates in separations/ environment** ***(LDRD strategic initiative idea)***

- **major collaborations at Argonne, UC, Northwestern,…**
  - **Notre Dame, Northwestern**
  - **APS/IPNS and CMT**
  - **international collaborations via the Actinide Facility**

Chemistry Division Overview: Group Activities

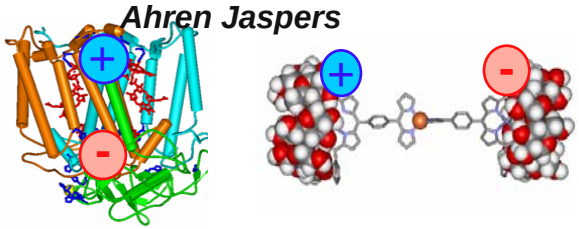
Bioinspired Energy Program

Bioinspired assemblies for energy production

Structural Dynamics Coupled to Photo-charge Transfer:

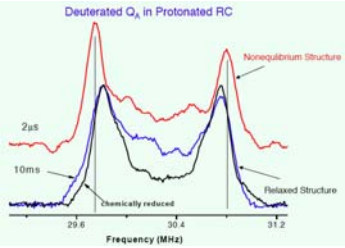
- **photosynthetic proteins and model systems**

Ahren Jaspers



Forefront Techniques:

- **time-resolved FT-EPR/ENDOR**

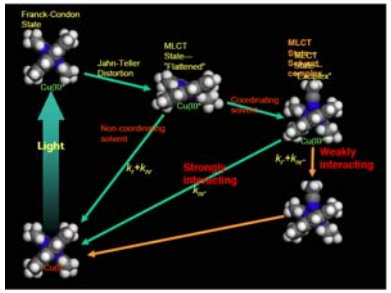


(electron-electron and electron-nuclear couplings)

- **time-resolved X-ray (APS-postdoc/equipment)**

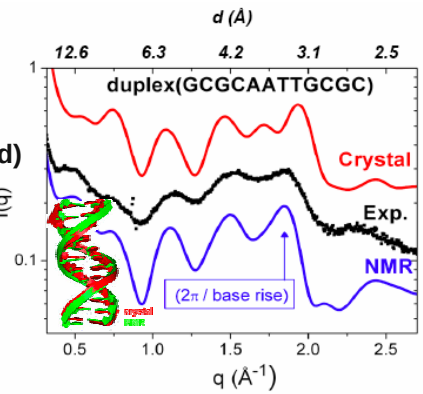
Time-Resolved EXAFS

(metal-ligand pair distances)



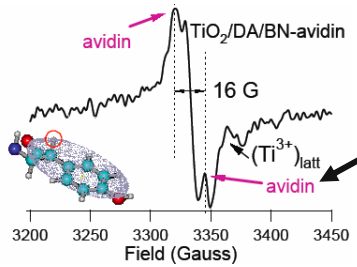
(Time-Resolved) Solution Diffraction

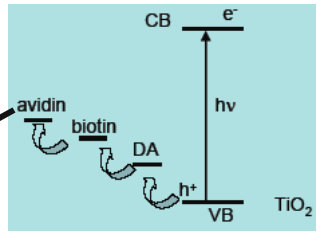
(all atom pair distances)



Nano/bio hybrids

- **drive bio-mimetic sequential charge transfer and separation**





Chemistry Division Overview: Group Activities

Photosynthesis Program (Hierarchical Photosynthetic & Bioinorganic Hybrid Systems)

- **major accomplishments in the past 5 years**
  - **pioneered time-resolved pump/probe laser/X-ray photochemistry measurements (involving both scattering and spectroscopy)**
  - **discovery of linkers between semi-conductor nanoparticles and organic/bio molecules that allows electrons/holes migration**
  - **pioneering of high field EPR techniques for photosynthesis**

- **major new directions for the next 5 years**
  - **Solar Center with Northwestern**
  - **harnessing 1ps APS X-rays in time-resolved studies *(LDRD strategic initiative idea)***
  - **study of response of structures to excess charge and excitation**

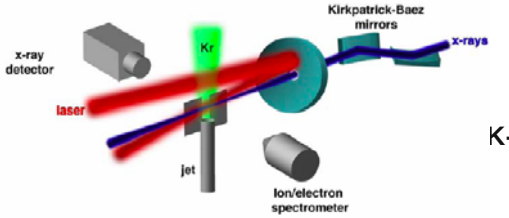
- **major collaborations at Argonne, UC, Northwestern,…**
  - **Northwestern (Solar Center), UC (Jim Norris, Michael Hopkins, Luping Yu, Norbert Scherer)**
  - **APS, CNM**

Chemistry Division Overview: Group Activities

Atomic Molecular and Optical Physics (AMOP) program

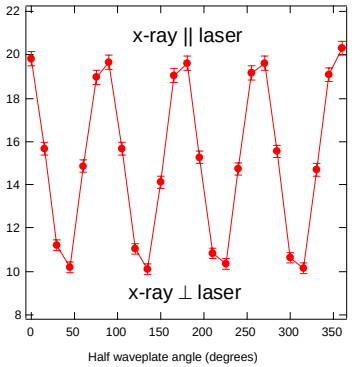
Understanding and controlling fundamental physical processes at high field strength

- **x-ray probes of ultra-fast processes in intense fields**

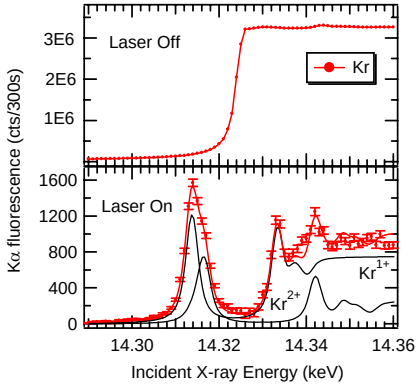


K-edge

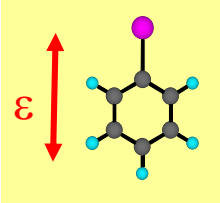
- **Orbital alignment in strong field ionization**



- **x-ray/laser interactions**



- **Laser-aligned molecules (APS test of LCLS application)**



- **New Theory Program**
  - **explained orbital alignment**
  - **synergistic with x-ray/laser overlap studies**

- **AMO LCLS end station & science development**

Chemistry Division Overview: Group Activities

Atomic Molecular and Optical Physics (AMOP) program

- **major accomplishments in the past 5 years**
  - **pioneered ultrafast-laser-induced modifications to x-ray processes**
  - **Developed a time-resolved x-ray microprobe to view atoms/molecules in extreme environments, e.g. at 10 V/Å comparable to Coulomb binding forces of electrons**

- **major new directions for the next 5 years**
  - **quantum control viewed via 1ps X-rays at APS:** ***(LDRD strategic initiative idea)***
    - **molecular behavior at atomic length/time scales**
    - **x-ray processes with ultrafast laser steering of electron motion**
  - **First observation of non-linear x-ray phenomena (at LCLS)**

- **major collaborations at Argonne, UC, Northwestern,…**
  - **AMO group is a Partner at APS (Sector 7) developing ultrafast/ultrasmall laser-x-ray techniques**
  - **UC (David Mazziotti), Ohio State University, Stanford, LBNL**

Chemistry Division Overview: Group Activities

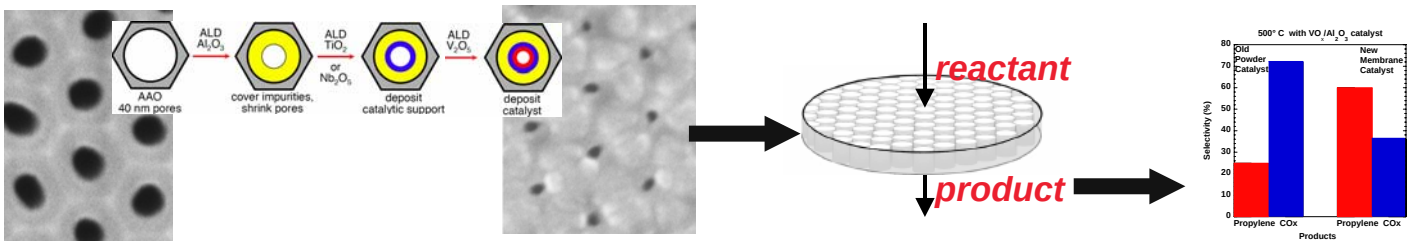
Institute for Catalysis in Energy Processes (ICEP)

Atomic and molecular-scale control of catalytic reactions for energy technologies

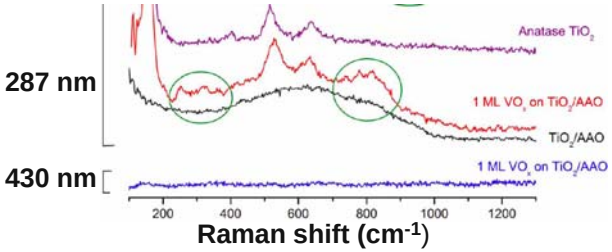
- **Program with Northwestern University directed by joint appointment Peter Stair**

- **By FY09 ICEP will combine 5 BES-funded ANL/NU programs -> 1 program with shared equipment / students and postdocs / common review**

- **One major focus on uniform nano-structured catalysts with:**
  - **substrate control**
  - **active site control**
  - **supporting theory**



- **Major shared instrumentation**
  - **operando Raman Spectroscopy Laboratory (LDRD)**
  - **powder/membrane catalytic reactor**
  - **APS operando x-ray studies**
  - **size selected cluster deposition**



Chemistry Division Overview: Group Activities

Institute for Catalysis in Energy Processes (ICEP)

- **major accomplishments in the past 5 years**
  - **demonstrated efficacy of nanoporous membranes as catalysts platforms**
  - **pioneered the application of X-rays to *in situ* studies of catalysis**
  - **among leaders in catalytic properties of size-selected clusters**

- **major new directions for the next 5 years**
  - **heterogeneous catalysis of cellulose conversion to fuels**
  - **establishment of an ANL Catalysis Center as a focus of basic/applied catalysis research at ANL** ***(LDRD strategic initiative idea)***

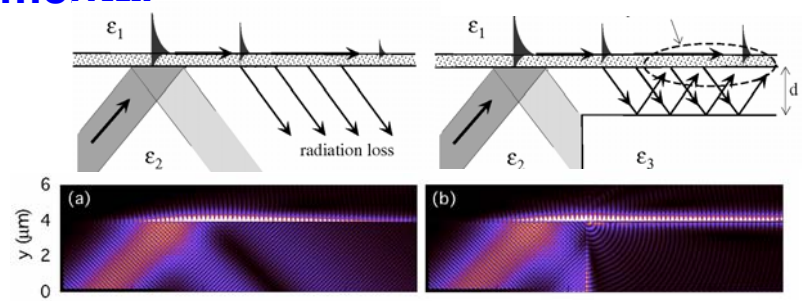
- **major collaborations at Argonne, UC, Northwestern,…**
  - **deep collaborations with CMT, ES, and MSD**
  - **first "center" with Northwestern**

Chemistry Division Overview: Group Activities

Computational Nanophotonics Program

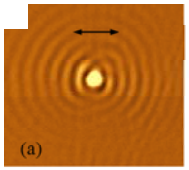
Controlling light on the nanoscale

- **synergistic effort with experimental programs in CNM, MSD**
- **regenerated plasmons**

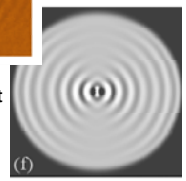


- **interferometric light transmission thru nanoholes**

7.5 μm² top surface NSOM images



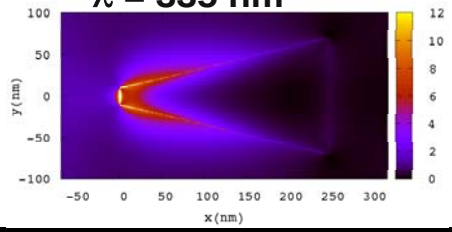
FDTD result



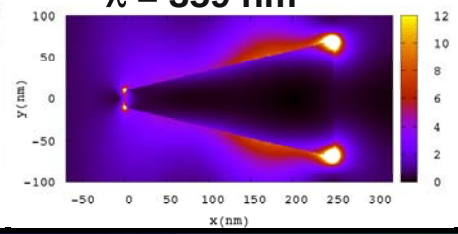
FDTD modeling shows interference between directly penetrating waves and SPPs leads to the fringes.

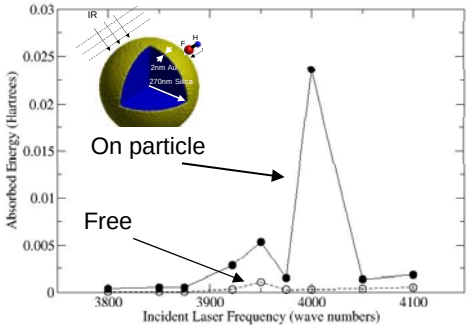
- **photoresponsive polymers for nanoscale imaging**
- **SPARC (surface plasmon assisted reactive chemistry)**

λ = 335 nm



λ = 359 nm





Chemistry Division Overview: Group Activities

Nanophotonics Program

- major accomplishments in the past 5 years
  - theoretical/experimental agreement on nanophotonic phenomena
    - polymer distortion in near field
    - mechanism of hole-driven plasmon waves
    - mechanisms of hole arrays for sensors
  - theoretical predictions currently under experimental study
    - extended plasmon propagation
    - light bending in sub-wavelength metal structures

- major new directions for the next 5 years
  - better multiscale merge of atomistic and continuum modeling of nanophotonics
  - Surface Plasmon Assisted Reactive Chemistry (SPARC) for catalysis, sensors,…

- major collaborations at Argonne, UC, Northwestern,…
  - UIUC (John Rogers), NU(George Schatz, Mark Ratner)
  - CNM (Gary Wiederrecht, Orlando Aucello)
  - MSD (Ulrich Welp)

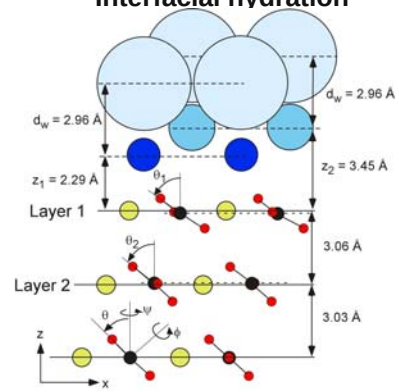
Chemistry Division Overview: Group Activities

Mineral-Fluid Interactions: X-ray Studies at APS

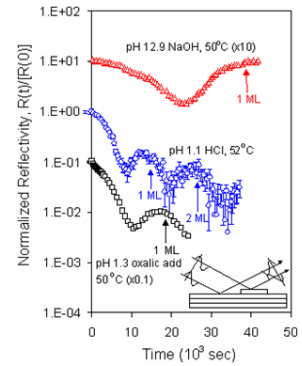
Interfacial processes fundamental to low-temperature geochemistry

- **Developing new fundamental insights**
  - **mineral surface hydration**
  - **mineral dissolution processes**
  - **ion adsorption structures**

Interfacial hydration

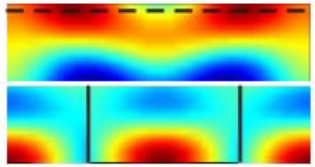


Mineral dissolution

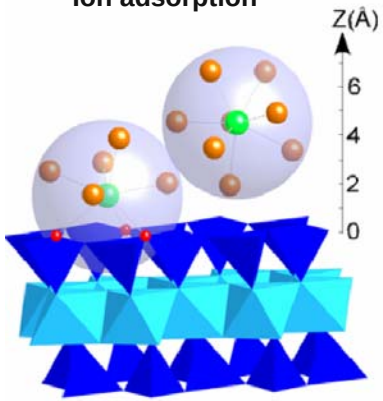


- **Application and development of new in-situ probes**
  - **scattering/spectroscopy (Resonant Anomalous X-ray Reflectivity)**
  - **model-free interpretation (phase and amplitude measurement)**

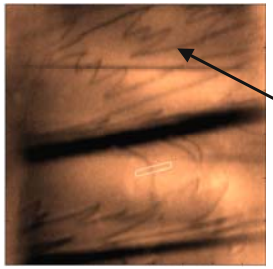
Ion adsorption



Ion adsorption



- **X-ray Reflection Interface Microscope**



Steps on the surface

Chemistry Division Overview: Group Activities

Mineral-Fluid Interactions: X-ray Studies at APS

- **major accomplishments in the past 5 years**
  - **understanding molecular mechanisms of water/ions interactions with mineral surfaces**
  - **new X-ray based approaches for studying interfaces**
    - **anomalous ion-species specific techniques**
    - **model-independent techniques**
  - **X-ray microscope**

- **major new directions for the next 5 years**
  - **interfacial properties in liquid/liquid separations**
  - **impurity distributions within minerals**

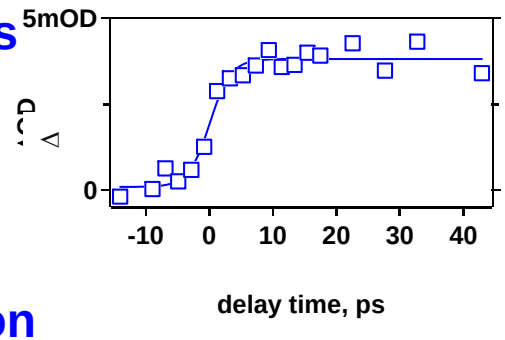
- **major collaborations at Argonne, UC, Northwestern,…**
  - **UIC (N. Sturchio, K. Nagy), NU (M. Bedzyk)**
  - **ORNL, Penn State, Illinois Water Survey, Vanderbilt multi-institutional project**

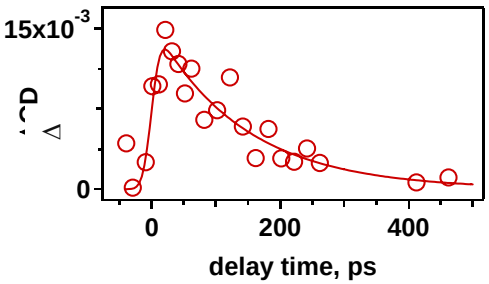
Chemistry Division Overview: Group Activities

Radiation and Photochemistry Group

High energy radiation effects on condensed media

- pioneering pico-sec radiation studies
  - terawatt fs laser
    + wakefield acceleration of electrons
  - observe e- thermalization/solvation

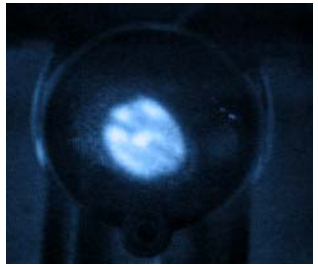




- femto-second laser photoionizaton studies in condensed media

- develop new uses / support for linear accelerator
  - positron generation:
    - one of the world's most intense slow positron sources with a flux of 3×10^7 slow positrons/sec
    - for studying voids, surface conditions
    - partial support out of Eglin AFB, many university collaborators

Positron Signal from Microchannel Plate



Chemistry Division Overview: Group Activities

Radiation and Photochemistry Group

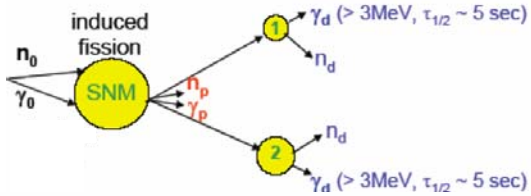
- major accomplishments in the past 5 years
  - pioneer terawatt laser wakefield accelerator for chemistry purposes
  - development of one of the world's most intense sources of slow positrons

- major new directions for the next 5 years
  - exploit laser wakefield acceleration for early radiation processes in liquids
  - mechanistic studies of radiation damage in materials
  - develop a surface studies program with positrons

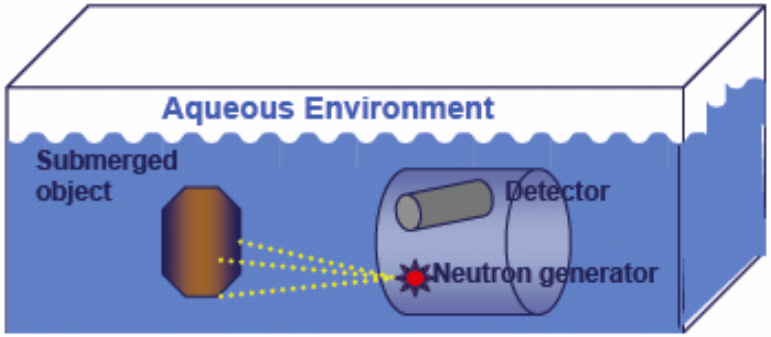
- major collaborations at Argonne, UC, Northwestern,…
  - collaborations with ANL Accelerator Institute (table top XUV source)
  - collaborations with Notre Dame (Radiation Lab), BNL, PNNL
  - collaborations in positrons with UC Irvine, U. of Kansas, Marquette

Chemistry Division Overview: Group Activities

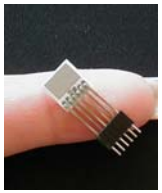
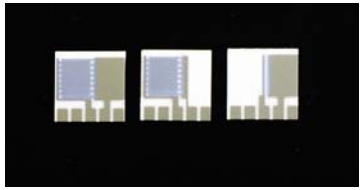
Chemical Analysis and Research Program



- **Neutron activation detection of WMD for Naval activities**
  - **Neutron activation analysis system with a combination of neutron sources and detectors**
  - **Monte Carlo N-Particle analyses to optimize system design**
  - **Measure underwater performance**
  - **Maritime demonstration**



- **Applied development/evaluation of sensors and electronic noses for chemical detection**
  - **Develop nanoparticle and macroparticle sensors**
  - **Characterize surface reactions using x-rays**
  - **Develop/test sensor arrays**
  - **Incorporate NRL's software for "real-time" monitoring of military bases**

- **Investigation of chemical system to remove arsenic from drinking water via X-rays**

- **New to CHM as our first applied program**

Chemistry Division Overview: Group Activities

Chemical Analysis and Research Program

- **major accomplishments in the past 5 years**
  - **important contributors to the establishment of ANL-ONR Office**

- **major new directions for the next 5 years**
  - **rapid expansion in neutron activation studies for WMD**
  - **win new funding for nanoparticle sensors**

- **major collaborations at Argonne, UC, Northwestern,…**
  - **APS, IPNS**
  - **University of Texas Nuclear Engineering Department**

Chemistry Division Overview: Summary

- Chemistry Division has a group structure
- Almost all groups have postdocs engaged in leading research
- Almost all groups collaborate with other divisions and major universities