Argonne NATIONAL LABORATORY

X-raying laser-aligned atoms and molecules

Motivation
Current experiments on laser-aligned atoms
Proposed experiments on laser-aligned molecules

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Collaborators

AMO Physics Group

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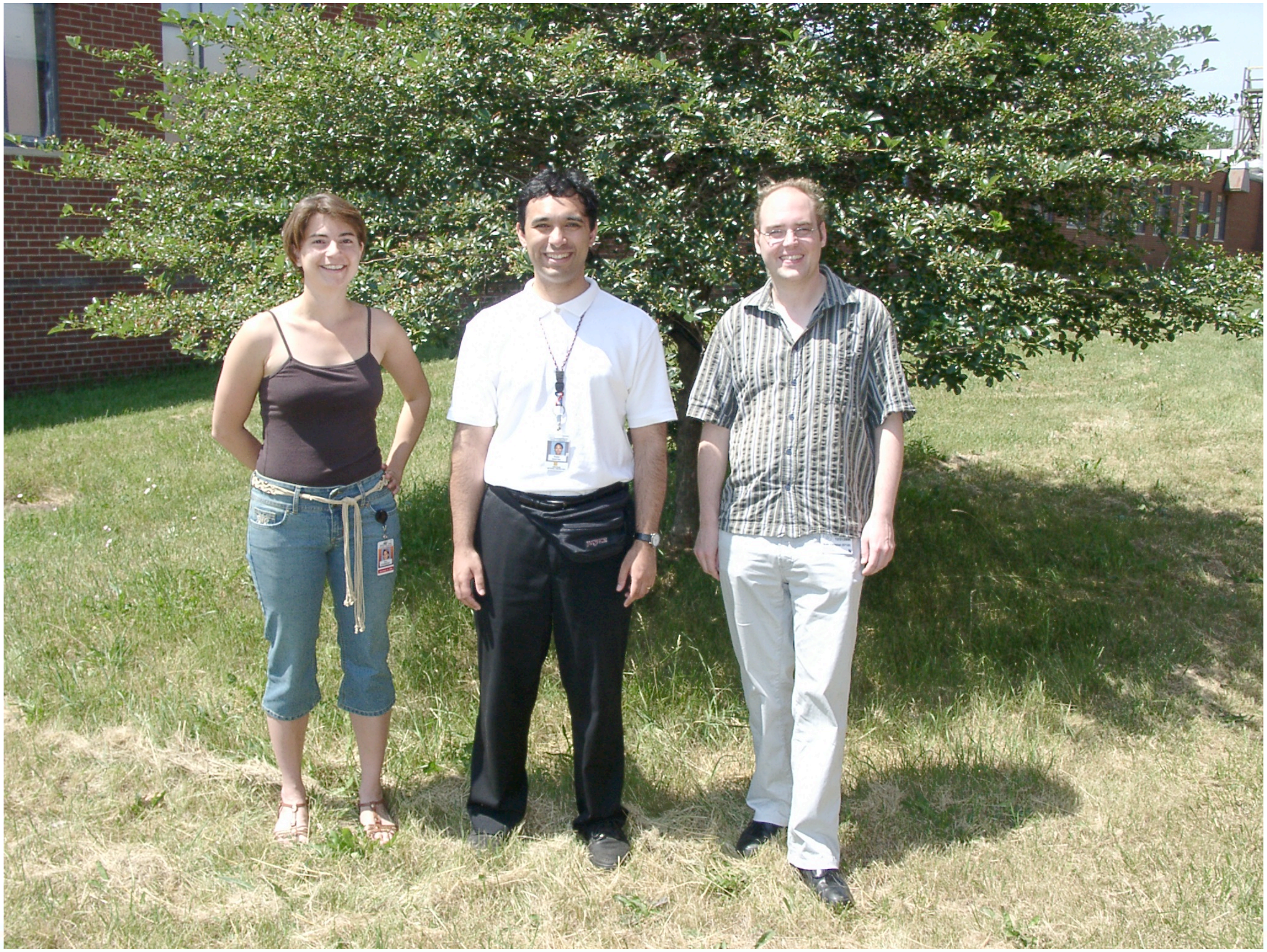
Advanced Photon Source

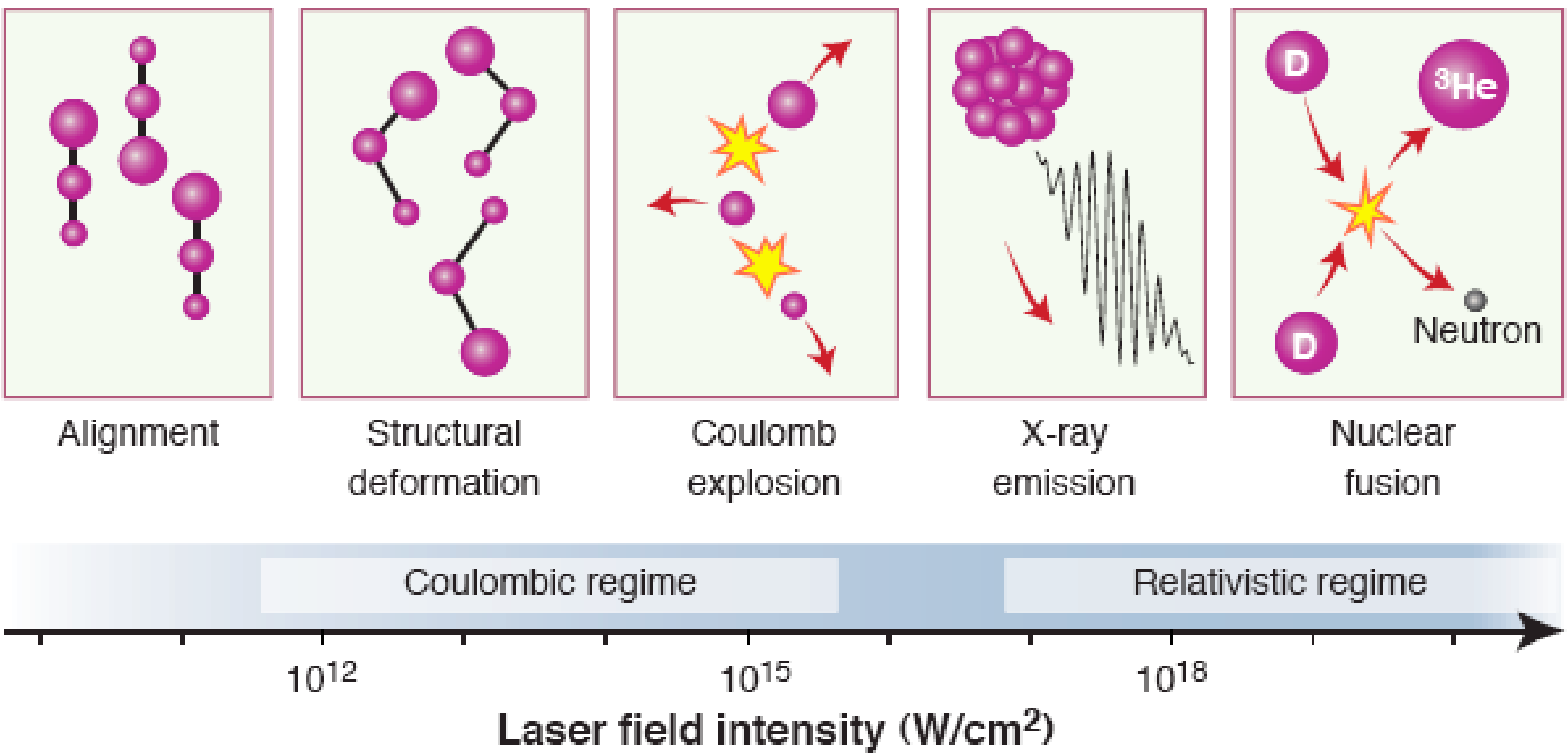
Eric Dufresne, Dohn Arms, Eric Landahl

Argonne AMO Group (Experimental Branch)

Argonne AMO Physics Group (Theory Branch)



What happens in strong optical fields?



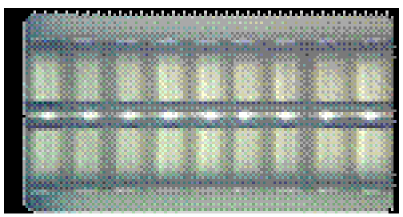
Yamanouchi, Science (2001)

Atoms in strong optical fields: I

High Harmonic Generation @ ~10^{15} W/cm^2

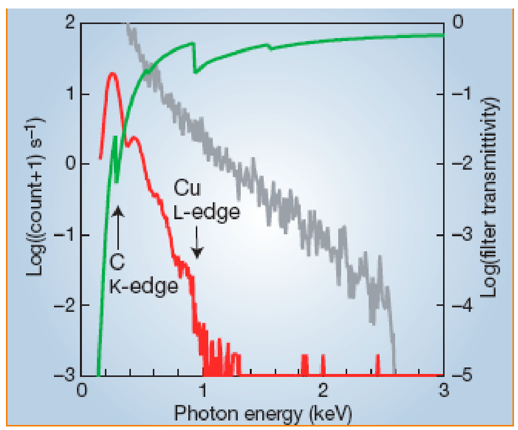
- *Tabletop source of coherent soft x-ray radiation*
- *The attosecond frontier*

Quasi-phase matching at 4nm and Harmonics from Ions (200-300 eV)

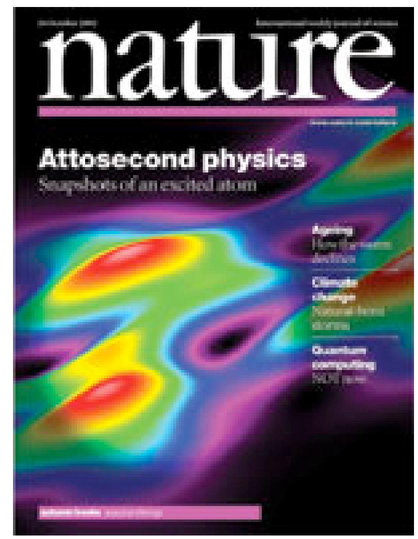


E. Gibson *et al.* Science (2003)

Coherent keV x-rays



Seres *et al.* Nature (2005)

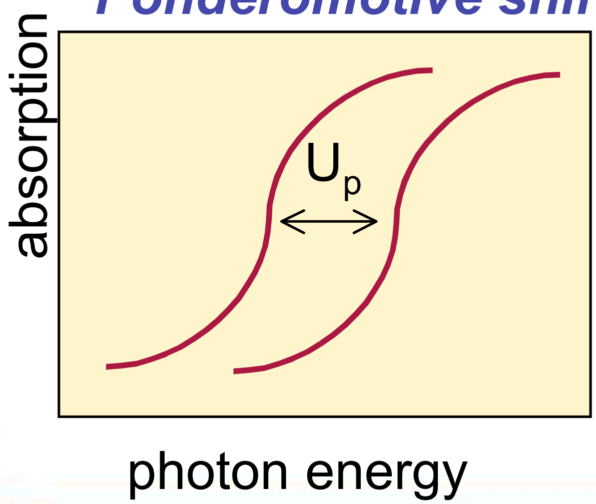


Atoms in strong optical fields: II

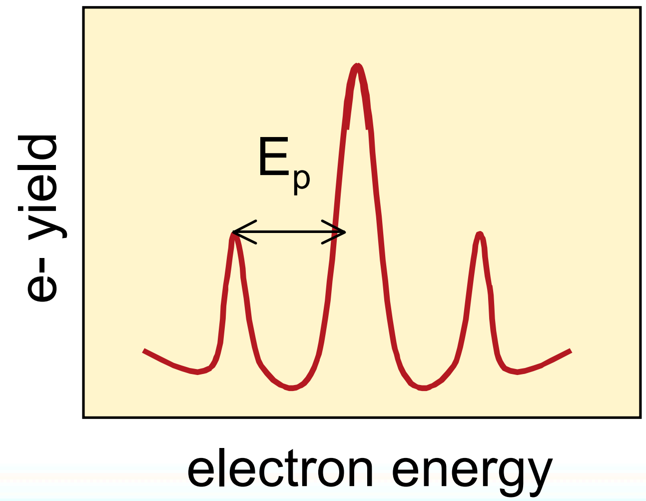
Motivations

- *Understand changes to x-ray processes in presence of strong laser fields*
- *Theoretical predictions*
  - *ponderomotive shift in threshold -> absorption spectrum*
  - *free-free transitions in continuum -> electron spectra*

Ponderomotive shift



Electron satellites



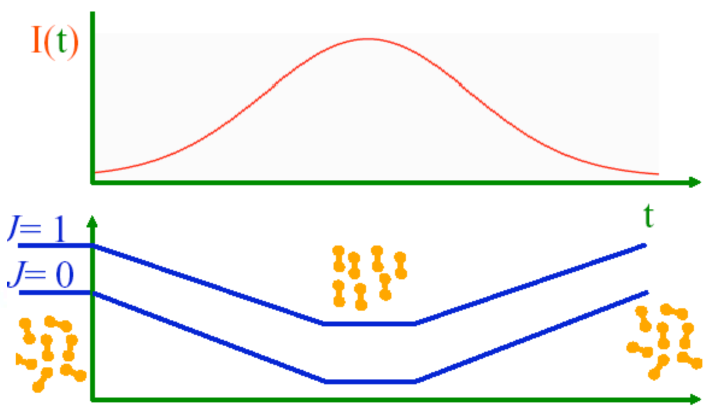
Molecules align in strong fields

Motivations

- *Stringent tests of photoionization and photodissociation*
- *Study behavior of molecules as fcn of strength of aligning potential*
- *Aligned molecules aid single biomolecule structure determination (Hajdu et al)*

Adiabatic alignment

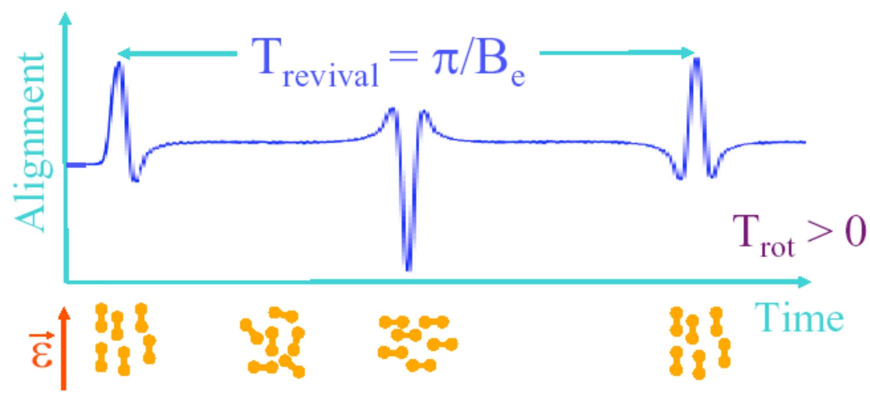
$\tau_{laser} > \tau_{rot}$



Friedrich & Hershbach, PRL 74, 4623 (95)

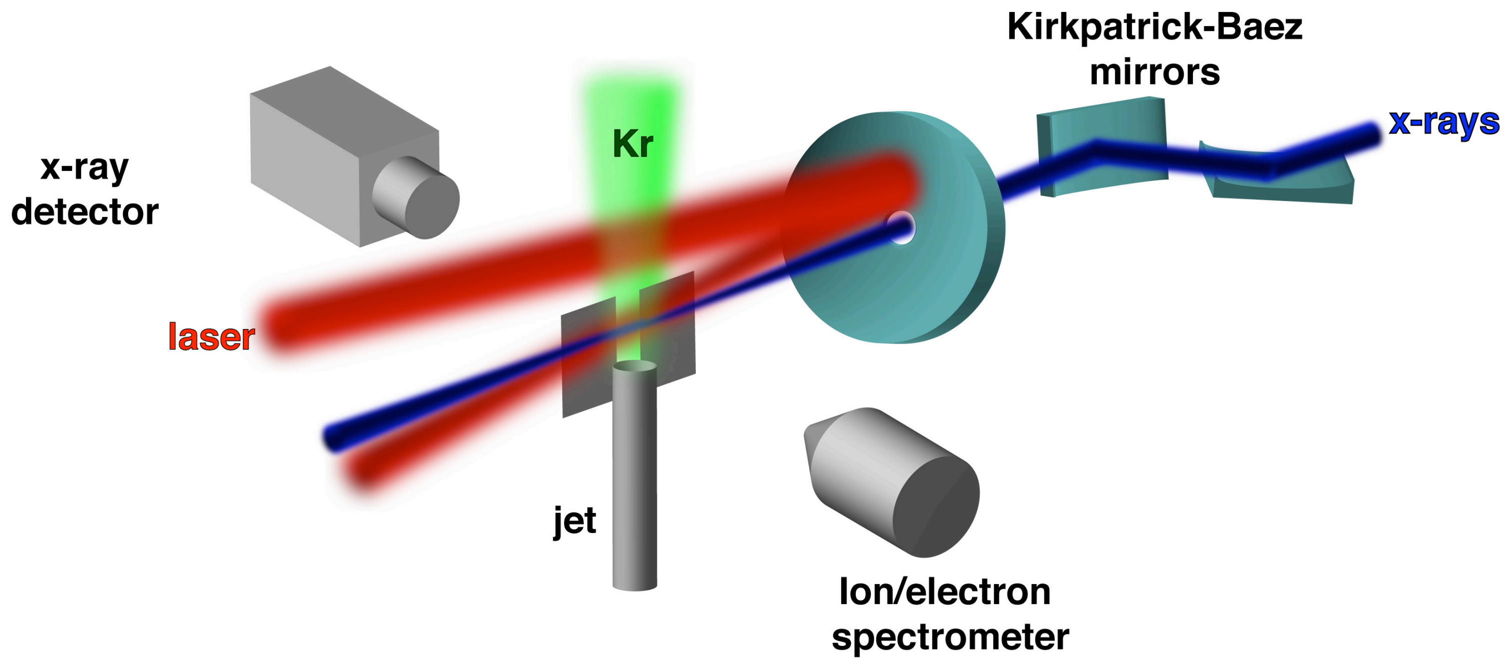
Field-free alignment

$\tau_{laser} < \tau_{rot}$



T. Seideman, PRL 83, 4971 (99)

X-ray microprobe of strong-field environment



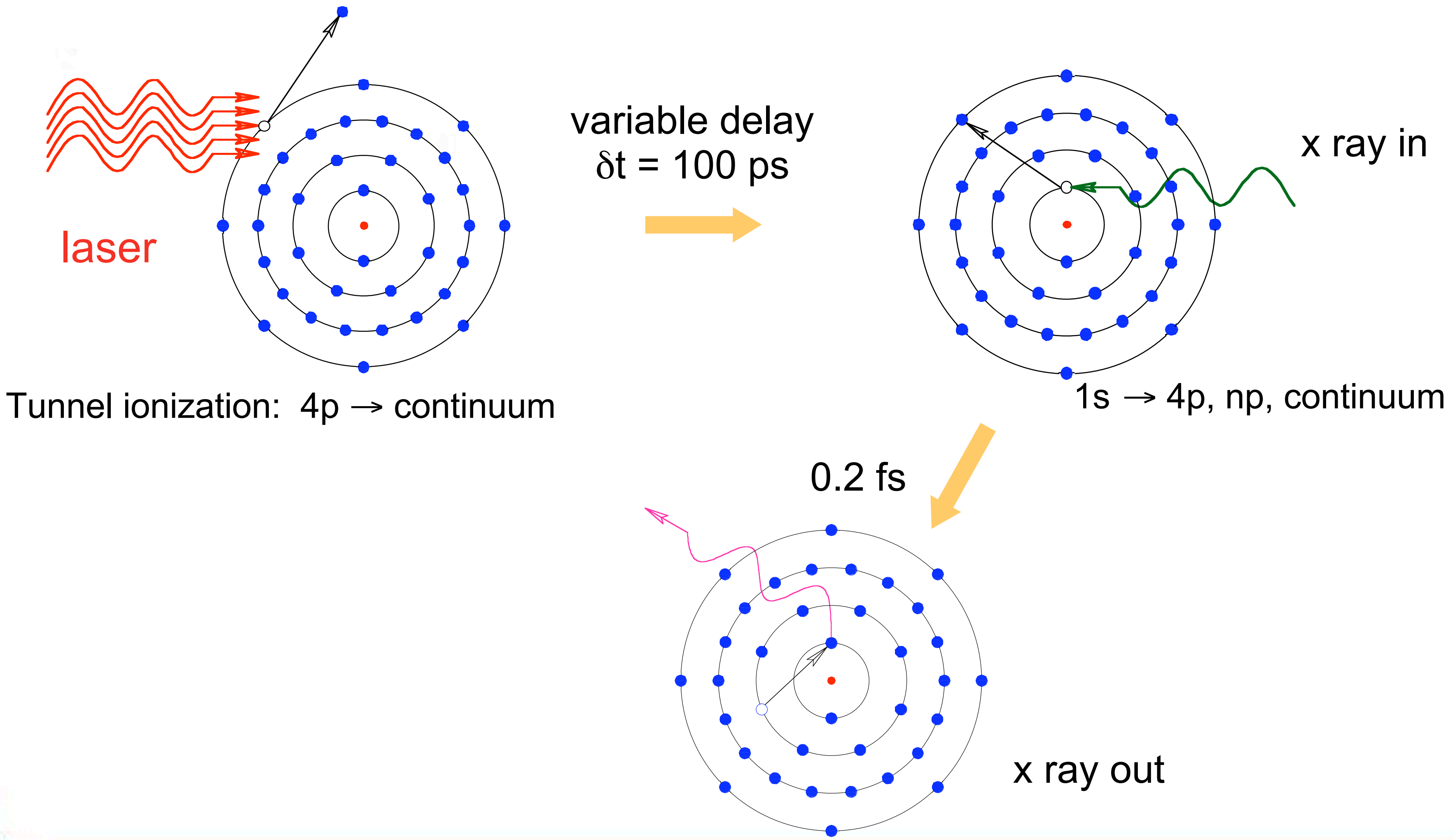
Laser: 30-100 micron focus, 2.5 W, 40 fs, 1-5 kHz, 800 nm

APS x-ray: ~10 micron focus, 10^6 x rays/pulse, 10^{-4} bandwidth, 100 ps

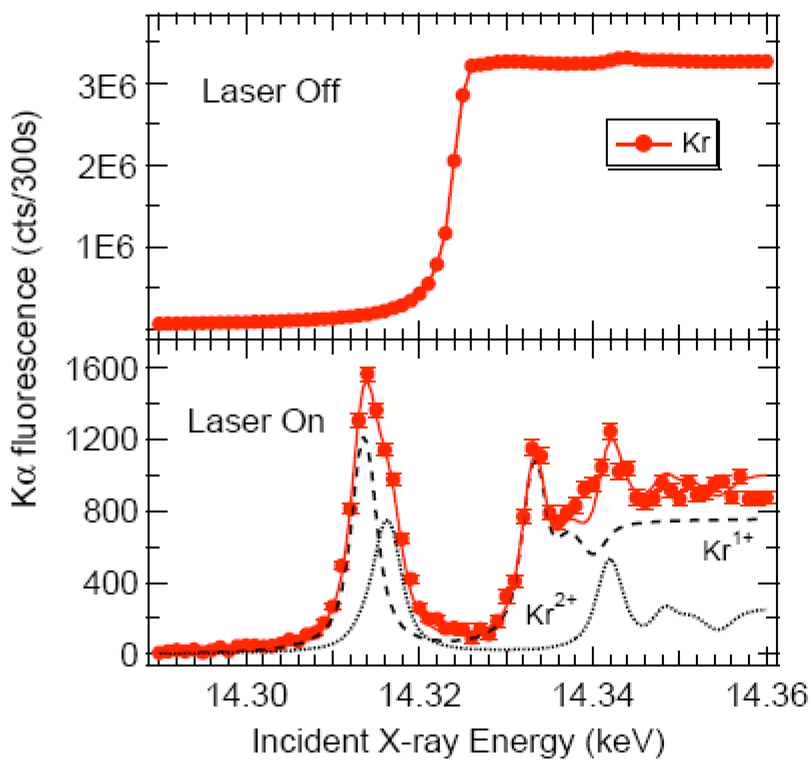
X-ray microprobe studies with atoms

- ***Isolated atom response***
  - *spectroscopic probe of the ions*
  - *initially ion velocities are thermal (~300 nm/ns)*
  - *x-ray in/x-ray out: sub-fs, collision-free signature*
- ***Collective ion dynamics - Coulomb Expansion***
  - *time and spatial resolved spectroscopy*
- ***Probe alignment in strong-field ionization***
  - *polarized x-ray spectroscopy*
  - *monitor & control alignment dynamics*
- ***X-ray / laser cross correlation of x-ray pulse duration***
  - *bunch length scaling $I^{1/3}$*
  - *intrinsic time resolution ~1/2 laser pulselength (20fs)*

X-ray probe of laser-produced Kr^+



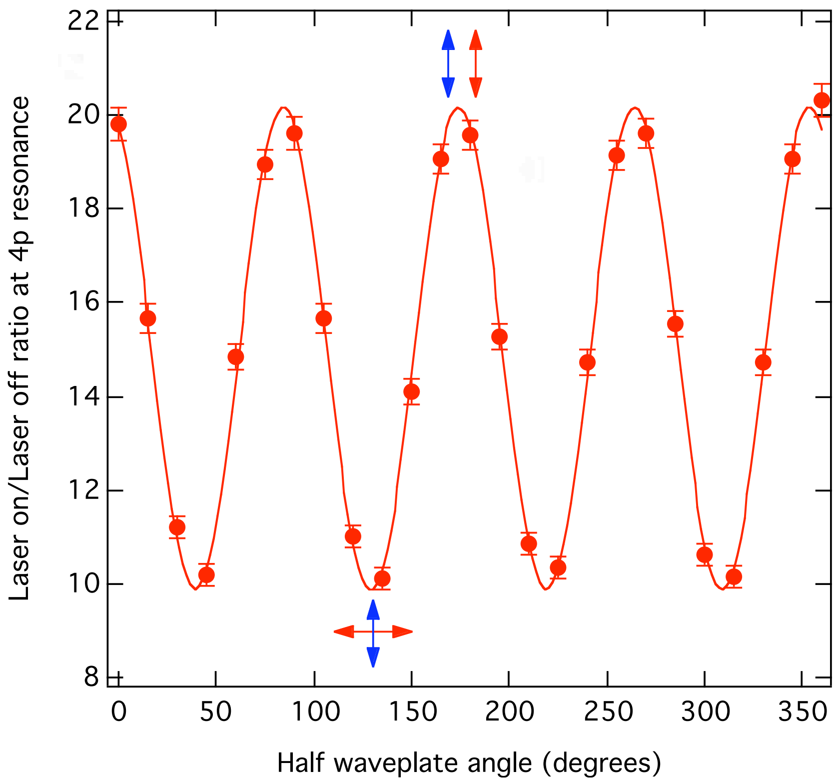
X-rays selectively probe Kr^{+}

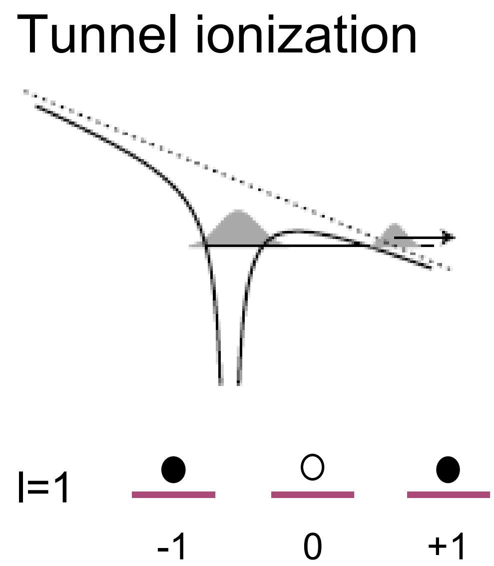


Experimental data:
3 parameter fit
Kr: Kr^{1+}:Kr^{2+}

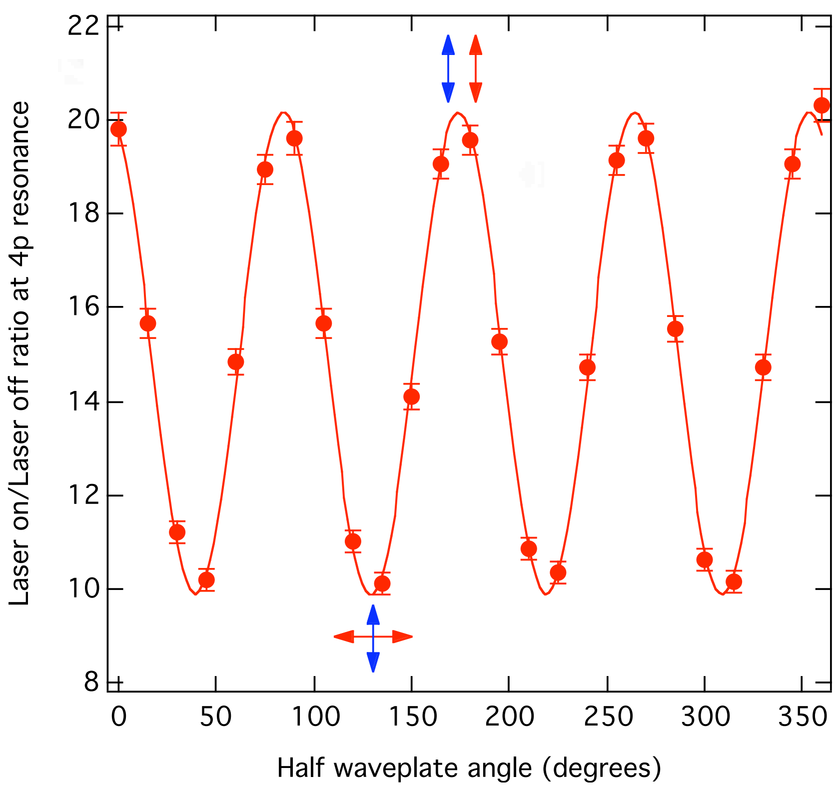
Kr ion theory: ab initio relativistic configuration interaction calcs
Pan, Beck, O'Malley JPB (2005)

Polarized resonant x-rays reveal alignment in tunnel-ionized Kr



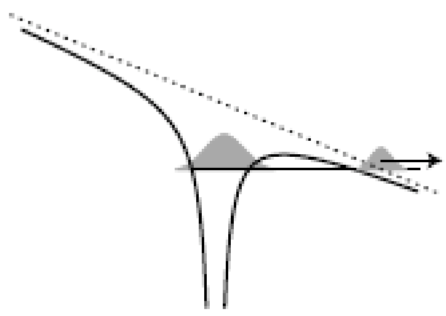


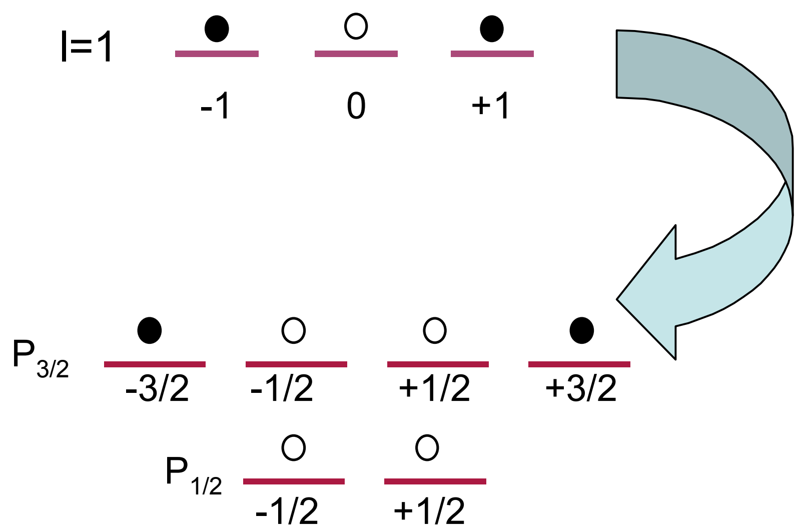
Polarized resonant x-rays reveal alignment in tunnel-ionized Kr



$$R = \frac{\text{parallel}}{\text{perpendicular}} = \frac{10\rho_0 + 4\rho_1}{4\rho_0 + 7\rho_1}$$

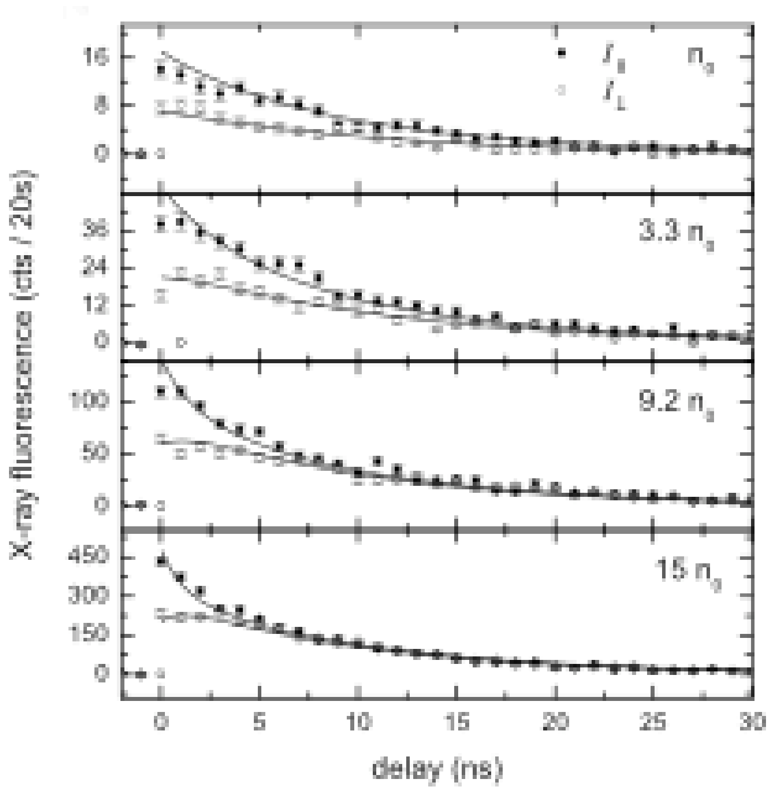
Tunnel ionization





Spin-orbit coupling required to explain the degree of alignment

Alignment dynamics 1:
Decay of alignment



n_0 = 1.3 x 10^{14} /cm^3

Observations:

- *Initial ratio ~2:1*
- *Disalignment density dependent*
- *Coulomb expansion timescale*

~10 ns at all densities, $n_{ion} = n_e$

Model disalignment:

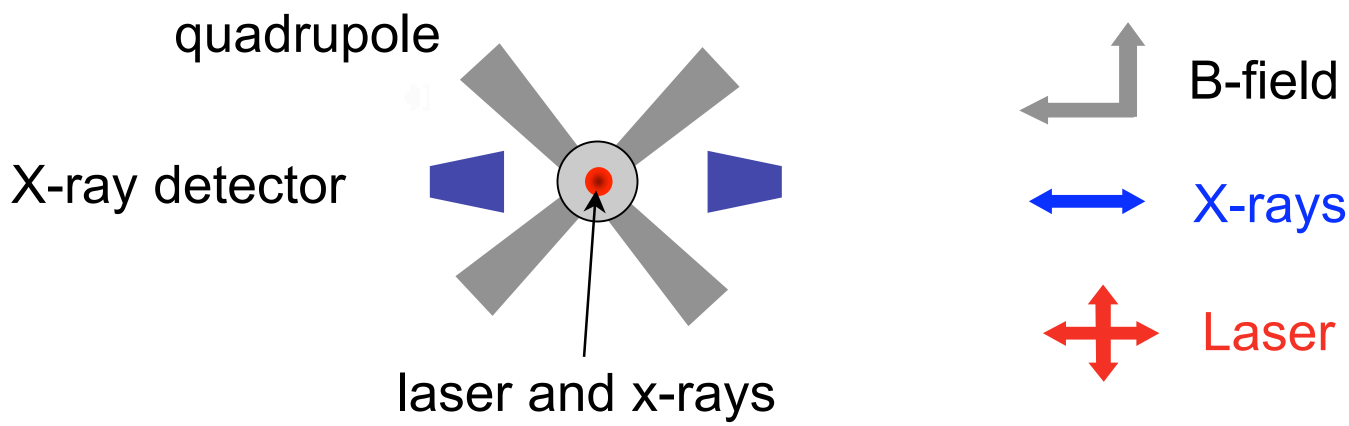
e^- - Kr^+ collns → m-sublevel transitions

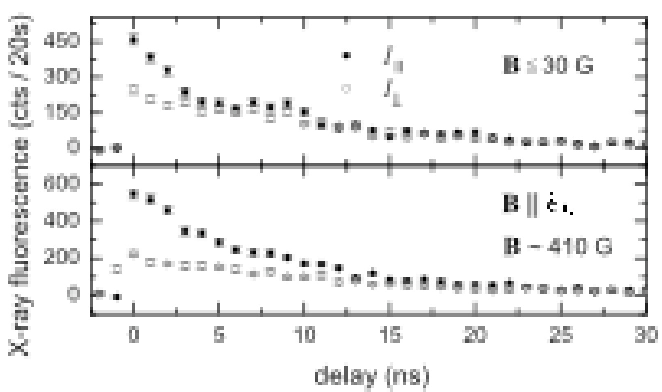
$$\hat{H} = -\frac{1}{2}\nabla^2 - \frac{1}{r} - \frac{1}{r^3}\sqrt{\frac{4\pi}{5}}(\mathbf{Q}_2 \cdot \mathbf{Y}_2)$$

Low KE electron-ion collisions

Alignment dynamics 2:

Preservation of alignment w/B-field





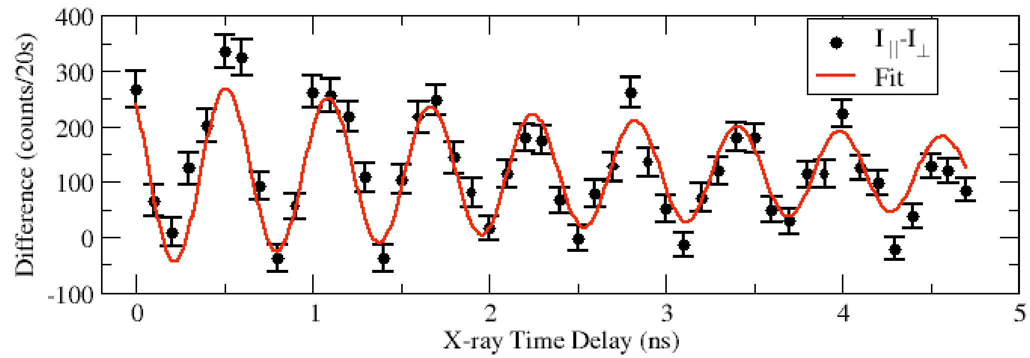
$n = 2.3 \times 10^{15}$ /cm^3

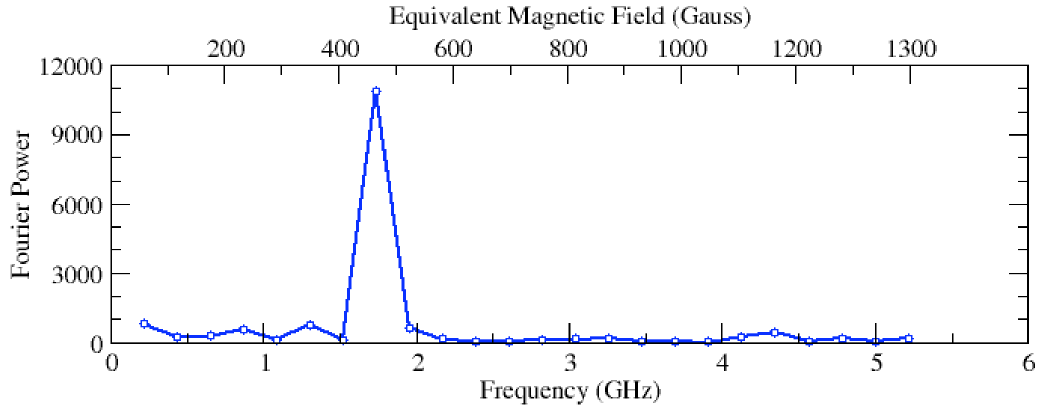
B = 0 G $\tau_{disalignment}$ ~ 2 ns

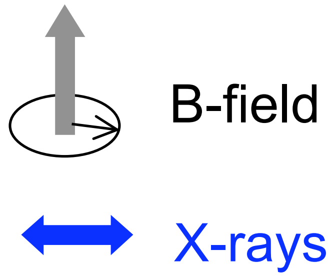
B ~410 G $\tau_{disalignment}$ ~ 8 ns

Alignment dynamics 3:

Coherent spin precession







Ω = $2g_j\,(p_{3/2})\,\mu_0$ **B**
= 3.73 MHz/G **B**

B ~ 450 G

Summary

•Versatile x-ray probe of atomic behavior in strong fields

- spectroscopic
- spatial
- time
- polarization

→

Isolated atom response
Collective ion behavior
Alignment dynamics
in a plasma environment

Molecules align in strong fields

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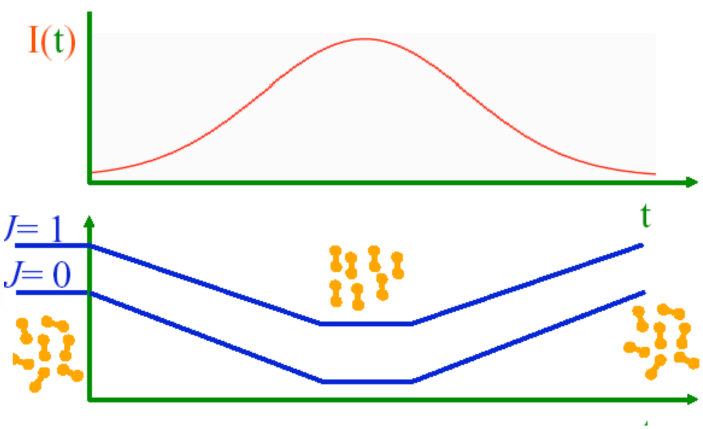

Friedrich & Hershbach, PRL 74, 4623 (95)

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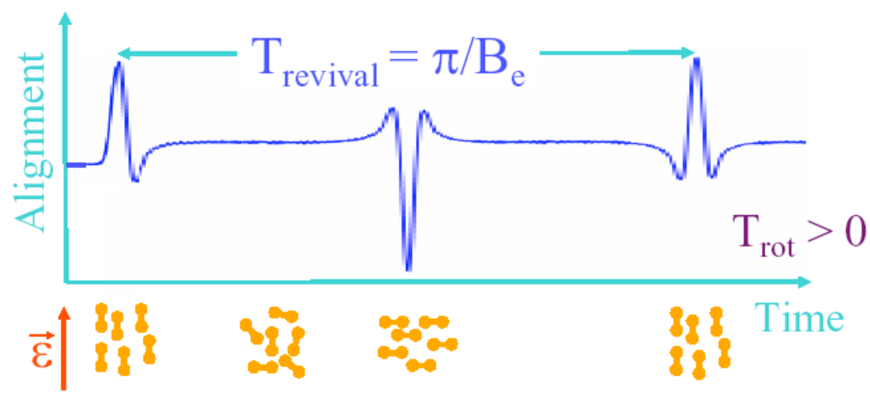

T. Seideman, PRL 83, 4971 (99)

Laser-aligned molecules

$$V_{int} = 1/2\ E(t)^2\ (\alpha_{||}\cos^2\theta + \alpha_{\perp}\sin^2\theta)$$

$$\Delta\alpha = \alpha_{||} - \alpha_{\perp}$$

X-ray advantages

- *no dynamic alignment by probe pulse (CE laser)*
- *degree-of-alignment thru near edge spectra*
- *change in structure by EXAFS (0.01Å)*
- *atomic resolution structures by diffraction*

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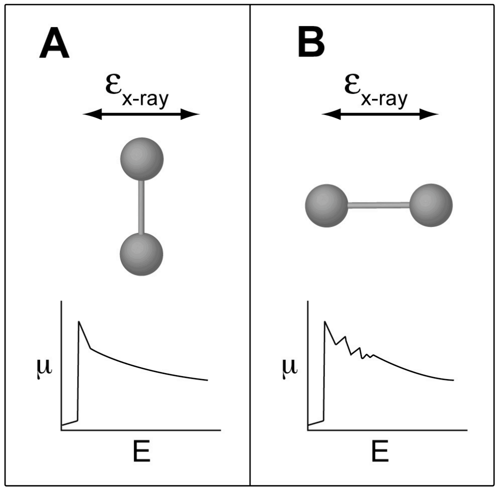
Br_2 EXAFS



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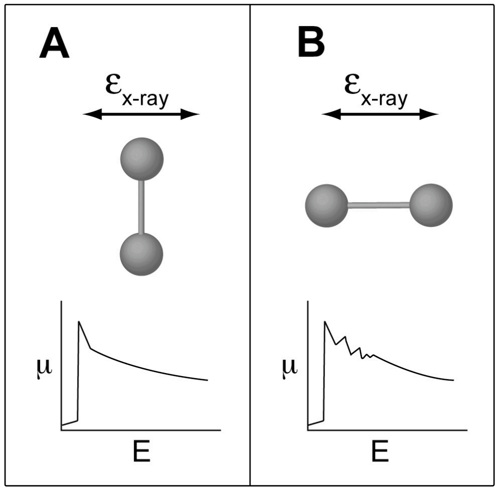
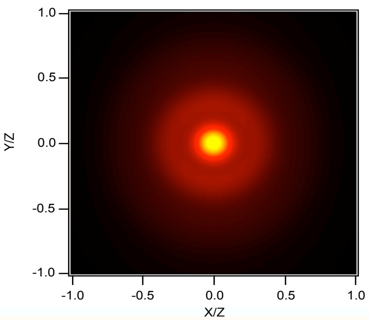
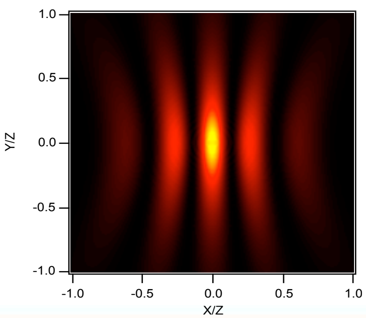


Isotropic

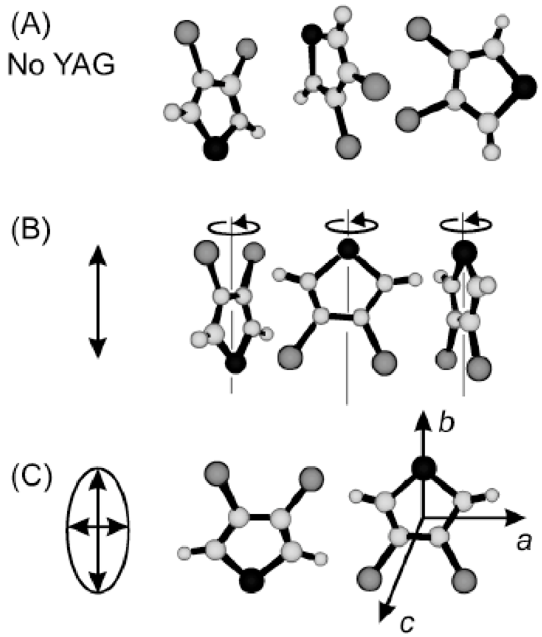


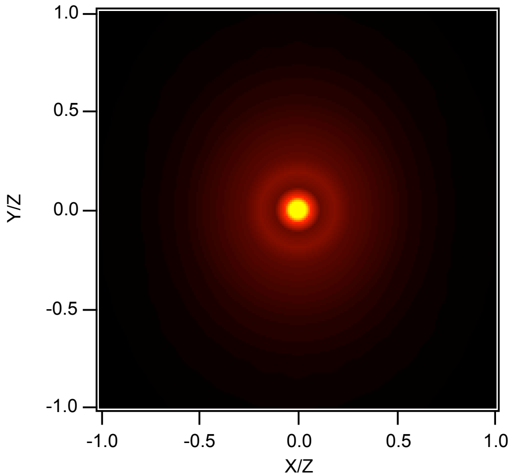


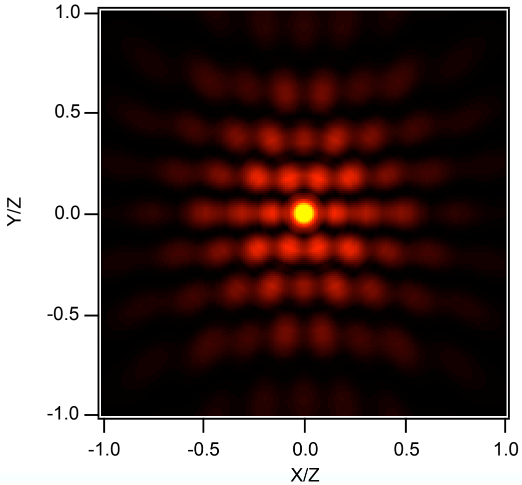
Aligned

Alignment of complex molecules

3-D alignment w/elliptically polz'd fields
3,4 dibromothiophene

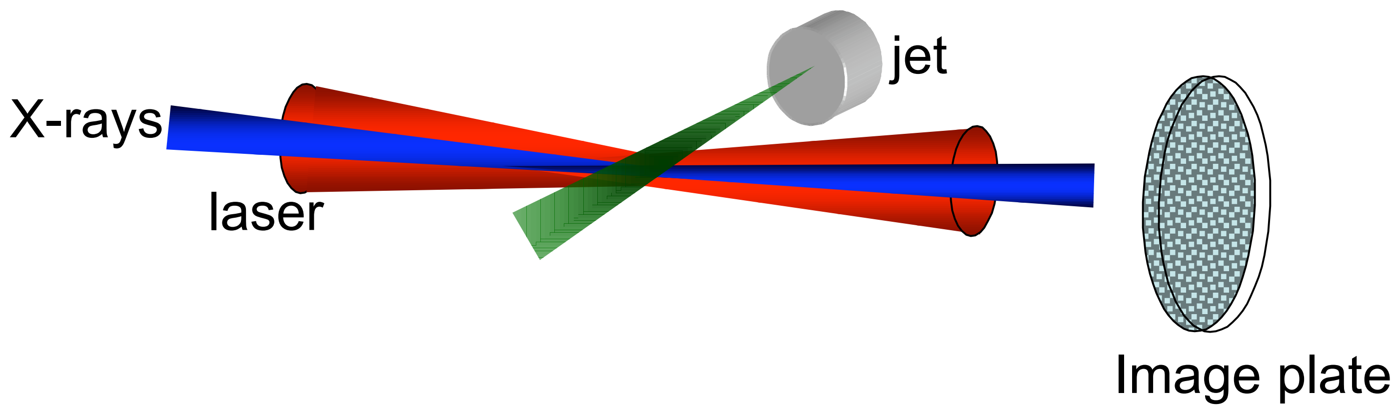




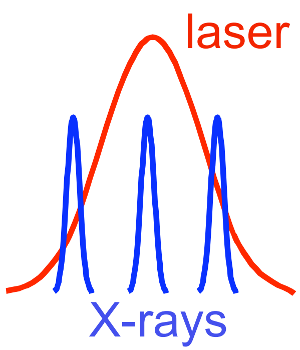


J.J. Larsen et al., PRL 85, 2470 (2000)

X-raying laser-aligned molecules



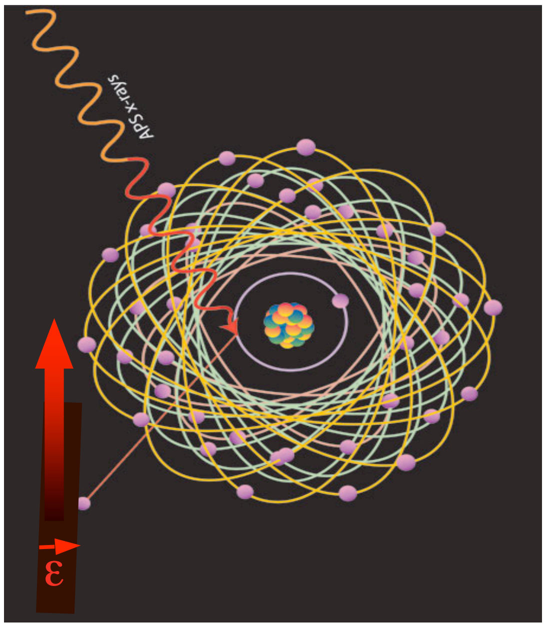
Aligning laser pulse ~120 ps, 10^{12}W/cm^2, 30μm,1 kHz
 # aligned molecules in volume ~ 10^6 - 10^{10}
APS: 10^6 x-rays/pulse, 0.01% BW, 100 ps, 1kHz
ERL: 10^8 x-rays/pulse, 0.1% BW, 50 fs, 1MHz
Time to acquire image of 3,4 DBT 1.5 hrs w/10^8/pulse



ERL: 1- probe evolution of adiabatic alignment
2- enable probe of impulsive alignment

Photon starved experiments

AMO

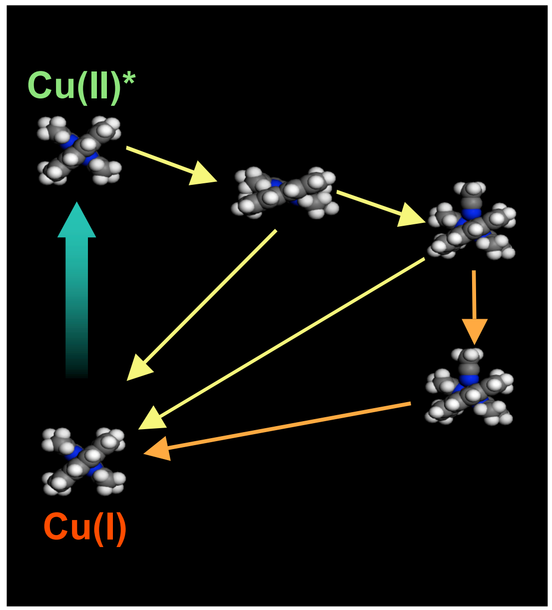


Spectroscopy

Isolated atoms & molecules in strong fields

~10^{14}/cm^3

Chemistry



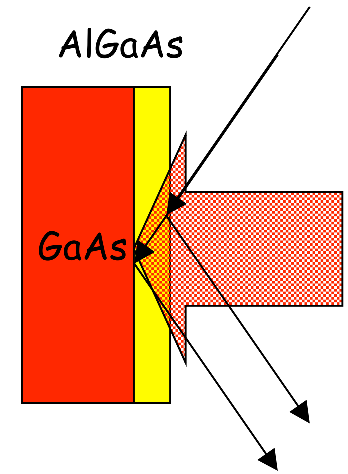
Spectroscopy

Transition state structures

Molecules in solution

~10^{18}/cm^3

Condensed Matter



Diffraction

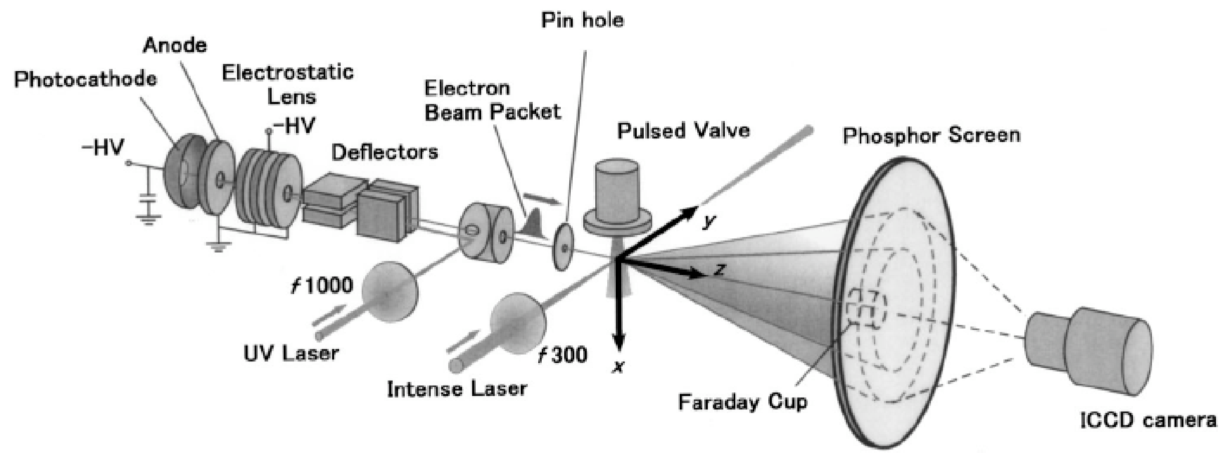
Scattering, spectroscopy

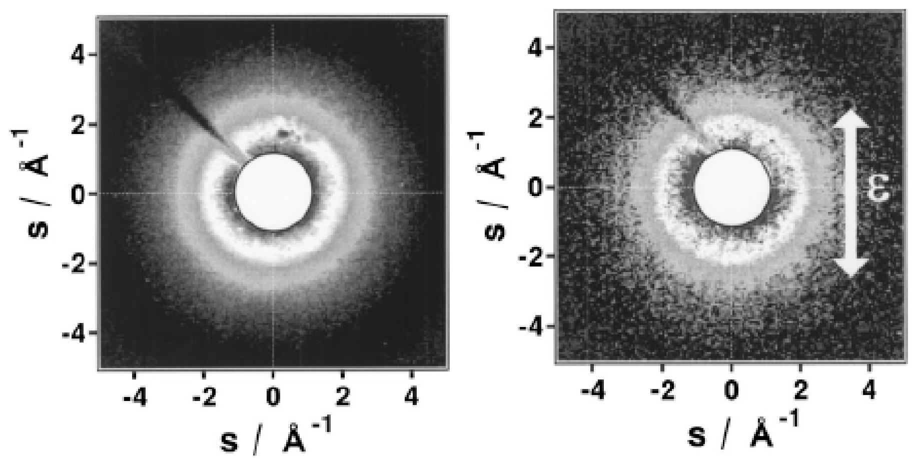
phonon dynamics,magnetism

Crystalline structures

~10^{22}/cm^3

Alignment of CS_2 probed by electron diffraction

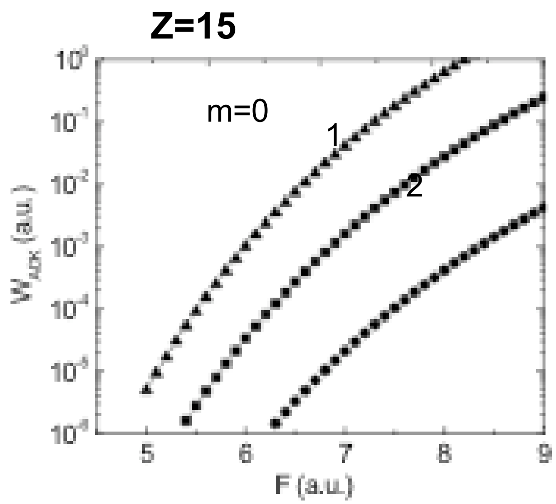


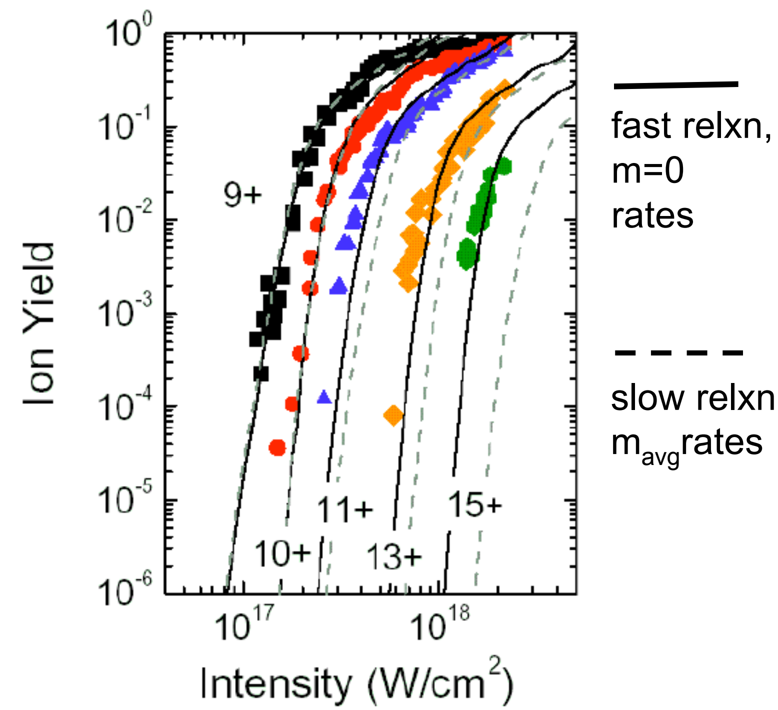


Hoshina et al. JCP 118, 6212 (2003)

No alignment observed in ultrastrong laser-field ionization with ion yield methods

- Relativistic intensity 10^{16}- 10^{18} W/cm^2, 40 fs
- Applicability of single active electron model
- Core relaxation between successive ionization steps





Gubbini et al, PRL 053602 (2005)

Impulsive alignment of molecules

Theory:

• Formation of ground state rotational wavepacket (coherent superposition of rotational eigenstates) through Raman excitation with short laser pulse.

• Subsequent time evolution gives aligned molecular states at well-defined time delays.

Seideman PRL **83**, 4971 (1999)

Ortigoso et al JCP **110**, 3870 (1999)

Experiment:

• Rosca-Pruna & Vrakking PRL 87, 153902 (2001).
Ti:sapphire pump 1-10 ps, probe 100 fs, 1.2 mJ. 50 Hz
Align I_2 in Ne

MEDI = multielectron dissociative ionization

Grand Challenge I

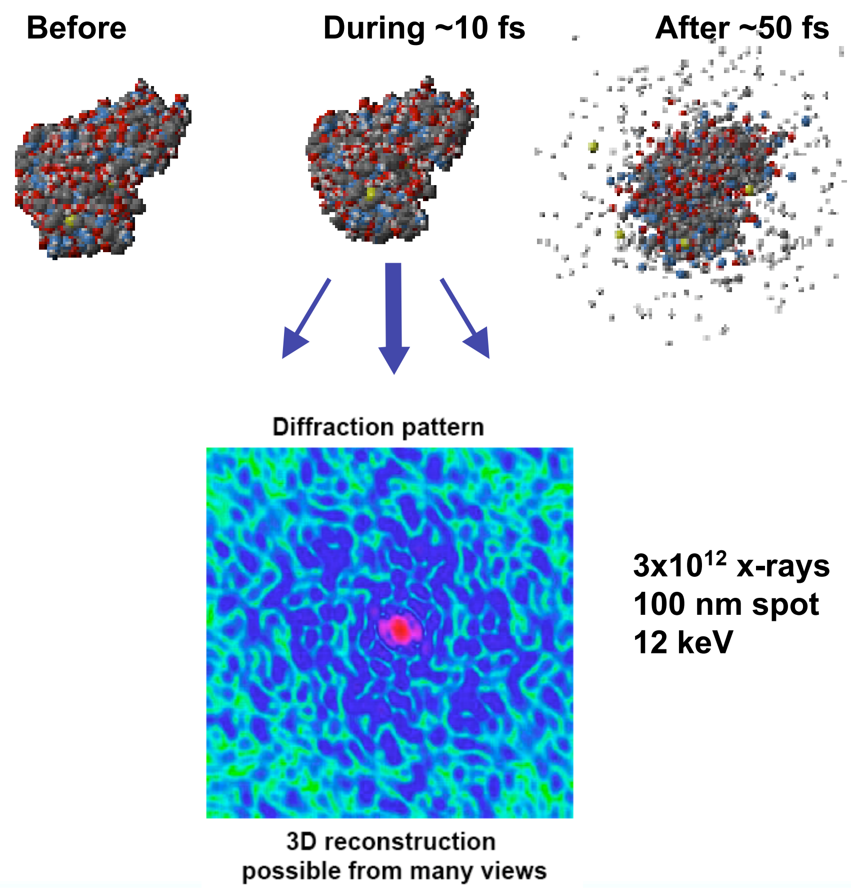
Biomolecule structure at atomic resolution w/o crystallization

Neutze, Wouts, van der Spoel, Weckert, Hajdu   Nature 406, 752 (2000)

- An important challenge is structure determination of ~10^6 human proteins which cannot be crystallized.

- Capture a diffraction pattern from a single molecule prior to Coulomb explosion.

- Unresolved issues -
  Coulomb explosion timescale
  Behavior at ~10^{22} W/cm^2 - 1Å



3×10^{12} x-rays
100 nm spot
12 keV