Workshop on Ultrafast X-ray Science

Advanced Photon Source
Argonne National Laboratory
June 4-5, 2002

Sponsored by:

MHATT-CAT,
the University of Michigan FOCUS Center,
and the Advanced Photon Source,

Workshop Organizers:

David Reis, University of Michigan
Roy Clarke, University of Michigan
Dennis Mills, Advanced Photon Source, Argonne National Laboratory

SUMMARY

On June 4th and 5th, 2002, a workshop, “Ultrafast X-ray Science”", was held at the Advanced Photon Source, Argonne National Laboratory. This event brought together leading experts and potential users in the production and application of pulsed x-ray beams. The workshop was sponsored by the University of Michigan-Howard University-Bell Labs Collaborative Access Team (MHATT-CAT), the Advanced Photon Source, and FOCUS, a new NSF Frontier Center at the University of Michigan devoted to the physics of coherent control.

This workshop was aimed at establishing a strong user base for the emerging field of time-resolved x-ray science through the marriage of ultrafast laser technology and synchrotron radiation. One of its main goals was to identify needs and opportunities among the community of potential users and to ensure broad access through the APS Independent Investigator program.

The agenda included several invited science presentations by potential users as well as technical presentations describing the unique capabilities available at the APS. In particular, the workshop tried to identify what experiments are best served by the characteristics of synchrotron radiation at the APS, and explored how to extend the capabilities of synchrotron based-facilities through advances in both instrumentation and the source

The workshop participants identified a range of exciting scientific opportunities in ultrafast x-ray science spanning physics, chemistry, and the earth- and life-sciences. The complementary needs of diffraction and spectroscopy were discussed in break-out sessions on the second day of the workshop, leading to recommendations from each group of scientists involved in these areas. An important aspect of the APS is the ability to use the standard bunch pattern for fast time-resolved studies. It was recommended that a fast chopper would extend the usefulness of this approach. Further development of advanced detectors, such as the picosecond x-ray streak camera, is a key requirement for progress in this field. The participants also stressed that beamline staffing devoted to ultrafast experiments would be essential for the success of a user program.

The high level of interest and enthusiasm expressed at this workshop are a reflection of new science possibilities at this frontier of x-ray science. There was a general consensus that the time-resolved expertise already accumulated at the APS could serve as a basis to build a strong, open-access user community in ultrafast x-ray science.

The organizers thank the following for their help in putting together this workshop: Michelle Murn and Siddharth Shah of the University of Michigan FOCUS Center, Susan Strasser, of the Advanced Photon Source User Office, Joan Brunsvold and Judy Benigno of Argonne Conference Services and Eric Dufresne, of MHATT-CAT. This workshop was made possible, free of fees, by generous support from FOCUS (NSF), MHATT-CAT (DOE contract #DE-FG02-99ER45743) and the APS (DOE contract #W-31-109-Eng-38).

Workshop on Ultrafast X-ray Science

Agenda

Monday, June 3rd

7:30–9:30 pm	informal reception, Argonne Guest House (snacks and cash bar)

Tuesday, June 4th

the Lodge, Building 600

7:30 am	continental breakfast

APS and MHATT-CAT SESSIONS

8:00 am	Introductory Remarks	David Reis
8:05 am	Welcome and Introduction to APS	Denny Mills
8:30 am	APS technical overview	Glenn Decker
9:00 am	Independent Investigator Information	Susan Strasser
9:20 am	Coffee Break	
9:45 am	MHATT-CAT Beamline Capabilities and Science	Roy Clarke, Eric Dufresne, Don Walko, Matt DeCamp

APS sector 7, Bldg 432-D

11:15–12:30 pm	tours of MHATT-CAT
12:30–2:00 pm	Lunch (Suggest Cafeteria or Argonne Guest House)

SCIENCE TALKS SESSION 1
the Lodge, Building 600

2:15–2:45 pm	Jorgen Larsson, Lund University Time-resolved X-ray diffraction investigations
2:45–3:15 pm	Aaron Lindenberg, U.C. Berkeley Time-resolved structural dynamics on the nano-scale
3:15–3:45 pm	Linda Young, Argonne National Lab X-ray photoionization of laser-dressed atoms

3:45-4:15 pm Coffee Break

SCIENCE TALKS SESSION 2
the Lodge, Building 600

4:15–4:45 pm	Reinhard Pahl, BioCARS Advanced Photon Source Time-resolved Macromolecular Crystallography at BioCARS
4:45–5:15 pm	Christoph Rose-Petruck, Brown University Ultrafast Laser Plasma Sources and X-ray Absorption Spectroscopy in Chemical Research
5:15–5:45 pm	Jean-Claude Kieffer, University of Quebec Ultrafast Instrumentation for Time-resolved Structural Studies *Withdrawn*
5:45–6:15 pm	Peter Sondhaus, Oxford University Time-resolved X-ray Diffraction from Coherent Phonons in Semiconductor Heterostructures

Argonne Guest House

7:30 pm dinner

Wednesday June 5
the Lodge, Building 600
BREAK OUT SESSIONS

8:30 continental breakfast
9:00 organize break out sessions
9:15–12:00 break out sessions (locations TBA)

12:00–2:00 lunch

2:00–3:45 re-convene and short presentation by break out groups
3:45–4:15 coffee break
4:15–5:00 continue discussions including recommendations.
5:00–5:15 concluding remarks
5:15– Informal proposal writing session

Workgroup "Spectroscopy"
Members:

Patrick Forget
Christoph Rose-Petruck
Roy Clarke
Matthew DeCamp
Phil Bucksbaum
Linda Young
Bernhard Adams

The bending magnet will be able to produce radiation above 5keV and below 45 keV without substantial beam line effort. This implies that transition metal complexes above Ti can be measured.
Many x-ray diffraction experiments we saw in the workshop could be performed by x-ray absorption spectroscopy (XAS).
A 256-pixel AVD could be valuable for x-ray detections. The desired detector properties should be:
Near-edge structure: 1eV resolution, 200 eV spectral range
EXAFS: 5 eV resolution, 1000 eV spectral range
Both requirements can be satisfied with the same detector by varying the sample detector distance
Having a chopper, enable the use of a LN2-cooled CCD camera as a detector instead of an AVD.

Scientific motivation

Fluorescence detection
Jim Penner-Hahn: Metalloprotein structural dynamics by EXAFS

Atomic physics: Rydberg state dynamics of atoms such as argon or potassium could be observed measuring the pre-edge structure of these elements after photo-preparation of the Rydberg states.

Plasma physics: Use the x-ray beam as a back-lighter to observe the temporal evolution of optically thick laser-induced plasmas. Goal could be to observe the plasma dynamics inside of a plasma that are usually inaccessible by emission x-ray spectroscopy because of the optical thickness of the plasma.

Chemistry: At low temporal resolution, we observe reaction rates, not structural dynamics, i.e. the motions of atoms.
XANES:
Coordination geometry of transitions metal coordination complexes
Pre-edge: Electronic structure of transitions metal coordination complexes
Catalysis
Solid-state physics:

Charge carrier dynamics in photo-excited semiconductors
Many x-ray diffraction experiments we saw in the workshop could be performed by x-ray absorption spectroscopy (XAS).

X-ray fluorescence could be coupled efficiently to a streak camera, transmission <=10^{-4}. Such a setup is particularly useful for low concentration samples.

Absorption detection
Good at rather concentrated samples, i.e. most non-biological, non-surface systems.

Transmission detection setups should have the following optical arrangement:
BM or undulator -> beam chopper -> focusing mirror -> x-ray focus in the sample -> crystal spectrometer -> detector
This arrangement has the following advantages:
Diffraction will occur in the sample. This reduces the spectral resolution of the setup because it blurs the beam on the detector. Thesefore, it is important that a) the sample sits in the x-ray focus AND that the crystal spectrometer follows the sample region.

Workshop on Ultrafast X-ray science, APS June 4-5, 2002

Diffraction experiments

Contributors:
Aaron Lindenberg
Peter Sondhauss
Jon Tischler
Ben Larson
Soo-Heyong Lee
Adrian Cavalieri
David Fritz
Dohn Arms
Eric Dufresne
Roberto Merlin
Dave Reis

The high brightness undulator beamline at MHATT-CAT coupled to the ultrafast laser has the potential to host a unique user programme allowing the studies of non-equilibrium structural dynamics. A number of experiments were discussed, they include Gain measurements for acoustic phonons in semiconductors, control of acoustic phonons using layered targets, studies of structural dynamics in nanocrystals and studies of the structure of water under high-pressure, high-temperature conditions, not readily available in laboratories today.

The discussed experiments are listed below, names in parenthesis are contact people for the different experiments. They are sorted under three headlines. Experiments on the 100 ps timescale are accessible today. Experiments in the 1-100 ps timescale will require a streakcamera. A streakcamera is under development at MHATT-CAT and can hopefully be commissioned during 2002. Experiments and improvements of experiments feasible today requiring additional upgrade of the instrumentation were also considered.

Experiments on the 100 ps timescale

Thermal transport in strong gradients (Ben Larson)

Liquid dynamics (Aaron Lindenberg)

Studies of liquids at high pressure-high temperature (Aaron Lindenberg)

Experiments on a faster timescale
requires a streakcamera
Acoustic phonons - quantitative numbers, amplitudes. (Roberto Merlin, Jorgen Larsson)

Control the phonons by multilayer systems (Peter Sondhauss)

Optical phonons (Roberto Merlin)

Liquid/nano-particle dynamics (Aaron Lindenberg)

Measurement of electron-phonon coupling times (Jorgen Larsson)

Look for acoustic phonon gain in semiconductors (Roberto Merlin)

Other capabilities
Sub-ns laser 1J allows for shockwave experiments

A chopper would make better use of photon flux for powder diffraction experiments.

User opinion and recommendations
The APS is currently the best source available for ultrafast X.ray studies. In particular, the standard filling pattern is well suited for time-resolved studies giving the APS an advantage over ESRF where required specialized filling modes reduces the available beamtime suited for time-resolved work. The high brightness X-ray undulator beamlines give higher flux and brightness than similar facilities at lower energy machines.
MHATT-CAT encompasses leading groups in the field of ultrafast science. It would be natural to let MHATT-CAT lead the development of ultrafast X-ray science at APS. However, in order to launch a successful user program, a dedicated beamline scientist with experience in working with ultrafast lasers will be needed.

Participants:

Bernhard	Adams	Experimental Facilities Division	Argonne National Lal
Dohn	Arms	Advanced Photon Source	Argonne National Lal
Dale	Brewe	APS/USR	Argonne National Lal
Philip	Bucksbaum	Department of Physics	University of Michig
Adrian	Cavalieri	Department of Physics	University of Michig
Lin	Chen	Chemistry Division	Argonne National Lal
Codrin	Cionca	Applied Physics Department	University of Michiga
Roy	Clarke	Applied Physics Program	University of Michiga
Rob	Crowell	Chemistry Division	Argonne National Lal
Steve	Davey	APS/AOD	Argonne National Lal
Matthew	DeCamp	Department of Physics	University of Michig
Glenn	Decker	APS/AOD	Argonne National Lal
Eric	Dufresne	APS/USR	Argonne National Lal
Robert W.	Dunford	Chemistry Division	Argonne National Lal
Patrick	Forget	INRS	University of Quebec
David	Fritz	Department of Physics	University of Michiga
David	Gosztola	Chemistry Division	Argonne National Lal
Jesse	Guzman	Department of Physics	University of Michig
Elliot	Kanter	Chemistry Division	Argonne National Lal
Bertold	Kraessig	Chemistry Division	Argonne National Lal
Bennett C.	Larson	Solid State Division	Oak Ridge National L
Jorgen	Larsson	Physics Department	Lund Institute of Tecl
Soo-Heyong	Lee	Department of Physics	University of Michiga
Aaron	Lindenberg	Physics Department	University of Califorr
Roberto	Merlin	Physics Department	University of Michiga
Dennis M.	Mills	Advanced Photon Source	Argonne National Lal
Reinhard	Pahl	APS/USR	University of Chicago
David	Reis	Department of Physics	University of Michiga
Christoph	Rose-Petruck	Chemistry Department	Brown University
Peter C.	Sondhauss	Atomic and Laser Physics Department	Oxford University
Steve	Southworth	Chemistry Division	Argonne National Lal
Vladimir	Stoica	Applied Physics Department	University of Michiga
Susan Barr	Strasser	APS Operations Division	Argonne National Lal
Marion C.	Thurnauer	Chemistry Division	Argonne National Lal
Jon	Tischler	APS/USR	Oak Ridge National L
Donald	Walko	APS Operations Division	Argonne National Lal
Linda	Young	Chemistry Division	Argonne National Lal

Workshop on Ultrafast X-ray Science
June 4th–5th, 2002
Argonne National Lab

thanks to:

•Advanced Photon Source

•Applied Physics Program,University of Michigan

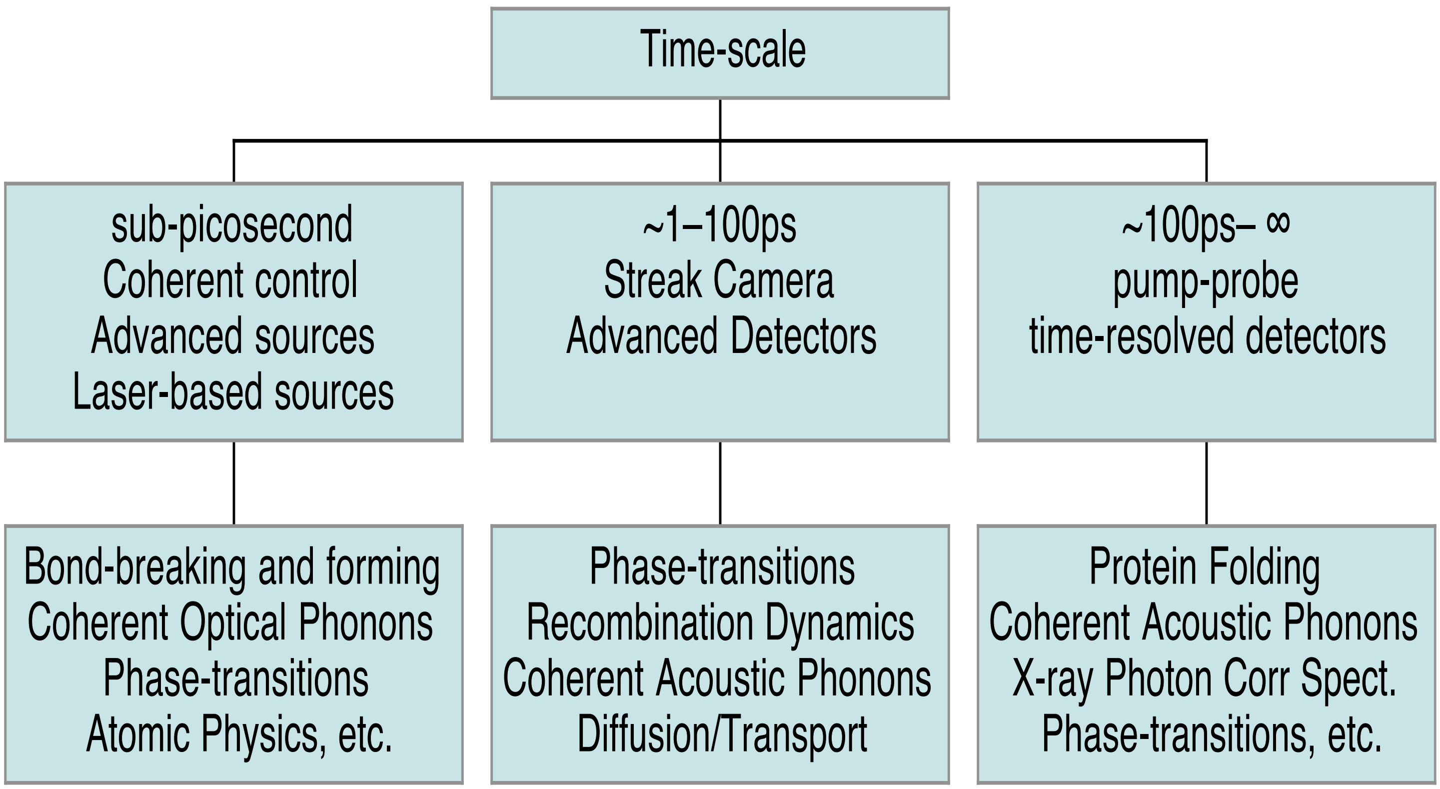
•FOCUS Center, University of Michigan

and especially:

Judy Benigno
Joan Brunsvold
Siddharth Shah
Susan Strasser

and…all our speakers and participants

Time-resolved X-ray Science



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Charge

•Help Develop a strong User Base for time-resolved x-ray experiments at synchrotrons

• Identify most important experiments that can be done on the existing MHATT-CAT beamlines and what advances are necessary to continue producing world class science

•Identify user need in order to complete these experiments (especially in terms of beamline support and instrumentation

Welcome and APS Overview

Dennis M. Mills
Deputy Associate Laboratory Director
Advanced Photon Source

Workshop on Ultrafast X-ray Science
June 4-5, 2002

Advanced Photon Source
ARGONNE NATIONAL LABORATORY

What is the Mission for the APS?

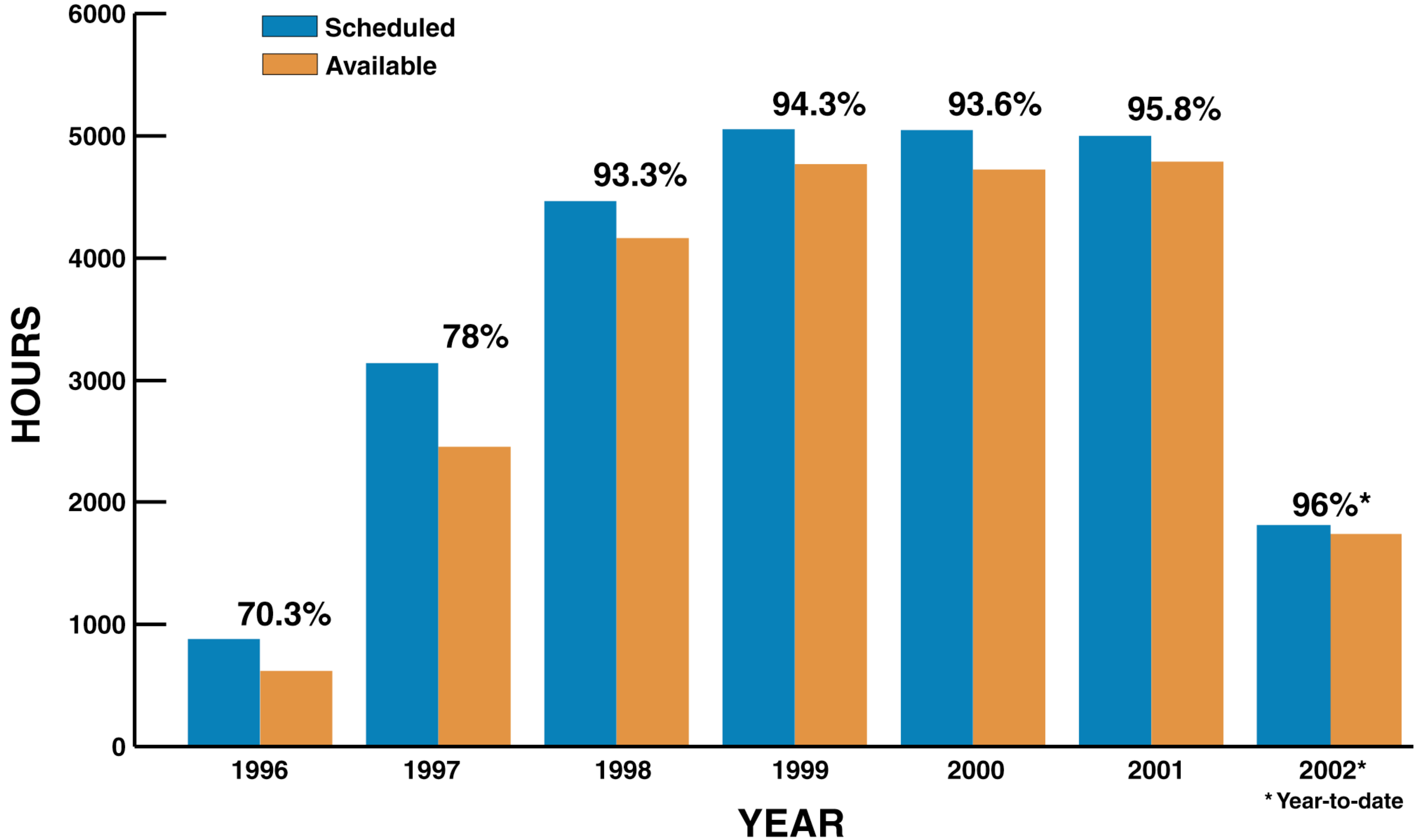
The mission of the Advanced Photon Source is to deliver world-class science and technology by operating an outstanding synchrotron radiation research facility accessible to a broad spectrum of researchers.

- Increase the effectiveness of the facility
- Keep the machine reliable and innovative
- Improve partnership with all users

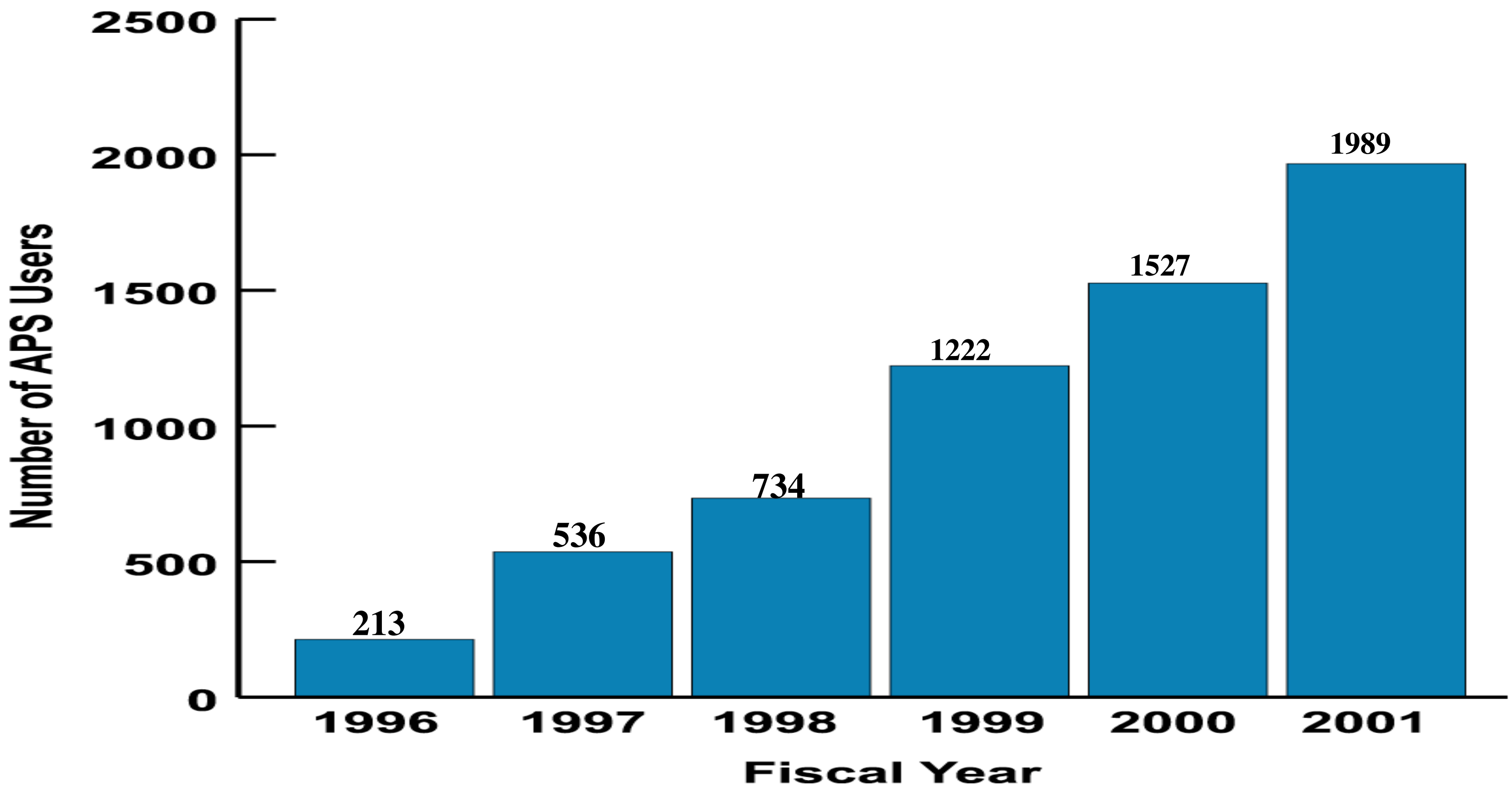
APS Update

- APS continues to provide 5000 user hours per year of the highest quality beam:
  - over 97% availability in 2002 (to date)
  - 31 hours mean time between faults in 2002 (to date)

- The user population continues to grow with
  - over 4800 badged users
  - almost 2000 unique users in 2001


Scheduled
Available
HOURS
6000
5000
4000
3000
2000
1000
0
70.3%
78%
93.3%
94.3%
93.6%
95.8%
96%*
1996
1997
1998
1999
2000
2001
2002*
* Year-to-date
YEAR

Advanced Photon Source
Argonne National Laboratory



Advanced Photon Source

Argonne National Laboratory

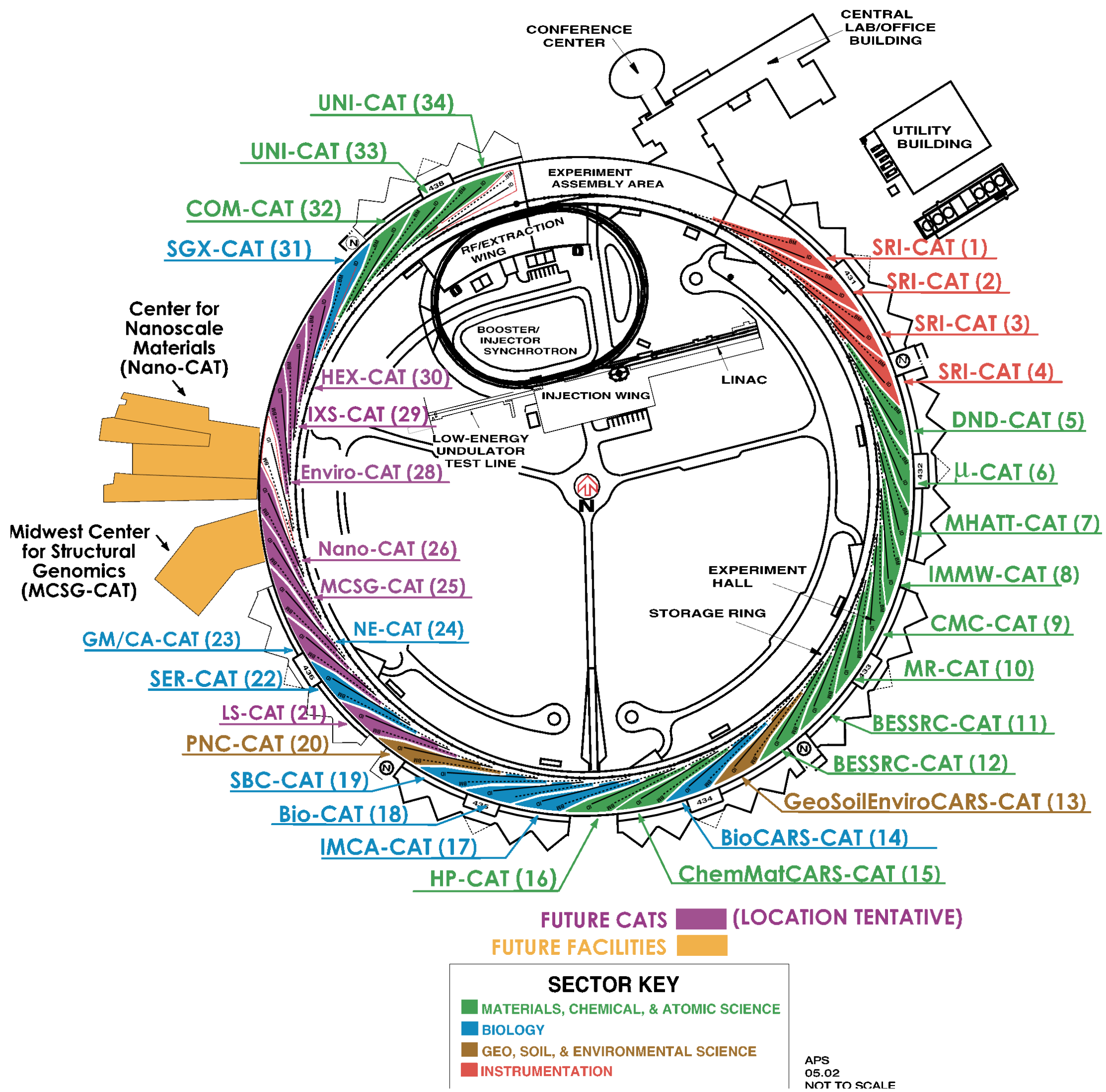
MOUs signed:

- GM/CA CAT
- NE CAT

LOIs from:

- LS CAT
- IXS CAT
- Nano CAT
- HEX CAT
- MCSG CAT

2 empty sectors

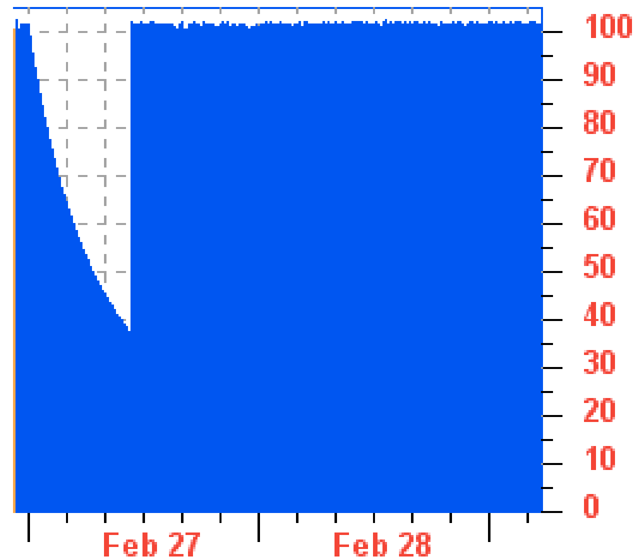


Present Operational Parameters

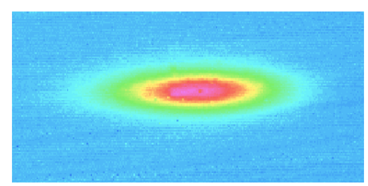
- 7 GeV and 100 mA
- Top-off 75% of the time
- 3.5 nm-rad horizontal emittance and 2% coupling
- At the insertion device source point:
  - Horizontal source size/divergence: 250 μ / 16 μrad
  - Vertical source size/divergence: 17 μ / 4 μrad

Operational Enhancements

- Top-up
  - constant current
  - increased ampere-hours
  - better stability!



- Reduced emittance
  - smaller source size and divergence
  - increased brilliance!



R&D for Future Accelerator Upgrades

- Paths to increased brilliance:
  - Higher current
  - Lower emittance
  - Optimized insertion devices (period, helical, length, etc.)

- Tailoring the source to the user:
  - Tandem undulators
  - Longer straight sections (and longer IDs)
  - Variable beta functions (local control of source size and divergence)

Other Enhancements Under Consideration

- Copper-coated titanium ID chambers
  - Reduce resistive wall effect to allow higher single-bunch current
  - Stronger permitting wider horizontal apertures for cleaner top-up and greater lattice flexibility

- Variable permanent magnet multipoles
  - shorter, stronger and more stable/reliable

- Low-emittance booster to allow smaller ID gaps and cleaner, less visible injection

Time-resolved Studies at the APS

- Several CATs have as part of their scientific program time-resolved studies.
  - MHATT CAT
  - Basic Energy Sciences Synchrotron Radiation Center (BESSRC) CAT
  - BioCARS (a part of CARS CAT)
  - A University- National Lab-Industry (UNI) CAT
  - Pacific Northwest Consortium (PNC) CAT
  - Synchrotron Radiation Instrumentation (SRI) CAT
  - Biophysics (Bio) CAT
  - IBM McGill MIT (IMM) CAT

Time-resolved Studies at the APS

- The APS has attempted to accommodate the wide range of time-scales that researchers are interested in through the selection of:

  – “standard” filling pattern

  – special operations mode (SOM) ≈ 4 weeks per year

(more on this from the next speaker)

Short Term Goals for the APS

- Maintain high quality of delivered beam in terms of reliability, stability, etc.

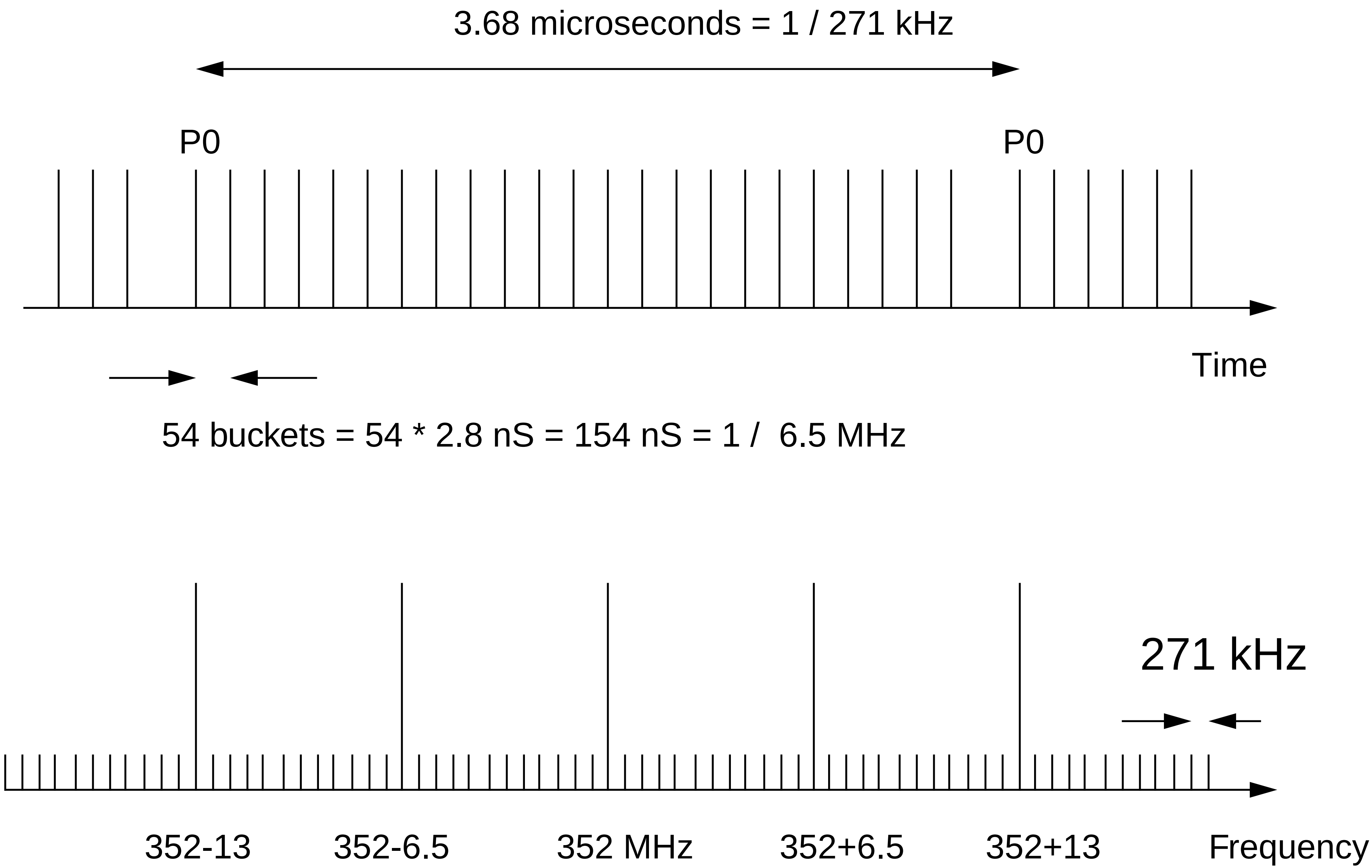
- Enhance the APS support to CATs to improve the operations of beamlines.

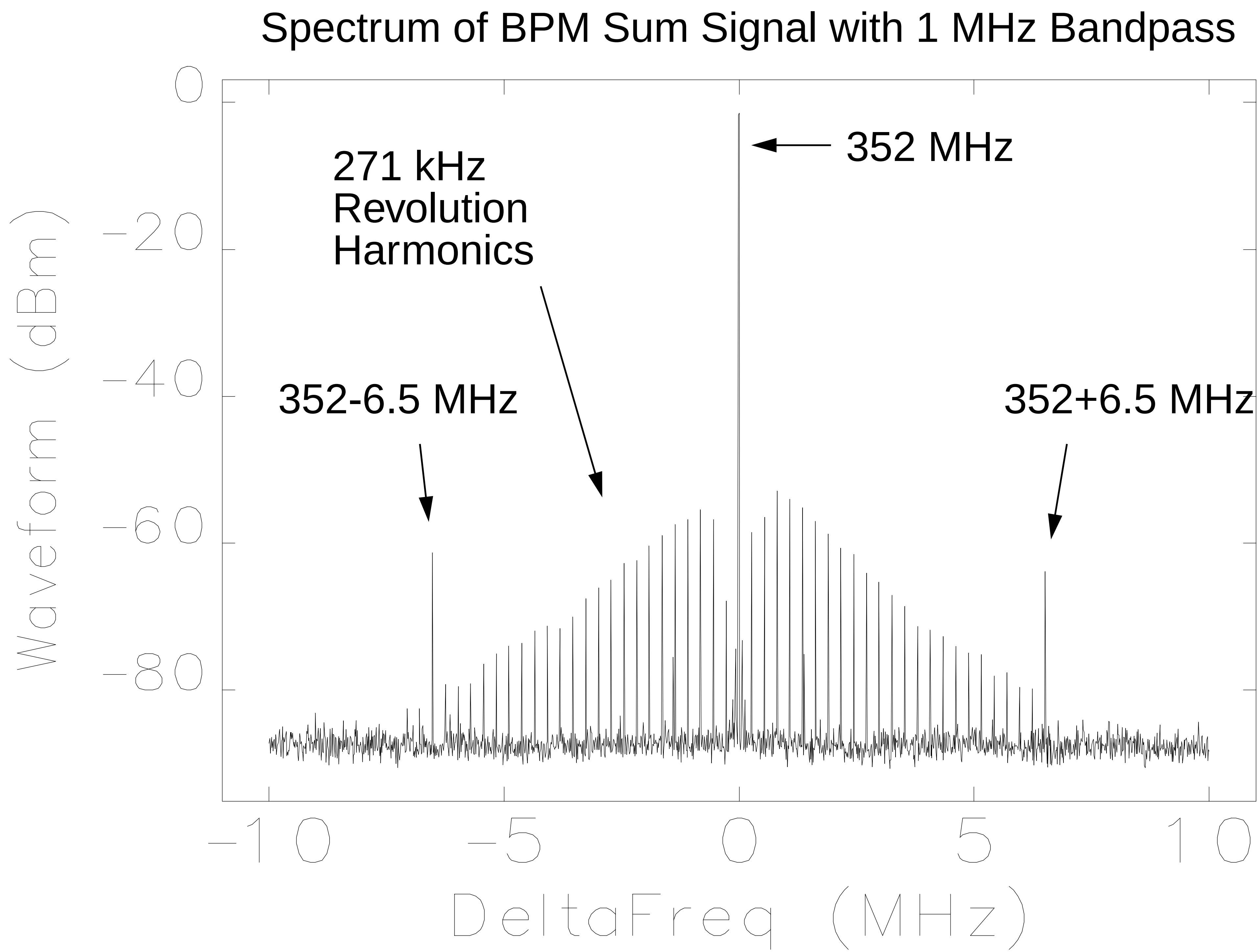
- In partnership with the CATs, foster and develop a robust Independent Investigator (II) program.

Storage ring timing status, capabilities and issues

Glenn Decker

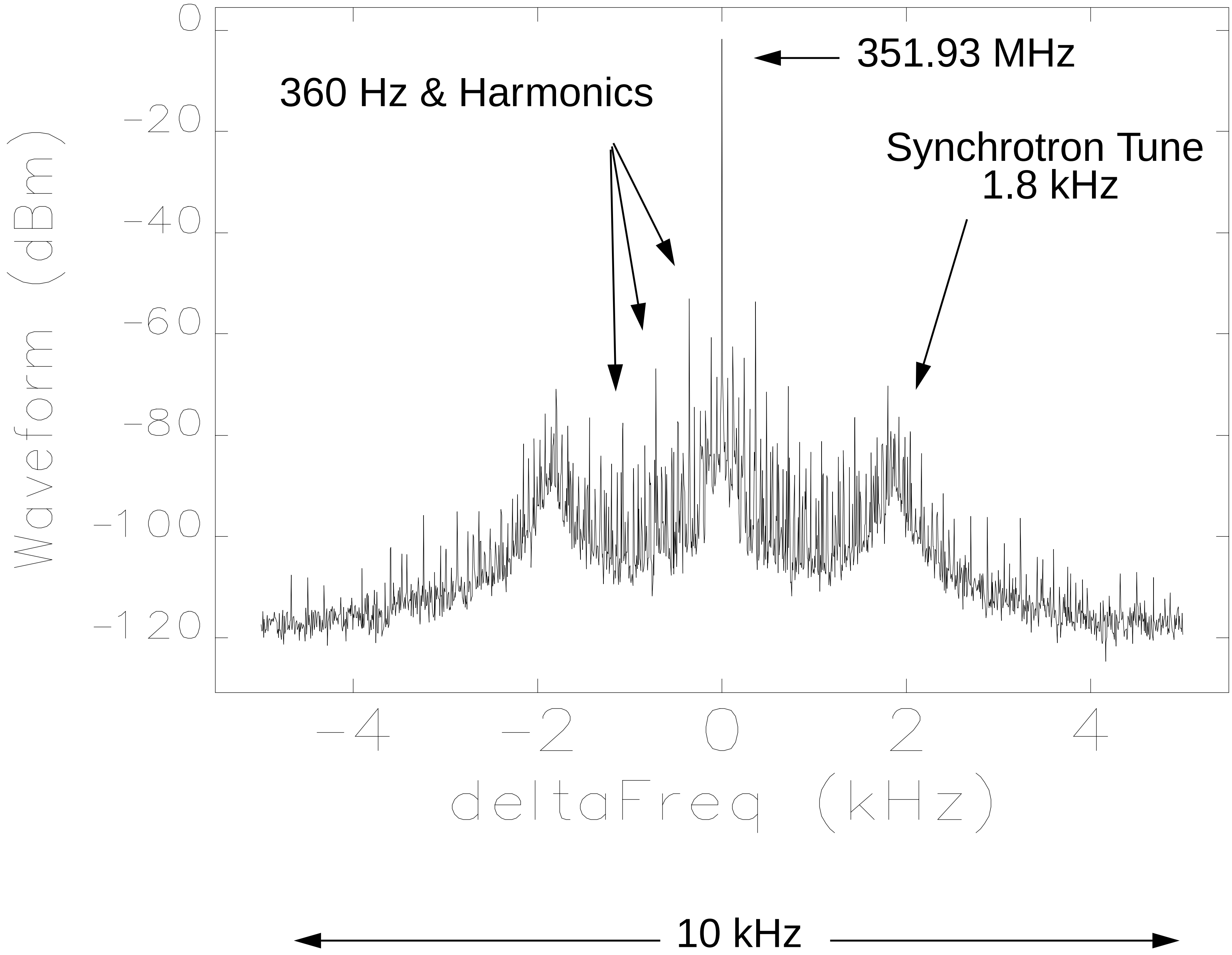
24 -1 singlets fill pattern



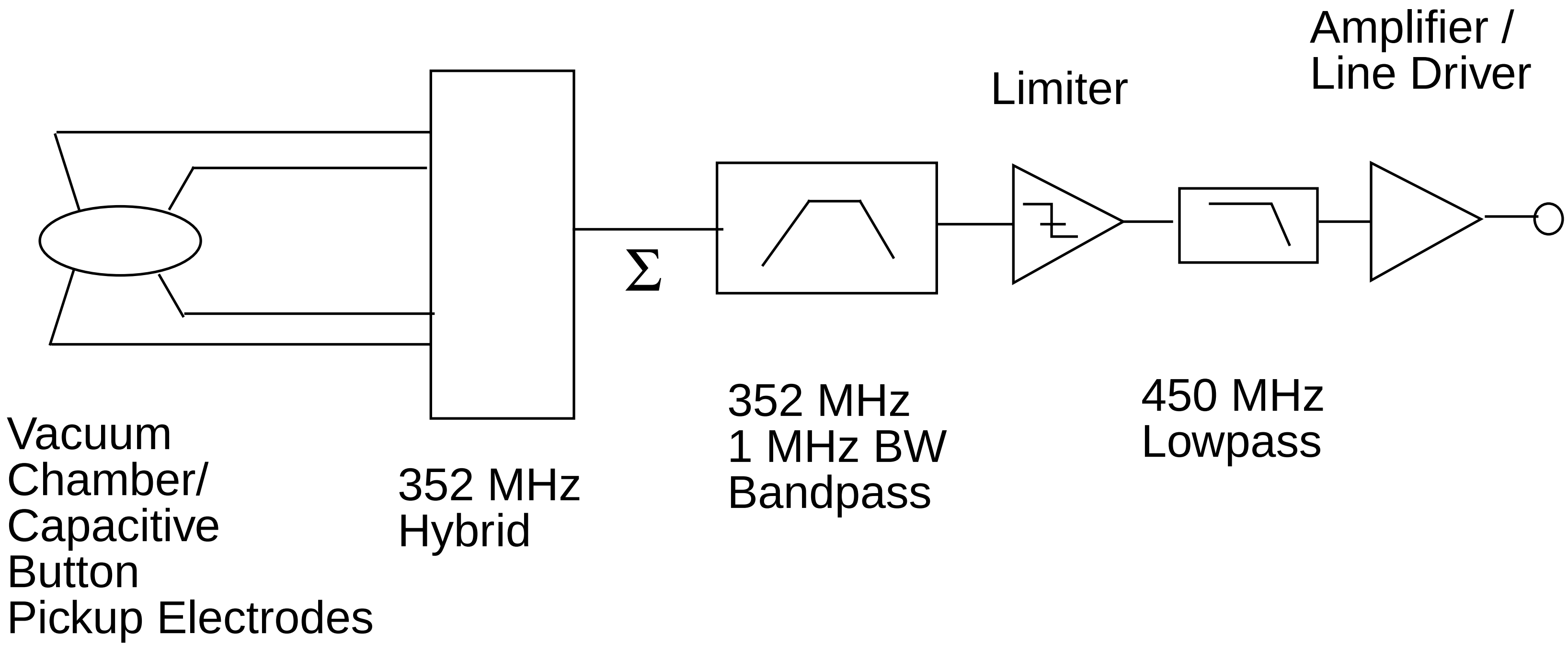


Spectrum of BPM Sum Signal with 1 MHz Bandpass
352 MHz
271 kHz
Revolution
Harmonics
352-6.5 MHz
352+6.5 MHz
0
−20
−40
−60
−80
Waveform (dBm)
−10
−5
0
5
10
DeltaFreq (MHz)

Phase Noise on Beam



A prototype beam-derived 352 MHz clock has been provided to 7-ID for the purpose of phase locking a laser to the beam. I think it works:



Points to keep in mind:

The accelerator’s longitudinal phase space satisfies a pendulum equation; the small amplitude resonance frequency is about 1.8 kHz. This is affected by the RF voltage, which is typically 9.4 MV - higher voltage increases the synchrotron tune. There is a spread in this tune - different particles have different tunes.

The dominant source of phase noise is the 100 kV high voltage power supply which supplies the accelerating voltage to the high power klystrons. 360 Hz and harmonics are prevalent. The fifth harmonic of this unfortunately is exactly 1.8 kHz.

Harmonics of 60 Hz are present, but smaller than the 360 Hz lines.

The “synchronous phase” is determined by the beam energy loss per turn - it has a weak dependence on insertion device gap - mostly only those with very small gaps that change a lot. Nominal energy loss per turn is 5.5 MV with all gaps open; approx. 6.5 MV with all gaps closed to minimum gap.

Every time the “decker distortion” takes place, the rf frequency has to change by several hundred Hz to compensate.

The rf frequency tracks the earth tide, which has 5 Hz p-p amplitude and approx. 12 hour period resulting from the moon passing overhead / underhead.

Longitudinal phase / energy oscillations couple directly into horizontal beam motion as determined by the “dispersion function”. Different energy particles follow slightly different horizontal orbits. The low emittance accelerator configuration has non-zero dispersion at the ID source points, unlike the original high emittance. The amount of horizontal motion due to phase noise is typically only a few microns rms, to be compared with the 0.25 mm rms horizontal beam size.

Fast coupled-bunch instabilities can occur such that different bunches will arrive at different phases. We try not to let that happen. These things are typically driven by narrow band higher order rf cavity modes. With 154 nS bunch spacing this has not been a problem.

High single bunch current induces microwave instability. In particular, longitudinal shape oscillations of a single bunch with associated harmonics of the synchrotron tune develop. An increase in energy spread is also seen. We typically cannot run with more than about 4.5 to 5 mA in a single bunch. The small aperture insertion device vacuum chambers are the culprit causing this effect.

Horizontal and vertical beam stability above about 30 Hz is not corrected. We have about 10 microns rms motion integrating up to 750 Hz. It only gets worse if you integrate up to the 3.25 MHz bunch repetition Nyquist frequency. We sample the beam once per turn (271 kHz), and the noise in the 135 kHz band is near 20 microns rms in both horizontal and vertical, comparable to the vertical beam size.

MHATT-CAT Overview

Roy Clarke*, University of Michigan
royc@umich.edu

*Director, Michigan/Howard/Bell Labs Collaborative Access Team (MHATT-CAT)

Workshop on Ultrafast Science,
Advanced Photon Source, June 4, 2002

MHATT-CAT Sector 7

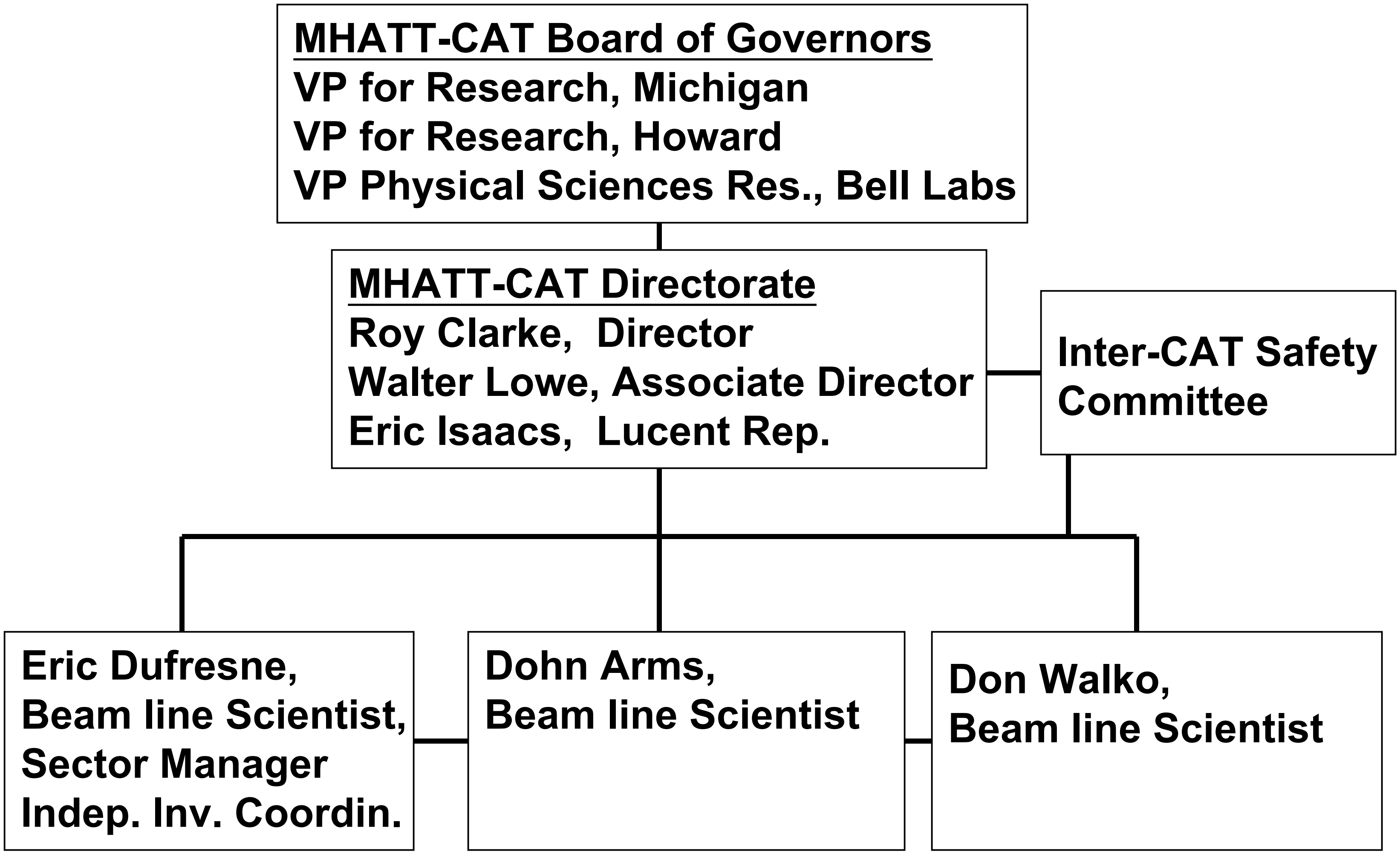
Center for Real-time X-ray Studies

MHATT-CAT's mission:
To develop a productive, open-access center for world-class research in x-ray science exploiting the unique characteristics of the APS, especially timing and brilliance.

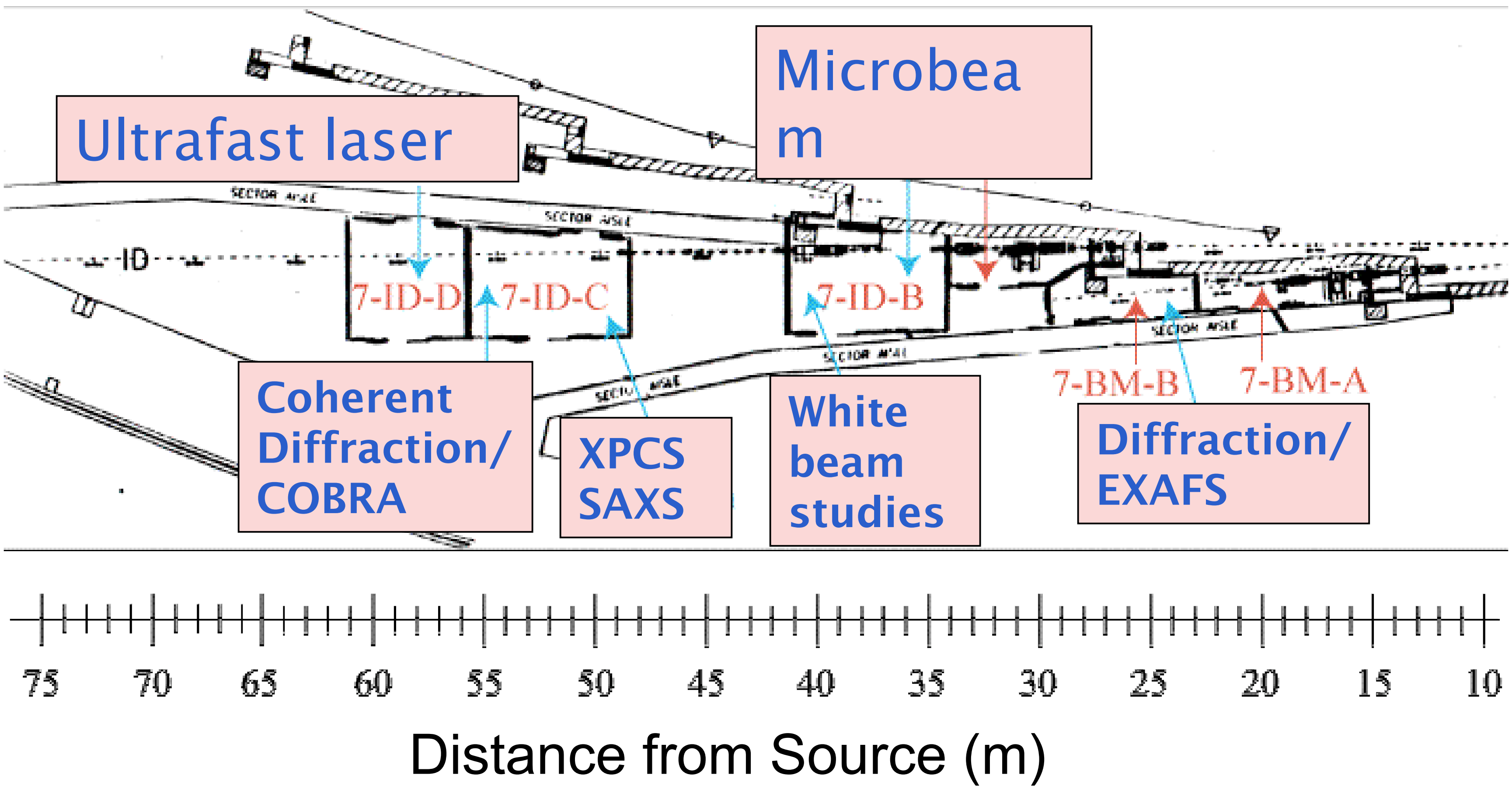
Collaborative Partnership:

- **University of Michigan, Ann Arbor, Michigan**
- **Howard University, Washington DC**
- **Lucent Technologies/Agere, Bell Labs, Murray Hill, NJ**

MHATT-CAT Management Structure



MHATT-CAT Sector 7 Floor Plan



Independent Investigator Access

MHATT-CAT is committed to making its facilities widely available

ID beamline is declared operational and will begin Independent Investigator operations in October 2002

BM beamline will be open for II access immediately upon commissioning (~summer 2003)

Science Program Highlights

MHATT-CAT has done seminal work in four new fields of x-ray science:

- ultrafast diffraction
- microbeam diffraction/XFS
- X-ray photon correlation spectroscopy
- Coherent Bragg Rod Analysis (COBRA)

The CAT structure is key to this science

The MHATT-CAT collaboration brings to APS:

- Expertise of leading program in Ultrafast Optics (**NSF Frontiers of Physics Center: FOCUS)**
- Sub-ps timing and synchronization
- ultrafast detector expertise (streak cameras)
- complementary state-of-the-art x-ray focusing techniques:
    - pre-figured K-B mirrors (ORNL/Howard)
    - zone-plates (e-beam lithography)

        aiming to reach ~10's nm scale

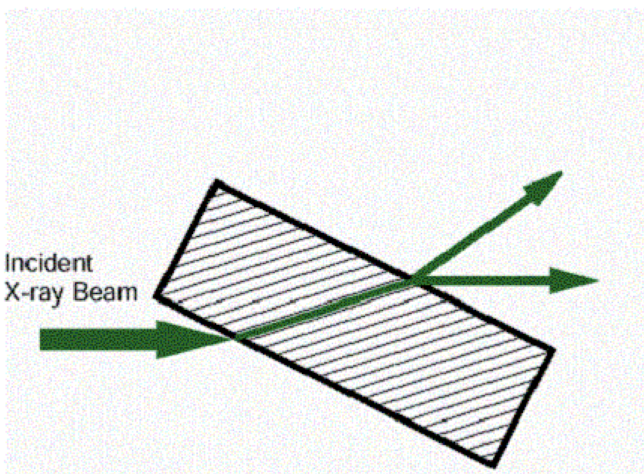
- Unique method for direct structure determination of 2D systems: MHATT/PNC/BESSERC collaboration
- Extremely broad range of materials research
- Culture of student education and training

MRC 2: Ultrafast x-ray diffraction at MHATT-CA

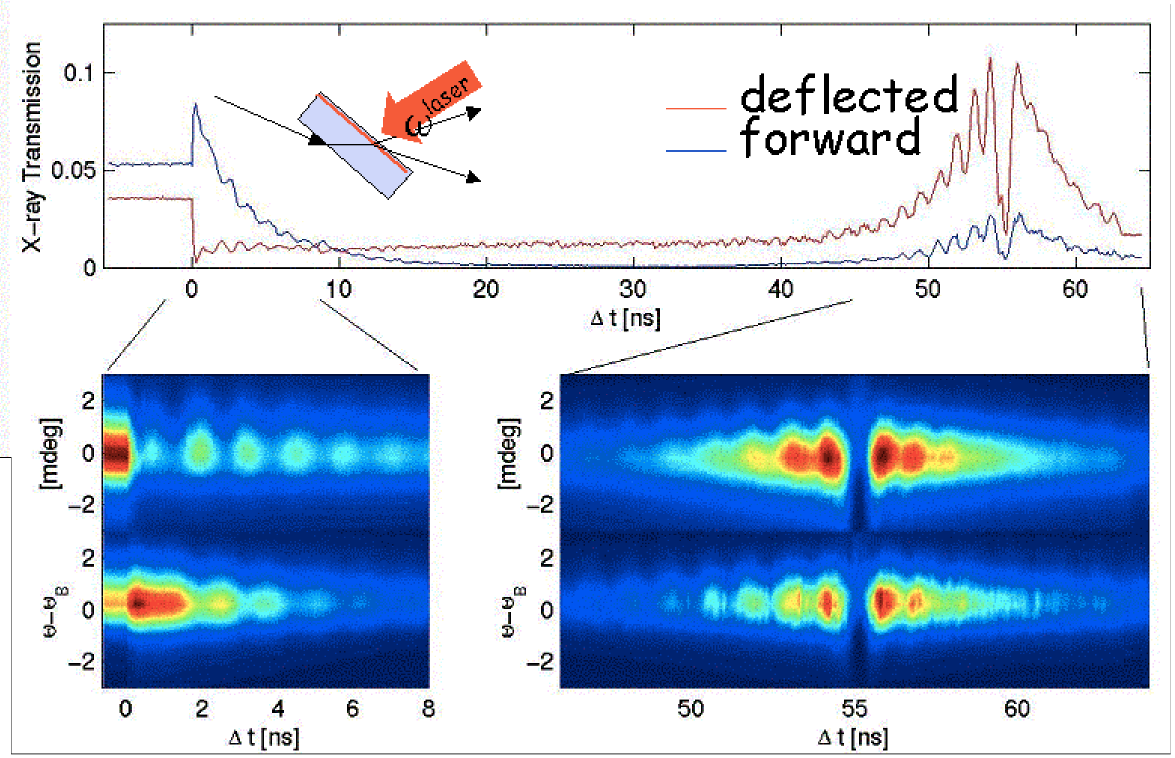
NATURE | VOL 413 | 25 OCTOBER 2001

Coherent control of pulsed X-ray beams

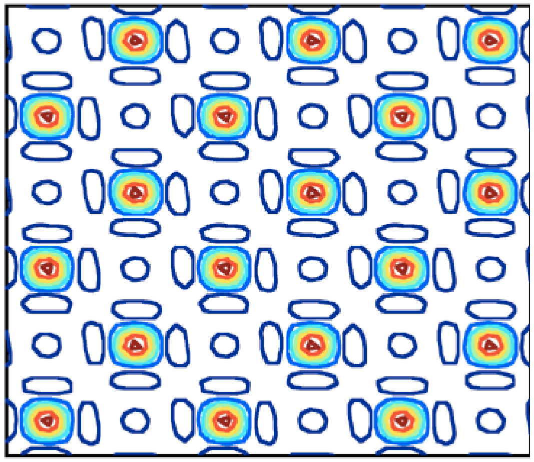
M. F. DeCamp*, D. A. Reis*, P. H. Bucksbaum*, B. Adams†,
J. M. Caraher*, R. Clarke*, C. W. S. Conover‡, E. M. Dufresne*, R. Merlin*,
V. Stoica* & J. K. Wahlstrand*



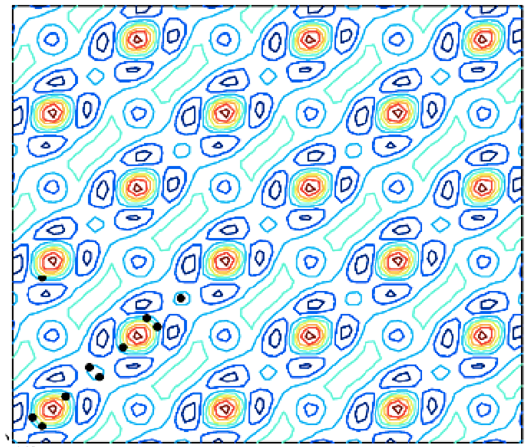
Ultrafast x-ray switch uses coherent acoustic phonons



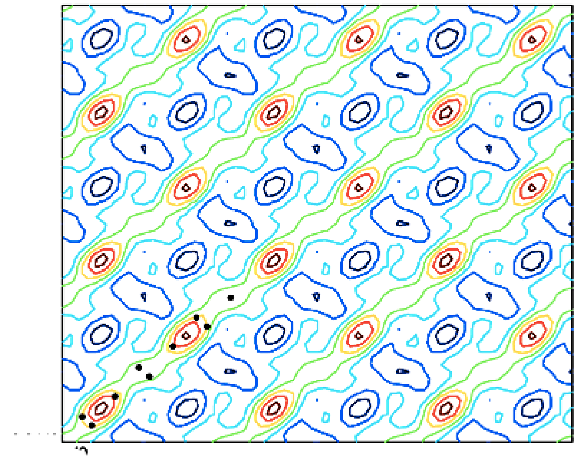
Mapping heterostructure interfaces



GaAs



5^{th} ML

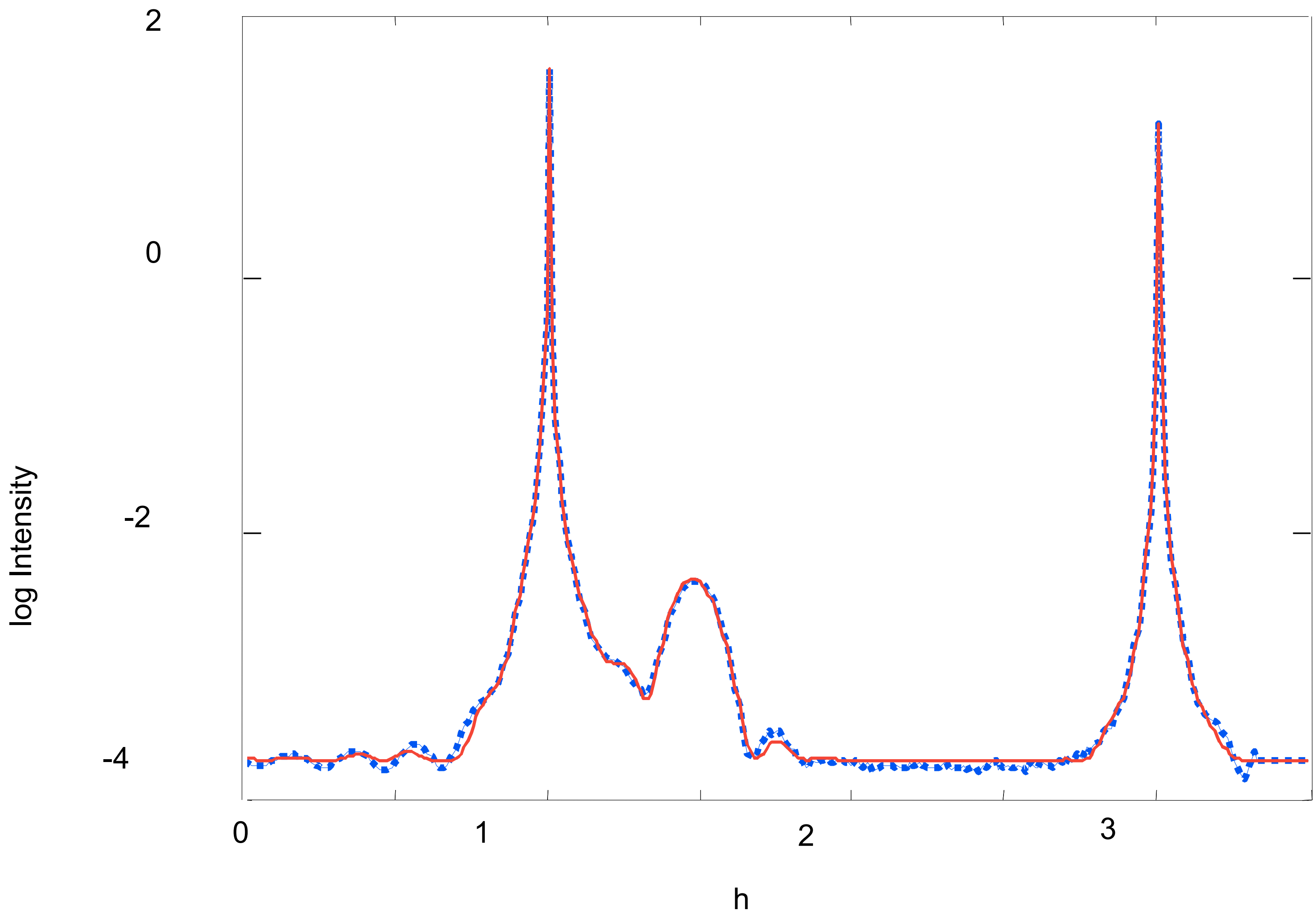


17^{th} ML

Coherent Bragg Rod Analysis (COBRA) electron density maps of epitaxial Gd_2O_3 on GaAs

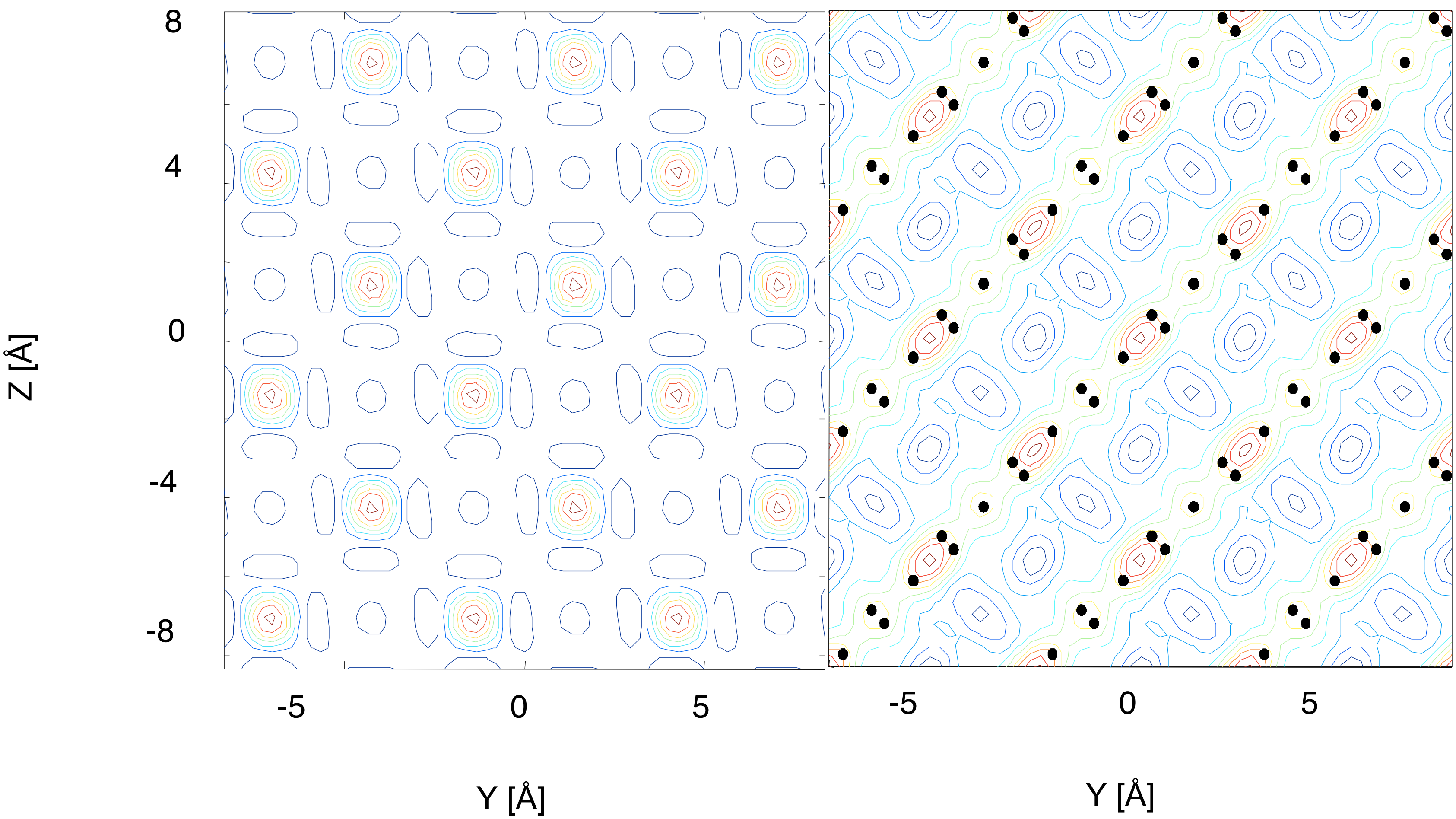
Yizhak Yacoby, Associate Member of MHATT-CAT
Ron Pindak, MHATT-CAT and NSLS
Roy Clarke, MHATT-CAT and U Michigan
Ed Stern, PNC-CAT and U Washington

Now extending collaboration to ferroelectric thin films, adding:
Xiaoqing Pan (Michigan, MHATT) and Brian Stephenson (MSD/BESSERC)



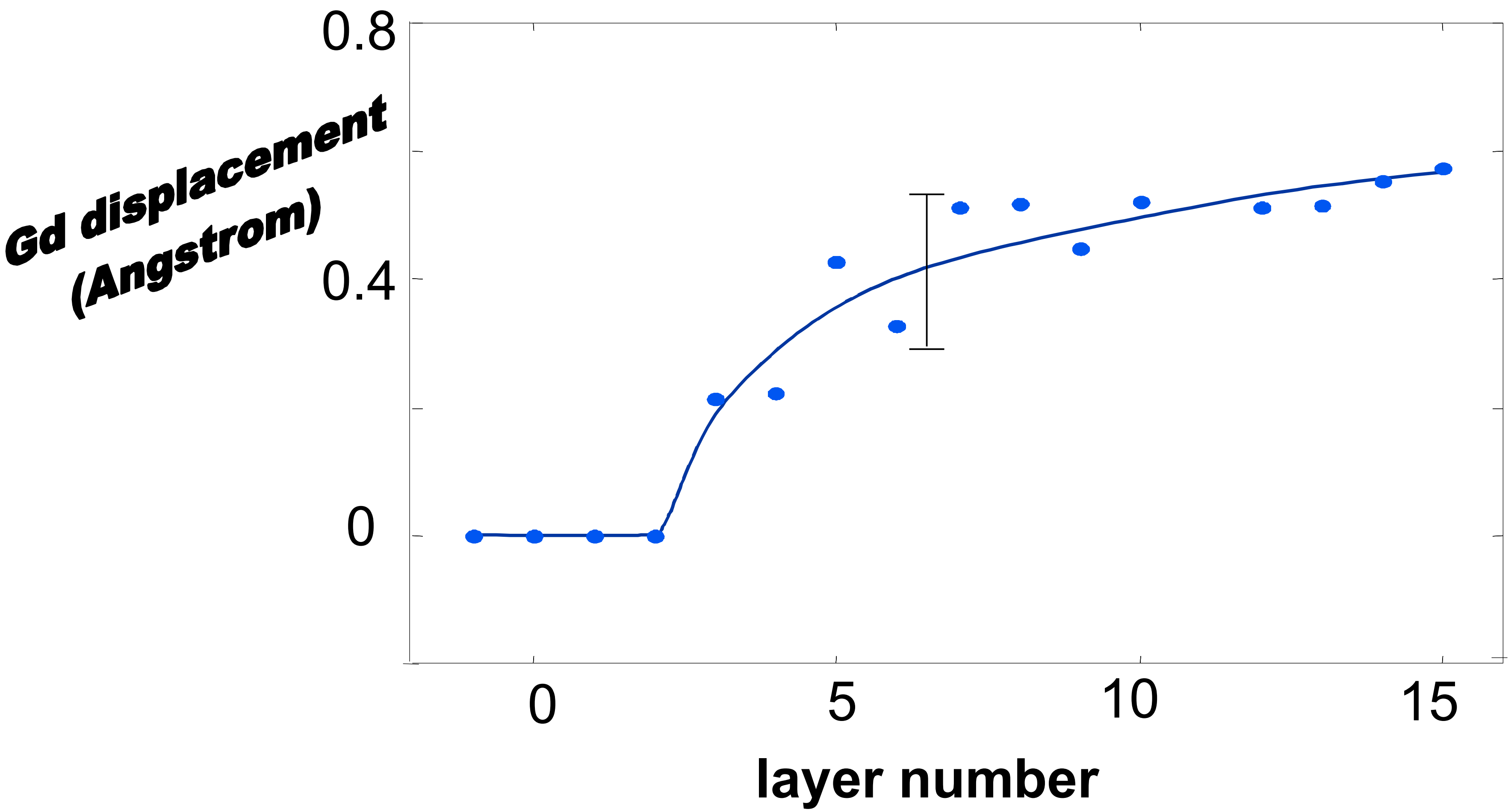
Bragg rod data obtained by COBRA technique
Blue trace (dots): experimental data; red trace: structure factor obtained from coherent Bragg rod analysis.

COBRA Map of GaAs – Gd_2O_3 interface



9 ML below (GaAs)   8ML above Gd_2O_3

Gd positions lock in to Ga/As close to interface



Confirms passivation function of Gd oxide epilayer

Examples of Independent Investigator Research at Sector 7

- Relaxation dynamics of magnetic polymers using XPCS (collaboration with Ford Research, Dearborn, MI)

- Micro-fluorescence mapping of mineral intrusions (Dan Core, Geosciences Dept., Michigan)

- Development of Li-metal x-ray lenses (Nino Pereia, Ecopulse Inc., Fairfax, VA)

- Study of failure mode precipitates in concrete (Dick Livingston, Federal Highway Commission)

- ……+ many more

All of these users new to the synchrotron field

Bend Magnet Line (7-BM) and Plans for future

Characteristics:

High critical energy (~ 20 keV)   continuous spectrum
Extends to very high energies (>100 kev)
Impressive brightness (10^{15} uu's)

Status

All major beam-forming components installed, collimating mirror and vertical focusing mirror to be added; Huber 6 circle available

Plans

User science requiring white beam (Laue, time resolved)
EXAFS and anomalous diffraction at high energy edges
Possibly bring a laser beam into BM-B for pump-probe
Biology interest in focused beam microfluorescence
Target for operational readiness of BM line: Summer 2003

THE DYNAMICS OF MAGNETORHEOLOGICAL ELASTOMERS STUDIED BY SYNCHROTRON RADIATION SPECKLE ANALYSIS

William F. Schlotter, Codrin Cionca, Sirinivas S. Paruchuri, Jevne B. Cunningham, Eric Dufresne, Steve B. Dierker, Dohn Arms and Roy Clarke
University of Michigan, Ann Arbor

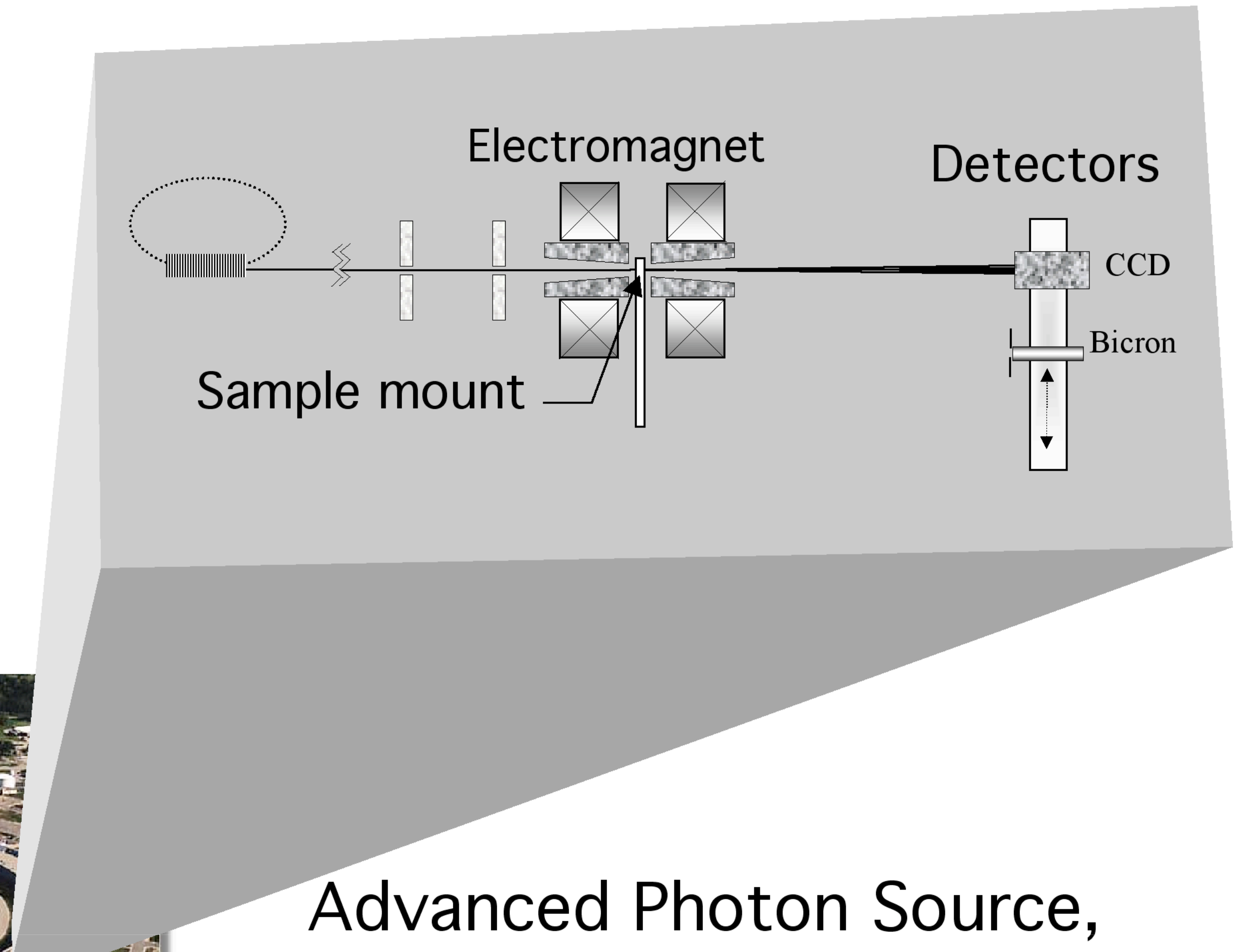
John M. Ginder and Mark E. Nichols
Ford Motor Company, Research Laboratory, Dearborn, MI

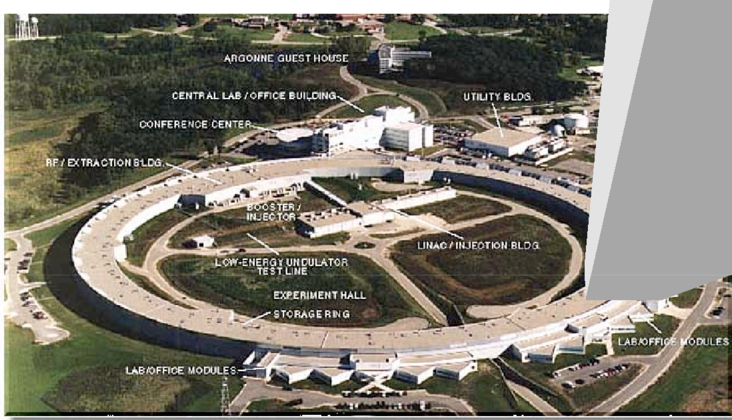
- Novel method for probing dynamics of MR materials
- X-ray photon correlation spectroscopy
- Small angle x-ray scattering setup using high brilliance synchrotron radiation (coherent x-rays!)

Small Angle Scattering Setup

MR sample is rigidly mounted while direction of magnetic field is alternated

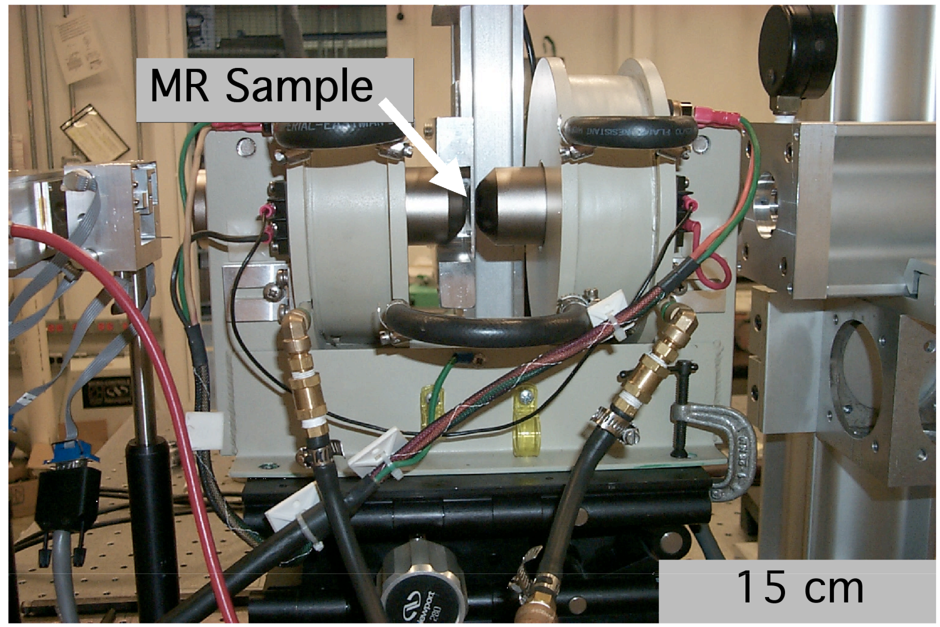
Coherence slits and guard slits allow only the transversely coherent component of the beam to pass.

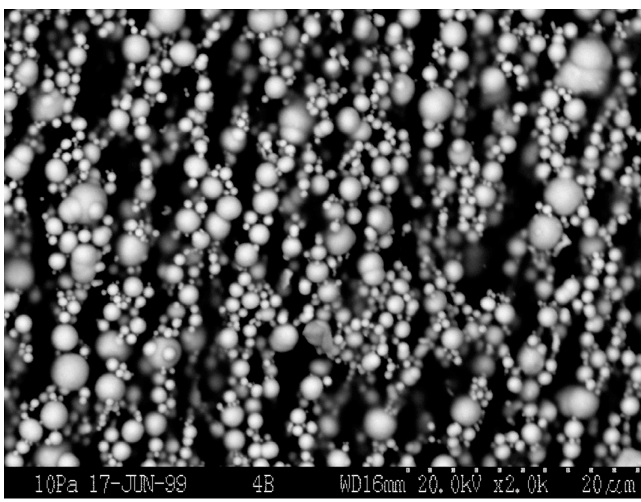




Advanced Photon Source,
Argonne National Lab
Illinois, USA

Magnet and Sample

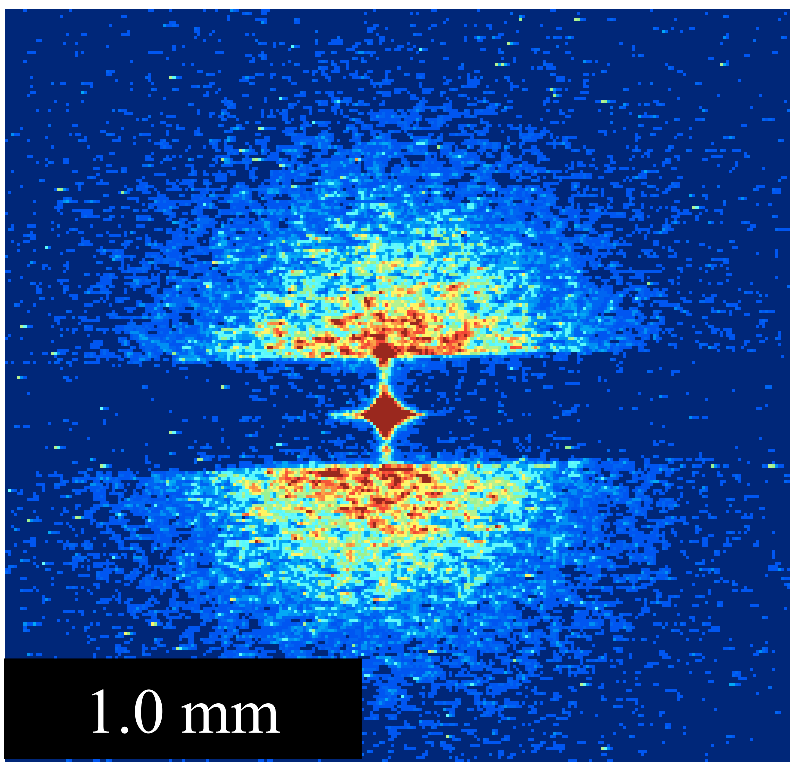




SEM of magnetic polymer

X-ray Speckle Movies

- CCD has efficient parallel data acquisition with msec capture time
- TI, low noise, high sensitivity CCD with 7.4μm pixel width in a 658 x 494 array was controlled by a 12-bit QMAX 650 camera.



•The ability to access very small scattering angles

•Enhanced statistics by averaging over annular regions of fixed *q*

•Time dependent speckle patterns were acquired as movies or 3D matrixes

•Obtained relaxation behavior of magnetic nano particle array embedded in polymer matrix.

Bringing in new Synchrotron Users

- Important goal of MHATT-CAT to build a diverse community of users at APS
- MHATT-CAT continues to expand its user base
  Past 2 years brought in new users from:

  - 9 universities
  - 4 national laboratories
  - 1 government dept. (Federal Highway Administration)
  - 3 industrial labs, and 3 small businesses

  delivering over 300 shifts of beam time to >50 new users

- Formal Independent Investigator program will begin **October 2002** on 7-ID (B-D). II plan in preparation.

Summary

- MHATT-CAT aims to provide advanced synchrotron facilities for time-resolved x-ray measurements

- The hard x-ray undulator beam line at Sector 7, APS, outfitted with a kHz femtosecond laser, offers unprecedented opportunities for ultrafast diffraction studies

- High brilliance X-ray optics and ultrafast detectors combine to make new science possible in the picosecond and sub-picosecond regime

- MHATT-CAT welcomes proposals for innovative science in the ultrafast area.

7ID-B overview

D. Walko

7-BM, MHATT-CAT

Science: Condensed Matter/Materials Science, Biology, Geology

Methods:
- time-resolved techniques
- EXAFS/general spectroscopy
- thin film/surface diffraction
- general diffraction

… for experiments where the brilliance and coherence of an ID line are not needed

Capabilities:

Monochromatic beam:
- Si(111) double-crystal mono, energy range 7-27 keV
- focussing via toroidal mirror, spot size ~25 μm x 100 μm

Pink beam:
- energy spectrum up to 20 keV
- focussing via toroidal mirror, spot size ~25 μm x 100 μm
- for Laue diffraction and experiments with special optics (e.g., multilayers)

White beam:
- full bending magnet spectrum
- for high-energy x-ray experiments

7-BM-A Optical Components

MHATT-CAT

- White beam slits

define beam size

- Lead collimator

- Collimating mirror

meridionally bent to collimate x rays, to make vertical divergence less than angular acceptance of mono

Rh coating, 3.2 mrad angle of incidence, for cutoff energy of 20 keV

- Monochromator

Si(111) double-crystal, similar to 7-ID mono

larger flux and higher energy resolution due to collimating mirror

- Focusing mirror

toroidal shape (with meridional bender) to focus horizontally to 0.1 mm and vertically to 0.025 mm

Rh coating and 3.2 mrad angle of incidence, like first mirror

- P6 shutter

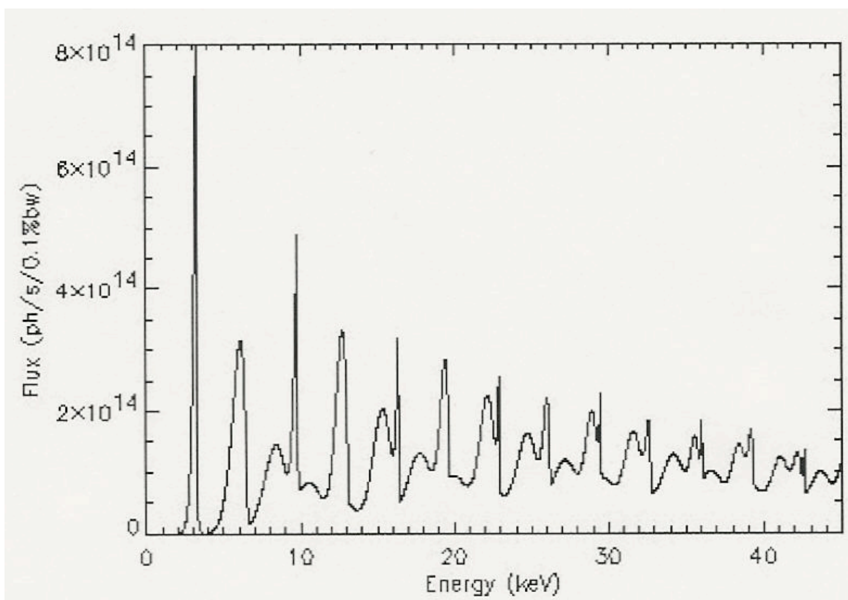
passes monochromatic or white beam while blocking other

Bending Magnet vs. Insertion Device

MHATT-CAT

	Horizontal Source Size	Vertical Source Size	Horizontal divergence	Vertical Divergence
Bending Magnet	145 μm	36 μm	6 mrad	47 μrad
Insertion Device	255 μm	4 μm	15.3 μrad	1.0 μrad

Comparison of ID and BM spectra



MHATT-CAT

Comparison of operating modes, 7-BM, MHATT-CAT

Mode	Energy Range	Bandpass	Power Density (W/mm^2)	Beam Size on Sample (mm)
Focused Mono Beam	4 – 20 keV	0.014% at 10 keV	1	0.025 (V) □ 0.1 (H)
Unfocussed Mono Beam	4 – 27 keV	0.076% at 10 keV	10^{-5}	4 (V) □ …
Pink Beam	5 – 20 keV	4	1.6□10^4	0.025 (V) □ 0.1 (H)
White Beam	5 – 100 keV	N/A	0.6	4 (V) □ …
7-ID beam (white beam)	5 – 100 keV	2.5%	200	0.65 (V) □ 2 (H)

MHATT-CAT

Why build a Bending Magnet line?

• Experiments where the brilliance and coherence of an ID line are not needed
• Experiments which need true white beam or pink beam (featureless spectrum)

Possible Examples:

- EXAFS/general spectroscopy
- thin film/surface diffraction
- general diffraction
- time-resolved techniques

Science: Condensed Matter/Materials Science, Biology, Geology

Frontiers in Optical Coherent and Ultrafast Science

The University of Michigan
Artes · Scientia · Veritas
1817

Ultrafast experiments at MHATT-CAT

Matt DeCamp

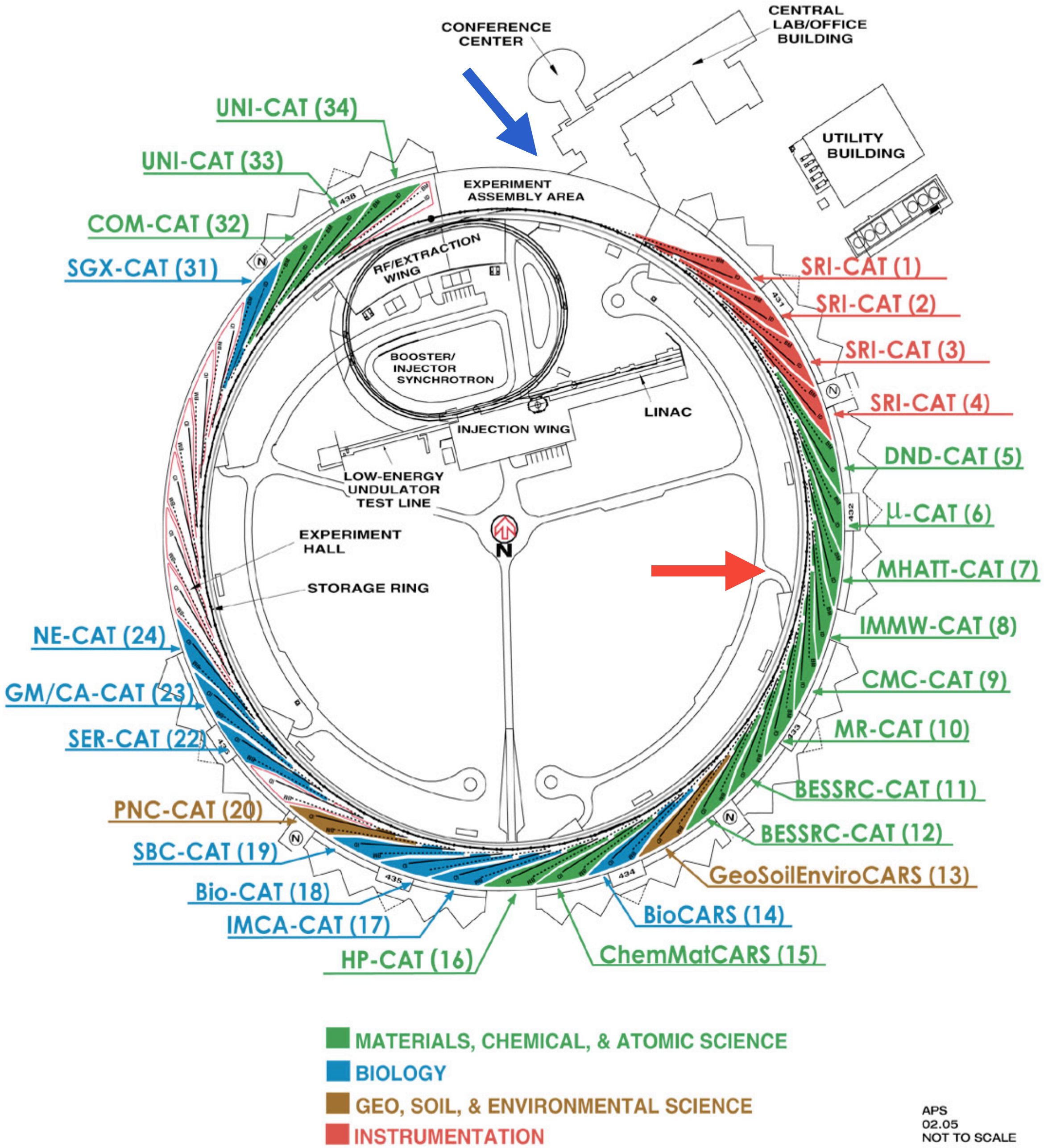
FOCUS Center, University of Michigan

MHATT-CAT
Center for Real-Time X-Ray Studies

Advanced Photon Source
Argonne National Laboratory

Outline

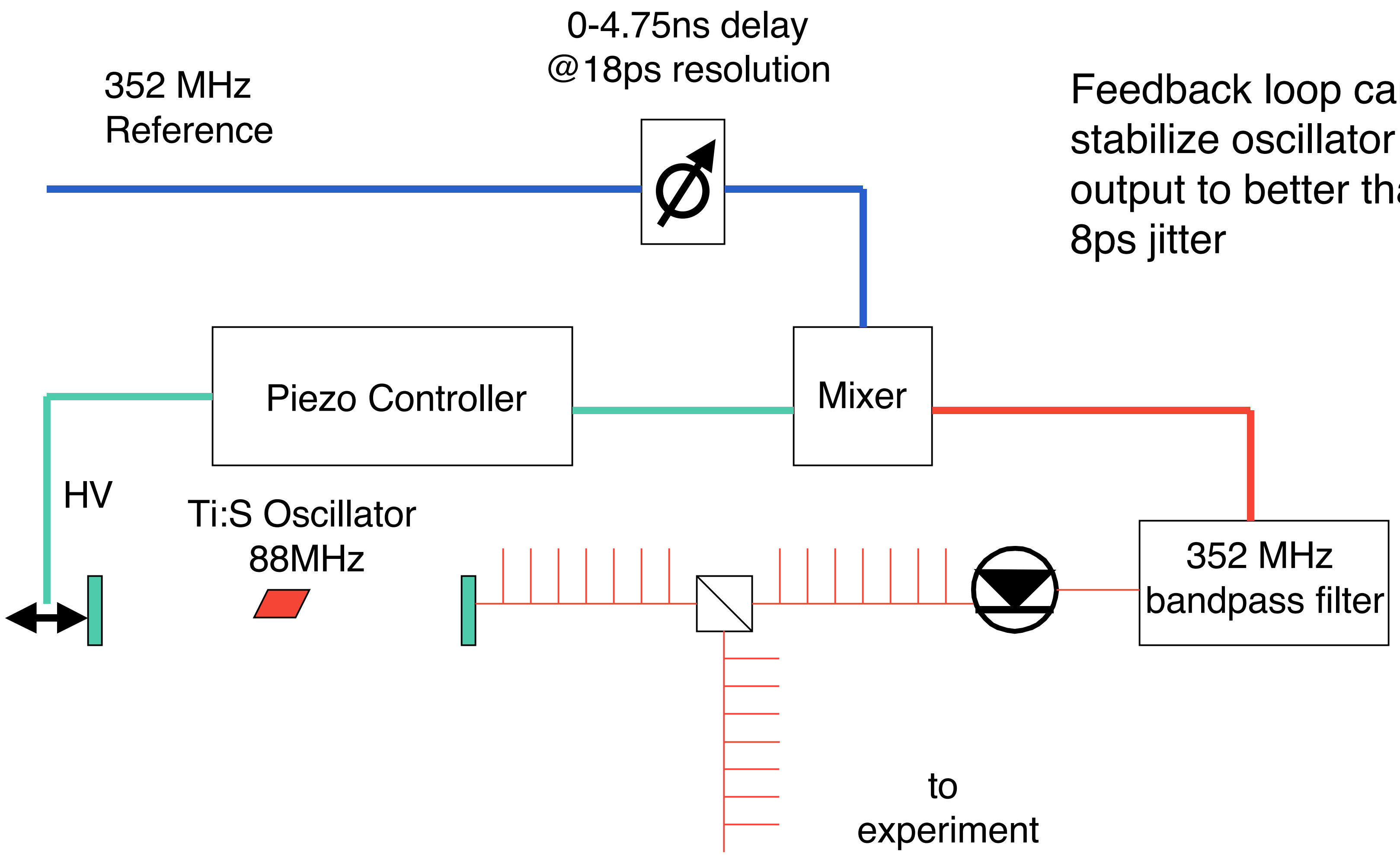
- MHATT-CAT specific issues for ultrafast time-resolved experiments
- Equipment available time-resolved studies
- Recent ultrafast time-resolved experiments at MHATT-CAT



352 MHz signal from control room which is locked to RF accelerators ~20ps timing jitter. Effective 100ps x-ray pulse.

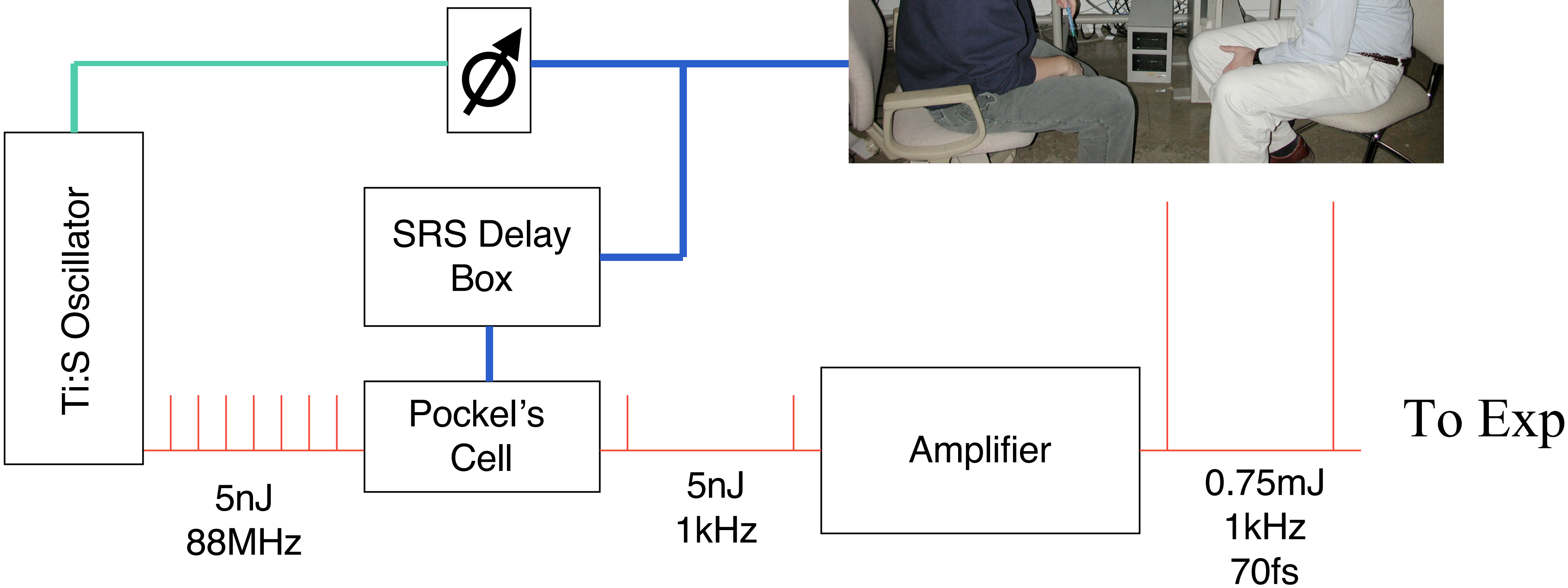
Upgrade summer 2002 Beam Position Monitor ~8ps timing jitter. Effective 80ps x-ray pulse.

Locking the oscillator



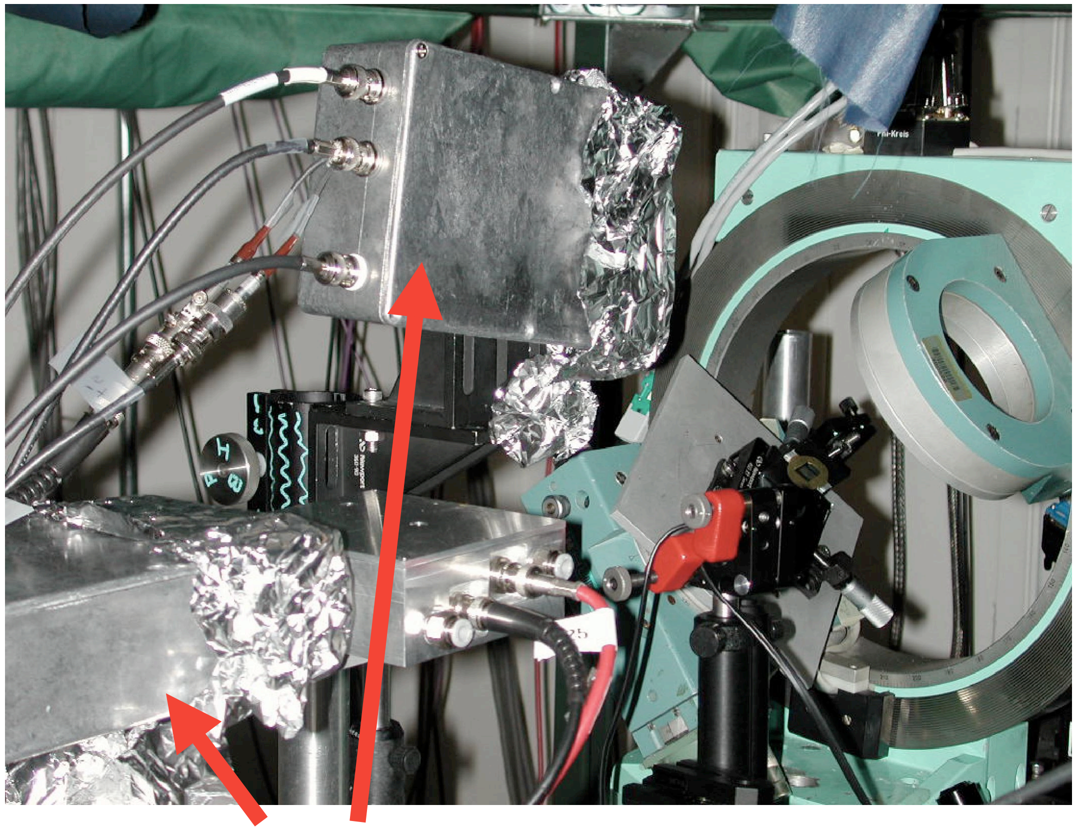
Feedback loop can stabilize oscillator output to better than 8ps jitter

Amplifier system and timing



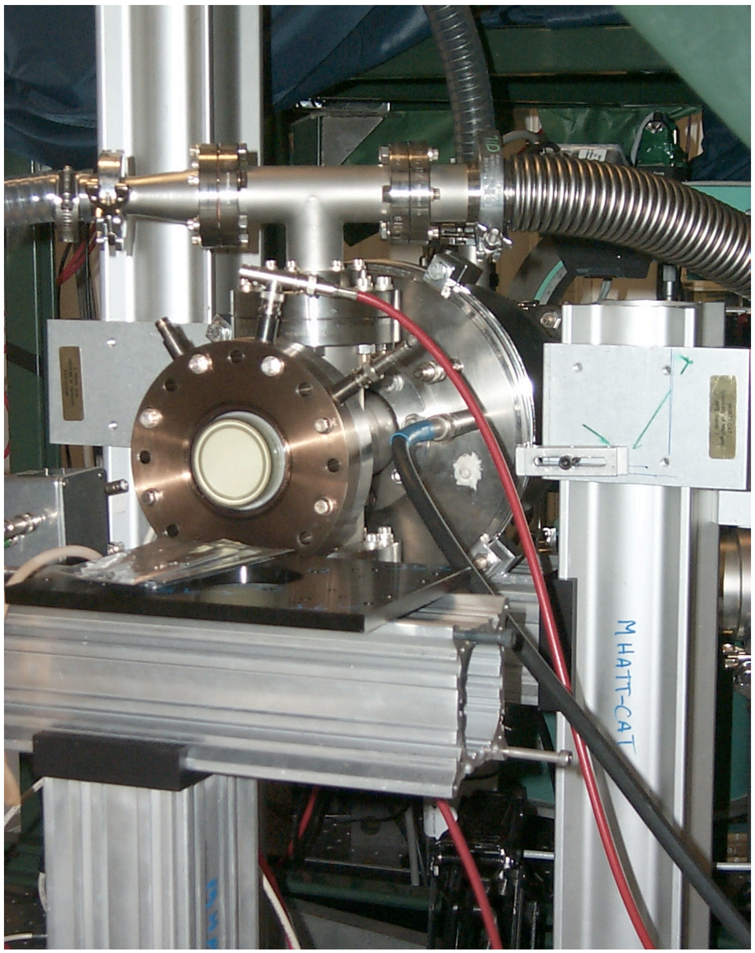
Time delays limited only by the amplifier repetition rate (-1 to 1ms)
Timing precision limited by the digital phase shifter (~18ps)

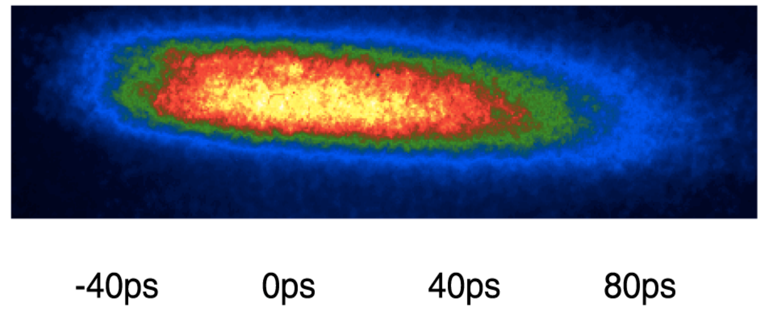
Time-resolved detectors (APD)



APD is fast enough to electronically gate a single x-ray pulse.
Timing resolution limited to x-ray pulse width (~100ps).
Use multiple APDs to sample many diffraction peaks simultaneously.

Time-resolved detectors (Streak Camera)

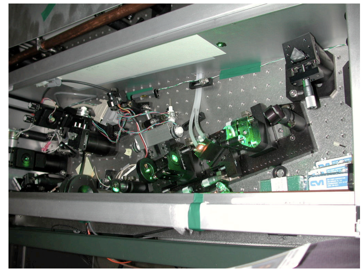


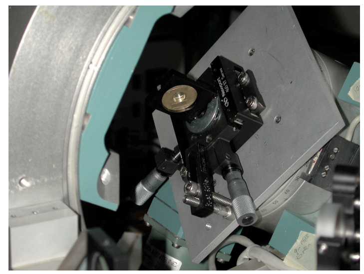


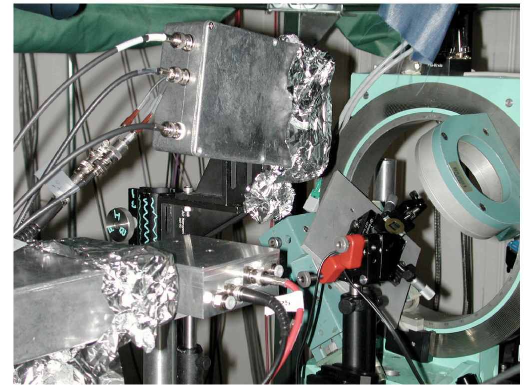
Streak camera has a time resolution of 1-2 ps

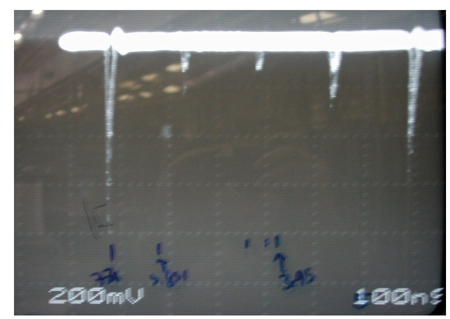
Permanent streak camera instillation date: Fall 2002

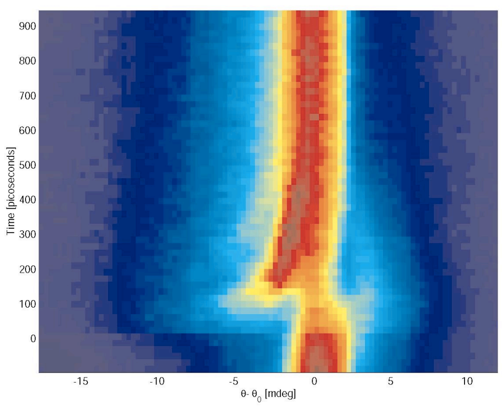
Data Acquisition











7ID-D

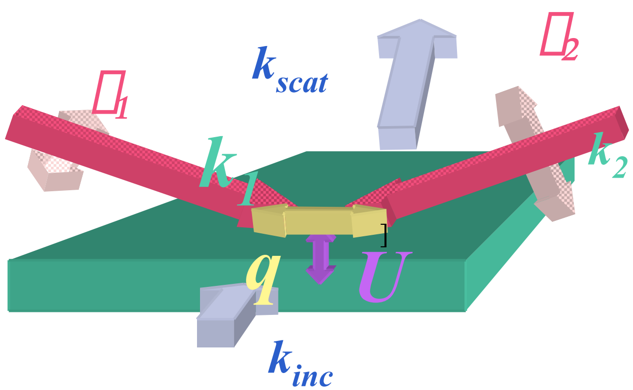
Last hutch on beamline
Allows for experimental setup at ‘all’ times
Possible ‘pink’ beam accessible

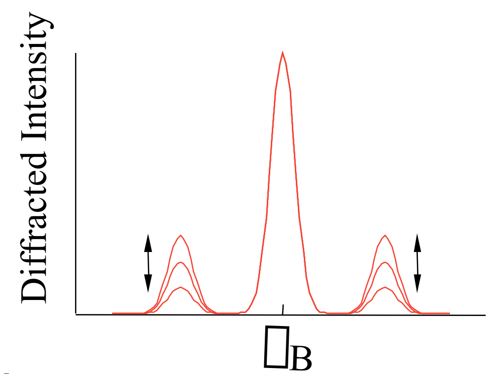
Motivation: an Ultrafast Bragg Switch for X-Rays

Goal: sub-picosecond control of x-ray pulses from synchrotrons.
Use coherent control of optical phonons to switch x-rays on and off in 1/2 phonon period

Bragg Side-bands

Transient Grating: Create coherent superlattice using optical phonons and change the Bragg condition



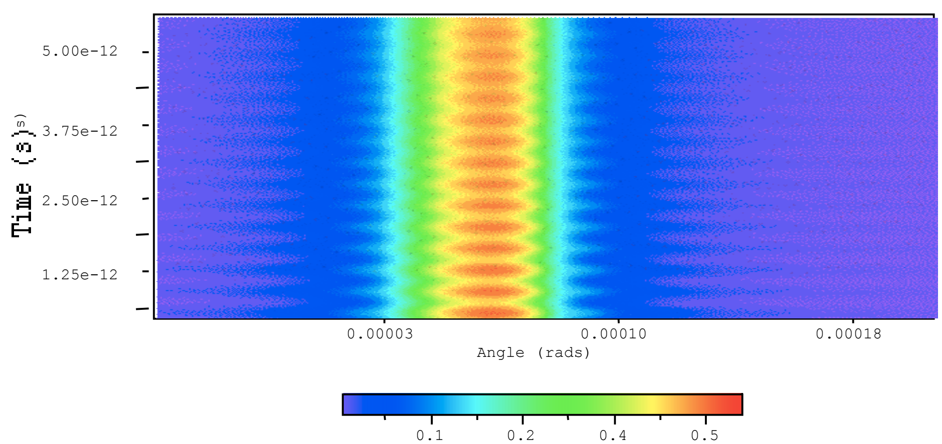


Symmetry breaking

Modify structure factor using “zero-wavector” coherent optical phonons

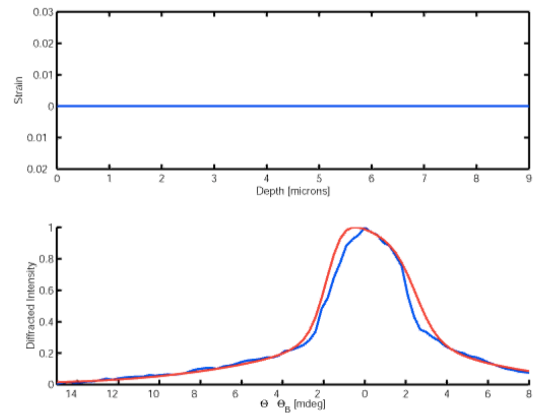
Lattice oscillation up to 10^{-2} have been measured using optical techniques
DeCamp et al. *PRB*, **64** 092301 (2001).
Hase et al. *PRL*, **88** 067401 (2002).

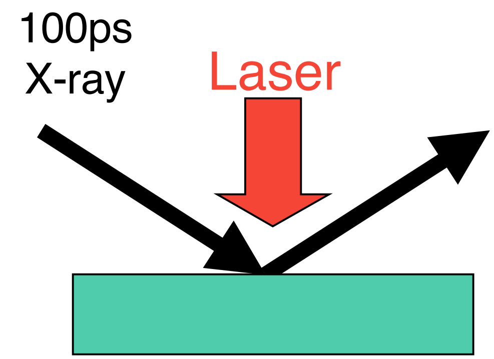
Early results in x-ray diffraction
Sokolowski-Tinton et al., CLEO (2002).



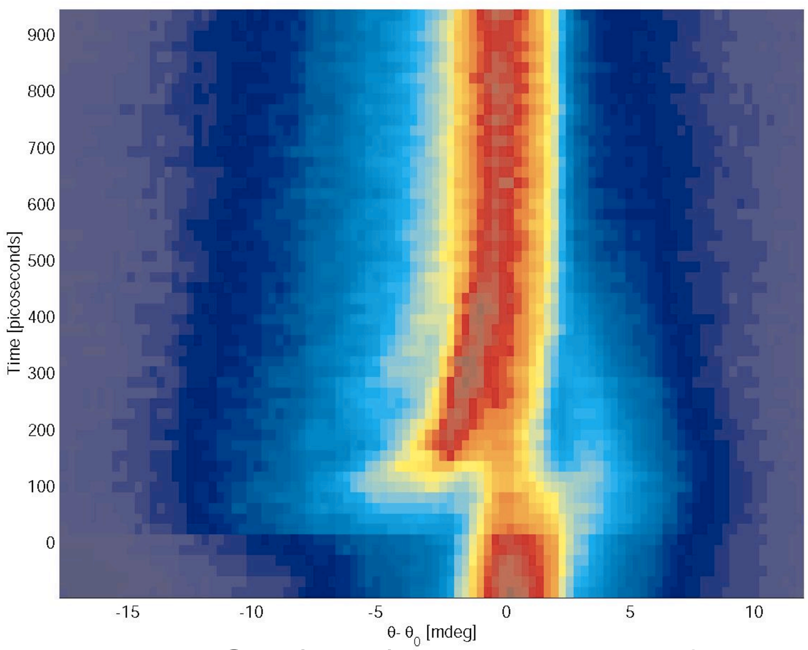
Time-resolved Bragg Diffraction

Phonon component underestimated by Thomsen model



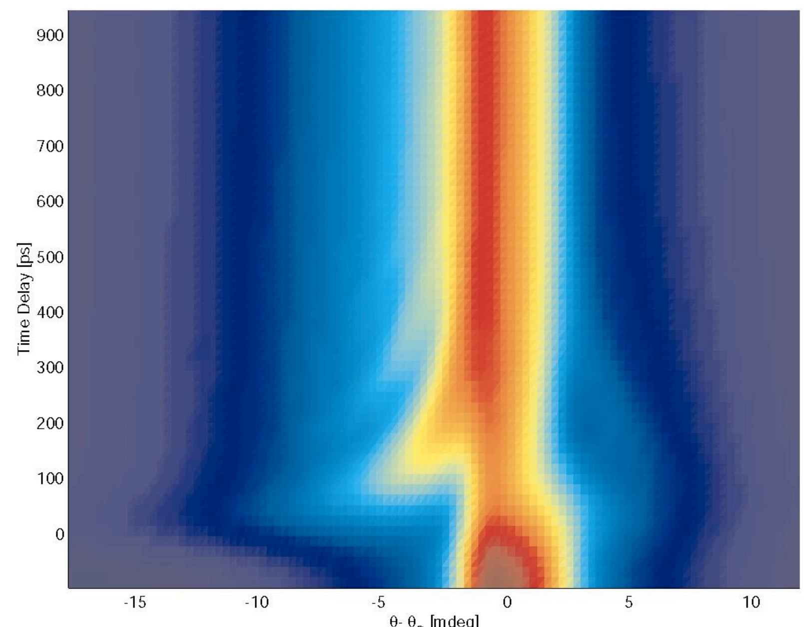


Data



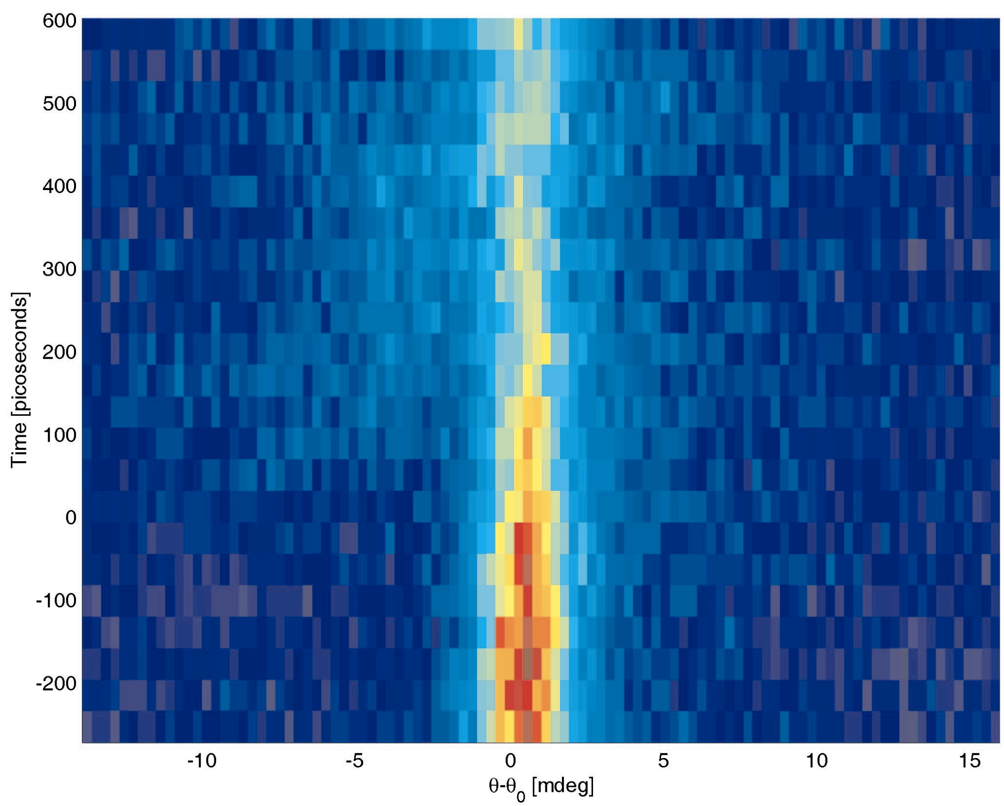
InSb (111), 840 nm, 70 fs,
~10 mJ/cm² laser pulse

Simulation



Reis *et al. PRL.*
86 3072 (2001)

Time-resolved 'forbidden' Bragg Diffraction



Reis *et al.* *PRL*, **86** 3072 (2001)

InSb (222), 840 nm, 70 fs, ~10 mJ/cm^2 laser pulse

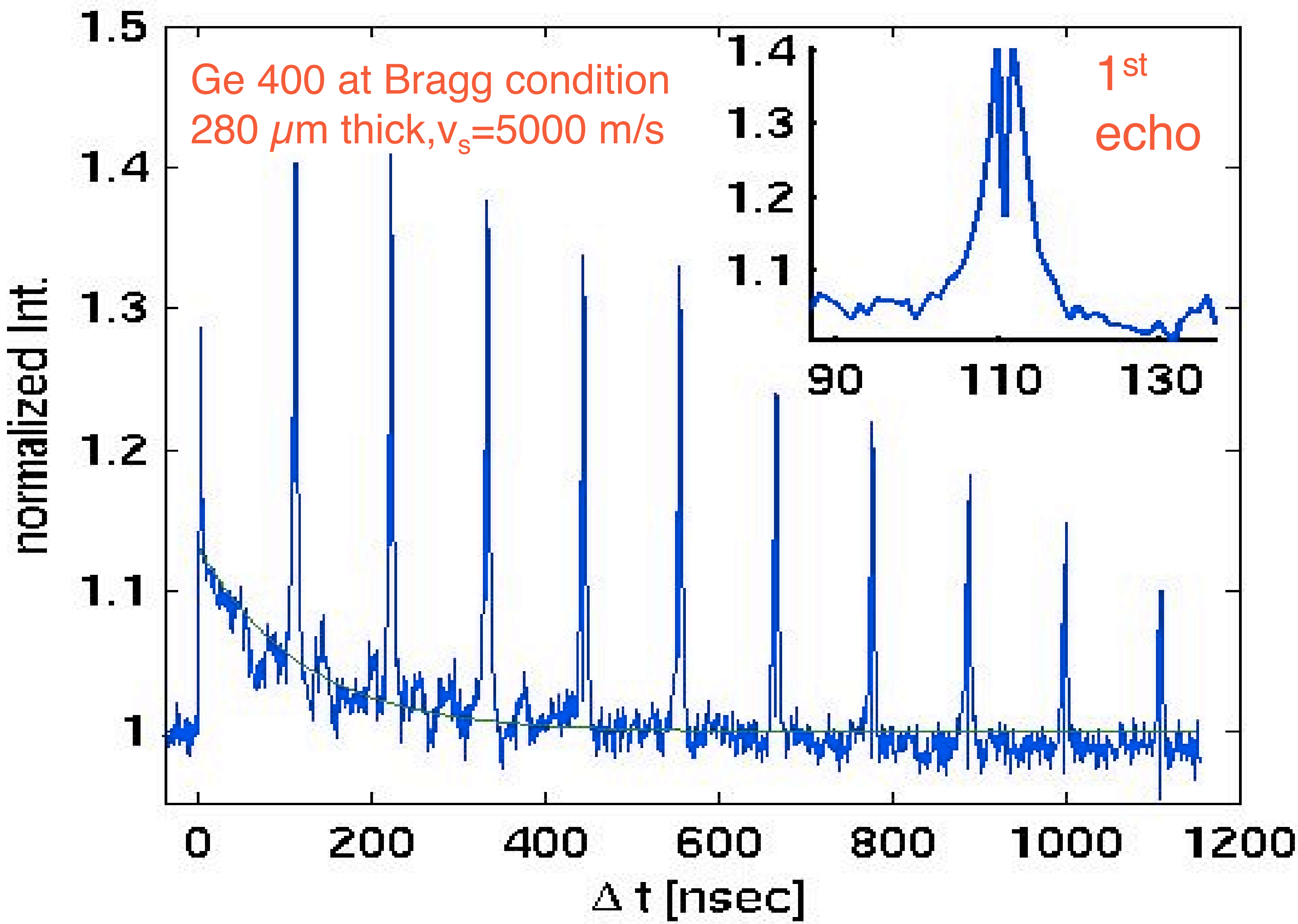
Utilize high brightness of APS to study structural dynamics with quasi-forbidden Bragg reflection.

Advantages of using 'forbidden' reflections:

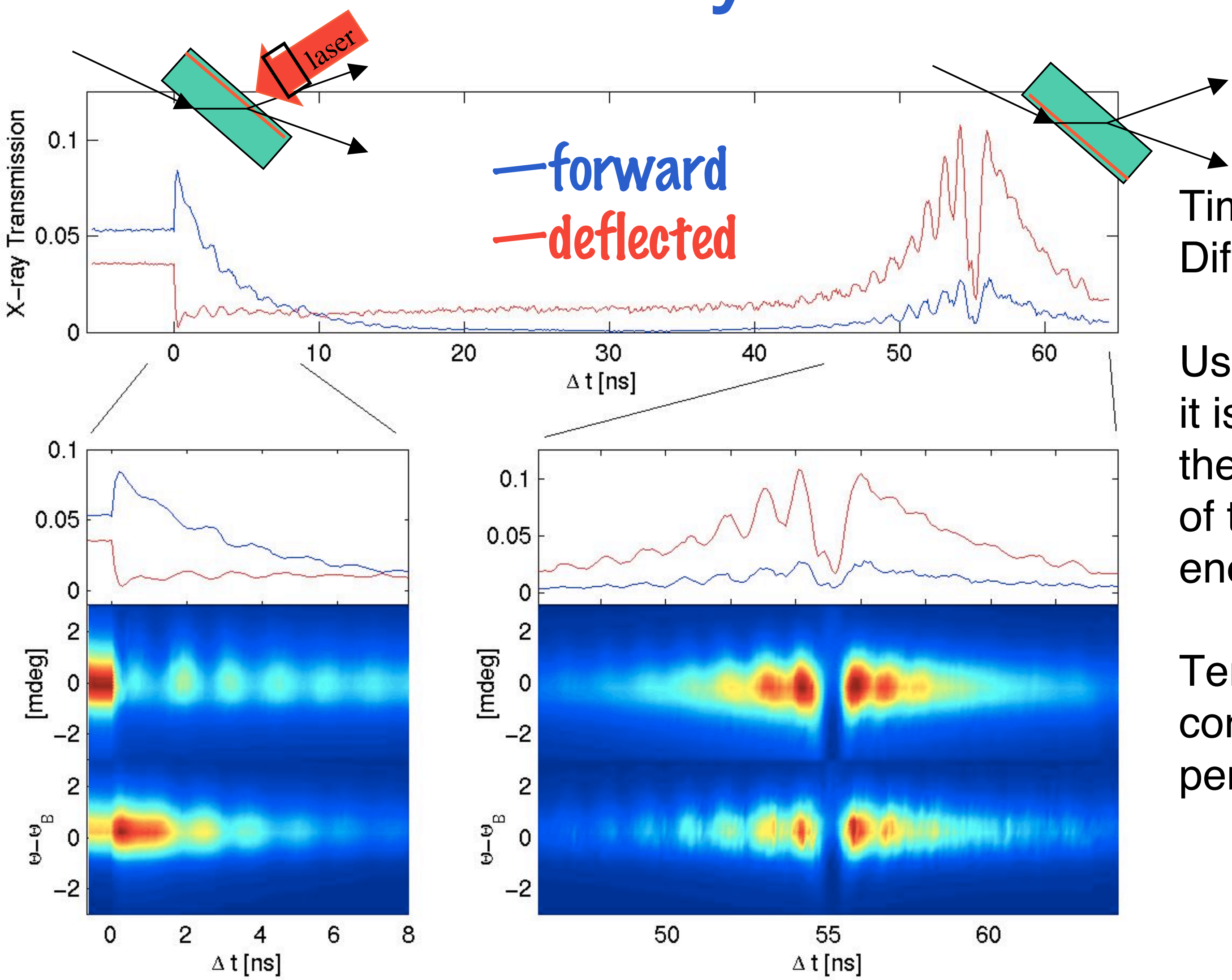
- Very sensitive to structural changes
- Very little background
- Large probe depth
- Wave vector selectivity

acoustic echoes & thermal diffusion

(l o n g time delays)



X-ray Control



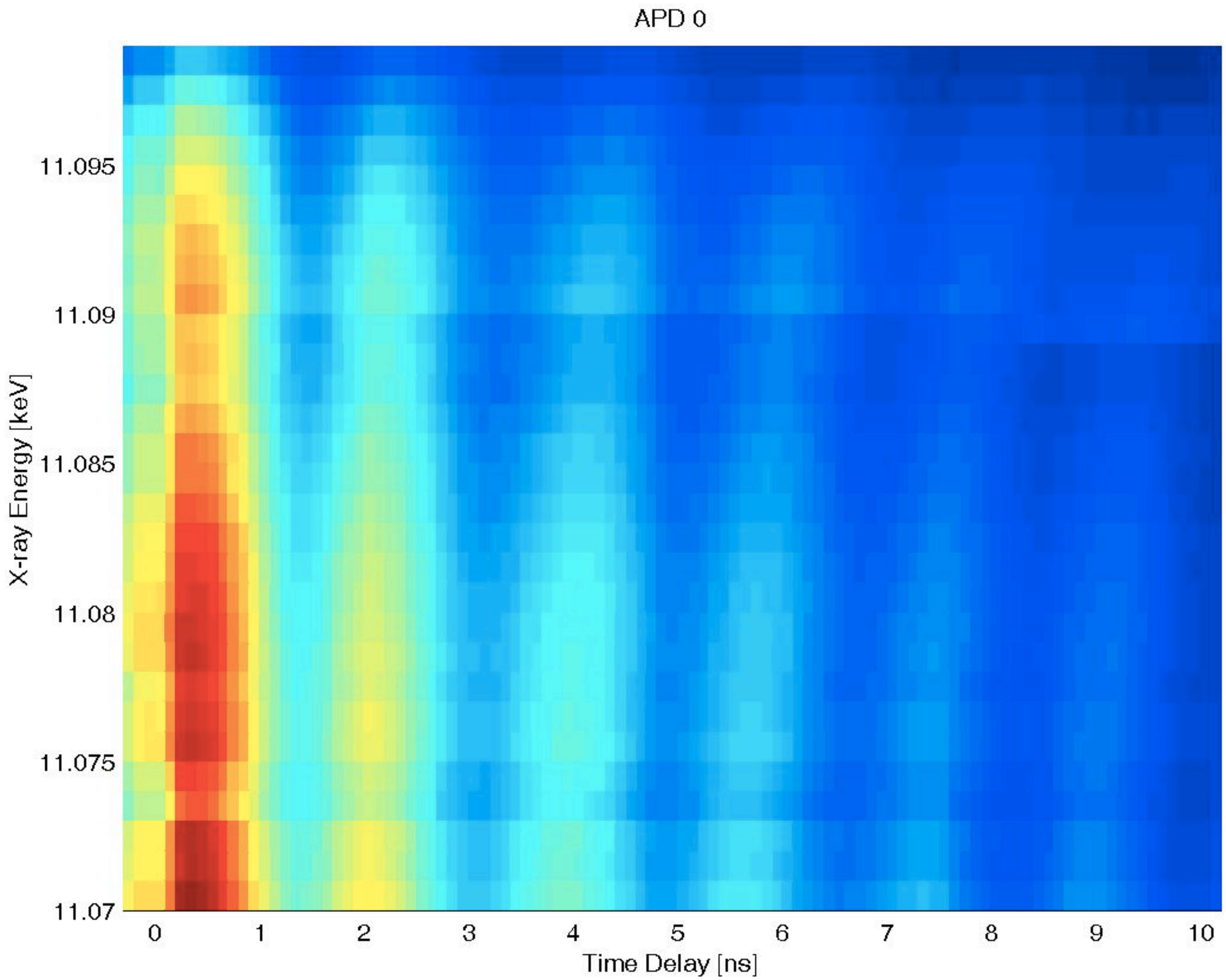
Time-resolved Laue Diffraction.

Using acoustic pulses, it is possible to control the intensity and direction of transmitted x-ray energy.

Temporal oscillations correspond to the pendellösung effect.

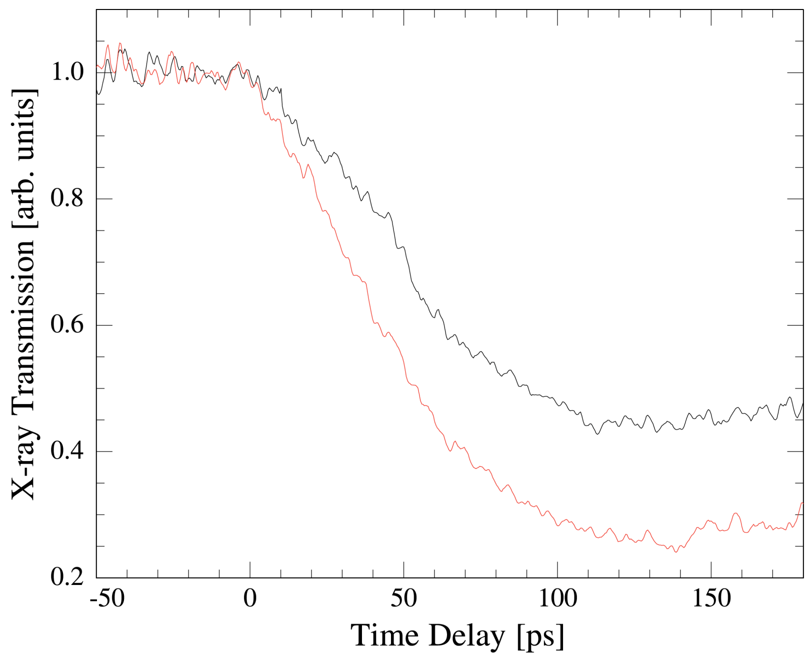
DeCamp et al. *Nature*, **413** 825 (2001)

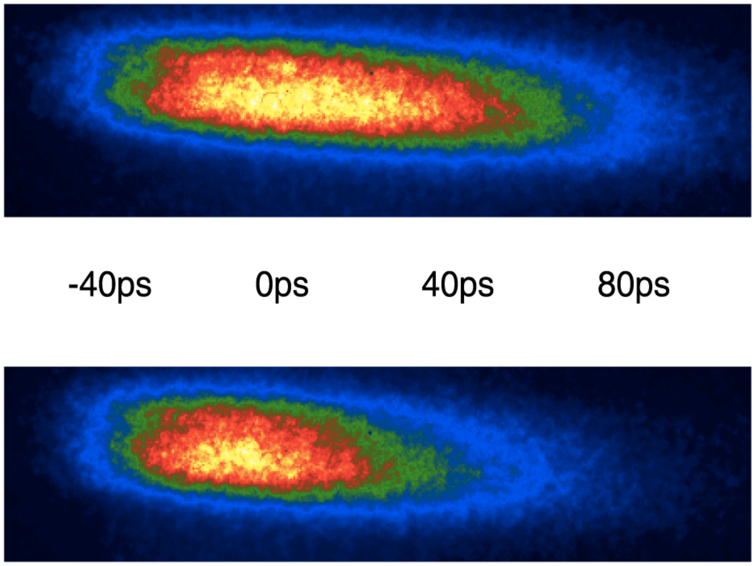
Utilizing energy flexibility of synchrotron radiation



The pendellösung period changes as the x-ray energy approaches the Ge K absorption edge.

Fast X-ray Switch

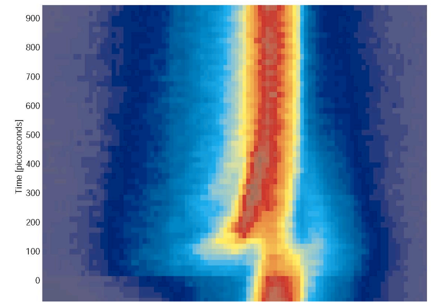




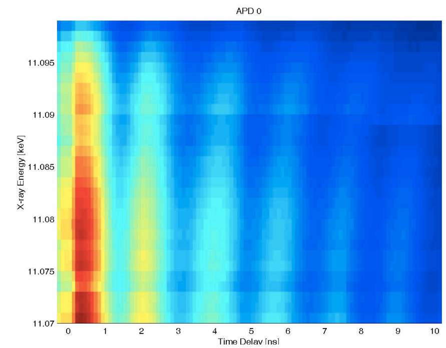
In current form reduces effective x-ray pulse width from 100ps to ~60ps
With timing upgrade: 80ps to ~40ps (summer 2002)
Ultimate goal: sub-picosecond x-ray switch using optical phonons

Summary

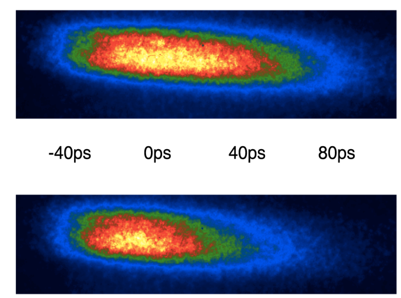
• Successfully using 100ps x-rays to study laser induced lattice dynamics



• Beginning to use available x-ray spectrum with ultrafast laser source

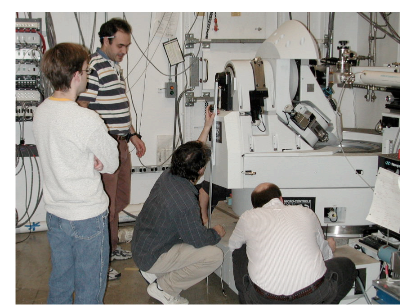


• Late summer 2002 will have 40-80ps x-ray source



• Late 2002 a permanent picosecond x-ray streak camera will be available

• Available to outside users starting fall 2002



Time-resolved X-ray diffraction investigations

Jörgen Larsson
Lund Institute of Technology
Lund, Sweden

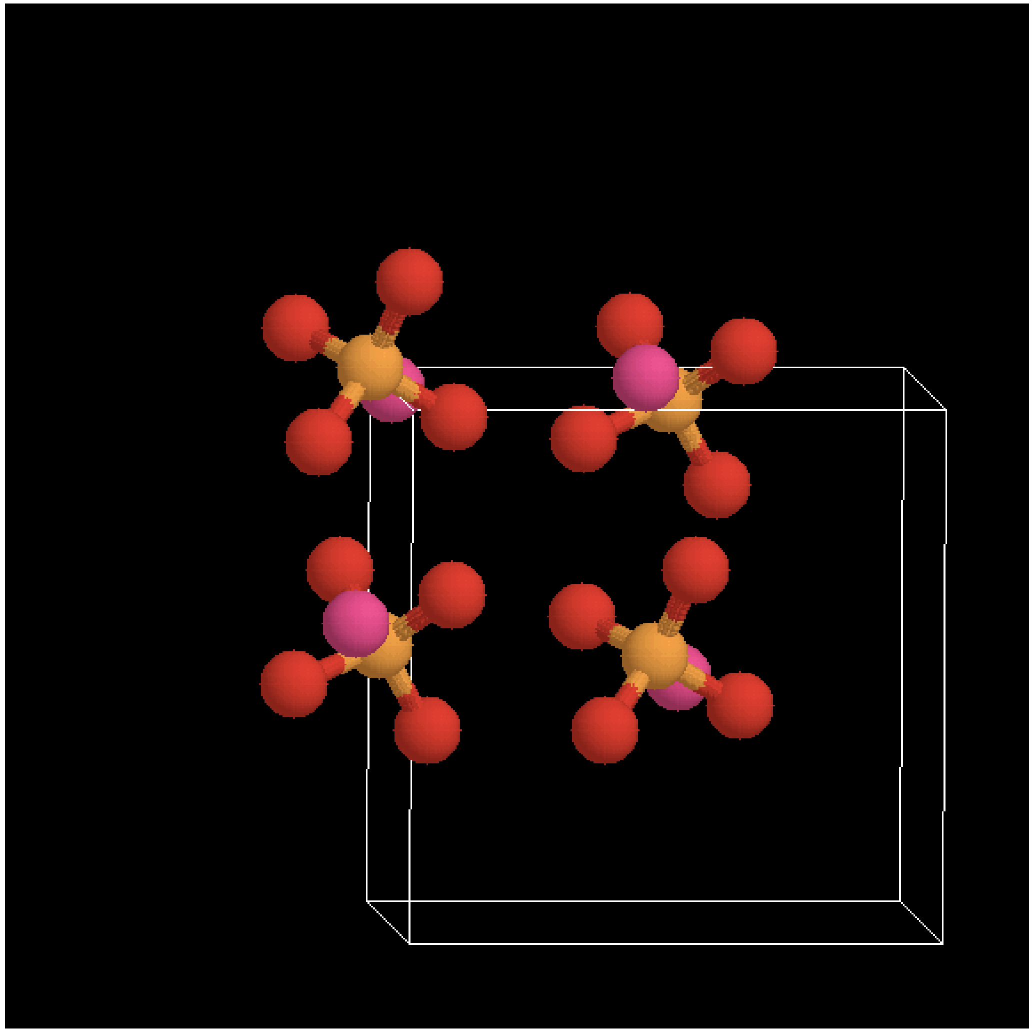
Time-resolved X-ray diffraction investigations

- Domain reversal in KDP
- Single-shot time-resolved rocking curves
- Beamline D611 at MAX II

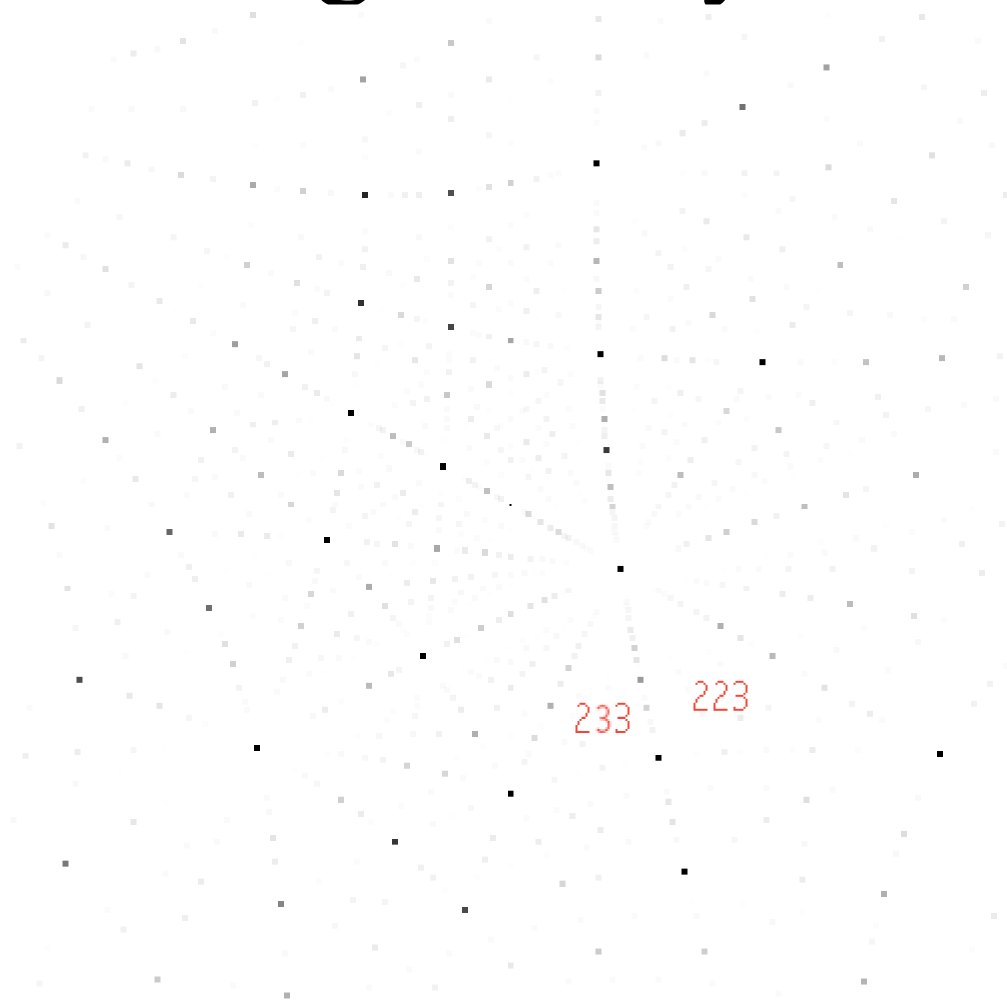
Domain reversal in KDP

- Jörgen Larsson, Ola Synnergren, Michael Harbst, Lund
- Peter Sondhauss, Justin Wark, Oxford
- Aaron Lindenberg, Phil Heimann, LBL

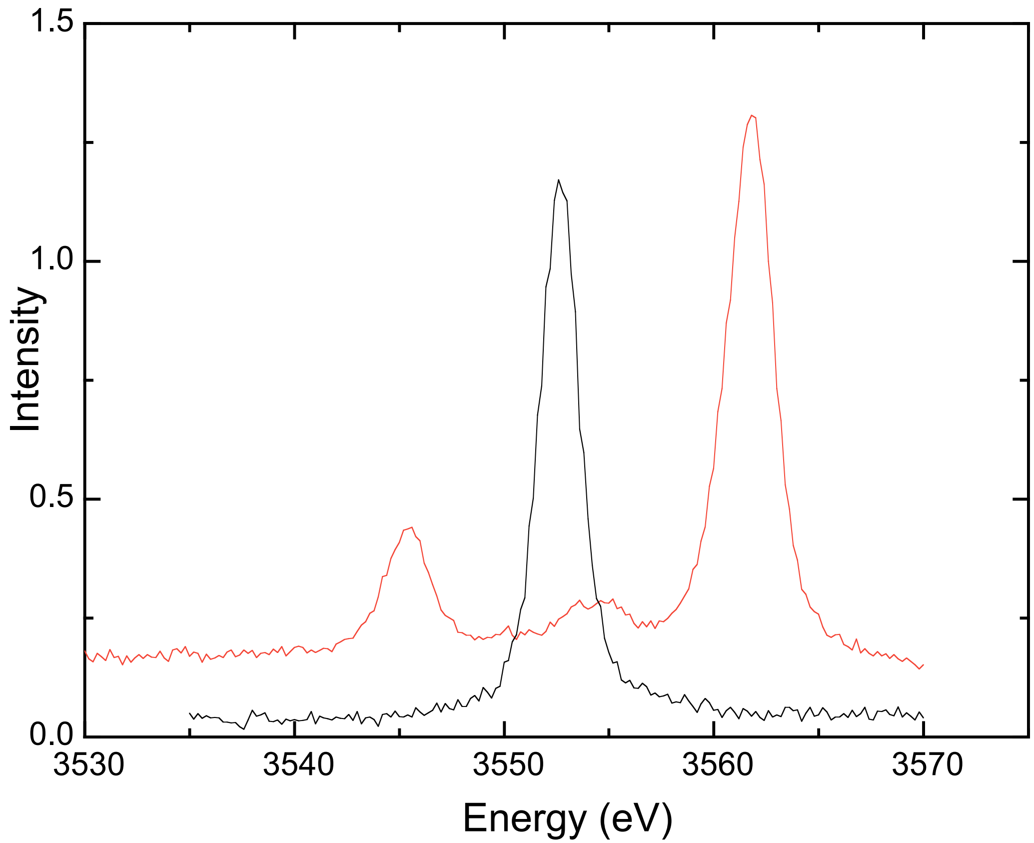
KD*P



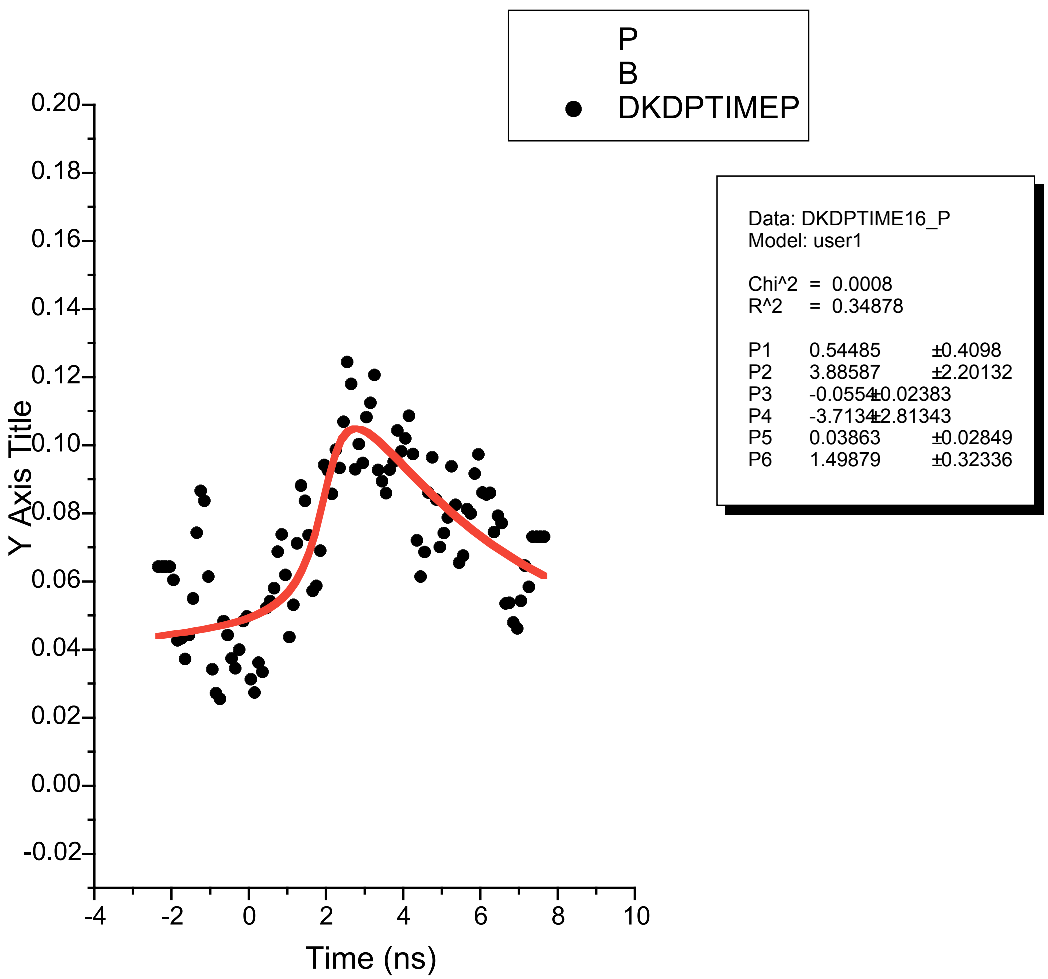
Laue simulation for chosen geometry



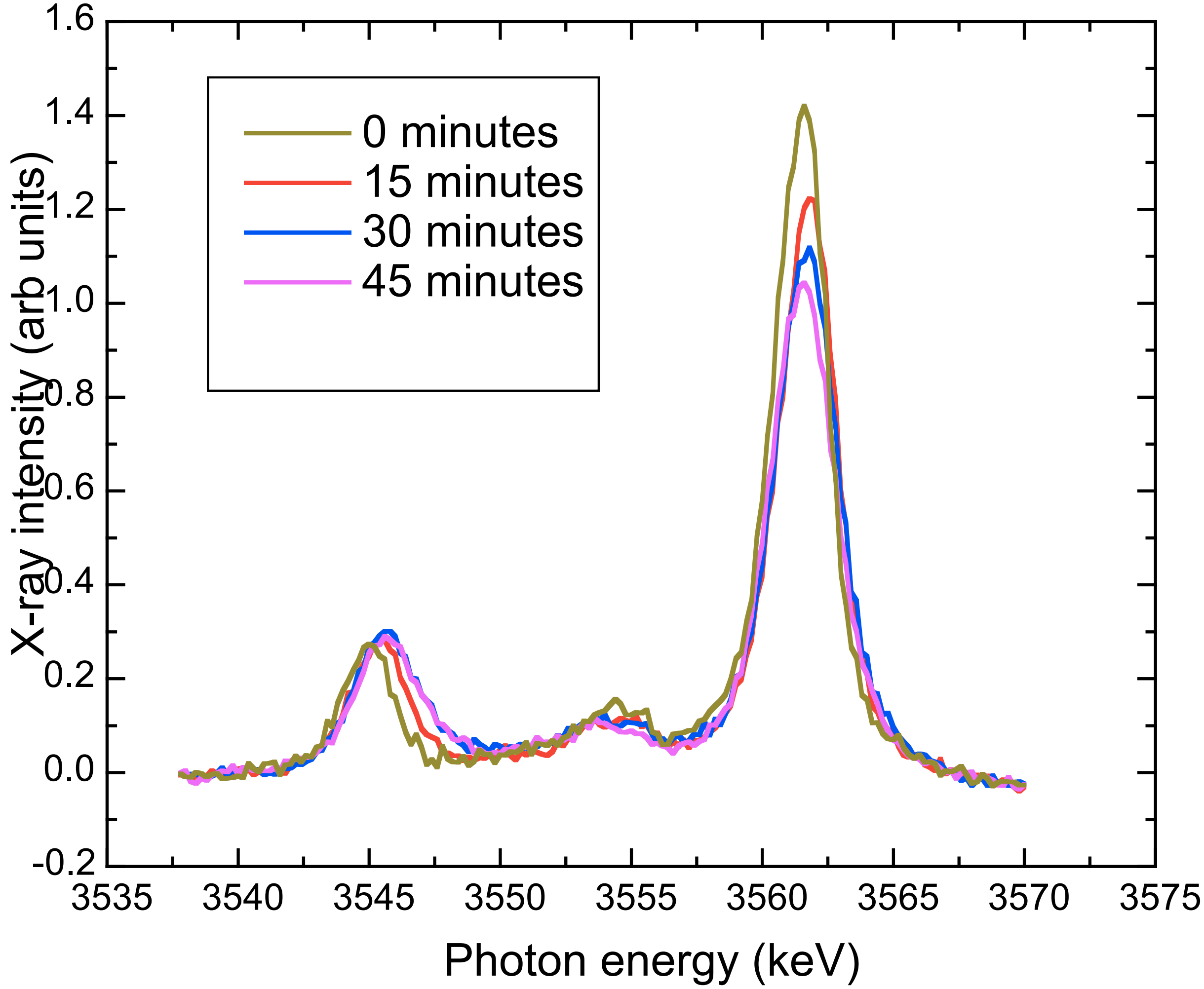
KD*P above and below Tc



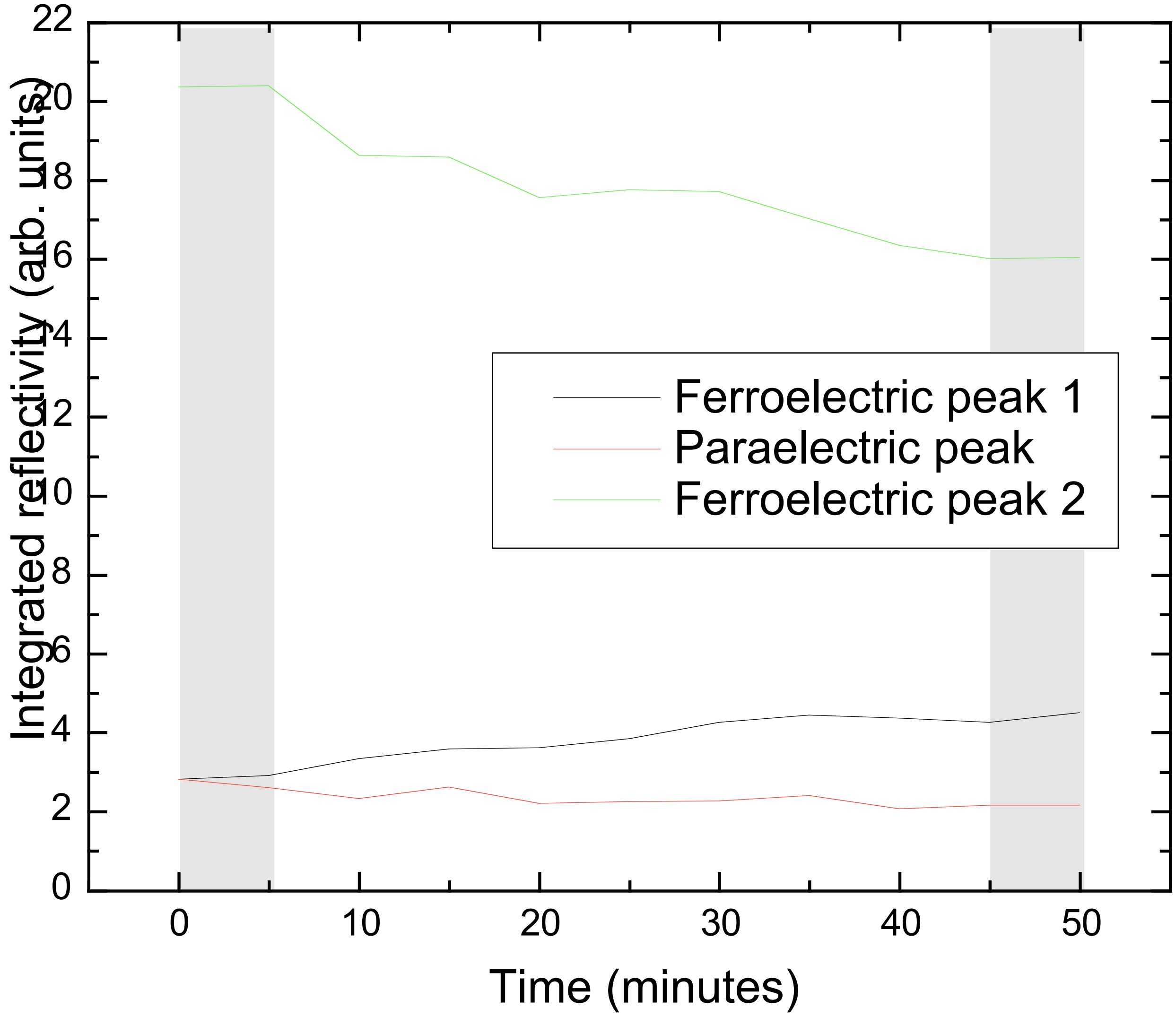
Time-resolved recording of the intensity of the paraelectric phase in KD*P at 210 K



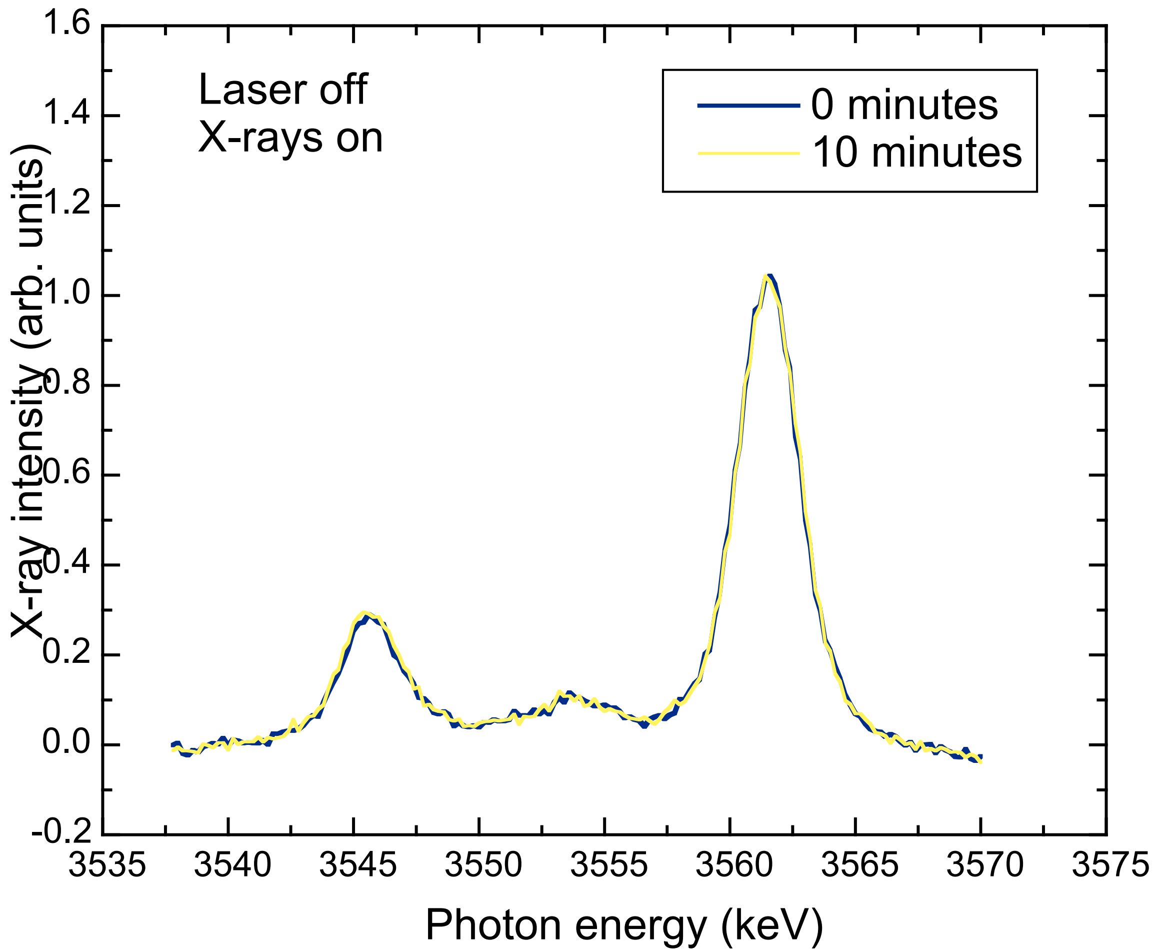
Domain reversal in KD*P



Integrated reflectivity



X-rays are not responsible

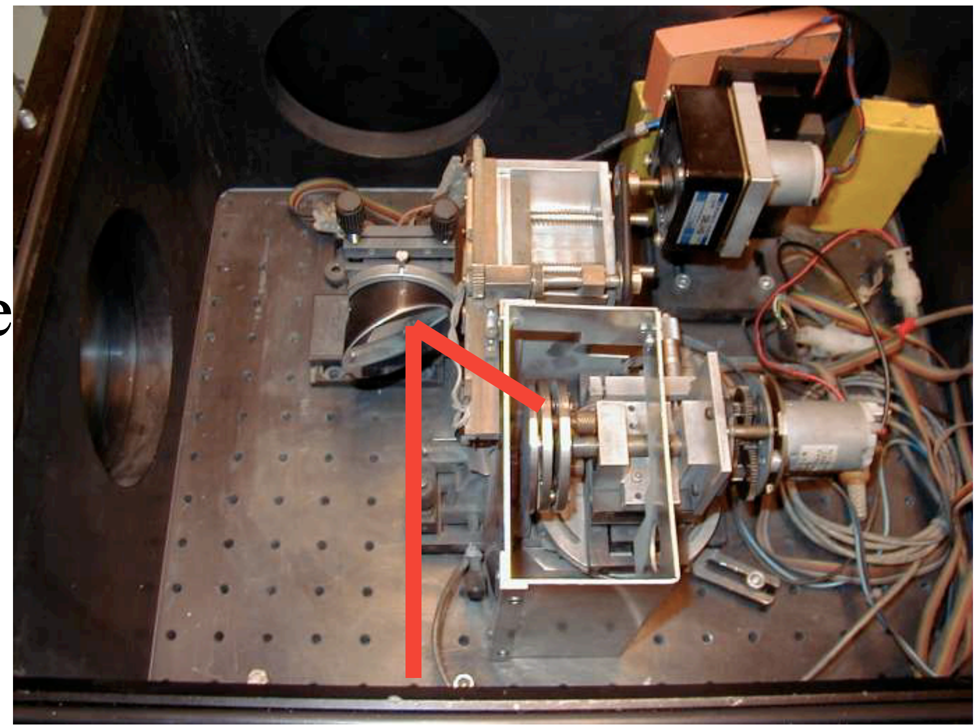


Single-shot time-resolved rocking curves

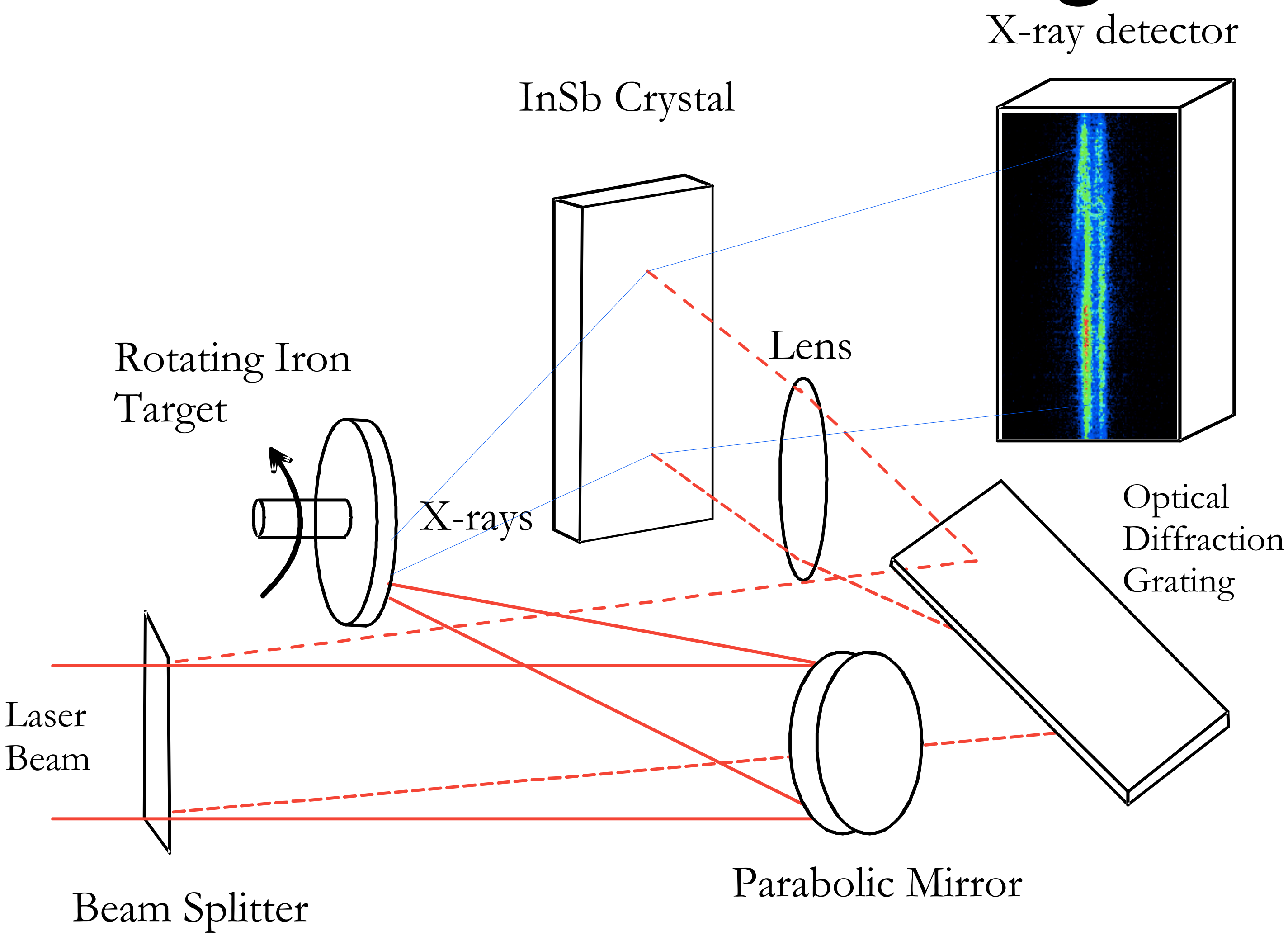
- Jörgen Larsson, Ola Synnergren, Michael Harbst, Lund
- Gergeley Katona, Richard Neutze, Göteborg
- Remco Wouts, Uppsala

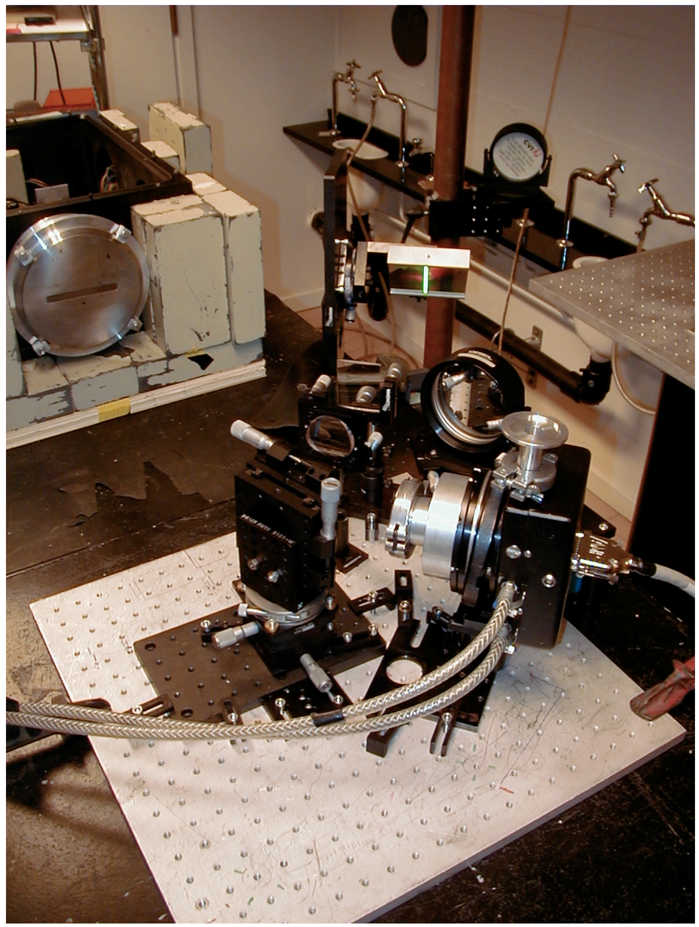
Solid target laser-plasma source

- 10 Hz
- 200 mJ driving laser
- 10^8 photons per pulse at 6 keV
- <3 ps pulse duration @6 keV

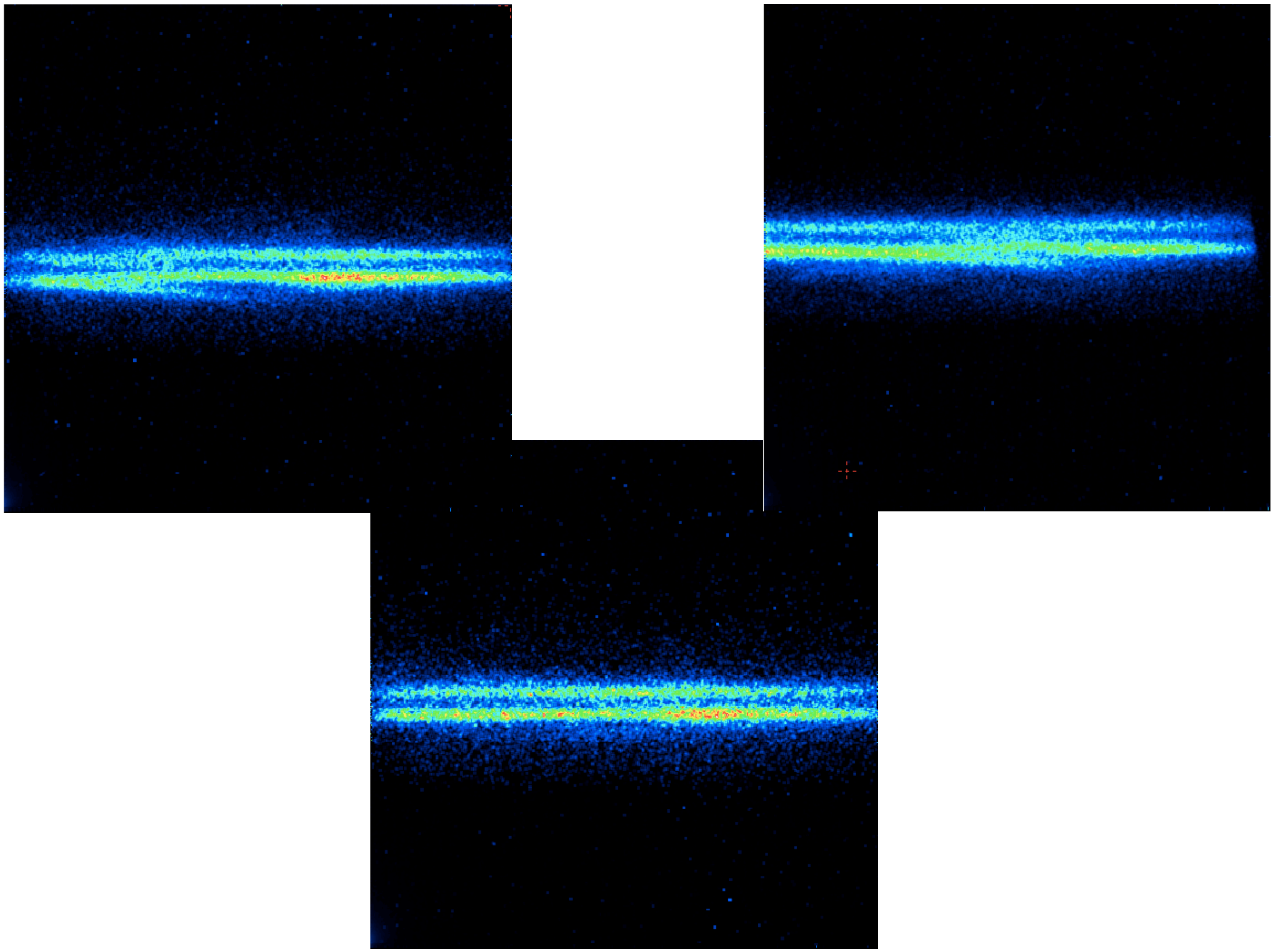


Set-up for acquisition of single-shot time-resolved rocking curve

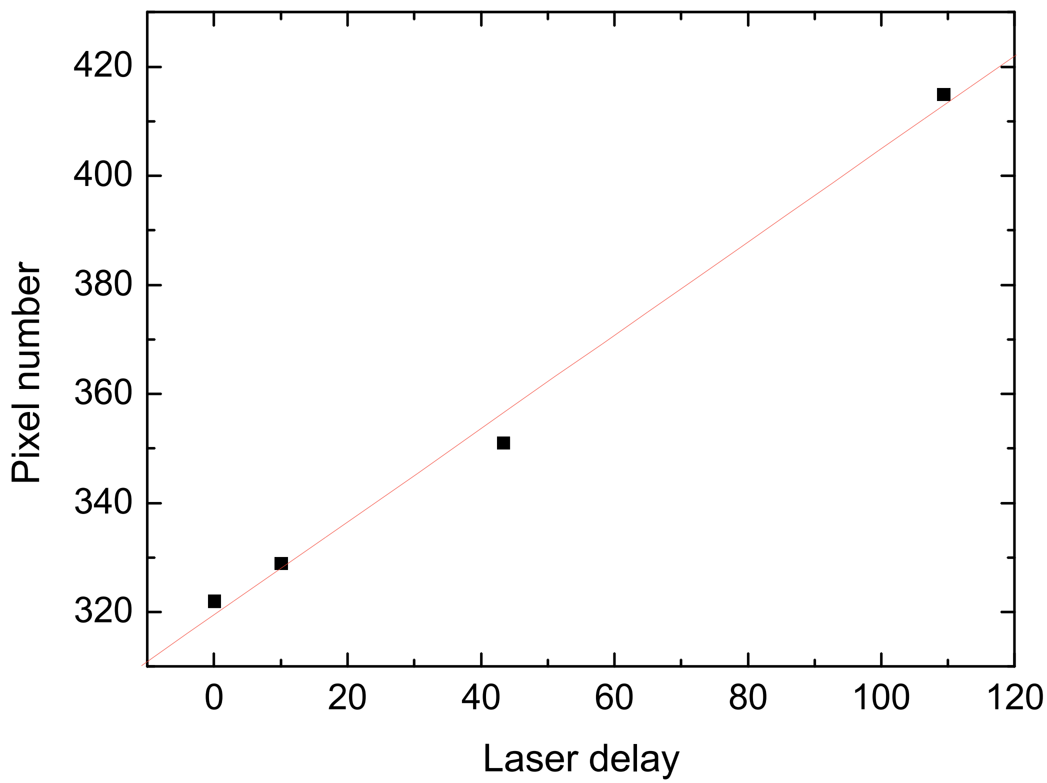




Time-resolved rocking curves



Calibrating cross-correlator



Cross-correlation

- No moveable parts
- Insensitive to shot-to-shot noise
- Removes the requirement of focusing optics
- Applicable at FEL

Time-resolved Structural Dynamics on the Nano-scale

Aaron Lindenberg

UC Berkeley Physics
Advanced Light Source, LBNL

Summary

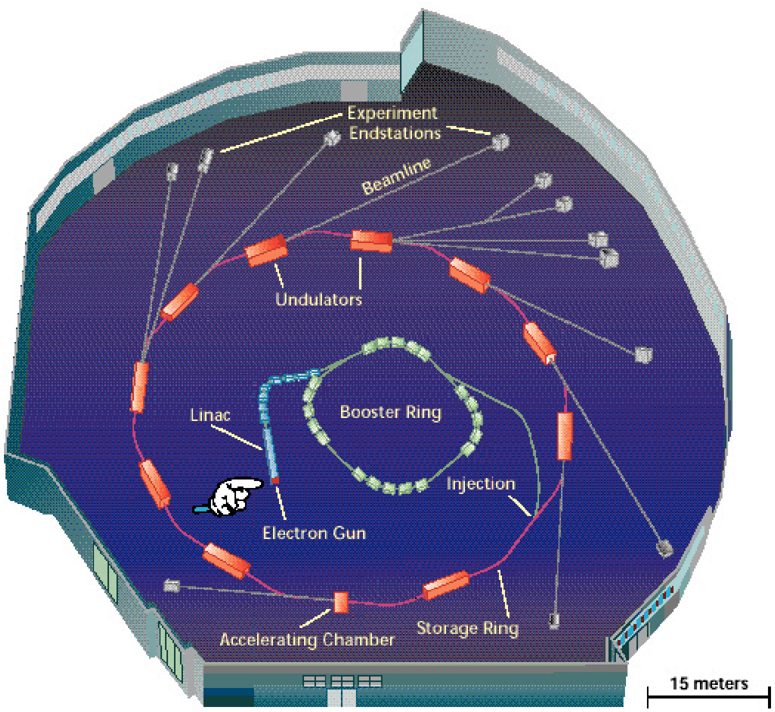
- Review of experiments at the ALS
  - Experimental setup
  - Phonon spectroscopy in InSb (brief)

- Phase transitions in semiconductor nanocrystals
  - Description of initial time-resolved diffraction measurements

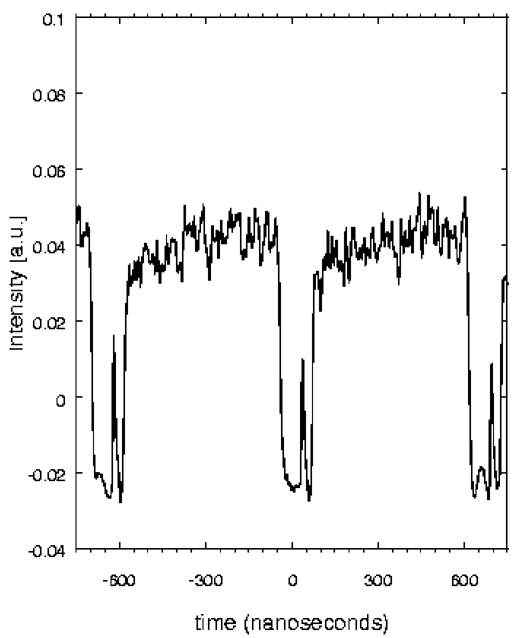
- Dynamics in H20
  - Diffraction measurements in the solid and liquid phase
  - NEXAFS probing of Hydrogen bonding

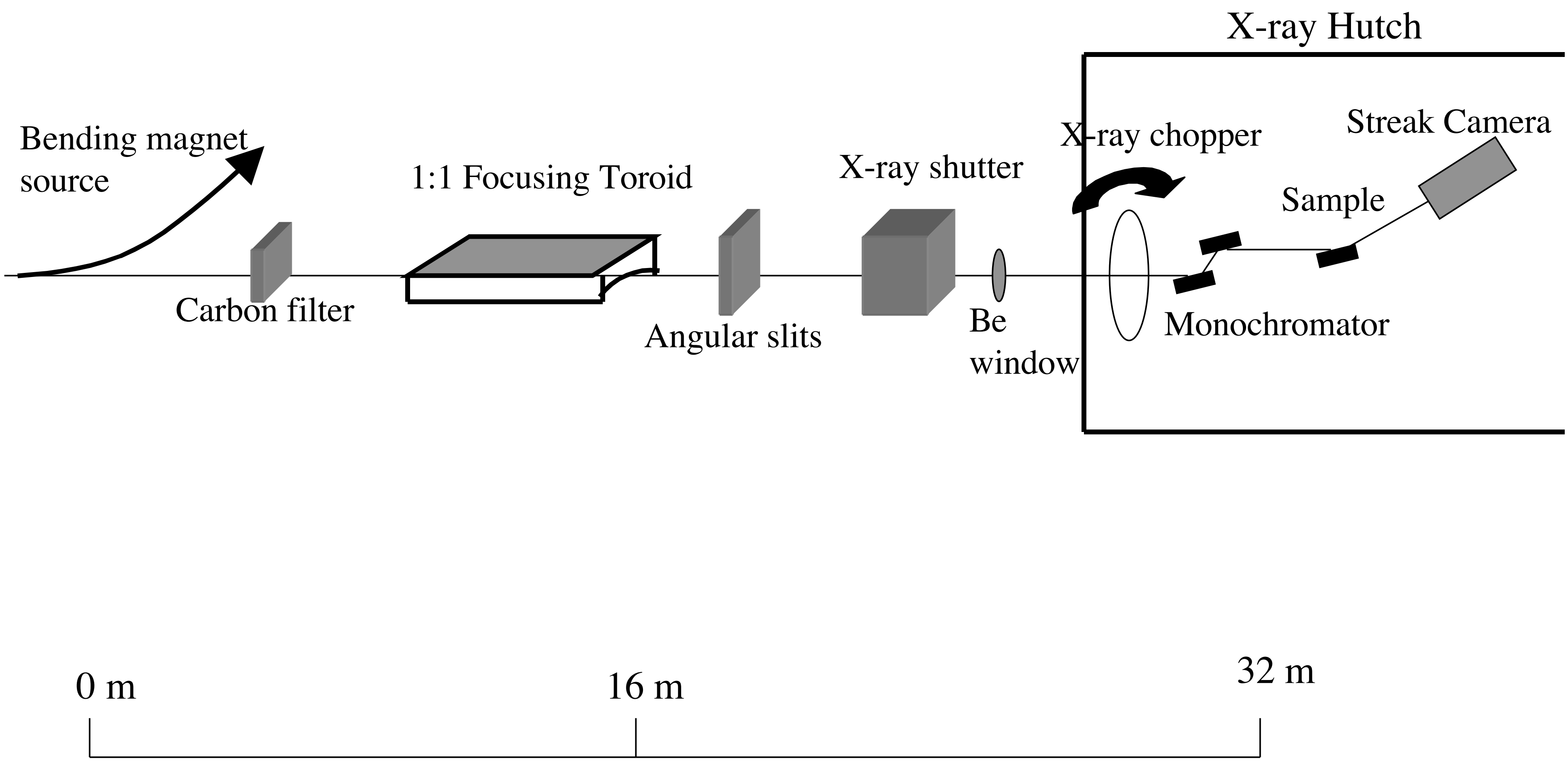
Experimental setup

The Advanced Light Source:



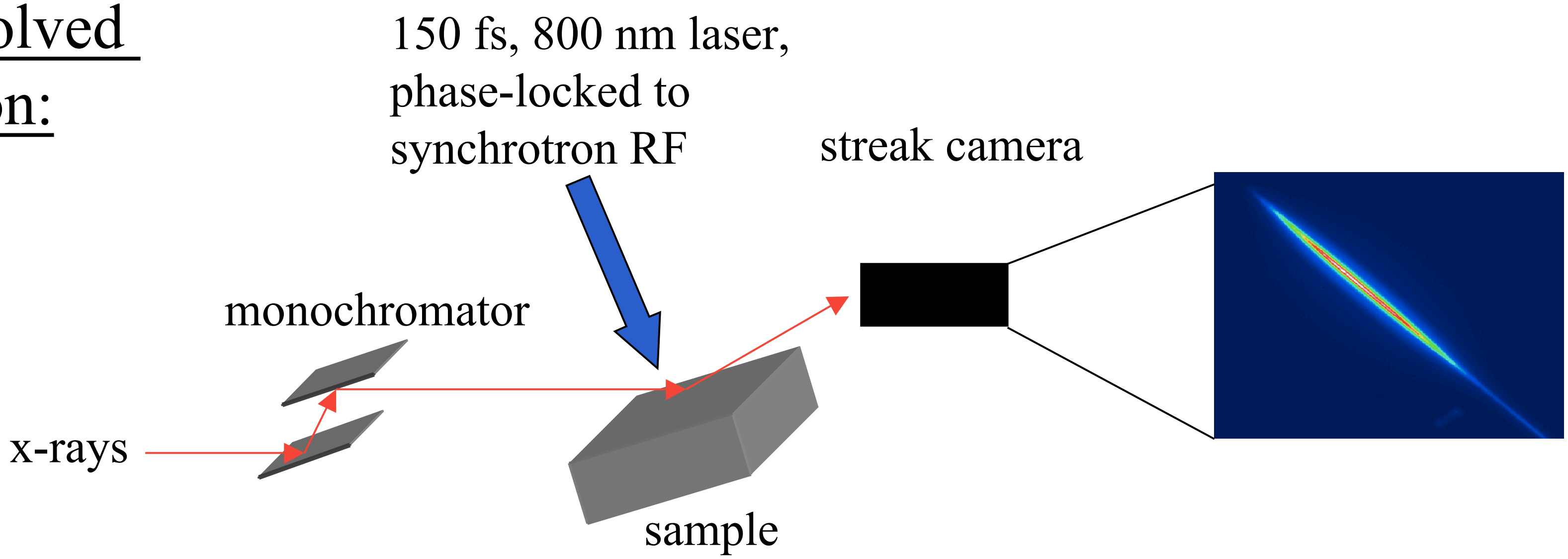
• 1.9 GeV electron beam
• electron bunches separated by 2 ns
• 656 ns roundtrip time



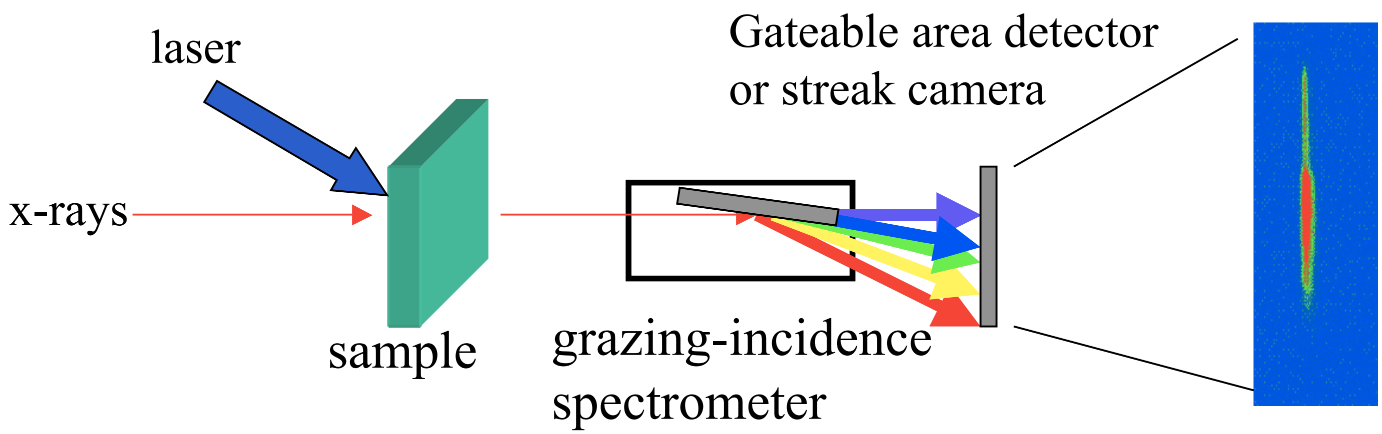

X-ray Hutch
Bending magnet
source
1:1 Focusing Toroid
X-ray shutter
X-ray chopper
Streak Camera
Sample
Carbon filter
Angular slits
Be
window
Monochromator
0 m
16 m
32 m

Experimental Setup

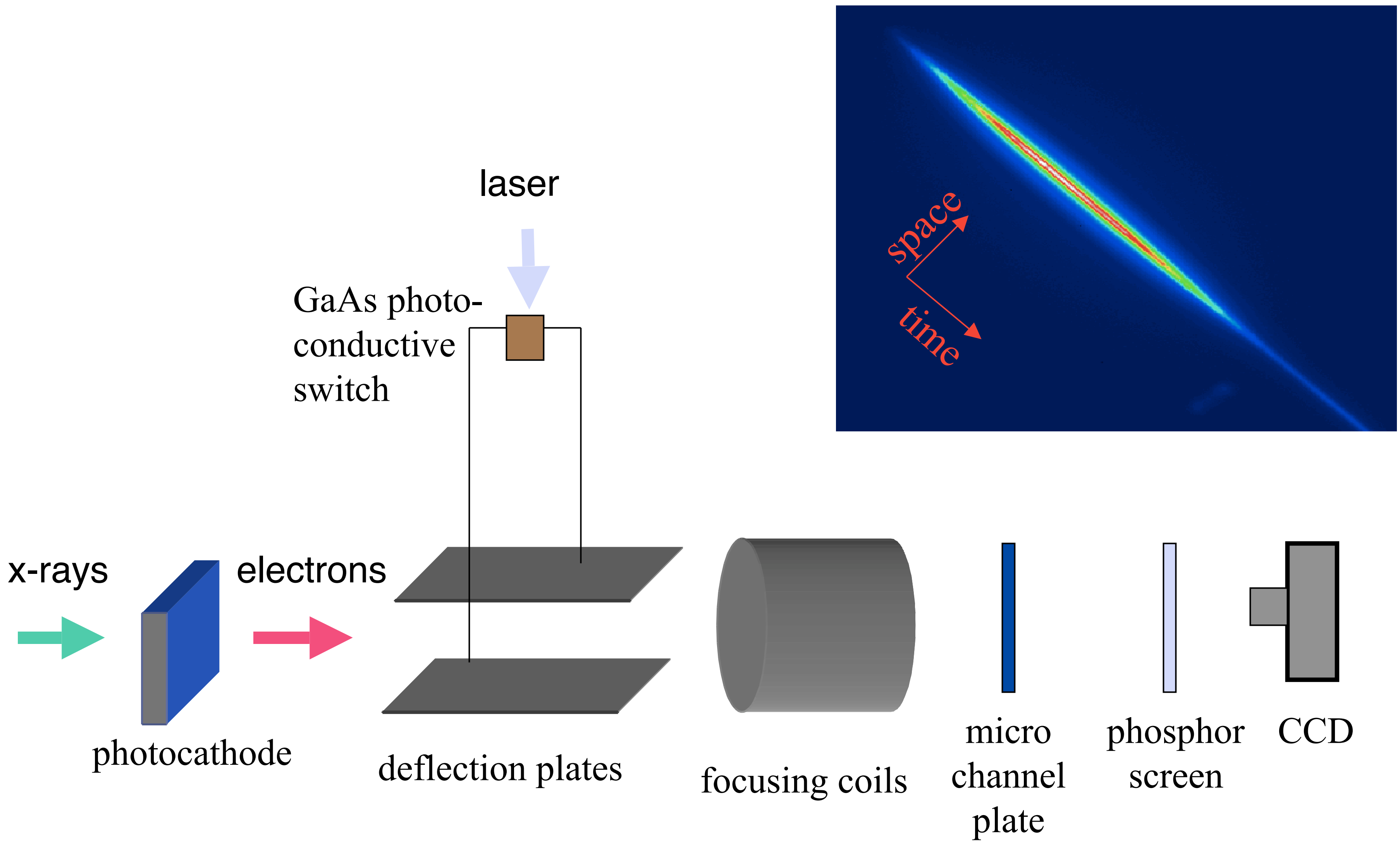
Time-resolved Diffraction:



Time-resolved Absorption:



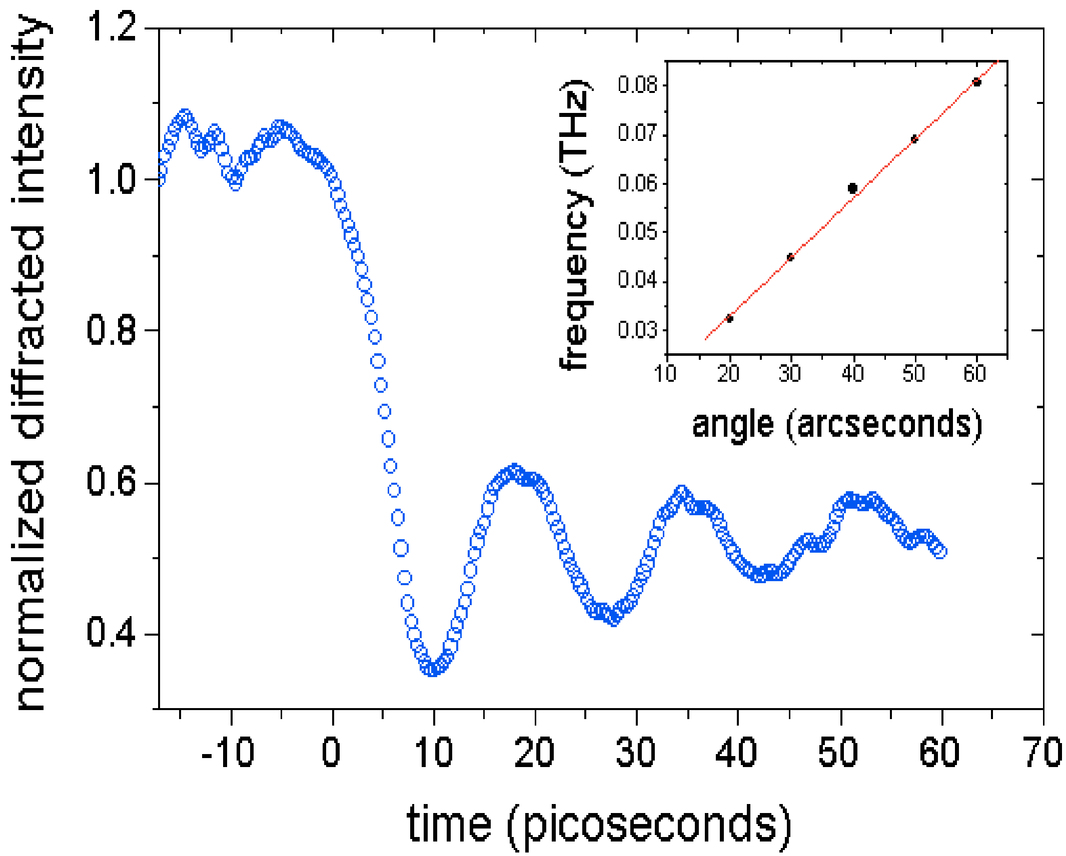
Picosecond x-ray streak camera detector



temporal resolution < 2 ps

Probing the q=9 x 10^5 cm^{-1} acoustic mode in InSb

□□=40 arcsec

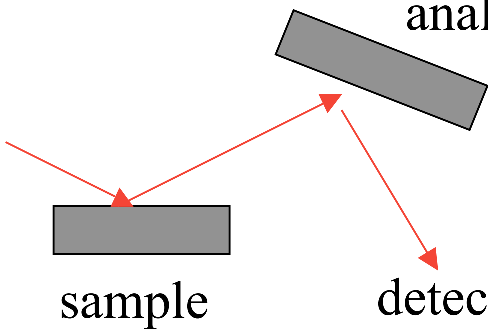


A.M. Lindenberg *et al., Phys Rev. Lett*. **84**, 111 (2000)

X-ray diffraction as a probe of vibrational excitations in solids

Frequency domain picture

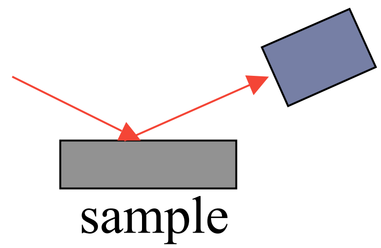
Inelastic x-ray scattering



Resolve the change in energy of an x-ray photon scattered off a phonon, at a given momentum transfer q

Time domain picture

Time-resolved x-ray diffraction



Resolve the modulations in time of the diffracted x-ray intensity, induced by a phonon of wave-vector q

Motivations for studying dynamics in nano-scale systems

•Nanocrystals lie on the boundary between atomic-scale and bulk systems.

•Model systems for the study of phase transitions

•What are the structural pathways these systems follow as they undergo solid-solid phase transitions?

•Can nano-scale systems be used to create new, metastable states of matter.

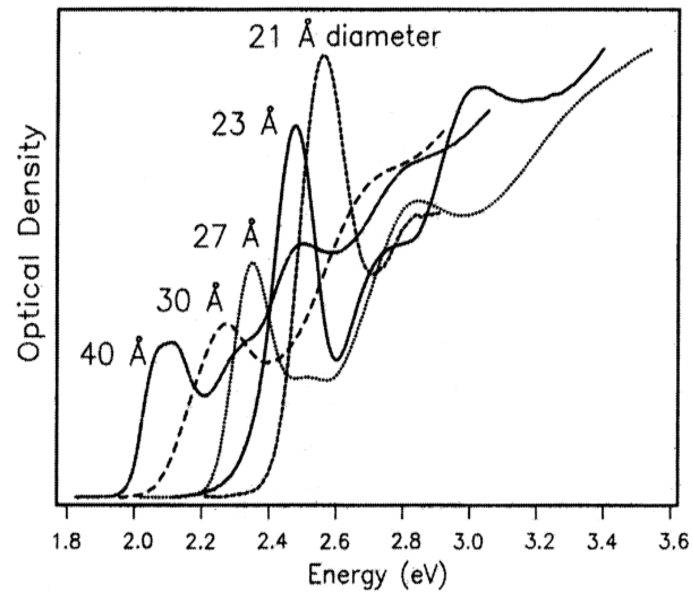
•Localization of energy -> High energy density physics

• Time-resolved methods as a technique for probing transient phases at high temperatures and pressures.

Properties of semiconductor nanocrystals

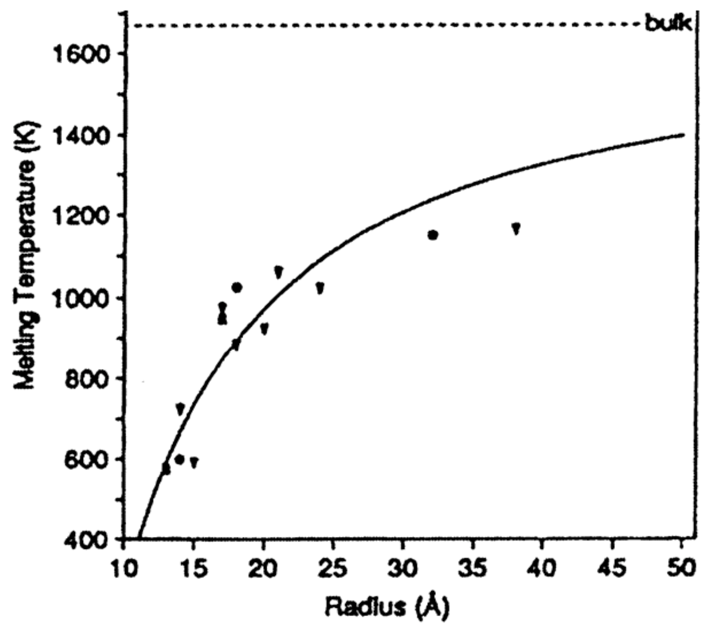
Electronic properties

•Quantum size-effects due to confinement of electrons/holes. Interesting nonlinear effects…



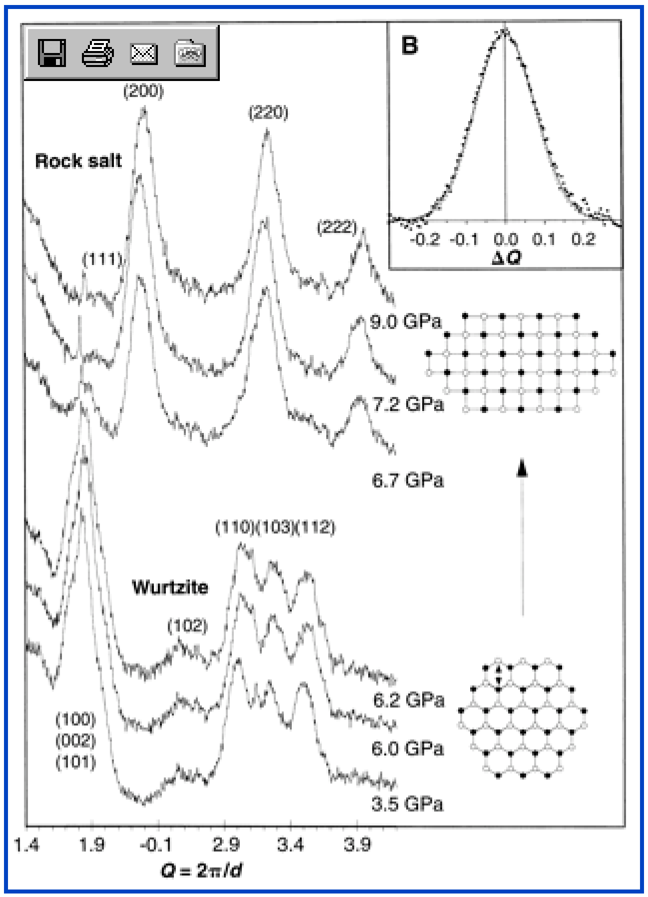
Structural properties

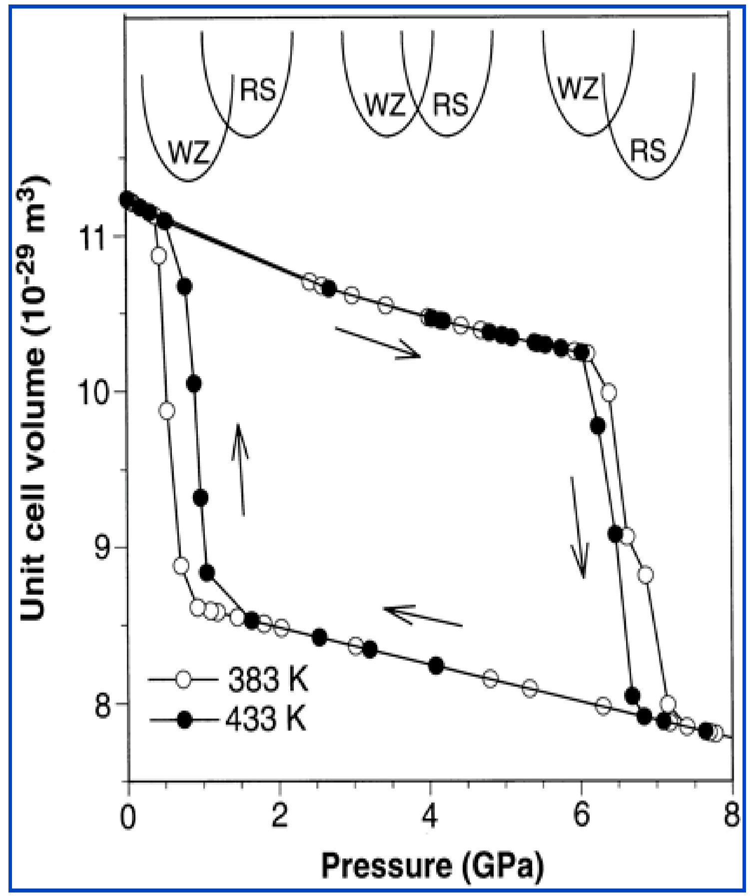
• ~30 % of the atoms in a single nanocrystal are on the surface: Implies size-dependent solid-solid and solid-liquid phase transitions



Properties of semiconductor nanocrystals

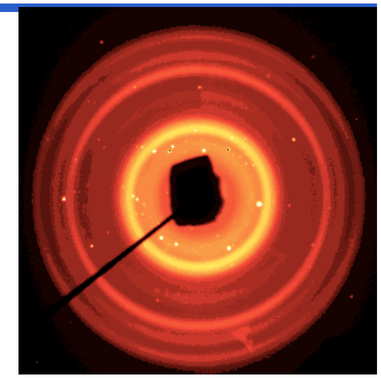
•A coherent transformation along well-defined pathways – a molecular isomerisation

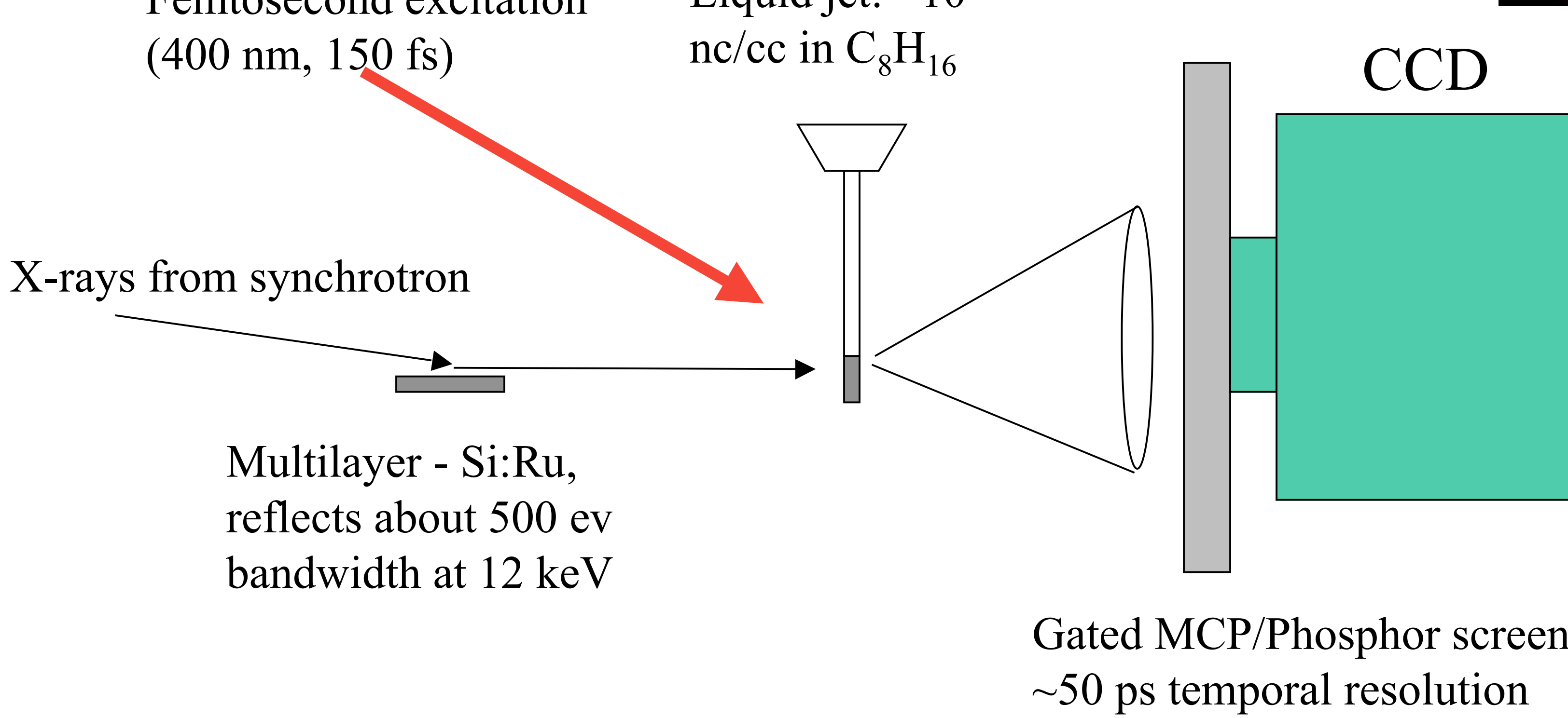




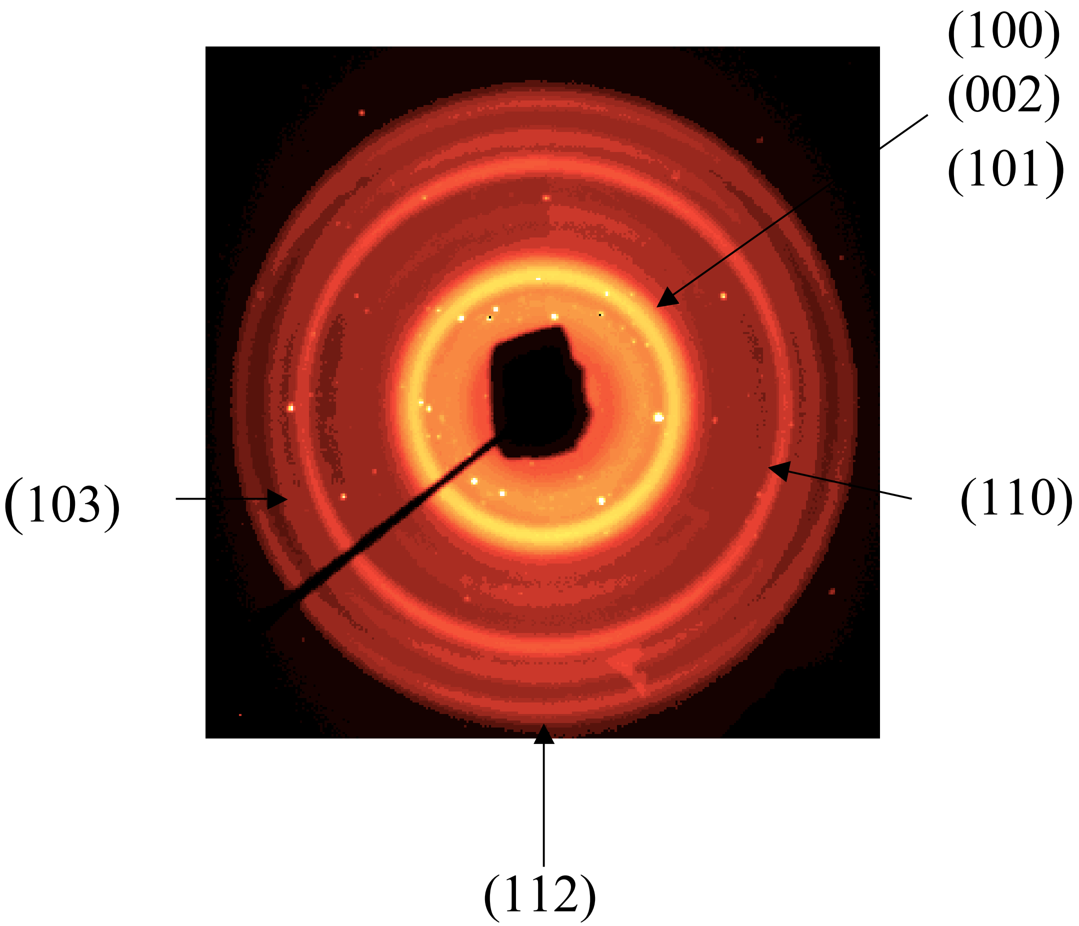
C. Chen et al, Science, **276**, 398.

Experimental Setup

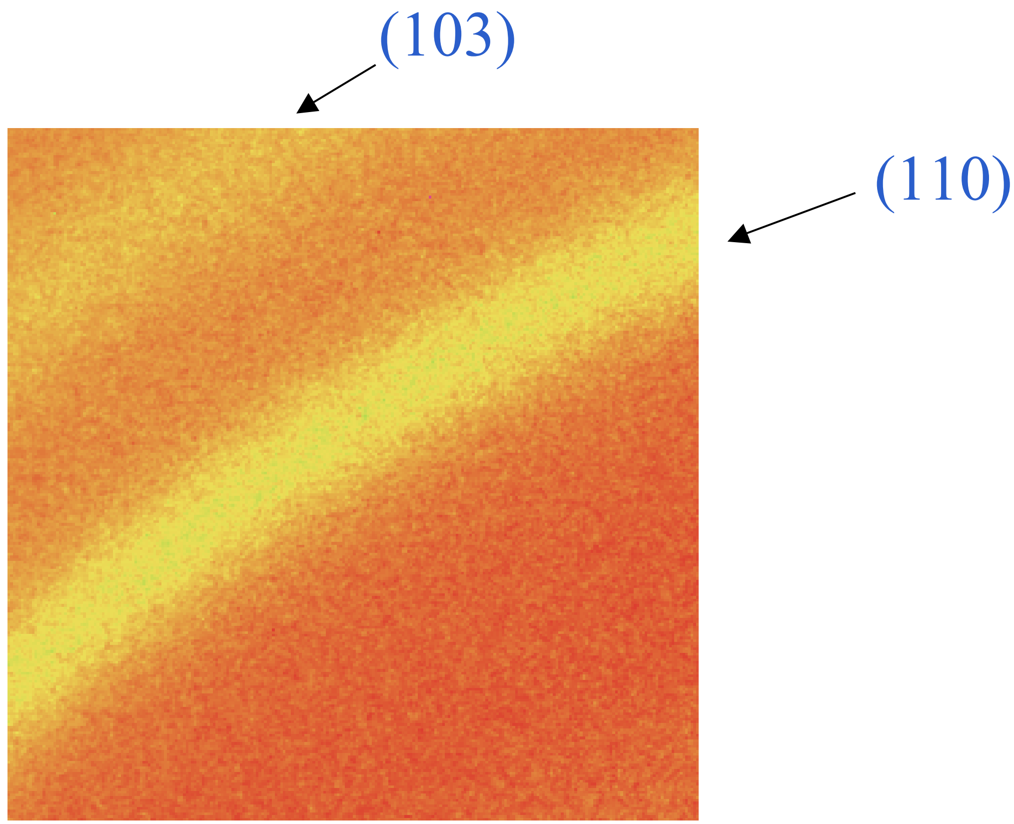




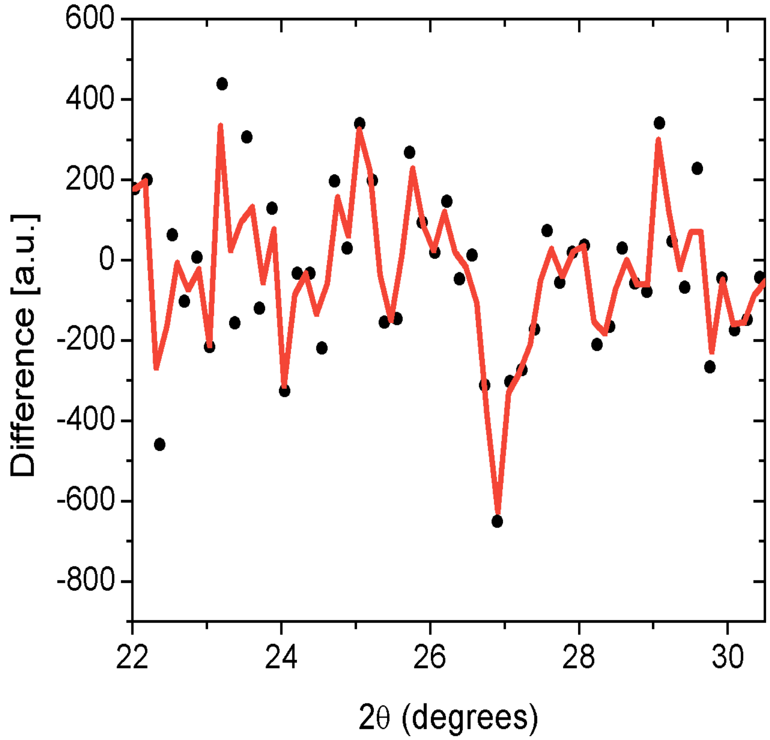
4.2 nm spherical CdSe nanocrystal diffraction pattern



CdSe nanorod diffraction pattern



Temporal difference map at t=10 ns

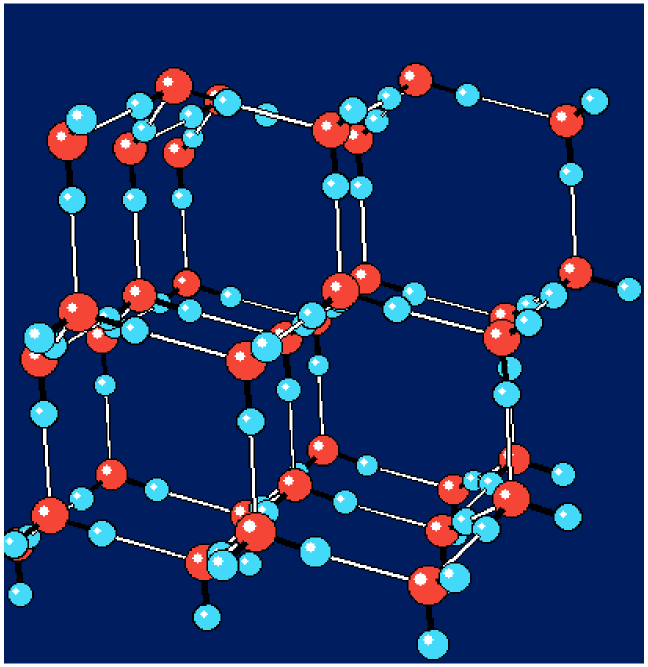


• Implies complete cancellation of the (110) diffraction peak. Simplest explanation: Disordering/melting.

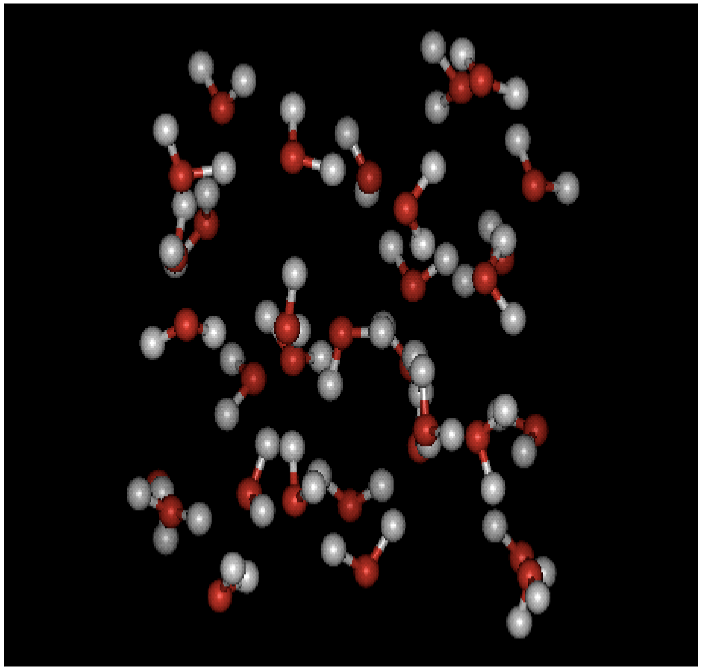
• Future experiments at the APS using a complete harmonic from undulator should give more detailed information.

The structure of water

(Ice)



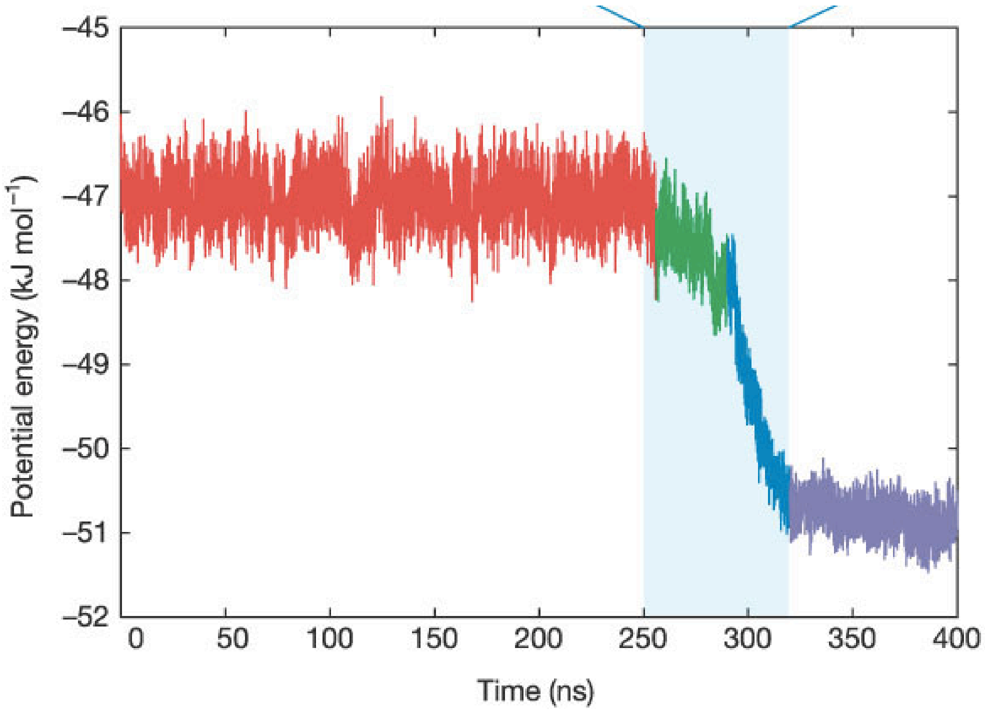
(Water)



•Weak hydrogen bonding implies fast intrinsic dynamics

Water-Ice Crystallization Dynamics

MD simulation:



Matsumoto et al., *Nature,* **416**, 409 (2002)

X-ray absorption spectroscopy of water

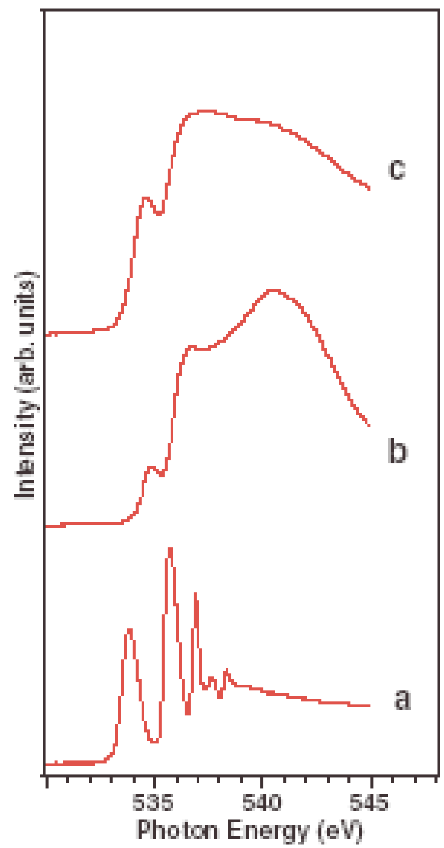


Figure 1. Experimental XAS spectra of water in (a) gas phase, (b) ice, and (c) liquid.

S. Myenni *et al. J. Phys. Cond. Matter* **14**, L213 (2002)

- Pre-edge peak gives a direct probe of hydrogen bonding in all phases
- Can we impulsively disrupt H-bonding through infrared excitation?

Motivations for studying liquid water

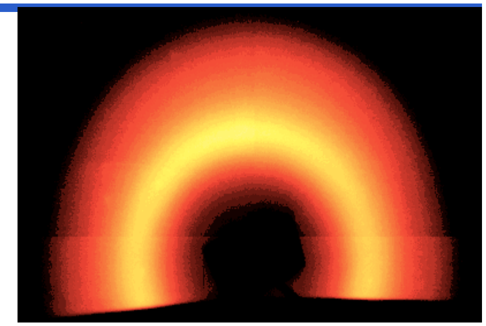
•What are the nature of the short wavelength collective excitations in the liquid state?

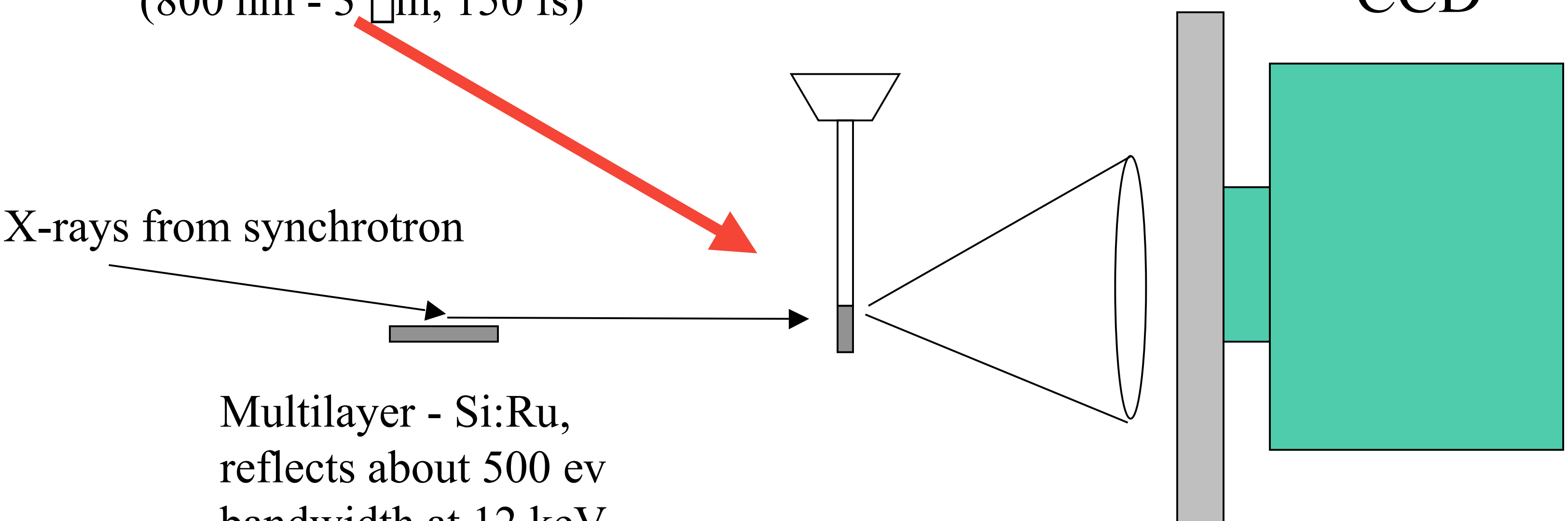
•At high Q, the dynamical response of H2O becomes solid like, due to strong hydrogen bonding on short length scales (Normal vs. Fast sound).

•Small Q region – scattering related to fluctuations (strong in a gas, weak in a liquid)

•New structural phases created at high excitation conditions? (PRL 82, 4110 (1999))

Experimental Setup





DC measurements

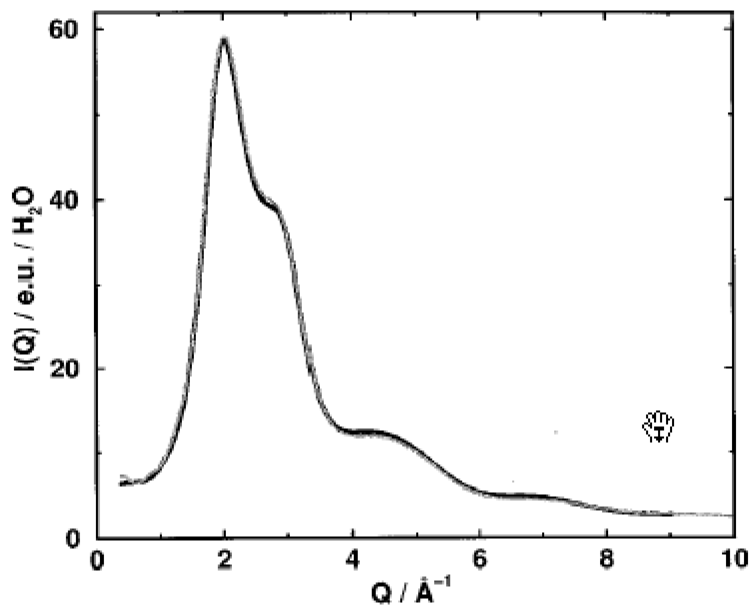
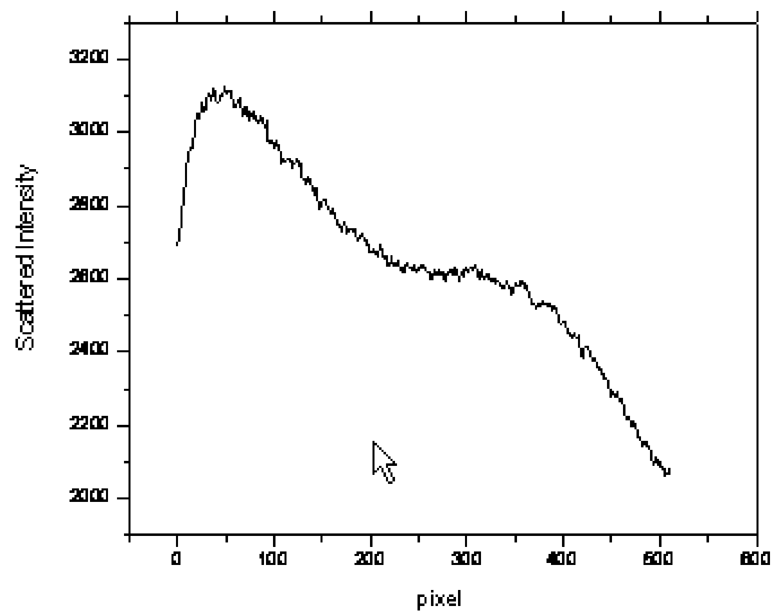


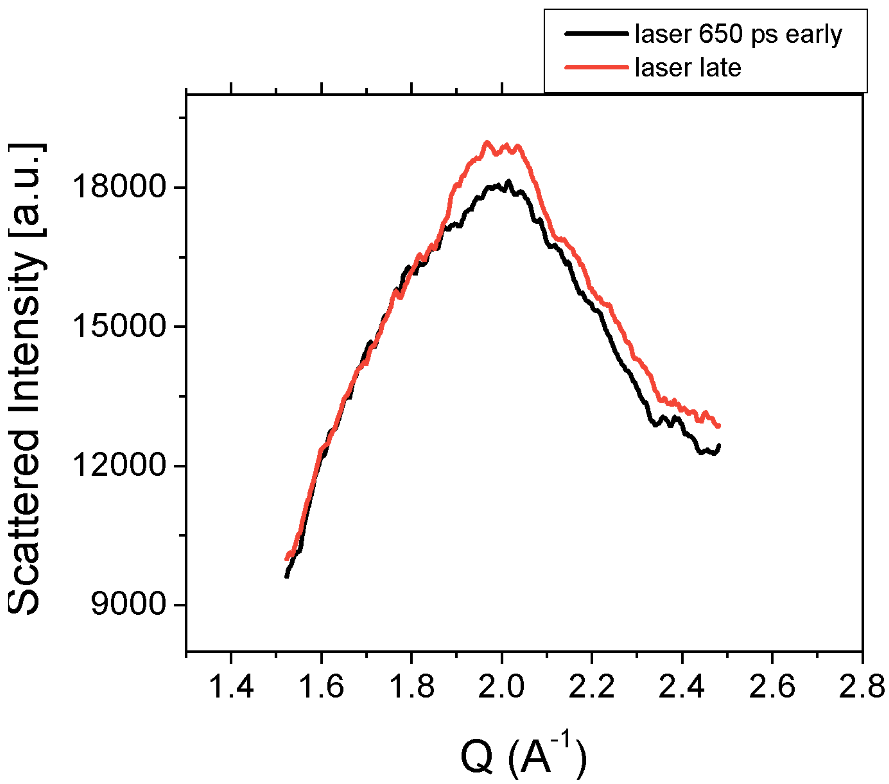
FIG. 4. Predicted x-ray scattering for the $g_{OO}(r)$ in Fig. 3 compared with experiment. Legend: Hura *et al.*, x-ray (black line); best fit (gray line).

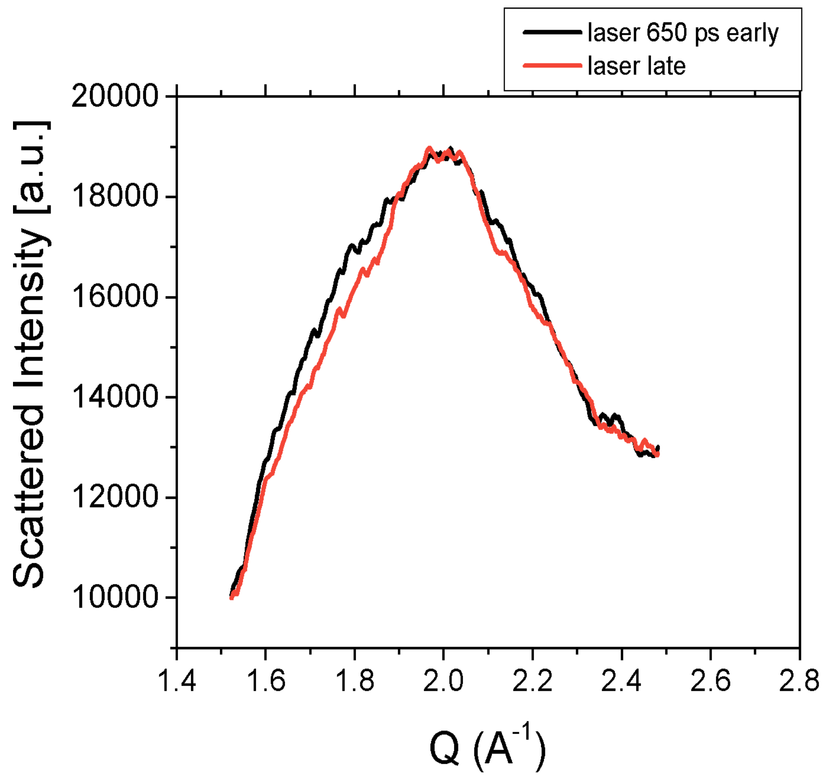
(J. Chem. Phys. **113**, 9153)



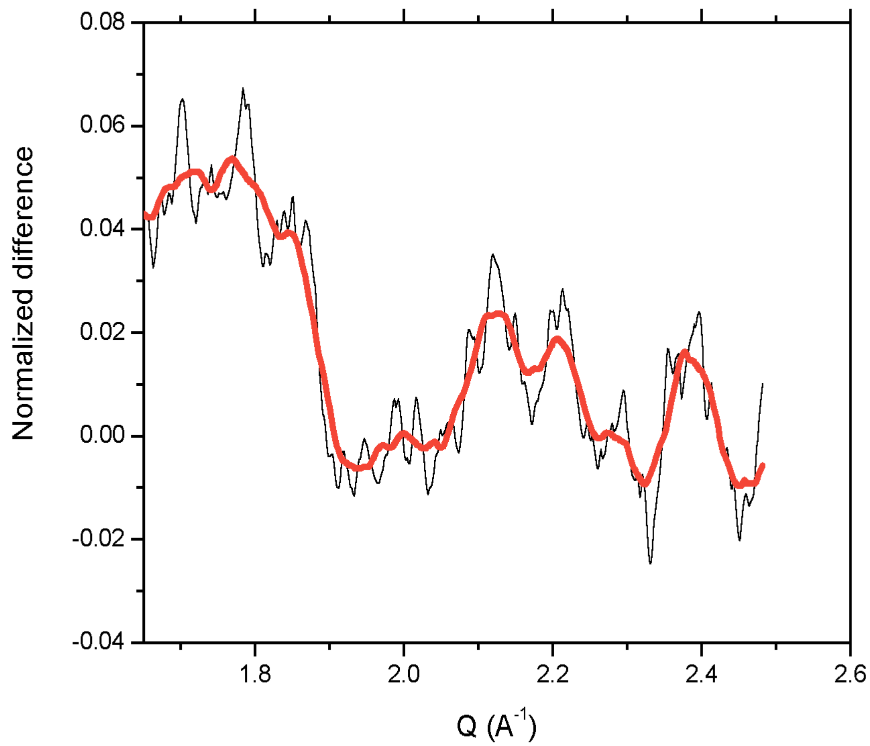
MCP data at 12 keV from 300 um water jet

Time-resolved measurements near the scattering peak in liquid H_2O





Difference Map at t=650 ps



Induced sideband at Q~1 nm^{-1}?

Conclusions

Potential Experiments at Sector 7:

•Time-resolved x-ray diffraction probing phase transitions in semiconductor nanocrystals. Evidence for a phase transition to a disordered state in CdSe nanocrystals. What are the time-scales? What is the dependence on shape?

•Water-ice crystallization dynamics. Can we follow the transition from liquid water (with associated diffraction rings) to crystalline ice (with associated Bragg spots) in real time?

•Dynamics in the liquid state. What are the time-scales for intrinsic structural rearrangements?

[1]University of California, Berkeley
[2]Lawrence Berkeley National Laboratory (Advanced Light Source)
[3]Kansas State University
[4]Lawrence Livermore National Laboratory
[5]Oxford University

[1]S.L. Johnson
[1]A. MacPhee
[2]P.A. Heimann
[3]Zenghu Chang
[4]R.W. Lee
[5]J.S. Wark
[1]R.W. Falcone

X-ray photoionization of laser-dressed atoms

- Understand x-ray photoionization from free atoms

- What happens with external field present ?
  - Low dressing fields (F≤0.03 a.u.) : free-free transitions
  - High dressing fields ??

    Ultrafast x-ray probe
    Clusters - collective electron motion?

- Can we modify the inner-shell decay dynamics in atoms ?

Linda Young
MHATT/CAT Workshop
June 4-5, 2002

Outline

- Theoretical predictions for x-ray + laser photoionization
  - Total cross sections
  - Angular distributions

    Hydrogen atom

    Laser fundamental = □; x-ray = n□

- Experimental observations

    Glover et al - He, $□_L$+ n $□_L$

    Recent attosecond metrology

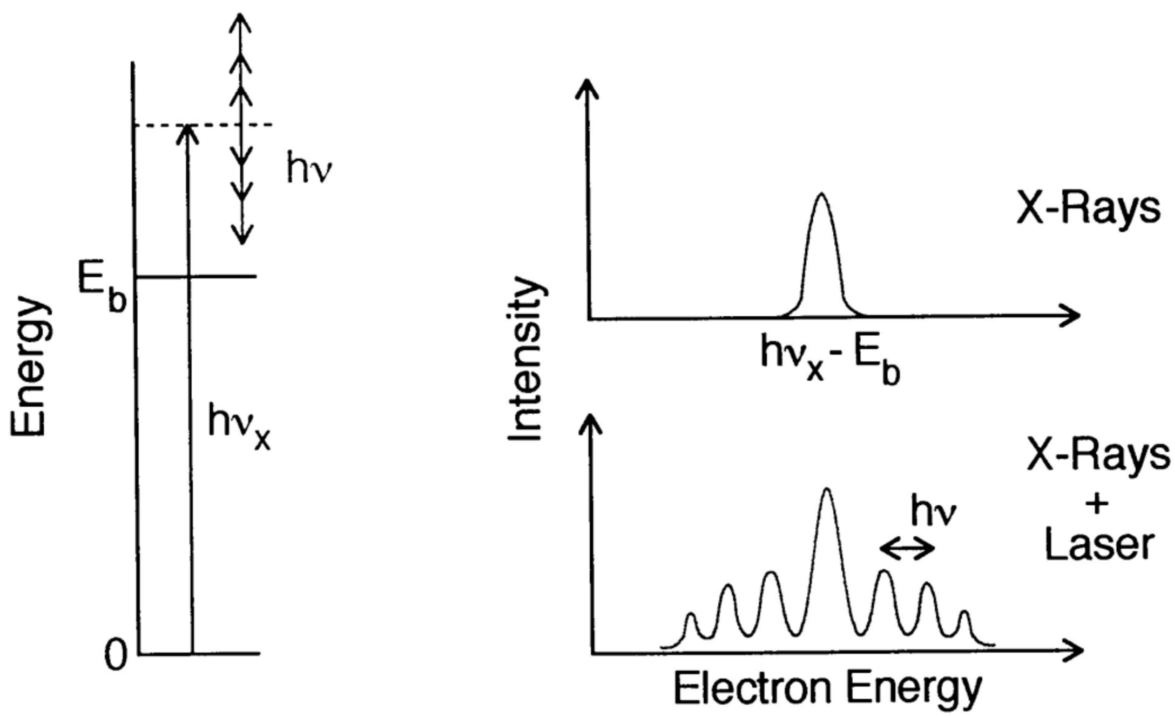
- Our wrinkles: tunable x-ray energy

    adjustable relative polarizations

    hard x-rays

    full angular distribution

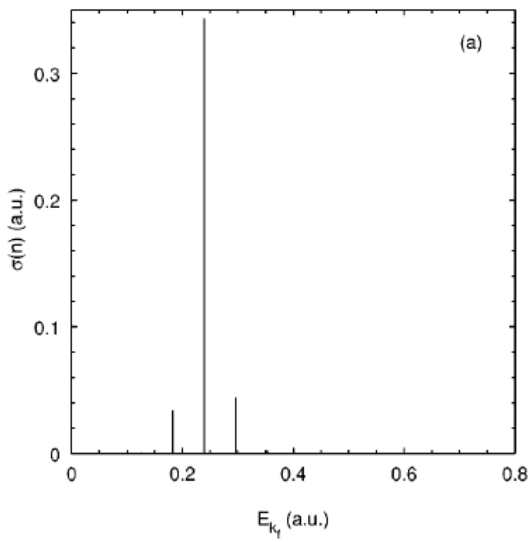
Laser Modification of X-Ray Photoionization



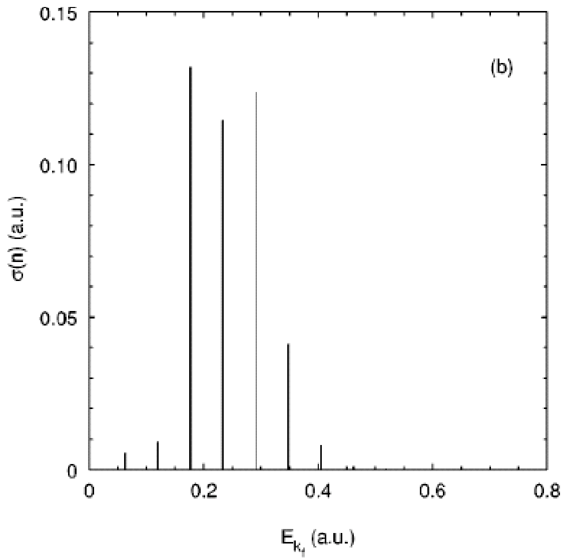
Total Cross Sections: Sideband Intensity v Laser Dressing Field

Hydrogen, $E_L = 1.55$ eV, $E_x = 13\ E_L$

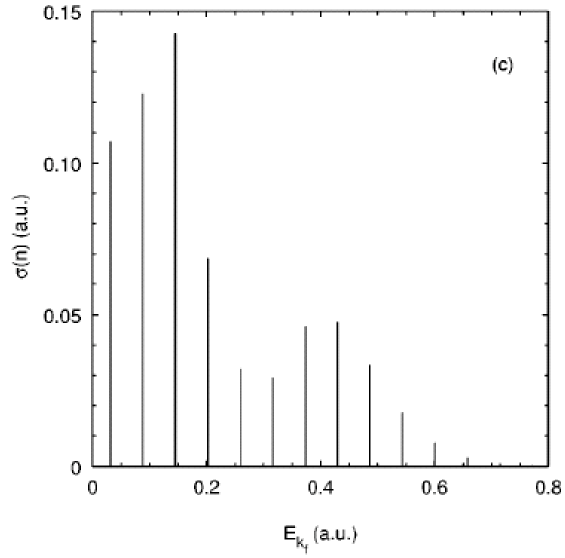
$I_L = 5 \times 10^{11}$ W/cm^2



$I_L = 3 \times 10^{12}$ W/cm^2



$I_L = 1.75 \times 10^{13}$ W/cm^2



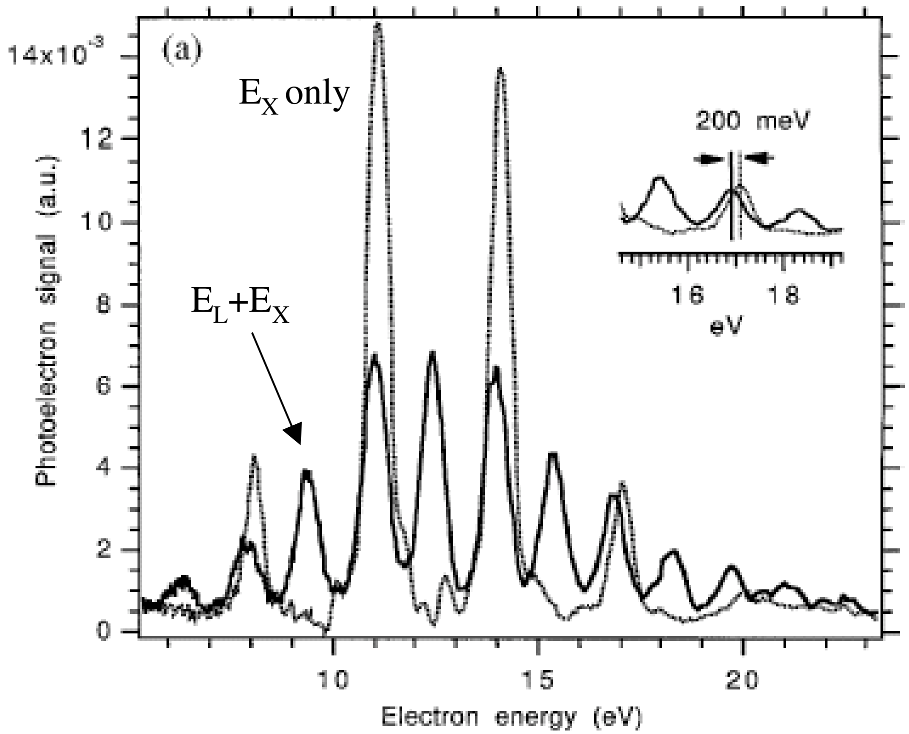
Véniard, Taïeb & Maquet PRL (1995)

Milosevic & Ehlotzky PRA (1998)

PRL (1996)

Observation of Laser Assisted Photoelectric Effect and Femtosecond High Order Harmonic Radiation

T. E. Glover,[1] R. W. Schoenlein,[1] A. H. Chin,[2] and C. V. Shank[1,2]
[1] *Materials Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory, Berkeley, California 94720*
[2] *Department of Physics, University of California at Berkeley, Berkeley, California 94720*
(Received 6 November 1995)



He: 4π collection of photoelectrons

E_L= 1.55 eV, E_X = $21E_L$ to $27E_L$

I_L = 7.4 x 10^{11} W/cm^2

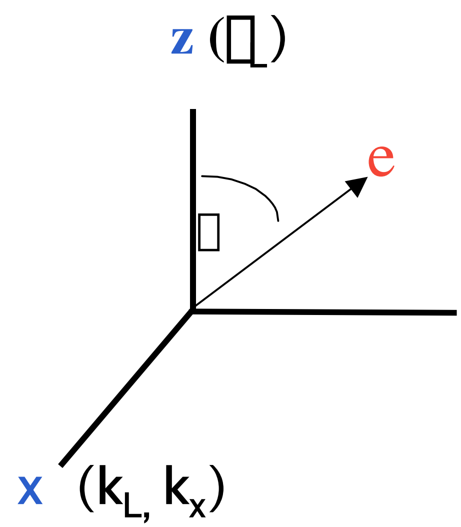
- Sideband intensity agrees well w/ simple model
- Sideband shift of photoelectron energies due to pondermotive energy

$E_e = E_X$ - IP - U_P

where $U_p = I_L/4E_L^2$

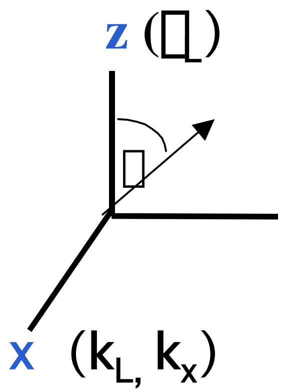
(U_p = 0.6 eV for 1.55 eV, 10^{13}W/cm^2)

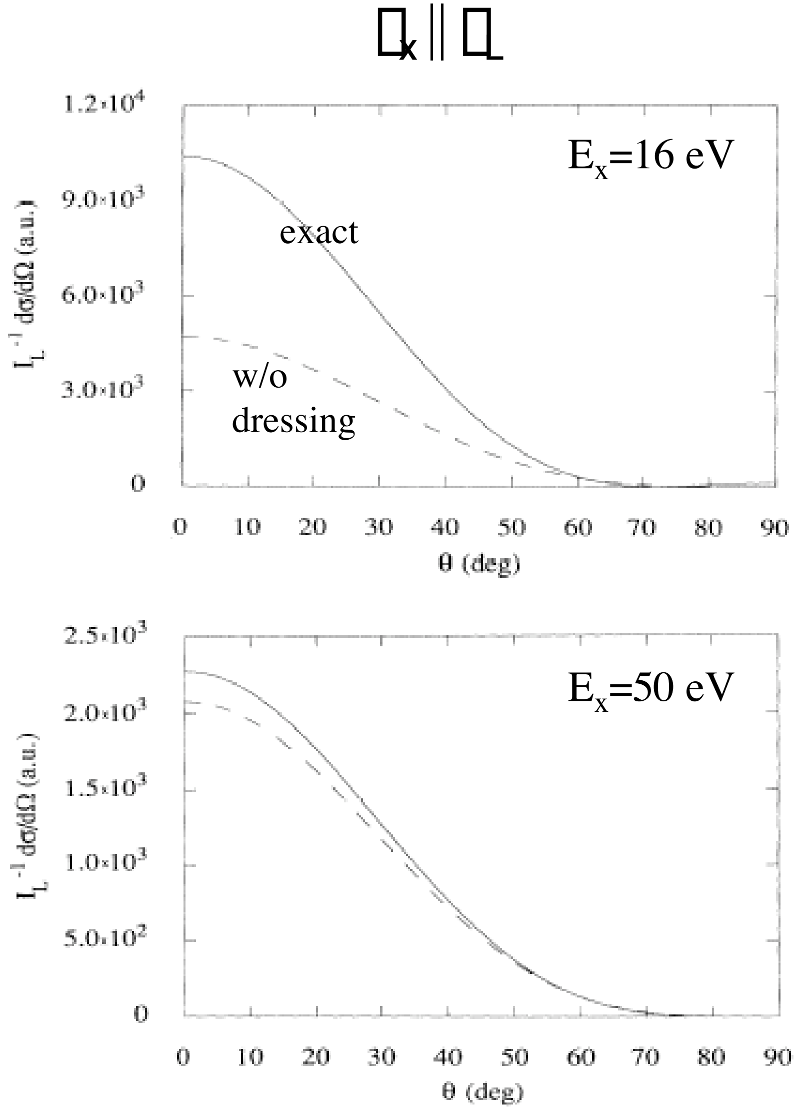
Angular distributions for x-ray + laser photoelectron spectroscopy



- Co-propagating laser and x-ray ($k_L \parallel k_x$)
- Polarization vectors either parallel or perpendicular: $\square_L, \square_x$

Perturbative regime: Target dressing effects





$E_L = 1.14$ eV

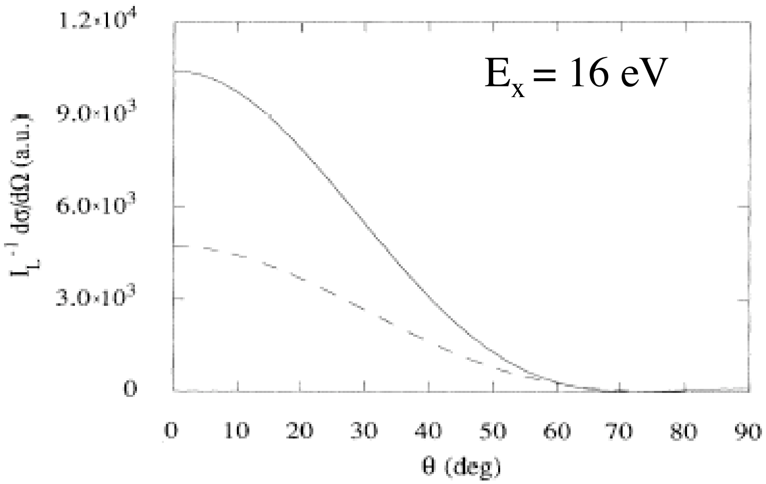
IP = 13.6 eV

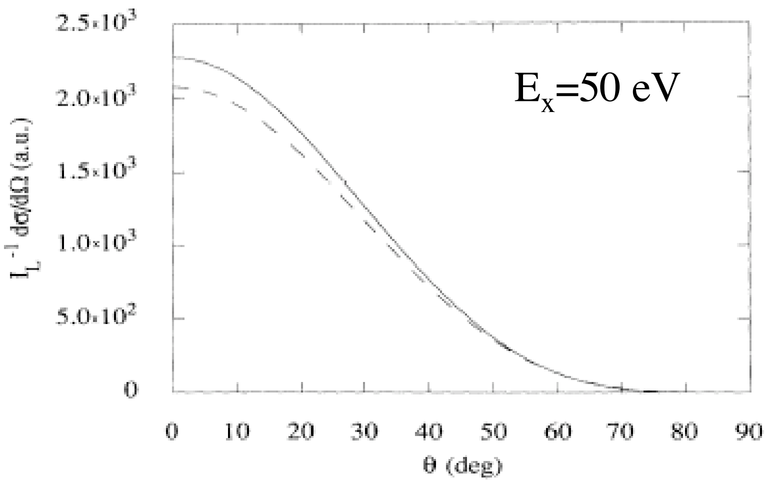
Cionga et al

PRA 47,1830 (1993)

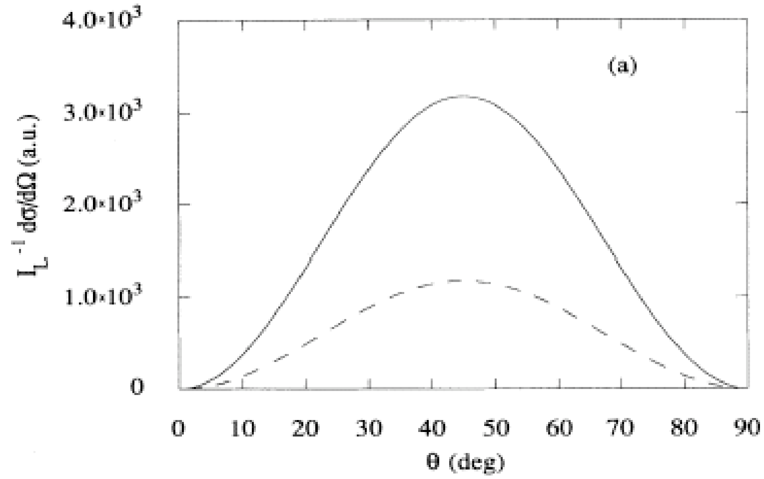
Relative polarization effects on angular distribution

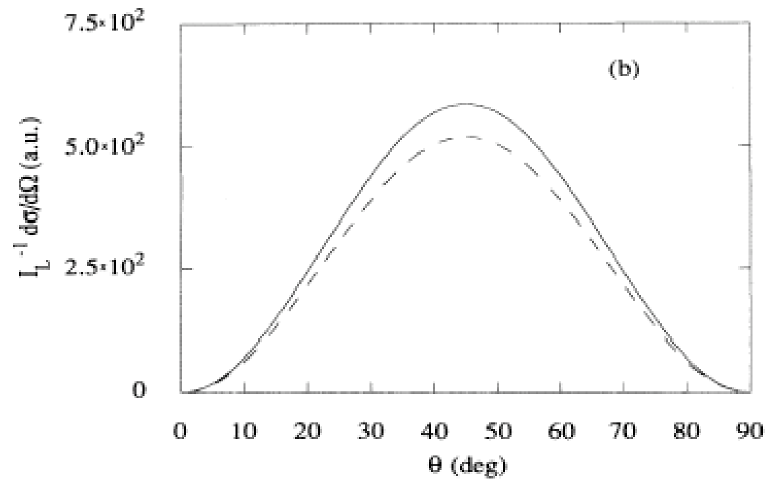
$\square_x \parallel \square_L$





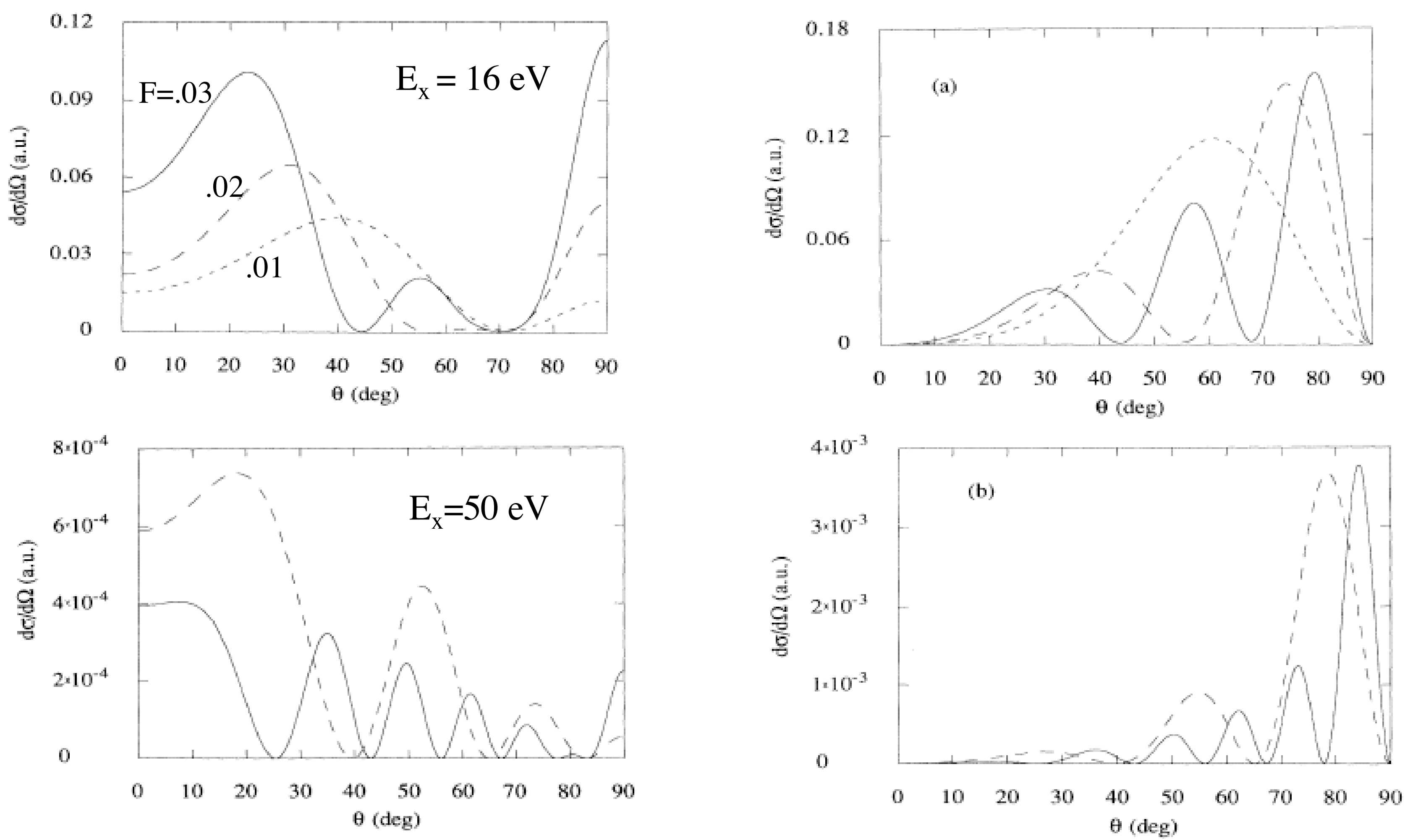
$\square_x \perp \square_L$





Polarization effects at higher dressing intensities

$\square_x \parallel \square_L$ $\square_x \perp \square_L$

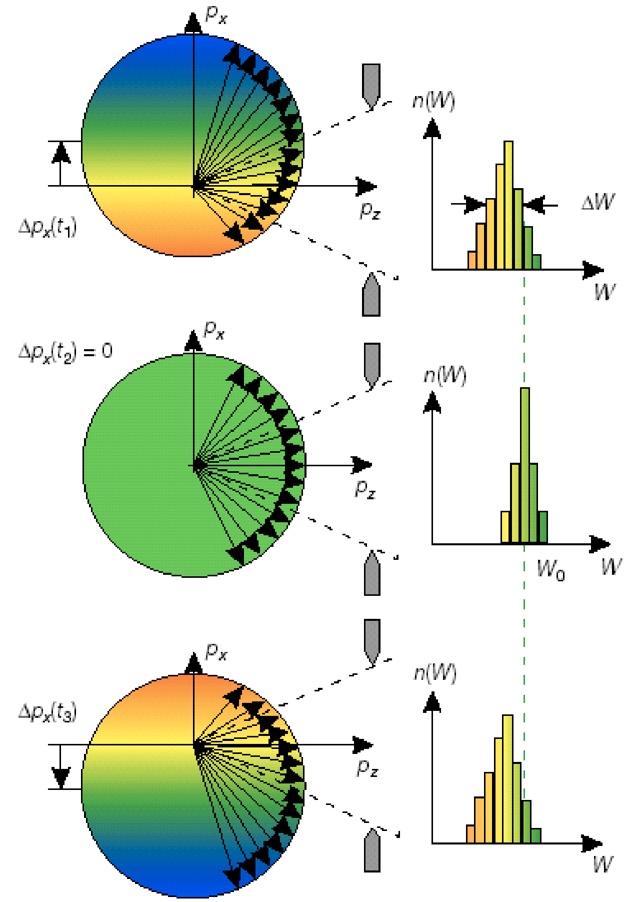


X-ray photoionization of laser-dressed atoms

- Attosecond metrology

  - control photoelectron energy by light field (not cycle-averaged intensity)
  - measure □W vs t_{delay} at “optimal” angle

Kr 4p photoelectron 60-80 eV
detection cone <40°
x- ray: 90 eV (≈14 nm)
650±150 as
optical: 1.6 eV (≈750 nm)
7 fs, 5×10^{13} W/cm²



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Considerations for APS X-ray photoionization of laser-dressed atoms

Possible projects:

- Photoelectron spectroscopy: map satellite intensities and angular distributions
- Measure threshold shift due to U_p directly
- Investigate near-threshold region

Technical Issues:

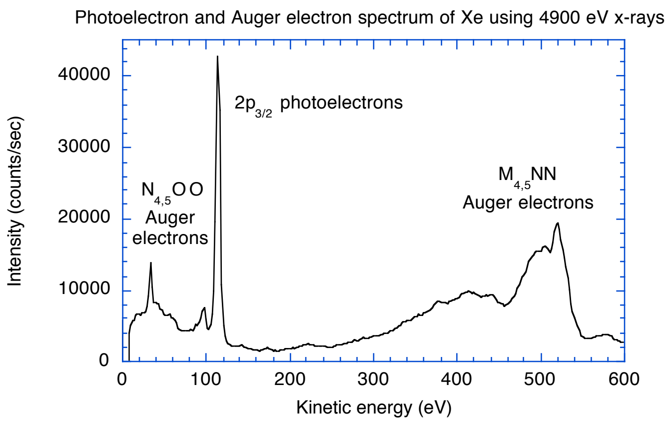
APS undulator: 5×10^{12} x-rays/s (20 bunch -> ≈ 10^6 x-rays /bunch) ≈ 60 ps
$\Delta E/E \approx 10^{-4}$ $3 < E < 30$ keV

Laser: Nd:YLF, 527 nm (2.35 eV), 1mJ/pulse, 1kHz, $\Delta t \approx 20$ ps
Ti:sapphire, 800 nm (1.55 eV), 1 mJ/pulse, 1 kHz, 100fs ≤ Δt

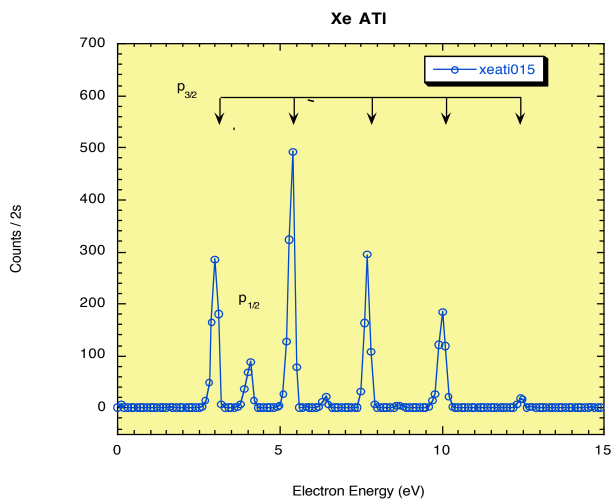
Photoelectron spectrometer (CMA): collection $\Omega = 0.07(4\pi)$
$\Delta E/E = 2.5\%$ $5 < E < 8000$ eV

Target: $nl \approx 10^{12}/cm^3$
Ar- K (3.2 keV, 100 kb), Xe-L (4.7-5.4 keV, 200 kb), Kr-K(14.3 keV, 20 kb)
Kr-L (1.6-1.9 keV, 600 kb)
$\sigma nl = 6 \times 10^{-7}$: 0.6 events/pulse x 1000 pulses/s x .07 = 42 cts/s

Examples: Electron Spectroscopy w/CMA at APS



BESSRC undulator



Nd:YLF laser
527 nm

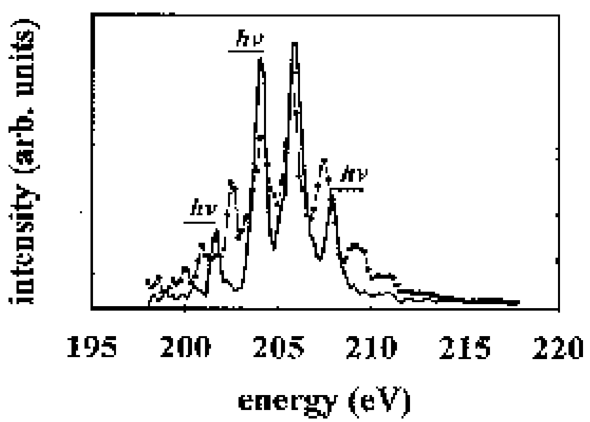
Steve Southworth, Dave Ederer, Anneli Munkholm,
Lin Chen, LindaYoung

Related & Ongoing Work

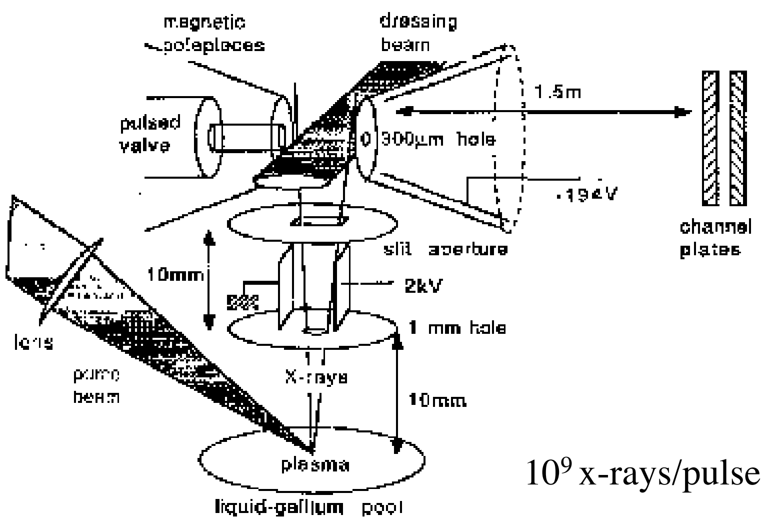
Laser-assisted Auger decay

J. M Schins *et al* PRL 73, 2180 (1994).

J. M. Schins *et al* PRA 52, 1272 (1995).



$I_L < 10^{12} W/cm^2$



10^9 x-rays/pulse

Modification of charge state distribution by laser dressing (ALS)

M. Hertlein, A. Belkacem, M. Prior, B. Feinberg, J. Roesch, J. Maddi, T.E. Glover, G. Ackerman (DAMOP2002)

Ion-electron coincidence spectroscopy (e^-: start, ion: stop): Rate <1 Hz, 10^4 x-rays/pulse

Charge state distributions in presence and absence of laser

Unexpected time dependence on nanosecond scale of charge state yield associated w/ electrons

Summary

- X-ray photoelectron spectroscopy of laser-dressed atoms feasible

  Tune x-ray energy -> cleaner spectra, near threshold effects

  Variable polarization -> map non-intuitive angular distributions

  Single x-ray + tunable -> directly map IP shift

  Hard x-rays -> complex atoms fit w/ simpleman's theory

- Transition to stronger-dressing fields when shorter x-ray pulses available

Ultrafast laser plasma x-ray sources and x-ray absorption spectroscopy in chemical research

Workshop on Ultrafast X-ray Science,
Advanced Photon Source, Argonne National Laboratory
June 2002

C. Rose-Petruck

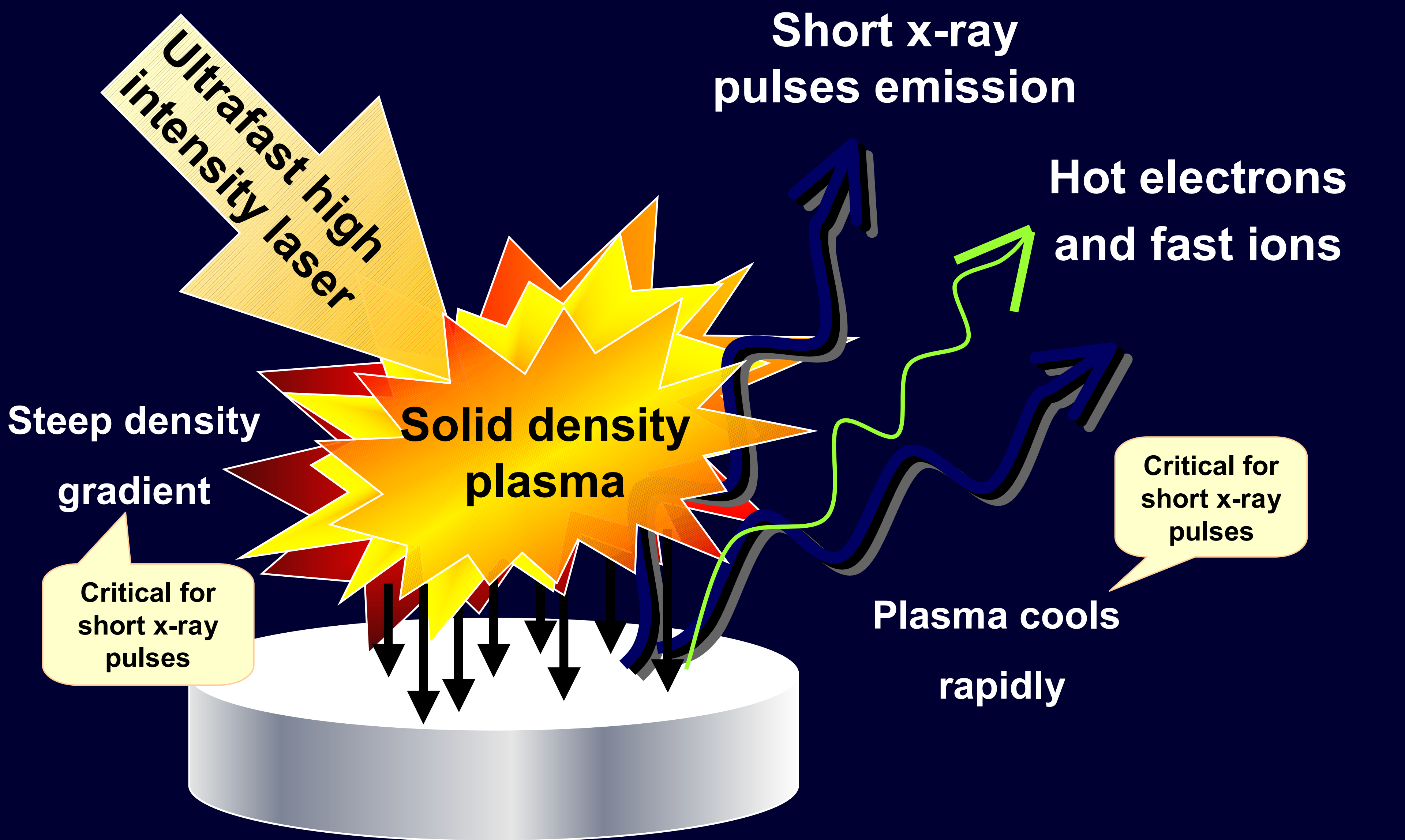
Department of Chemistry,
Brown University, Providence, R.I.

www.rosepetruck.chem.brown.edu

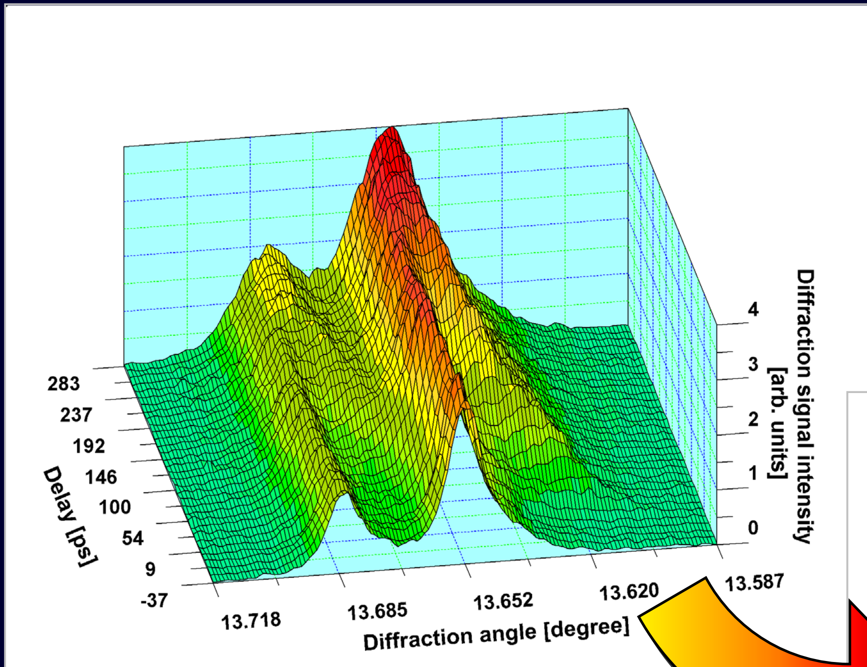
Outline

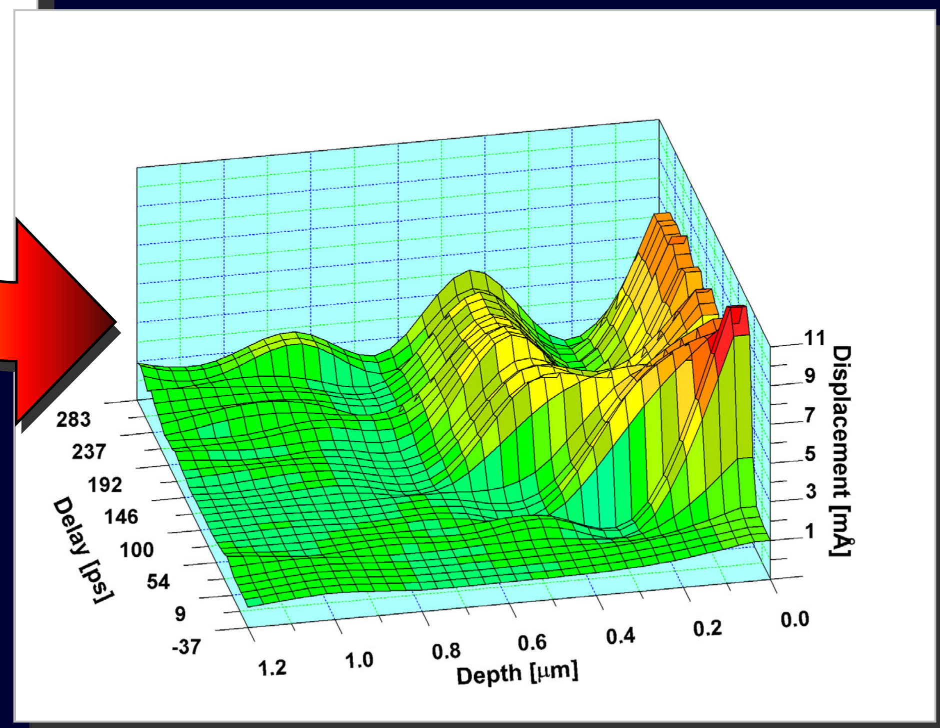
- High-brightness, tabletop x-ray sources
  - High-average power lasers for x-ray generation
  - Ultrafast x-ray sources
- X-ray and IR Absorption Spectroscopy of solvated $Fe(CO)_5$
  - X-ray absorption spectroscopy for Fe-CO distances
  - IR-spectroscopy of $Fe(CO)_5$
- Other X-ray applications

Ultrafast laser solid interaction



Picosecond-milliångström resolution structural dynamics in GaAs(111) single crystals





C. Rose-Petruck, R. Jimenez, T. Guo, A. Cavalleri, C. W. Siders, F. Raksi, J. A. Squier, B. C. Walker, K. R. Wilson, C. P. J. Barty, Nature 398, 310 - 312 (1999)

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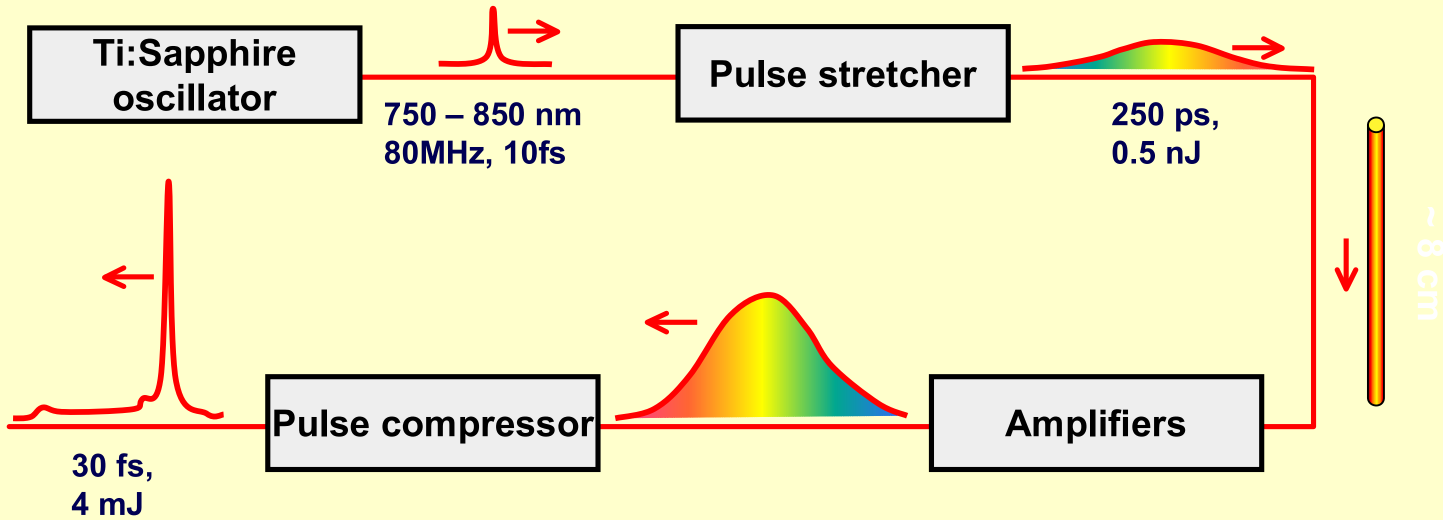
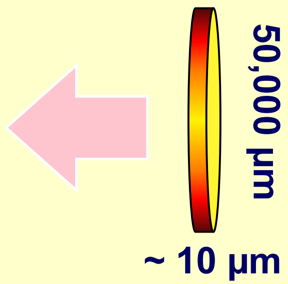
Desirable properties of laser systems for ultrafast x-ray generation

- Laser pulses: ~40 fs, ~ 4 mJ / pulse
  - Laser pulse interaction with solid density plasma
  - Intensity >10^{17} W/cm^2
- High ns, ps-pulse contrast
- Low ASE
- Good long-term stability and low shot to shot fluctuations, in particular of the beam profile
- Turn-key operation

Chirped Pulse Amplification laser system



Pancake of light

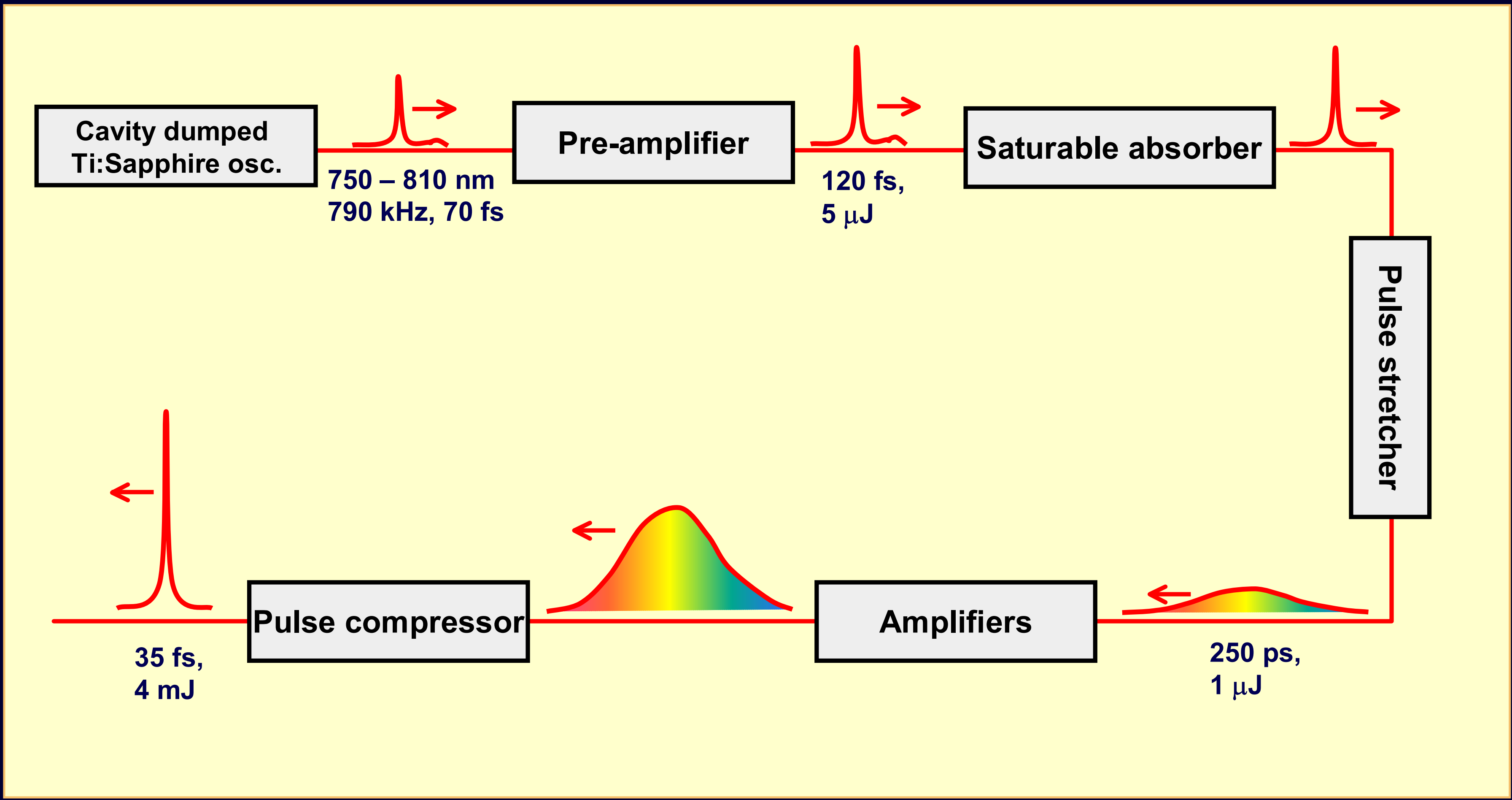


Important laser characteristics:

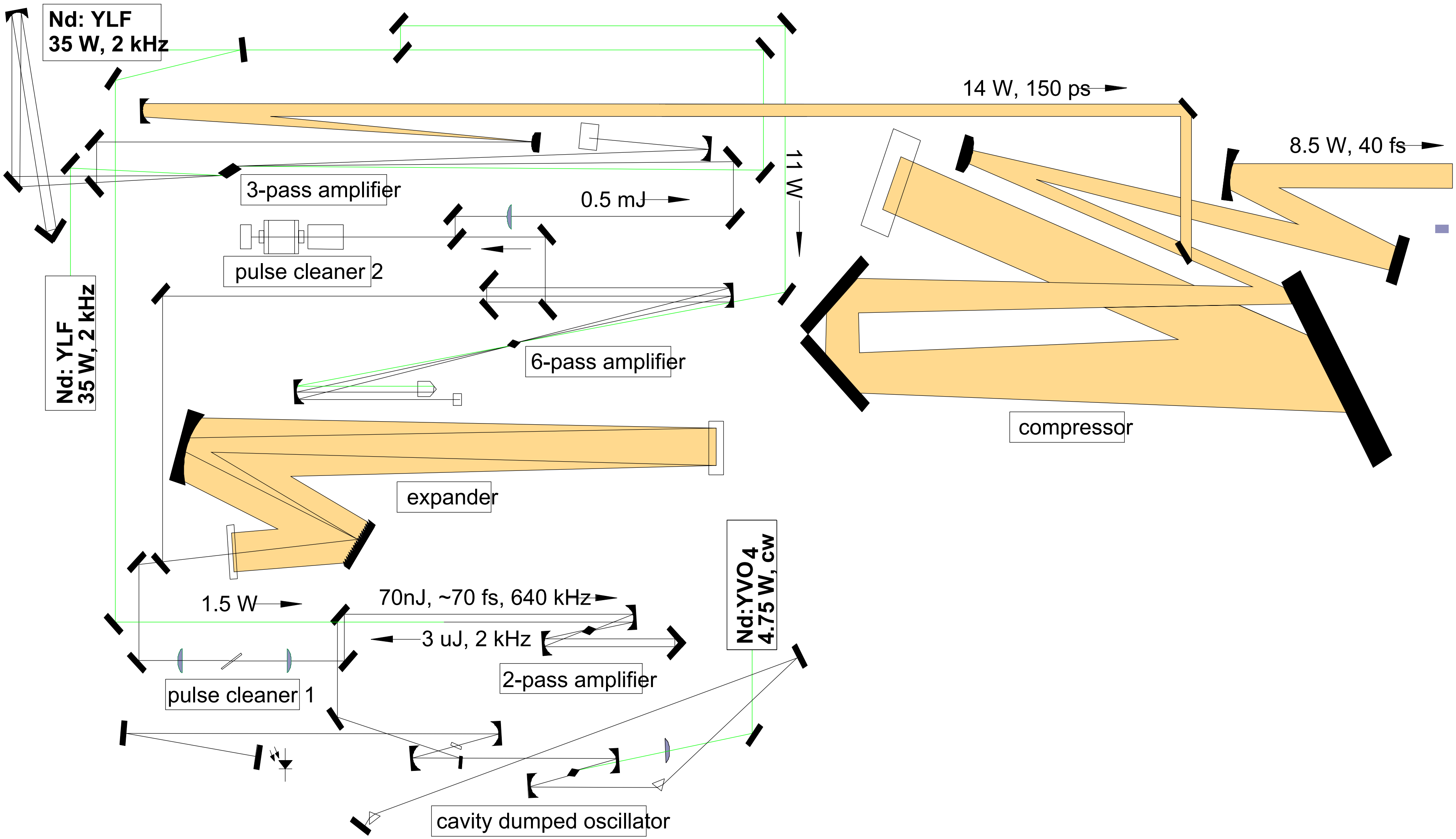
Maintain broad spectral band width during amplification

Precise dispersion control

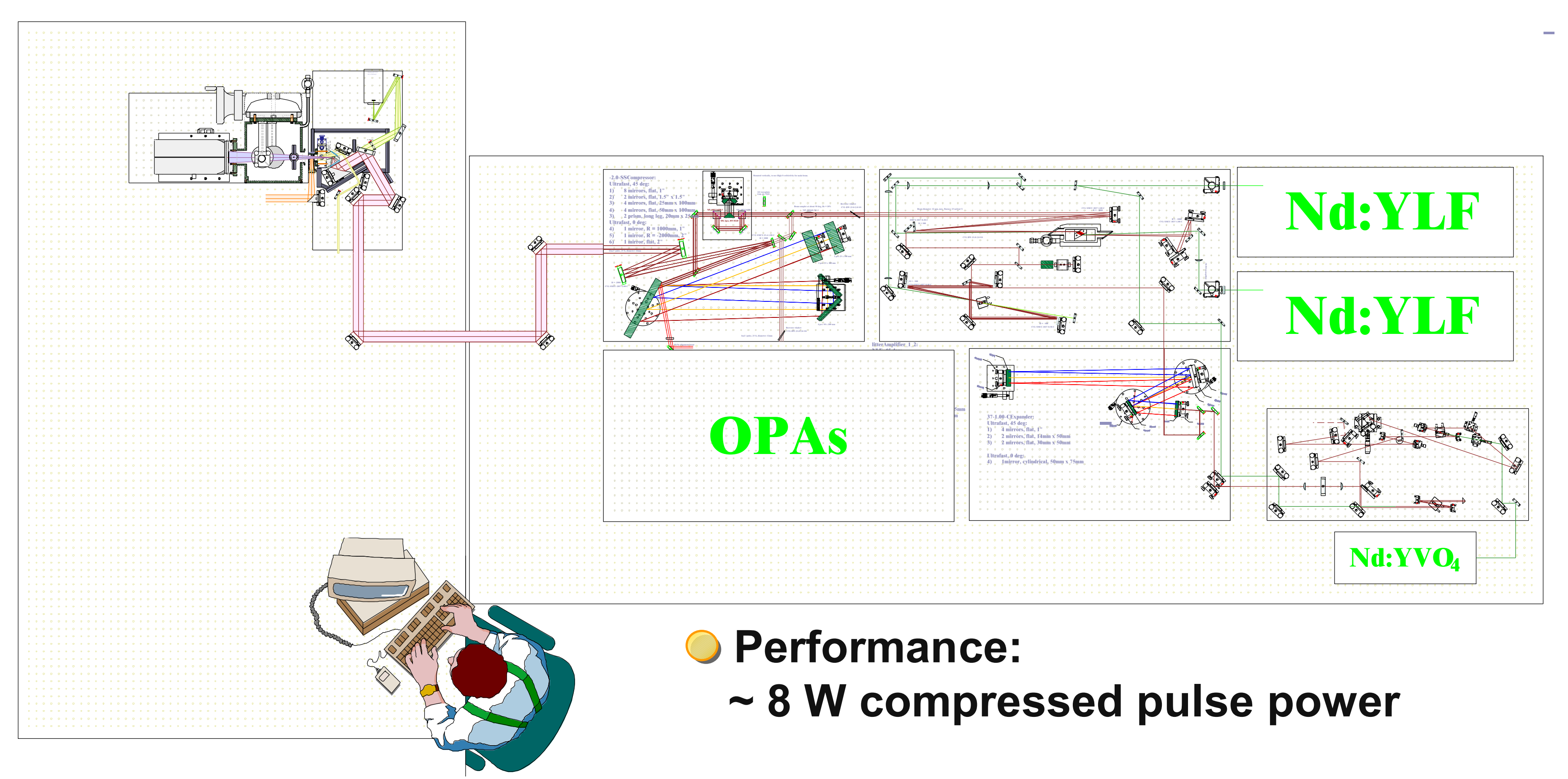
CPA laser system with high-intensity pulse injection



Chirped Pulse Amplification Laser

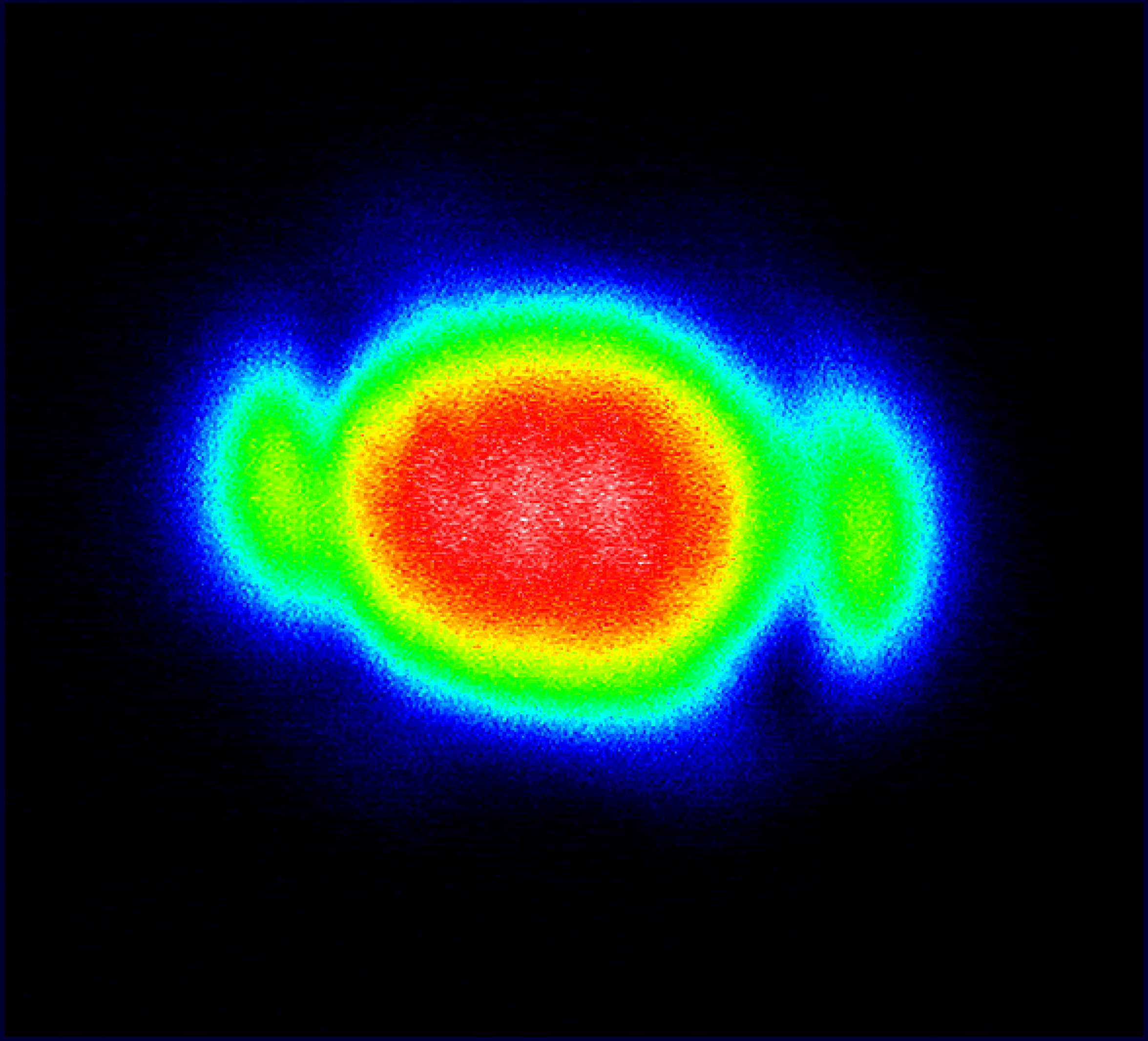


Ultrafast lasers for laboratory-based plasma x-ray sources can be compact

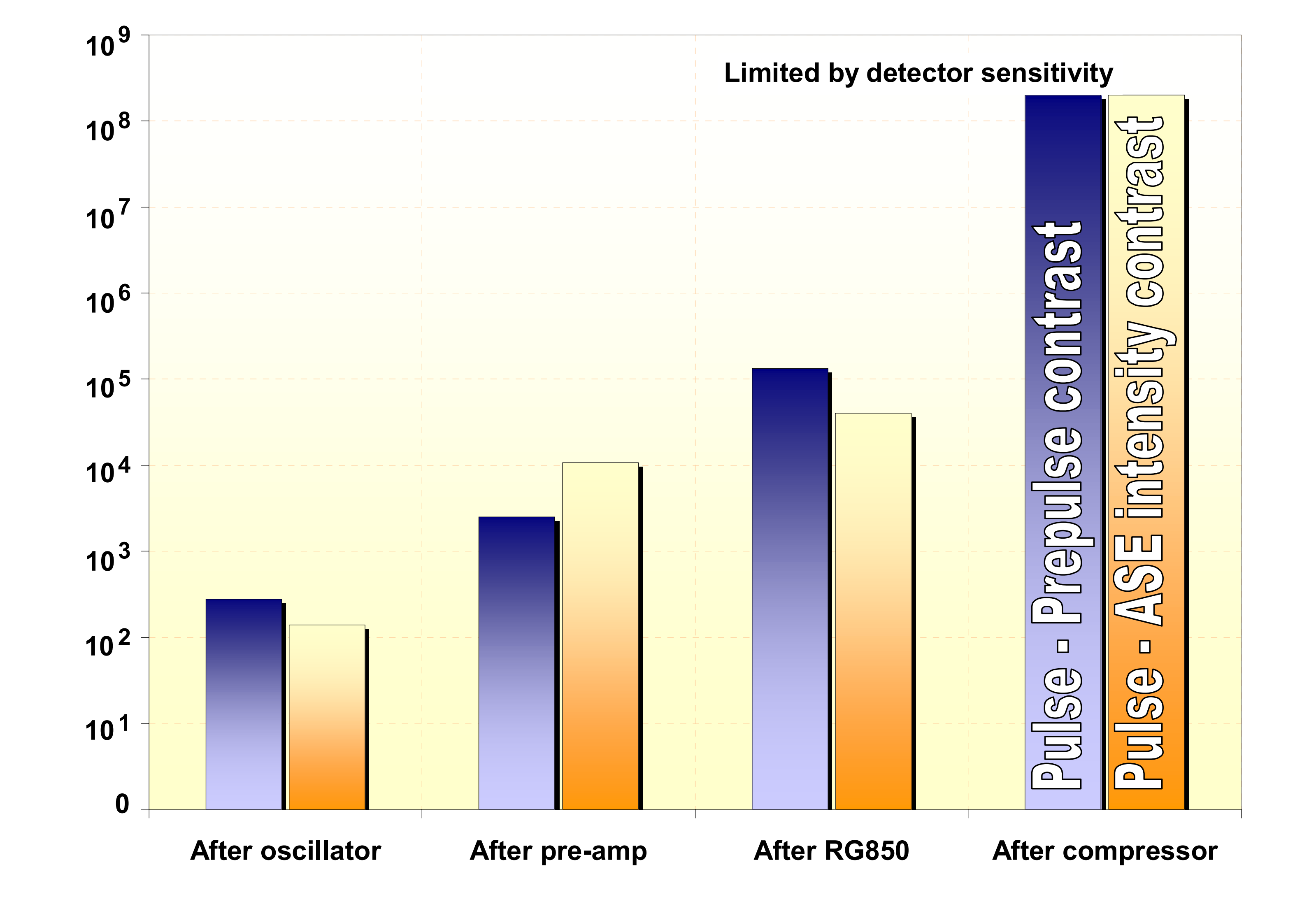


- **Performance:**
  ~ 8 W compressed pulse power

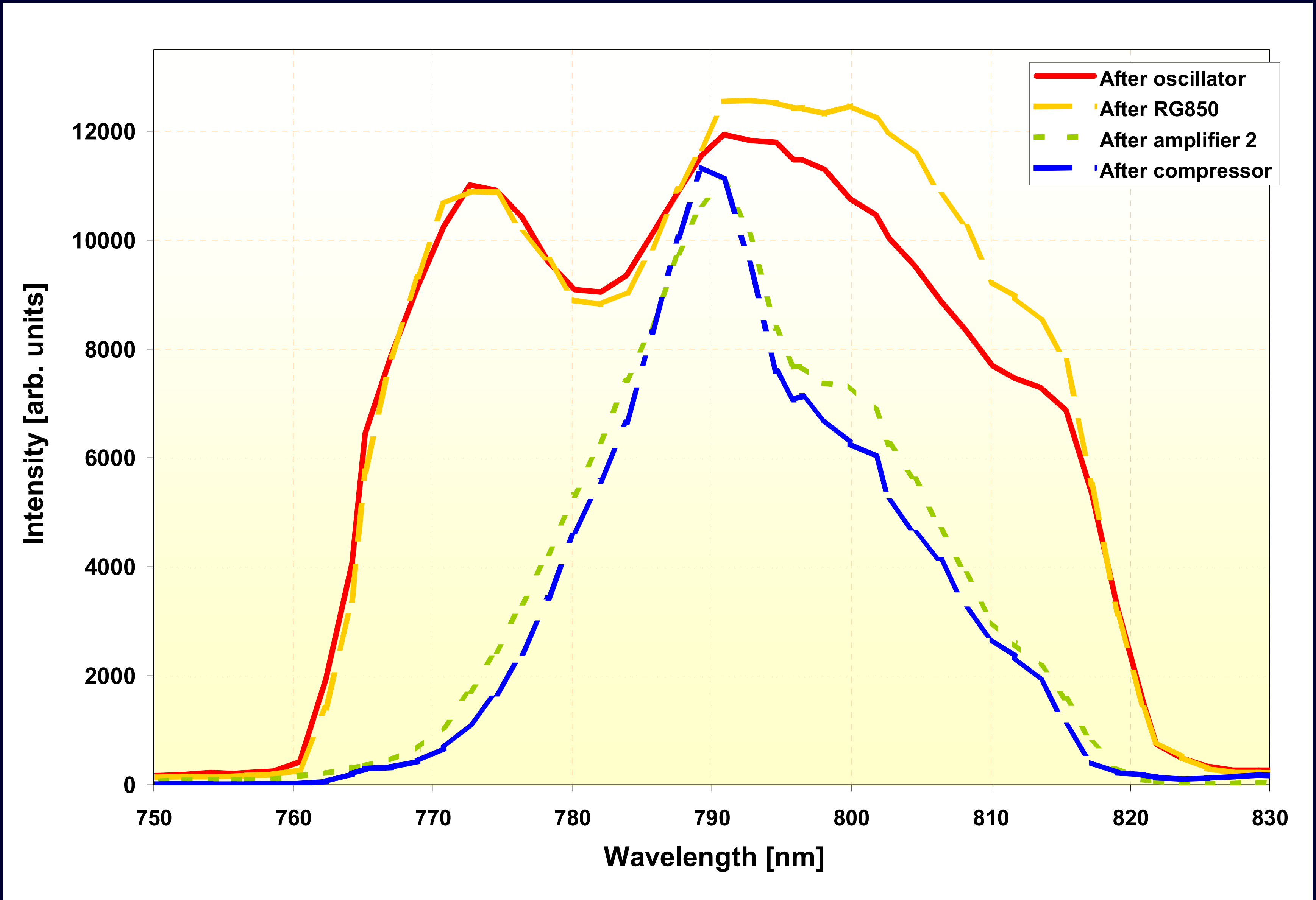
Beam profile of the compressed beam at 8.5 W



Pre-pulse and ASE intensity contrast at various locations in the laser system



Laser Spectra at various locations in the laser system



Ultrafast x-ray pulses for x-ray absorption spectroscopy

Laser pulse 800 nm, 40 fs, 4 mJ, 2KHz

Average laser power before compr. 11W

Average compressed power 7W

Focus diameter ~ 10 μm

Laser intensity ~ 1×10^{17} W/cm^2

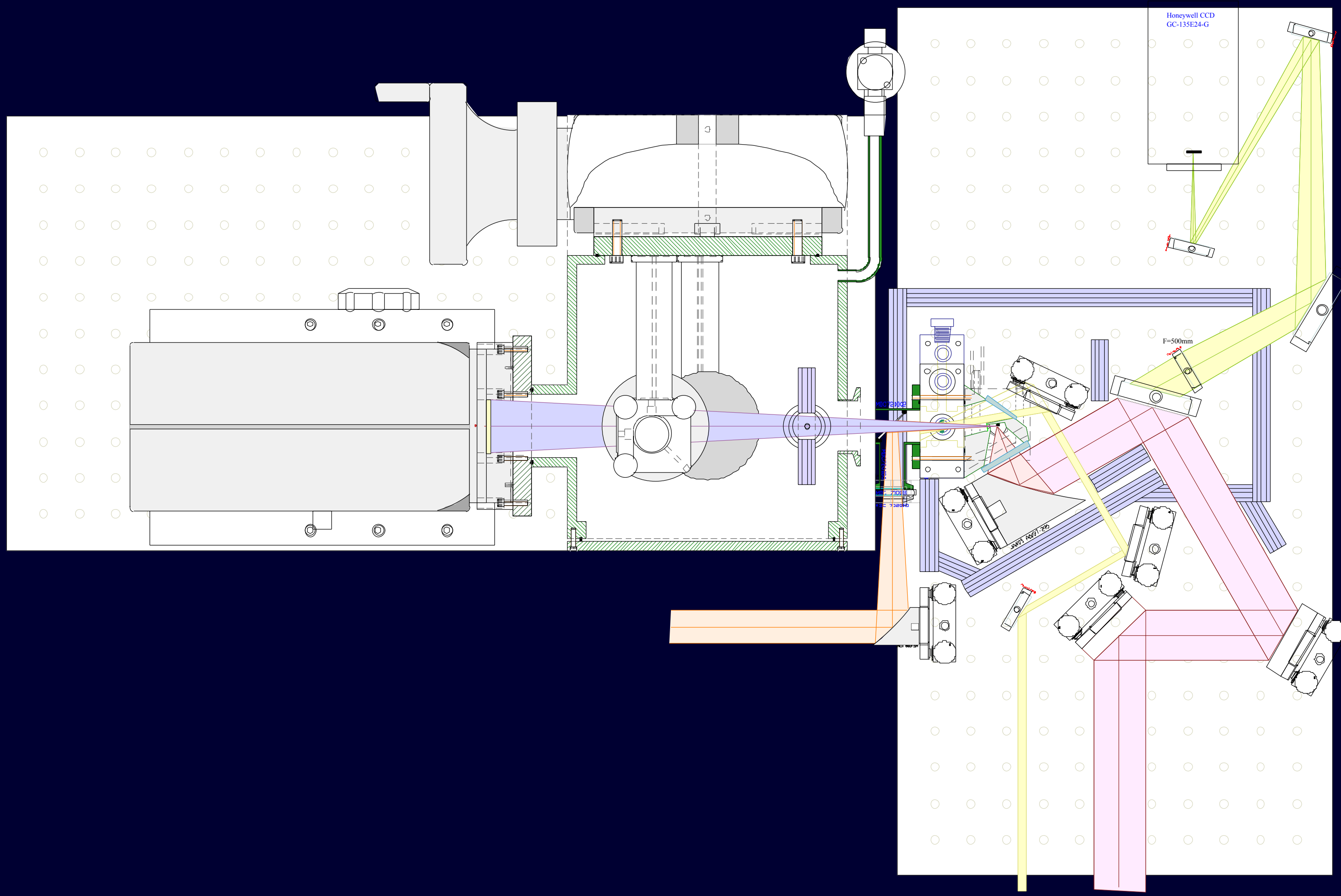
X-ray flux @ 8keV ~10^8 photons / (s 4π sr)

X-ray flux @ 3keV ~10^{13} photons / (s 4π sr keV)

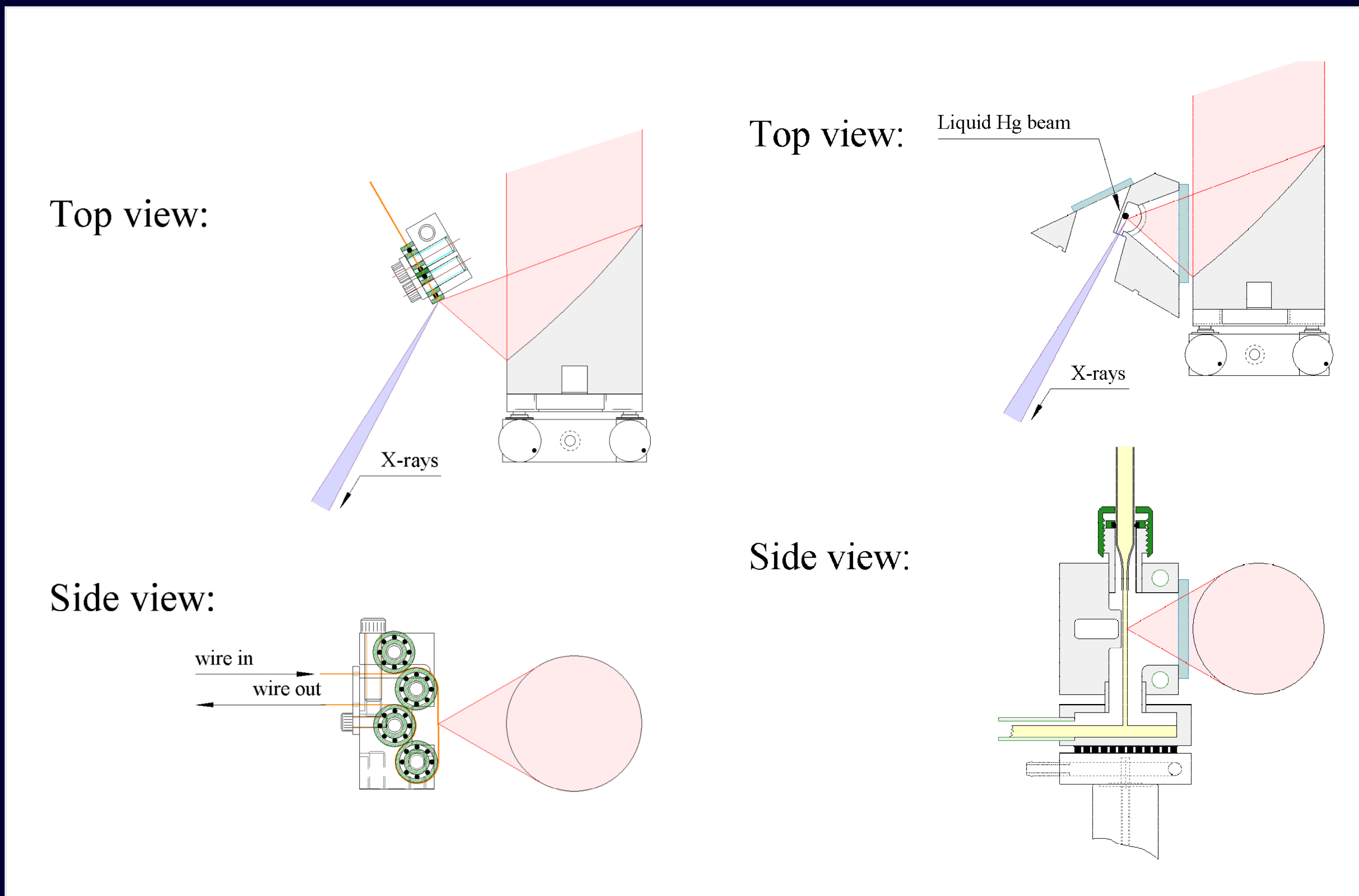
Conversion efficiency 2×10^{-6} % @ 8keV

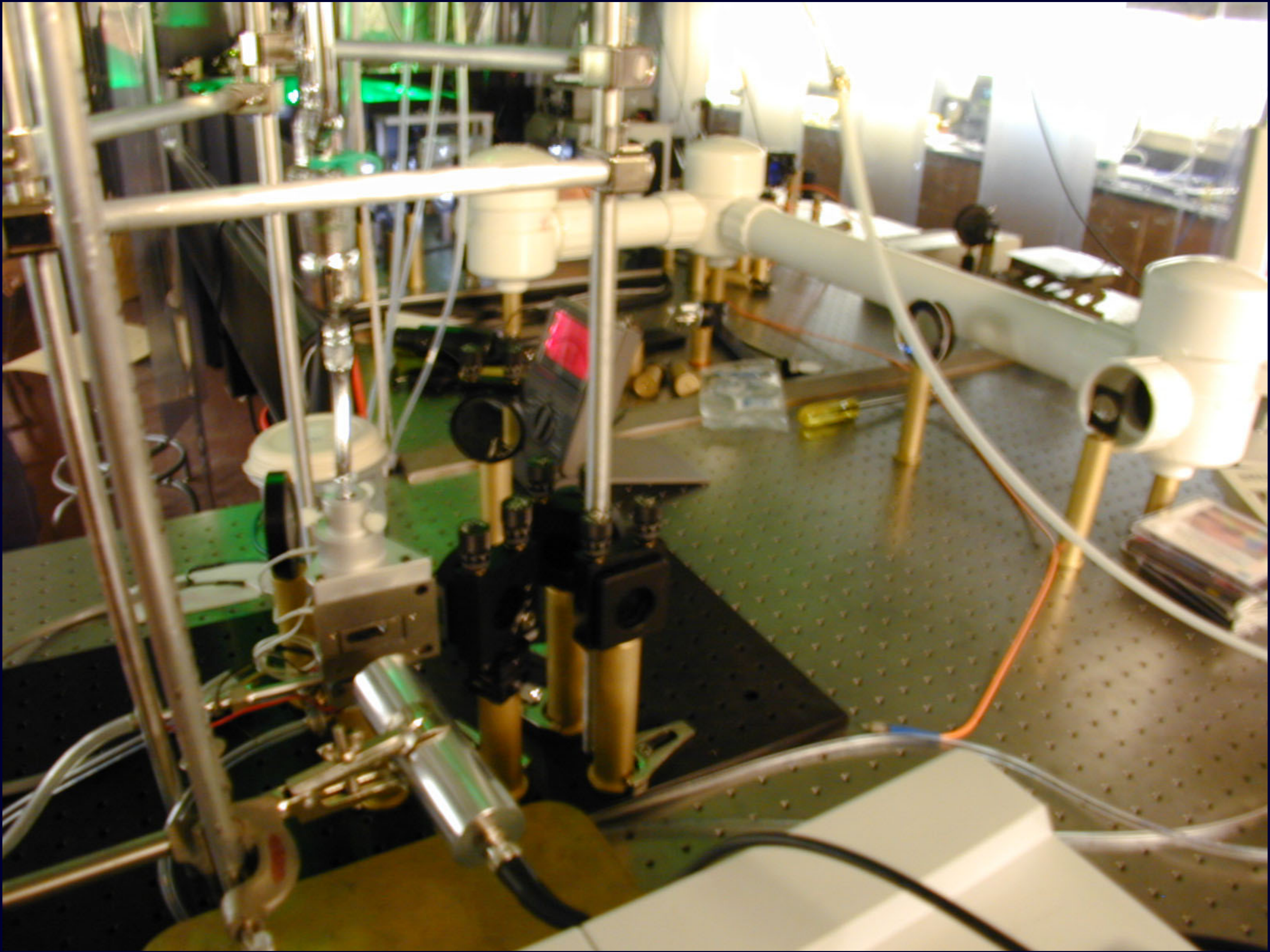
0.5 % @ 3keV

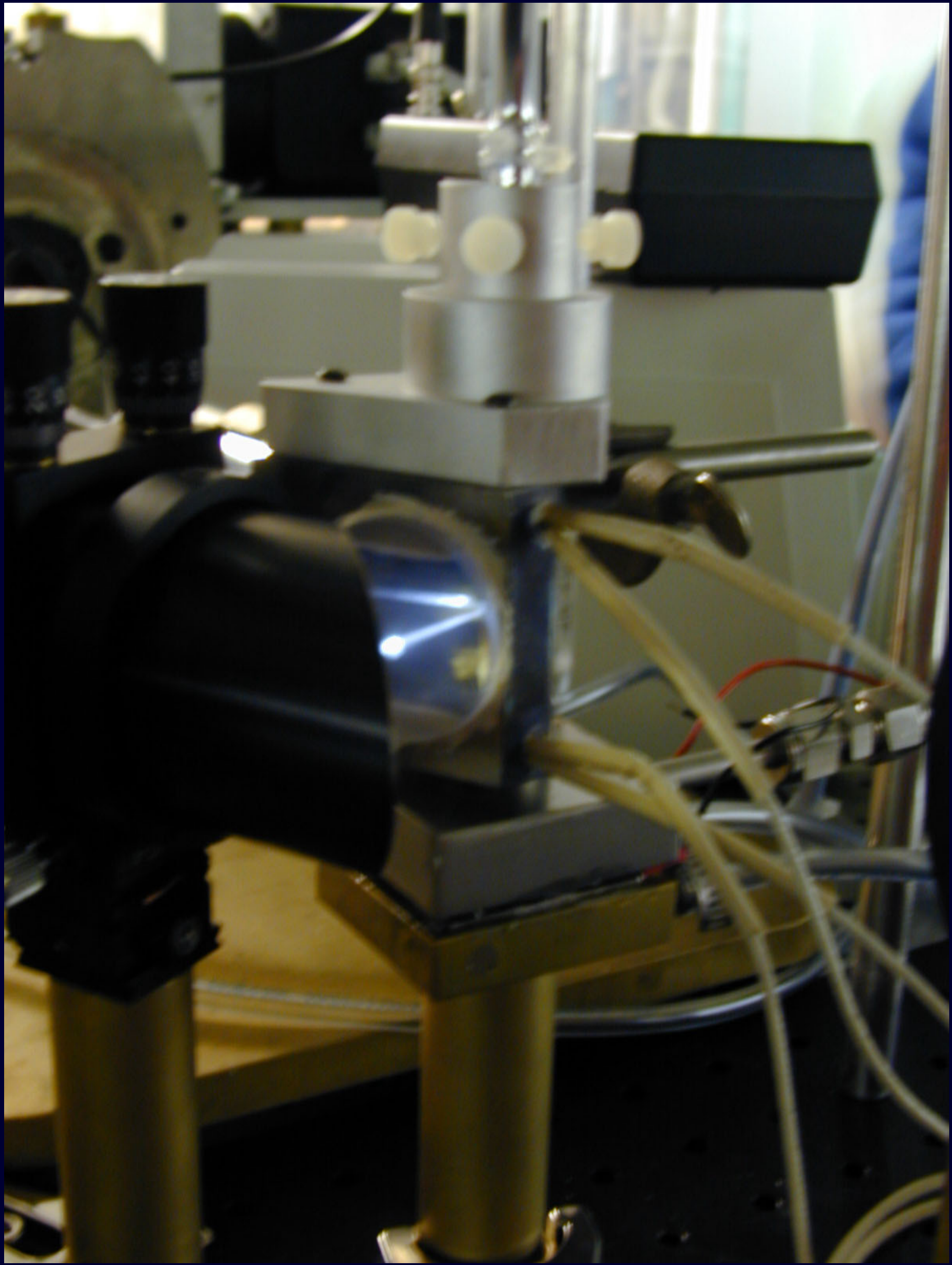
Ultrafast x-ray absorption spectrometer



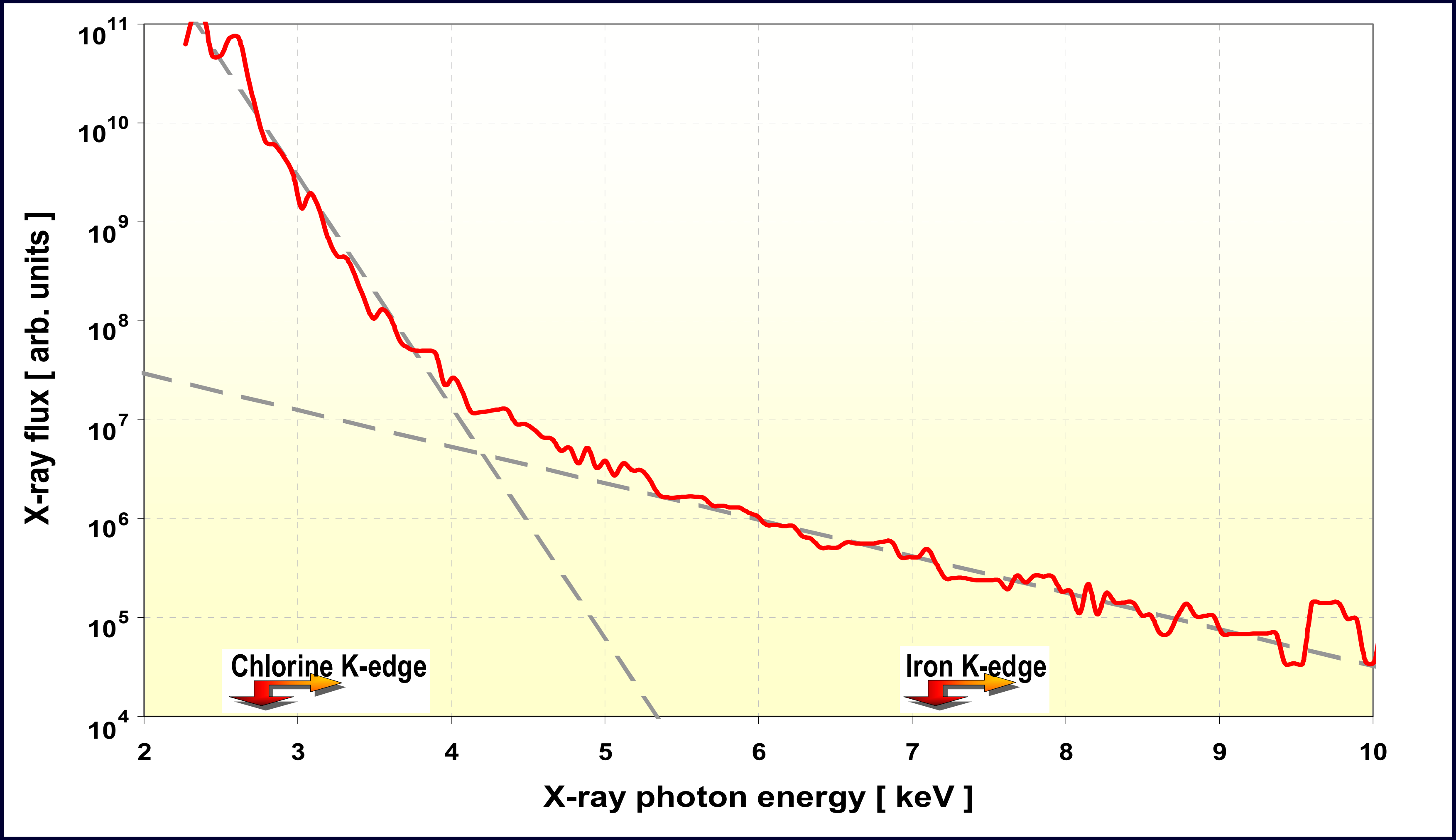
Design of solid and liquid metal targets





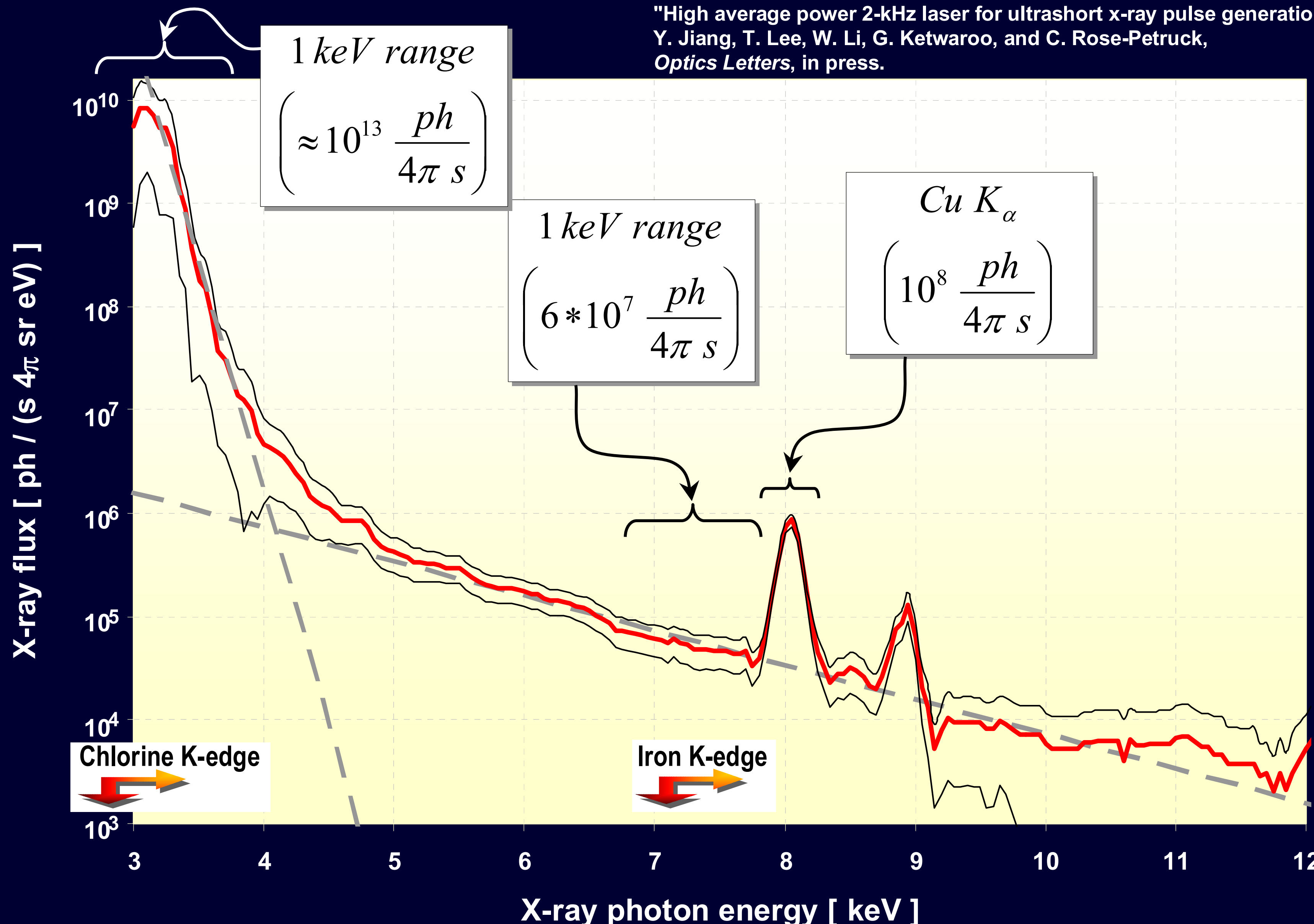


X-ray emission spectrum measured from a liquid mercury target in Ne



X-ray emission spectrum of 2-kHz source

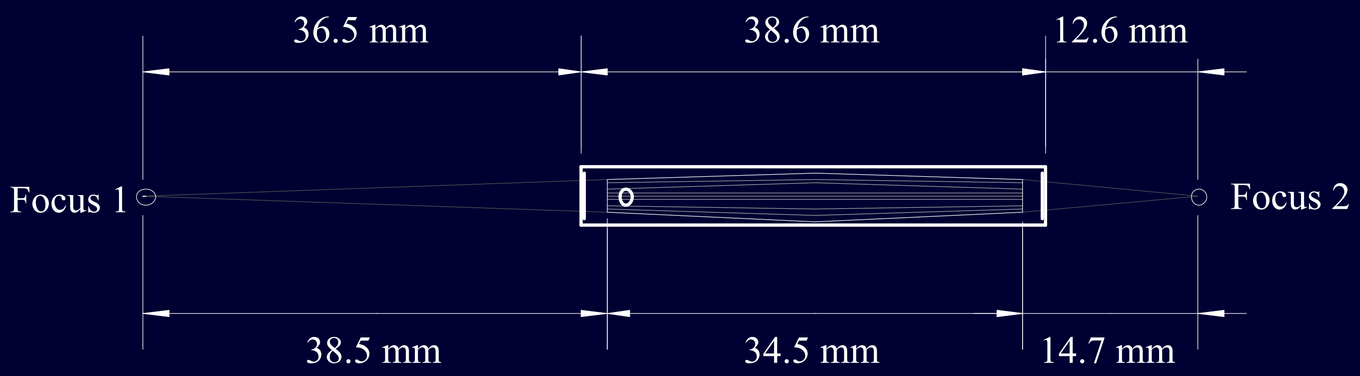
"High average power 2-kHz laser for ultrashort x-ray pulse generation", Y. Jiang, T. Lee, W. Li, G. Ketwaroo, and C. Rose-Petruck, *Optics Letters*, in press.



Comparison of various x-ray sources for x-ray absorption spectroscopy

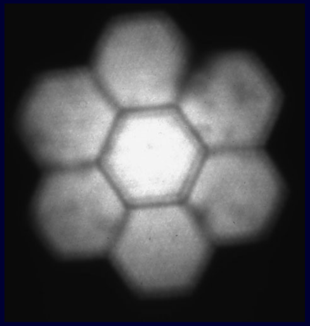
X-ray absorption spectrometer with ...	Average x-ray flux on sample [ph / keV s]	Peak x-ray flux on sample [ph / ps keV s]
12 kW rotating anode tube	6×10^{9}	6×10^{-3}
3rd generation synchrotron @ 1kHz pump laser rep. rate	$10^{9} – 10^{13}$ (sbm)	$10^{7} – 10^{11}$ (sbm)
Tabletop ultrafast x-ray sources (10 W)	10^{9}	$10^{9} – 10^{10}$

X-ray lens mounted on 3D Translation Stage

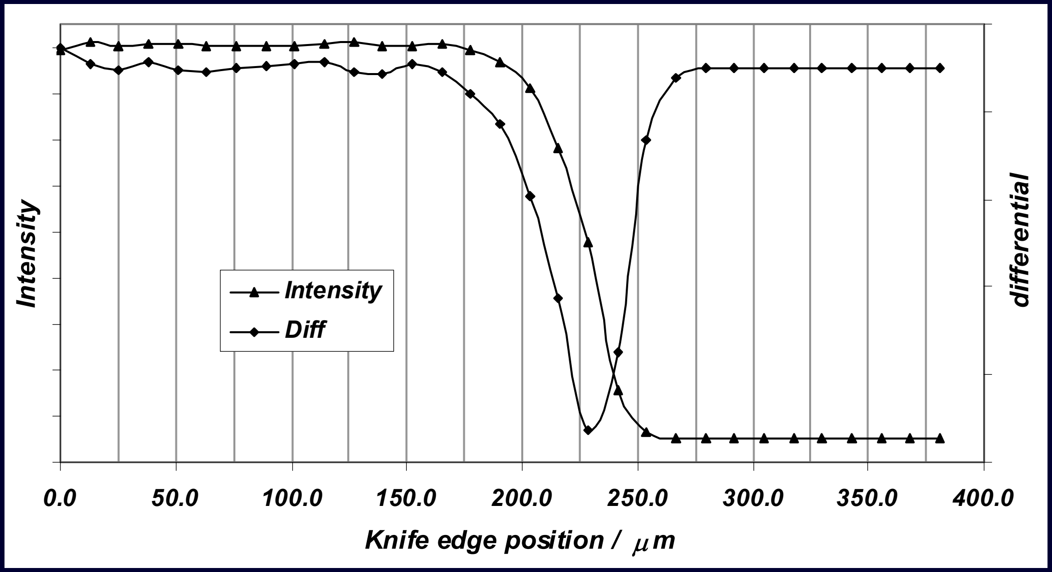


	Focus 1	Focus 2
Focal Length	38.5±0.5 mm	14.7±0.1 mm
Diameter	1.7 mm	1.4 mm
ϕ	0.044 rad / 2.5°	0.095 rad / 5.4°

Performance Test of X-ray lens



Pattern of the X-rays transmitted through the X-ray lens



Knife edge experiment to measure the focal spot size.
The experiment is performed at 10 kV, 0.1 mA, with 1 second exposure time.
Focal spot size ~35um (33um at 6.5~10.2kV)

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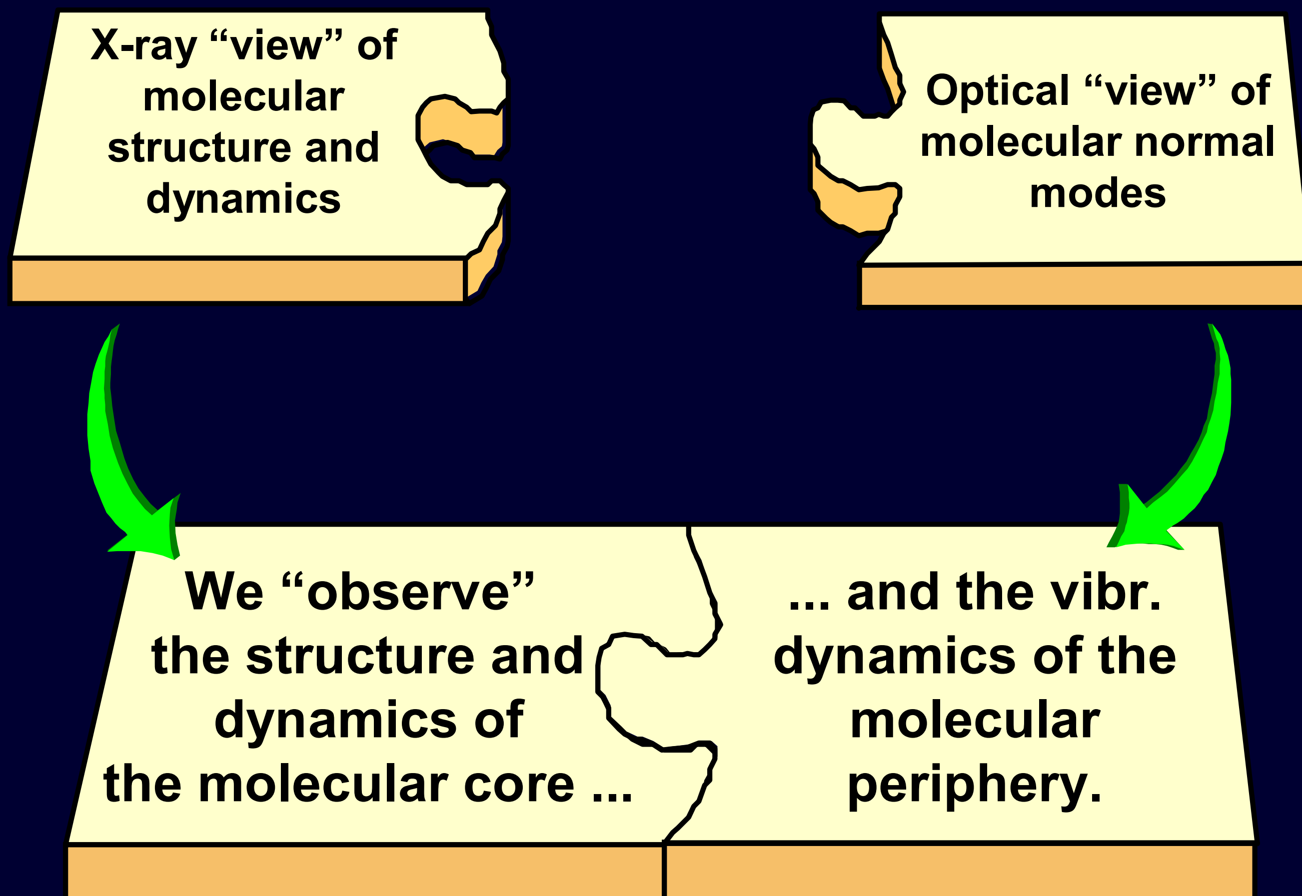
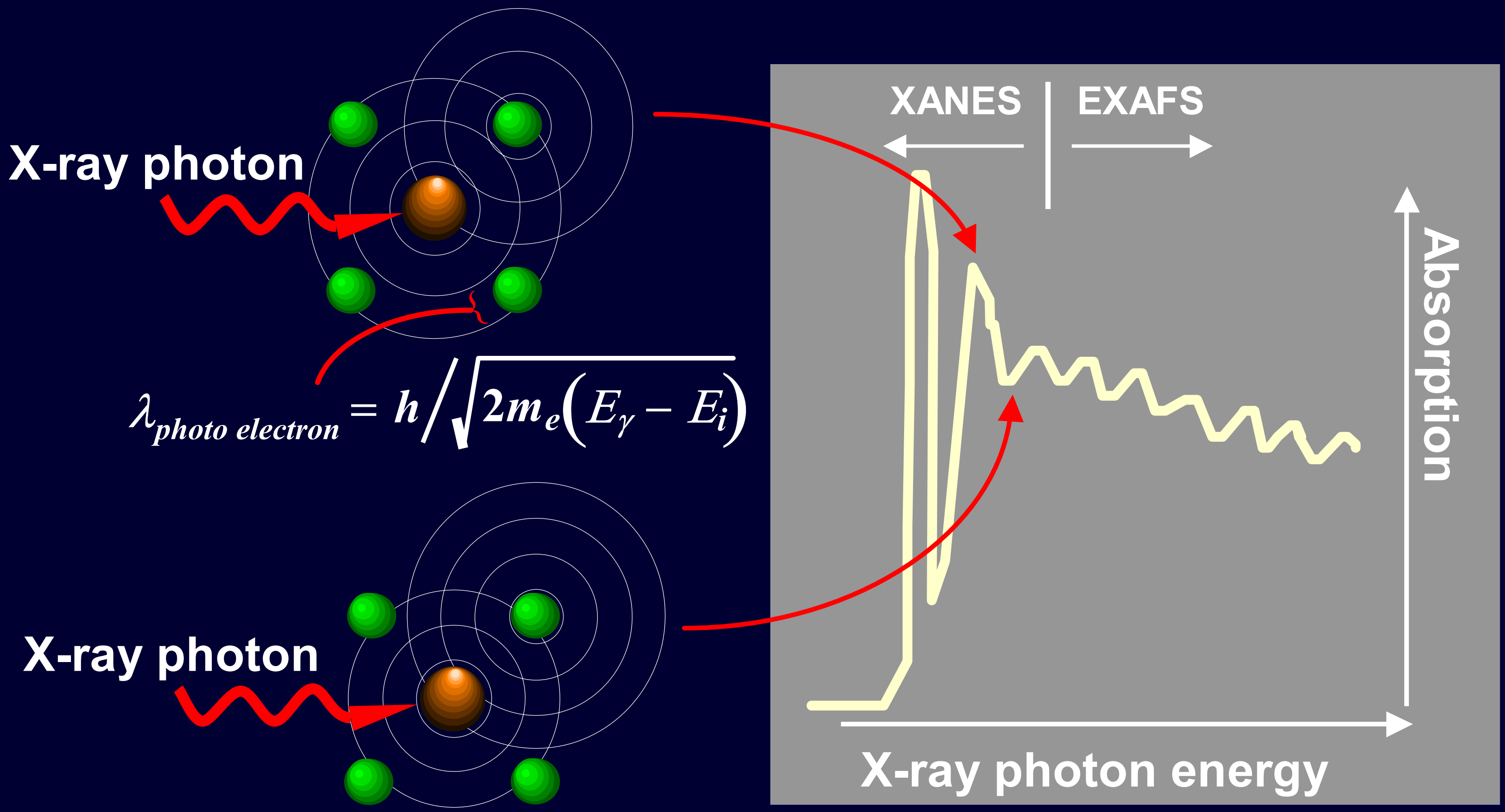
X-ray and IR absorption spectroscopy of solvated molecules

X-ray and infrared spectroscopy: Complementary methods probing structural dynamics and molecular normal modes

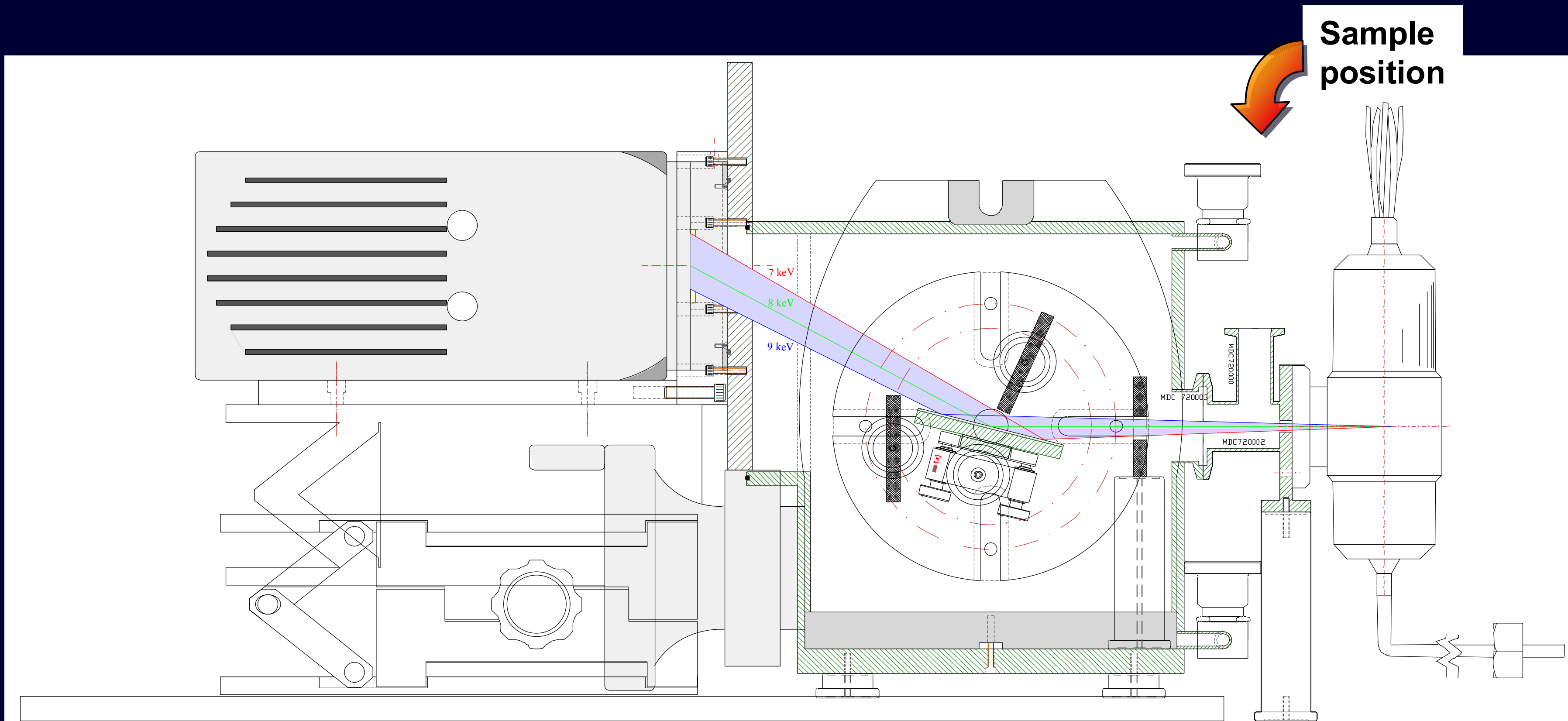


X-ray absorption spectroscopy probes the local environment

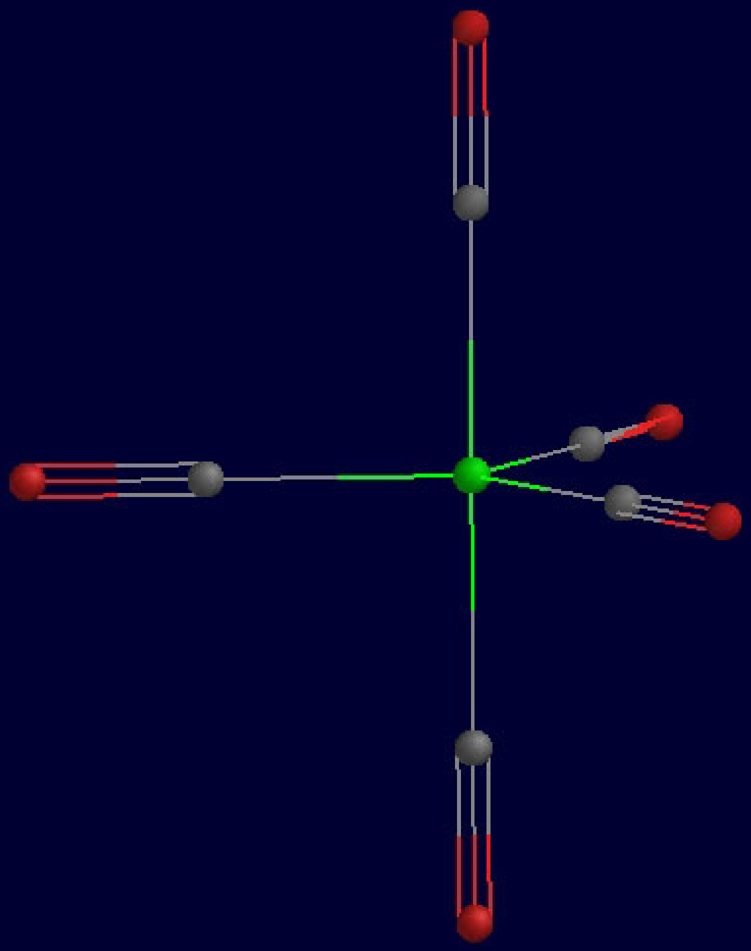


$$\lambda_{photo\ electron} = h \Big/ \sqrt{2m_e\left(E_\gamma - E_i\right)}$$

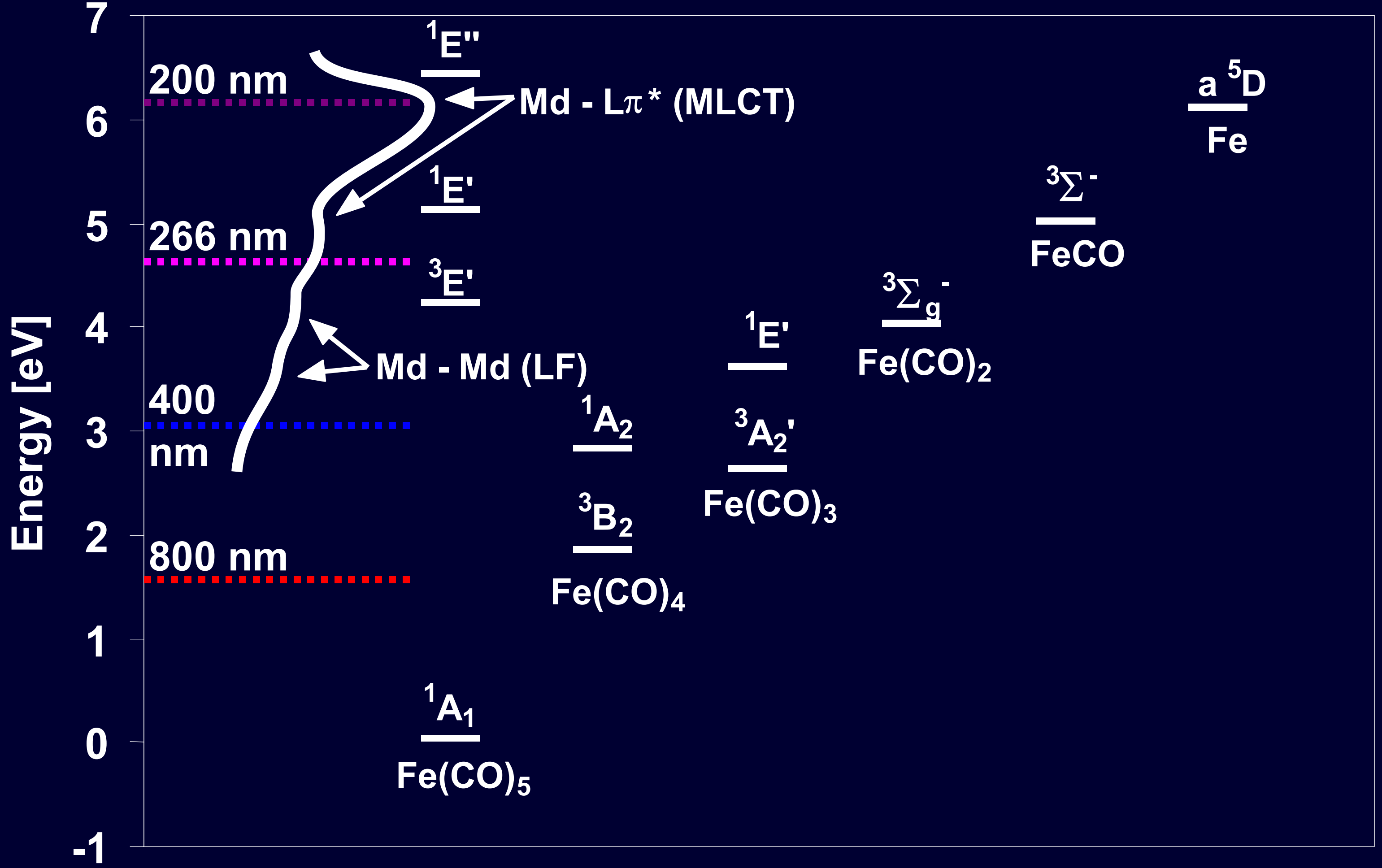
Setup for Single Crystal Static X-Ray Absorption Measurement



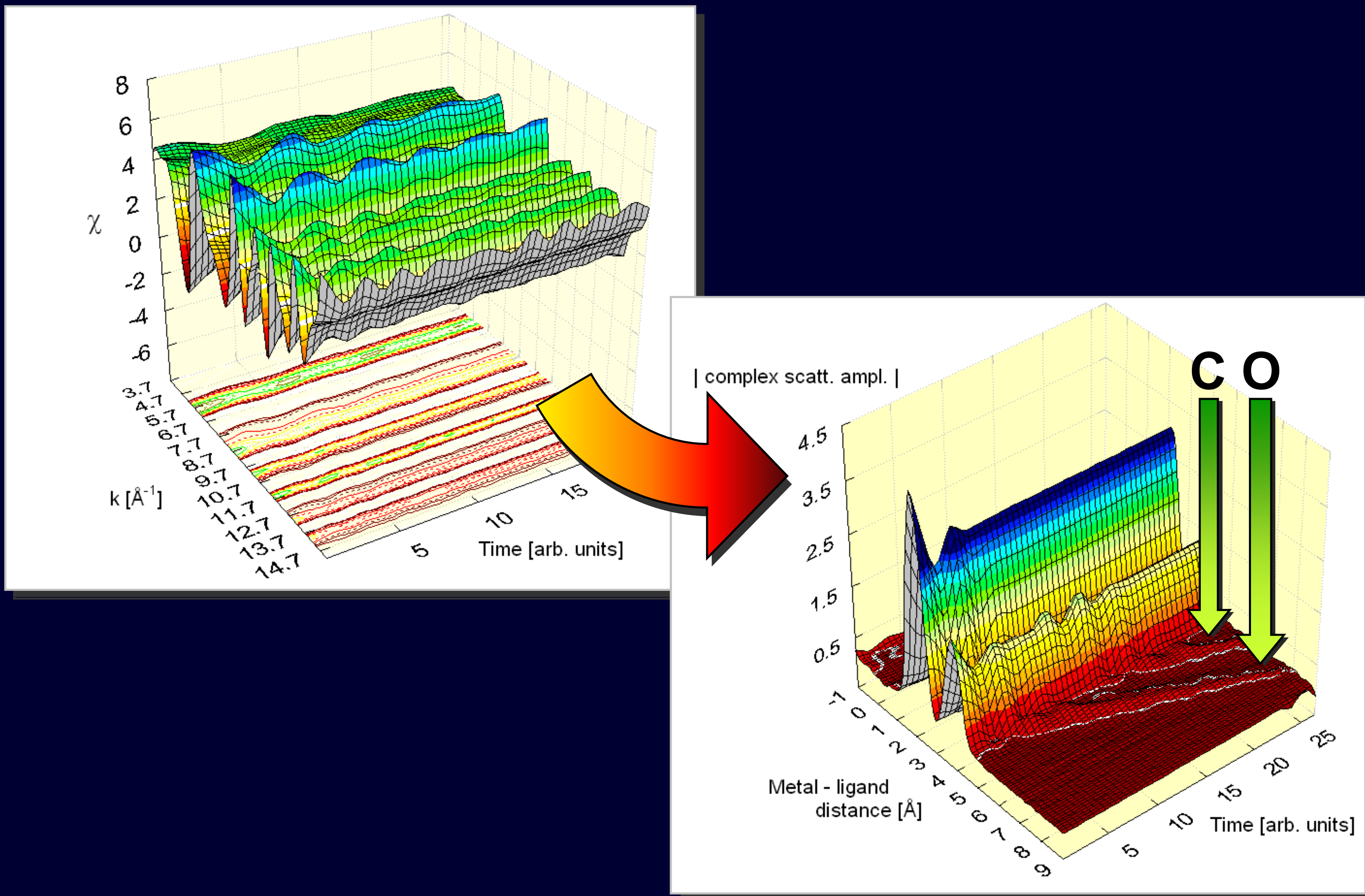
$Fe(CO)_5$



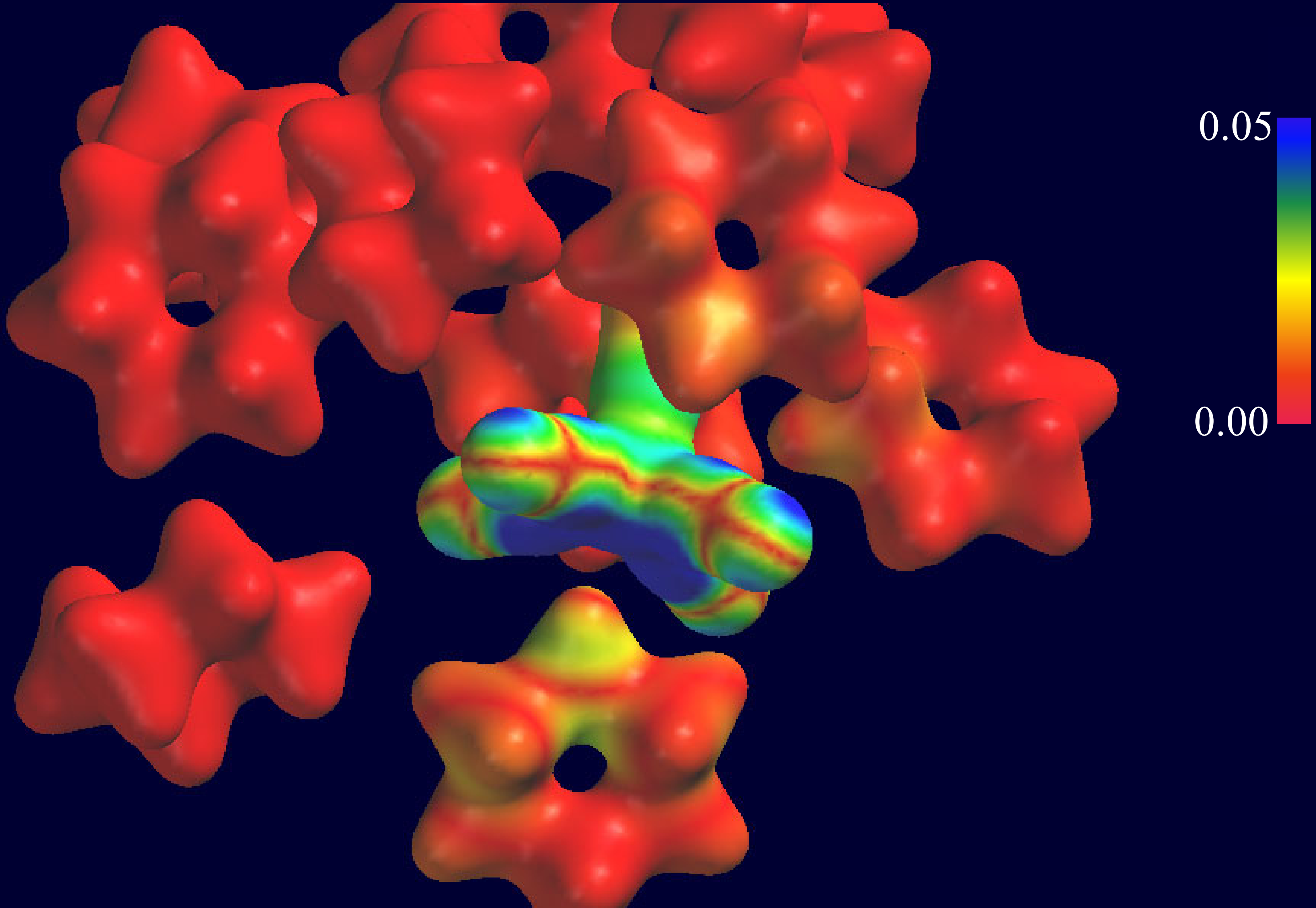
$Fe(CO)_5$ energy levels



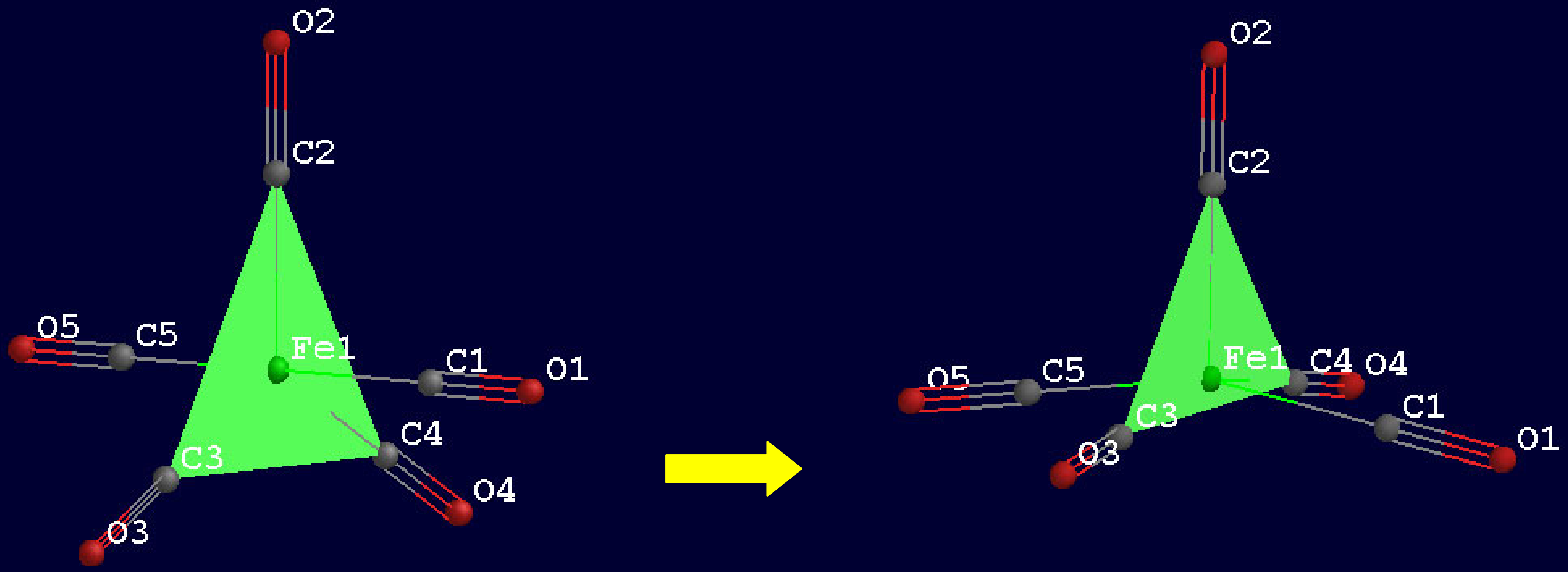
The limited spectral x-ray window should be sufficient to measure photoinduced elimination of a single CO-ligand from $Fe(CO)_5$



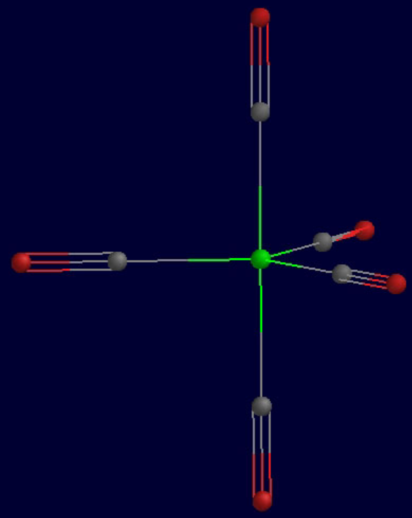
Isovalue electron density surface mapped with HOMO density for $Fe(CO)_5$-10 C_6H_{12}



Structure Change of $Fe(CO)_5$

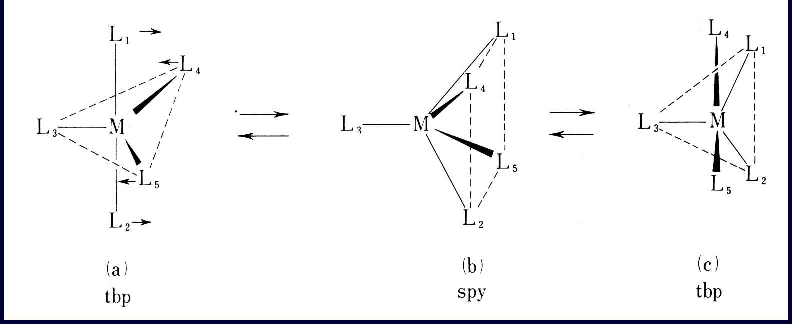


$Fe(CO)_5$



$D_{3h} \rightleftharpoons C_{4v}$

Berry pseudorotation mechanism



D_{3h} C_{4v} D_{3h}

transition state

The energy barrier between C_{4v} and D_{3h} is 1kcal/mol in experimentally (NMR)[1] and 2.3kcal/mol in theoretical calculations[2, 3].

$$\Delta G = - RT \ln K$$

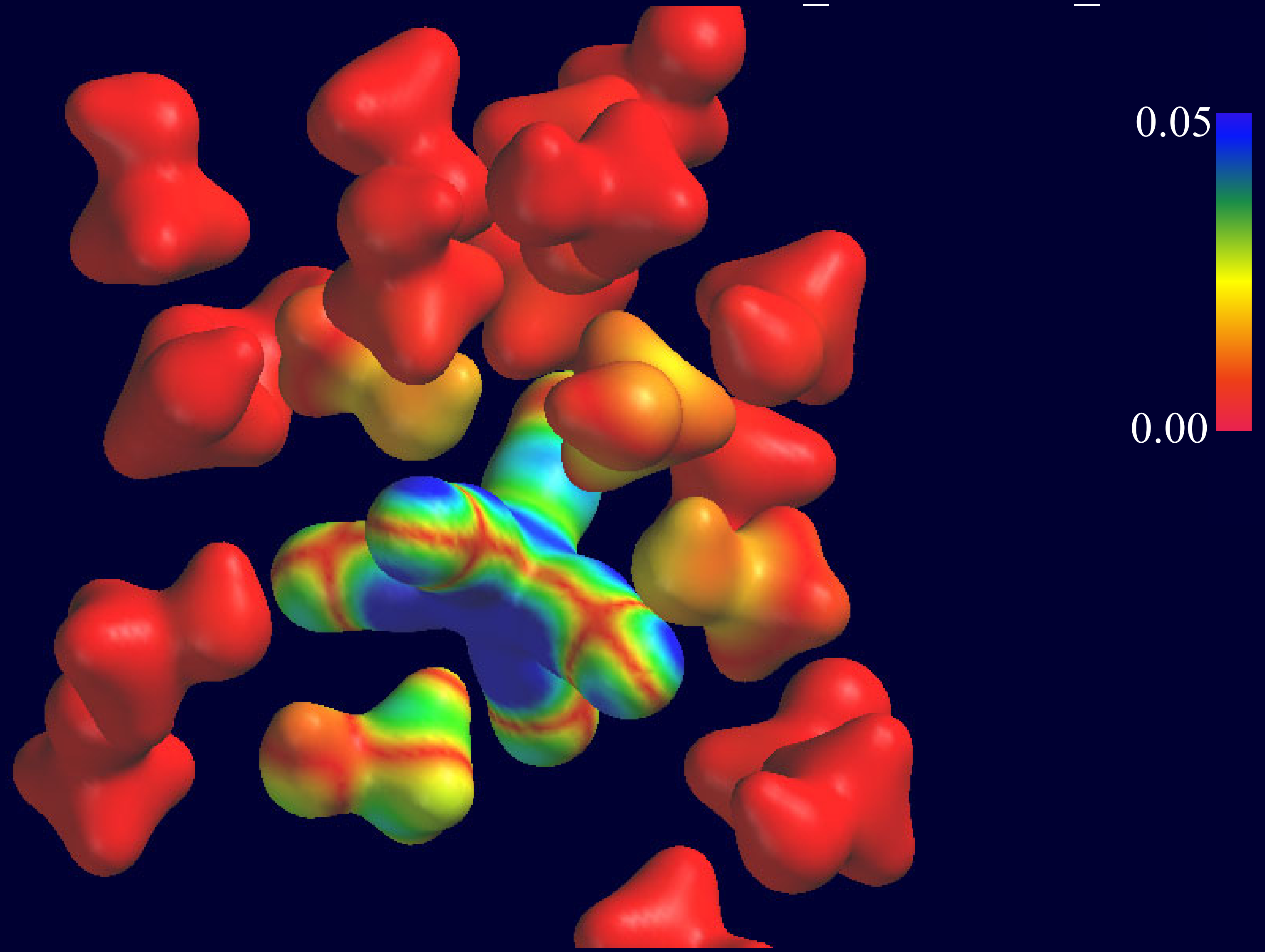
ΔG = 2.3 kcal/mol, then K = 2%,

if ΔG = 1 kcal/mol, then K = 9.8%.

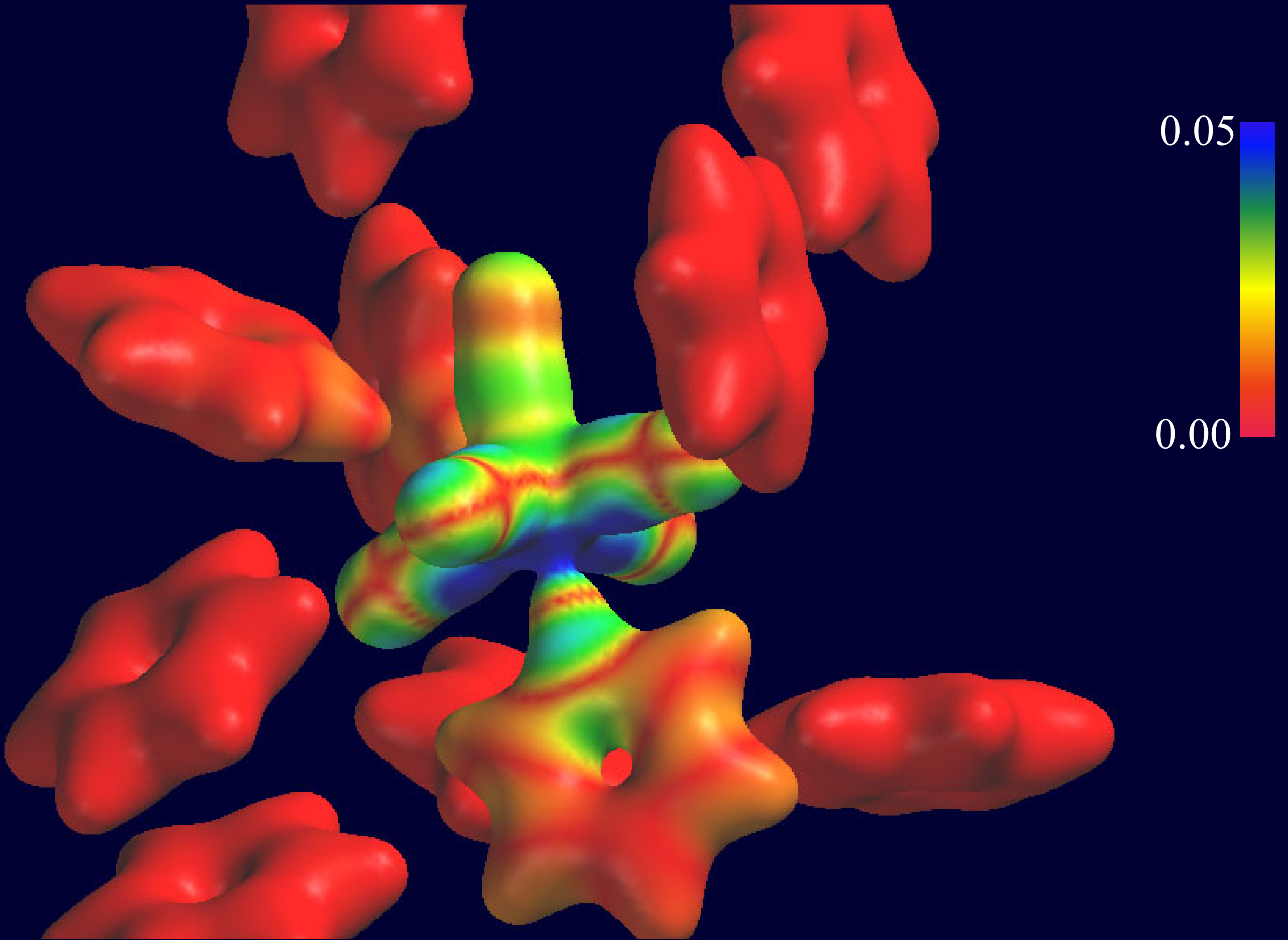
=> At least 2% of $Fe(CO)_5$ in C_{4v}-configuration.

1. Spiess, H. W.; Grosescu, R.; Haeberlen, U. Chem. Phys. 1974, 6, 226
2. J. H. Jang, J. G. Lee, and H. Lee, J. Phys. Chem. A 1998, 102, 5298-5304
3. Demuynck, J.; Strich, A.; and Veillard, A., Nouveau J. de Chim., 1977, 1(3), 217-228

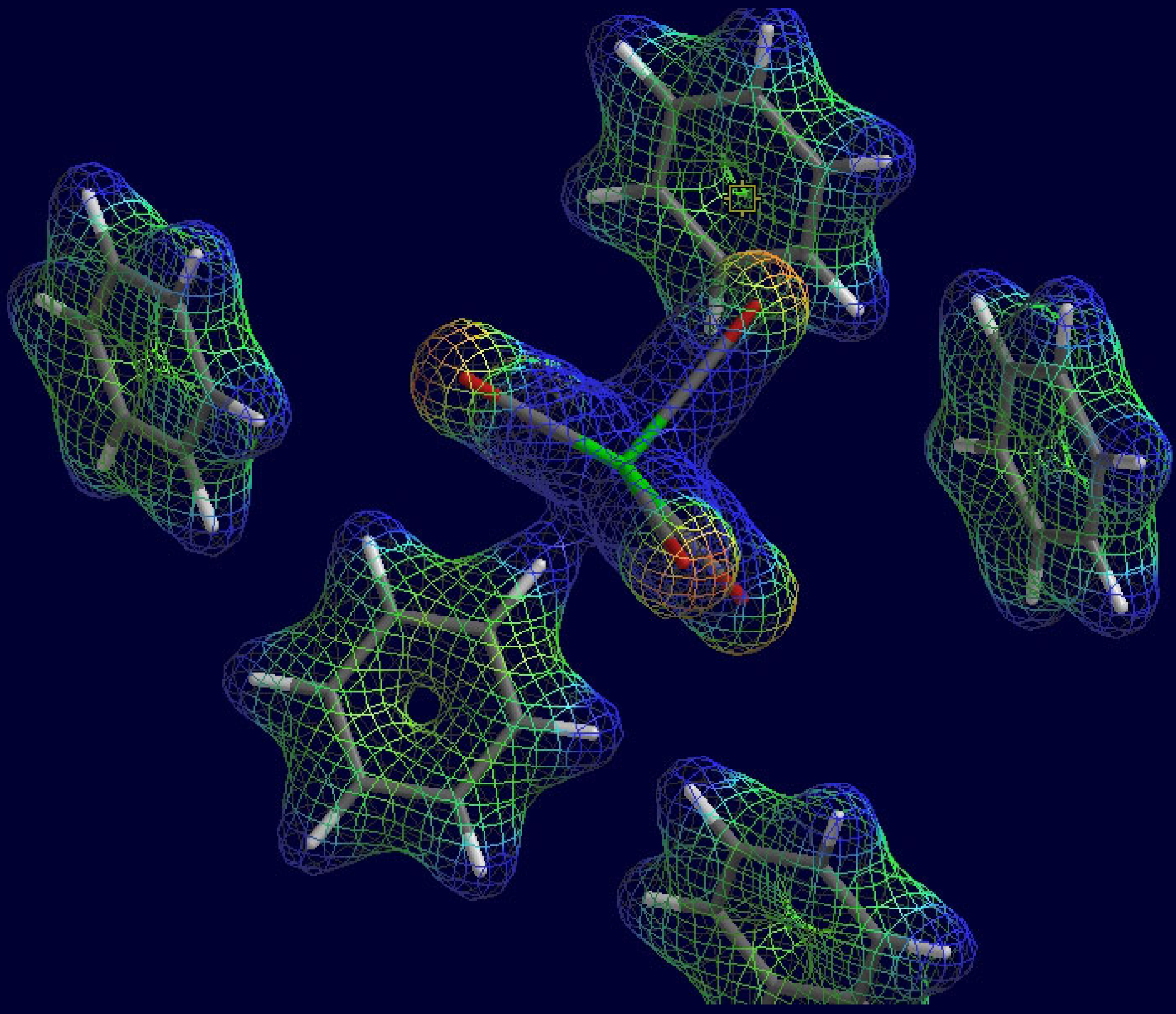
Isovalue electron density surface mapped with HOMO density for $Fe(CO)_5$ -20 CH_3OH



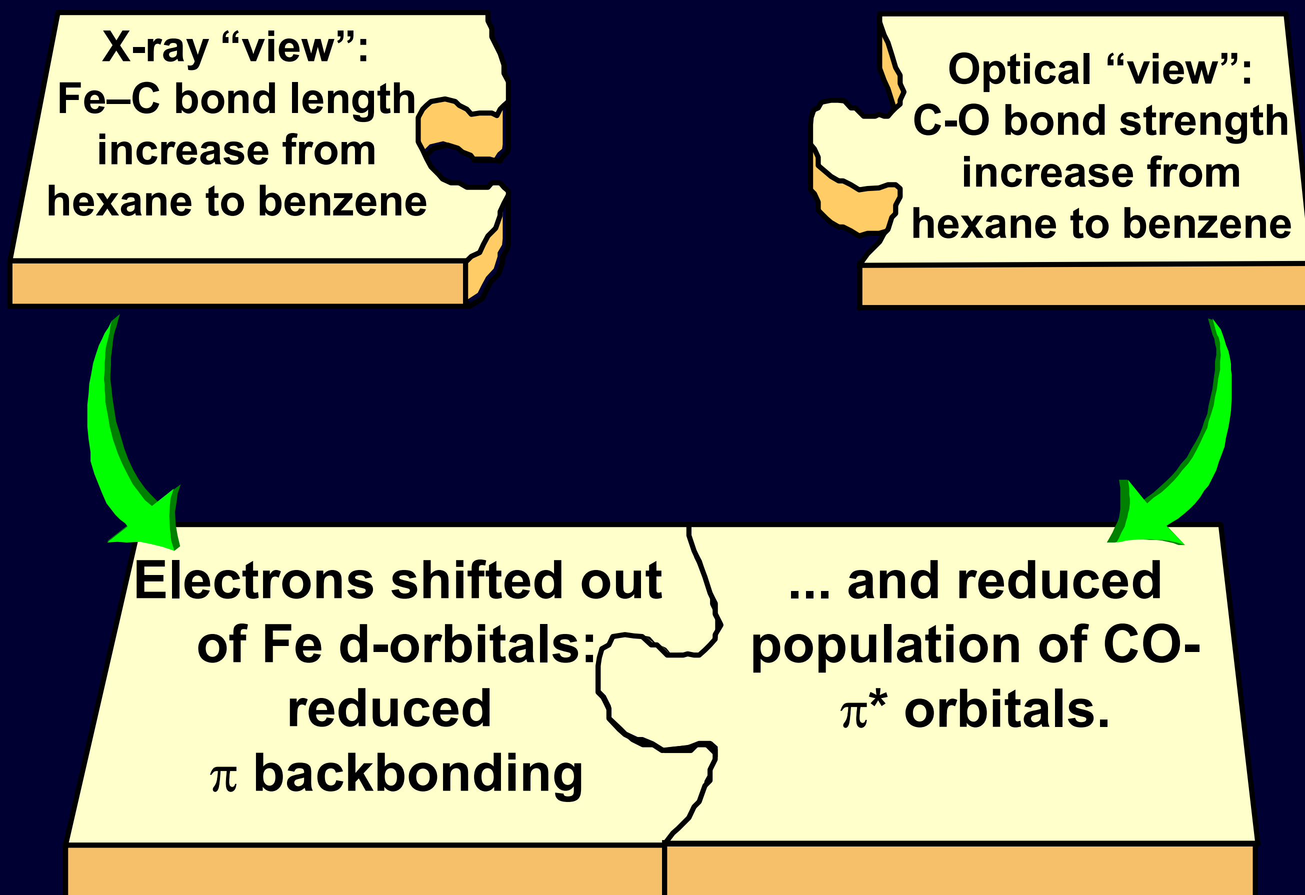
Isovalue electron density surface mapped with HOMO density for $Fe(CO)_5$ -10 C_6H_6



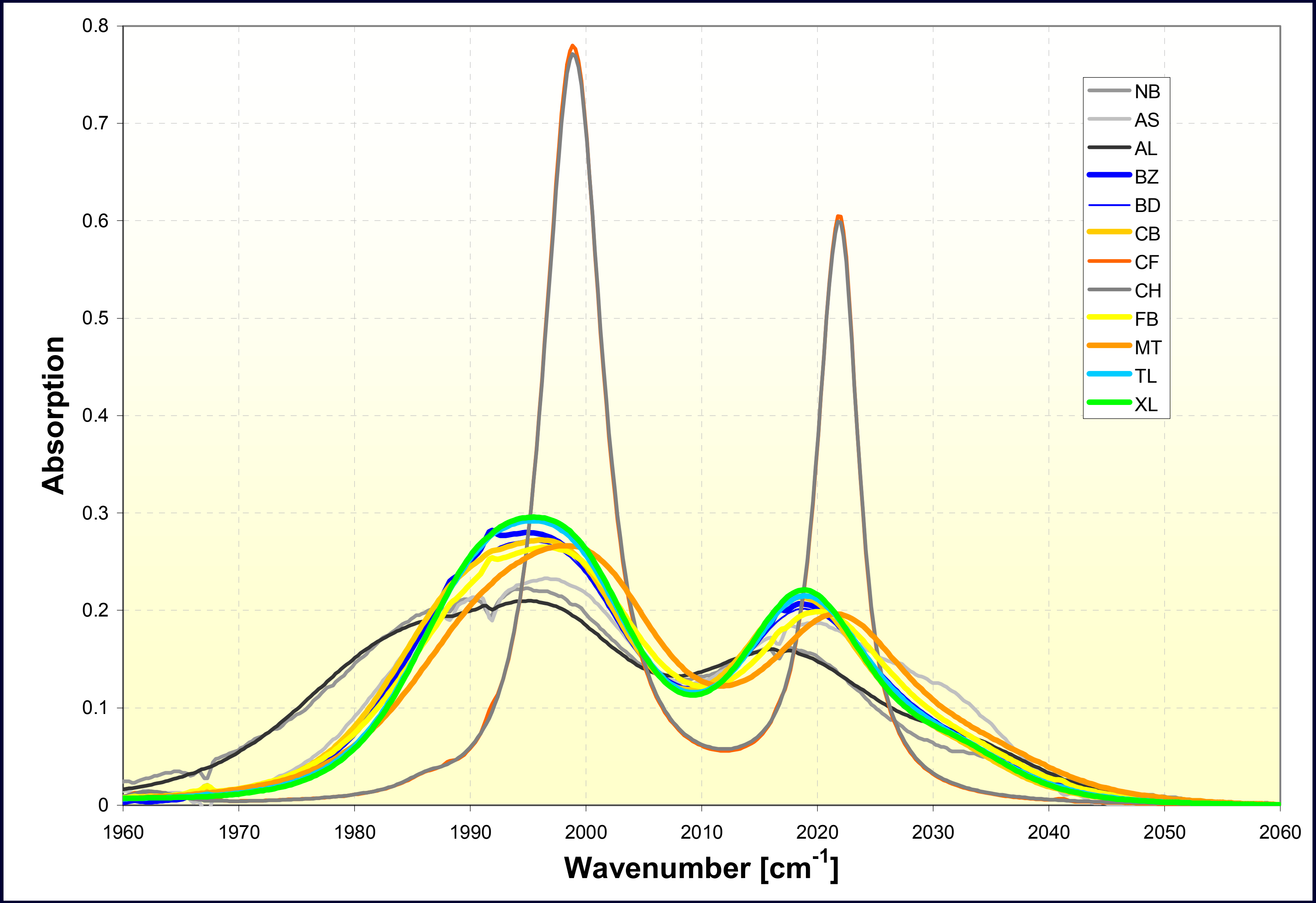
Isovalue electron density surface mapped with HOMO density for $Fe(CO)_5$ - 5 C_6H_6



X-ray and infrared spectroscopy: Complementary methods probing structural dynamics and molecular normal modes



Measured IR absorption spectra of solvated $Fe(CO)_5$



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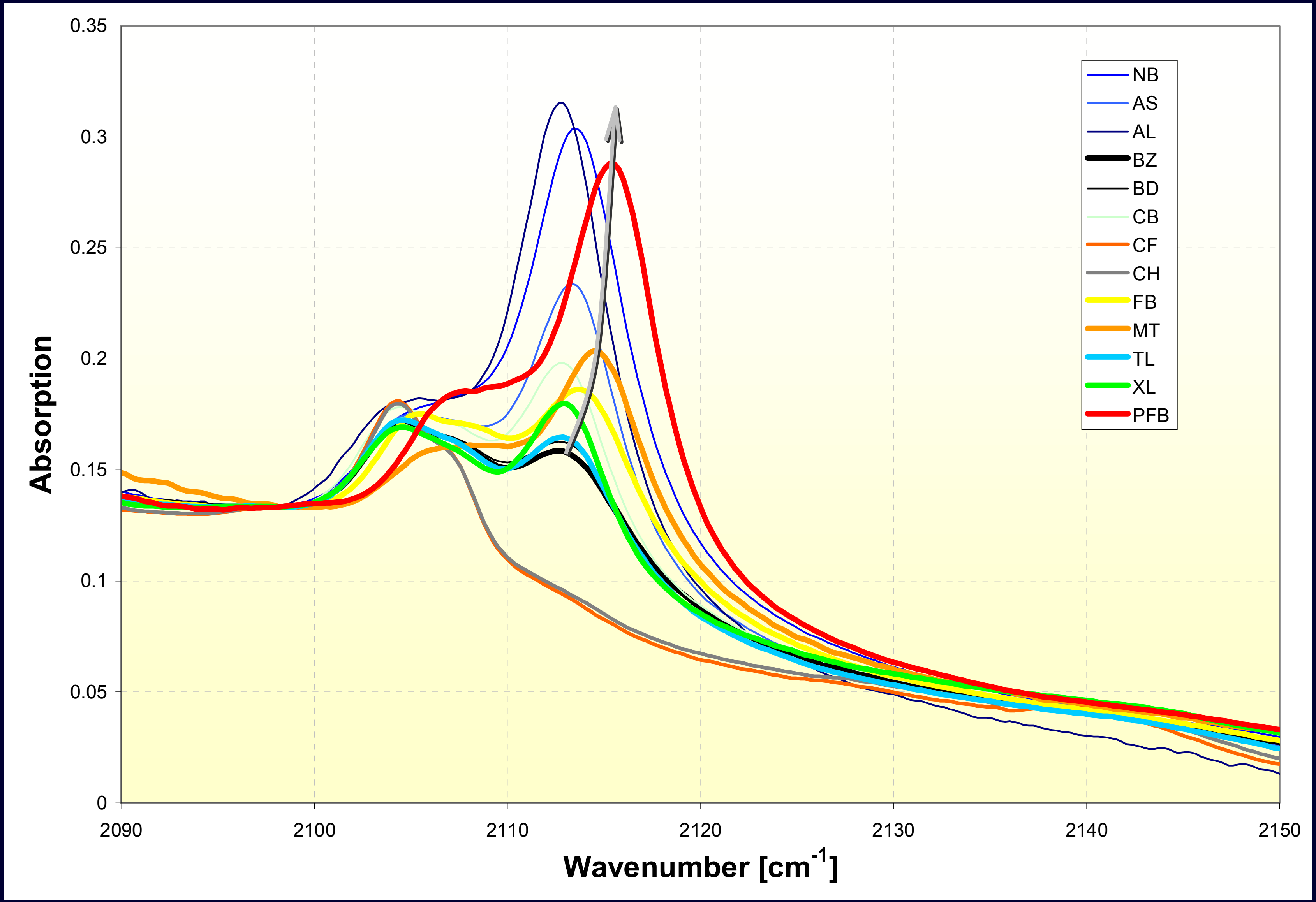
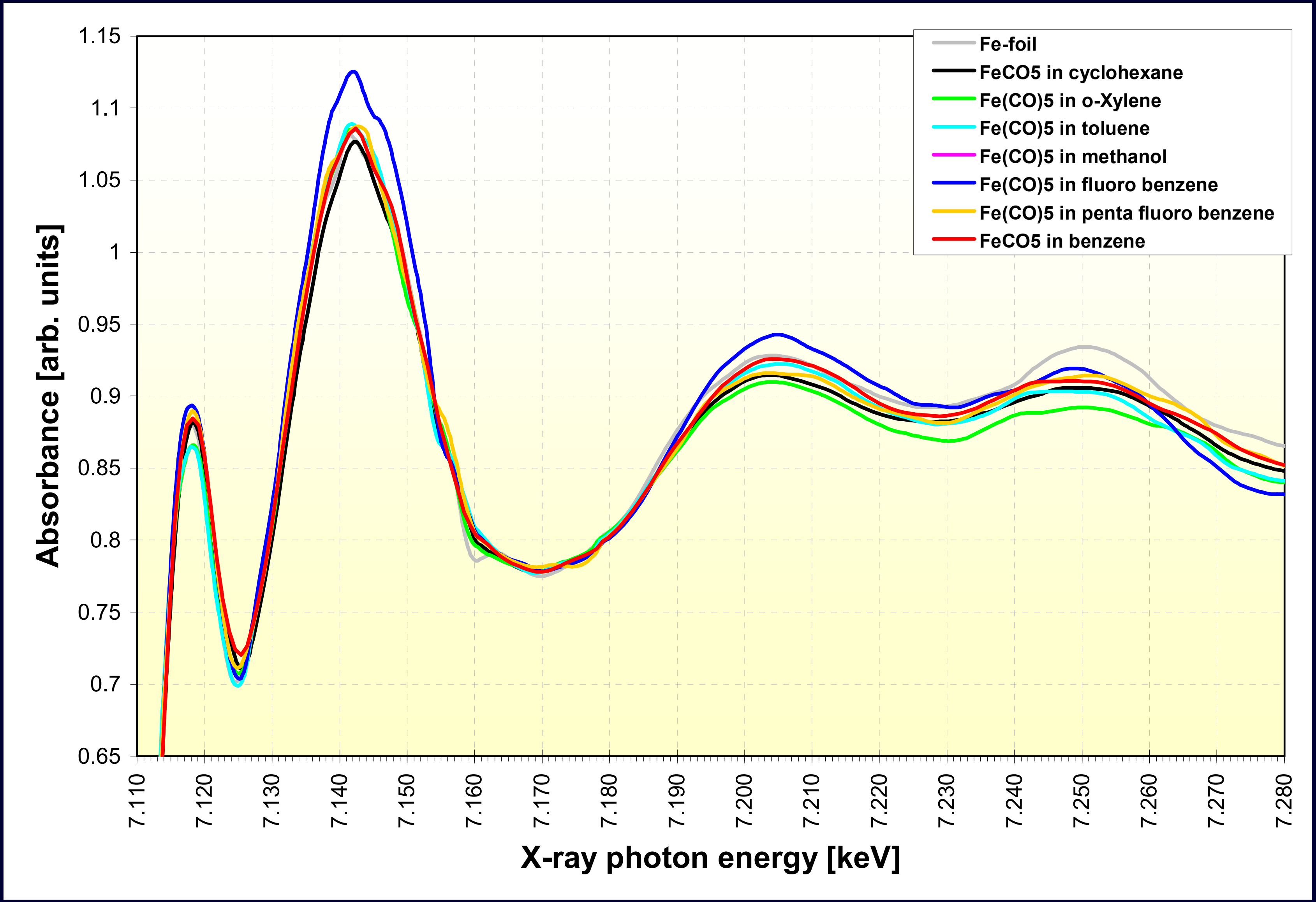


Both Measured and calculated x-ray absorptions of $Fe(CO)_5$ in solvents



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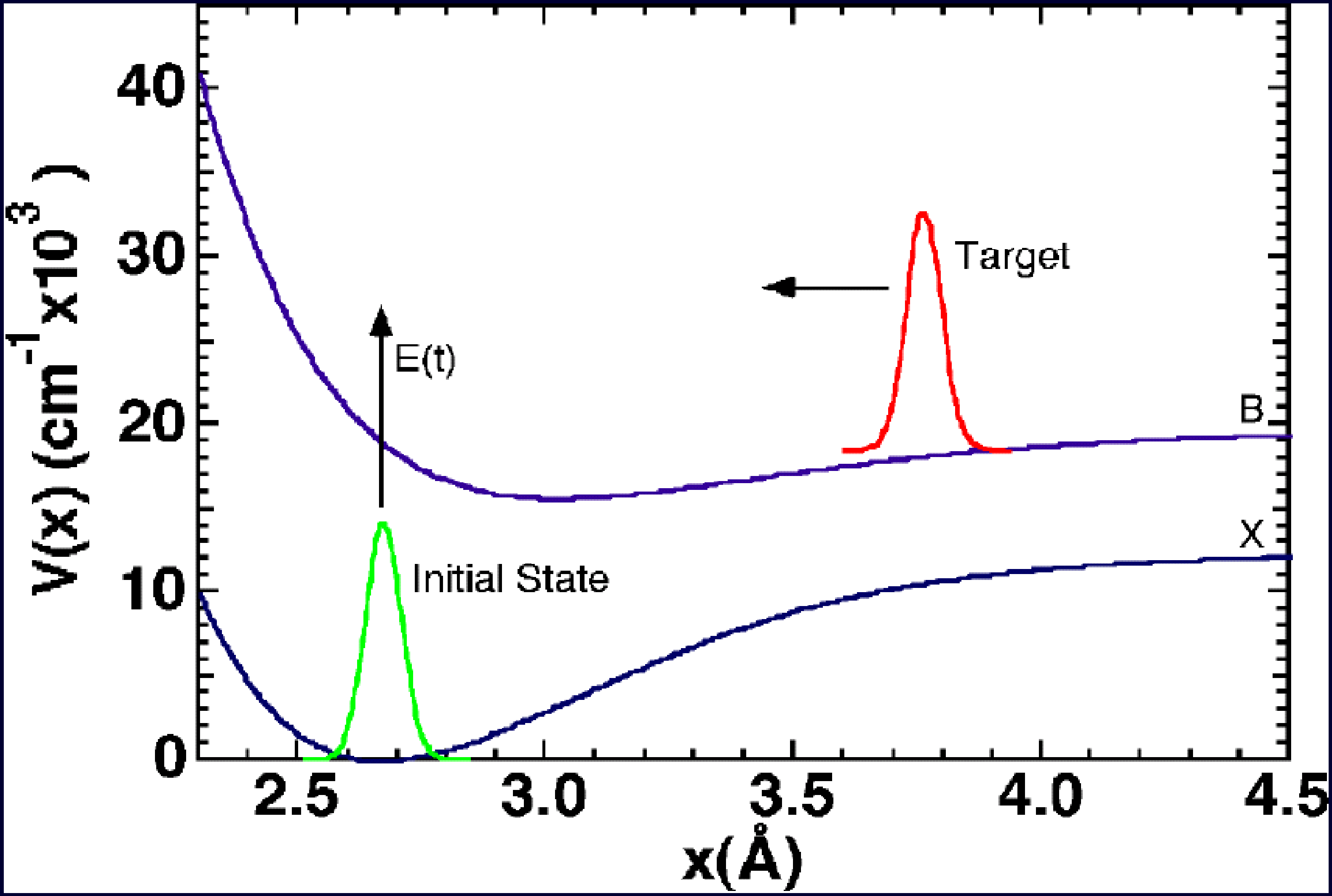
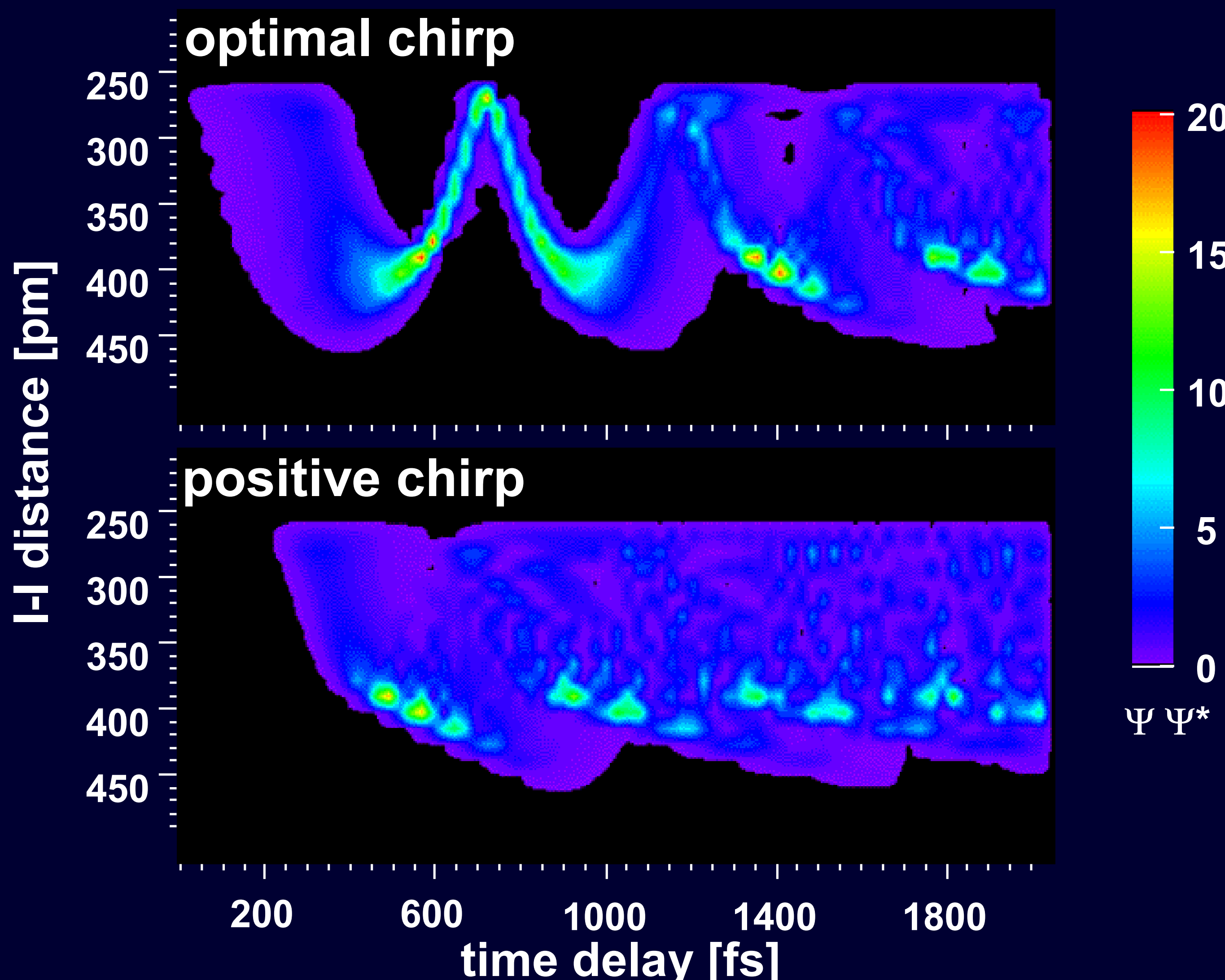
Schematic of I_2 reflectron

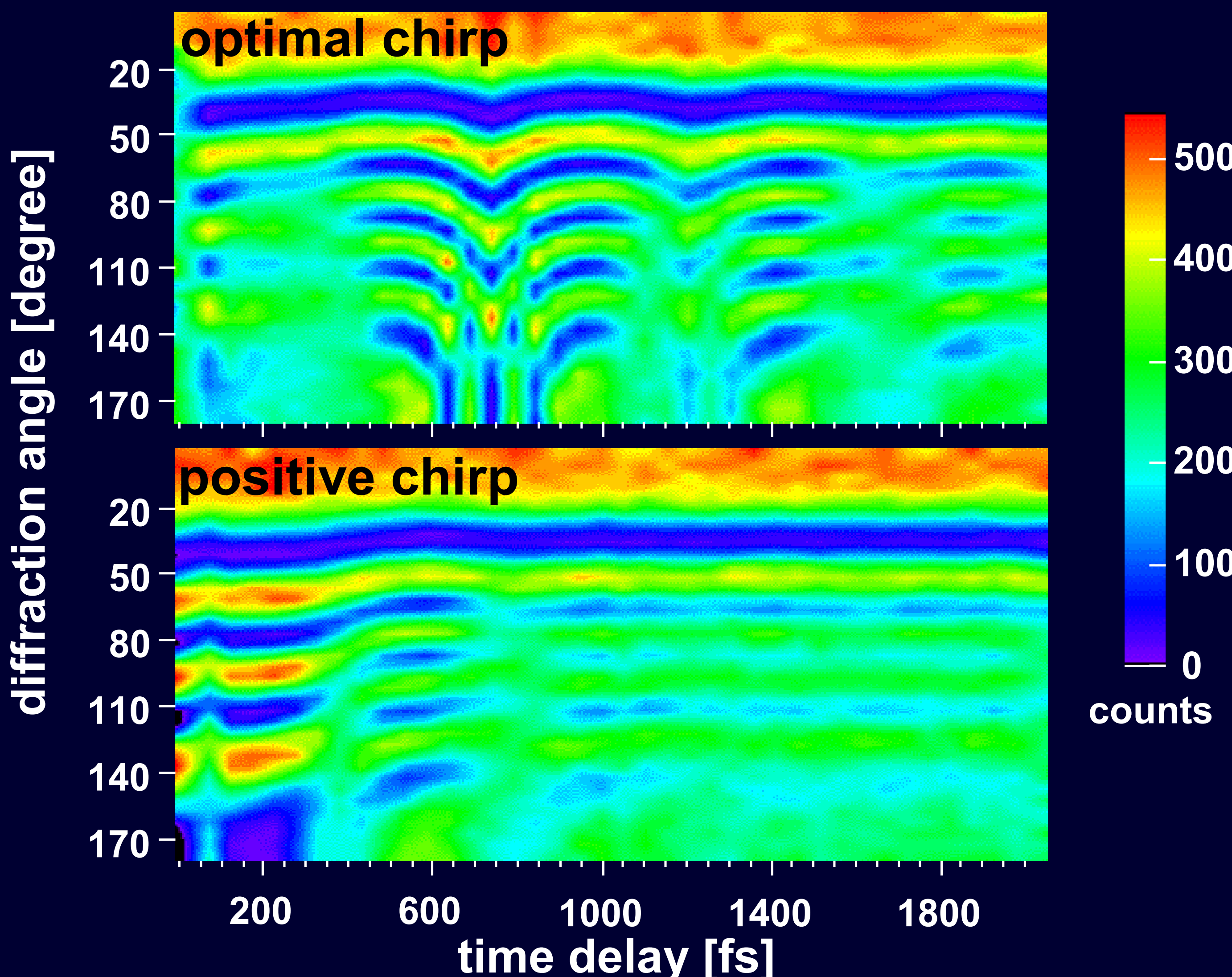


Kohler, B.; Krause, J. L.; Ráksi, F.; Rose-Petruck, C.; Whitnell, R. M.; Wilson, K. R.; Yakovlev, V. V.; Yan, Y.; Mukamel, S. ***J. Phys. Chem.*** **1993**, *97*, 12602.

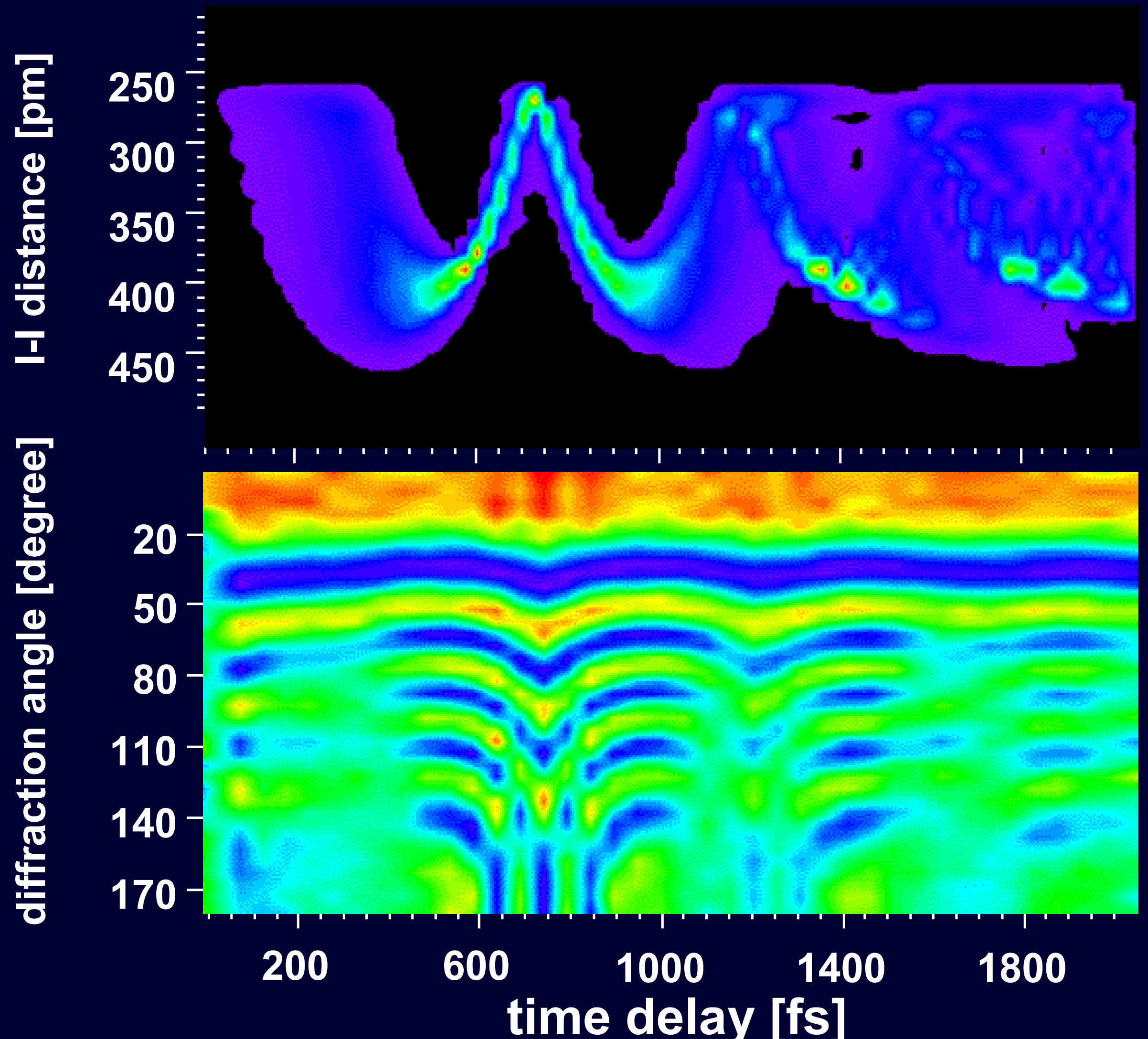
I-I distance probability distributions



Diffraction pattern for Cu-K_α x-ray pulse



Optimal Chirp



Summary

- High-contrast, high average power laser pulses generated through saturable absorber filtering.
- Concept usable for upgrading of regular kHz laser systems.
- Kilohertz x-ray source: X-ray flux at 3keV: 10^{13} ph / s 4π keV
- Ligand motions of $Fe(CO)_5$ are, in principle, observable
- Equilibrium structure of $Fe(CO)_5$ in liquid phase strongly influenced by solvation effects
- The combined applications of IR and x-ray absorption spectroscopy is benefitial

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Klaus Stollberg

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www.rosepetruck.chem.brown.edu

Time Resolved X-ray Diffraction from Coherent Phonons in Semiconductor Heterostructures

P.C. Sondhauss, A. Allen, J.S. Wark

Oxford University, UK

M. Harbst, J. Larsson, O. Synnergren

Lund University, Sweden

G. Naylor, A. Plech, K. Scheidt, M. Wulff

ESRF, Grenoble, France

June 11, 2002

Overview

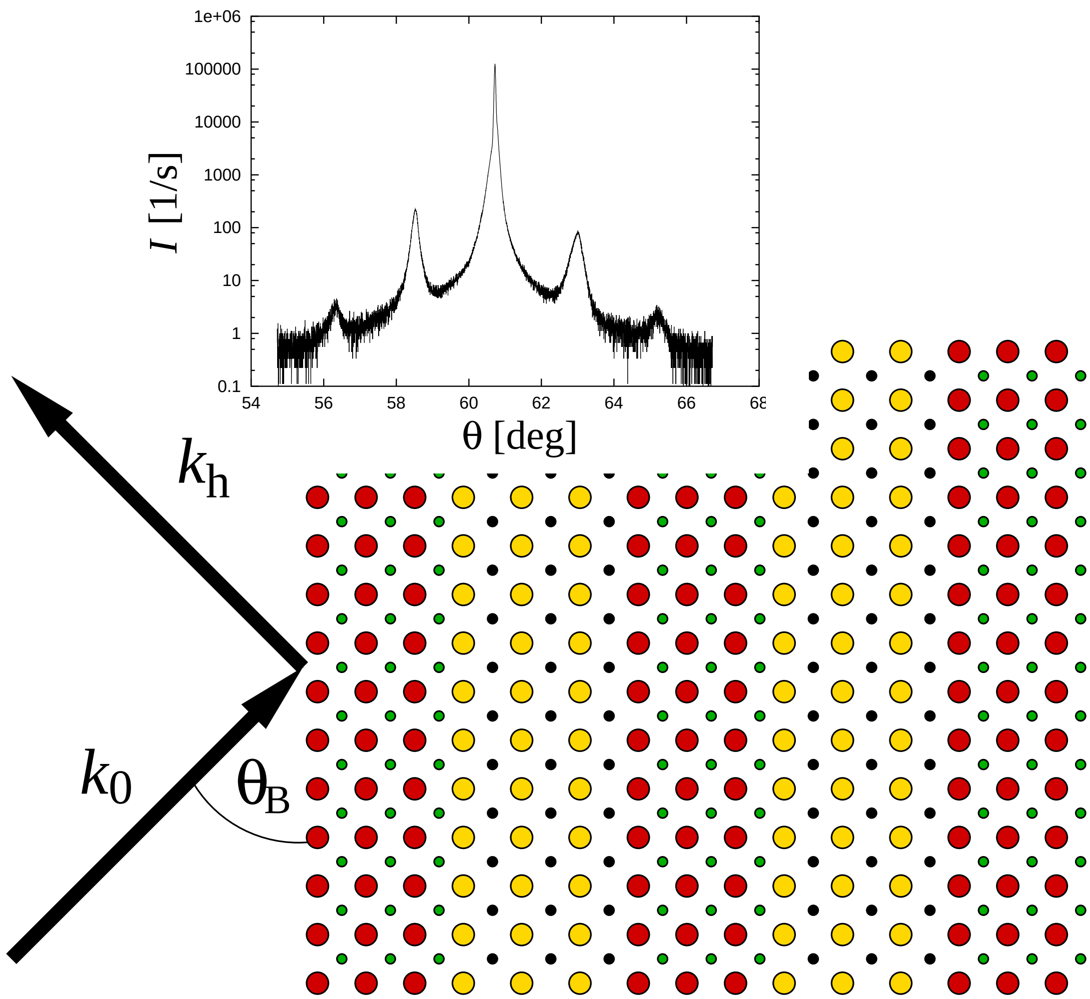
Part 1: Time Resolved X-ray Diffraction from Coherent Phonons in Semiconductor Heterostructures

- Super-lattices
- Strain in a laser heated super-lattice
- Simulated rocking curves
- Synchrotron experiment

Part 2: Picosecond X-ray Diffraction Studies of Shocked Crystals

What are super-lattices?

Epitaxial layers: lattice long-range order preserved over multiple layers $\rightarrow$ **superstructure**
analogous to optical phonons if $a = a'$ and $\chi_h \neq \chi'_h$
analogous to acoustic phonons if $a \neq a'$ and $\chi_h = \chi'_h$



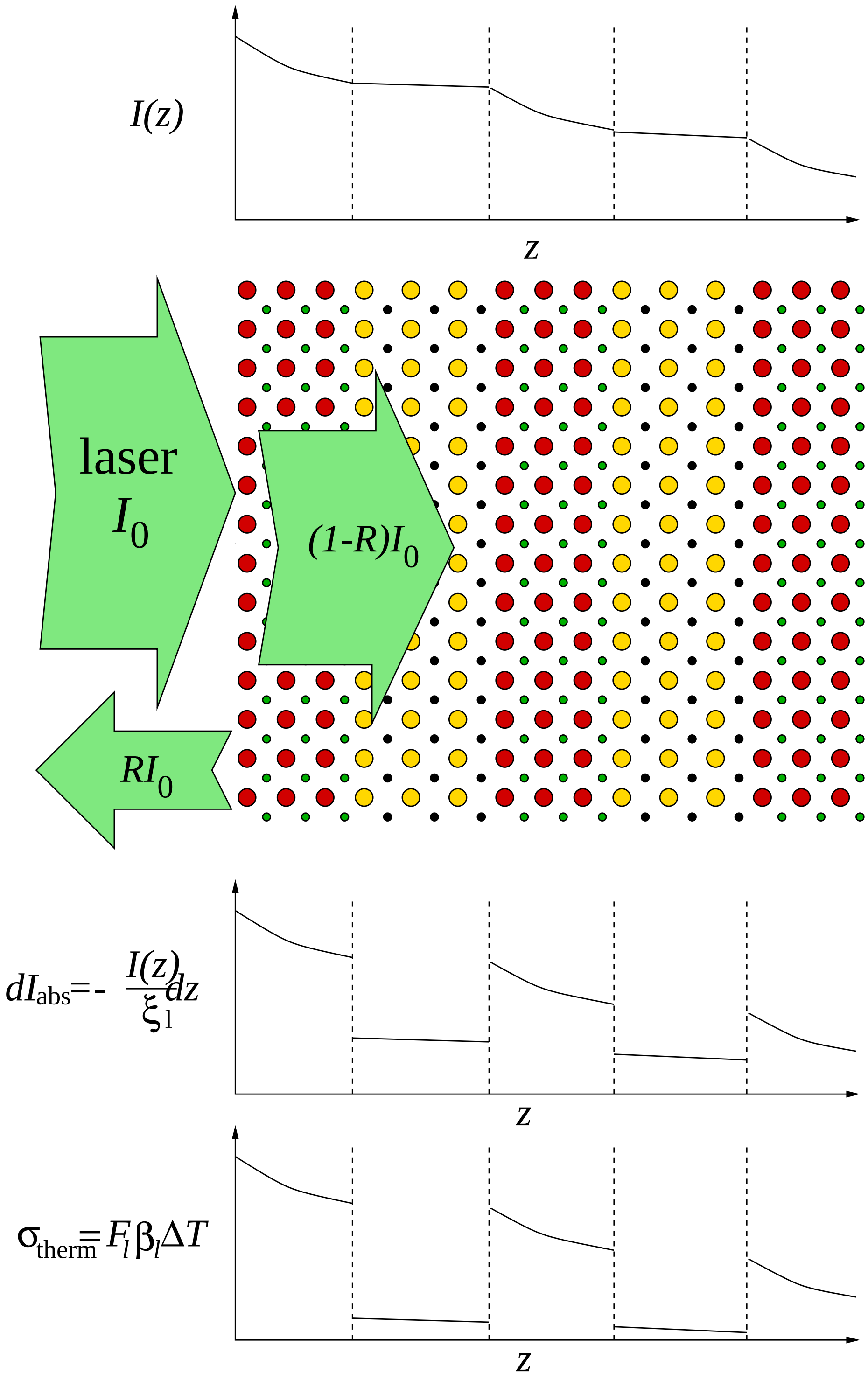
Why super-lattices?

Coherent control: enhancing certain phonon modes

- Resonance effect if $v' \lesseqgtr v$ (reflection @ interfaces)
- "Periodical" thermal stress profile if e.g.
  laser absorption depth $\xi' \lesseqgtr \xi$
  or
  thermal expansion coeff. $\beta'_{\mathrm{therm}} \lesseqgtr \beta_{\mathrm{therm}}$

More information about propagating strain profile if diffraction features from different materials can be distinguished.

laser generated strain in a super-lattice @ $t = 0$



A model for the strain

Displacement in layer l described by **inhomogeneous wave-equation**

$$\frac{\partial^2 u_l}{\partial z^2} - \frac{1}{v_l^2}\frac{\partial^2 u_l}{\partial t^2} = g_l(z)$$

Inhomogeinity $g_l(z) \propto \partial_z \sigma_{\mathrm{therm}}(z)$ (initial thermal stress).

Initial condition: no initial displacement, nor movement

$$u_l(z,0) = 0 \quad , \quad \partial_t u_l(z,0) = 0$$

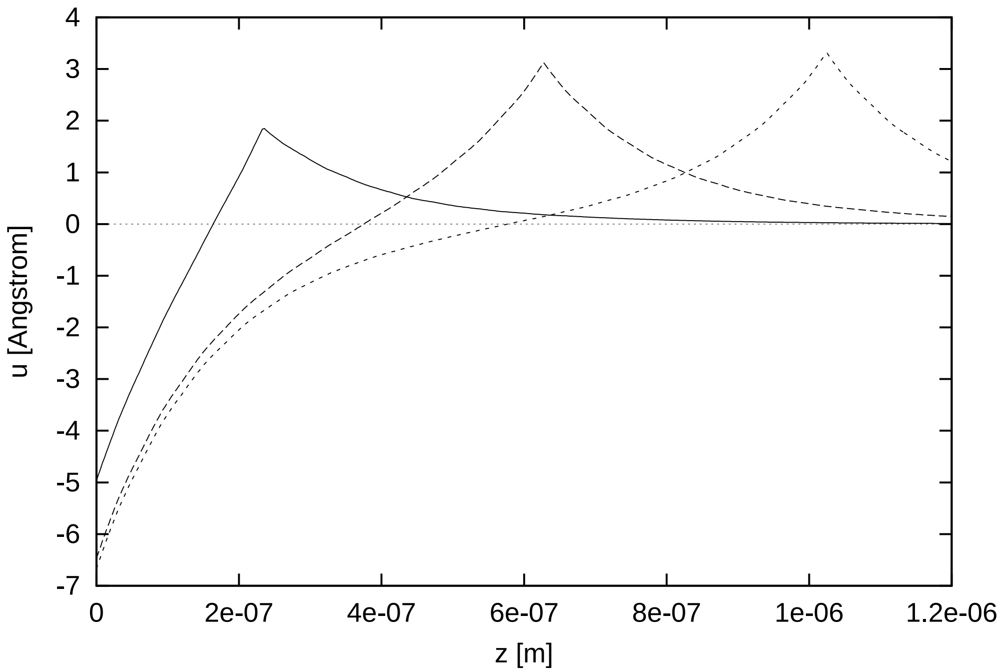
Boundary conditions: free surface @ $z = 0$ and continuous displacement @ interfaces $z = z_l$

$$\sigma_0(0,t) = 0 \quad , \quad u_l(z_l,0) = u_{l-1}(z_l,0)$$

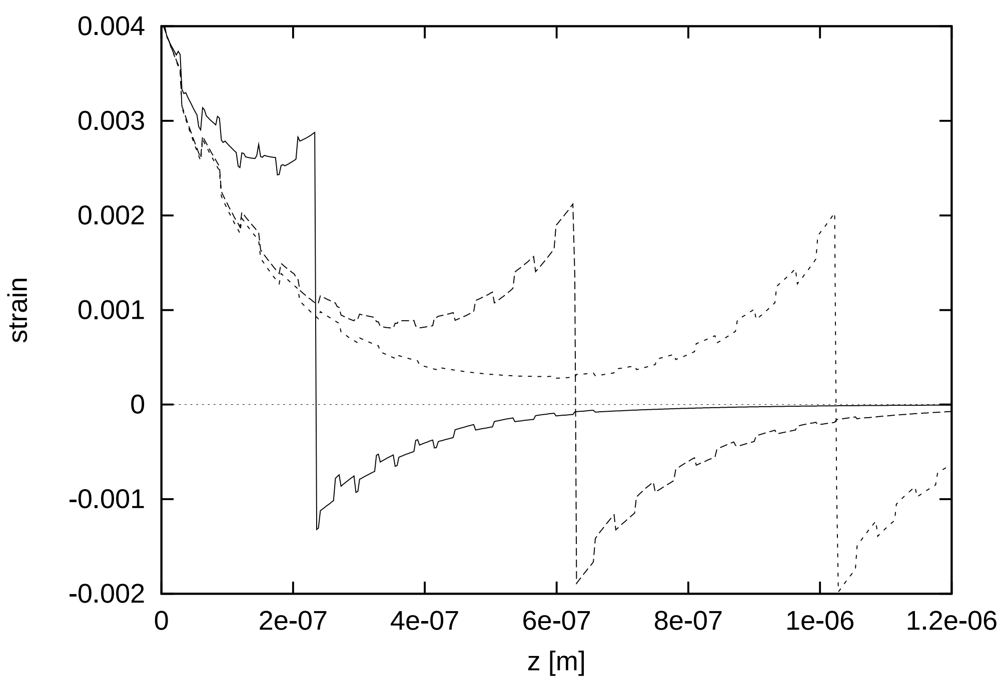
Analytic solution possible.

Displacement and strain

displacement $u(z, t)$:

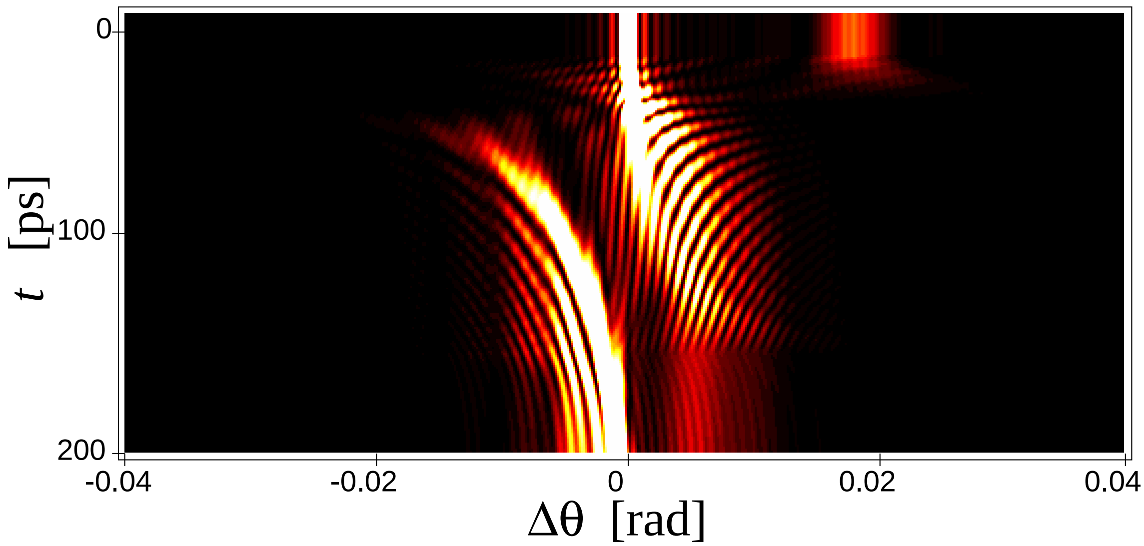


strain $\eta(z, t) = \partial_z u(z, t)$:



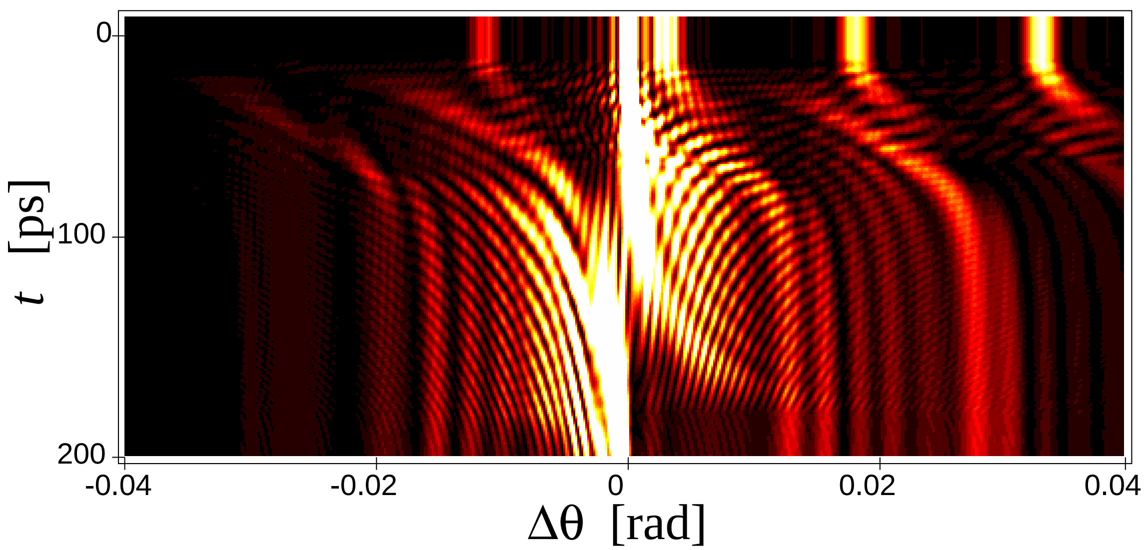
Rocking curves

- 50×4 nm GaSb/InAs layers on GaAs substrate
  laser flux: 8 mJ/cm^2



layer-peak disappears about 20 ps after laser shot

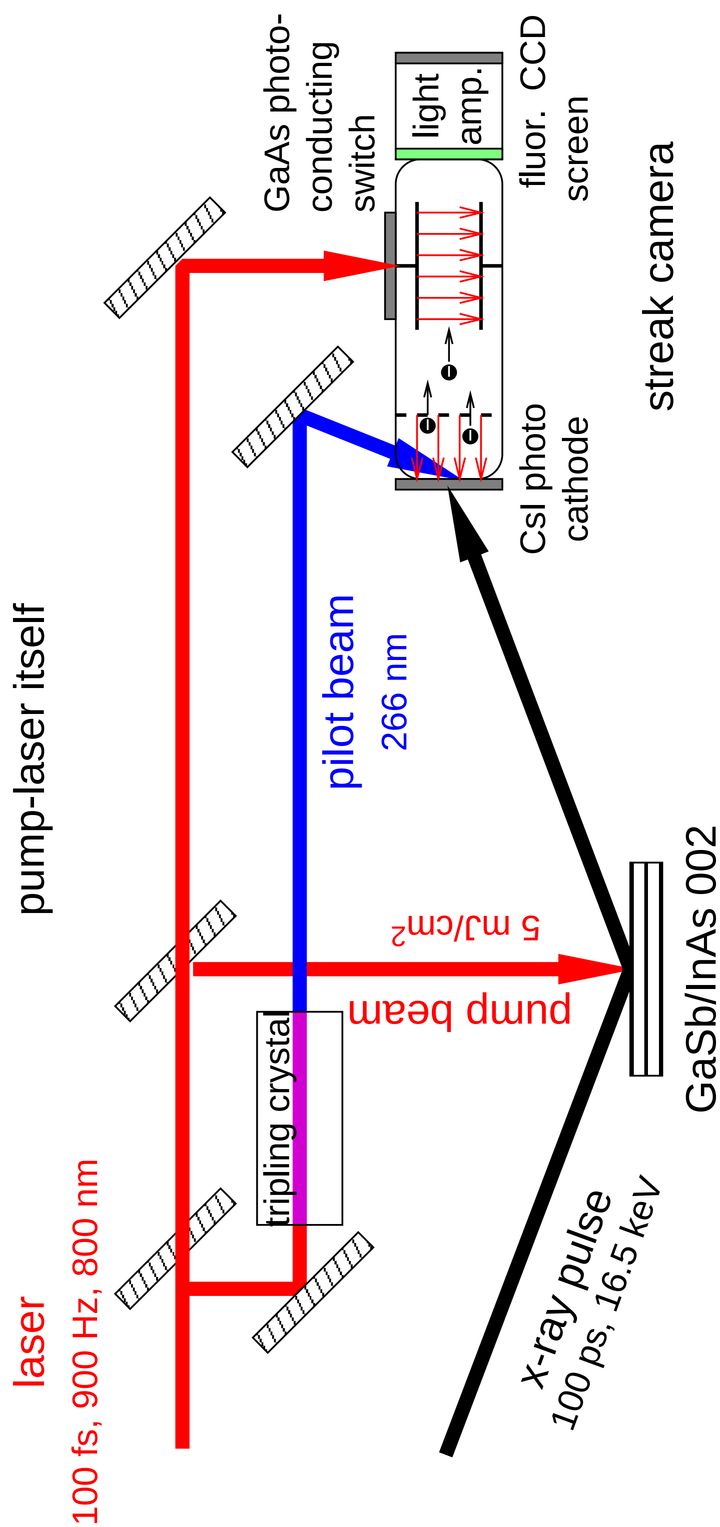
- 7×60 nm GaSb/InAs layers on GaAs substrate
  laser flux: 8 mJ/cm^2



features from close sidebands overlap → complicated time dependence.

Experimental setup

synchronisation of laser-pump and x-ray probe by optically triggering the streak camera with the pump-laser itself



Picosecond X-ray Diffraction Studies of Shocked Crystals

J.S. Wark, A. Allen, P. Sondhauss, J. Hawreliak,

K. Rosolankova, B. Lings

Oxford University, UK

T.R. Boehly

Rochester University, U.S.A.

P.S Lomdahl, T.C. Germann, B.L. Holian,

K. Kadau, D. Paisley, D. Swift, G. Kyrala

Los Alamos National Laboratory, U.S.A.

D.H. Kalantar, B.A. Remington, J. Belak,

E. Bringa, K. Budil, M. Caturla, J. D. Colvin,

M. Kumar, K. T. Lorenz

Lawrence Livermore National Laboratory, U.S.A.

M.A. Meyers, M. Schneider, B.K. Kad

University of California at San Diego, U.S.A.

June 11, 2002

Overview

Part 2: Picosecond X-ray Diffraction Studies of Shocked Crystals

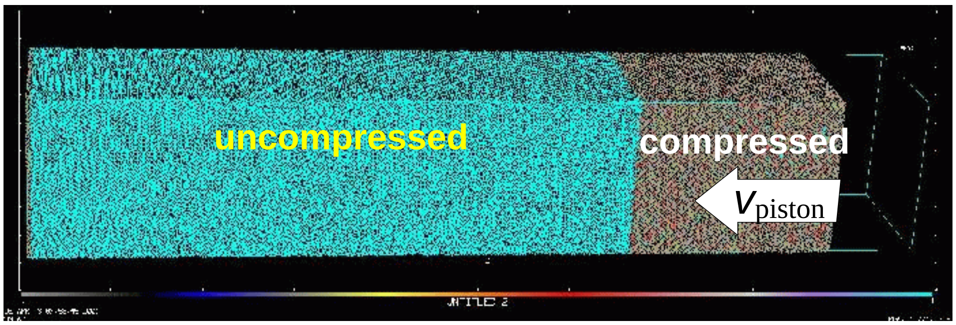
- What happens in a shocked crystal?
- Why is it relevant?
- Ideas for synchrotron experiments

Introduction

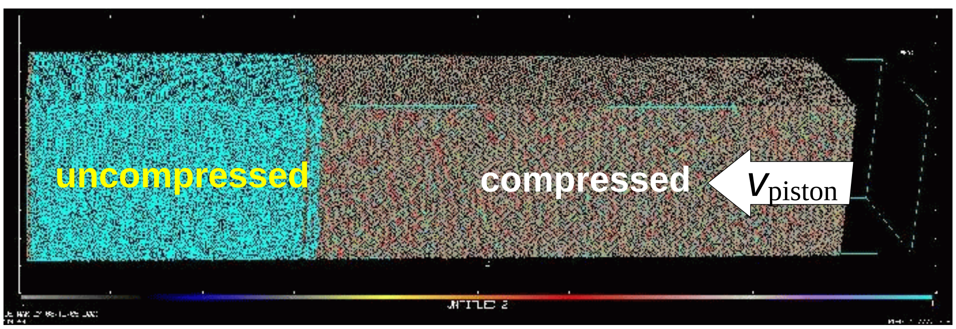
Laser shock experiments provide unique insight on basic deformation mechanisms at high strain rate.

Non-equilibrium molecular-dynamics simulations:

weak shock: $v_{\text{piston}} = 1.09$ km/s $\rightarrow$ 13% compression
compression still elastic



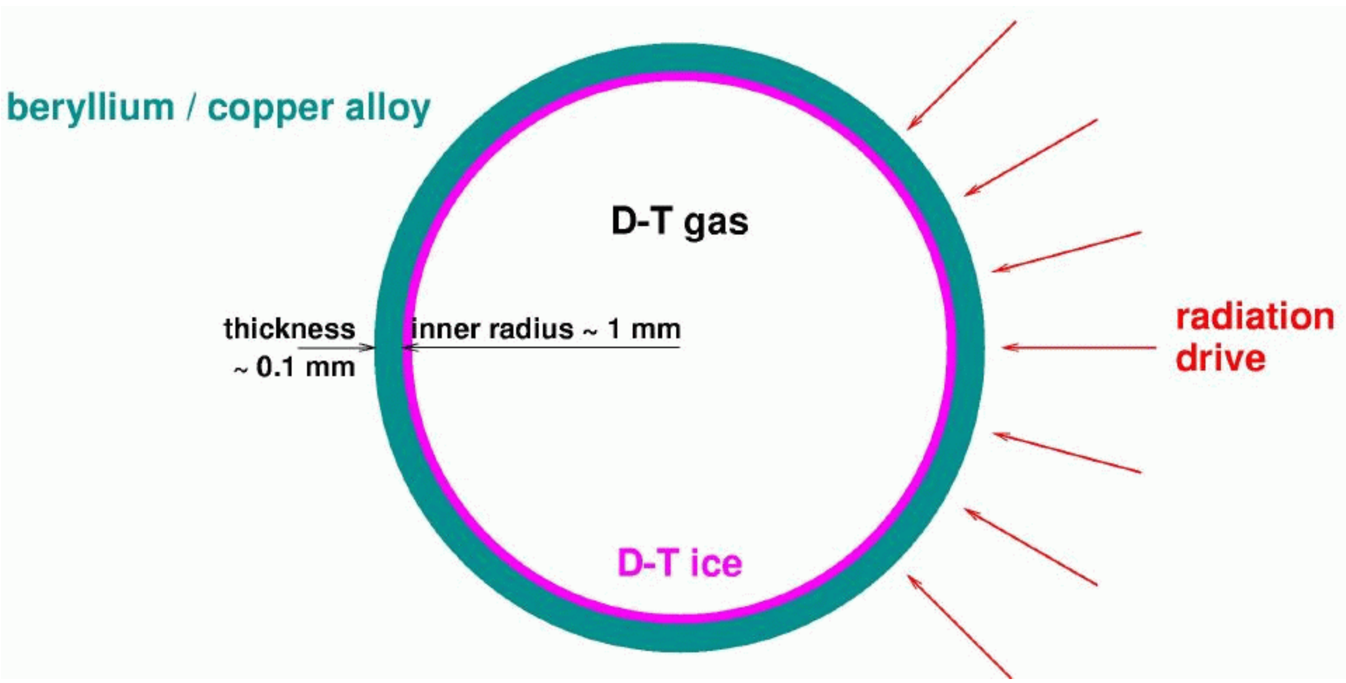
stronger shock: $v_{\text{piston}} = 1.91$ km/s $\rightarrow$ 22% compression
starts out elastic



just after plastic deformation begins, rarefaction wave propagates backwards from the shock front.

Motivation

Experiments are relevant to ICF designs relying on Be capsules.

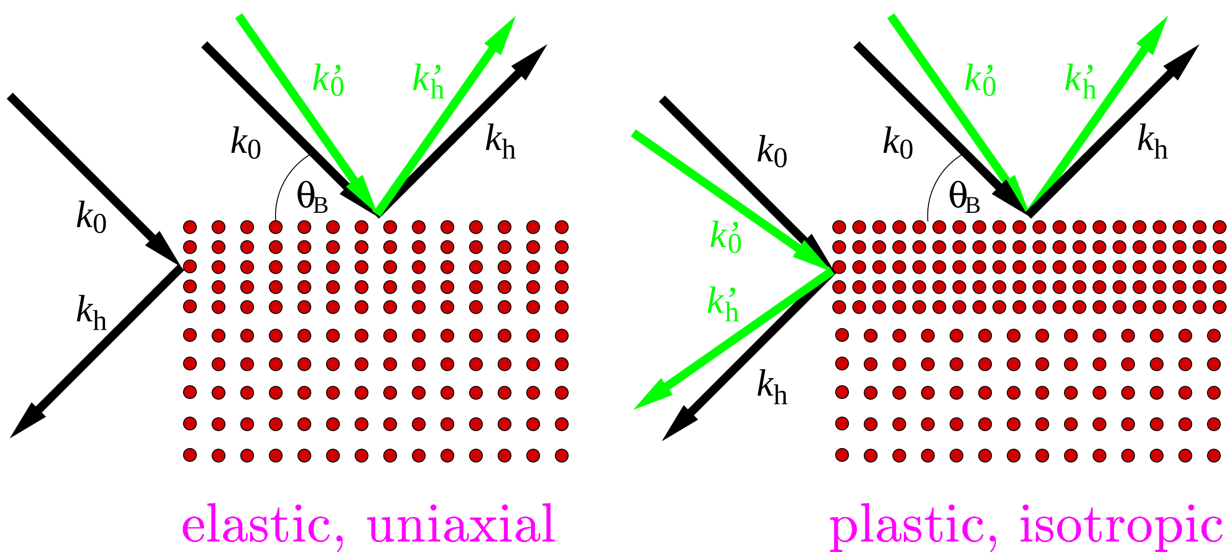


Shock wave inhomogeinities in Be
→ inhomogeinity in fuel compression
→ faster growth of instabilities
→ less compression → reduced yield

Uniaxial versus isotropic compression

- Elastic compression is largely 1-D
- Plastic compression is expected to become isotropic, due to lattice rearrangements (stacking faults)

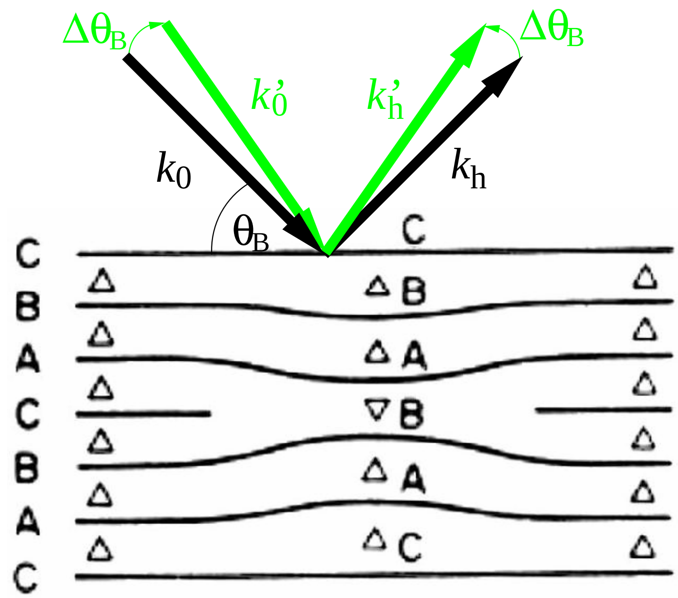
Time resolved diffraction may allow to observe the transition from uniaxial elastic to isotropic plastic compression.



elastic, uniaxial   plastic, isotropic

Stacking faults

Sign and modulus of $\Delta\theta_B$ due to stacking faults depends on diffraction order



→ comparing the shifts of two different orders (e.g. 111 a. 222) allows to determine simultaneously **compression** and **stacking fault density**.

Conclusions

- A model for strain in a laser-heated super-lattice has been developed
- Semiconductor heterostructures give the freedom to modify the temporal response of the rocking curve
- Laser shock experiments provide unique insight on basic deformation mechanisms at high strain rate.
- Experiments may be relevant to ICF designs that rely on Be capsules.
- Laser shock experiments can be performed at a synchrotron, provided a laser is available (≈ 1 ns, few Joule). Nobody has done this so far.