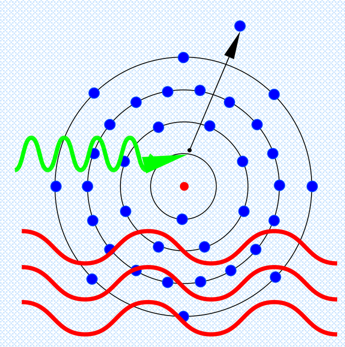
EIT in laser-dressed atoms probed by x rays

Christian Buth,* Robin Santra, Linda Young

Laser Dressing and X-ray Absorption

X-ray probe of laser-dressed atoms

- Atoms are in the field of an **optical laser**, $800\,\mathrm{nm}$ (**Ti:Sapphire**)
- Probed by x rays
- Laser dressing barely influenced by x rays
- Laser is of **moderately high intensity** $10^{13}\ \mathrm{W\,cm^{-2}}$
  - Ground state atomic electrons are neither excited nor ionized
  - Only final states are modified
- Keldysh parameter for Rydberg orbitals (here Ne $3p$): $\gamma=\sqrt{I_{3\mathrm{p}}/(2U_{\mathrm{p}})}=1.5$ => **Strong field regime**



Quantum electrodynamic description of atoms

- Non-relativistic **quantum electrodynamics** in electric dipole approximation

$$\hat{H}=\hat{H}_{\mathrm{AT}}+\hat{H}_{\mathrm{EM}}+\hat{H}_{\mathrm{L}}+\hat{H}_{\mathrm{X}}+\hat{W}=\hat{H}_0+\hat{H}_1$$

- **Hartree-Fock-Slater** one-electron model

$$\hat{H}_{\mathrm{AT}}=-\frac{1}{2}\hat{\nabla}^2+V_{\mathrm{HFS}}(r)$$

- **Free electromagnetic field** for two-modes (laser plus x rays)

$$\hat{H}_{\mathrm{EM}}=\omega_{\mathrm{L}}\hat{a}_{\mathrm{L}}^{\dagger}\hat{a}_{\mathrm{L}}+\omega_{\mathrm{X}}\hat{a}_{\mathrm{X}}^{\dagger}\hat{a}_{\mathrm{X}}$$

- **Interaction** of electrons with laser- or x-ray-light $\lambda=\mathrm{L},\mathrm{X}$

$$\hat{H}_{\lambda}=\vec{x}\cdot\mathrm{i}\sqrt{2\pi V^{-1}\omega_{\lambda}}\ [\vec{e}_{\lambda}\hat{a}_{\lambda}-\vec{e}_{\lambda}^{*}\hat{a}_{\lambda}^{\dagger}]$$

- Continuum electrons treated with complex absorbing potential $\hat{W}$

Refraction of x rays

- **Atomic polarizability** due to x rays

$$2\,\mathrm{Re}\,E_{I,2}=-\frac{1}{4}\alpha(\omega_{\mathrm{X}})$$

- Polarization of gas medium

$$P(\omega_{\mathrm{X}})=n_{\mathrm{nd}}\alpha(\omega_{\mathrm{X}})E(z,t)$$

- Solve **Maxwell wave equation** with plane wave for $E(z,t)$

$$\frac{\partial^2 E(z,t)}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2 E(z,t)}{\partial t^2}=\frac{4\pi}{c^2}\frac{\partial^2 P(z,t)}{\partial t^2}$$

- **Index of refraction**

$$n(\omega_{\mathrm{X}})\equiv\frac{k\,c}{\omega_{\mathrm{X}}}=1+2\pi\,n_{\mathrm{nd}}\alpha(\omega_{\mathrm{X}})$$

Laser-atom interaction

- Hamiltonian for the atom in the laser field **[no x rays so far]**

$$\hat{H}_0=\hat{H}_{\mathrm{AT}}+\hat{H}_{\mathrm{EM}}+\hat{H}_{\mathrm{L}}+\hat{W}$$

- Direct product basis set of atomic orbitals $\psi_{n,l,m}(\vec{r})$ and laser Fock states with μ laser photons absorbed $\mu>0$ (emitted $\mu<0$)

$$|\Phi_{nlm\mu}\rangle=|\psi_{n,l,m}\rangle\ |N_{\mathrm{L}}-\mu\rangle$$

- Diagonalization yields **laser-dressed atomic energy levels**

$$(\boldsymbol{H}_0^{(m)})_{nl\mu,n'l'\mu'}=\langle\Phi_{nlm\mu}|\hat{H}_0|\Phi_{n'l'm\mu'}\rangle$$

$$\boldsymbol{H}_0^{(m)}\vec{c}_F^{(m)}=E_F^{(m)}\vec{c}_F^{(m)}$$

X-ray photon absorption

- Decaying core excited state with complex Siegert energy
- Relaxes by **Auger decay** [Ne 2.4 fs] and **x-ray fluorescence** [Kr 240 as] => extra width $E_{F,0}^{(m)}=E_F^{(m)}-\mathrm{i}\,\Gamma_{1s}^{\exp}/2$
- X-ray probe $\hat{H}_1\equiv\hat{H}_{\mathrm{X}}$ is a **weak, one-photon** process => Non-Hermitian Rayleigh-Schrödinger perturbation theory
- Initial state $|I\rangle=|\psi_{1,0,0}\rangle\ |N_{\mathrm{L}}\rangle\ |N_{\mathrm{X}}\rangle$ and laser-dressed, core-excited final states $|F^{(m)}\rangle=\sum_{n,l,\mu}c_{F,n,l,\mu}^{(m)}|\Phi_{nlm\mu}\rangle\ |N_{\mathrm{X}}-1\rangle$

$$E_{I,0}=\langle I|\hat{H}_0|I\rangle,\qquad E_{I,1}=\langle I|\hat{H}_1|I\rangle=0$$

$$E_{I,2}=\sum_{F,m}\frac{\langle I|\hat{H}_1|F^{(m)}\rangle\langle F^{(m)}|\hat{H}_1|I\rangle}{E_{I,0}-E_{F,0}^{(m)}}$$

$$\Gamma=-2\,\mathrm{Im}[E_{I,0}+E_{I,1}+E_{I,2}]\ \Rightarrow\ \sigma_{1s}=2\frac{\Gamma}{J_{\mathrm{X}}}$$

Absorption of x rays

$$\sigma_{1s}(\omega_{\mathrm{X}},\vartheta_{\mathrm{LX}})=\sigma_{1s}^{\parallel}(\omega_{\mathrm{X}})\cos^2(\vartheta_{\mathrm{LX}})+\sigma_{1s}^{\perp}(\omega_{\mathrm{X}})\sin^2(\vartheta_{\mathrm{LX}})$$

$$\sigma_{1s}^{\parallel}(\omega_{\mathrm{X}})\equiv\sigma_{1s}^{0}(\omega_{\mathrm{X}}),\qquad\sigma_{1s}^{\perp}(\omega_{\mathrm{X}})\equiv\sigma_{1s}^{1}(\omega_{\mathrm{X}})$$

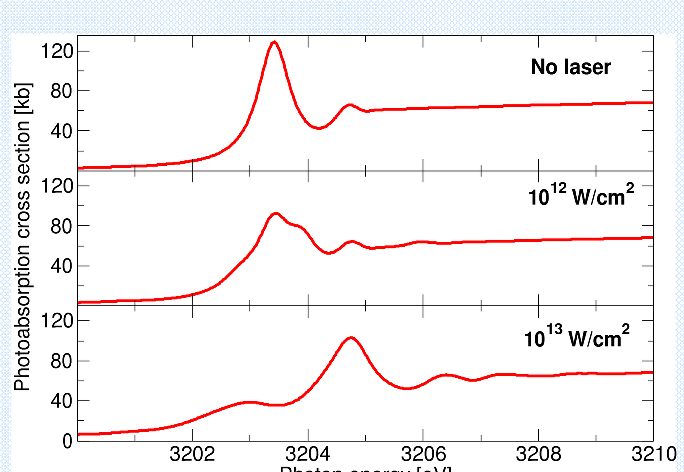
$$\sigma_{1s}^{|m|}(\omega_{\mathrm{X}})=\frac{8\pi}{3}\alpha\,\omega_{\mathrm{X}}\,\mathrm{Im}\left[\sum_F\frac{(d_F^{|m|})^2}{E_{F,0}^{|m|}-E_{1s}-\omega_{\mathrm{X}}}\right]$$

- Atom is **cylindrically deformed** along the laser axis
- Dependence on angle between polarizations ϑ_{LX}
- Atomic properties described by $\sigma_{1s}^{\parallel}(\omega_{\mathrm{X}})$, $\sigma_{1s}^{\perp}(\omega_{\mathrm{X}})$
- Radial dipole matrix element between initial and dressed final state $d_F^{|m|}$; energy of K edge E_{1s}

Argon and Krypton

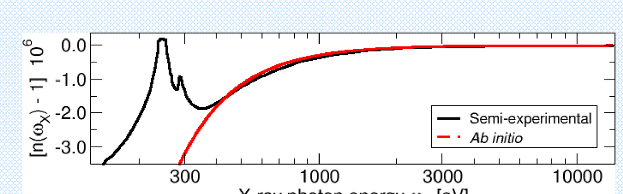
Argon K edge: absorption

- X-ray absorption on $1s\rightarrow 4p$ resonance
- Laser dressing with $800\,\mathrm{nm}$ light
- Parallel x-ray and laser polarizations
- Line width $\Gamma_{1s}=0.66\,\mathrm{eV}$
- **Strong suppression** of absorption on resonance

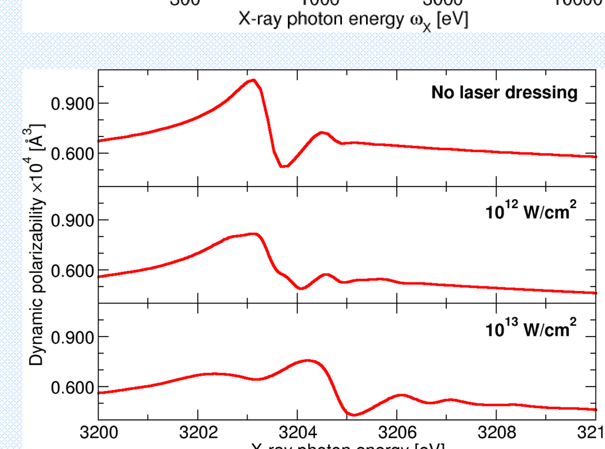


Argon K edge: refraction

- X-ray refractive index with $\hat{H}_{\mathrm{X}}=\alpha\,\vec{p}\cdot\vec{A}_{\mathrm{X}}+\alpha^2\vec{A}_{\mathrm{X}}^2/2$

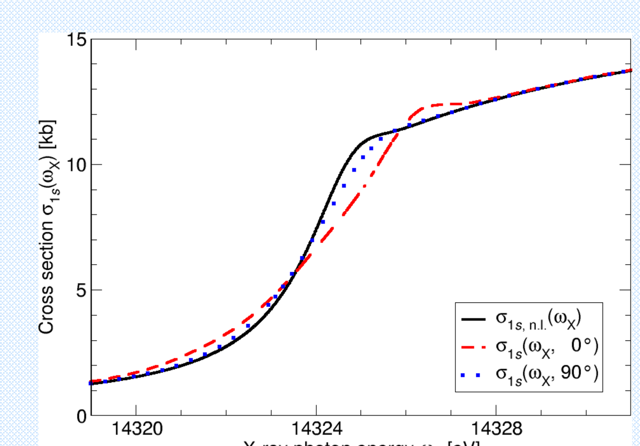


- Polarizability with $\hat{H}_{\mathrm{X}}$ in length form
- Dispersion **reduced** by laser dressing (with $800\,\mathrm{nm}$ light at $10^{13}\ \mathrm{W\,cm^{-2}}$)
- No slow x rays!



Krypton K edge

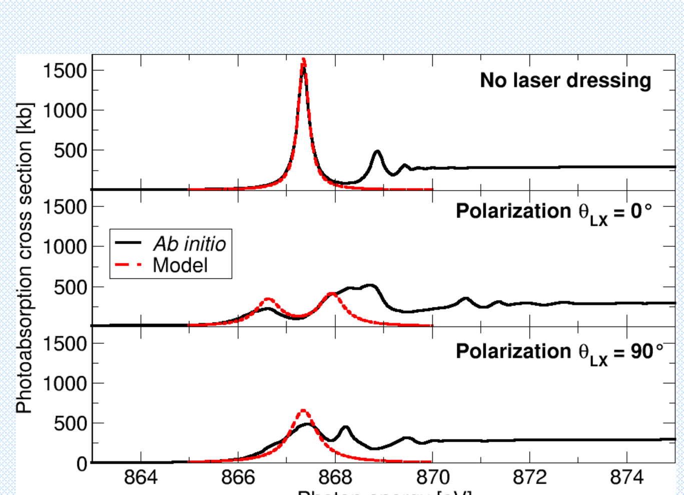
- Laser dressing with $800\,\mathrm{nm}$ at $10^{13}\ \mathrm{W\,cm^{-2}}$ on the $1s\rightarrow 5p$ transition
- Laser influences cross section in the **vicinity** of the K edge
- **Largest effect** for parallel polarization in relation to no laser
- **Moderate** effect (20%) due to the line width $\Gamma_{1s}=2.7\,\mathrm{eV}$



Neon

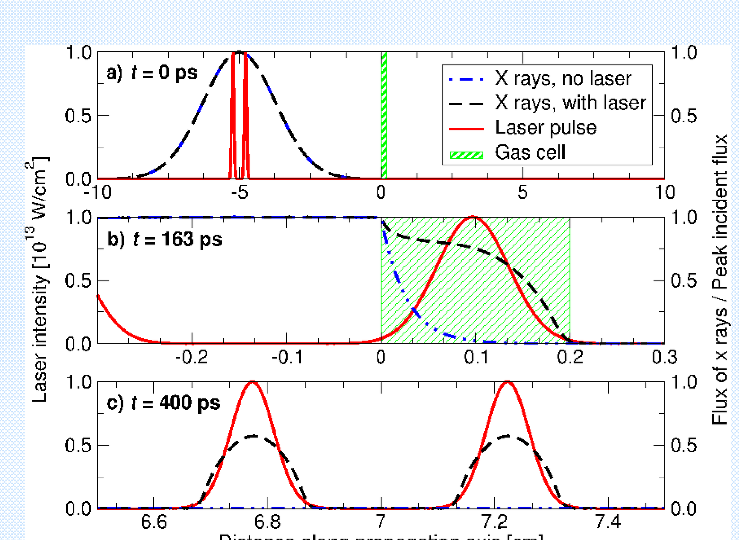
Neon K edge

- Laser dressing with $800\,\mathrm{nm}$ at $10^{13}\ \mathrm{W\,cm^{-2}}$
- Rydberg series clearly resolved due to a **low line width** $\Gamma_{1s}=0.27\,\mathrm{eV}$
- For **parallel polarizations** transparency at the $1s\rightarrow 3p$ transition
- Dominant physics from three levels: $1s$, $3s$, and $3p$



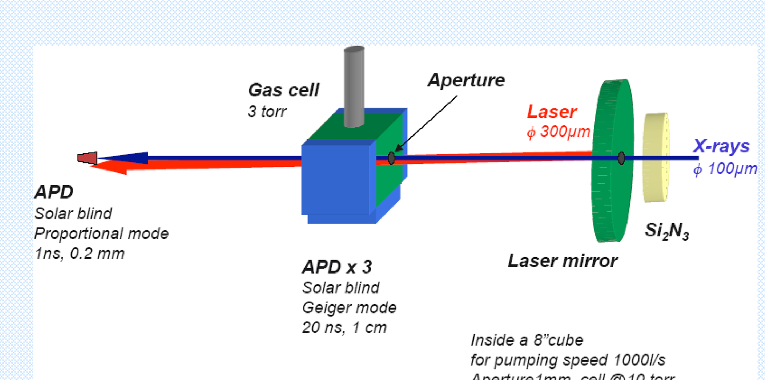
Ultrashort pulse shaping of x rays

- Laser pulse shape is **imprinted** on x rays
- **Femtosecond** x-ray pulses
- All x-ray pump-probe experiments
- Amplitude modulation only



Schematic experimental setup of two-color neon experiment

- Experiment under way at Lawrence Berkeley National Laboratory
- Overlap x rays and laser beams both in **space** and **time**
- Need **ultrafast x-ray source** for neon experiment



*Present Address

Department of Physics and Astronomy
Louisiana State University
Baton Rouge, Louisiana 70803, USA

Email: cbuth@phys.lsu.edu

References

1. Buth, Santra, Young, Phys. Rev. Lett., 98, 253001 (2007)
2. Buth, Santra, Phys. Rev. A 75, 033412 (2007)
3. Buth, Santra, Cederbaum, Phys. Rev. A 69, 032505 (2004)

A U.S. Department of Energy laboratory managed by UChicago Argonne, LLC

Alexander von Humboldt Stiftung/Foundation