Detectors For Ultrafast X-ray Spectroscopy

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Femtosecond X-ray Science at the ALS

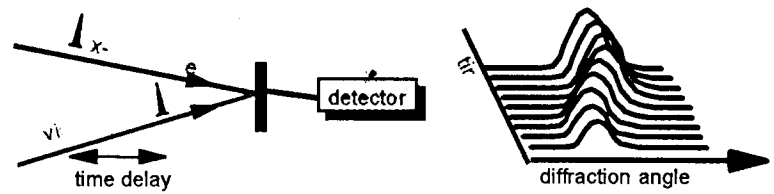
Structural Dynamics in Condensed Matter

fundamental time scale for atomic motion
vibrational period: h/kT ~ 100 fs

- ultrafast chemical reactions
- ultrafast phase transitions
- surface dynamics
- ultrafast biological processes

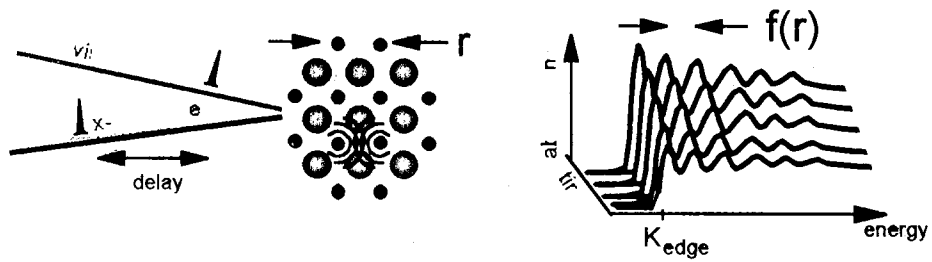
Rapidly emerging filed of research
Physics, Chemistry and Biology

time-resolved x-ray diffraction



ordered crystals - phase transitions, coherent phonons

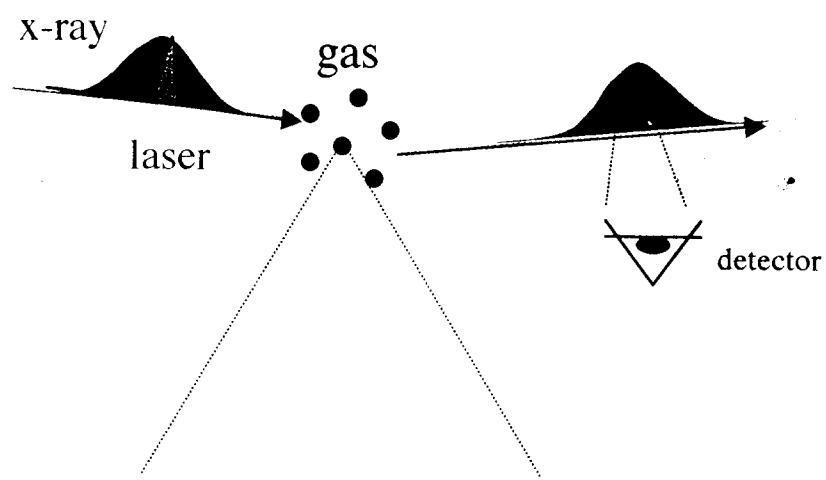
time-resolved EXAFS, NEXAFS, surface EXAFS



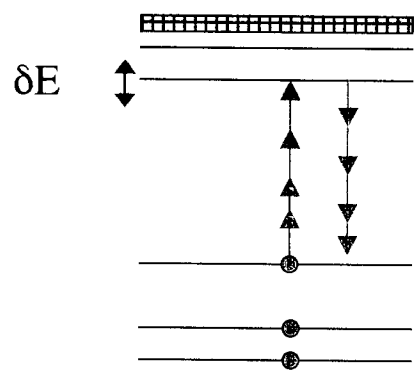
complex/disordered materials - chemical reactions
surface dynamics
bonding geometry

High Field Atomic Structure

fast detectors to study novel structure of atoms in intense fields



stark shifts

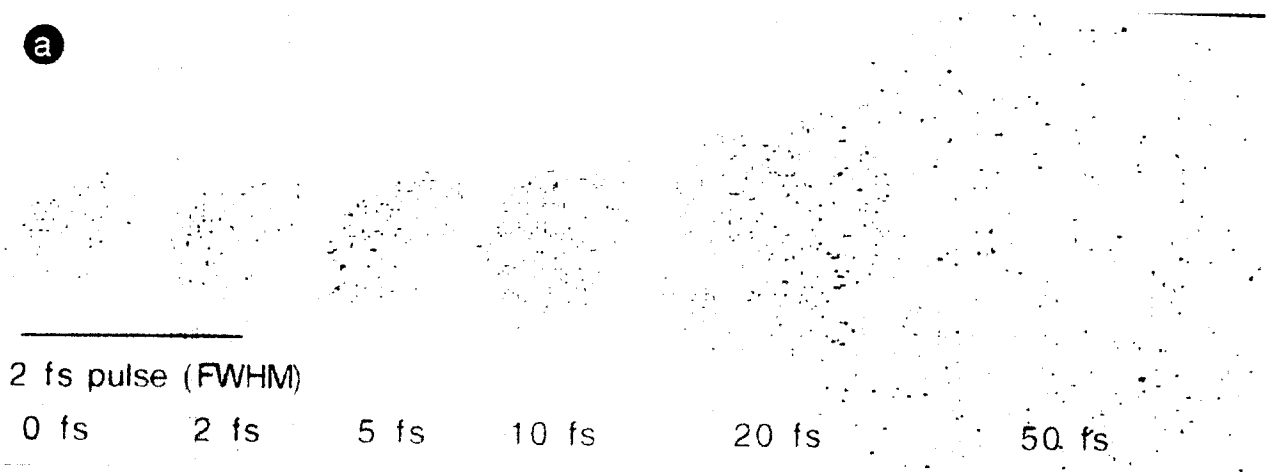


multiphoton mixing of states

A Need for < 100 fs time-resolution

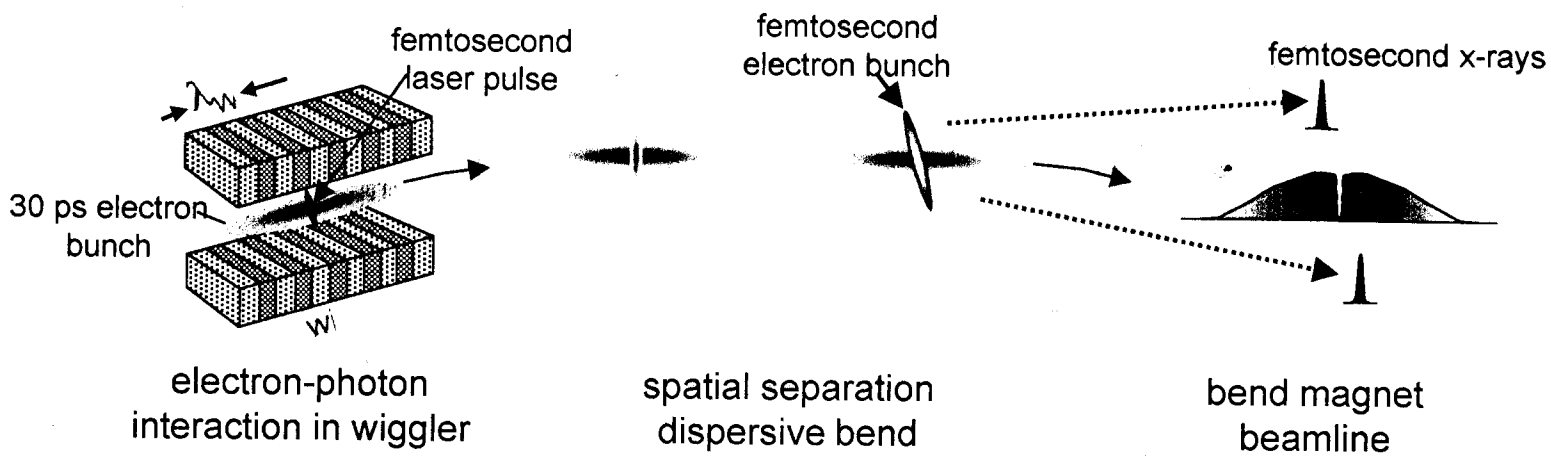
(femtosecond molecular vibrations, single molecule imaging LCLS...)

(Radiation-damage-induced explosion of T4 lysozyme.)



X-ray Spectroscopy @ 100 fS

(beam slicing)



needs

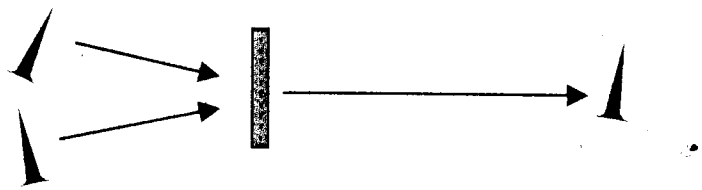
low fs x-ray flux =>

a need for high quantum efficiency, gatable (1 ns) detectors

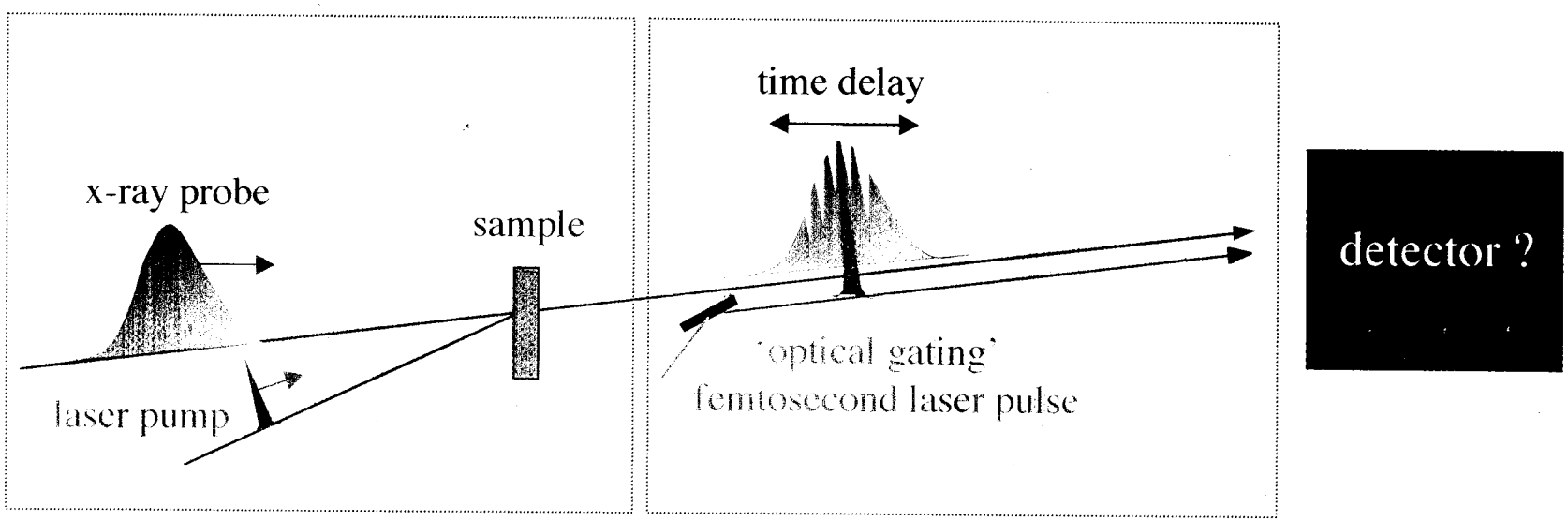
X-ray Spectroscopy @ 1-10 fs ??

Visible regime : optical - optical nonlinear processes

(frequency mixing)



Fast detectors via x-ray/laser nonlinear processes

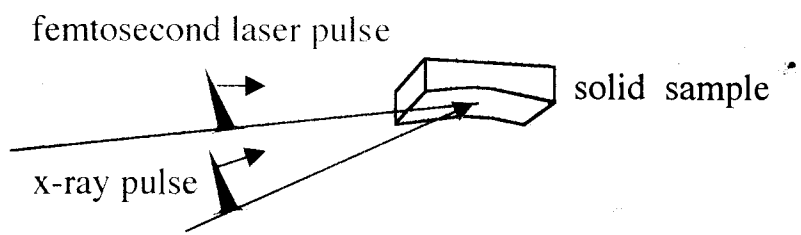


- Time-resolution limited only by laser pulse (<10 fs)

Detector mechanism : interesting fundamental science

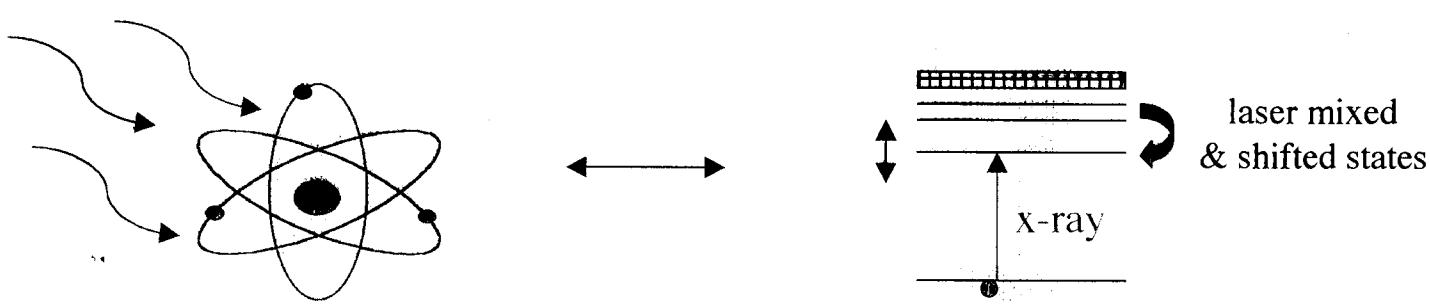
Understand intense laser-solid interaction

Highly non-equilibrium hot & dense matter; novel phase transitions : how are x-ray interactions modified ?



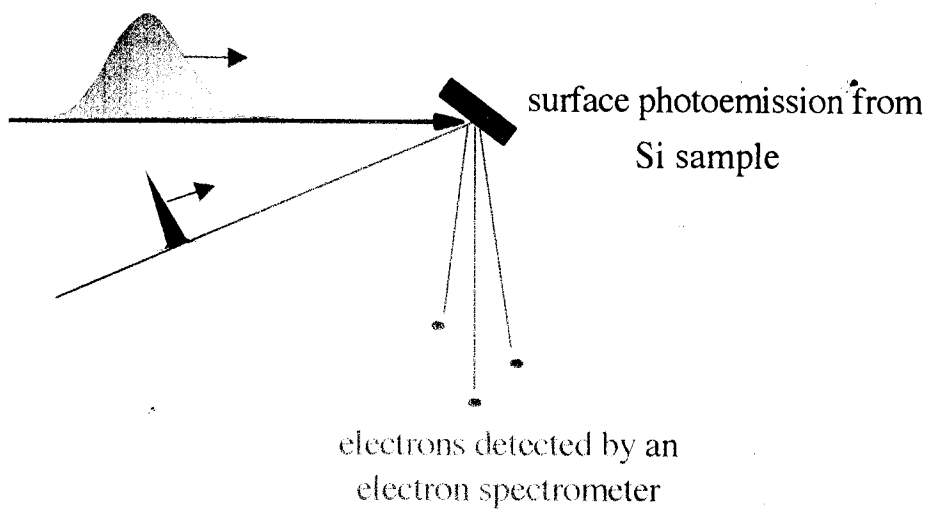
Understand atomic structure in an intense laser field

Synchrotron x-rays can probe novel atomic structure created by an intense laser pulse



Initial ALS experiments

Observe laser-perturbed XPS in a solid

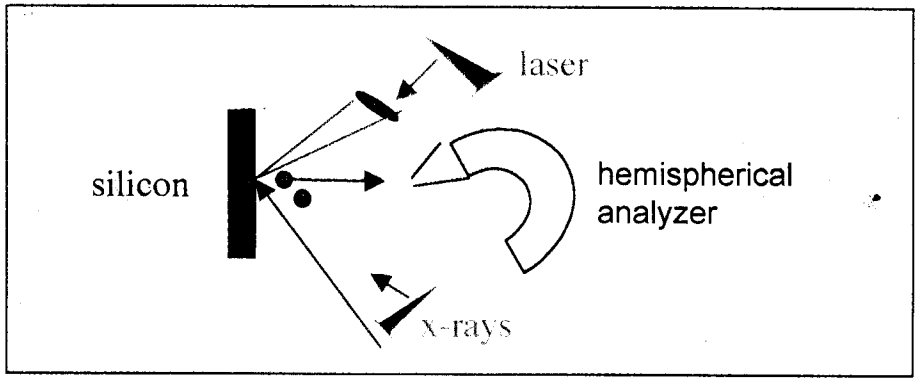


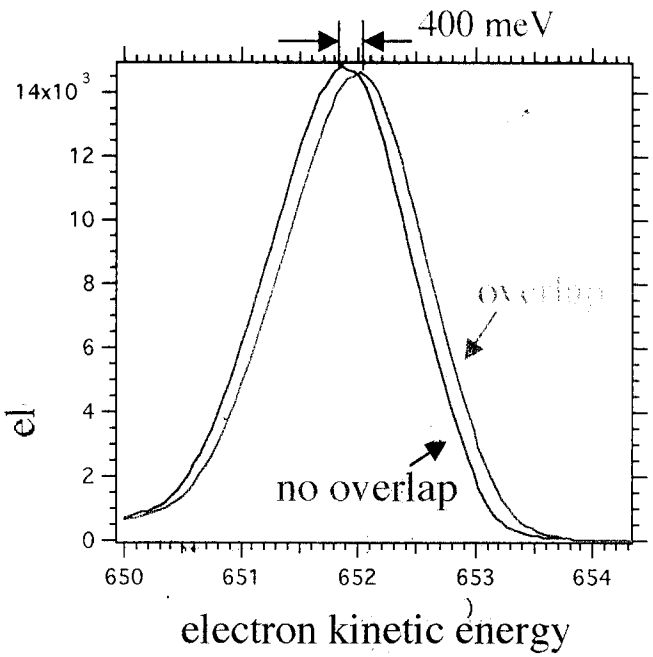
Choose solid rather than a gas since :

better vacuum compatiblity, high yield, physics not yet demonstrated
(@ available beamline)

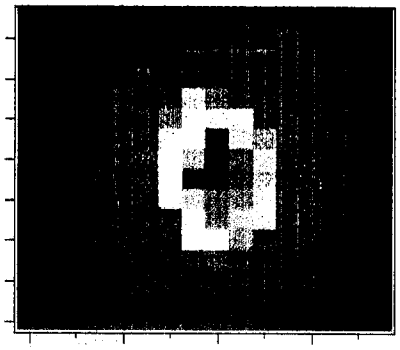
Low power experiments : Laser-induced Si 2p XPS shift

When laser & x-ray spots are overlapped, migration of laser generated carriers causes an XPS shift.

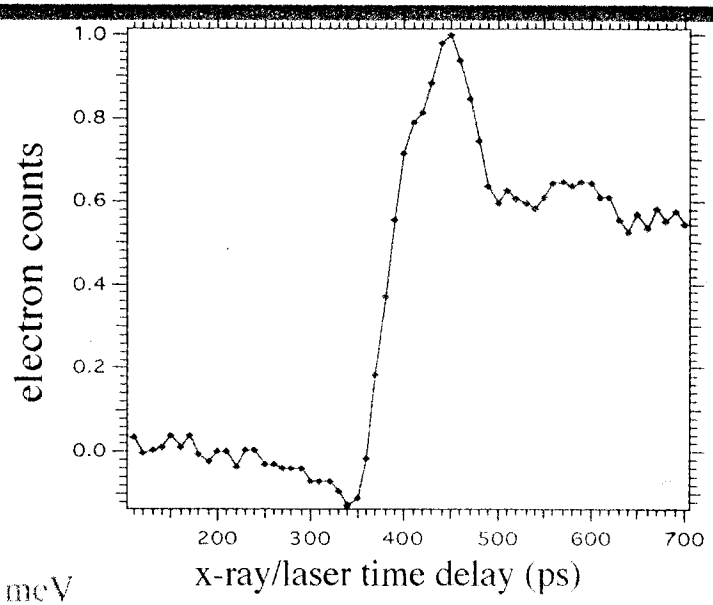


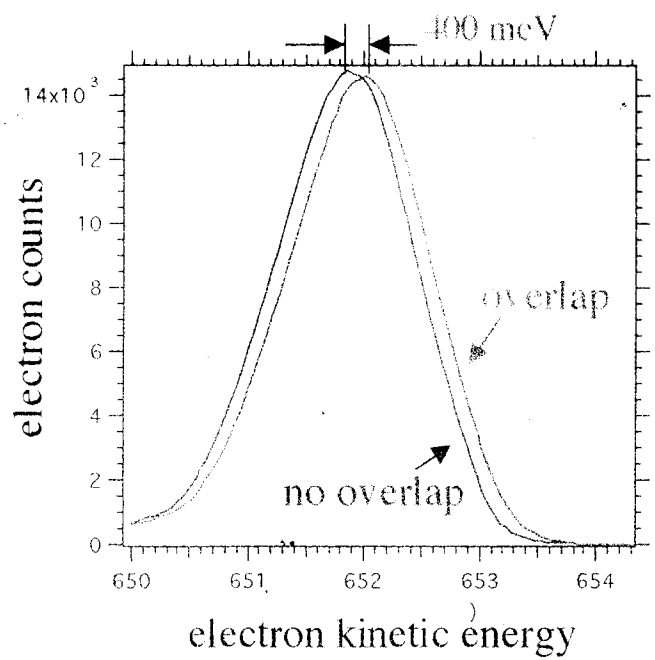


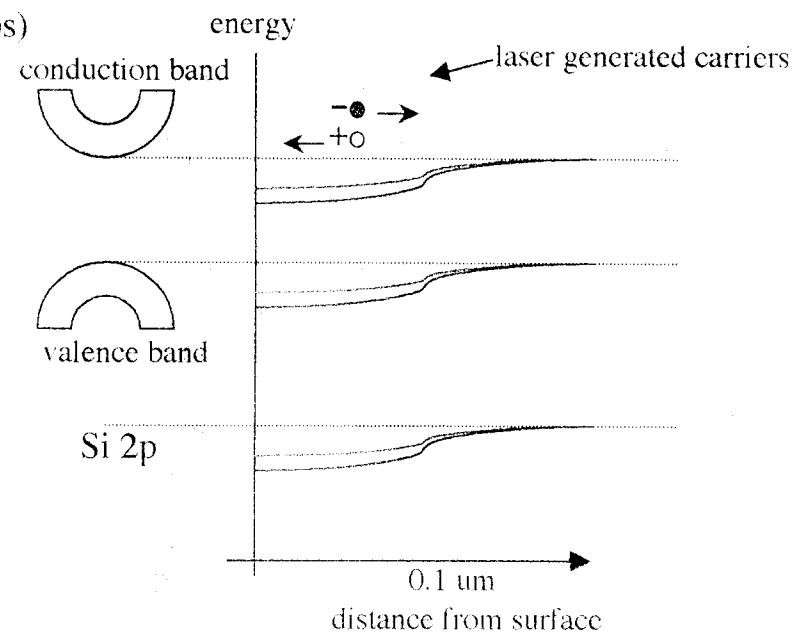
X-ray/Laser spatial cross-correlation



Low power experiments : dynamics of Si 2p shift

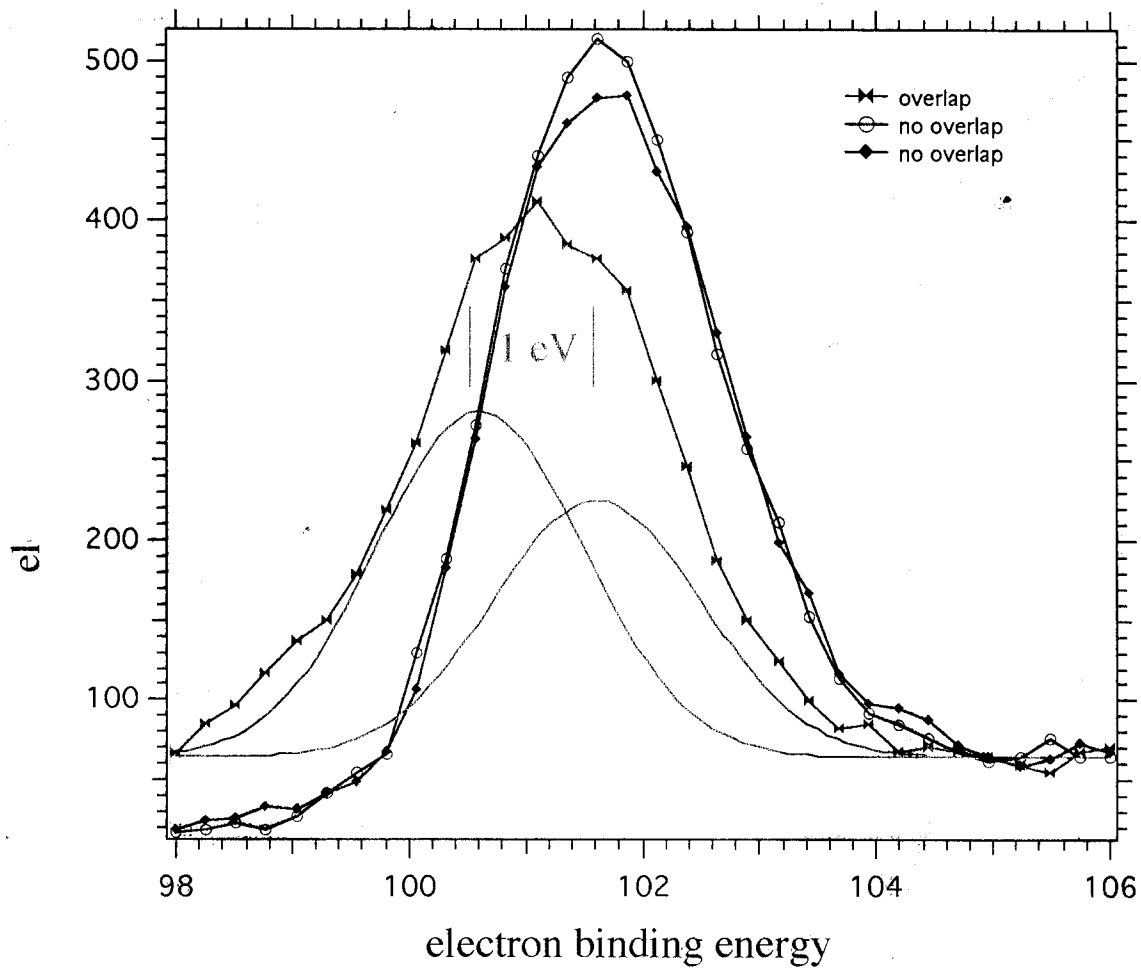






Photoelectron spectrum at high power

FWHM = 2.1 eV

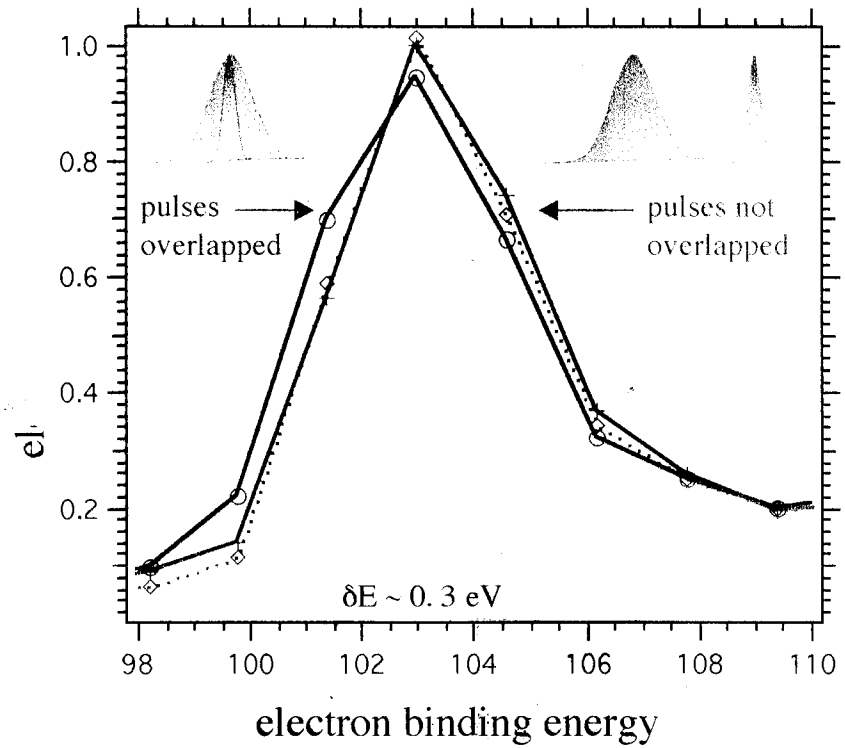


High power experiments : Laser-perturbed Si 2p spectra

Laser : 1-100 J/cm^2, 10^{10} -10^{15} W/cm^2

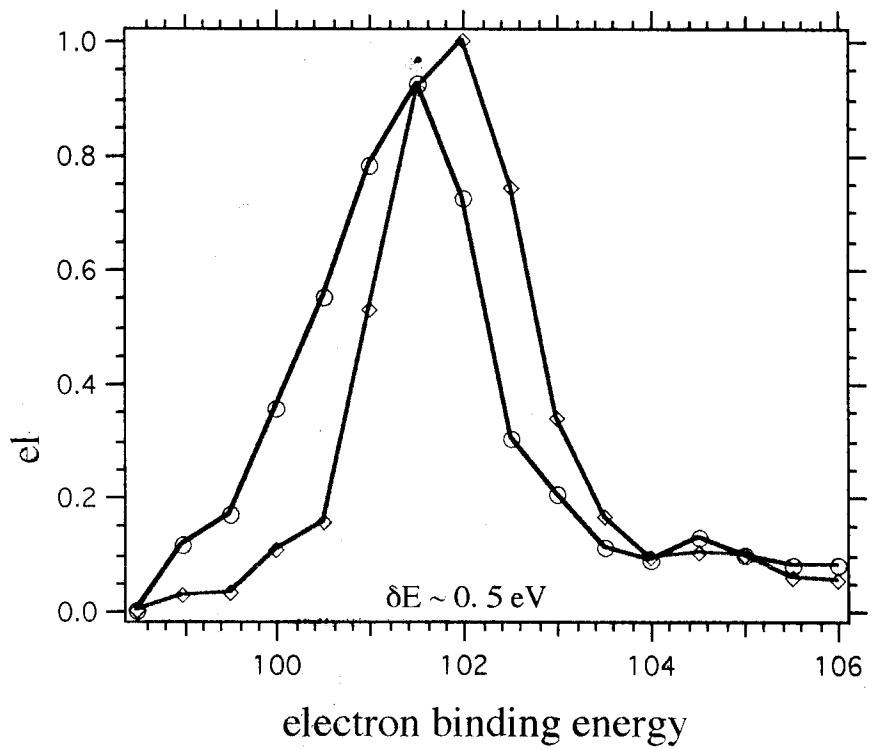
100 uJ

I ~ 2×10^{12} W/cm^2



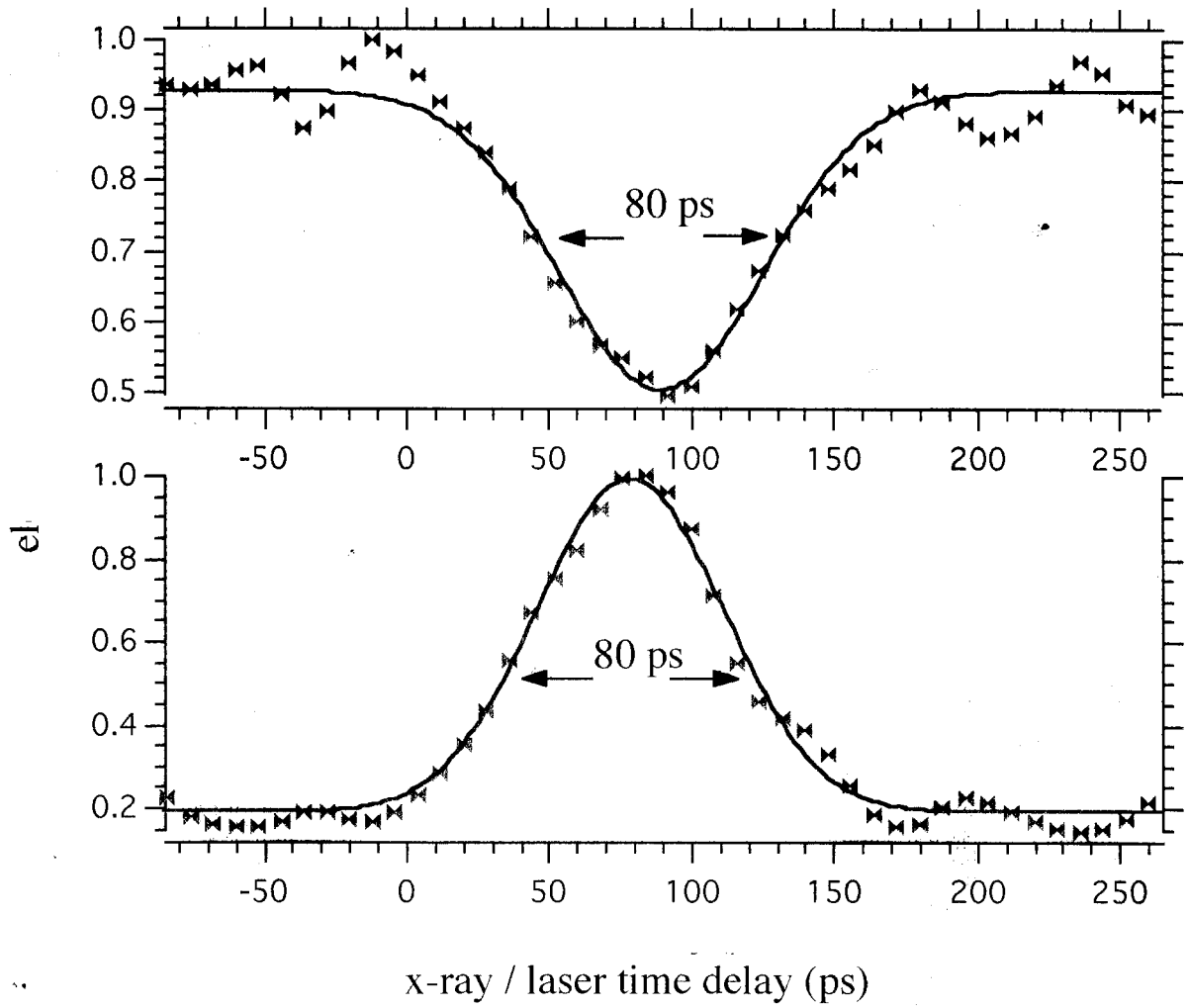
300 uJ

I ~ 6×10^{12} W/cm^2



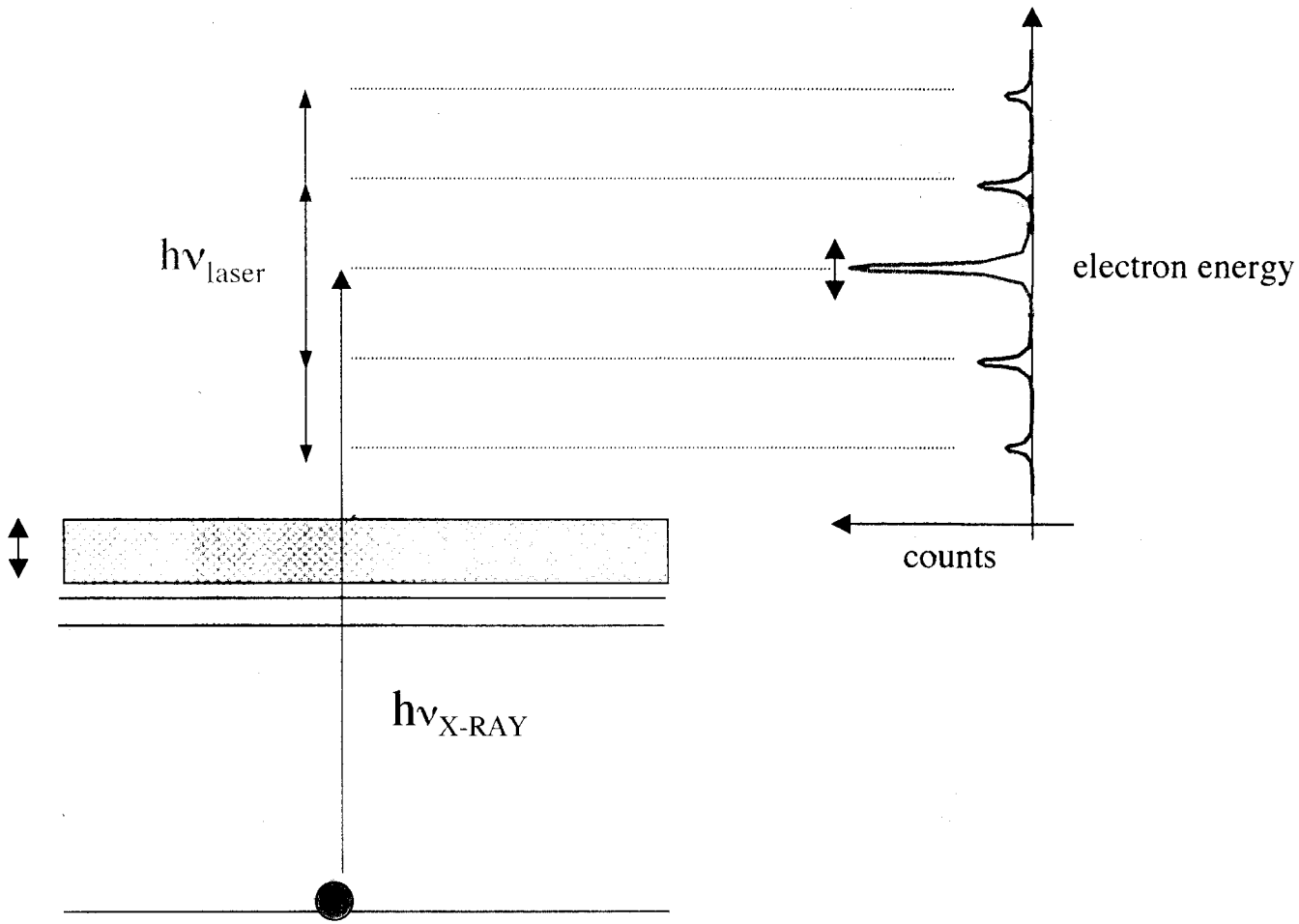
·Shift is NOT due to : (1) low power carrier dynamics, (2) AC stark shift, (3) sample charging, or (4) continuum scattering.

X-ray/laser cross-correlation



Optical gating via continuum scattering

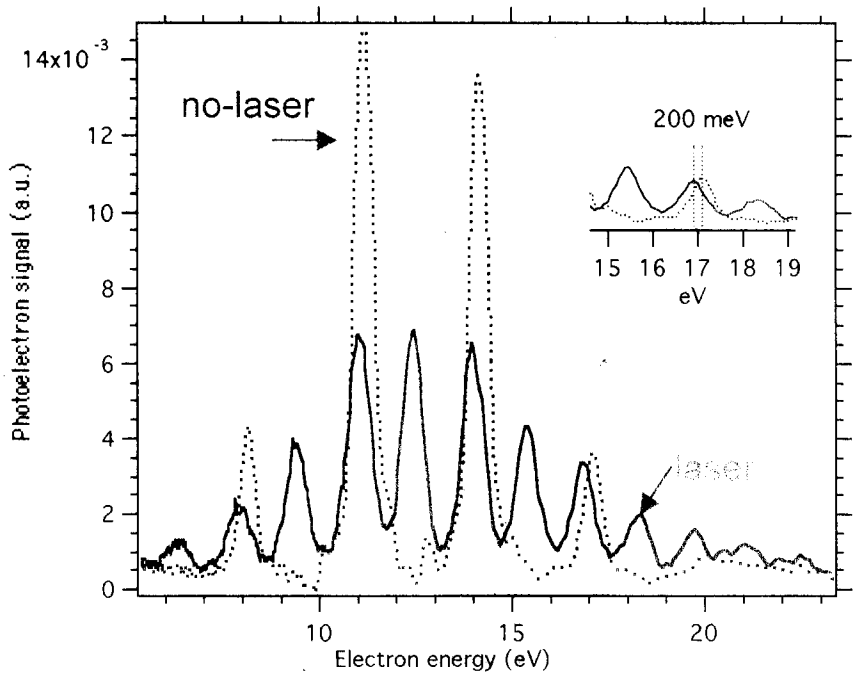
Look for laser induced modifications to XPS : (1) optical scattering and (2) AC stark shift



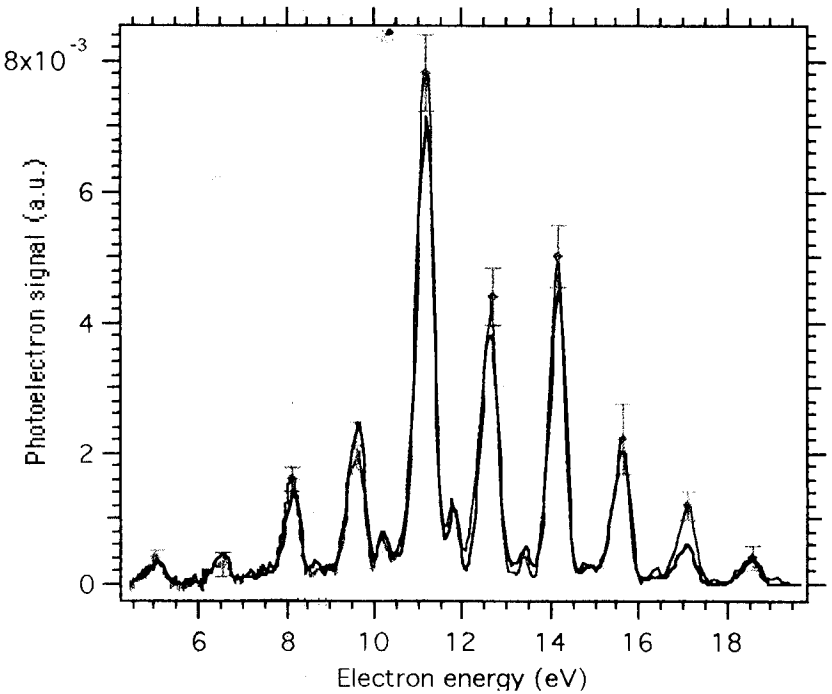
Advantage of this approach : no x-ray resonances => works for many x-ray wavelengths

Gas phase experiments : data

Photoelectron Spectra



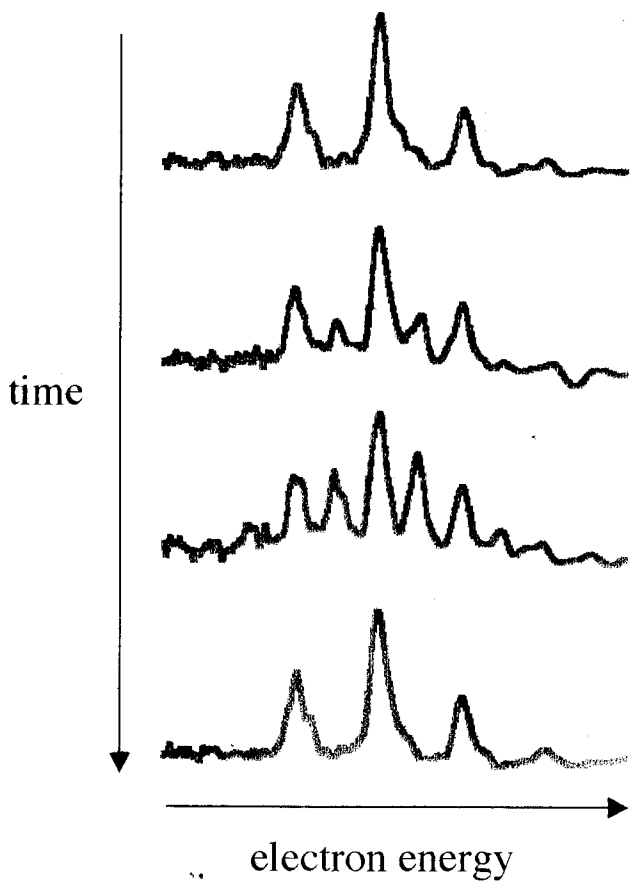
Comparison with theory



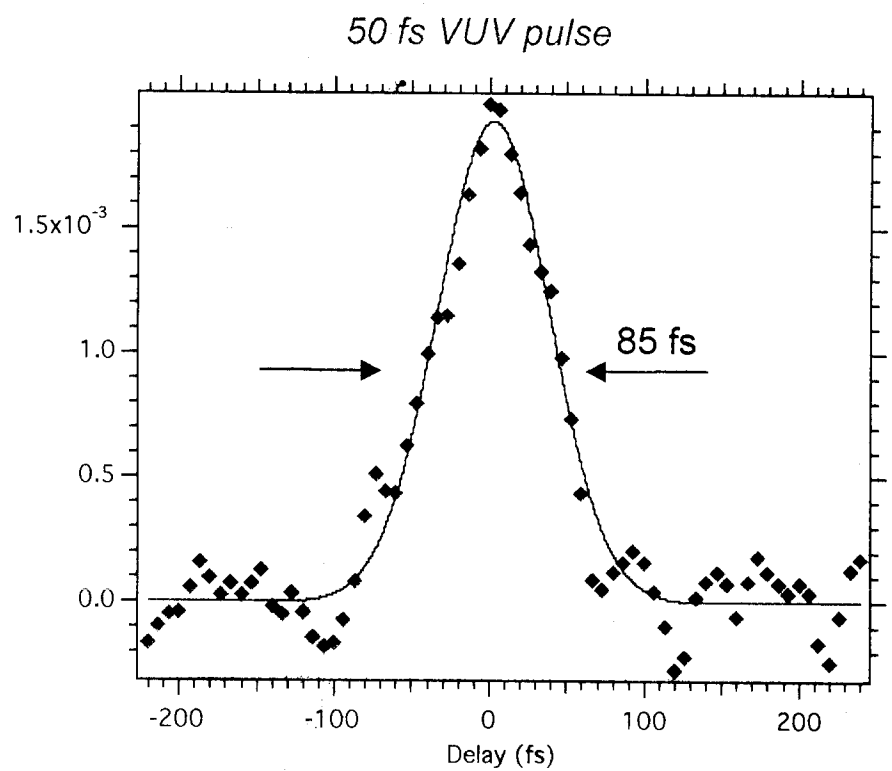
Glover, Schoenlein, Chin, Shank, Phys. Rev. Lett. 76, 2468 (1996)

Gas phase experiments : data

Photoelectron Spectra vs time



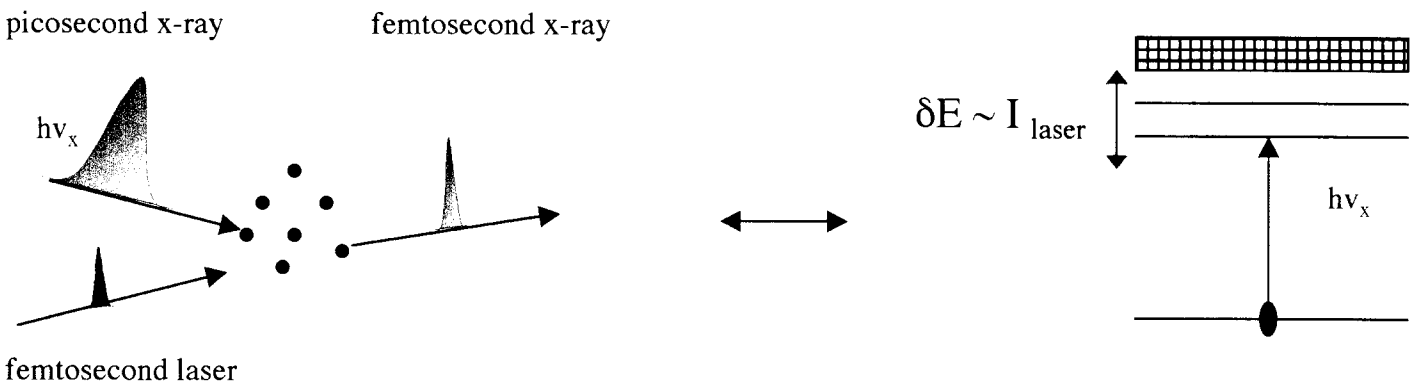
Laser-VUV cross-correlation Measurment



Glover, Schoenlein, Chin, and Shank, Phys. Rev. Lett. 76, 2468 (1996)

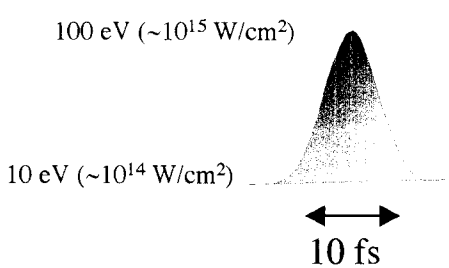
Modulation of x-ray absorption : X-ray Filters and Detectors

Time-filter via Stark shifts



absorption probability = $N\sigma L \sim 1$ @ 1 atm, 10^{-19}cm^2, L=1 cm

Rapidly shift through a resonance



$\delta E \sim I_{laser} \sim 100$ eV/10 fs = 1eV/0.1 fs

Summary

Two-color techniques

- As in optical spectroscopy, two-color (x-ray + laser) processes will play an important role in x-ray spectroscopy @ 1-10 fs regime.

Condensed matter (Si)

- High power : rapid phase-transition permits x-ray/laser cross-correlation.

Gas phase

- Demonstrated continuum scattering as an x-ray/laser two-color diagnostic.
- Detectors based on control of atomic structure ?