SIMES

STANFORD INSTITUTE FOR MATERIALS & ENERGY SCIENCE

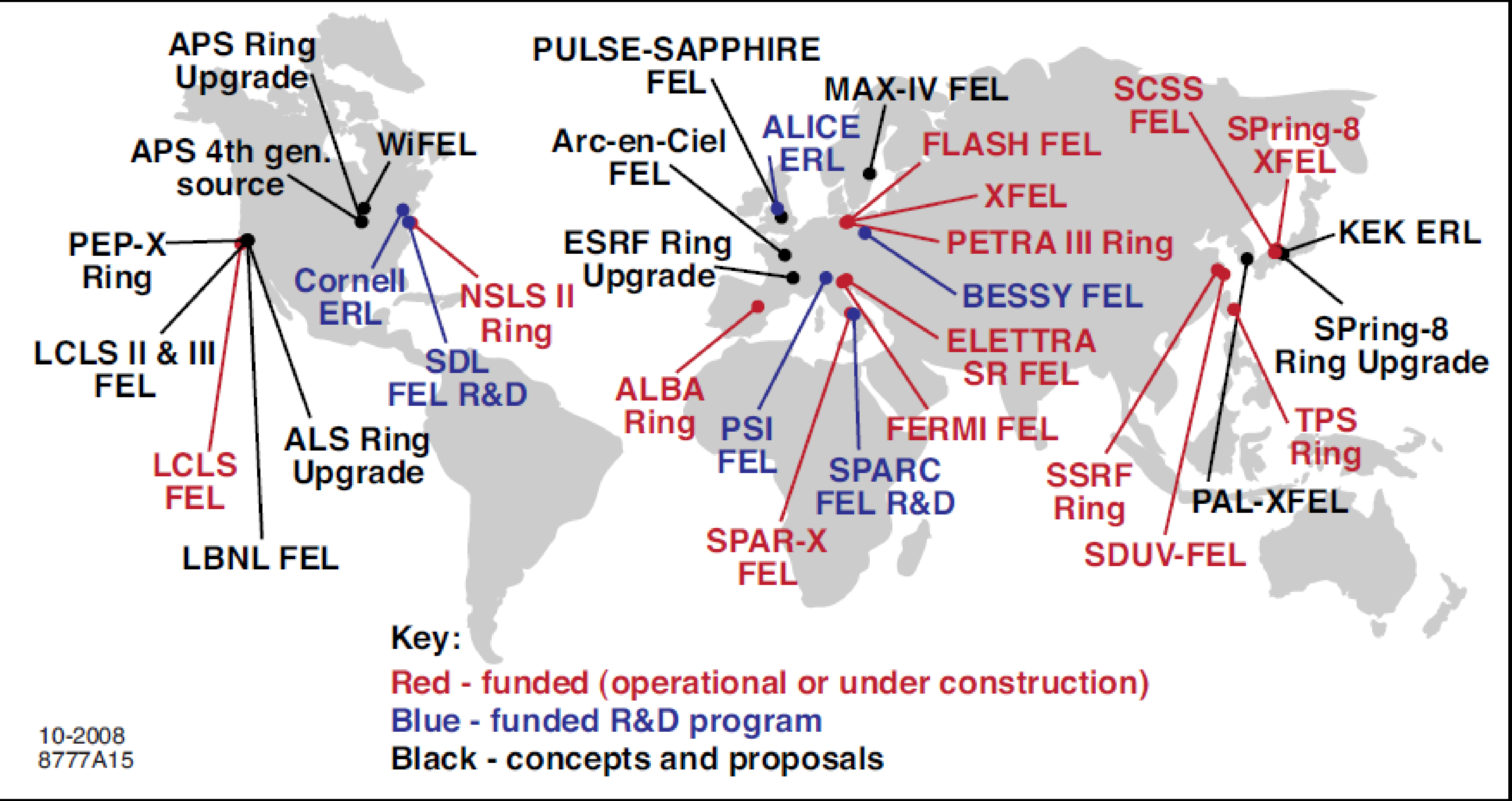
T. P. Devereaux

Spectroscopy and Light-Matter Interactions

Collaborators – B. Moritz, Z.-X. Shen (SLAC/Stanford), J. K. Freericks (Georgetown), Z. Hussain (LBNL), R. T. Scalettar (UC-Davis).

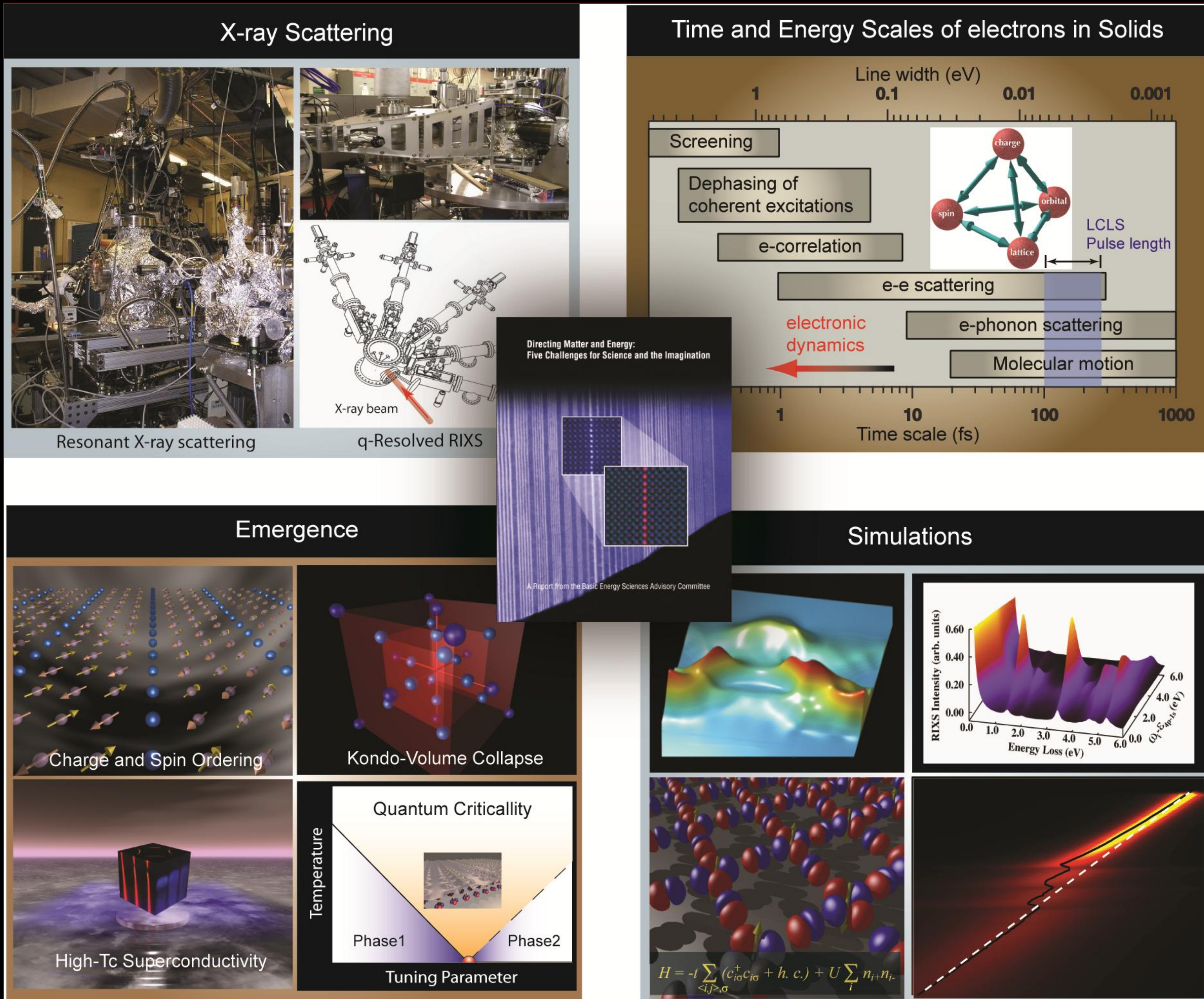
Machines used: Bassi, Jacquard, Franklin, Hopper.

New & future light sources

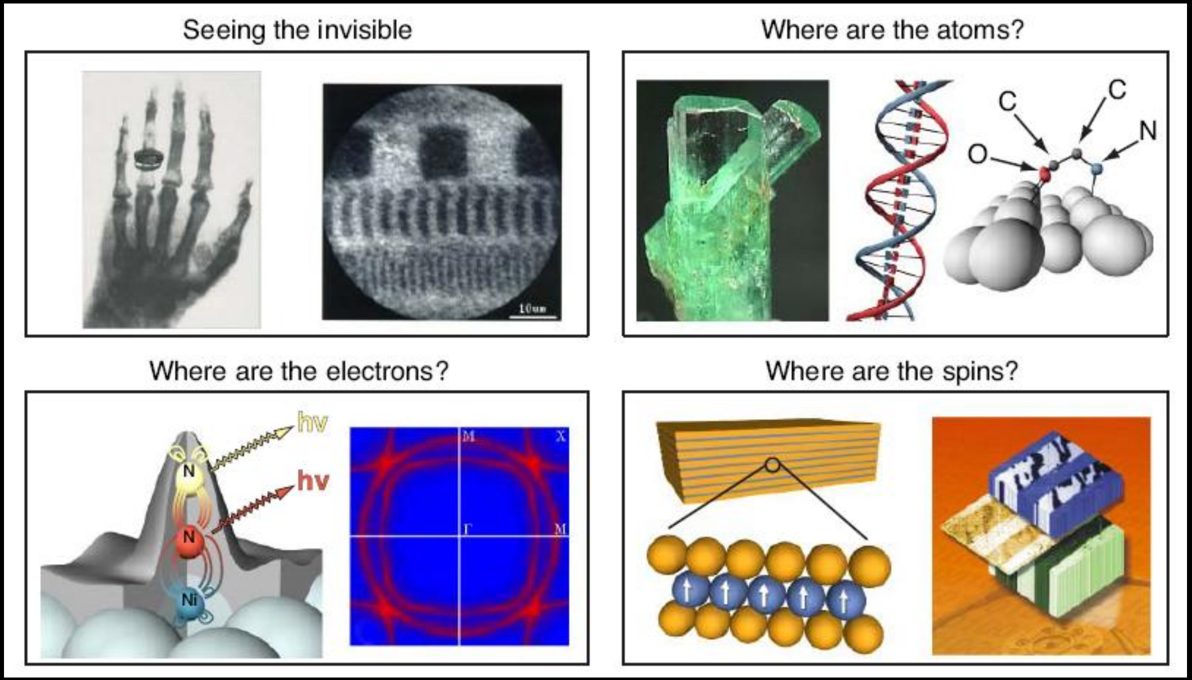


A field in expansion

Using x-rays to study materials science

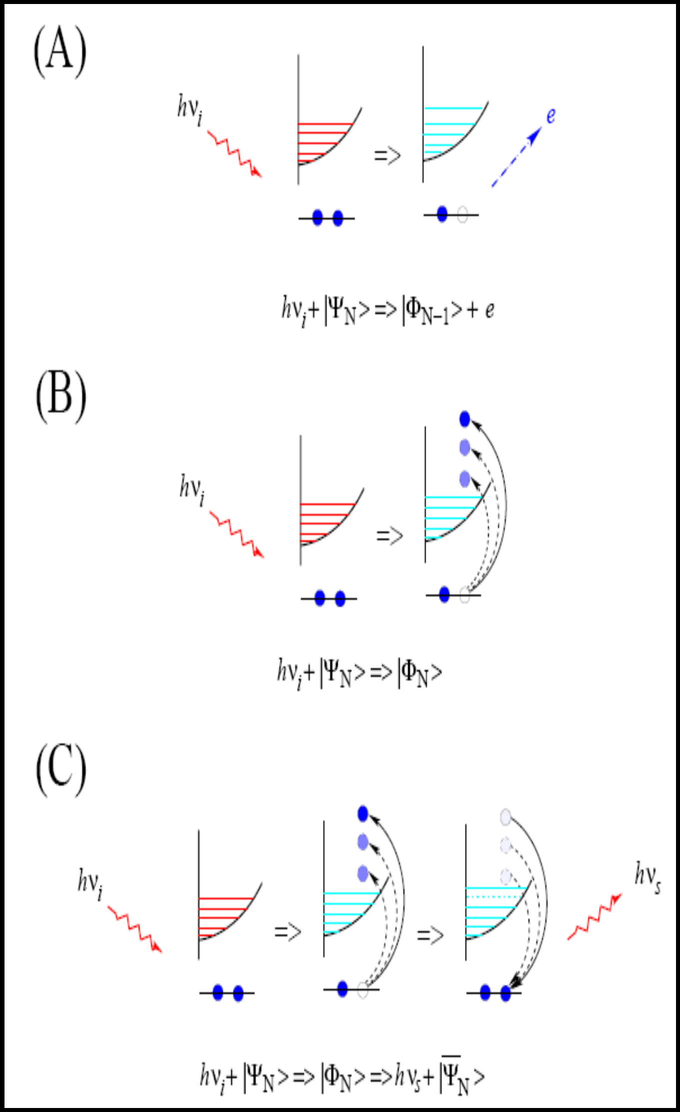


X-rays capabilities



- Driving chemical transformations by controlled optical or infrared pulses and understanding the atomic and electronic transformation with x-rays.
  - to capture, with snapshots on the femtosecond timescale, the making and breaking of chemical bonds and the crucial transition-state intermediates in chemical reactions.
- Understanding of the origins of nanoscale charge and spin order and their dynamics in correlated materials through high-resolution energy- and time-dependent x-ray spectroscopies.
  - to visualize through ultrafast x-ray motion pictures the performance limits of materials, e.g., the speed limit of reliable switching of a spintronics device.

Photon Spectroscopies…

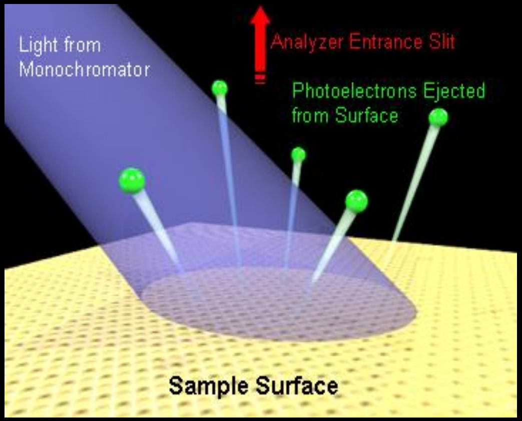


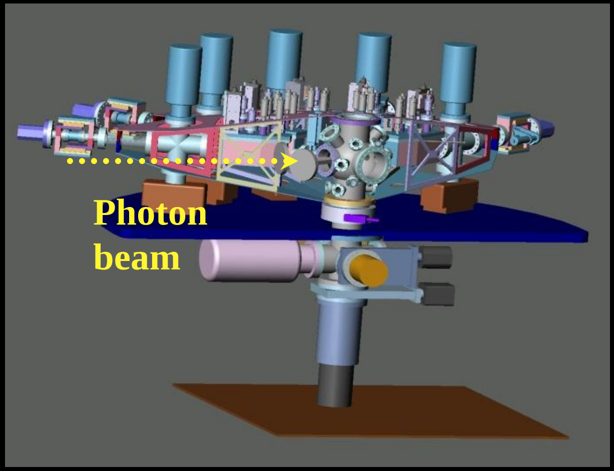
Photoemission (PES, XPS) – measures occupied density of states

Absorption (XAS) – measures unoccupied density of states

Inelastic Light Scattering (IXS) – measures excitations & dynamics

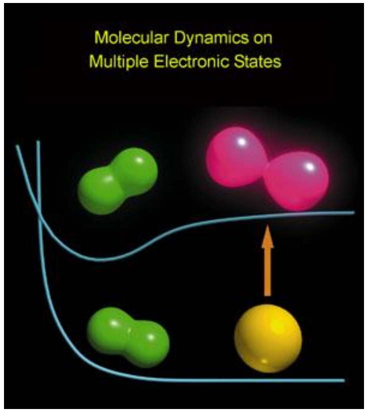
Technique





q-resolved RIXS endstation

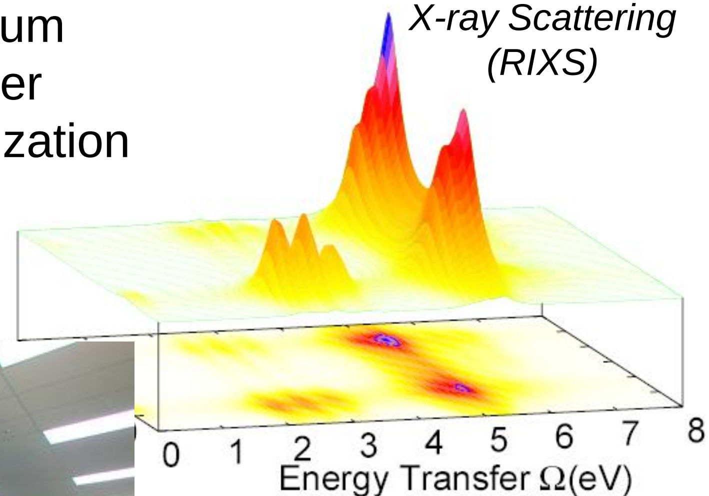
Computational Approaches to Photon Science



Reaction Dynamics

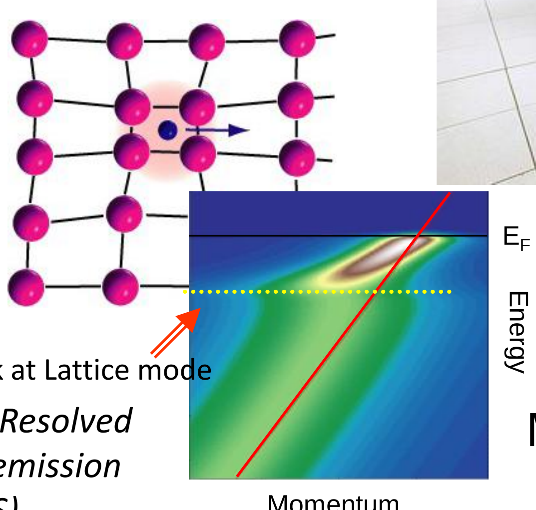
Quantum
Cluster
Diagonalization

Resonant Inelastic X-ray Scattering (RIXS)



Quantum
Molecular
Dynamics

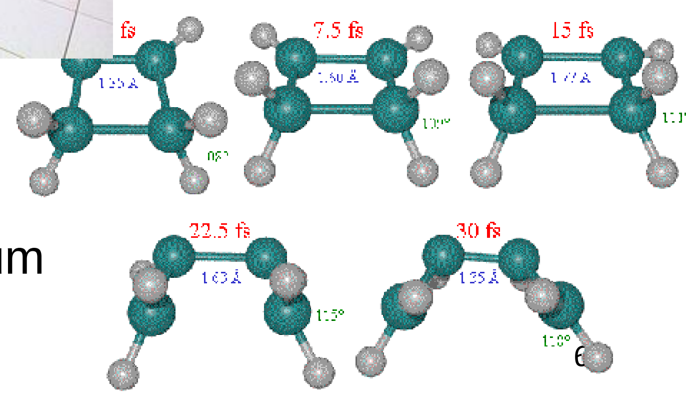
Femtosecond spectroscopy



Angle-Resolved Photoemission (ARPES)

Quantum
Monte Carlo

Time-
dependent
non-equilibrium
dynamics



Quantum Monte Carlo for Spectroscopy

Auxiliary Field QMC and MEM

Finite Temperature, Imaginary Time Green's Function Formalism

Trotter-Suzuki Decomposition

$$\beta = L\Delta\tau$$

Hubbard Stratonovich Transformation

$$e^{-U\Delta\tau(n_\uparrow-\frac{1}{2})(n_\downarrow-\frac{1}{2})} = \frac{1}{2}e^{\frac{U\Delta\tau}{4}}\sum_s e^{\lambda s(n_\uparrow-n_\downarrow)}$$

Sample the Auxiliary Field HS variables with "Boltzmann weight" equal to the product of two determinants

White *et al* – *Phys. Rev. B* **40**, 506 (1989)

Limitations:

- Trotter Error ($\Delta\tau^2$)
- Fermion Sign Problem

$$G(K,\tau) = \int d\omega \frac{A(K,\omega)e^{-\tau\omega}}{1+e^{-\beta\omega}}$$

Maximum Entropy Method (MEM)

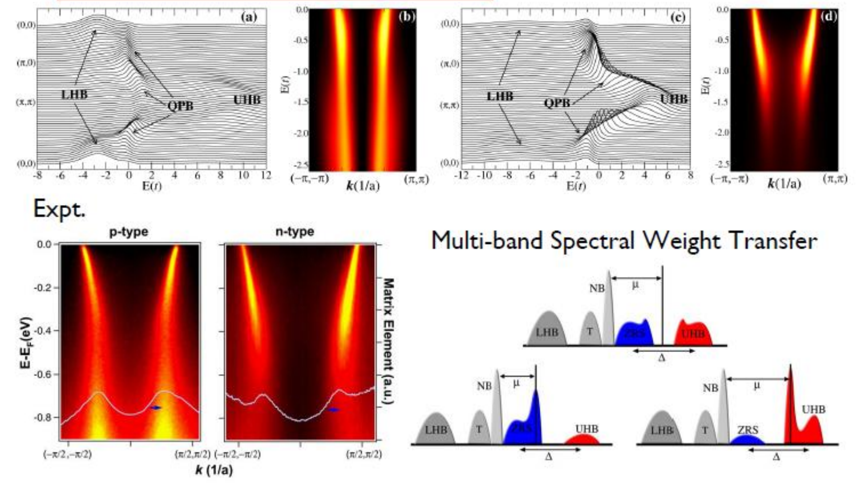
$$Q = \alpha S - \frac{1}{2}\chi^2$$

$$S = -\int d\omega A(\omega)\ln[A(\omega)/m(\omega)]$$

- Stochastically Sample $A(K,\omega)$
- Maximize Q for each α
- Search for α_{max} that generates the Q_{max}

Jarrell *et al* – *Phys. Reports* **269**, 133 (1996)

DQMC - Electron vs. Hole Doping

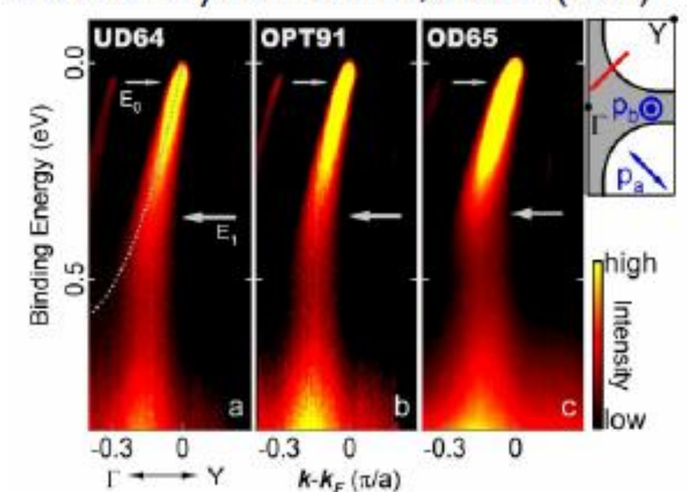


- good statistics needed for post-processing.
- not memory/storage intensive.
- Markov - "perfectly" parallel under weak scaling ~ 128-256 procs.

Limitations

- resolution in momentum space.
- warm-up overhead limits scalability.
- could be mitigated by storing and swapping configurations.

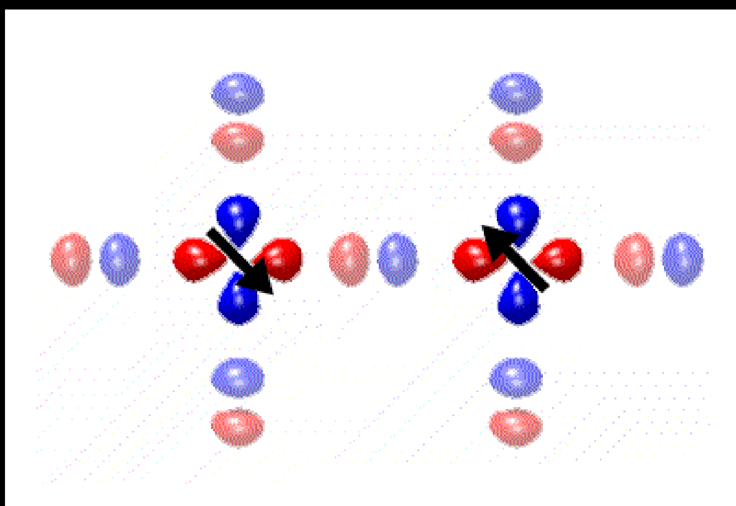
Graf *et al* - *Phys. Rev. Lett.* **98**, 067004 (2007)

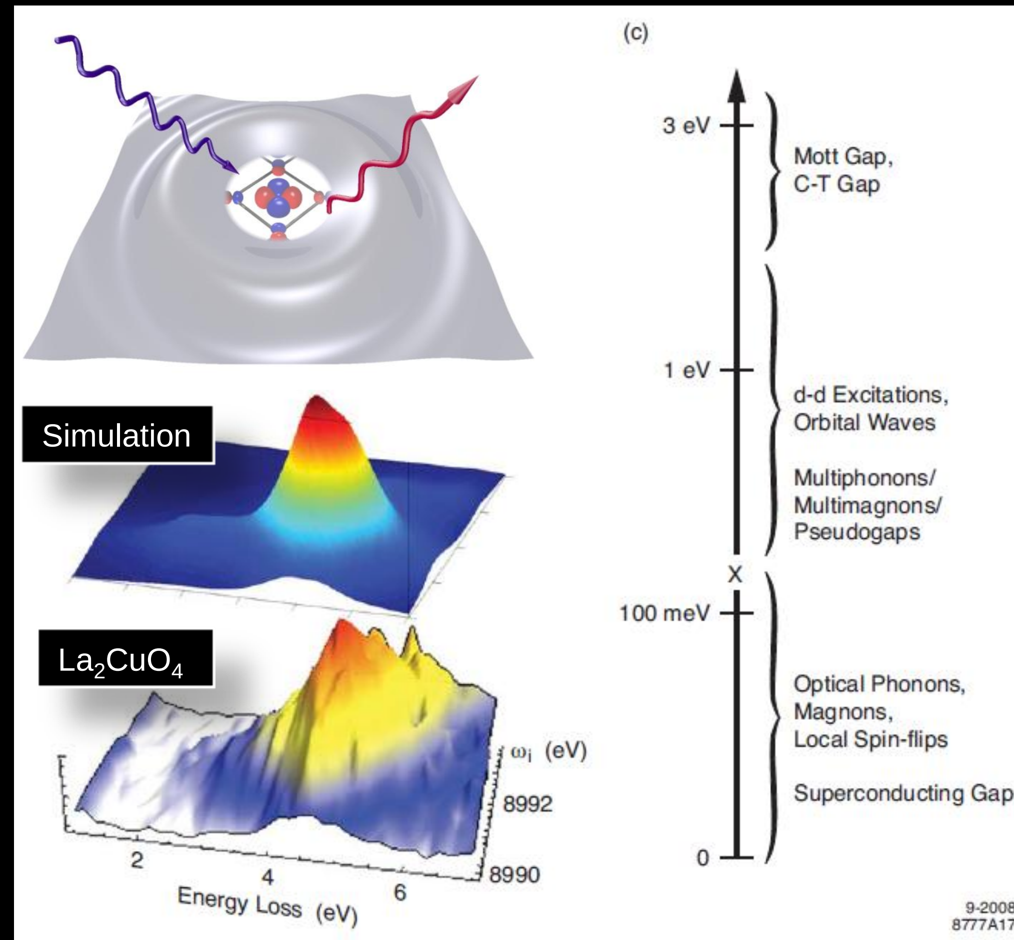


Exact Diagonalization Routines for X-ray spectroscopy

Quantum Clusters + DFT
Sparse Matrix ED
Hilbert spaces ~ 10^8

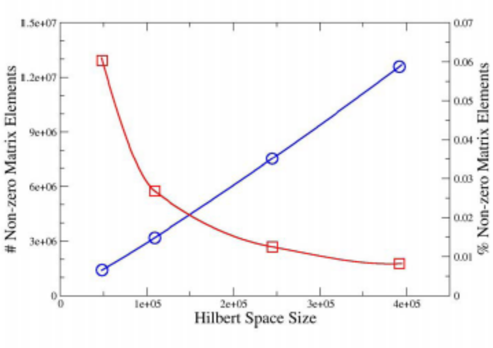
Energy loss: $\omega=\omega_2-\omega_1$
Momentum transfer: $q=k_2-k_1$
Resonance: $\omega_1 \sim \omega_{edge}$

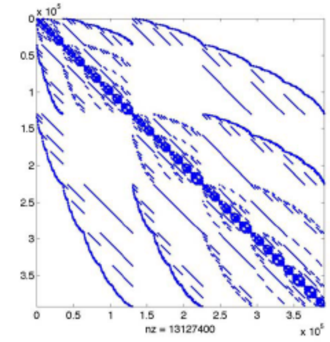




$$I(\omega_I, \Omega = \omega_I - \omega_F) \propto \sum_f \left| \sum_i \sum_{4p} \frac{\langle \psi_f | \psi_{ci} \rangle \langle \psi_{ci} | \psi_0 \rangle}{E_{ci} + \epsilon_{4p-1s} - E_0 - \hbar\omega_I - i\Gamma_1} \right|^2 \times \delta\left(E_F - E_I - \hbar\Omega\right)$$

Sparse Matrix ED

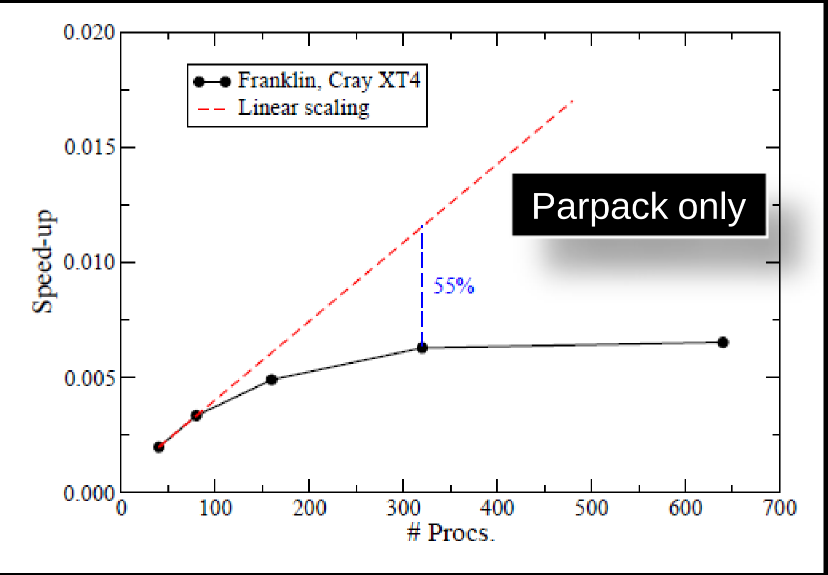




Exact Diagonalization ~ 14,000

Sparse Matrices – ARPACK/PARPACK Libraries

- Ground/Excited States
- Core-hole States
- DFT – Materials Specificity

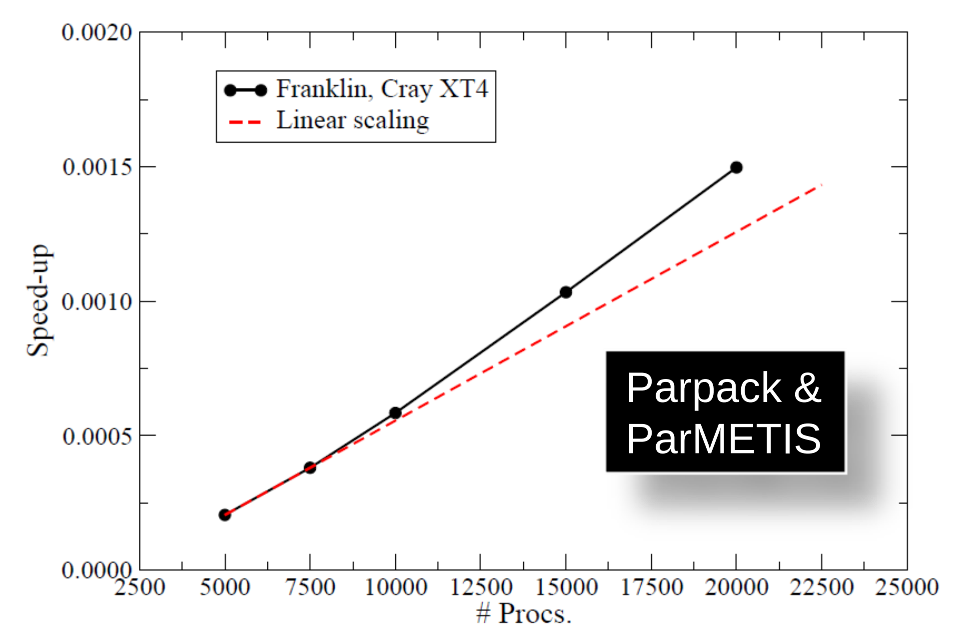


-PARPACK: Parallel Lanczos-Arnoldi iterative matrix diagonalization.

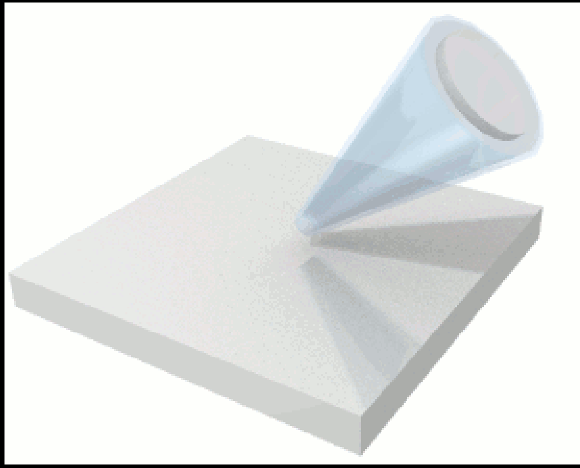
-Superlinear strong scaling up to ~10K procs. w/ ParMETIS: *parallel graph partitioning and fill-reducing matrix ordering .*

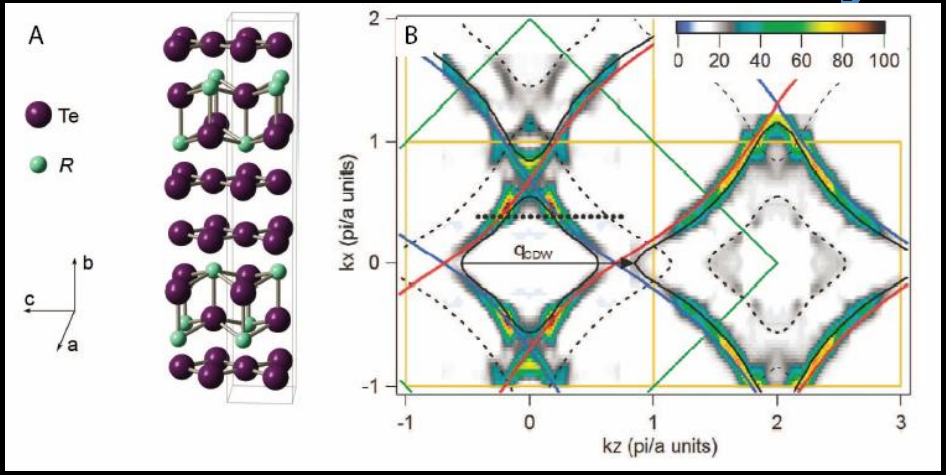
Limitations

- exponential scaling with Hilbert space.
- memory limited.

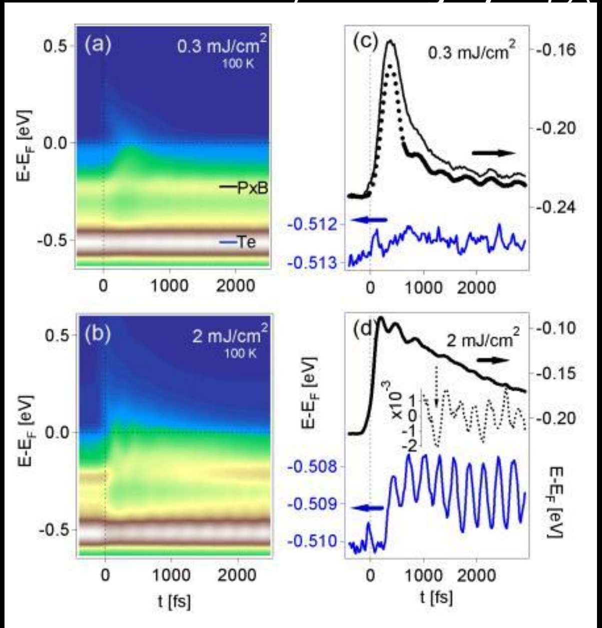


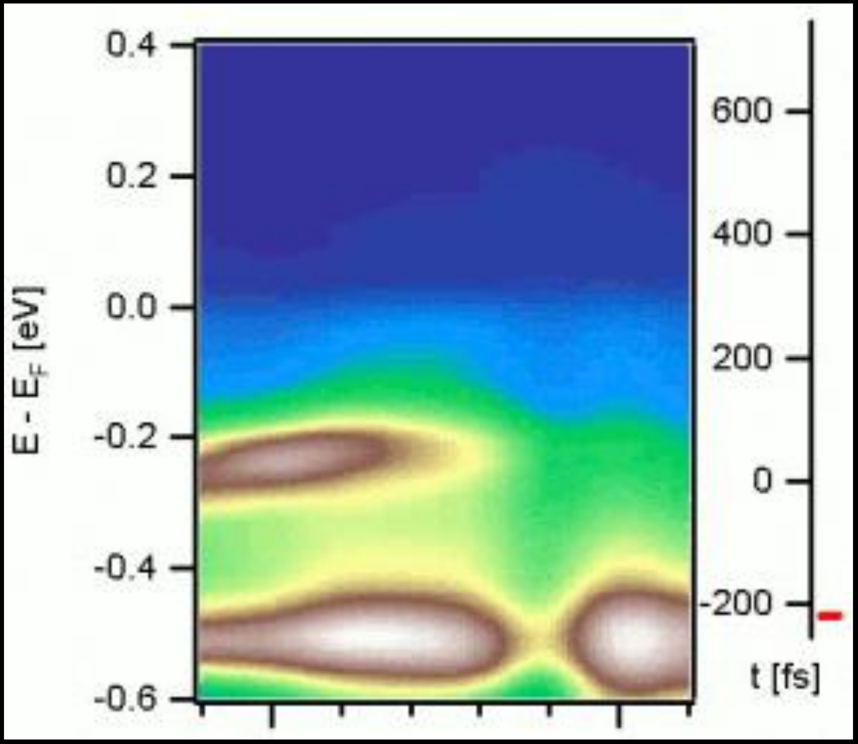
Science Driver: Time-resolved pump-probe photoemission in charge density wave material $TbTe_3$





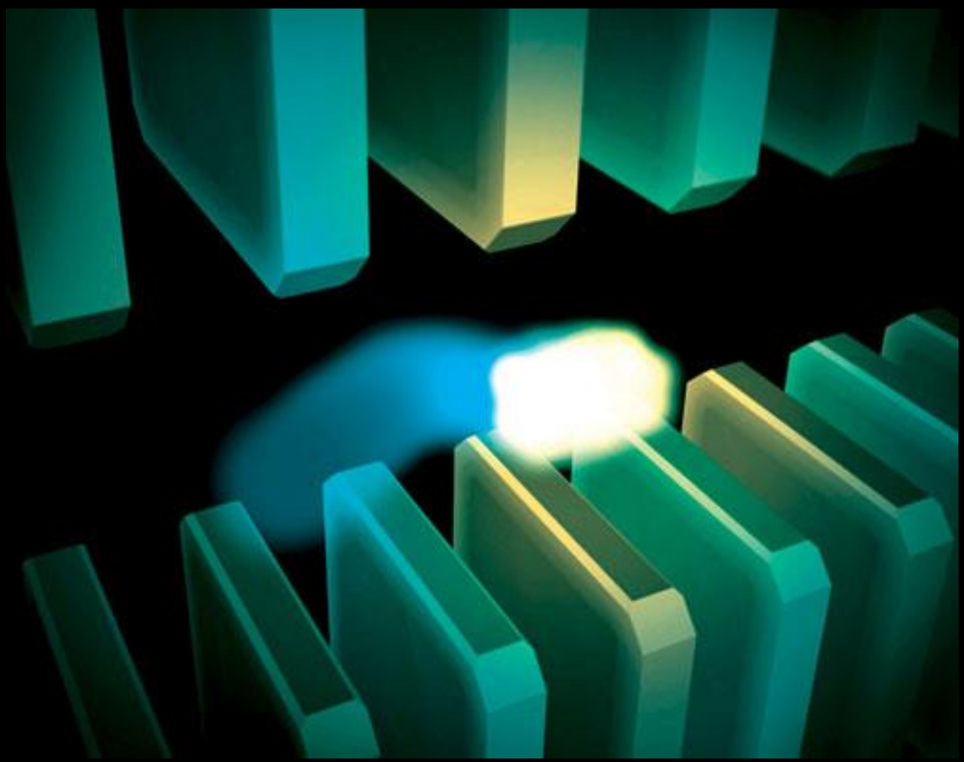
F. Schmitt et al, Science ***321,*** *1649 (2008)*

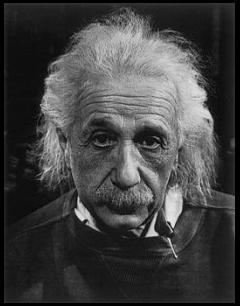




- Energy, momentum, and time-resolved spectral function.
- Spectral weight shifts dramatically; CDW gap "closes".
- Oscillation frequencies set by CDW & lattice phonons.

LCLS – a new era of science



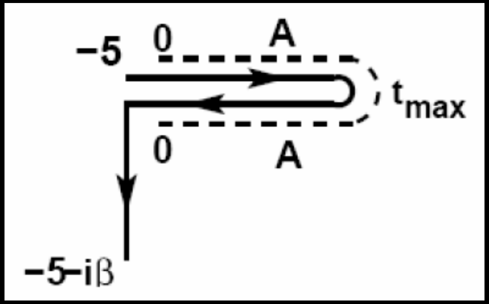


Relativistic electron bunches see length contracted alternating magnets

- World's longest linear accelerator producing the first hard x-ray free electron laser 1.5 Å wavelength.
- Billion times brighter than any other light source.
- pulse width 10-100 femtoseconds (or less).
- Experiments underway, open for users Fall 2009.

Time-domain non-eq. spectroscopies

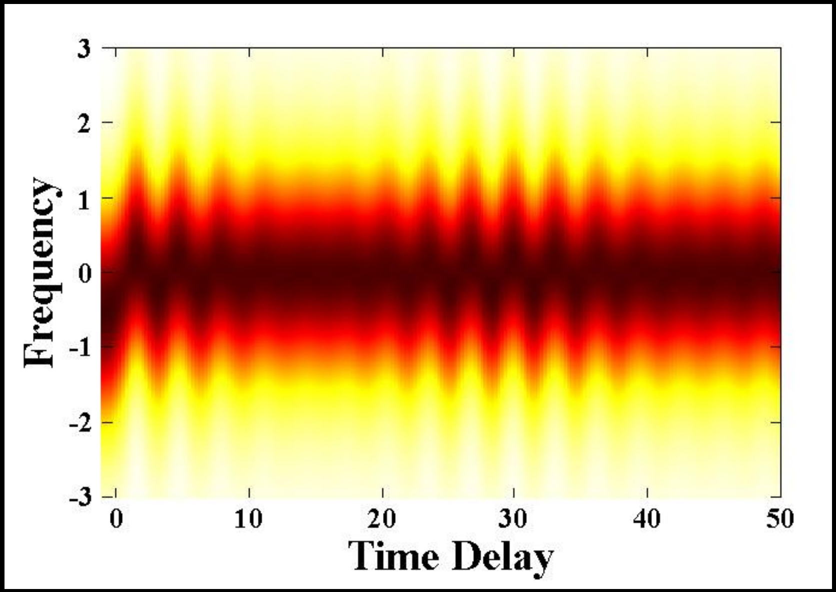
2.25M cpu-hrs
INCITE Award

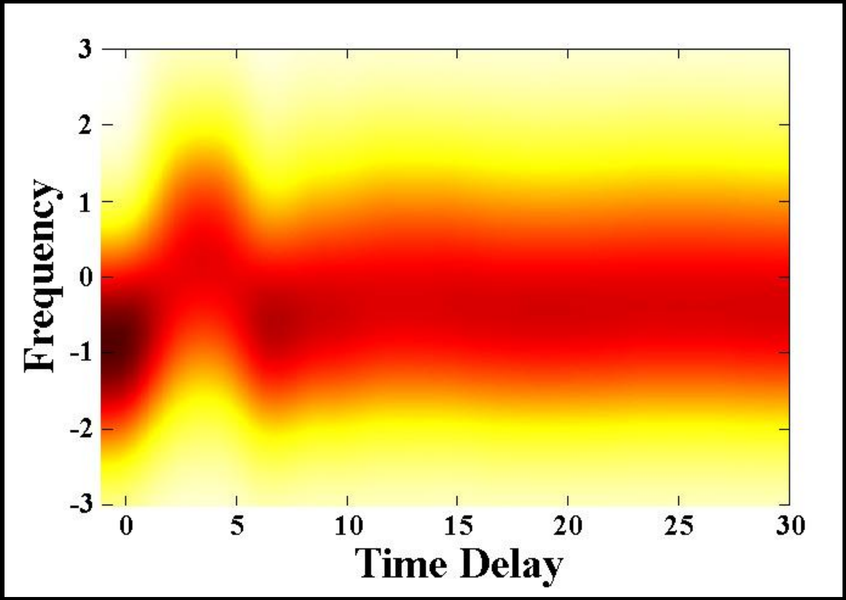


Double-time Greens function on Keldysh contour using DMFT – include fields exactly to all orders (Moritz, Freericks, TPD) arXiv:0908.1807

Bloch oscillations in metal U<Uc

Bloch oscillations damped in insulator U~Uc

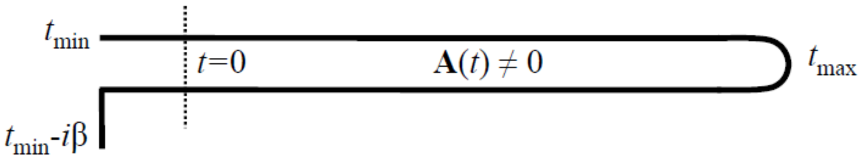




Pump-probe DMFT formalism

Nonequilibrium DMFT

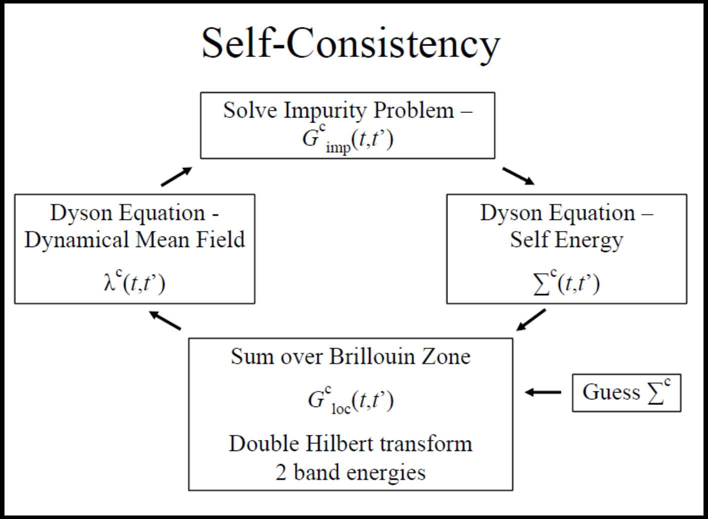
- Recent formulation of DMFT for nonequilibrium problems
  Freericks *et al.*, *Phys. Rev. Lett.* **97**, 266408 (2006)
- Evaluate the contour-ordered, double-time Green function on the Kadanoff-Baym-Keldysh contour



- Lesser Green function:

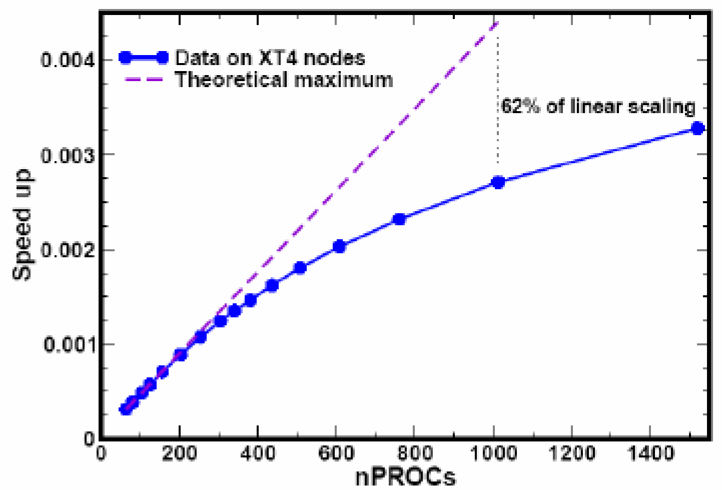
$$G_{ij}^{<}(t,t') = iZ_{eq}^{-1}\mathrm{Tr}[e^{-\beta H_{eq}} c_j^{\dagger}(t')c_i(t)]$$

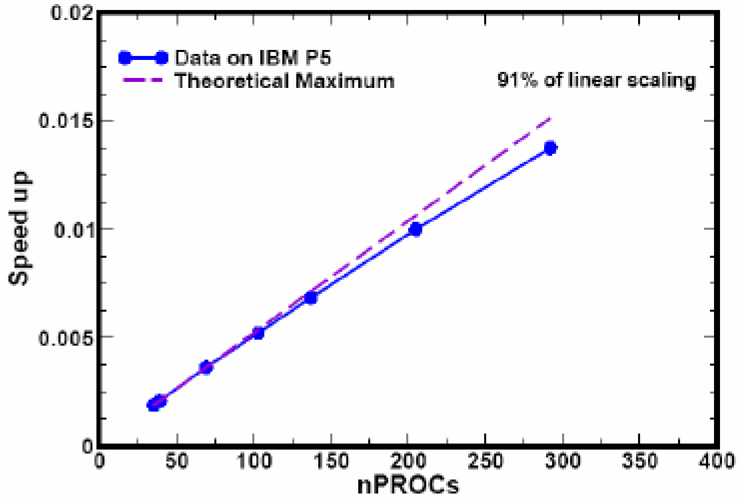
- Include the effects of strong driving field through Peierls' substitution - $\boldsymbol{k} \rightarrow \boldsymbol{k} - e\mathbf{A}(t)$



- Iterative solution of coupled set of integral equations.
- critical elements – local Green's function: matrix inversion and multiplication of dense complex matrices.
- wall-times limit the number of iterations per run.
- GPUs can be utilized (simple BLAS and LAPACK).

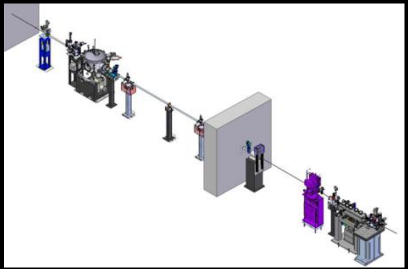
Performance and Resources



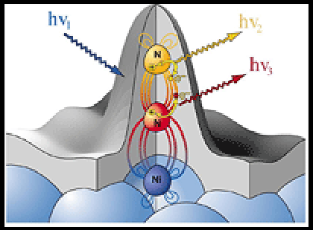


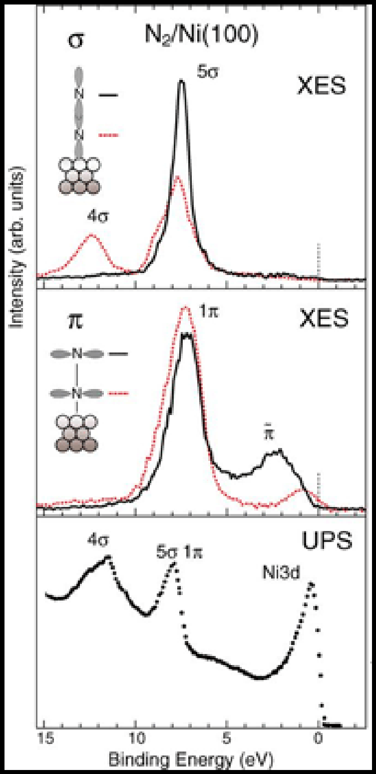
- Performs similarly on a number of platforms –
  Cray XT3, Cray XT4, HP Beowulf, Sun Opteron, SGI Altix
- Dependent on cache size
- Message passing bottleneck overcome with a recursive binary gather to improve scaling beyond 1000 processors
- INCITE award - 2,250,000 CPUhrs.

Soft x-ray consortium @ LCLS to begin spring 2010.

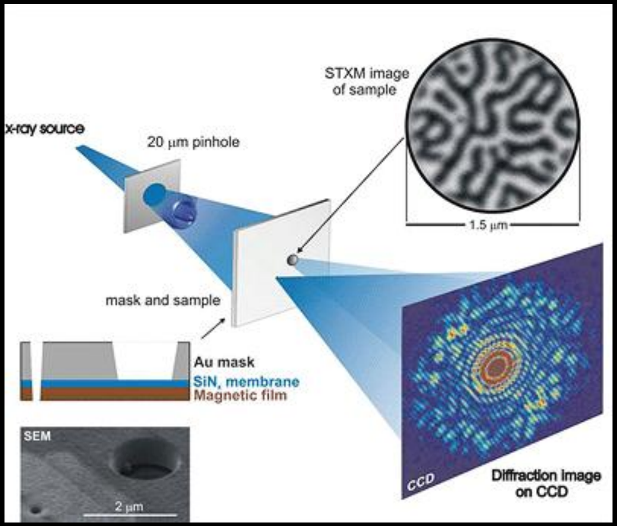


Pump-probe ultrafast chemistry

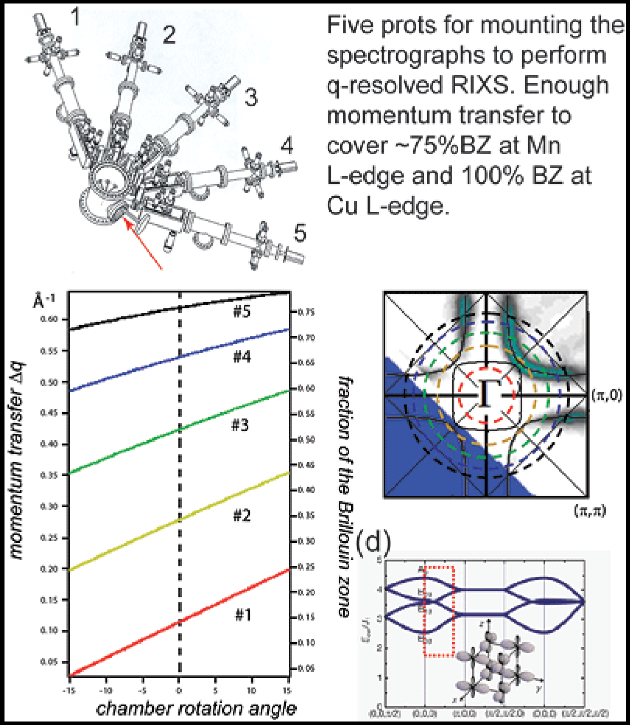




Magnetic imaging



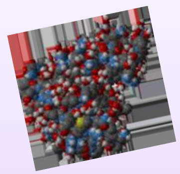
X-ray scattering spec.



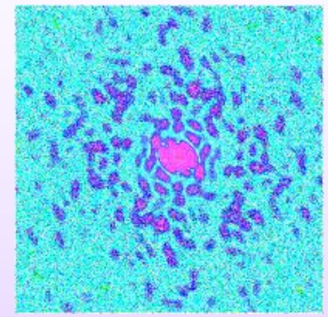
Coherent Imaging of Single Molecules

• *Diffraction from a single molecule:*

single LCLS pulse



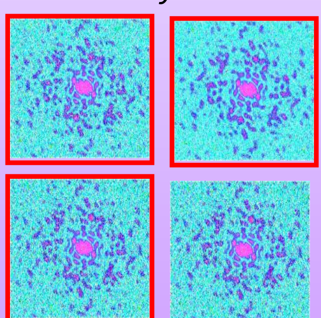
unknown orientation



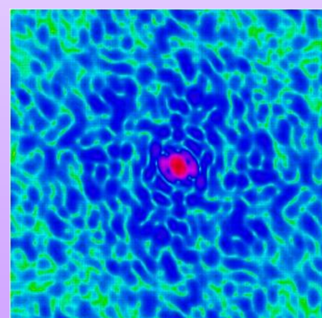
noisy diffraction pattern of unknown orientation

• *Combine 10^5 to 10^7 measurements into 3D dataset:*

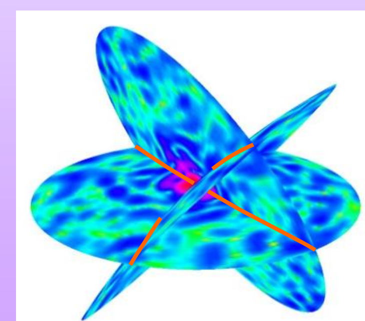
Classify/sort



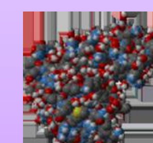
Average



Alignment



Reconstruct by Oversampling phase retrieval



The highest achievable resolution is limited by the ability to group patterns of similar orientation

Gösta Huldt, Abraham Szöke, Janos Hajdu (J.Struct Biol, 2003 02-ERD-047)

Miao, Hodgson, Sayre, PNAS 98 (2001)

CXI Peak Data Rate and Volume

For CXI Detector: 1.16 Mpixel & 14 Bit encoding & at 120 Frames/sec			
	Average (in 2009)	Lower Limit	Upper Limit
Peak Rate (Gigabit/s)	1.95	1.95	1.95
Success Rate (%)	10%	5%	100%
Ave. Rate (Gigabit/s)	0.20	.097	1.95
Daily Duty Cycle (%)	25%	10%	100%
Accumulation (Terabyte/day)	0.53	0.11	21
Yearly Uptime (%)	25%	10%	100%
Accumulation (Petabyte/year)	0.048	0.0038	7.7

require high performance and high capacity data acquisition and management system

Longer Term Trend

For CXI Detector: pixel number x2/3 years			
	Average (in 2009)	2012	2015
Peak Rate (Gigabit/s)	1.95	3.90	7.80
Success Rate (%)	10%	30%	50%
Ave. Rate (Gigabit/s)	0.20	1.17	3.90
Daily Duty Cycle (%)	25%	50%	75%
Accumulation (Terabyte/day)	0.53	6.31	31.6
Yearly Uptime (%)	25%	75%	90%
Accumulation (Petabyte/year)	0.048	1.73	10.4

Pixels will go up, machine will be more stable and measurement techniques more refined

Summaries

- Algorithm development needed on fundamental issues (50x compute power?).
- Memory/core increase, latency reduction very useful.
- GPU attractive.
- Large data set analysis tied to light scattering facilities.