Atomic and molecular physics using ultrafast x-rays

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Ultrafast X-ray Summer School
DESY Hamburg
23 Jun 2011

Outline

- Why are we here? What is so exciting about ultrafast x-rays in atomic and molecular physics?
- Review some basic x-ray processes in atoms
- Extension to the strong-field regime for x-rays
  - Optical-control of x-ray processes
  - X-ray induced processes
- Quiz

Some General References:

Ultrafast "optical" lasers and strong field studies

- "Intense few cycle laser fields: frontiers of nonlinear optics"
  T. Brabec & F. Krausz, Rev Mod Phys **72**, 545 (2000)

- "Attosecond physics"
  F. Krausz & M. Ivanov, Rev Mod Phys **81**, 163 (2009)

Basic concepts of x-ray atom interactions

- "Concepts in x-ray Physics"
  R. Santra J Phys B 42, 023001 (2009)

- Electron Spectrometry of Atoms using Synchrotron Radiation
  V. Schmidt (Cambridge University Press, 1997)

SACLA X-ray free electron laser sets new record

June 13, 2011

RIKEN and the Japan Synchrotron Radiation Research Institute (JASRI) have successfully produced a beam of X-ray laser light with a wavelength of 1.2 Angstroms, the shortest ever measured. This record-breaking light was created using SACLA, a cutting-edge X-ray Free Electron Laser (XFEL) facility unveiled by RIKEN in February 2011 in Harima, Japan. SACLA (SPring-8 Angstrom Compact free electron LAser) opens a window into the structure of atoms and molecules at a level of detail never seen before.

Compare the evolution of high intensity optical and x-ray sources

Hign-intensity at optical wavelengths

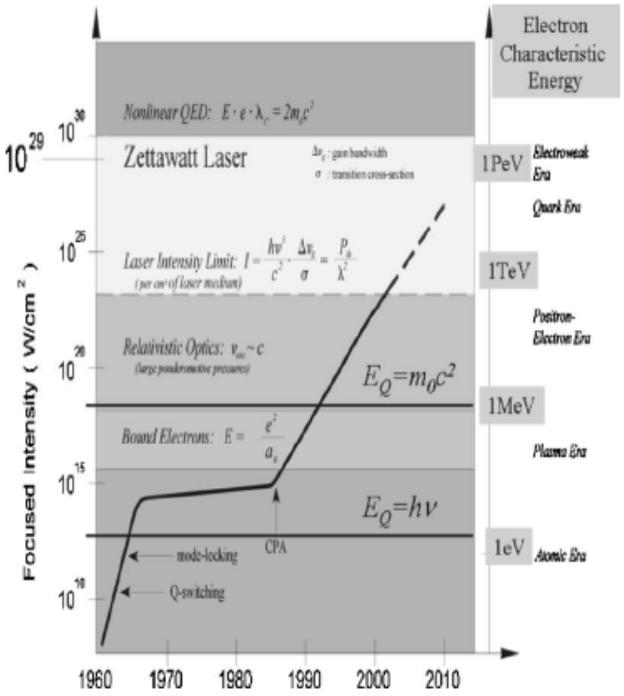
- high harmonic generation
- tabletop coherent x-ray radiation
- attosecond pulses

High-intensity at x-ray wavelengths

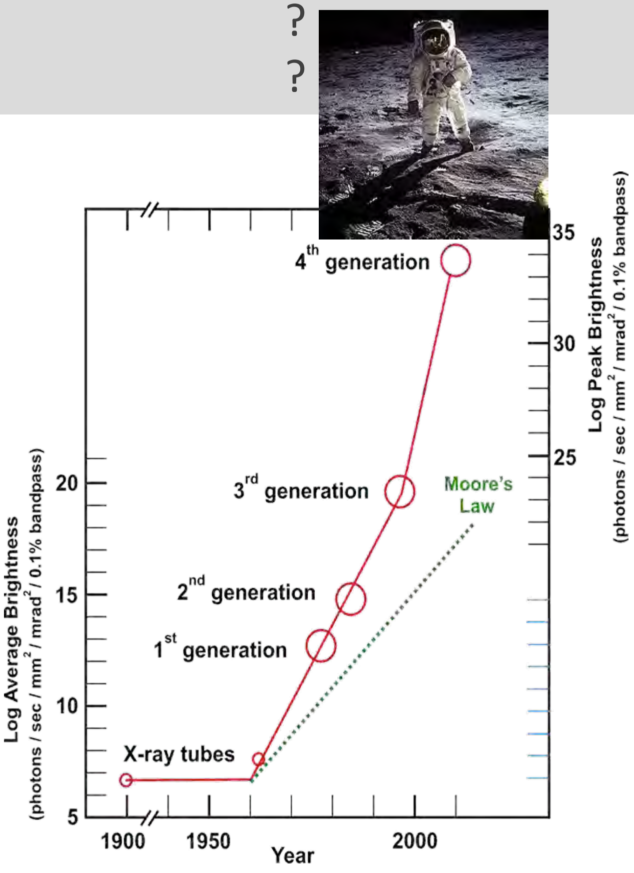
?

?

?



G. Mourou RMP 2006



D. Moncton, George Brown

Contrast optical and x-ray interactions at high intensity

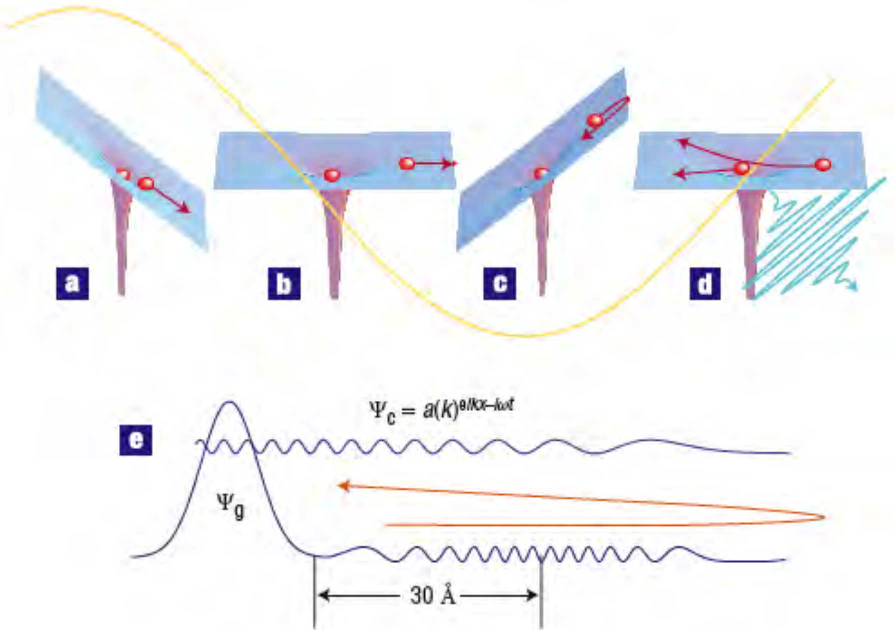
At long wavelengths - laser-driven electron dynamics is dominant
… not so at short wavelengths

electron ponderomotive energy (au)

$$U_p = I/4\omega^2$$

displacement

$$\alpha = E/\omega^2$$



Graphic from Corkum & Krausz
Nature Physics (2007)

Ti:sapphire laser (1.55 eV) PW/cm^2
$U_p \sim 60$ eV & $\alpha \sim 50$ au

LCLS (800 eV) 100 PW/cm^2
$U_p \sim 25$ meV & $\alpha \sim 0.003$ au

Parameters – intense optical lasers vs x-fel

Ti: sapphire

photon energy:	1.5 eV
number of photons:	5 x 10^{15}/shot
pulse energy:	1 mJ
pulse duration:	30 fs
focused spot size:	1 µm
flux:	5 x10^{35} cm^{-2} s^{-1}
intensity:	10^{17} W/cm^2

period:	2.7 fs
number of cycles:	10
ponderomotive energy:	6000 eV
displacement:	1000 au

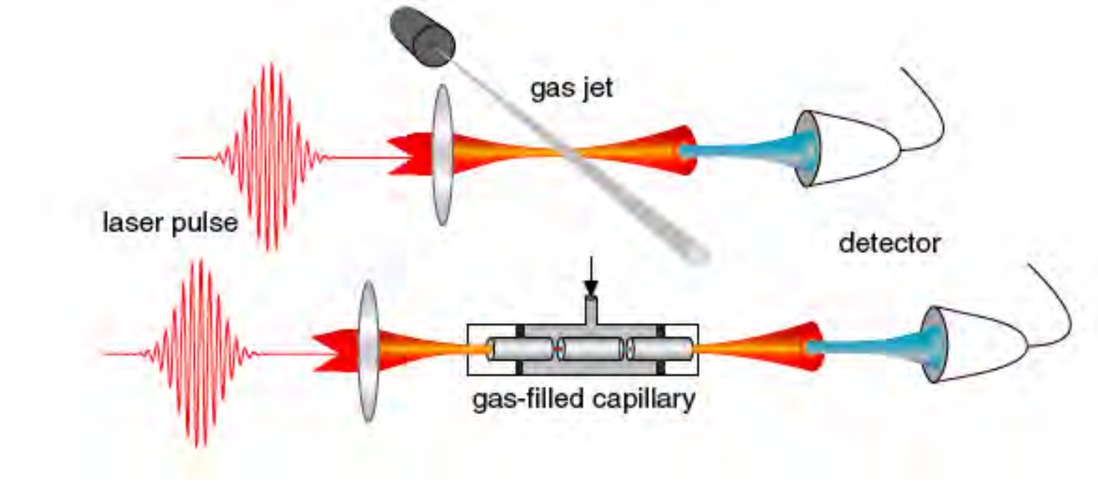
LCLS

photon energy:	800 eV
number of photons:	10^{13}/shot
pulse energy:	1 mJ
pulse duration:	100 fs
focused spot size:	1 µm
flux:	10^{33} cm^{-2} s^{-1}
intensity:	10^{17} W/cm^2

period:	2 as
number of cycles:	40,000
ponderomotive energy:	25 meV
displacement:	0.003 au

Use short pulse optical lasers for extreme nonlinear optics, i.e. generation of high harmonics

High Harmonic Generation: a tabletop ultrafast x-ray source

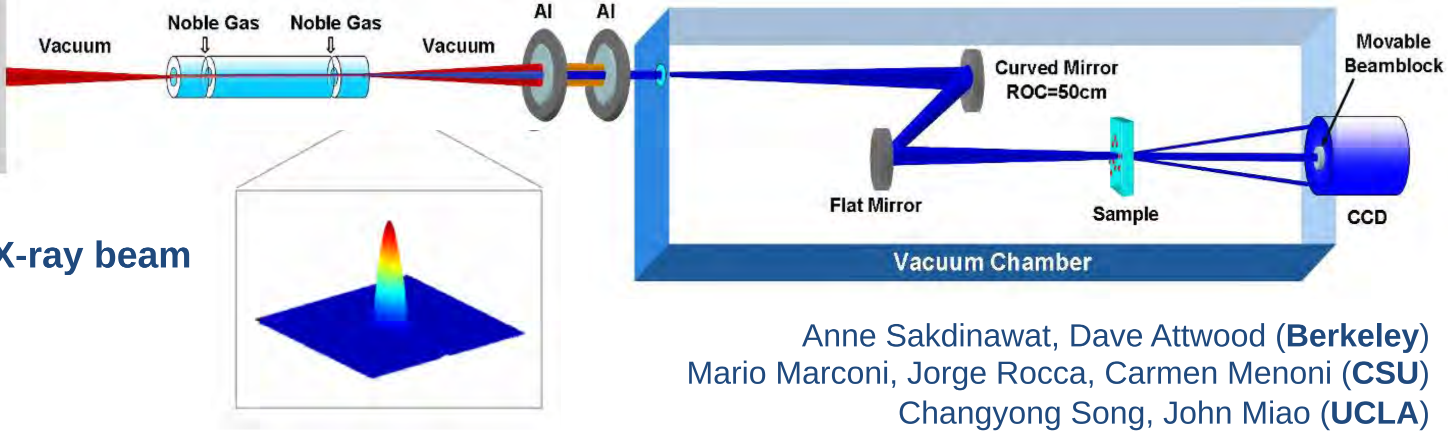


Coherent, collimated, ultrafast (down to attoseconds), tabletop
But - typical conversion efficiency from Ti:sapphire 10^{-5}/harmonic
Frontiers – shorter, more controlled pulses, shorter wavelengths

Reviews: T Pfeifer, C Spielmann, G Gerber, Rep Prog Phys (2006)
P Agostini & L DiMauro, Rep Prog Phys (2004)

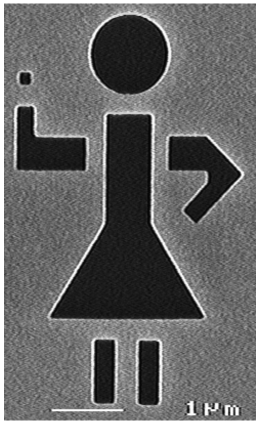
Coherent Diffraction Imaging using HHG beams resolution ≈ 90nm

High harmonic source (30nm)

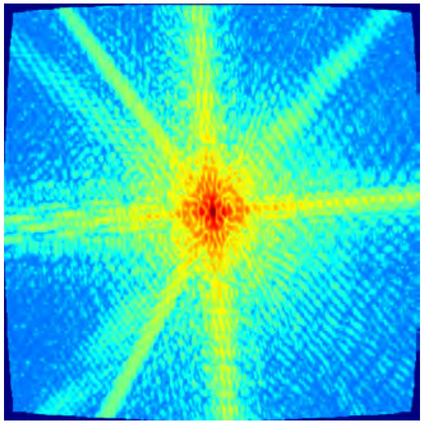


X-ray beam

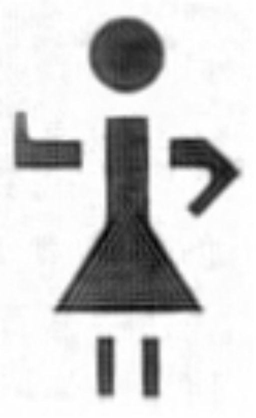
Anne Sakdinawat, Dave Attwood (**Berkeley**)
Mario Marconi, Jorge Rocca, Carmen Menoni (**CSU**)
Changyong Song, John Miao (**UCLA**)
Richard Sandberg, Daisy Raymondson, MM, HK (**JILA**)



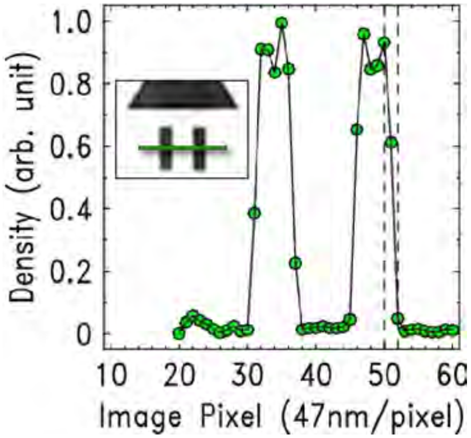
1 μm
sample (SEM image)



HHG diffraction pattern



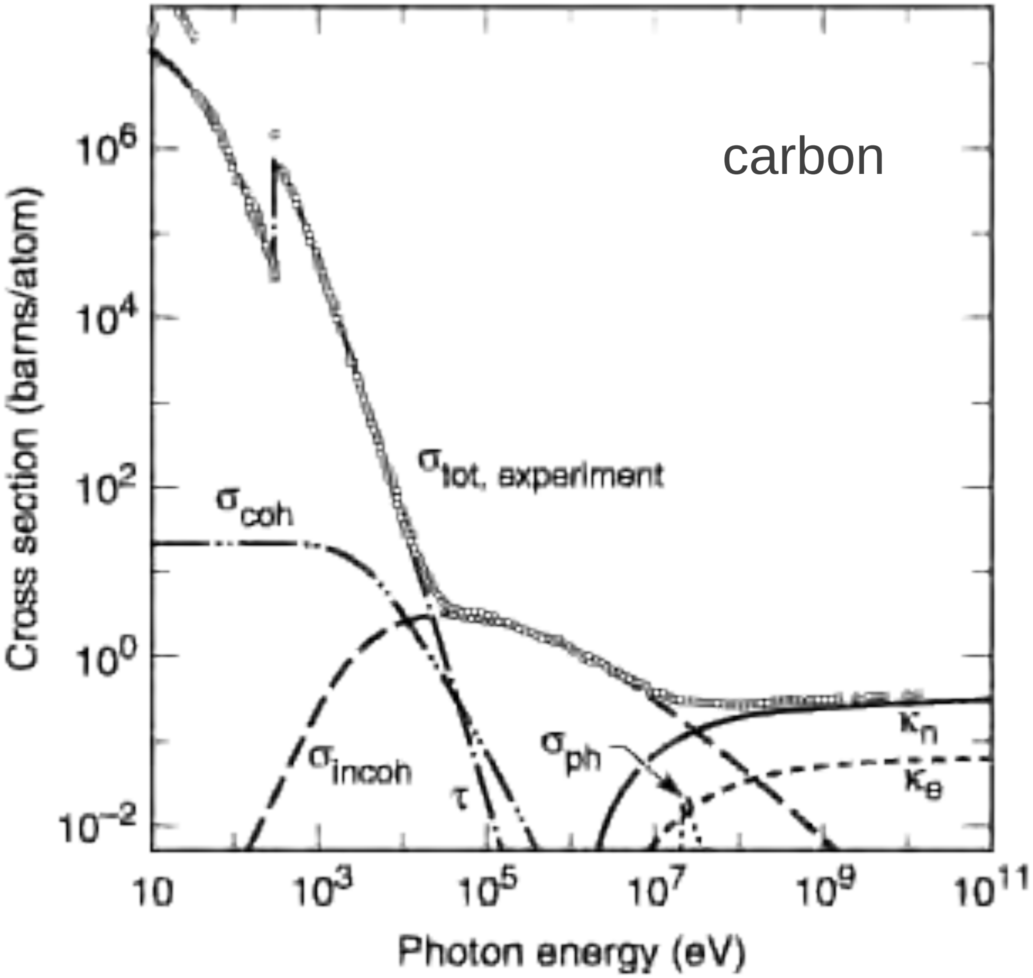
reconstruction



R. Sandberg et al, PNAS ***105****, 24 (2008); Nature Photonics* ***2****, 64 (2008)*

Basic x-ray processes in atoms

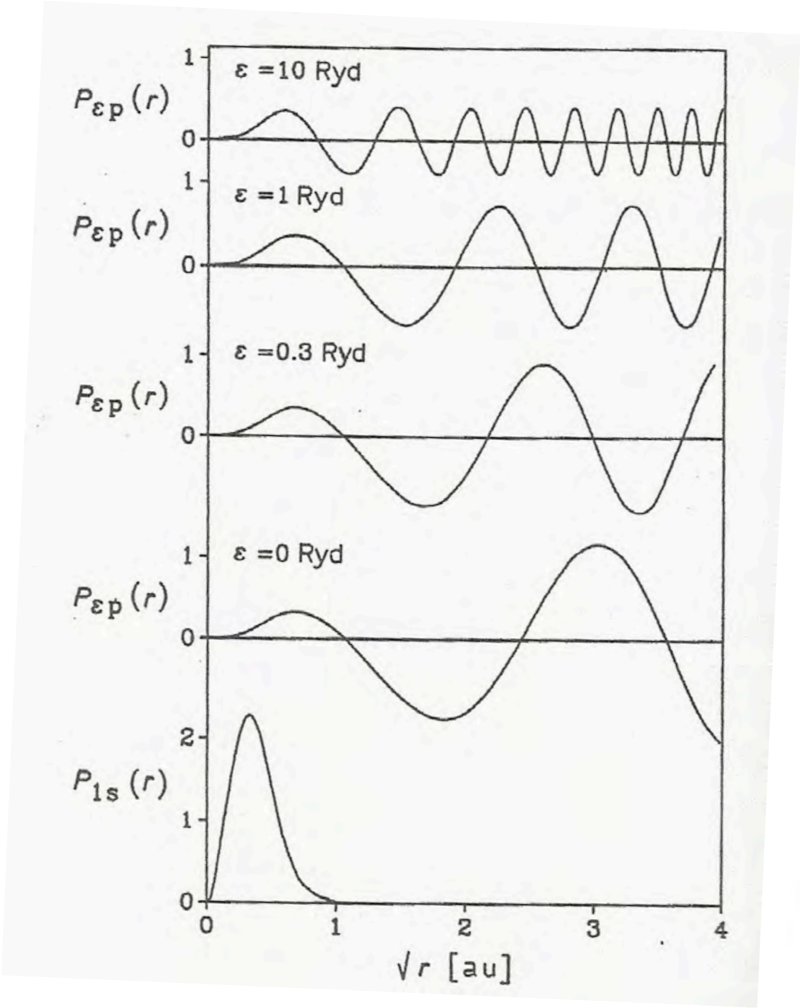
Fundamental x-ray atom interactions



Photoabsorption
Coherent/Rayleigh/Elastic Scattering
Incoherent/Compton Scattering
Pair Production
Photonuclear absorption

Construction of photoionization cross-sections

Radial functions in neon



$$\sigma_{1s} = \frac{8\pi^2}{3}\alpha E_{ph} R^2_{\epsilon p,1s}$$

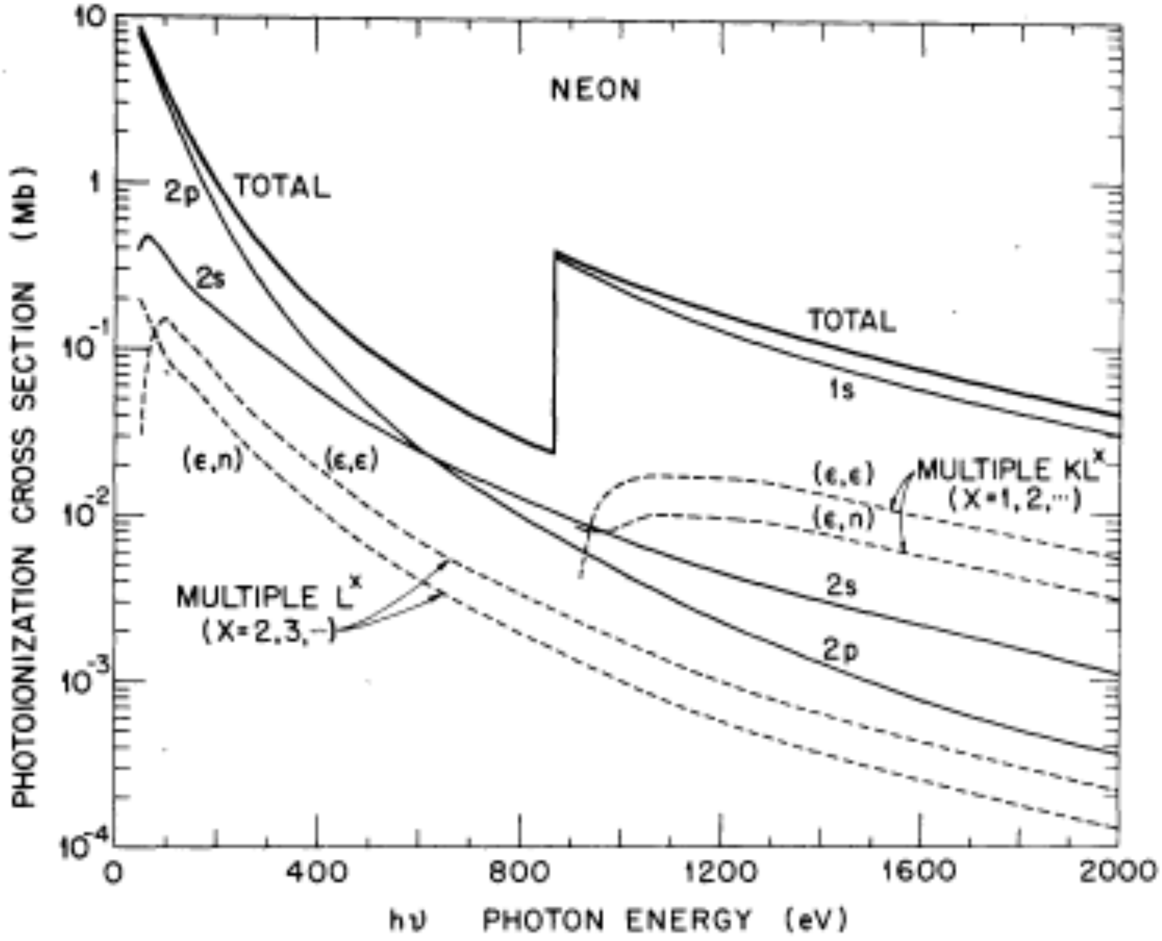
$$R_{\epsilon p,1s} = \langle R_{\epsilon p(r)} | r | R_{1s(r)} \rangle$$

$$= \int_0^\infty R_{\epsilon p(r)} r R_{1s(r)} r^2 dr$$

$$= \int_0^\infty P_{\epsilon p(r)} r P_{1s(r)} dr$$

$\sigma \propto$ overlap integral weighted by r

Dissection of the total photoabsorption cross section



$$E_{e,\,\mathrm{kin}}(\epsilon l) = h\nu - E_{nl}$$

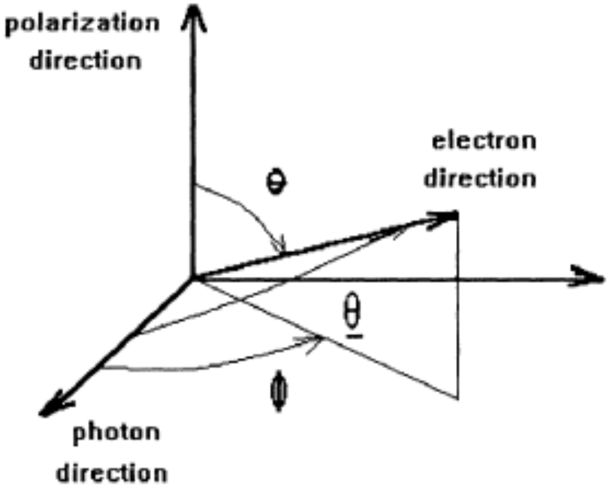
$$E_{e,\,\mathrm{kin}}(\epsilon l, n'l') = h\nu - E_{nl} - E_{n''l''\to n'l'}$$

$$E_{e,\,\mathrm{kin}}(\epsilon l, \epsilon' l') = h\nu - E_{nl} - E_{n''l''} - E^{*}_{e,\,\mathrm{kin}}(\epsilon' l', \epsilon l)$$

F. Wuilleumier & M. O. Krause PRA **10**, 240 (1974)

Photoelectron angular distributions

$$\frac{d\sigma}{d\Omega}(\vartheta) = \frac{\sigma}{4\pi}[1 + \beta P_2(\cos(\vartheta)]$$



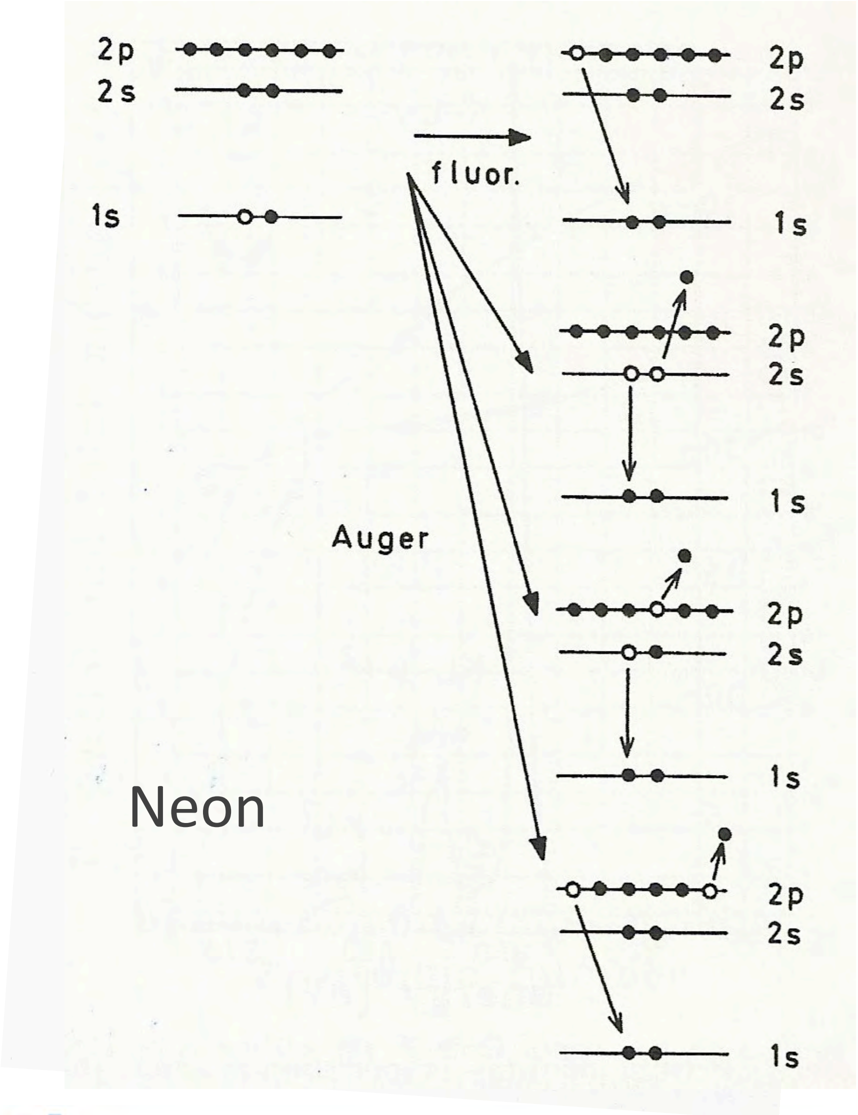
$$\beta_{1s} = 2$$

$$\beta_{2s} = 2$$

$$\beta_{2p} = \frac{2R^2_{\epsilon d,2p} - 4R_{\epsilon d,2p}R_{\epsilon s,2p}\cos(\Delta)}{R^2_{\epsilon s,2p} + 2R^2_{\epsilon d,2p}}$$

Angle independent measure of cross section at “magic” angle 54.7° where $P_2\cos(\theta) = 0$.

What’s after photoabsorption (1s hole creation)?



Neon

Radiative – fluorescence

Operator: dipole

Selection Rules

$$\Delta J = \pm 1, 0 \text{ , } J = 0 \rightarrow J = 0 \text{ forbidden}$$

Parity change

Non-radiative – Auger

Operator: Coulomb interaction

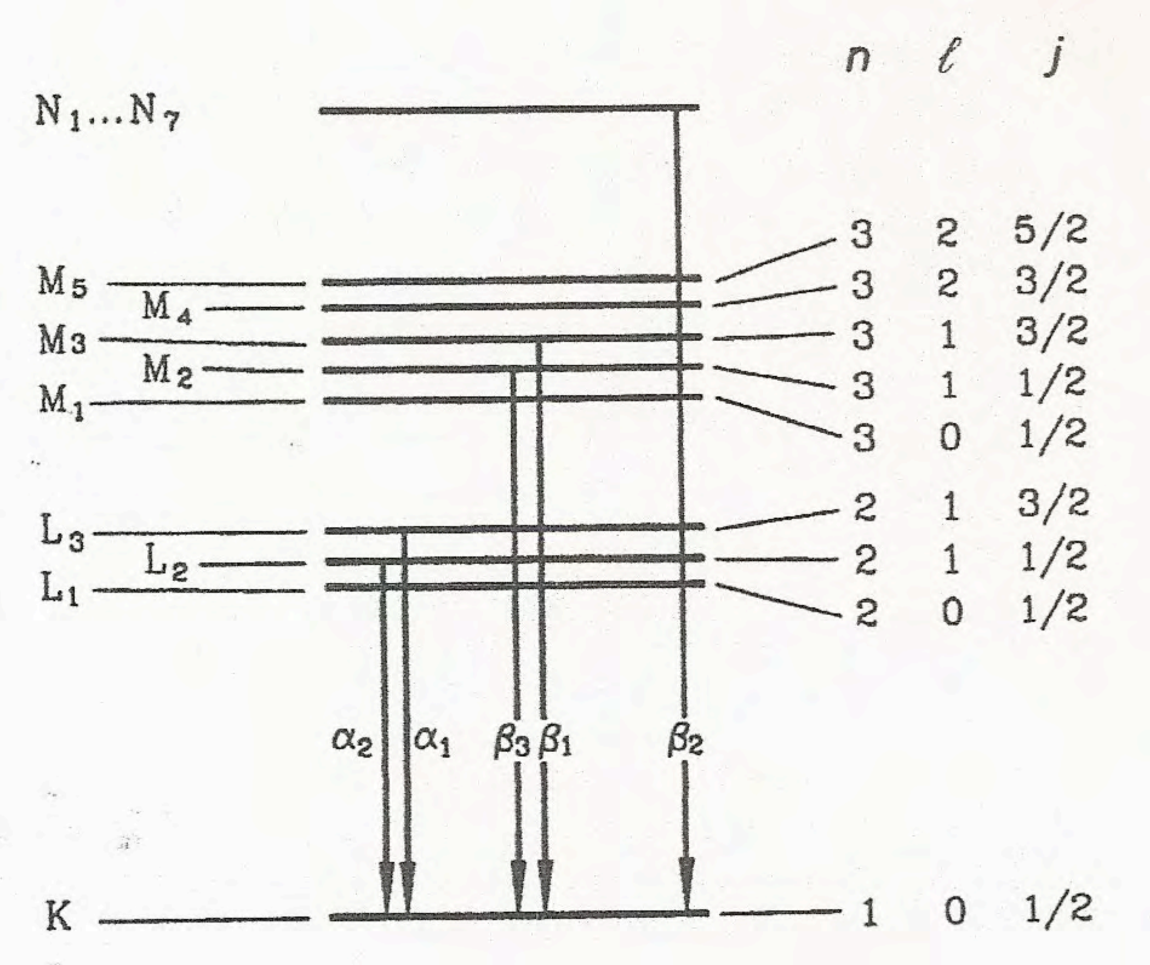
$$\mathrm{Op(Auger)} = \sum_{i<j} \frac{1}{r_{ij}}$$

Selection Rules

$$\Delta L = \Delta S = \Delta M_L = \Delta M_S = 0$$

No parity change

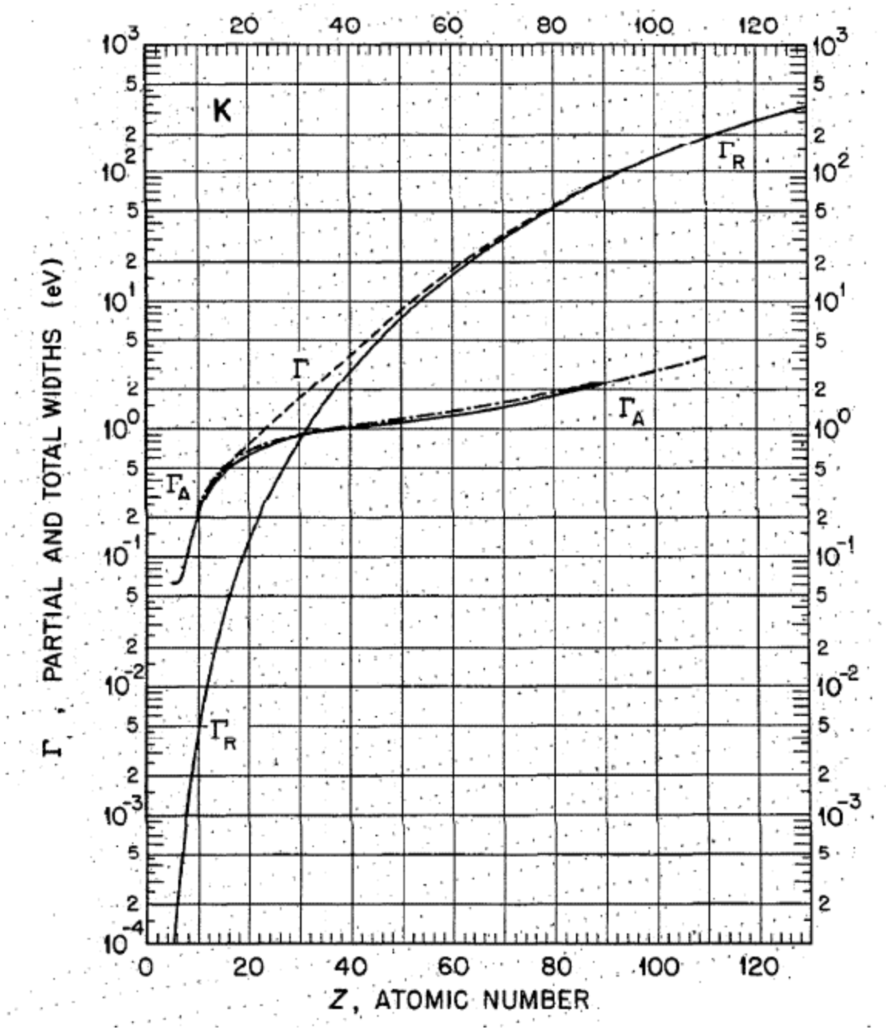
Nomenclature for inner-shell transitions



For example: Auger lines labelled K-$L_{2,3}L_{2,3}$, K-$L_{2,3}$,L_1, K-L_1L_1

Hollow atoms: KK-K$L_{2,3}L_{2,3}$…

Relative probability for radiative and Auger decay



M. O. Krause, JPCRD (1979)

$$\Gamma\tau = \hbar$$

$$\Gamma(1s) = \Gamma_R(1s) + \Gamma_A(1s)$$

$$(\hbar = 0.657eV\ fs)$$

Z-dependence

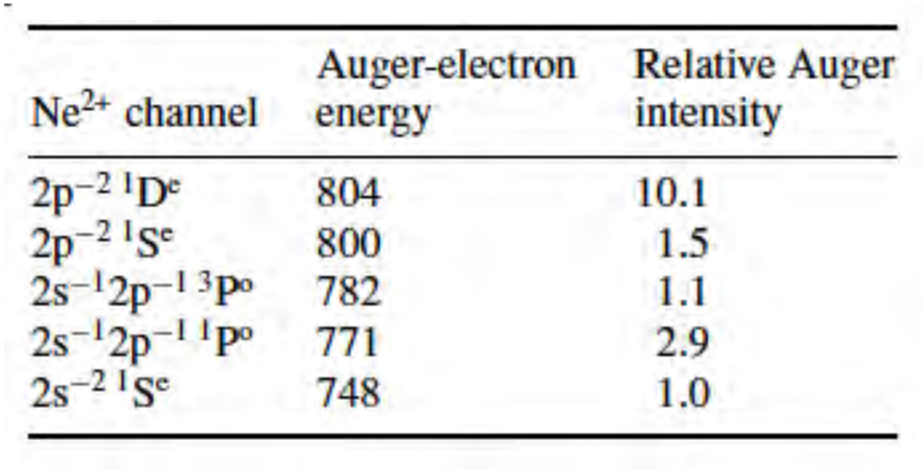
$$\gamma_R = \frac{4\omega_0^3}{3\hbar c^3}\frac{|\langle g, J||r||e, J'\rangle|^2}{2J'+1}$$

$$\omega_0 \propto Z^2, \langle r\rangle \propto 1/Z \implies \gamma_R \propto Z^4$$

Ab initio calculations of Auger rates

Neon K-LL transition

5 lines: Initial state = [1s $2s^2 2p^6$] = $^2S^e$

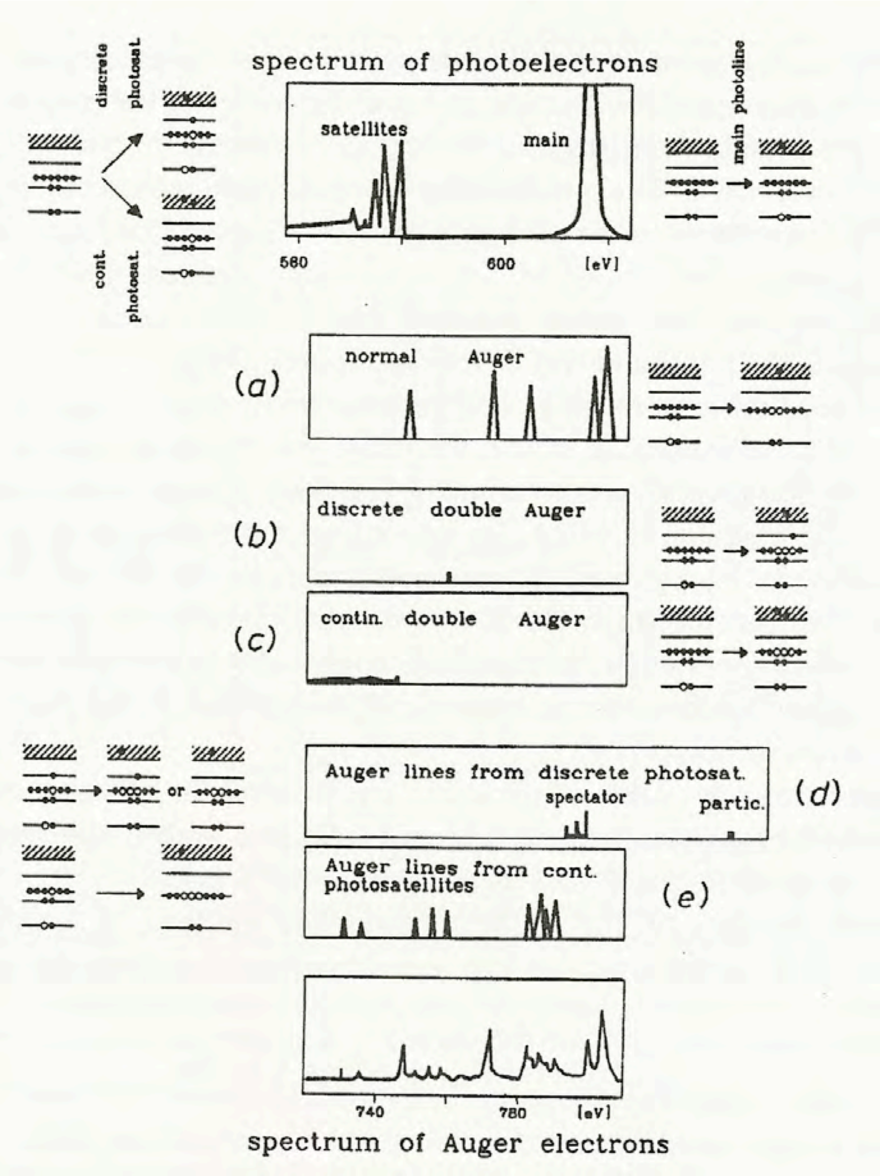


Ne^{2+} channel	Auger-electron energy	Relative Auger intensity
$2p^{-2}\ {}^1D^e$	804	10.1
$2p^{-2}\ {}^1S^e$	800	1.5
$2s^{-1}2p^{-1}\ {}^3P^o$	782	1.1
$2s^{-1}2p^{-1}\ {}^1P^o$	771	2.9
$2s^{-2}\ {}^1S^e$	748	1.0

H. Kelly, Phys Rev A **11**, 556 (1975) Hartree Fock + correlation
Ne neutral
C. Bhalla et al., Phys Rev A **8**, 649 (1973) Hartree-Fock-Slater (+ configuration mixing)
All charge states & configurations
M.H. Chen, Phys Rev A **44**, 239 (1991) Multiconfiguration Dirac Fock
[KK] energies and radiative & Auger transition rates vs Z

New toolkit: Sang-Kil Son & Robin Santra, Phys Rev A (2011) Hartree-Fock-Slater framework

Beyond the diagram lines: Example neon



From V. Schmidt

Categories of Auger lines
A: normal Auger from 1s photoionization

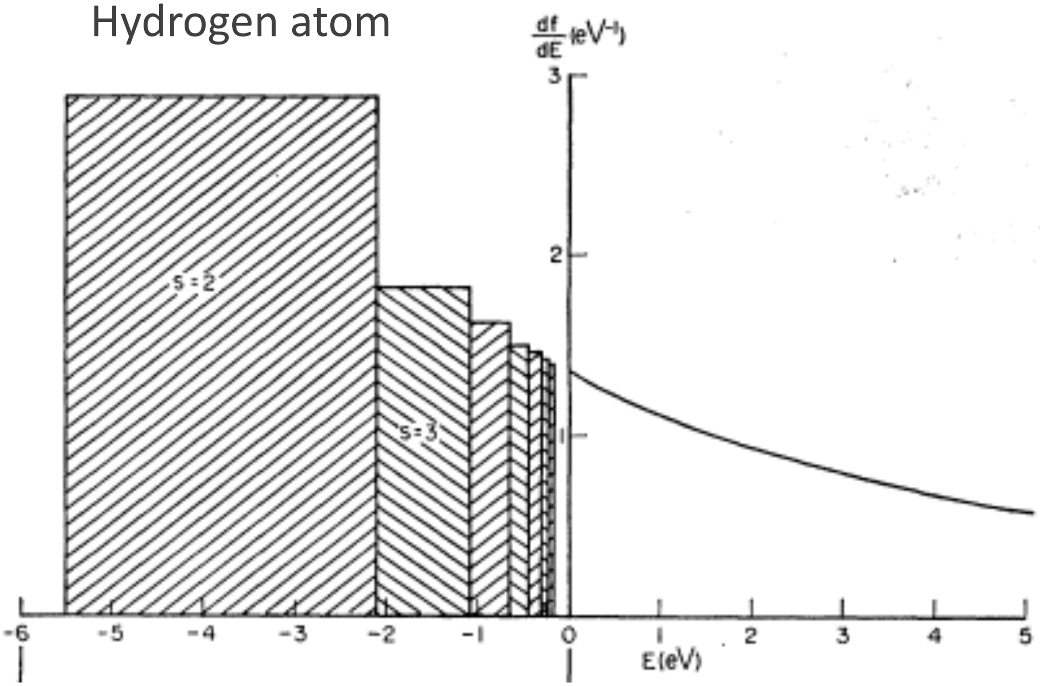
Bα & Bβ: satellite lines from 1s – nl excitations with spectator or participator transition

Cα & Cβ: KL* - LLL* arising from 1s, 2s- ∞, nl and 1s, 2p – ∞, nl two electron processes of ionization and excitation with subsequent Auger decay where excited electron is involved or spectating

D: KL – LLL Auger transitions from 1s,2s – ∞, ∞ and 1s, 2p – ∞, ∞ two electron processes with subsequent Auger decay

Distribution of absorption oscillator strength

Hydrogen atom



Fano & Cooper, RMP (1968)

Smooth transition: discrete to continuum

Discrete transitions:

Area = average oscillator strength

$$\bar{f}_{n'l',nl} = \frac{2}{3}\,\omega_{n'l',nl}\,\frac{l_{max}}{2l+1}\,|\langle n,l|r|n'l'\rangle|^2$$

$$\int_0^{\omega} \sigma(\omega)\,d\omega = 2\pi^2 r_0 c f_{ik}$$

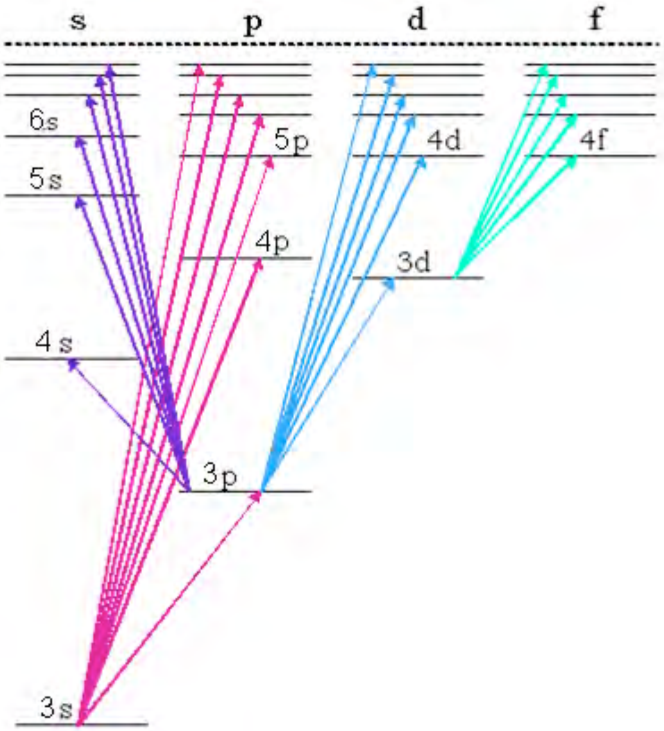
Continuum

$$\frac{d\bar{f}_{\epsilon'l',nl}}{dW} = \frac{2}{3}\,\omega_{\epsilon'l',nl}\,\frac{l_{max}}{2l+1}\,|\langle \epsilon'l'|r|nl\rangle|^2$$

$$\sigma = \frac{2\pi^2}{c}\cdot\frac{d\bar{f}_{\epsilon'l',nl}}{c\,dW}$$

An aside – bound-bound transitions & laser cooling

Grotrian Diagram for Sodium



Energy levels & oscillator strengths involved in laser cooling of sodium

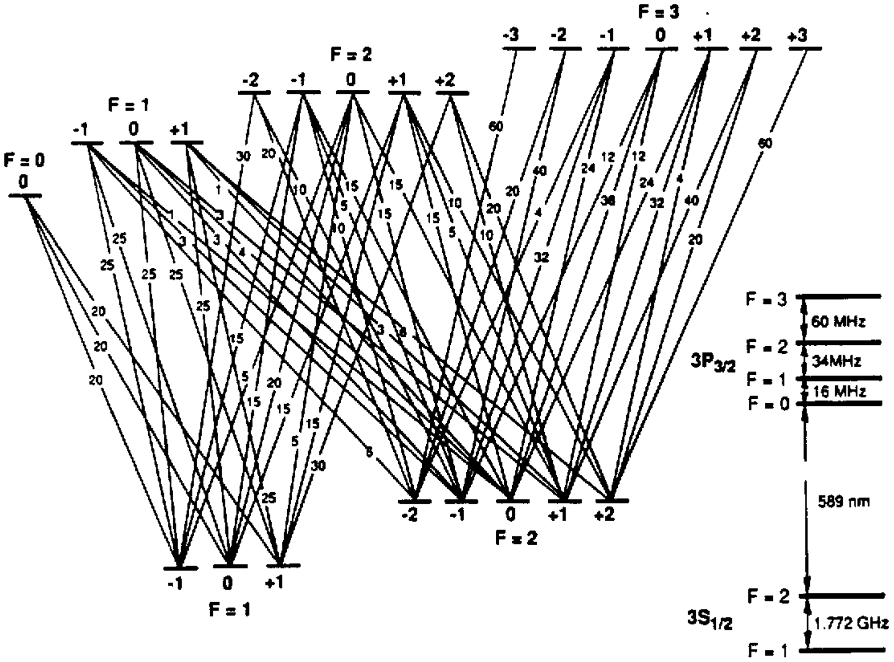


Fig. 2.2: Oscillator strengths and energy separations for the sodium $3S_{1/2} \rightarrow 3P_{3/2}$ hyperfine transitions.

Scattering factors & refractive index & susceptibility can be derived from absorption cross sections

$$f(E) = f_1(E) + i f_2(E)$$

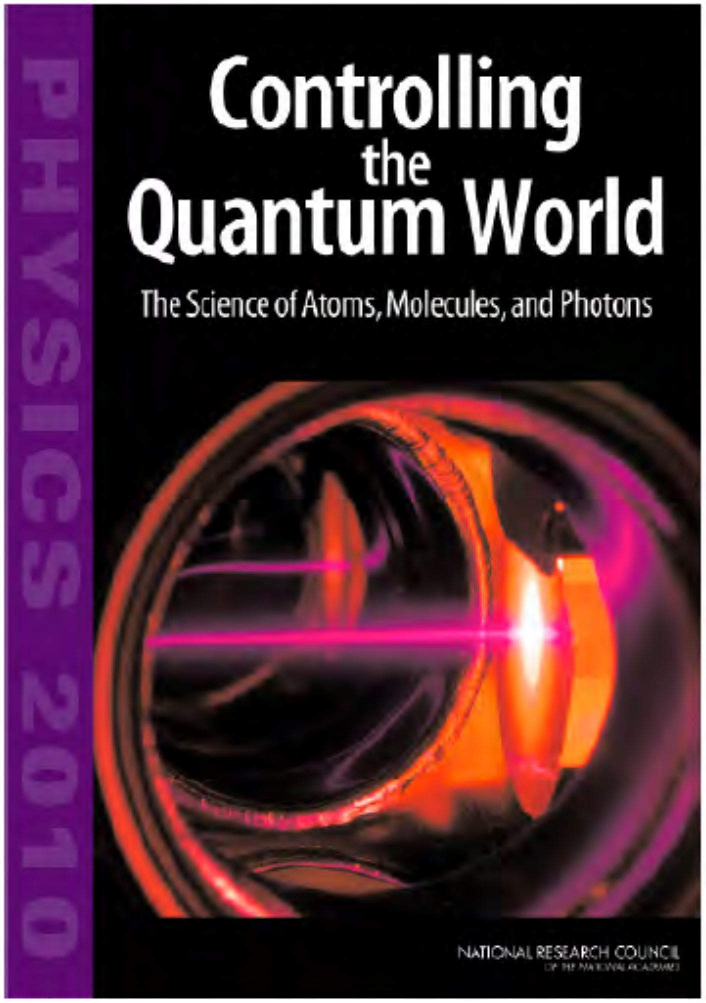
$$f_1(E) = Z + C \int_0^\infty \frac{\epsilon^2 \mu_a(\epsilon) d\epsilon}{E^2 - \epsilon^2}$$

$$f_2(E) = \frac{\pi}{2} C E \mu_a(E) \quad \text{where } C = 1/\pi r_0 hc$$

$$n = 1 - \delta - i\beta = \sqrt{1 + 4\pi\chi} \approx 1 + 2\pi\chi = 1 + 2\pi N \alpha$$

$$\delta = K f_1, \beta = K f_2 \text{ where } K = \frac{r_0 \lambda^2}{2\pi} \frac{N_A}{A} \rho$$

Optical control of x-ray processes



PHYSICS 2010

Controlling the Quantum World

The Science of Atoms, Molecules, and Photons

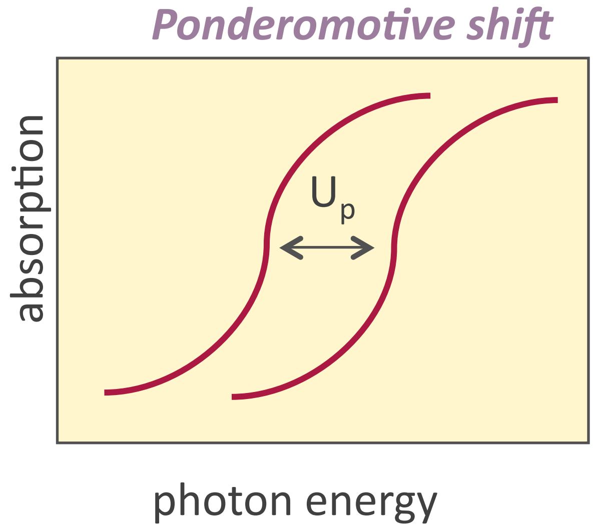
NATIONAL RESEARCH COUNCIL
OF THE NATIONAL ACADEMIES

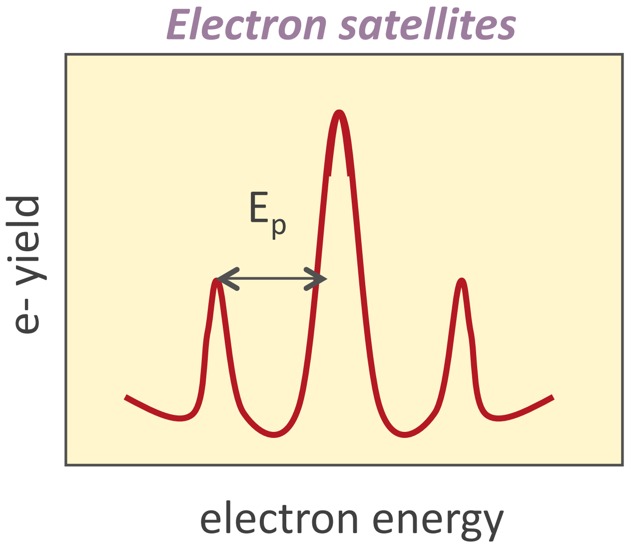
Modification of x-ray processes by strong optical fields

Motivations

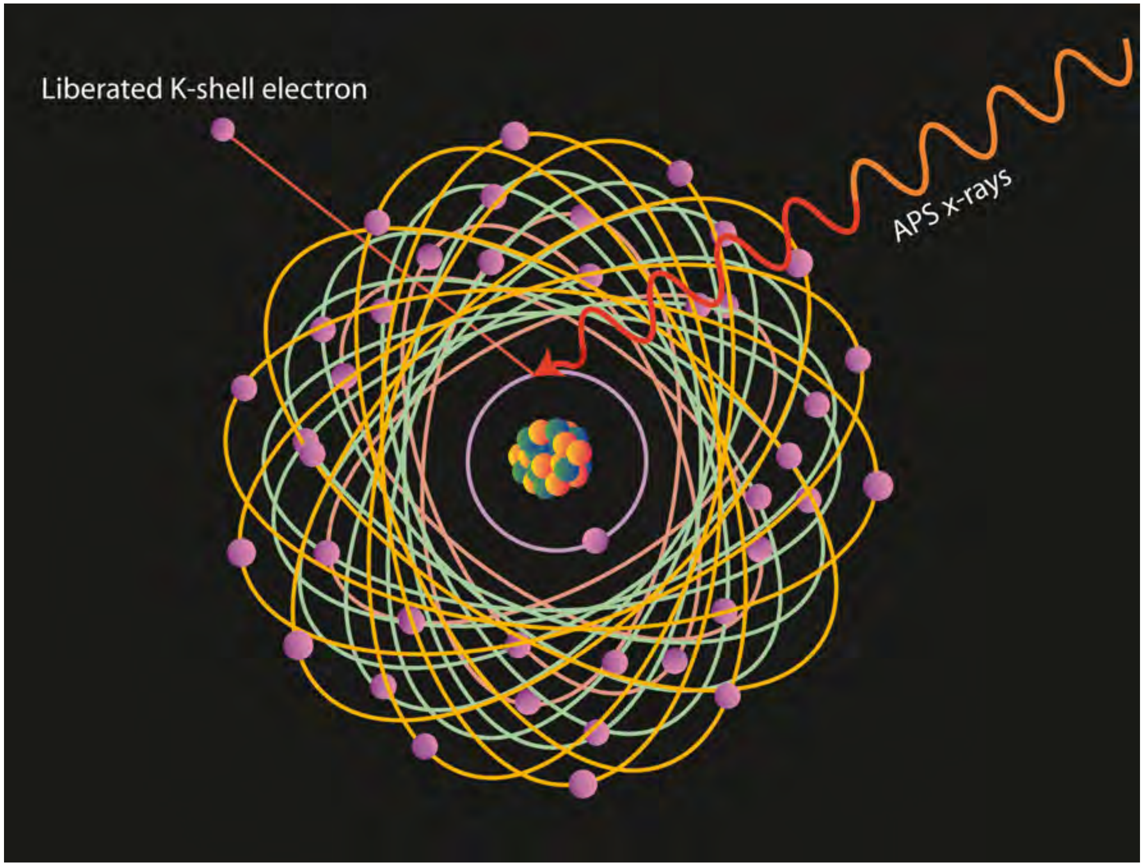
- Understand changes to x-ray processes in presence of strong laser fields
- Theoretical predictions

ponderomotive shift in threshold ⇒ absorption spectrum
free-free transitions in continuum ⇒ electron spectra



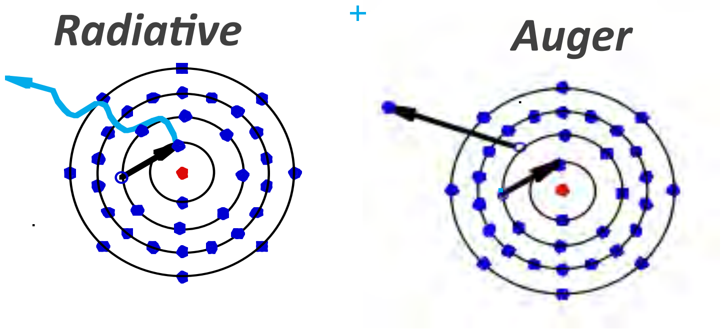


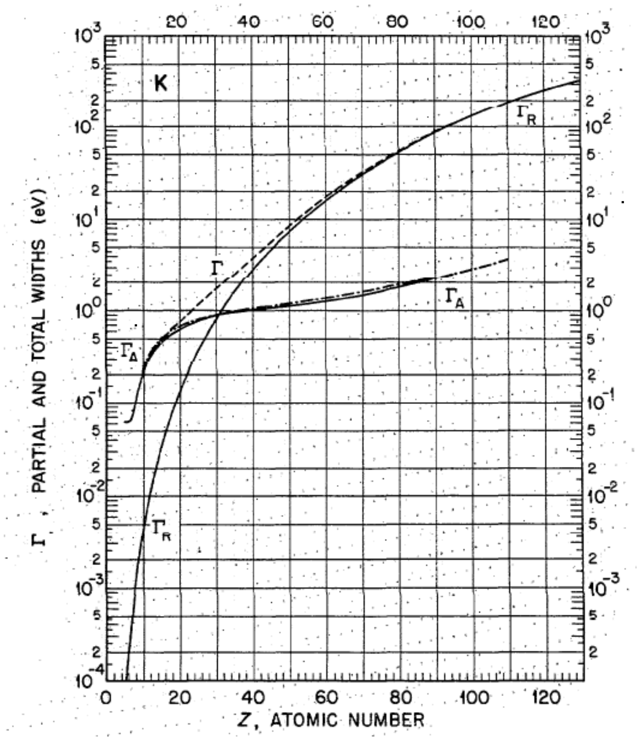
Can strong optical fields control x-ray processes?



- *Optical laser-induced processes must compete with inner-shell decay*
- *Typical inner shell decay width 1eV ⇒ 0.66 fs lifetime*

Intraatomic inner-shell decay vs laser-driven transitions





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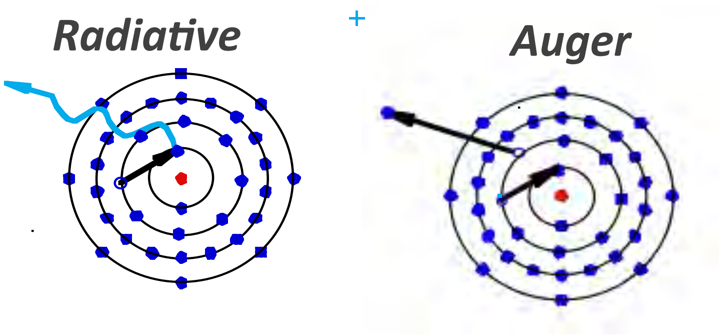
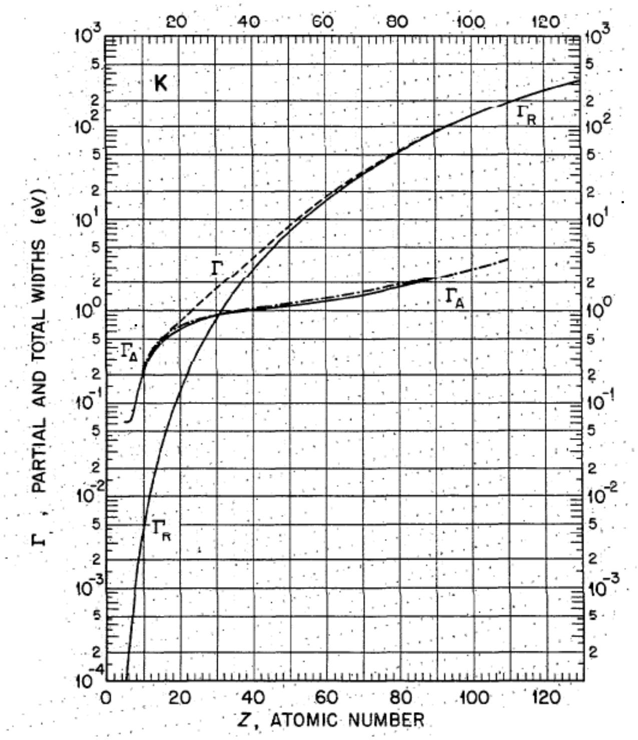
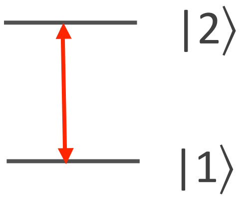




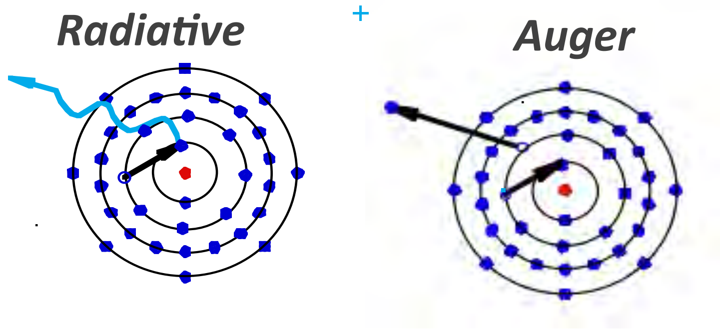
Rabi frequency

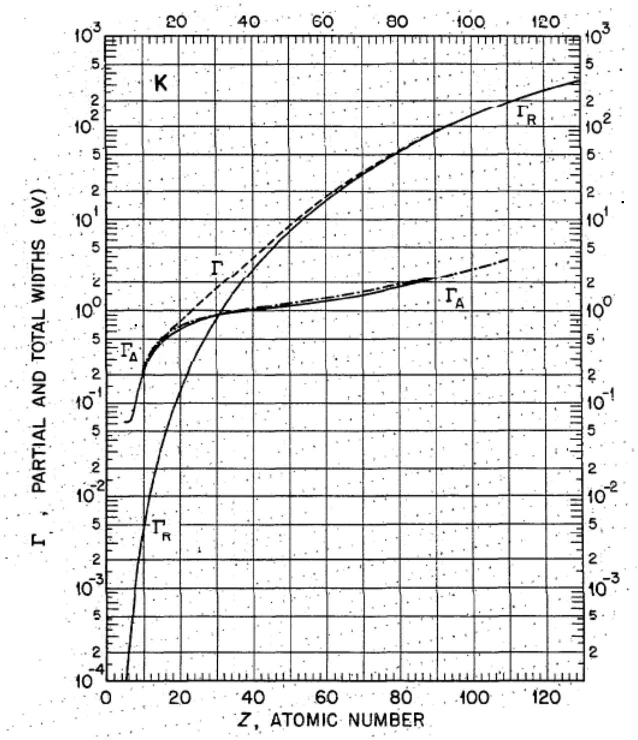
$$\Omega_{12} = \frac{\mu_{12}E}{\hbar}$$



$$\mu = er$$

Intraatomic inner-shell decay vs laser-driven transitions





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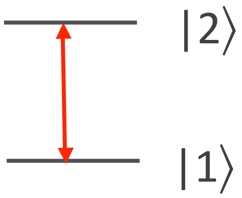


$\mu = er$

$\mu_{H1s\text{-}2p1/2} = 1.05\, ea_0$

Atomic Units : Hydrogen

Charge: electron charge = e

Length: Bohr radius a_0 = 0.529 Å

Velocity: Bohr velocity α c = 1/137 c

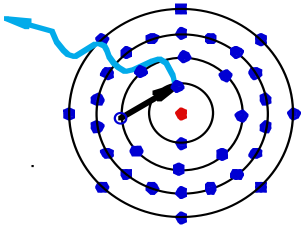
Time: length/velocity = 0.024 fs

Electric field: field at Bohr radius = 51 V/Å

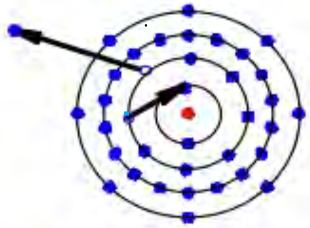
Intensity: 3.5 x 10^{16} W/cm^2

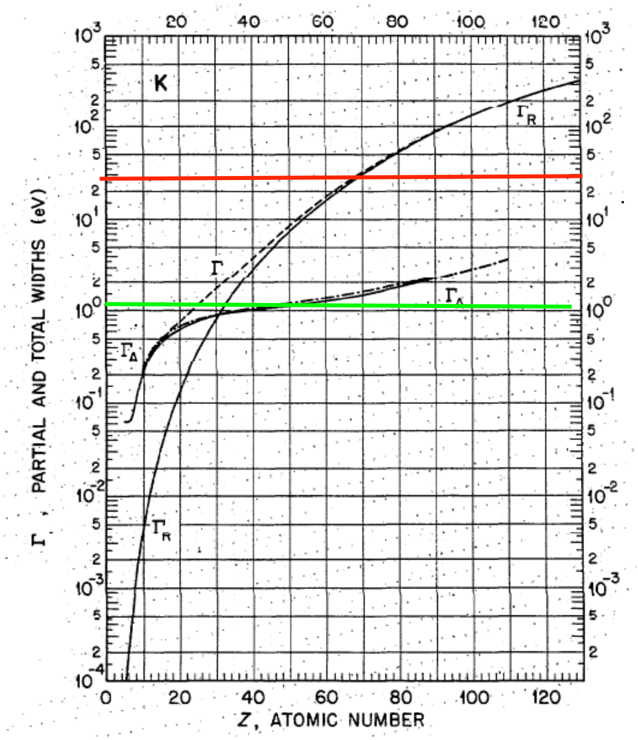
Intraatomic inner-shell decay vs laser-driven transitions

Radiative



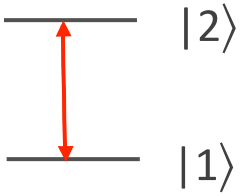
Auger





Rabi frequency

$$\Omega_{12} = \frac{\mu_{12}E}{\hbar}$$



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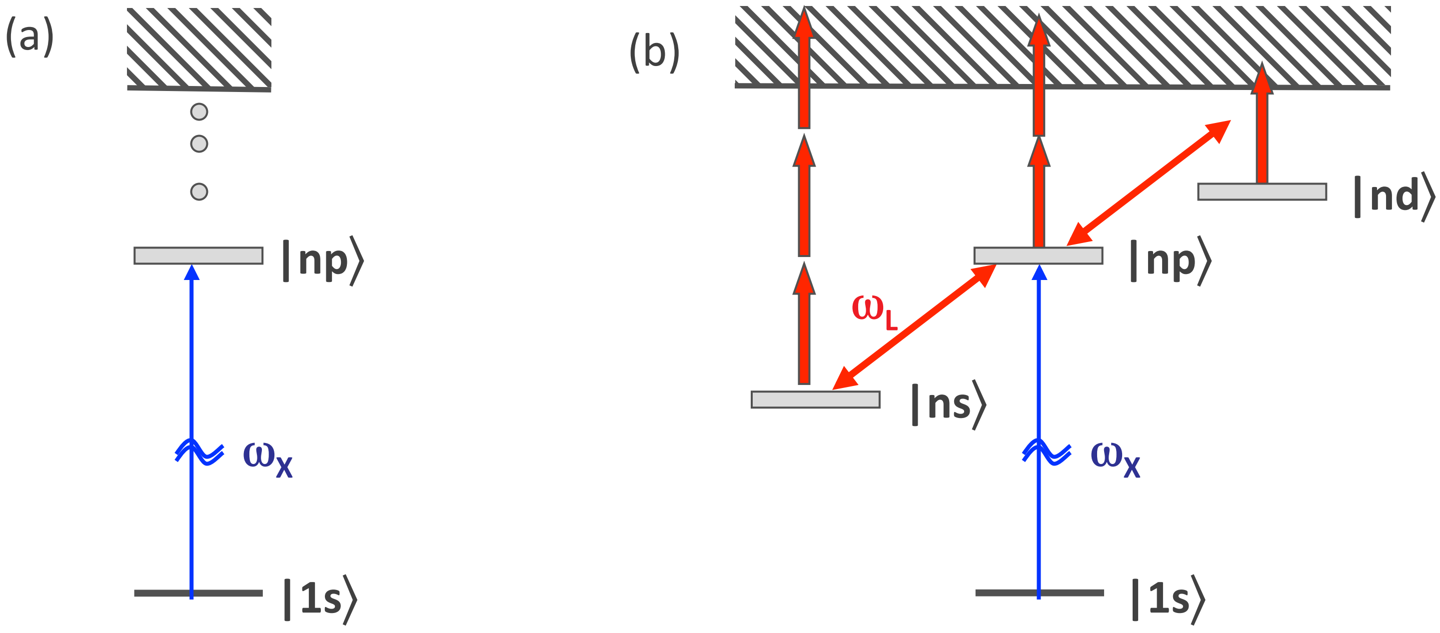
High intensity laser dressing of core-excited states



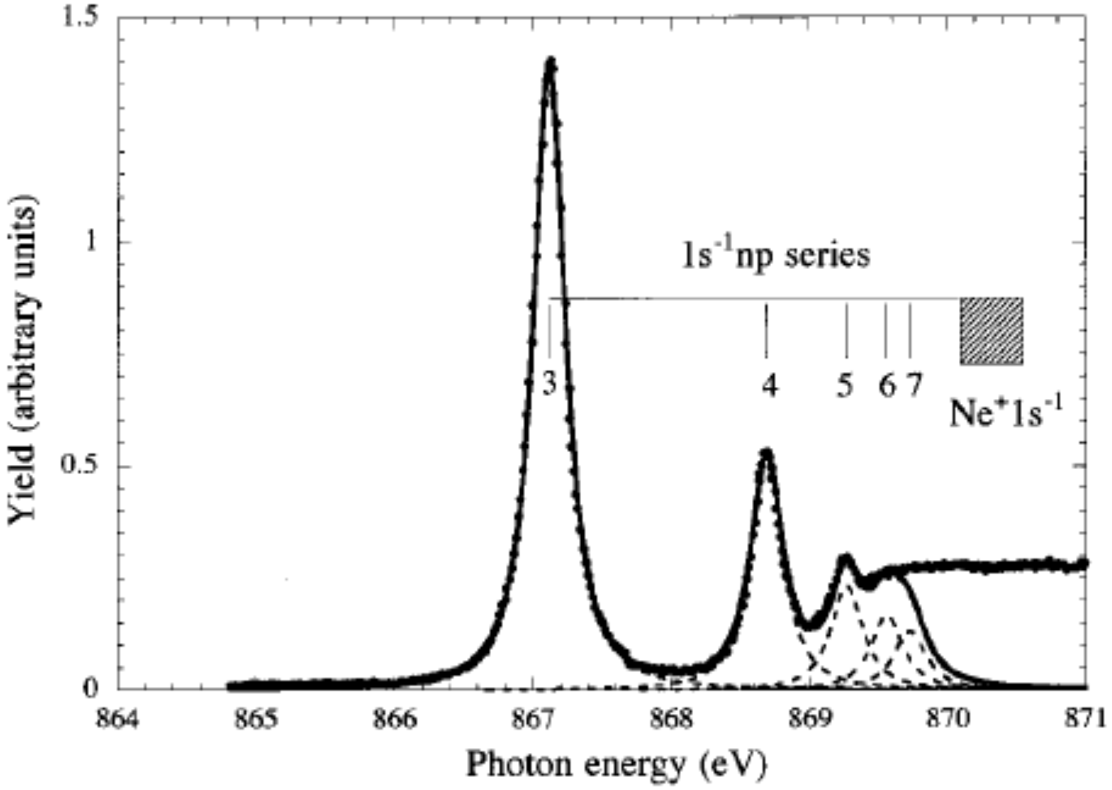
Laser-free

With strong-coupling laser

Control of x-ray absorption in neon

Ultrafast, reversible x-ray switch

Choice of neon



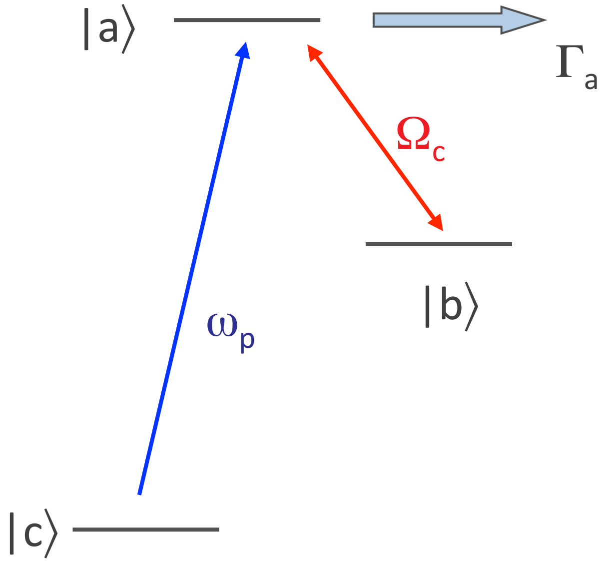
- Isolated resonance
- High IP = 21.6 eV

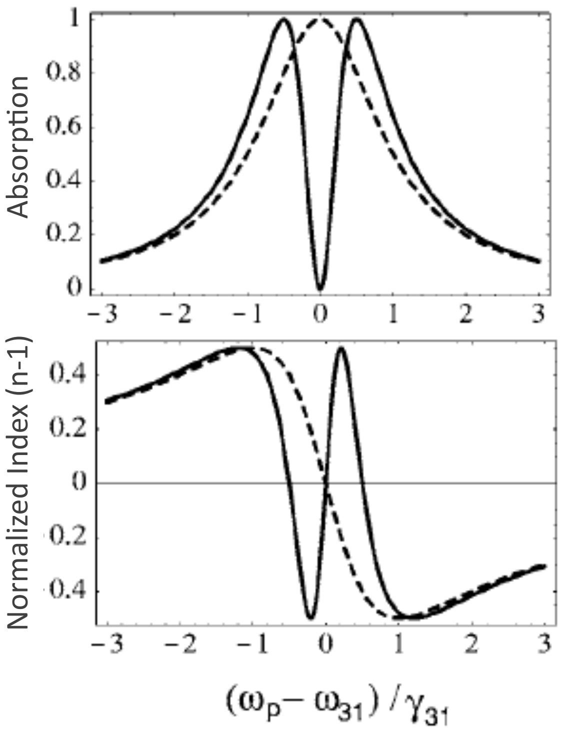
M. Coreno et al. PRA 99

Electromagnetically Induced Transparency (EIT)

One can make opaque resonant transitions transparent to laser radiation …

- S.E. Harris





$$v_{gr} = \frac{c}{n + \omega_p (dn/d\omega_p)}$$

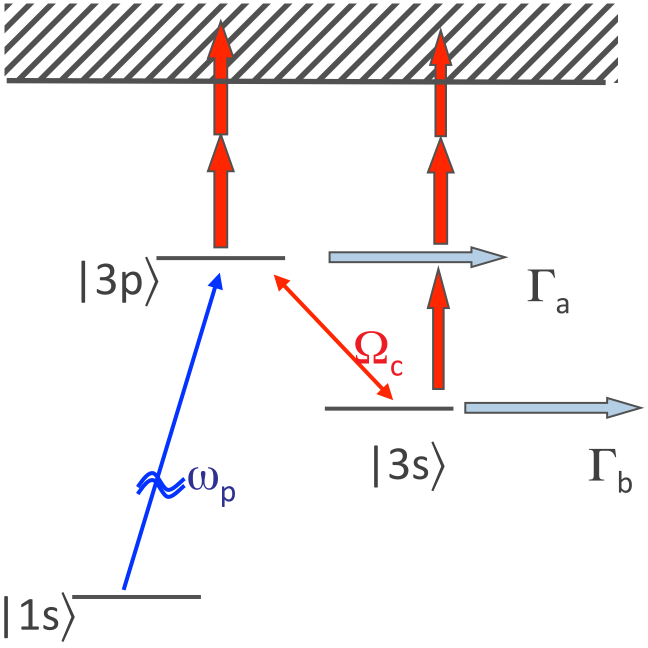
Light speed reduction to 17 metres per second in an ultracold atomic gas

Lene Vestergaard Hau*†, S. E. Harris‡, Zachary Dutton*† & Cyrus H. Behroozi*§

Nature **397**, 594 (1999)

S. E. Harris, Physics Today July 1997

Extend EIT concept to soft x-ray regime: Neon



Neon

Γ_{1s}~ 0.27 eV

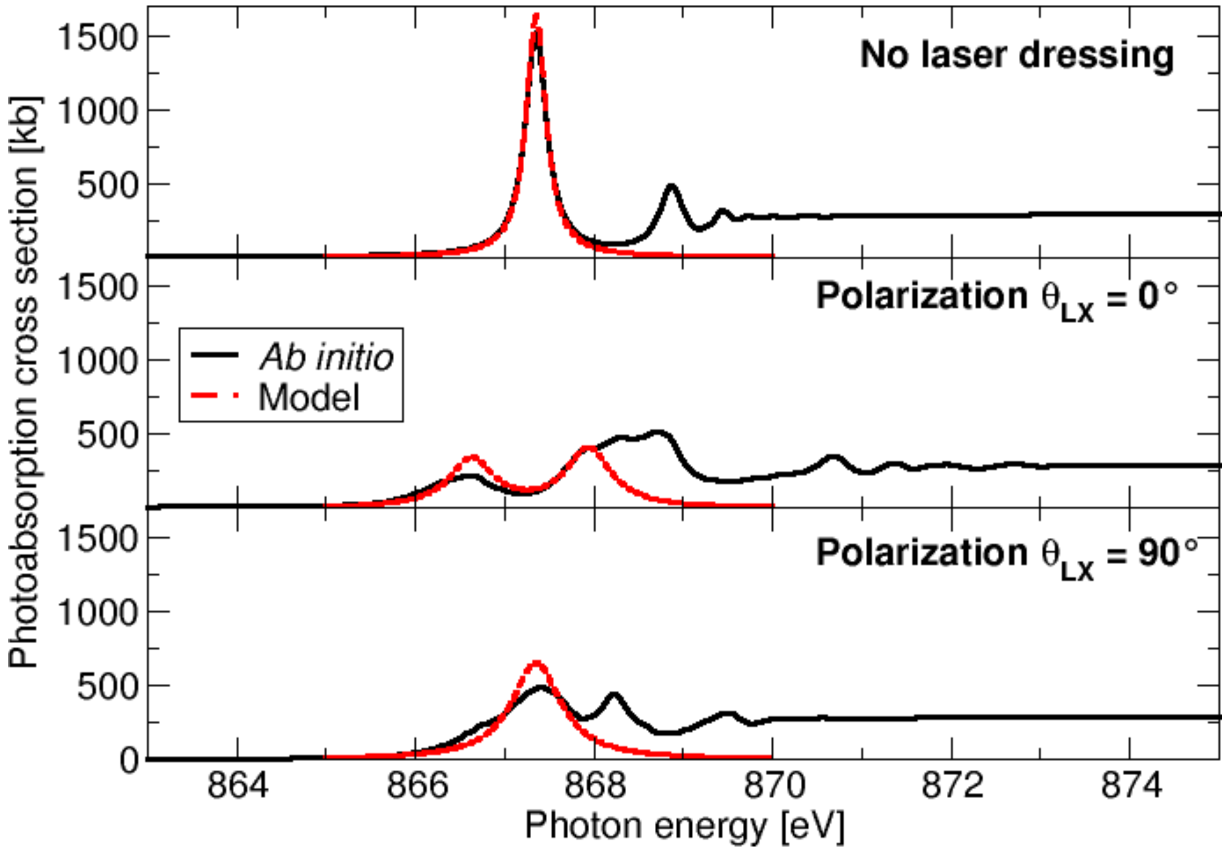
$\Delta_{3s\text{-}3p}$= 1.88 eV

Complexities

- Rapid Auger decay (2.4 fs)
- Laser induced ionization of core-excited states
- Existence of resonances at requisite coupling intensity
- $\tau_{Auger} \sim \tau_{Rabi} \sim \tau_{Laser}$

Buth, Santra, Young, PRL(2007)

Extend EIT concept to soft x-ray regime: Neon



Laser dressing intensity
10^{13} W/cm^2
800 nm(1.55 eV)

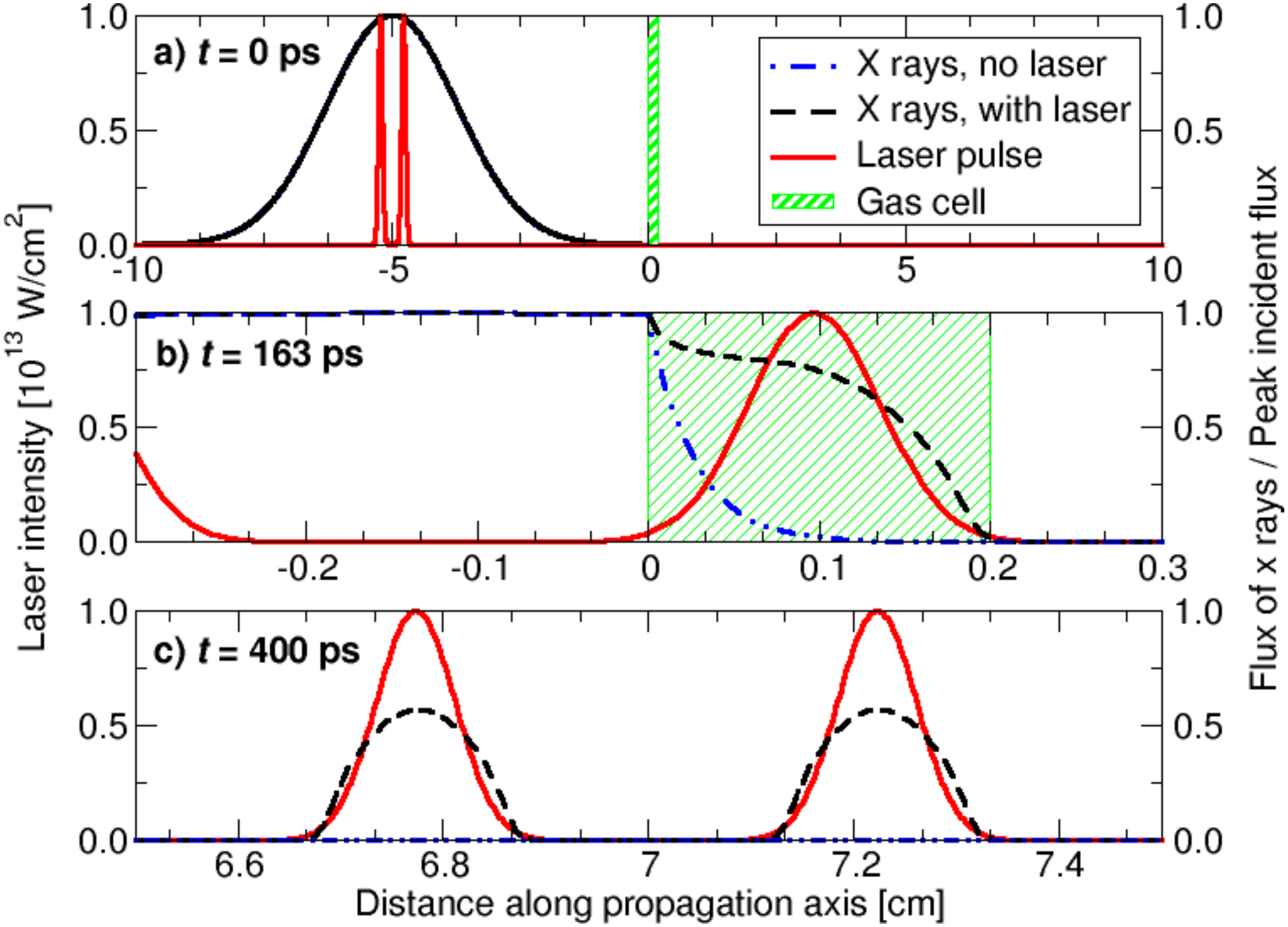
1 mJ/100fs/(300µm)^2

$$\frac{\sigma(\omega_X,0^\circ)}{\sigma_0} = \frac{4\Gamma_a^2\Delta_{LX}^2 + \Gamma_a\Gamma_b(\Omega_{ab}^2+\Gamma_a\Gamma_b)}{[\Omega_{ab}^2+\Gamma_a\Gamma_b-4\Delta_{LX}(\omega_{ac}-\omega_X)]^2+4[\Gamma_a\Delta_{LX}+\Gamma_b(\omega_{ac}-\omega_X)]^2}.$$

Buth et al PRL (2007)

Imprinting ultrafast laser pulse sequences on long x-ray pulses

1 atm Ne, 2-mm
10^{13} W/cm^2 @ 800 nm
$T_{No\ Laser}$ = 0.07%
T_{Laser} = 57%



Exptl demonstration: Controlling x-rays with light


Berkeley - ALS
10 - 2000 eV

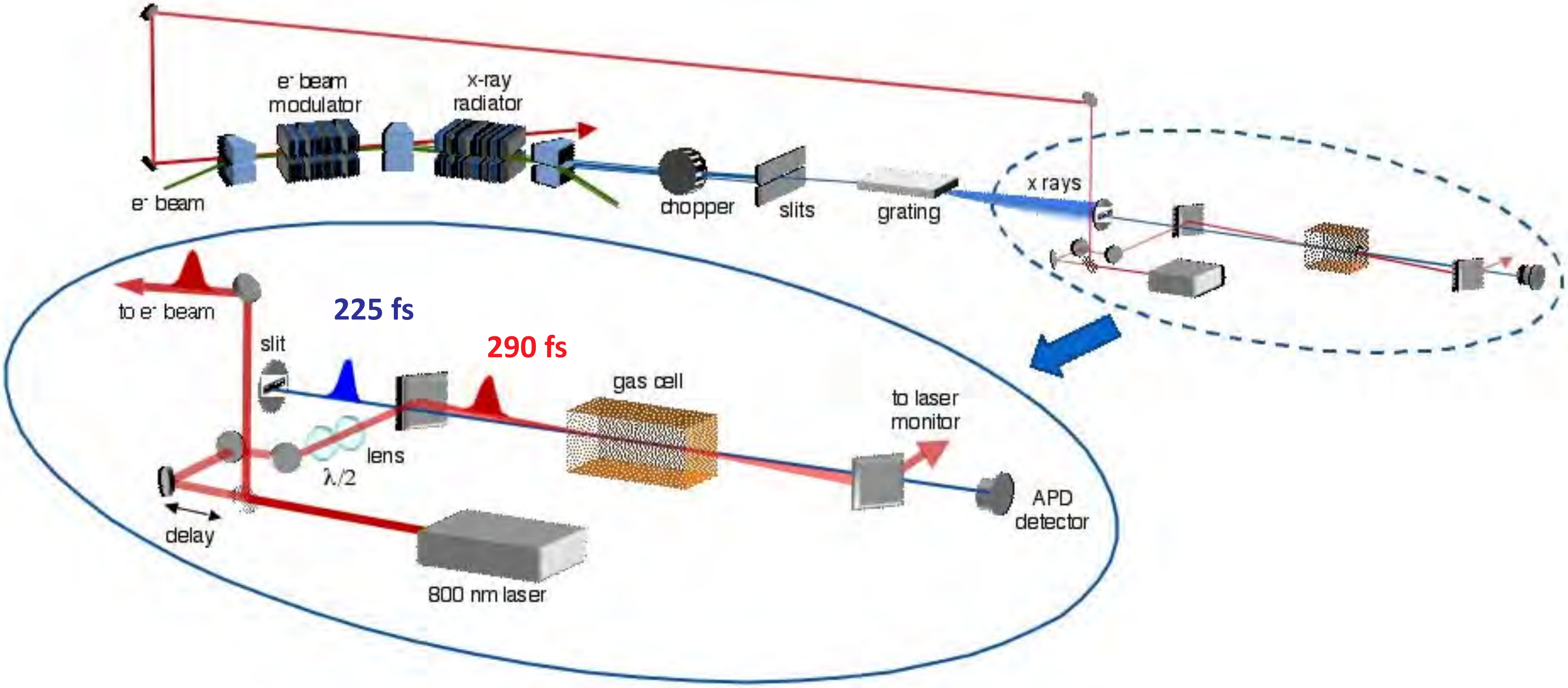
Femtosecond slicing beamline at Advanced Light Source

Co-located dressing laser (10^{13} W/cm^2) &
Monochromatic, tunable, short pulse (200 fs) soft x-rays (~870 eV)

Berkeley: T.E. Glover, M. Hertlein, T. Allison, J. van Tilborg, A. Belkacem, B. Rude
Argonne: E.P. Kanter, B. Krässig, R. Sanra, S.H. Southworth, H.R. Varma, L. Young

T. E. Glover et al. Nature Physics (2010)

ALS Femtosecond Spectroscopy Beamline & gas phase transient absorption apparatus



In situ characterization

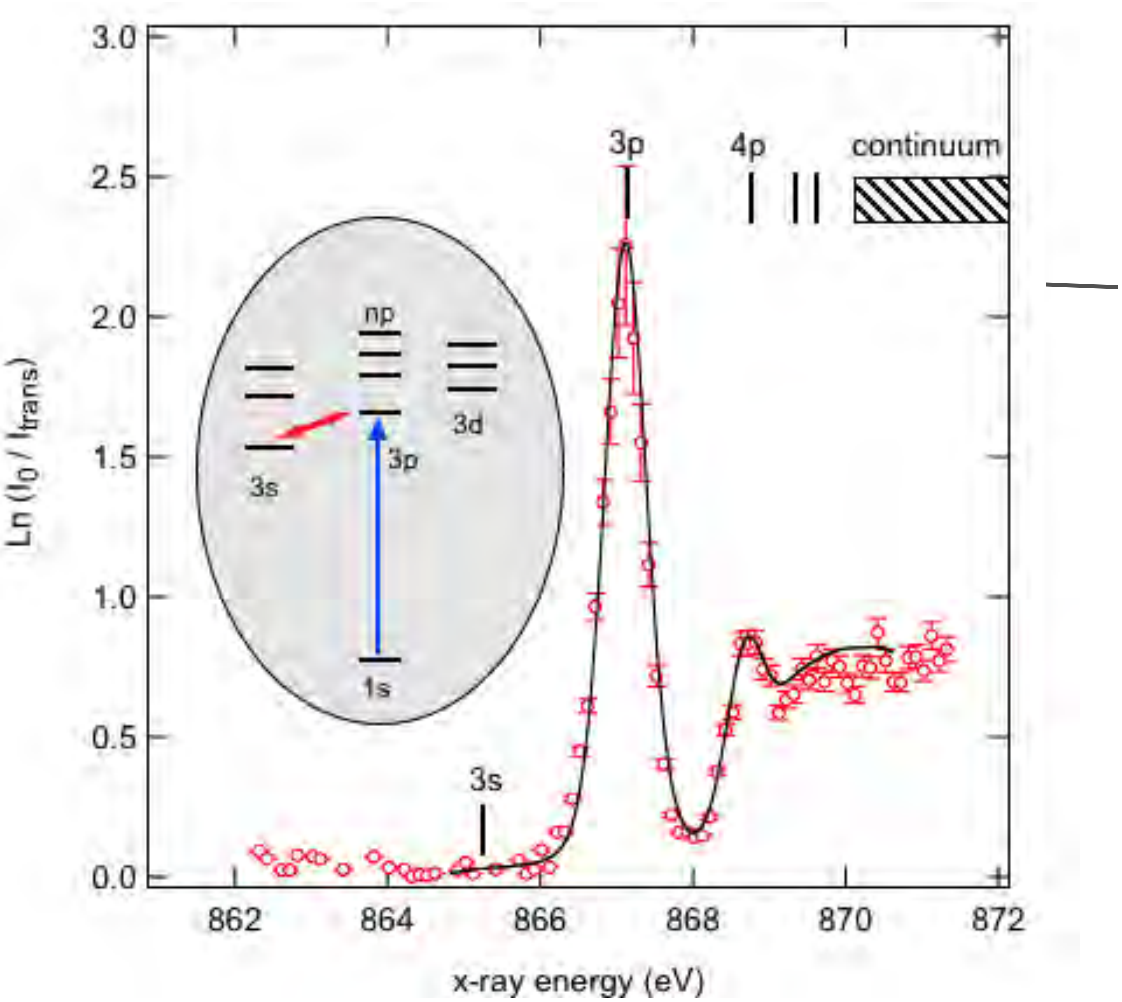
Starting overlap: ~3 ps, ~2 microns

x-rays: (54 × 84.5 μm) (H × V), 225 fs

laser: (80 - 150 × 160 - 195 μm) (H × V), 290 fs

laser pulse energy (1.12, 0.80, or 0.50 mJ)

Neon absorption spectrum

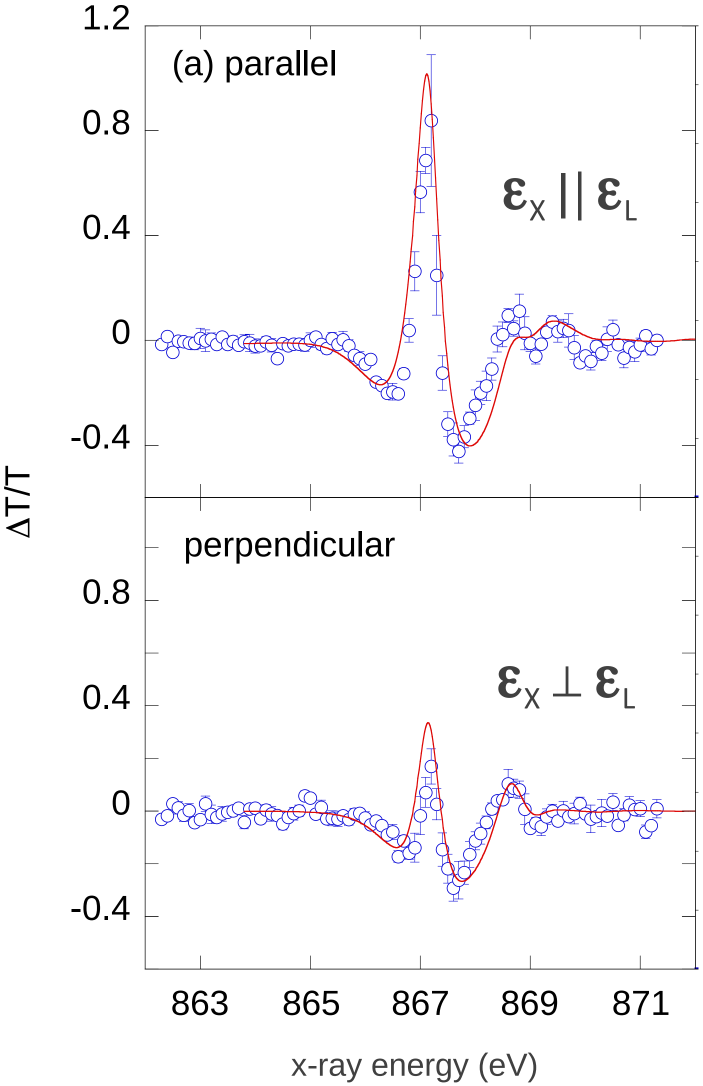


Convolve 0.24 eV Gaussian w/ best XANES
M. Coreno et al. PRA 99

Coupling laser
800 nm, linearly polarized
~10^{13} W/cm^2

Absorption spectrum w/ fs x-rays reproduces high resolution expt'l spectrum

Observe EIT for x-rays



- *large effect (200-300%)*

- *induced transparency parallel >> perpendicular*

- *1s-3p-3s subspace dominant*

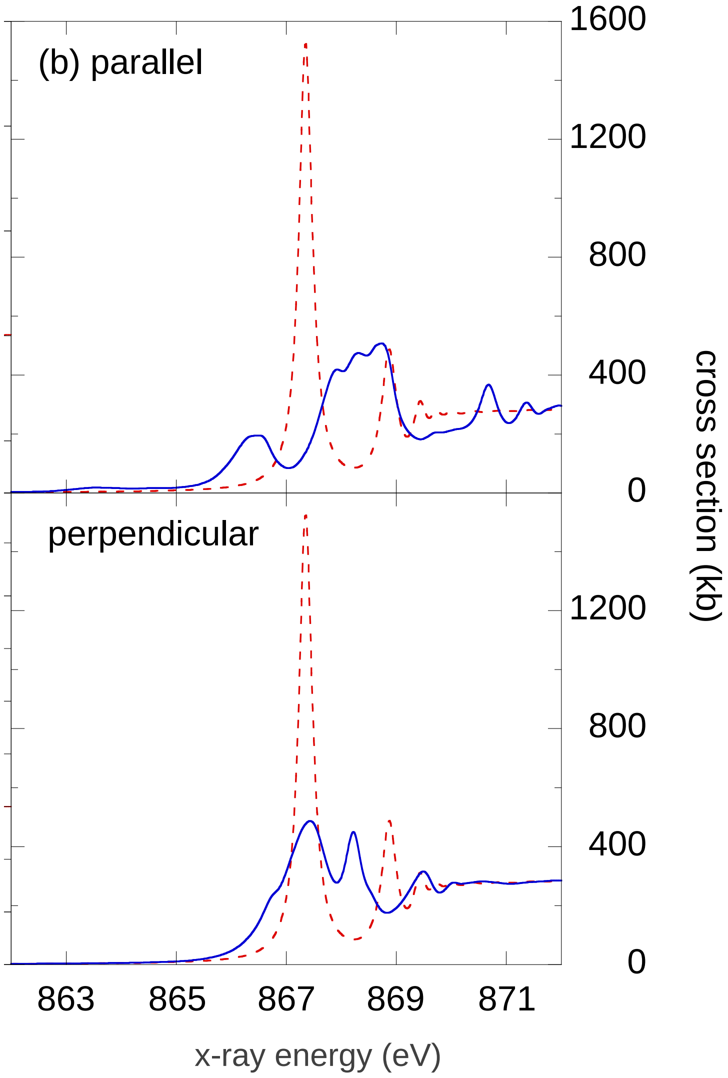
- *excellent agreement with theory simulation with no adjustable parameters*

Simulation of x-ray propagation through laser-dressed media

- *Ab initio x-ray cross sections as fcn of laser intensity 0.5×10^{11} - 3×10^{13} W/cm^2*

- *Co-propagate x-rays and dressing laser through the medium*
  - *-x-ray spatial grid*
    - *41x41 transverse*
    - *51 longitudinal*
  - *- 3900 time steps, 15.75 fs*
  - *- 864-875 eV, 0.02 eV steps*

- *X-ray transmission at each space time point*

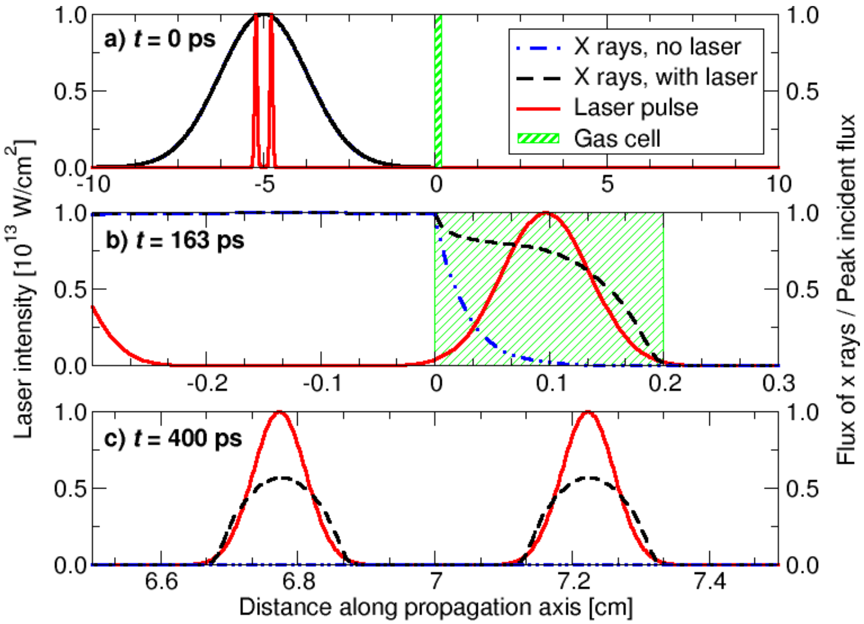


Summary and outlook - EIT for x-rays

- *Demonstrate control of photoelectric absorption of x-rays w/light*
  - *ultrafast, reversible x-ray switch*
  - *modifying the final state*
  - *predictive theoretical treatment*

- *Cross-correlation measurement of x-ray pulse width*

- *Imprint fs laser pulse shapes and sequences onto x-ray pulses*



- *Amplitude modulator for many wavelengths by multiplexing*
- *Extend to hard x-rays (Ar, Kr, Kr ions) - Buth & Santra PRA 08*
- *Control ratio of absorption to scattering*

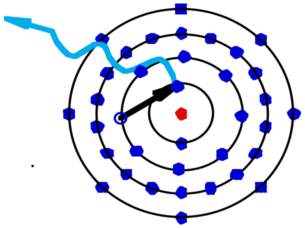
Strong field x-ray induced processes

This exposition, for which we have previously established a conceptual foundation, provides a natural introduction to the nonlinear phenomena which are currently under investigation. The reader will discover that the latter have really been ready for discovery for many years, awaiting the techniques which could make them observable. They have counterparts in phenomena long known to specialists, but the intense beams and the precision and sensitivity of measurement which are essential to accurate description and prediction have come only recently.

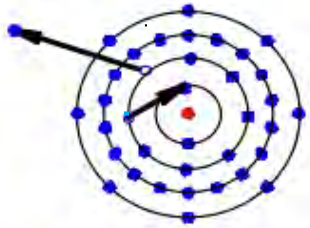
G. C. Baldwin
An Introduction to Nonlinear Optics (1969).

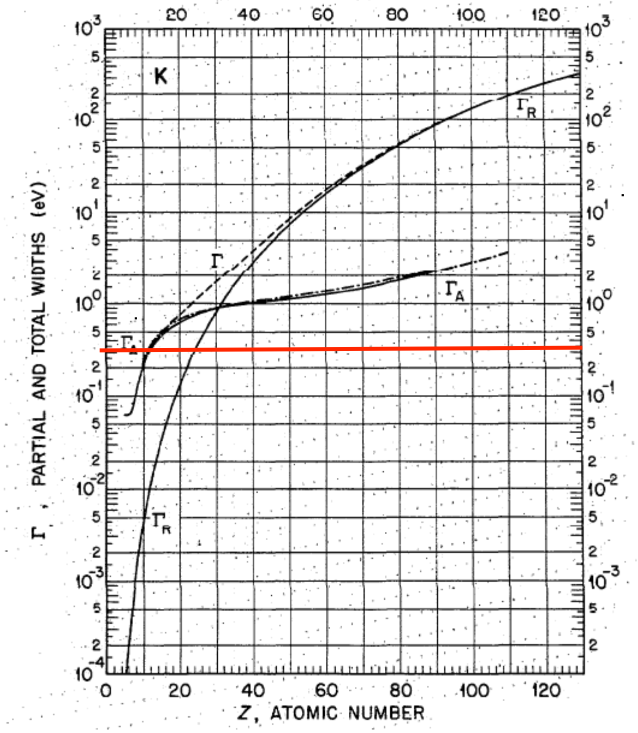
Intraatomic inner-shell decay vs x-ray–driven transitions

Radiative



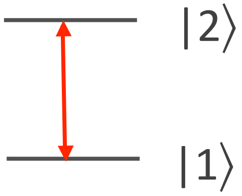
Auger





Rabi frequency

$$\Omega_{12} = \frac{\mu_{12}\mathrm{E}}{\hbar}$$



$\mu = er$

$1/\tau = \Omega_{12}$

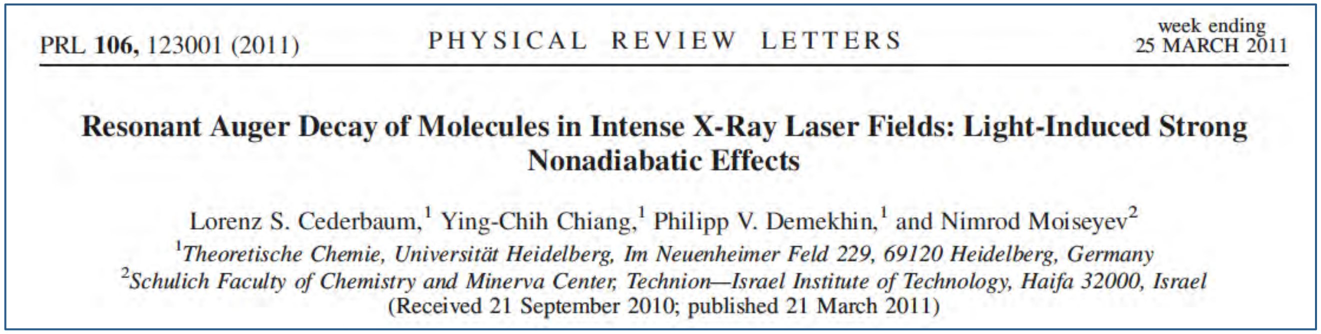
$E_{required} = 1/\mu_{12}\tau$

$\mu_{Ne\ 1s\text{-}3p} = 0.01\ ea_0$

$\tau_{Ne\ 1s^{-1}} = 2.4\ fs = 100\ a.u.$

$E_{Ne} \sim 1\ a.u.$

$I_{Ne} \sim 3.4\ x10^{16}\ W/cm^2$



PRL **106,** 123001 (2011) PHYSICAL REVIEW LETTERS week ending 25 MARCH 2011

Resonant Auger Decay of Molecules in Intense X-Ray Laser Fields: Light-Induced Strong Nonadiabatic Effects

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PHYSICAL REVIEW A **83**, 023422 (2011)

Strong interference effects in the resonant Auger decay of atoms induced by intense x-ray fields

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PHYSICAL REVIEW A **81**, 013812 (2010)

Propagation of a strong x-ray pulse: Pulse compression, stimulated Raman scattering, amplified spontaneous emission, lasing without inversion, and four-wave mixing

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Auger effect in the presence of strong x-ray pulses

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Exploration of strong-field x-ray interactions has just begun!