The Next Generation Light Source:
Facility Overview and
Applications for Understanding Correlated Materials

Bob Schoenlein

NGLS Initiative
MSD Ultrafast Materials Program
CSD Ultrafast X-ray Science Lab

How do the properties of matter emerge from the: correlated motion of electrons, & coupled atomic/electronic structure?

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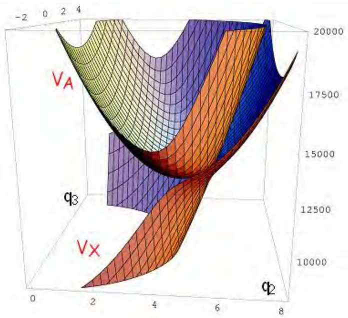
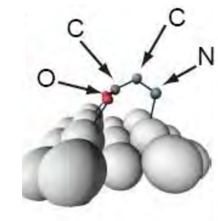


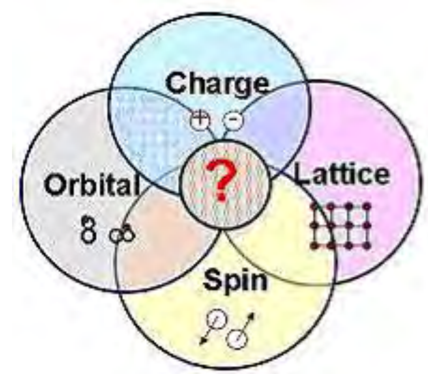
Understanding Chemical Reactivity

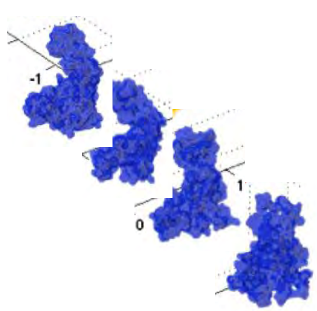
- beyond simple adiabatic potential energy surfaces
  Born-Oppenheimer approximation $\psi_{total} \neq \psi_{nuclear}\ \psi_{electronic}$
- charge transfer, catalysis, photosynthesis (natural and artificial)



Understanding Correlated Materials- Beyond Bloch

- beyond single-electron band structure
- charge correlation, nanoscale organization, charge/spin/lattice coupling
- superconductivity, colossal magnetoresistance, exotic properties





Imaging structure ⇒ imaging "function" in biological systems

- **structure ♦ dynamics ♦ function**
- identify conformational states - pathways connecting conformational states
- interacting proteins and molecular machines

Fundamental Time Scales in Condensed Matter

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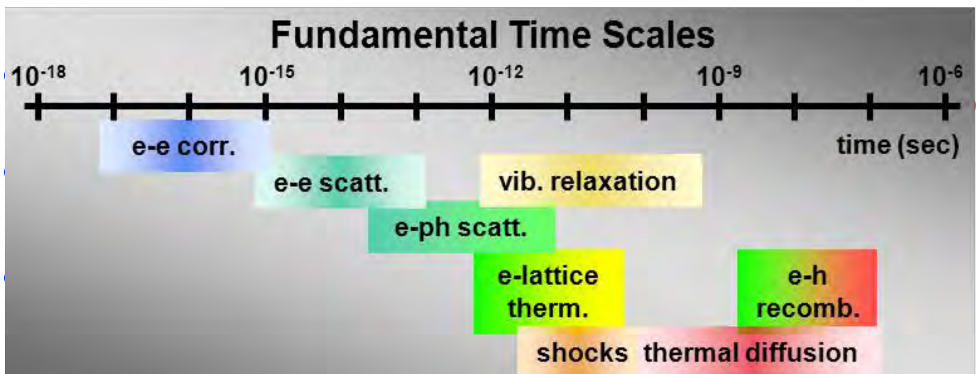
Atomic Structural Dynamics

atomic vibrational period: $T_{vib} = 2\pi(k/m)^{-1/2} \sim 100$ fs

$k \sim eV/a^2$ $m \sim 10^{-25}$ Kg



Sample recovery time ⇔ rep. rate

Electronic Structural Dynamics

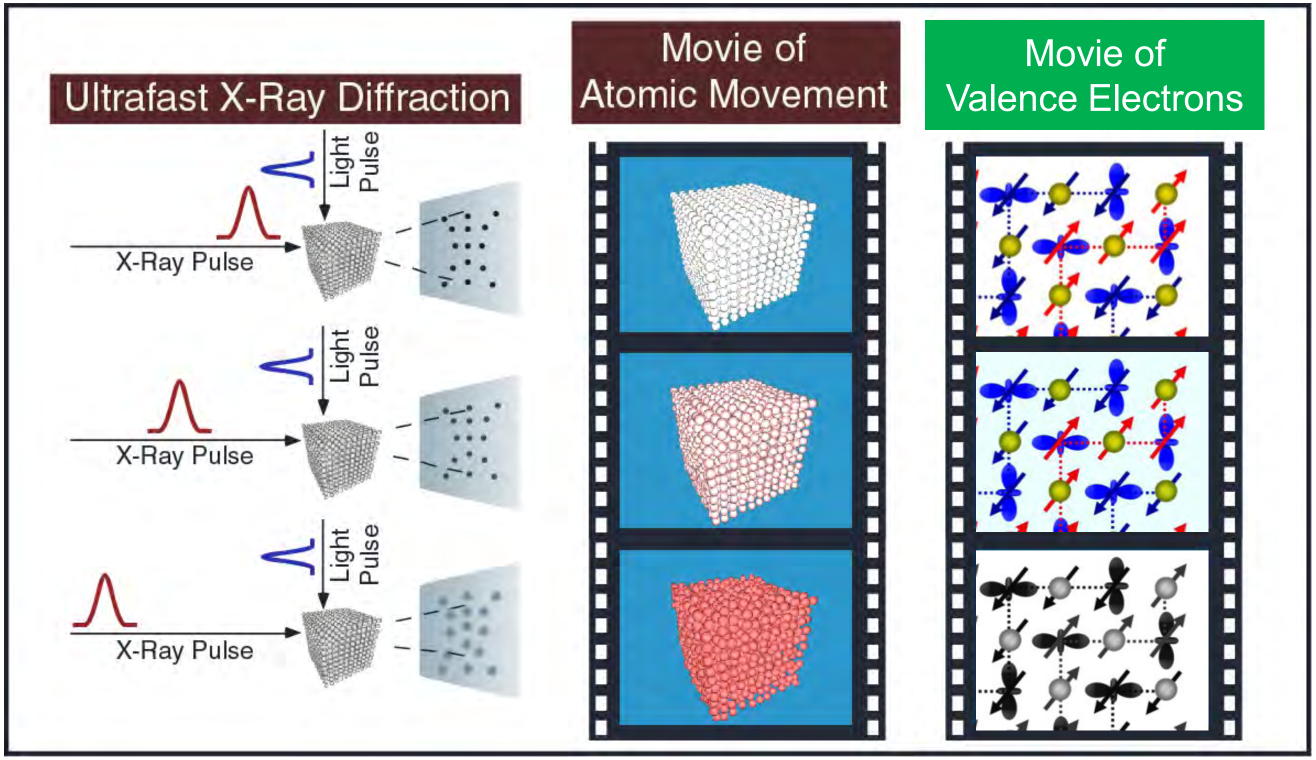
electron-phonon interaction ~ 1 ps
e-e scattering ~10 fs
e^- correlation time ~100 attoseconds (a/V_{Fermi})

- **bond dynamics, valence charge flow**
- **charge transfer**
- **electronic phase transitions**
  - correlated electron systems

Ultrafast Measurements:

- separate correlated phenomena in the time domain
- direct observations of the underlying correlations <u>as they develop</u>

Fundamental Time Scales in Condensed Matter



Ultrafast Measurements:

- ***separate correlated phenomena in the time domain***
- ***direct observations of the underlying correlations <u>as they develop</u>***

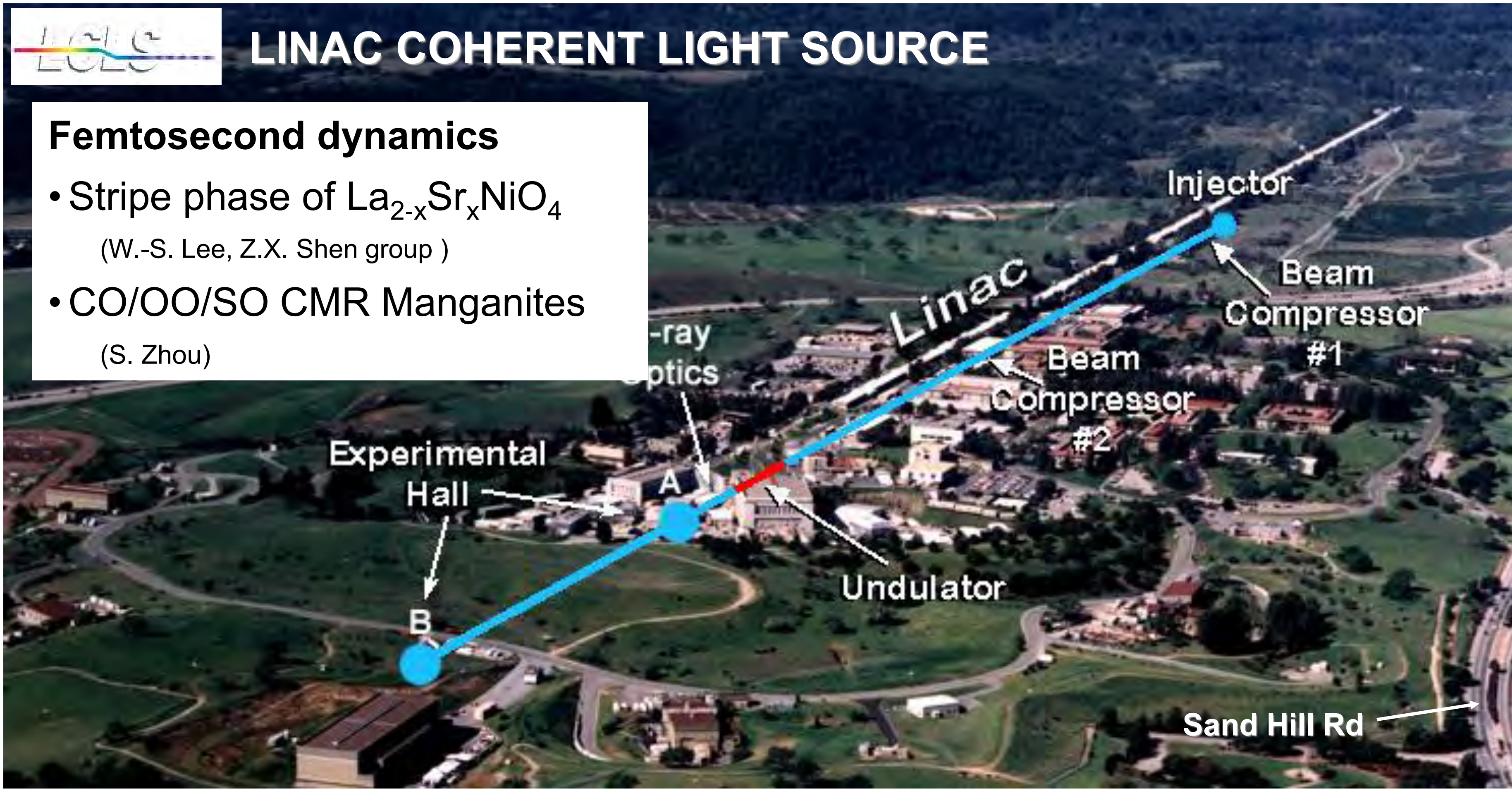
Future – Ultrafast X-ray Science

X-ray Free-Electron Lasers!

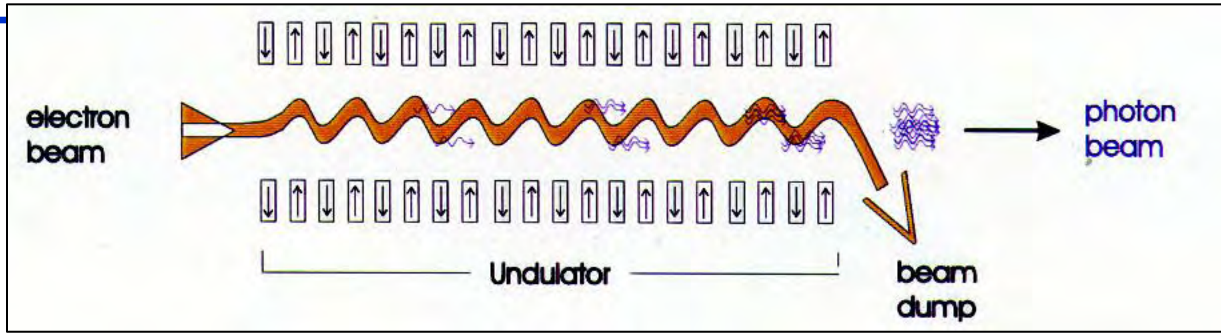
SLAC National Accelerator Laboratory

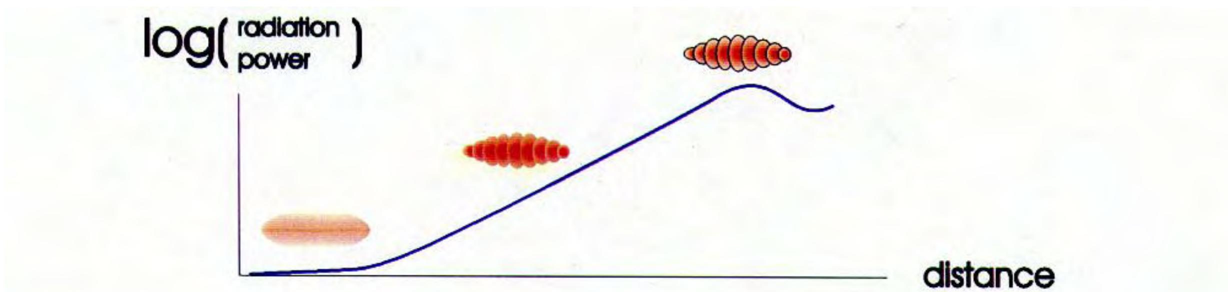
Femtosecond dynamics

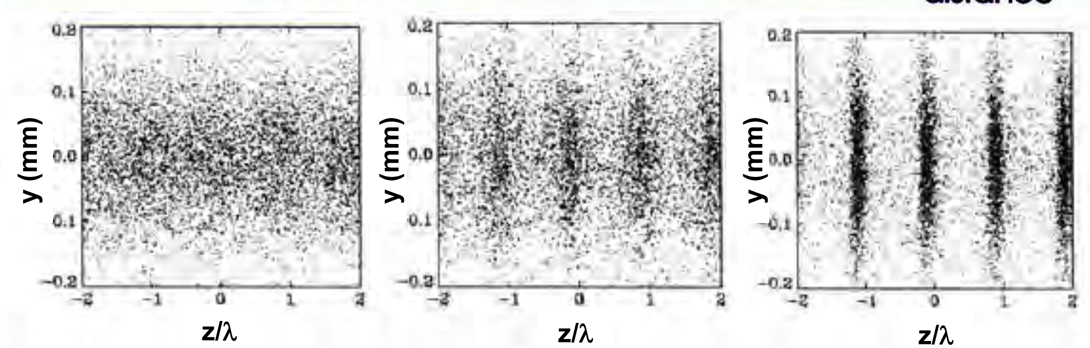
- Stripe phase of $La_{2-x}Sr_xNiO_4$
  (W.-S. Lee, Z.X. Shen group)
- CO/OO/SO CMR Manganites
  (S. Zhou)



Self-Amplified Spontaneous Emission





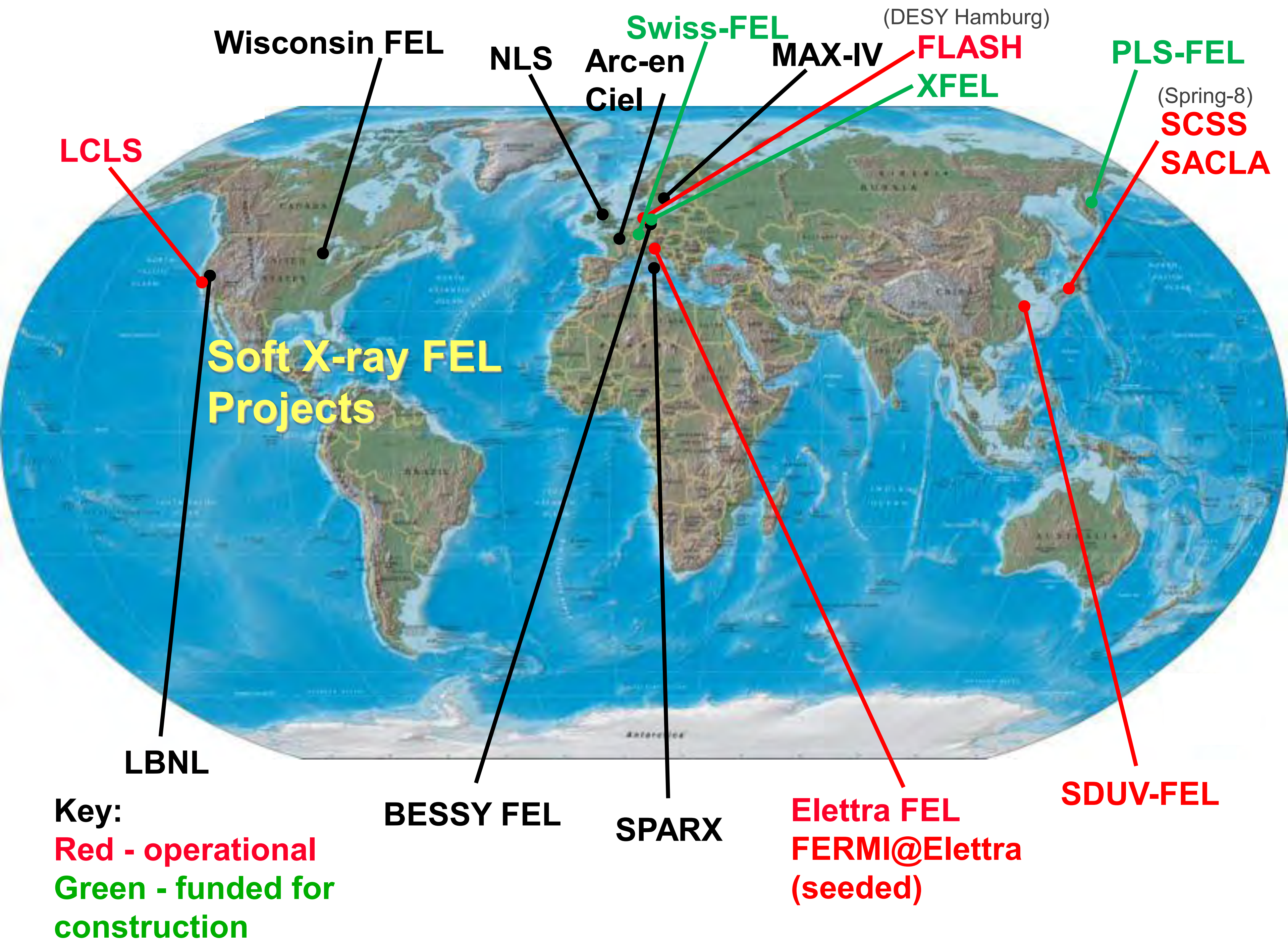


micro-bunching

↓

coherence

- Electrons are bunched under the influence of the light that they radiate
- Bunch dimensions are characteristic of the wavelength of the light (1-10Å)


Wisconsin FEL
NLS
Arc-en Ciel
Swiss-FEL
MAX-IV
(DESY Hamburg)
FLASH
XFEL
PLS-FEL
(Spring-8)
SCSS
SACLA
LCLS
Soft X-ray FEL Projects
LBNL
BESSY FEL
SPARX
Elettra FEL
FERMI@Elettra
(seeded)
SDUV-FEL
Key:
Red - operational
Green - funded for construction

The Next Generation Light Source

- High repetition rate X-ray laser with ultrafast pulses
- Significant impact on the DOE mission
  … urgent energy science: photosynthesis, combustion, catalysis…
- NGLS will probe the motion of molecules, atoms, and electrons
  On their natural time scales – femtoseconds (fs) and faster….
- With unprecedented resolution – nm (molecules) to Å (atoms)
- With chemical sensitivity – Carbon, Oxygen, Nitrogen…..
- Unique machine that will enable global leadership in critical areas
- Recent X-ray laser breakthroughs (LCLS) point the way…
  … ten year time horizon, NGLS is not an incremental advance

Worldwide, no source – operating today or under construction – will be able to provide all the scientific capabilities of NGLS

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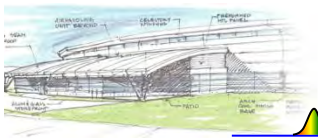
Storage Rings and X-ray Lasers

Today's storage ring x-ray sources



~ nanoseconds

nanojoule

~ picoseconds

Weak pulses at high rep rate

Today's x-ray laser sources

Europe X-FEL

~ milliseconds

millijoule

~ femtoseconds

comparable average power to SR

Intense pulses at low rep rate

Tomorrow's NGLS

~ microseconds

millijoule

~ attoseconds to femtoseconds

>100x average power

Intense pulses at high rep rate

We have engaged a broad science community over four years to develop a proposal for a Next Generation Light Source

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NGLS CD-0 Proposal

- **More than 150 contributors**
- **Representing >40 national and international research institutions**

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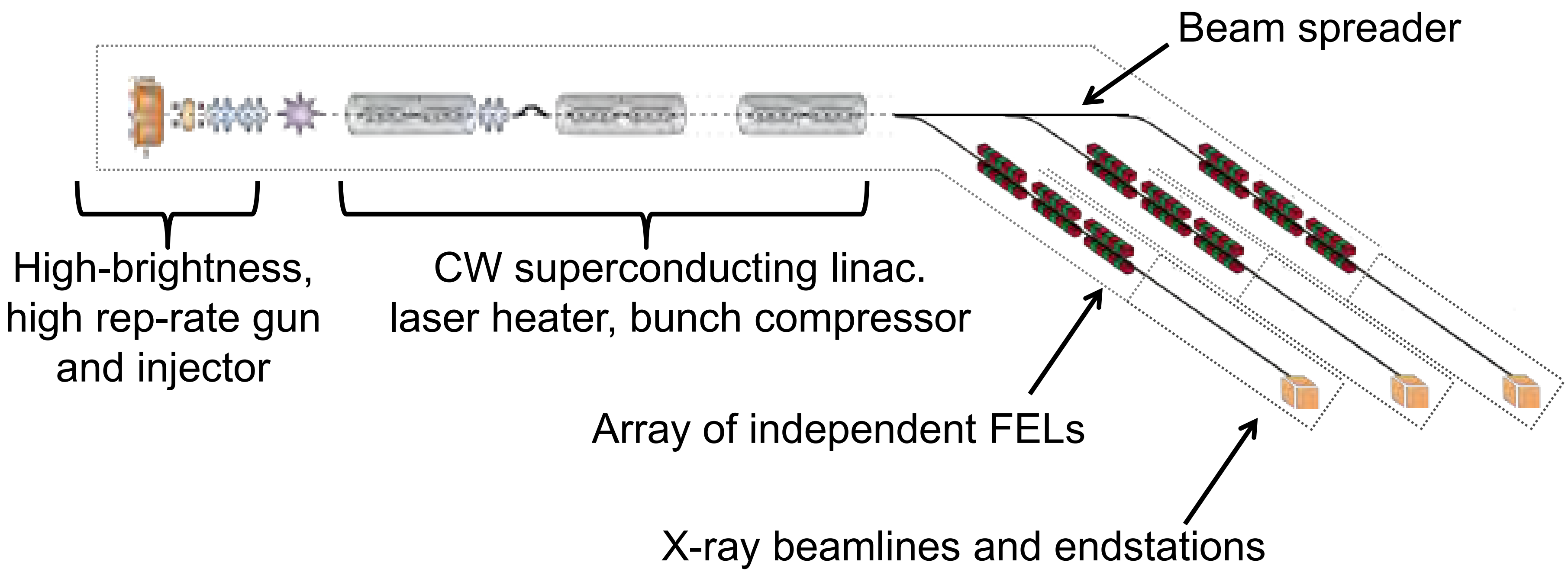
Next Generation Light Source - Unique Capabilities

- 1.8 GeV CW SC LINAC
- 3 FELs (seeded, 2 color, SASE) 280 eV – 1.2 keV



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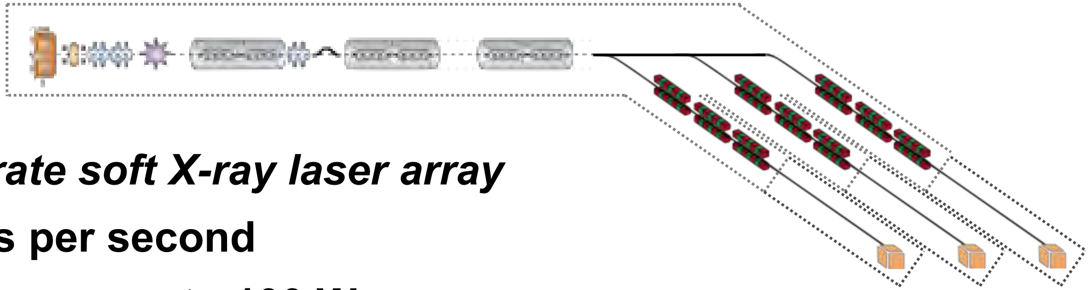

High repetition rate soft X-ray laser array

- **10^6 pulses per second**
- **average power up to 100 W**
- ***continuously* powered superconducting accelerator**

Spatially and temporally coherent X-rays (seeded)

- **ultrashort pulses from 250 as – 250 fs**
- **narrow energy bandwidth to 50 meV**

Tunable X-rays

- **adjustable photon energy from 280 eV – 1.2 keV**
  higher energies in the 3rd and 5th harmonics
- **polarization control**
- **moderate to high flux with 10^8 – 10^{12} photons/pulse**
- **Multiple pulse interrogation across the spectrum from THz to X-ray**

Three initial FEL beamlines to span the science case

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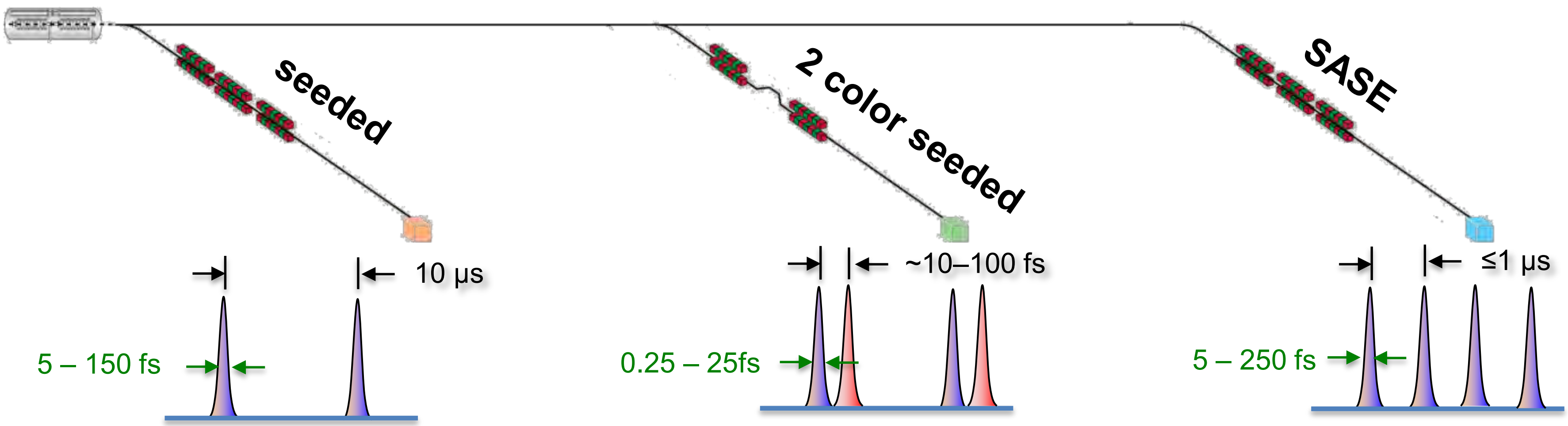

- Up to 100 kHz
- High resolution
- ~Time-bandwidth limited
- 10^{11} – 10^{12} photons/pulse
- 10^{-3} – 5×10^{-5} $\Delta\omega/\omega$

- Up to 100 kHz
- Ultra-fast
- 250 as pulses
- Two color
- 10^{8} ph/pulse

- Highest rep rate, MHz and greater
- High flux
- 10^{11} - 10^{12} photon/pulse
- 100 W

NGLS offers significant advances over current capabilities:

- More photons per unit bandwidth
- More photons per second
- Shorter pulses
  - With controlled trade-off between time and energy resolution

Broad Range of Energy Science Uniquely Enabled by NGLS

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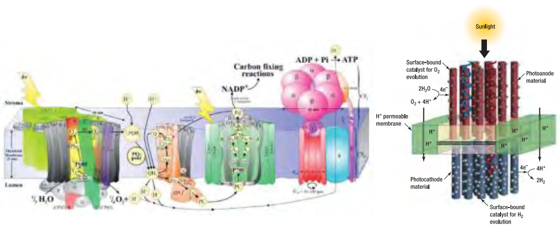
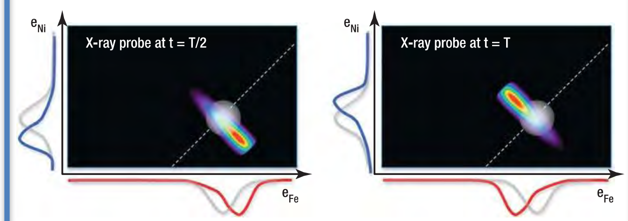
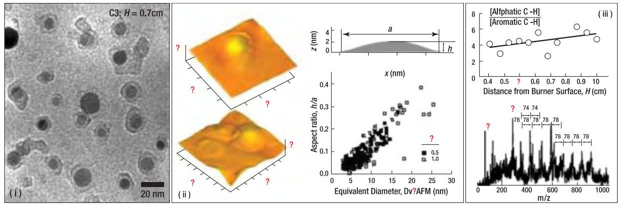
Natural and Artificial Photosynthesis



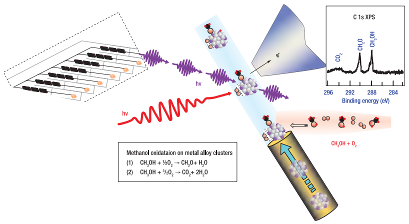
Fundamental Charge Dynamics



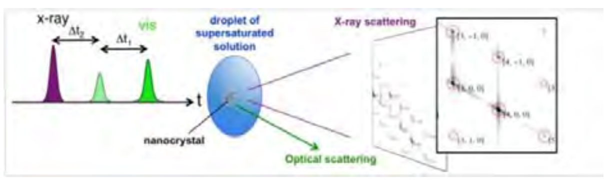
Advanced Combustion Science



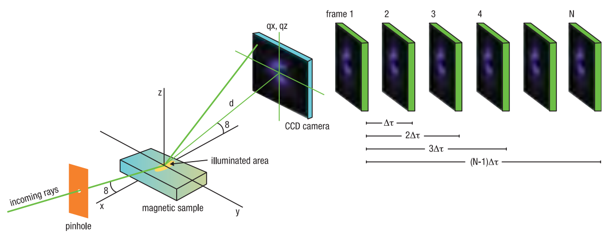
Catalysis



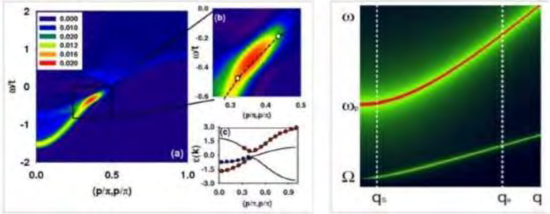
Nanoscale Materials Nucleation



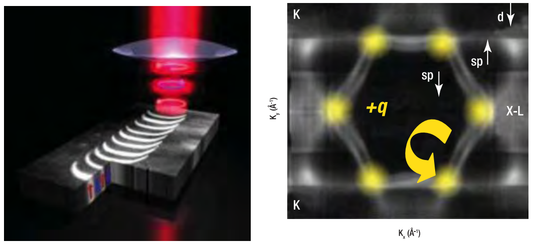
Dynamic Nanoscale Heterogeneity



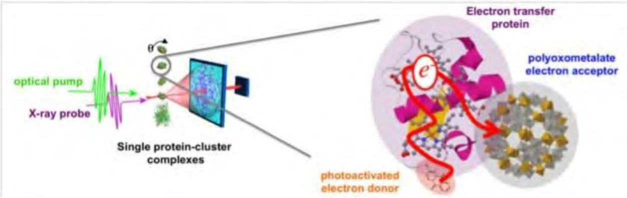
Quantum Materials



Nanoscale Spin and Magnetization



Bioimaging: Structure-to-Function



NGLS – Correlated Materials – Photoemission

Limitations of current approaches

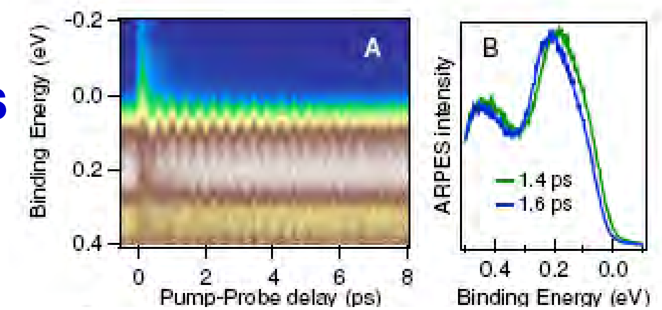
(1) ARPES
– limited dynamics (laser/HHG)

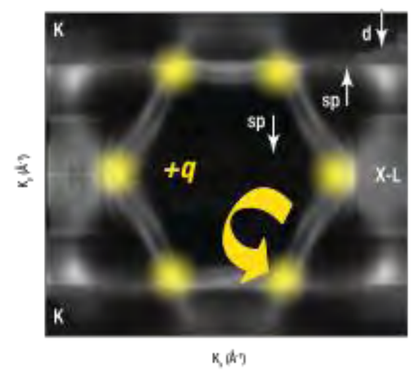
(2) Spin sensitivity
– limited by available flux

(3) Single particle spectral fctn. A(k,ω)
– ***not charge correlations***

NGLS:
- **High rep-rate**
- **Moderate flux/pulse**
- **High Average Brightness**
- **Time (atto), energy res.**
- **Tunability**
- **Polarization Control**

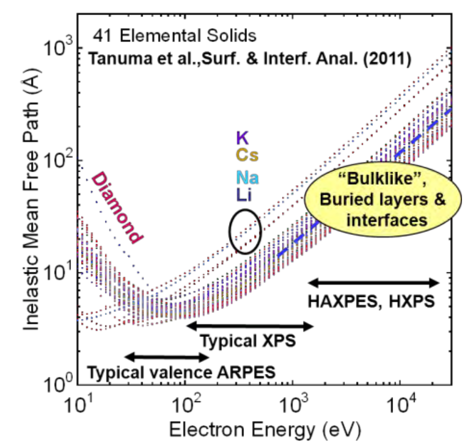
Time-resolved ARPES
Non-linear ARPES





Spin-resolved ARPES

Hard X-ray ARPES



Dynamic Probe of Electronic Structure: Time-resolved ARPES

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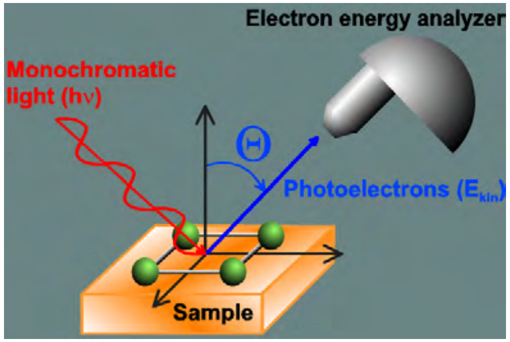
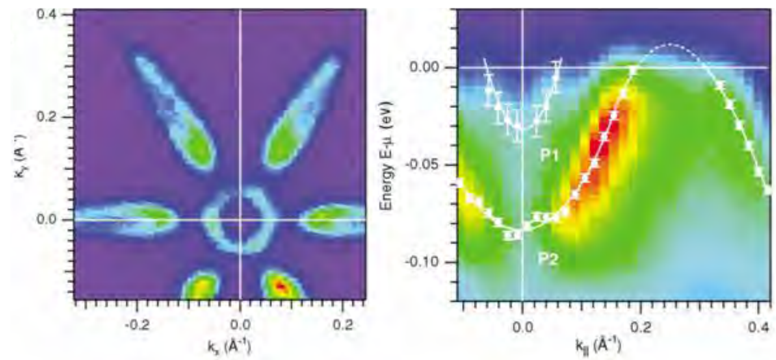
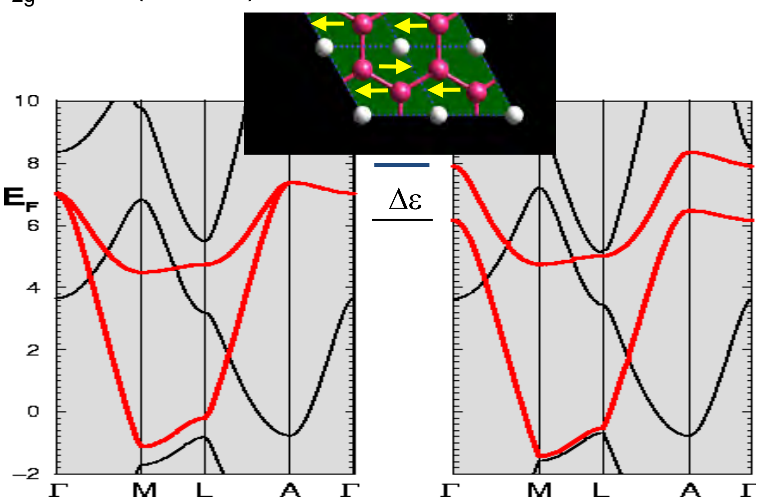


- Time evolution of single-particle spectral density function $A(k,\omega)$
- Dynamic band structure – response to tailored excitation
- Time-resolved – separation of correlated phenomena in time



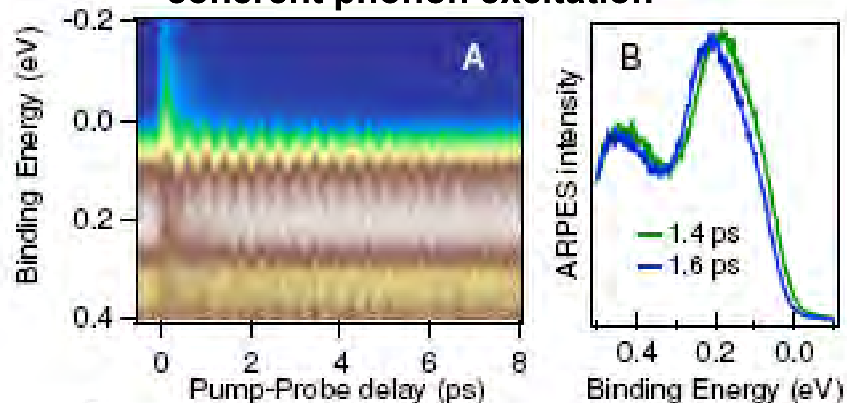
Bismuth occupied electronic density of states

MgB_2
Modulation of Electronic Structure
E_{2g} Mode (17 THz)



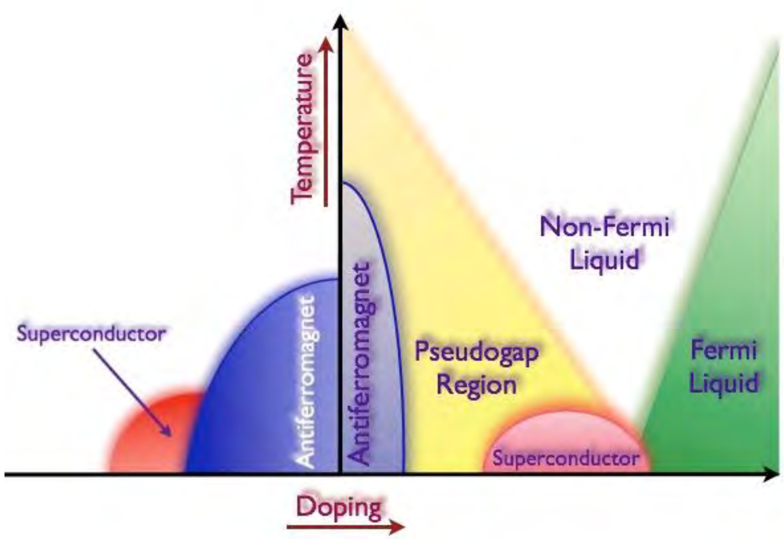
T. Yildrim et al., *PRL* (2001)

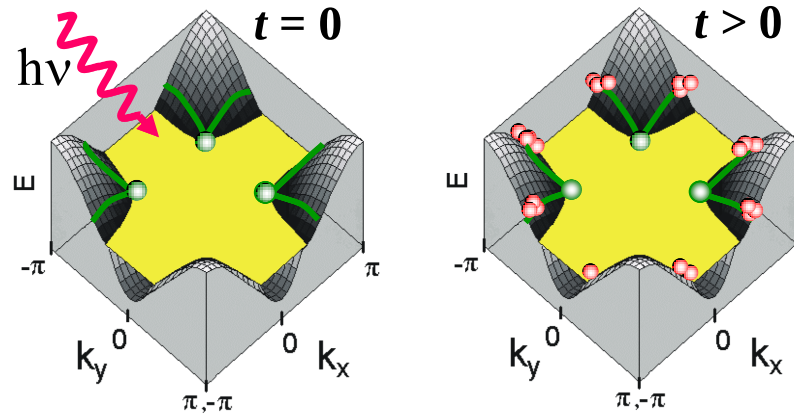
1T-TaS_2 - Modulation of Hubbard Gap coherent phonon excitation



Perfetti, et al. *PRL*, **97**, 067402 (2006)

High-T_c Superconductors





momentum-resolved
quasiparticle dynamics

Controlled perturbation of superconducting state (near-equilibrium)

- Identify specific modes associated with superconducting state
- Observe re-establishment of SC from near-by states
  (e.g. transient pseudogap)
- Resolve this process with *time*, *energy*, *spin*, and *momentum* resolution

Time and spin-resolved ARPES is a powerful now tool to understand a wide class of complex materials: topological insulators, CMR compounds, multiferroics, etc.

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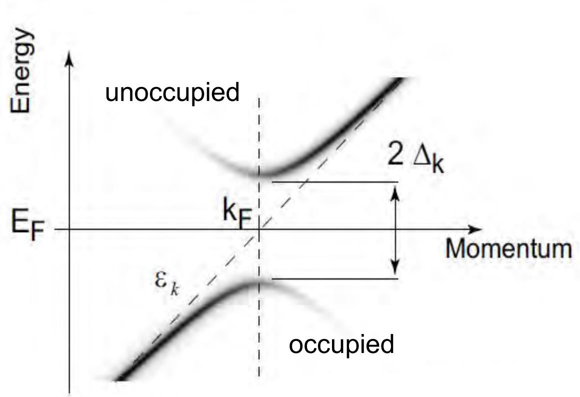
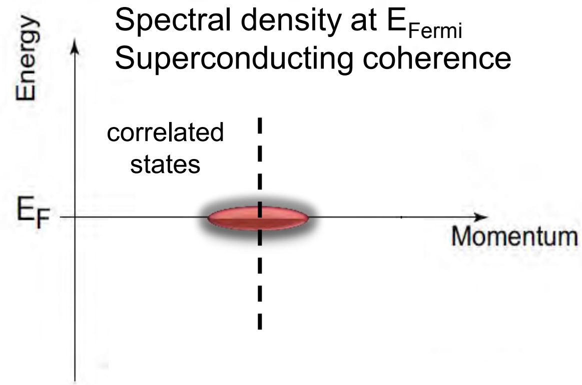
NGLS: High-T_c Superconductors (J. Orenstein)

Nonlinear ARPES – 2 photons in, 2 electrons out,
probes 2-particle ground state properties



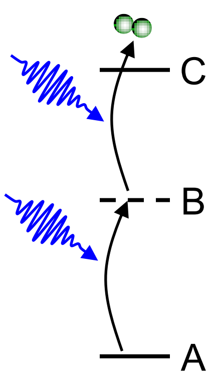
Spectral density – linear ARPES

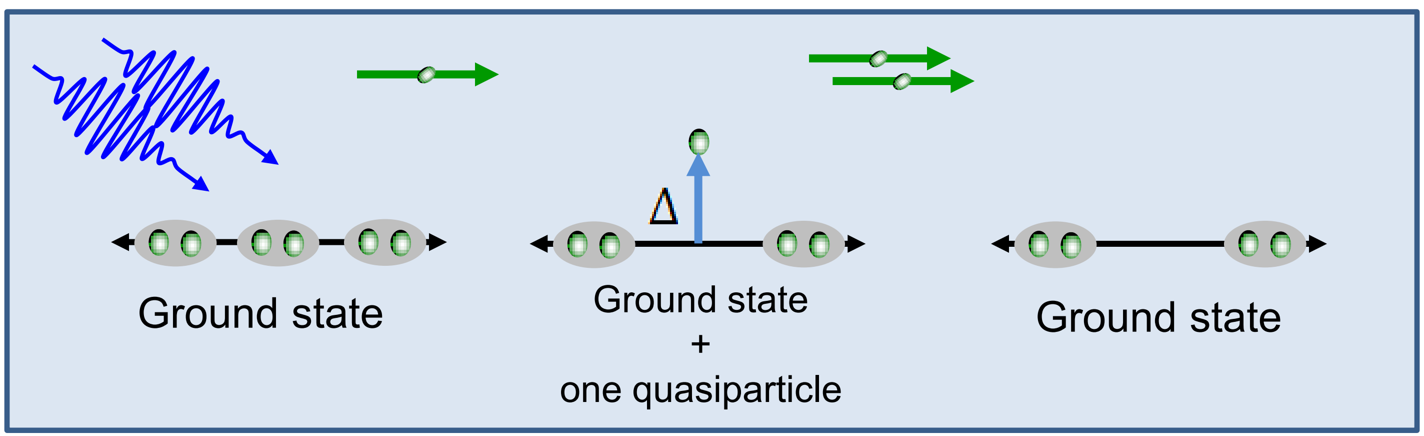


Spectral density – nonlinear ARPES

Current ARPES:
Single particle probe
– not charge correlations

Non-linear ARPES:
2 photons within
coherence length (~100 nm)
coherence time (~1 ps)
of paired electrons





Initial (A) **Intermediate (B)** **Final (C)**

NGLS and Properties of Topological Insulators

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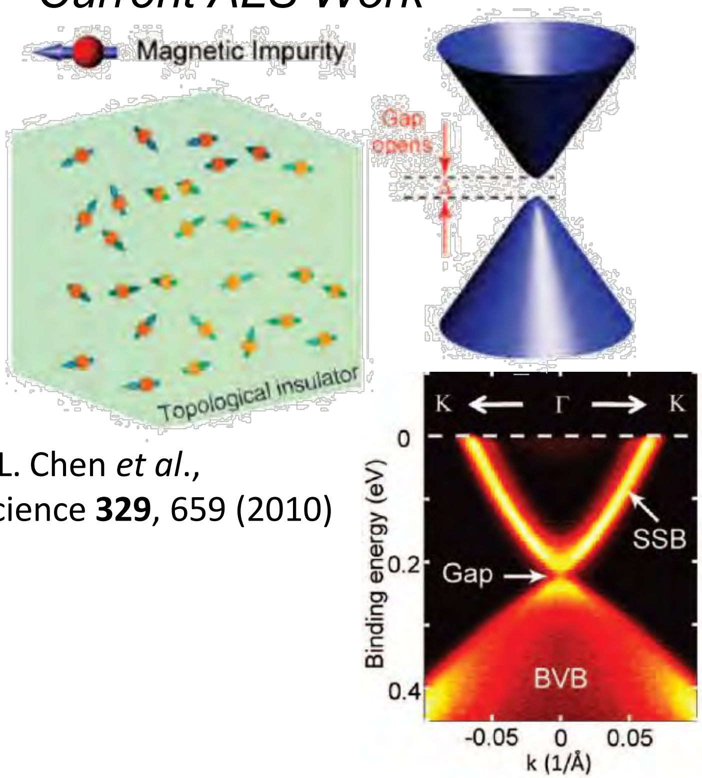
Protected metallic surface states – spin/orbit coupling, time reversal symmetry

Current ALS Work



Y.L. Chen *et al.*, Science **329**, 659 (2010)

ARPES (static) measures:

- formation of a gap in surface state
- due to broken **Time Reversal Symmetry**
- from chemical doping of magnetic impurities

Physical relationships between topological states with and without TRS are unknown:

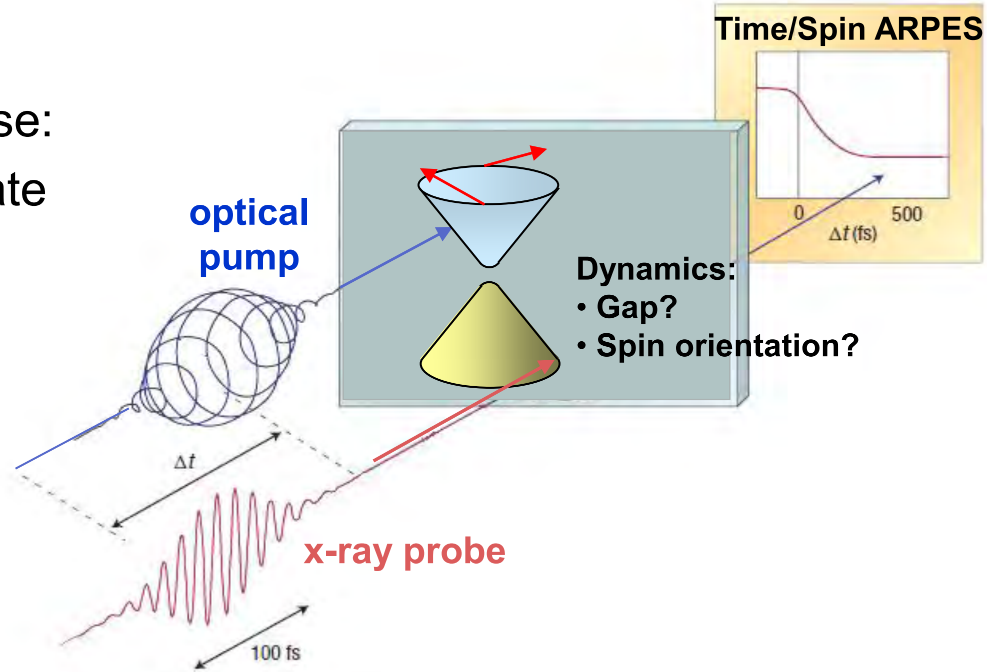
- What is precise effect of **TRS** on spin/electronic structure?
- Dynamics: How does the system recover once **TRS** returns?

- Time, spin, and momentum sensitive spectroscopy required to understand functional control of exotic properties
- Fsec to psec – Dynamics between nearby states of different fundamental symmetry

NGLS and Properties of Topological Insulators

High-intensity circular optical pump-pulse:

- breaks **TRS** via transient magnetic state
- w/o introduction of impurity scattering
- w/o altered chemical potential



Time, spin, and angle-resolved photoemission:

- *directly* measures the **spin-dependent electronic structure**
- follows evolution between states of different symmetry.

- Requires high repetition rate and high average power

- Requires energy and polarization tunability

Time-resolved ARPES at KeV Energies

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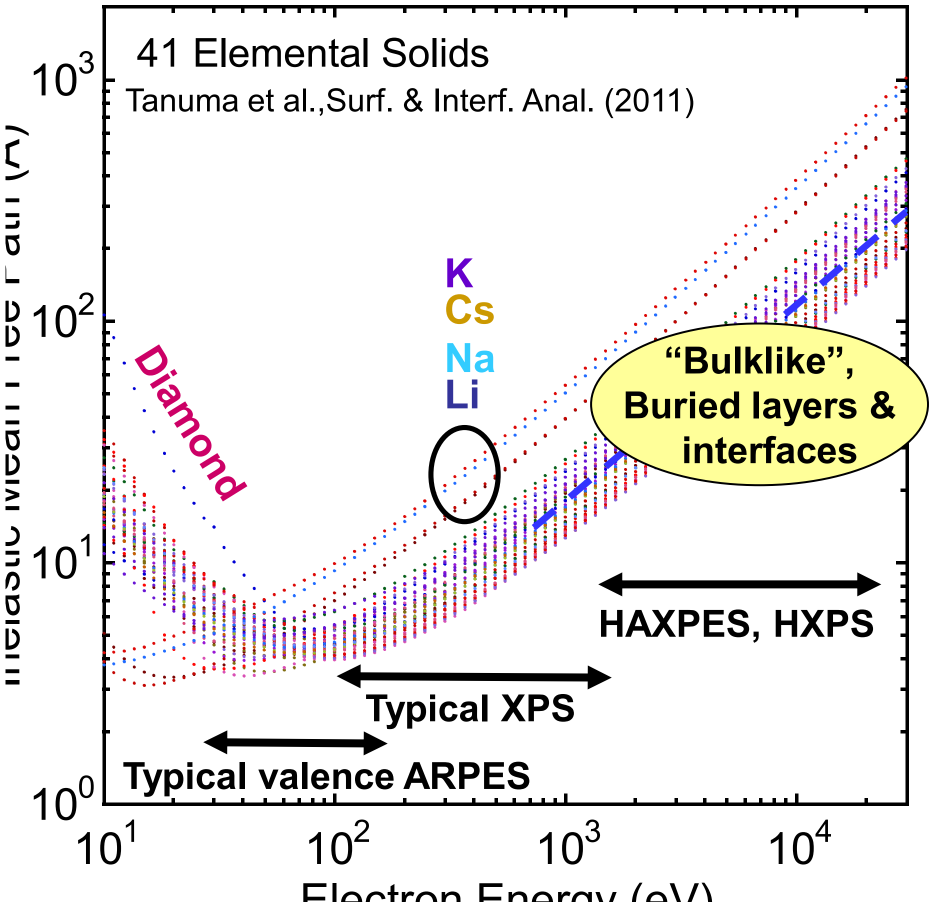
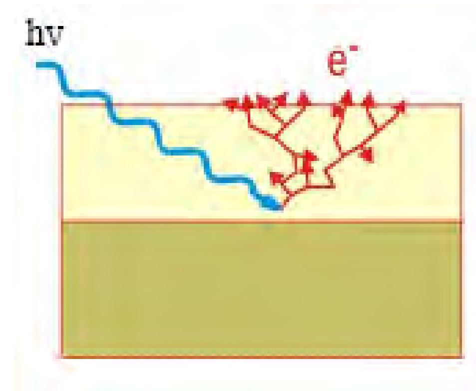
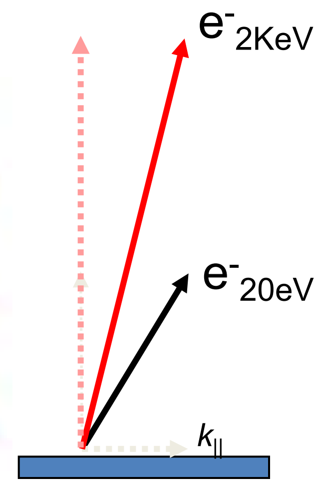
Electrons interact strongly
Surface Sensitivity – 5-20Å







- **KeV energy (10-100 meV res.)**
  - bulk penetration (VUV sources)
- **valence, core PES**
- **Space charge distortion of EDC**
  low flux/pulse ⇔ high rep. rate
- **Electron dynamics**
  rapid recovery ⇔ high rep. rate

See talk by Alex Gray (Fadley Group)

Probing Electron Correlation in Solids

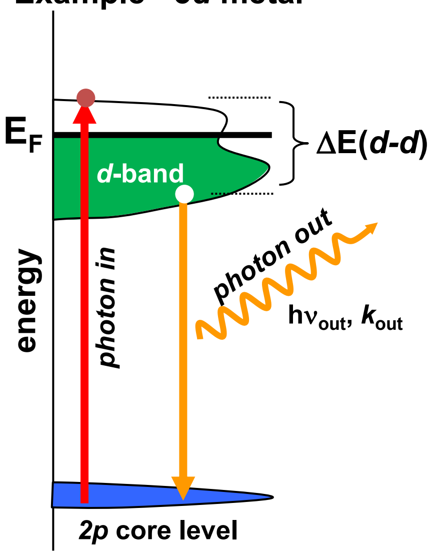
Inelastic X-ray Scattering

Electronic Structure:

Photoemission (ARPES)	**A(*k*,ω) - single-particle spec.**
Inelastic Neutron Scattering (INS)	**S(*q*, ω) - spin fluctuation spec.**
Inelastic x-ray scattering (IXS)	**S(*q*, ω) - density-density *correlation***

(X-ray Raman, q>0)

Example - 3*d* metal



Energy conservation:

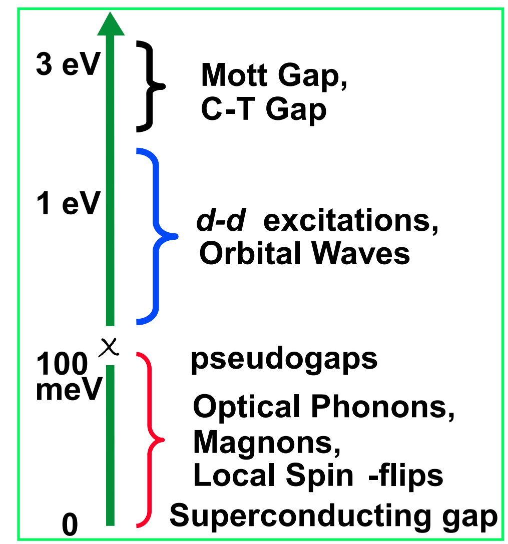
$\omega_{out} = \omega_{in} - \Delta E(d\text{-}d)$

Momentum conservation:

$k_{out} = k_{in} - \Delta k(d\text{-}d)$

Advantages:

- **Bulk sensitive**
- **Insulating samples (organics, bio-materials ….)**
- **External fields (magnetic, electric) pressure, optical excitation**
- **Probe optically 'forbidden' transitions**
- **Resonant IXS**
  - **signal enhancement 10^2-10^3**
  - **element sensitivity (buried interfaces)**
  - **soft x-ray 2*p*→3*d* (spin, orbital ordering …)**



Highly Optimized Photon Source – Scientific Impact

- High Average Flux – photons/sec/bandwidth

- High Resolution - throughput

Time-resolved RIXS: development of correlation S(q,ω) in response to tailored excitation

Probing Electron Correlation in Solids

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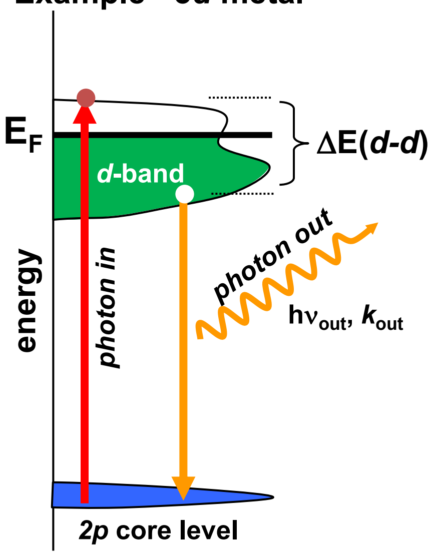

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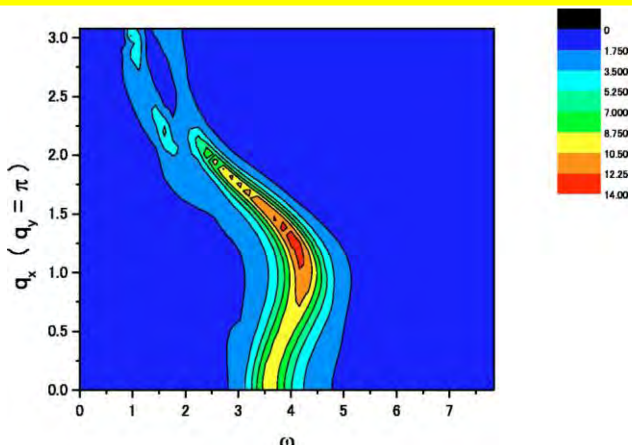
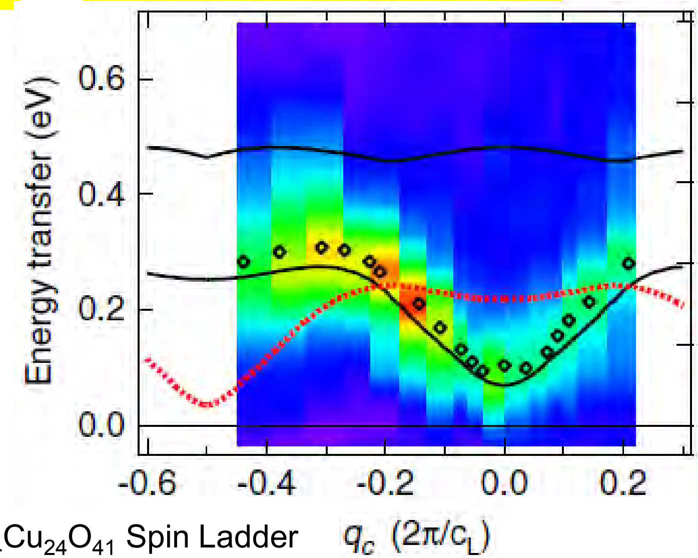


Spec. Function - ϕ Gauge Collective Mode
Doped Mott Insulator

Lee et al. PRB, 2003



$Sr_{14}Cu_{24}O_{41}$ Spin Ladder
Schlappa et al., PRL (2009)

Highly Optimized Photon Source – Scientific Impact

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Time-resolved RIXS: development of correlation S(q,ω) in response to tailored excitation

From Spontaneous Raman (RIXS)….
to Pump-Probe to Multidimensional Spectroscopy

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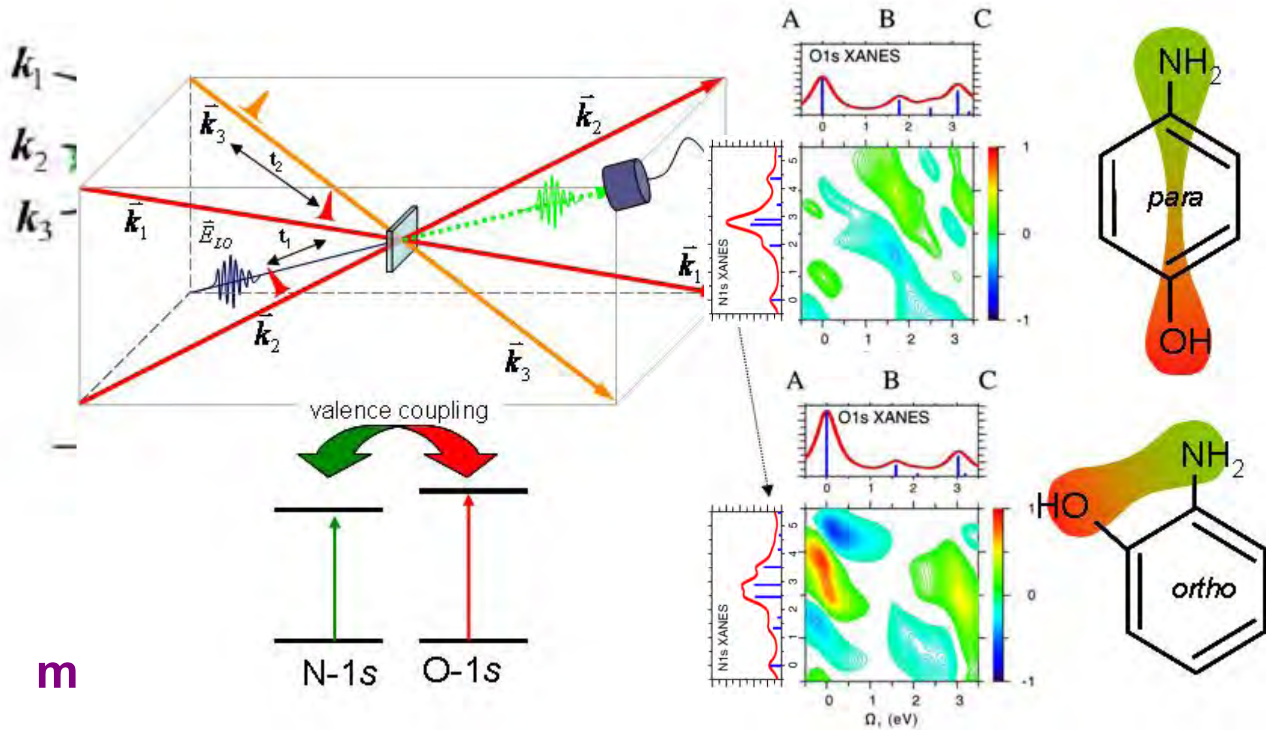
multidimensional spectroscopy has revolutionized fields of science



Mukamel et al, 2007

1991 Nobel Prize multidimensional NMR

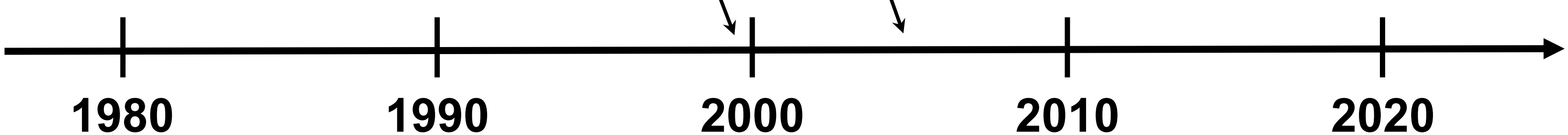
m

visible

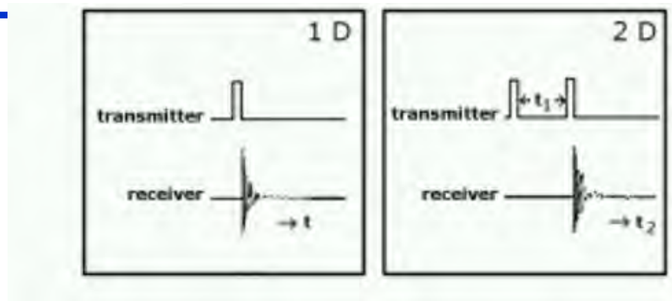
IR

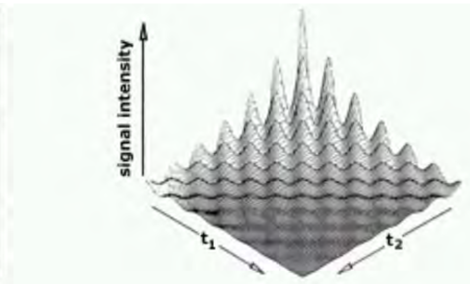
NMR

X-ray?



Multidimensional Spectroscopy NMR – Optical – X-rays





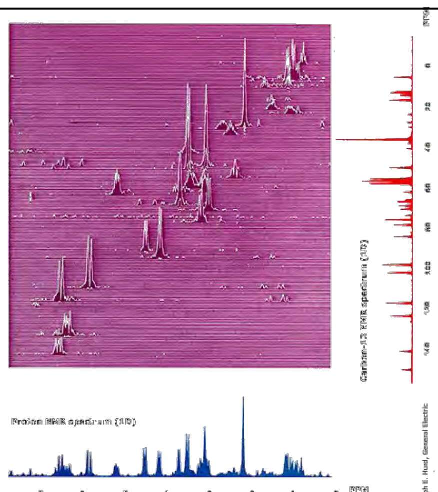
NMR (nuclear magnetic resonance)

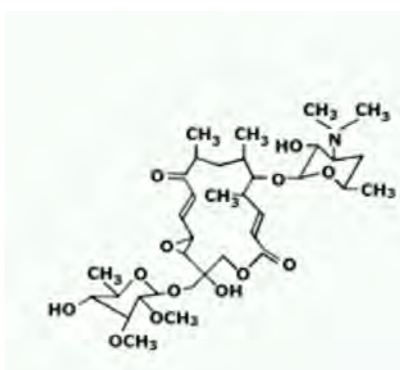
- RF pulse sequences couple to nuclear spins
- Measure ***nuclear resonances***, and correlations
- Fingerprint map of ***molecular structures*** and their spatial relationships

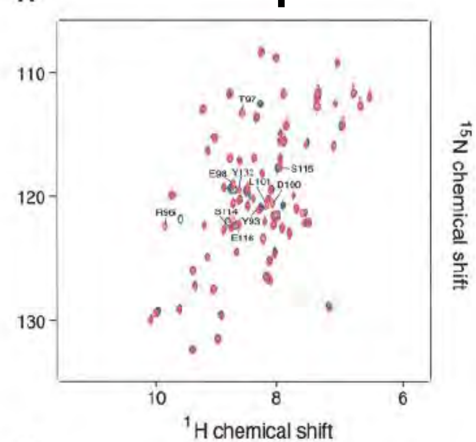
Optical/X-ray Multidimensional Spectroscopy

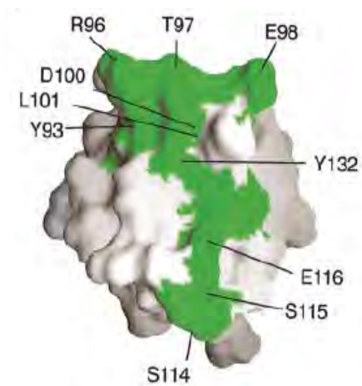
- Optical/X-ray pulses sequences couple to ***valence*** electronic states
- Measure ***electronic resonances***, and correlations
- Fingerprint map of ***valence electronic structure and correlation***
  - *- X-rays – element specific (correlation - different atomic sites)*
  - *- Time-resolution – attoseconds (evolution of electronic structure)*
  - *- Momentum resolution – probe entire Brillioun zone*

Multidimensional NMR Examples:





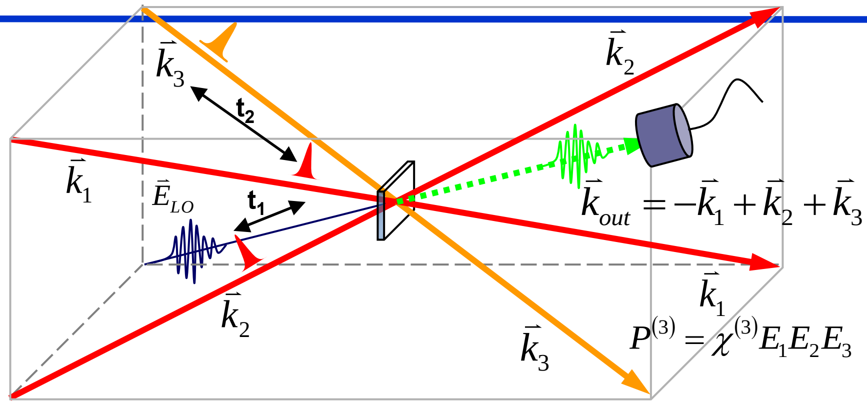




a next generation light source

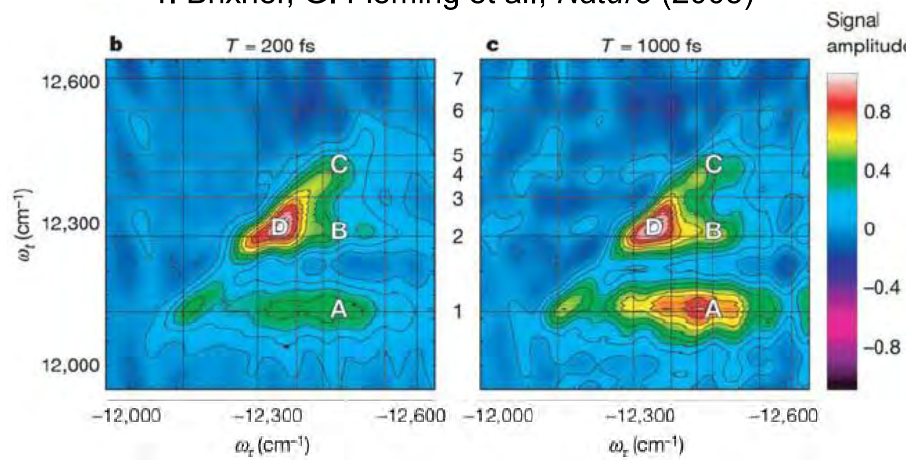
X-ray FELs

Multidimensional Spectroscopy in the Visible Regime



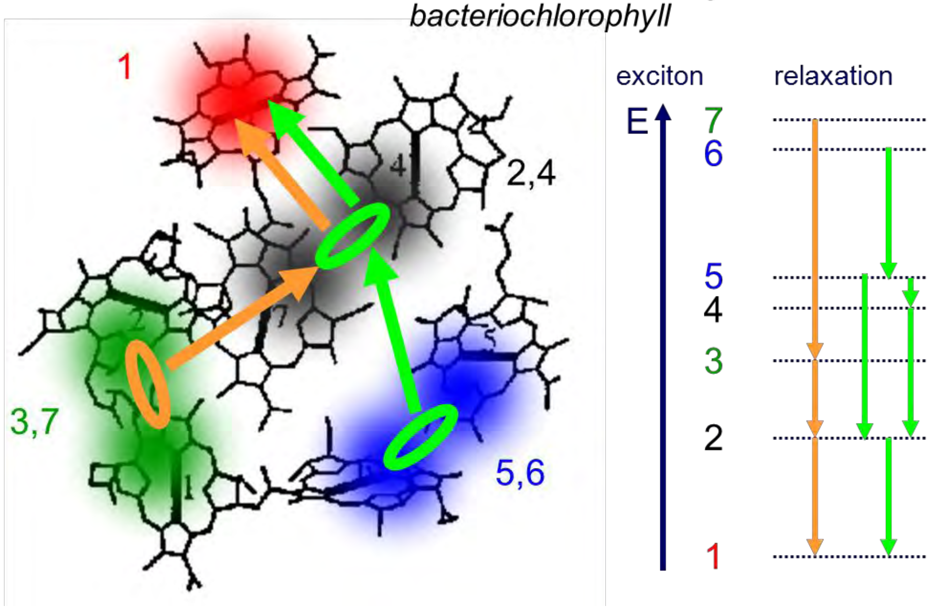
Mapping valence charge flow Photosynthesis

T. Brixner, G. Fleming et al., *Nature* (2005)



FMO: Exciton relaxation dynamics

bacteriochlorophyll



Disentangle coupled quantum systems

Follow the flow of valence charges

a next generation light source

X-ray FELs
Multidimensional Spectroscopy in the X-ray Regime

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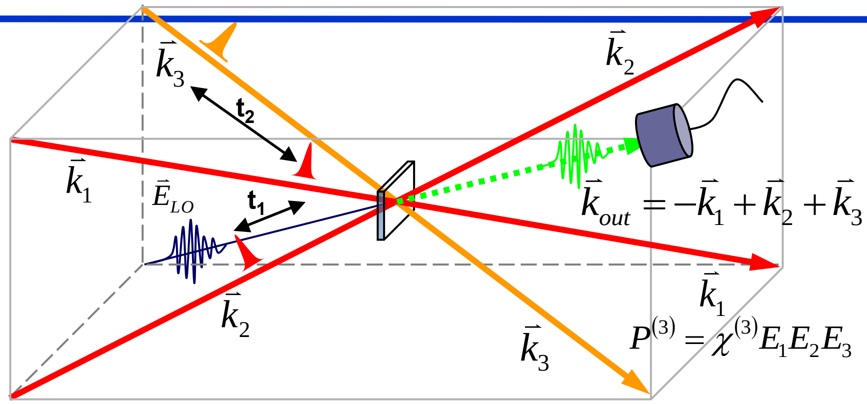

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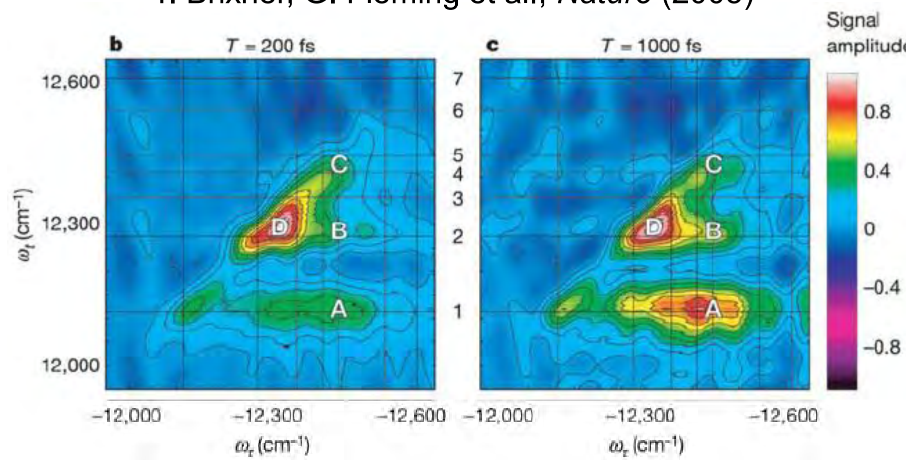
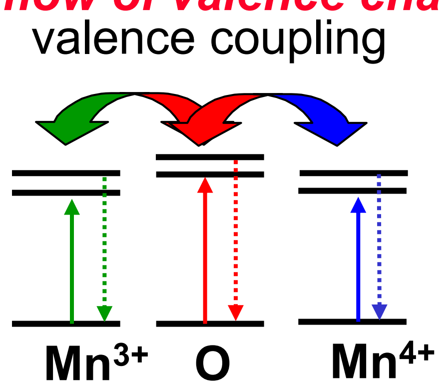
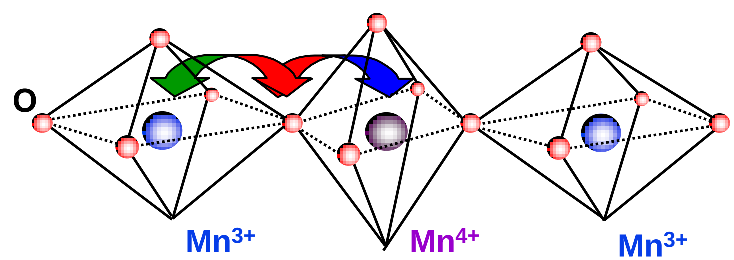


Disentangle coupled quantum systems

Follow the flow of valence charges





X-Rays – element specificity, attosecond resolution, momentum - Brillioun zone

Broad Range of Energy Science Uniquely Enabled by NGLS

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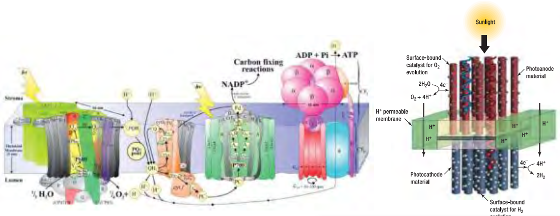
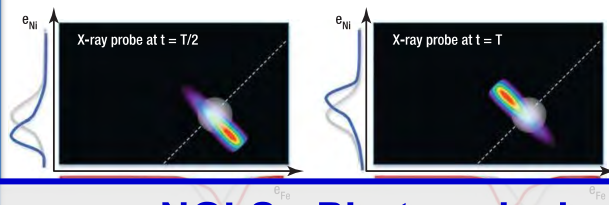
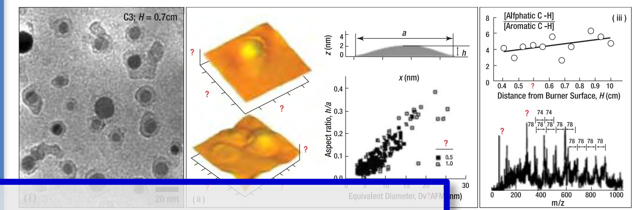
Natural and Artificial Photosynthesis



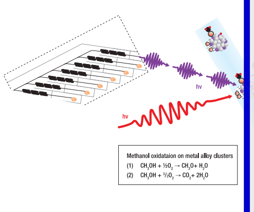
Fundamental Charge Dynamics



Advanced Combustion Science

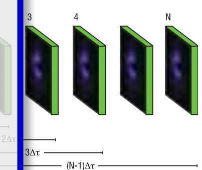


Catalysis



Nanoscale Materials Nucleation

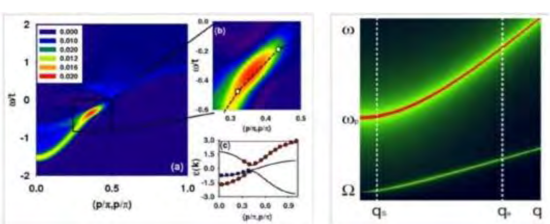
Dynamic Nanoscale Heterogeneity



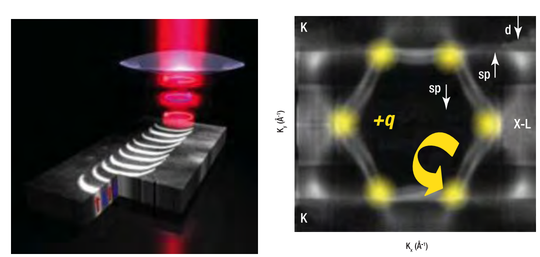
NGLS - Photoemission

Quantum Materials	**nonlinear PES, Time/Spin ARPES, HXPES**
Nano Spin/Magnetization	**Time/Spin ARPES, HXPES**
Nano-plasmonics	**Time/Spin ARPES, HXPES**
Fundamental Charge Dynamics	**COLTRIMS (gas-phase)**
Heterogeneous Catalysis	**Time-resolved Ambient Pressure PES**

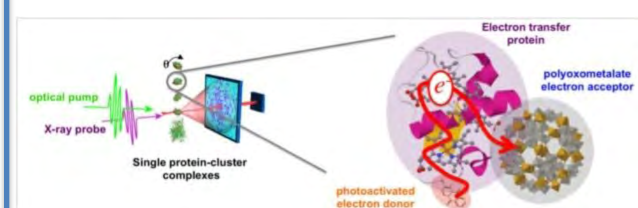
Quantum Materials



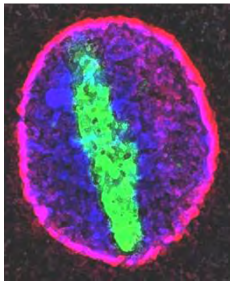
Nanoscale Spin and Magnetization



Bioimaging: Structure-to-Function

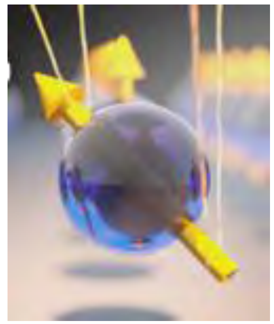


NGLS Broad Science Impact

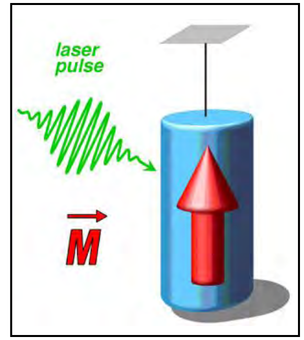


cellular imaging

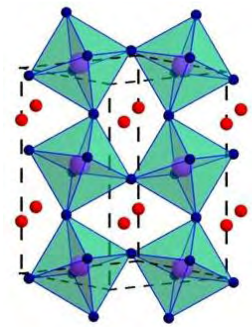
topological insulators



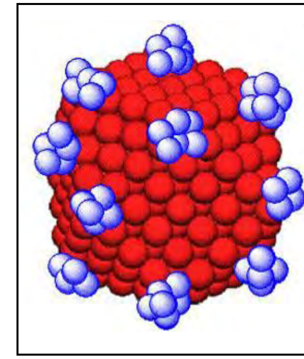
spin dynamics



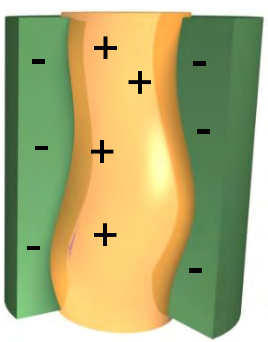
magnetization dynamics



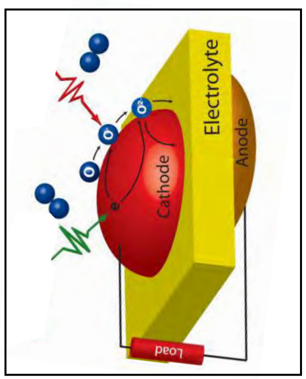
complex materials



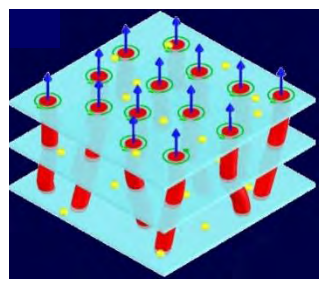
nano-synthesis and catalysis



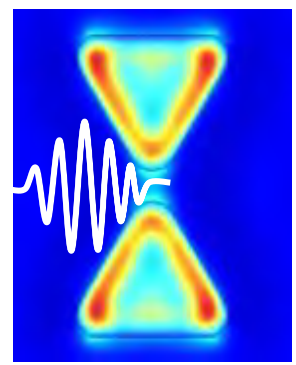
PV charge migration



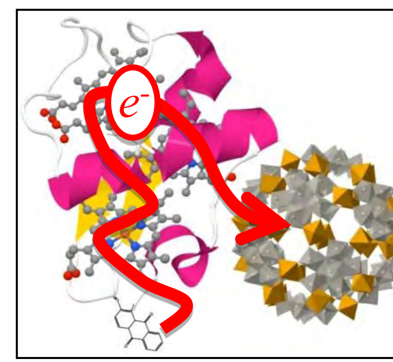
fuel cells



HTSC flux pinning



attosecond plasmonics



bio-geo electrochemistry

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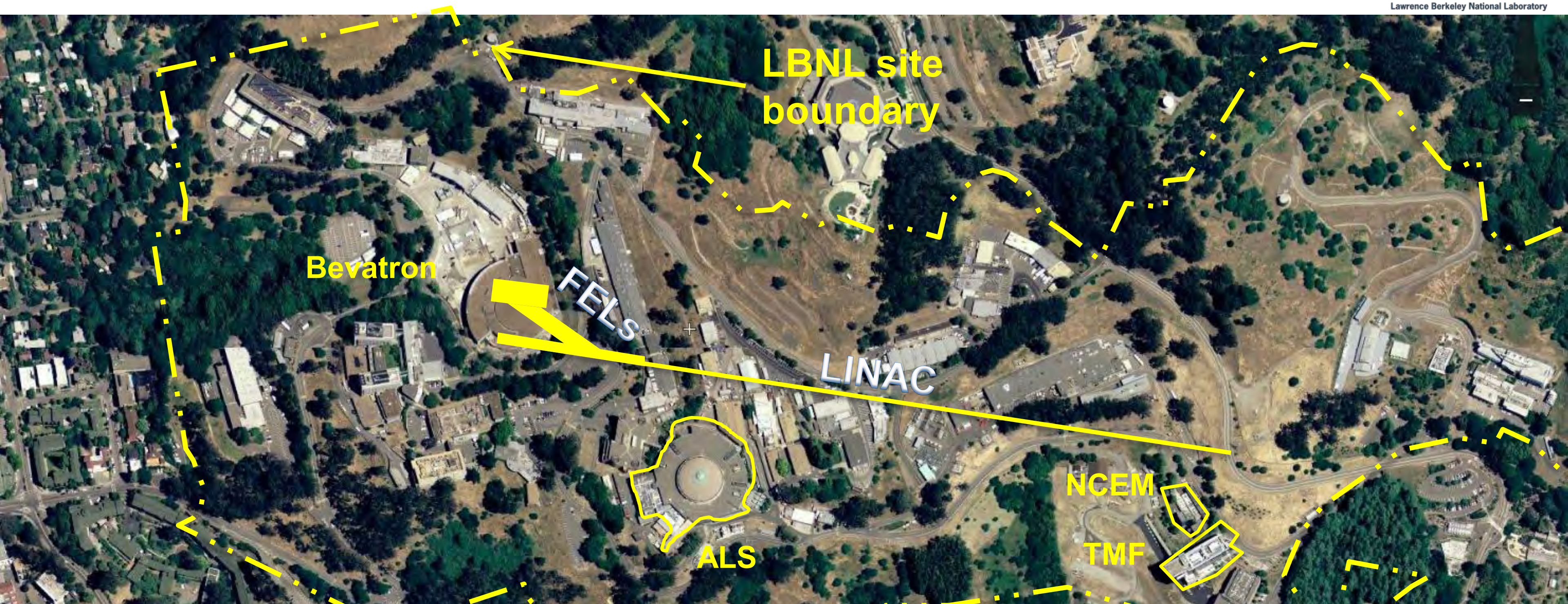
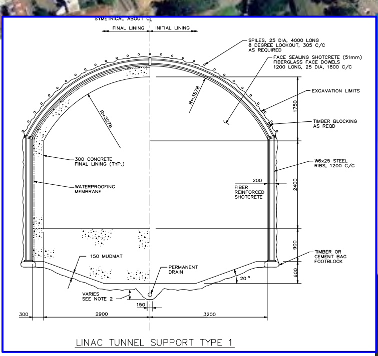
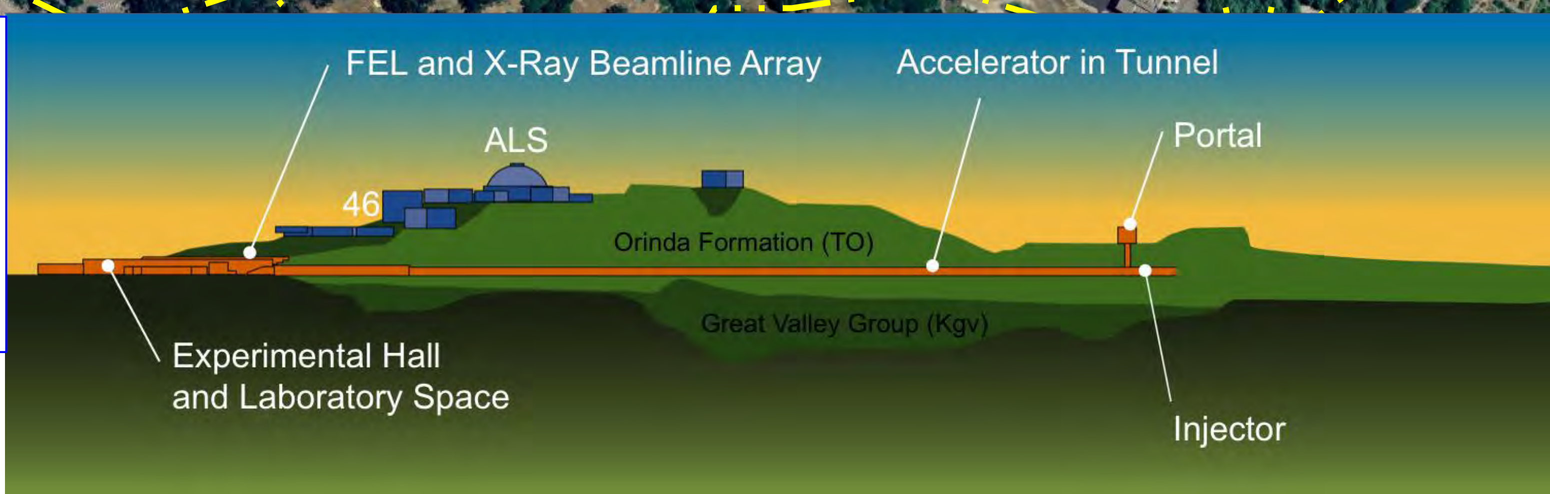
NGLS at the LBNL site

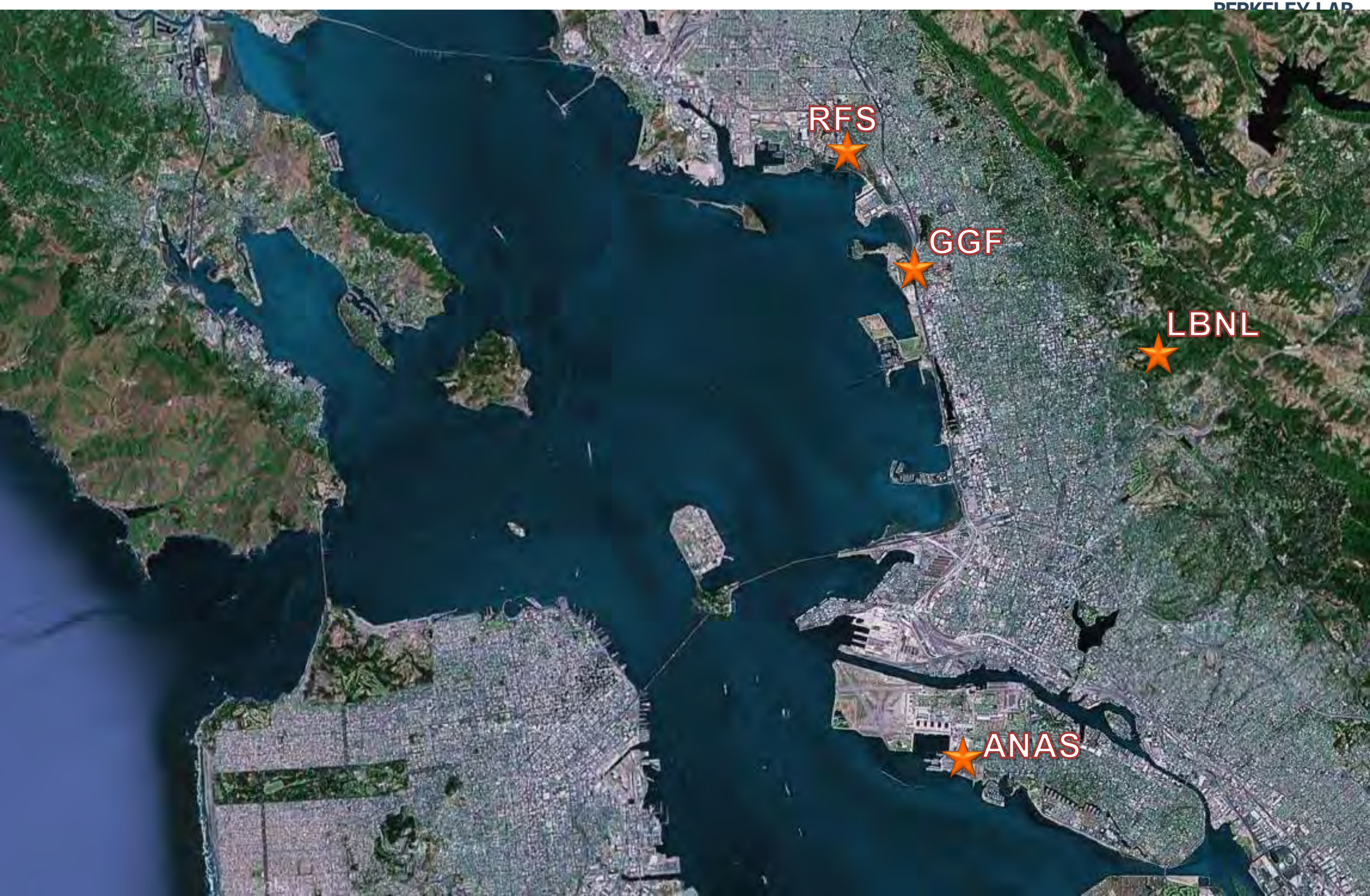




Mined tunnel



Alternate Sites



Next Generation Light Source - Unique Capabilities

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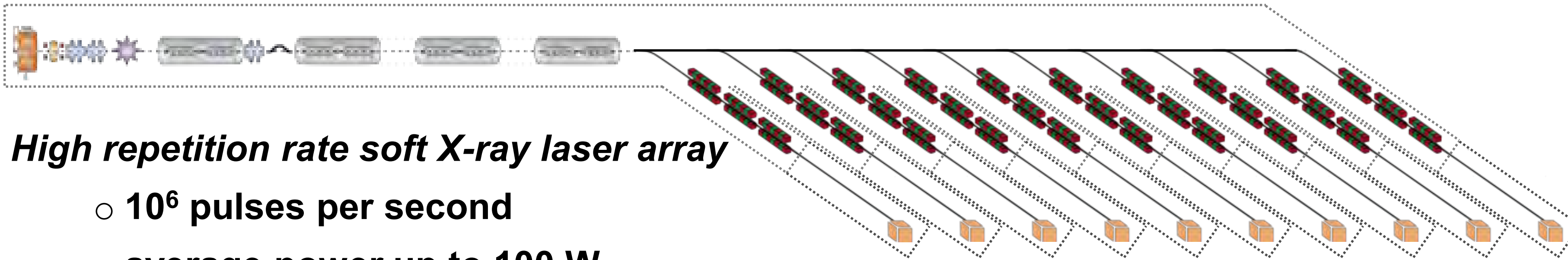
- 2.4 GeV CW SC LINAC
- 10 FELs ⇒ harder x-rays, optimized for specific science



High repetition rate soft X-ray laser array

- **10^6 pulses per second**
- **average power up to 100 W**
- ***continuously* powered superconducting accelerator**

Spatially and temporally coherent X-rays (seeded)

- **ultrashort pulses from 250 as – 250 fs**
- **narrow energy bandwidth to 50 meV**

Tunable X-rays

- **adjustable photon energy from 280 eV – 1.2 keV**
  higher energies in the 3rd and 5th harmonics
- **polarization control**
- **moderate to high flux with 10^8 – 10^{12} photons/pulse**
- **Multiple pulse interrogation across the spectrum from THz to X-ray**