



... for a brighter future



Alexander von Humboldt
Stiftung / Foundation



UChicago ▶
Argonne LLC



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

Electromagnetically induced transparency for x-rays by high intensity lasers

Christian Buth, Robin Santra, Linda Young

Atomic, Molecular, and Optical Physics Group
Chemistry Division

Lawrence Berkeley National Laboratory, Berkeley, California, USA
February 21, 2007

Contents

1. Introduction
2. Theory of laser-dressed atoms
3. Krypton
4. Neon
5. Conclusion

X-ray probe of laser-manipulated atoms

- Pump-probe-experiments with lasers and x-rays since the advent of synchrotron radiation sources
- Mostly weak laser
- **Review:** Wuilleumier, Meyer, **J. Phys. B 39, R425 (2006)**

- **Pump:** valence **ionization** by linearly polarized laser
 - Electrons are ejected predominantly along the laser axis
 - Hole orbital is aligned in ion
- **Probe:** electron **excitation** from *K*-shell to hole orbital
 - Background free; only absorption for ionized atoms
 - Different photoabsorption for parallel and perpendicular x-rays
- **Krypton atom:** Young *et al.*, **Phys. Rev. Lett. 97, 083601 (2006)**

Quantum state-resolved EUV probe of xenon ions

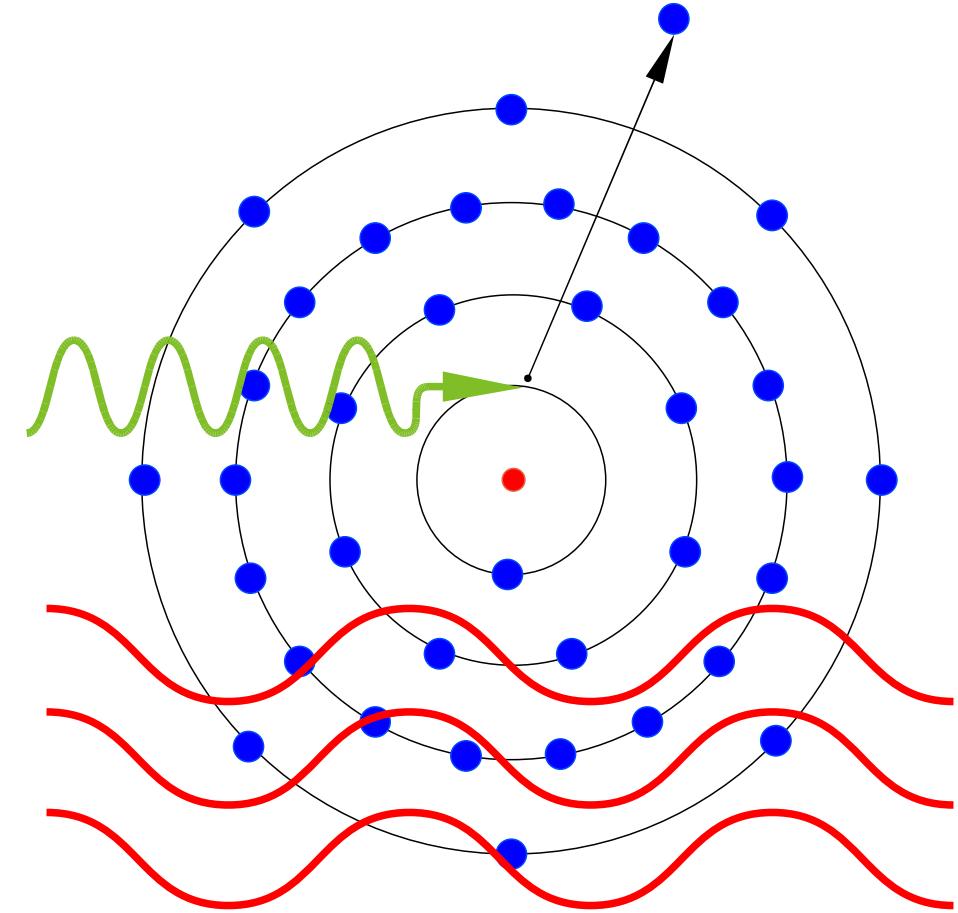
- **Pump:** Laser ionization of $5p$ valence electron of xenon atoms
- **Probe:** Excitation of $4d$ core electron to $5p$ valence hole for parallel and perpendicular polarizations
- **Experiment:** femtosecond high-order harmonic transient absorption spectroscopy of a xenon atom

- **Theory:** xenon is a heavy atom with appreciable spin-orbit coupling
- Strong-field tunnel ionization of $5p$ valence electron of xenon atoms
- Ionization produces a distribution of $|j, m\rangle$ quantum states
- Probabilities to find the accessible quantum states are calculated

[Loh et al., Phys. Rev. Lett. 98, 143601 (2007)]

X-ray probe of laser-dressed atoms

- Atoms are in the field of an optical laser
- Probed by x-rays
- Laser dressing barely influenced by x-rays



[Buth, Santra, Phys. Rev. A 75, 033412 (2007)]

Laser characteristics

- Assume an 800 nm (**Ti:Sapphire**) laser system
- Laser is of **moderately high intensity** $10^{13} \text{ W cm}^{-2}$
 - Atomic electrons are neither excited nor ionized
 - Only final states are modified
- **Keldysh parameter** for Rydberg orbitals (here Ne $3p$)

$$\gamma = \sqrt{\frac{I_{3p}}{2 U_p}} = 1.5 \approx 1$$

- **Strong field regime**
[$\gamma \ll 1$ adiabatic tunneling picture; $\gamma \gg 1$ perturbative multiphoton process]
- **Multiphoton** physics
- Need **sophisticated** theoretical treatment!

[**Buth, Santra, Young, Phys. Rev. Lett. 98, 253001 (2007)**]

Contents

1. Introduction
- 2. Theory of laser-dressed atoms**
3. Krypton
4. Neon
5. Conclusion

Quantum electrodynamic description of atoms

- Hartree-Fock-Slater one-electron model
[radial part of the wave function is given in a flexible finite-element basis]

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

- Non-relativistic quantum electrodynamics in electric dipole approximation
- Free electromagnetic field for the two-modes (laser plus x-rays)

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

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

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

[Butch, Santra, Phys. Rev. A 75, 033412 (2007)]

Resonance energies using complex absorbing potentials

- Complex absorbing potential (CAP) \hat{W} added to the Hamiltonian
- CAP derived from smooth exterior complex scaling
- Spectrum of Hamiltonian is modified by CAP
- Decaying state becomes a bound state with a complex Siegert energy

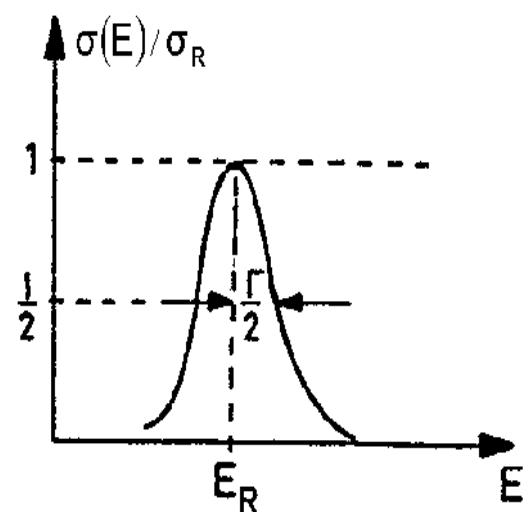
$$E_{\text{res}} = E_R - i \frac{\Gamma}{2}$$

- Wave function

$$\psi \propto e^{-i E_{\text{res}} t} \propto e^{-i E_R t} e^{-\frac{\Gamma}{2} t}$$

- Resonance position E_R and width Γ ; lifetime $\frac{1}{\Gamma}$

[Buth, Santra, Phys. Rev. A 75, 033412 (2007)]



Laser-atom interaction

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

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

- Direct product basis set of atomic orbitals and laser Fock states

$$|\Phi_{nlm\mu}\rangle = |\psi_{n,l,m}\rangle |\mu\rangle$$

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

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

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

[Butch, Santra, Phys. Rev. A 75, 033412 (2007)]

X-ray photon absorption

- Decaying core excited state with complex Siegert energy
- Relaxes [Kr 230 as, Ne 2.3 fs] by **Auger decay** and **x-ray fluorescence**
- Decay of 1s core hole is accounted for by $E_F^{(m)} = E_F^{(0)} - i\Gamma_{1s}^{\text{exp}}/2$
- X-ray probe $\hat{H}_1 \equiv \hat{H}_X$ is a **weak, one-photon** process
=> Non-Hermitian Rayleigh-Schrödinger perturbation theory
- Initial ground state $|I\rangle$ and laser-dressed core-excited final states $|F^{(m)}\rangle$

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

$$E_{I,2} = \sum_{F,m} \frac{\langle I | \hat{H}_1 | F^{(m)} \rangle \langle F^{(m)} | \hat{H}_1 | I \rangle}{E_{I,0} - E_{F,0}^{(m)}} \quad \Rightarrow \quad \sigma_{1s} = 2 \frac{\Gamma}{J_X}$$

$$\Gamma = -2 \text{ Im} [E_{I,0} + E_{I,1} + E_{I,2}],$$

[**Buth, Santra, Phys. Rev. A 75, 033412 (2007)** and **Buth, Santra, Cederbaum, Phys. Rev. A 69, 032505 (2004)**]

Total x-ray absorption cross section

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

$$\sigma_{1s}^{\parallel}(\omega_X) \equiv \sigma_{1s}^{|0|}(\omega_X), \quad \sigma_{1s}^{\perp}(\omega_X) \equiv \sigma_{1s}^{|1|}(\omega_X)$$

$$\sigma_{1s}^{|m|}(\omega_X) = \frac{8\pi}{3} \alpha \omega_X \operatorname{Im} \left[\sum_F \frac{(d_F^{|m|})^2}{E_F^{|m|} - E_{1s} - \omega_X} \right]$$

- Atom is **cylindrically deformed** along the laser polarization axis
- Dependence on angle between polarizations ϑ_{LX}
- Atomic properties described by $\sigma_{1s}^{\parallel}(\omega_X)$, $\sigma_{1s}^{\perp}(\omega_X)$
- Electron correlations and **non-dipole effects** ignored
=> manifest in deviations from the angular behavior

[Buth, Santra, Phys. Rev. A 75, 033412 (2007)]

Contents

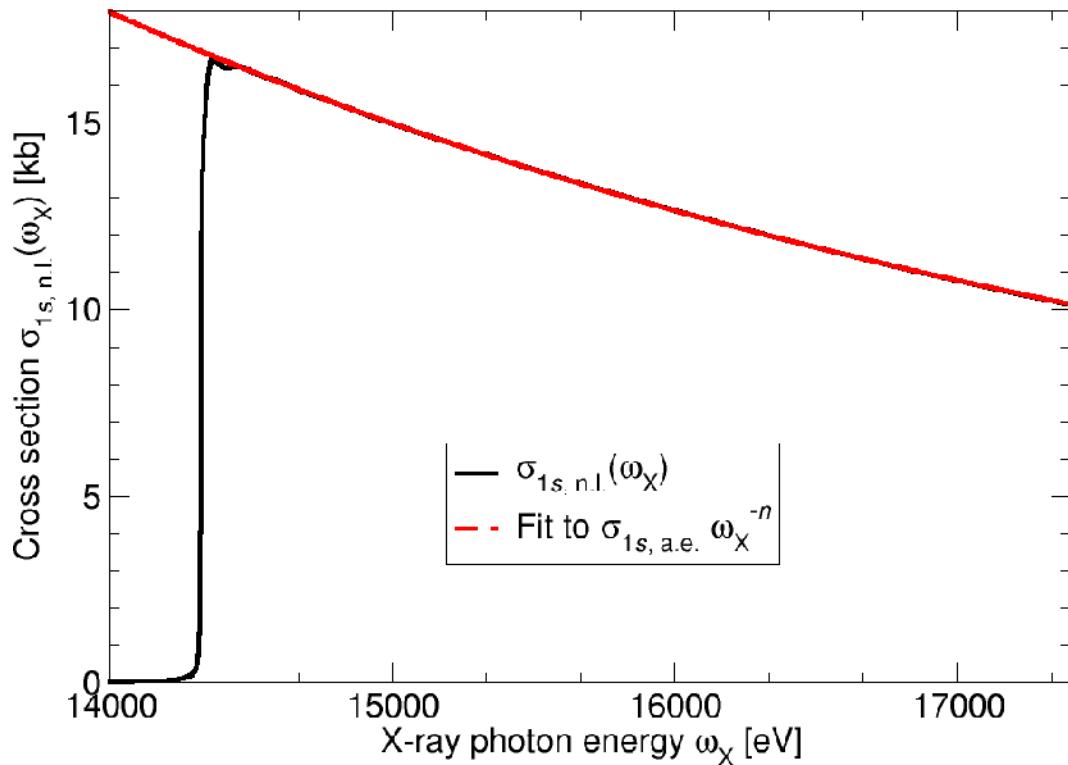
1. Introduction
2. Theory of laser-dressed atoms
3. **Krypton**
4. Neon
5. Conclusion

Krypton above the K-edge

- Without laser dressing; only x-ray absorption
- Above edge Bethe and Salpeter give

$$\sigma_{1s}(\omega_X) = \frac{\sigma_{1s, \text{a.e.}}}{\omega_X^n}$$

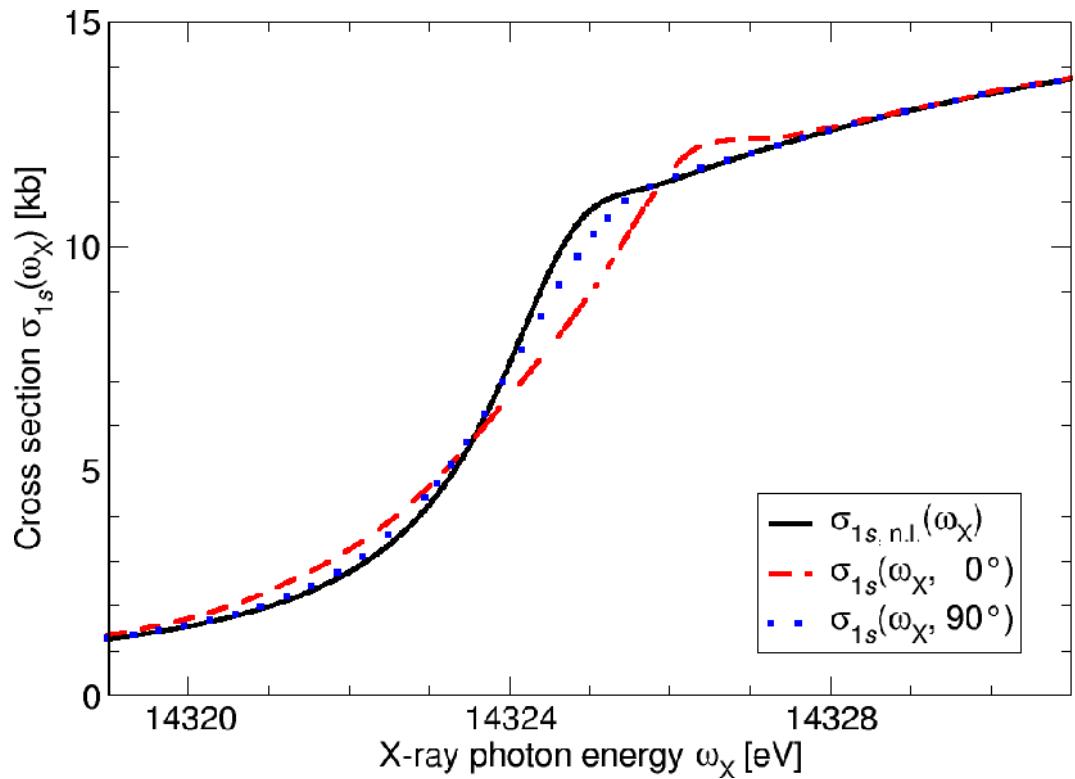
- Non-linear fit yields $n=2.63$
- For hydrogen $n=\frac{8}{3}=2.67$
- Test of Hartree-Fock-Slater model, radial finite-element basis, and CAP method



[Buth, Santra, Phys. Rev. A 75, 033412 (2007)]

Krypton K-edge

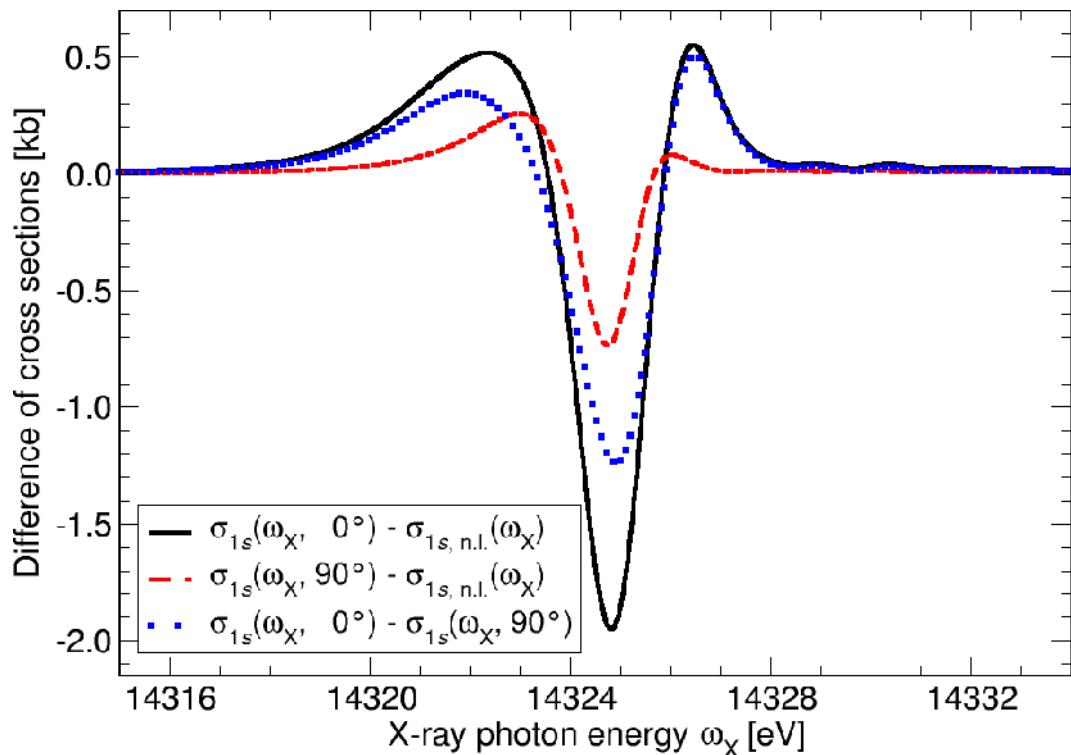
- Laser dressing with 800 nm at $10^{13} \text{ W cm}^{-2}$
- Laser influences cross section in the **vicinity** of the K-edge
- Dependence on the polarization between laser and x-rays
- **Appreciable** effect (20%)



[Buth, Santra, Phys. Rev. A 75, 033412 (2007)]

Differences of cross sections at the krypton K-edge

- Impact revealed by looking at differences
- **Largest effect** for parallel polarization in relation to the case without laser
- Shape understood: transition $1s \rightarrow 5p$ is **suppressed** by laser. Oscillator strength is redistributed to $5s$ and $4d$
- Reason for moderate effect: the **width** $\Gamma_{1s} = 2.7$ eV

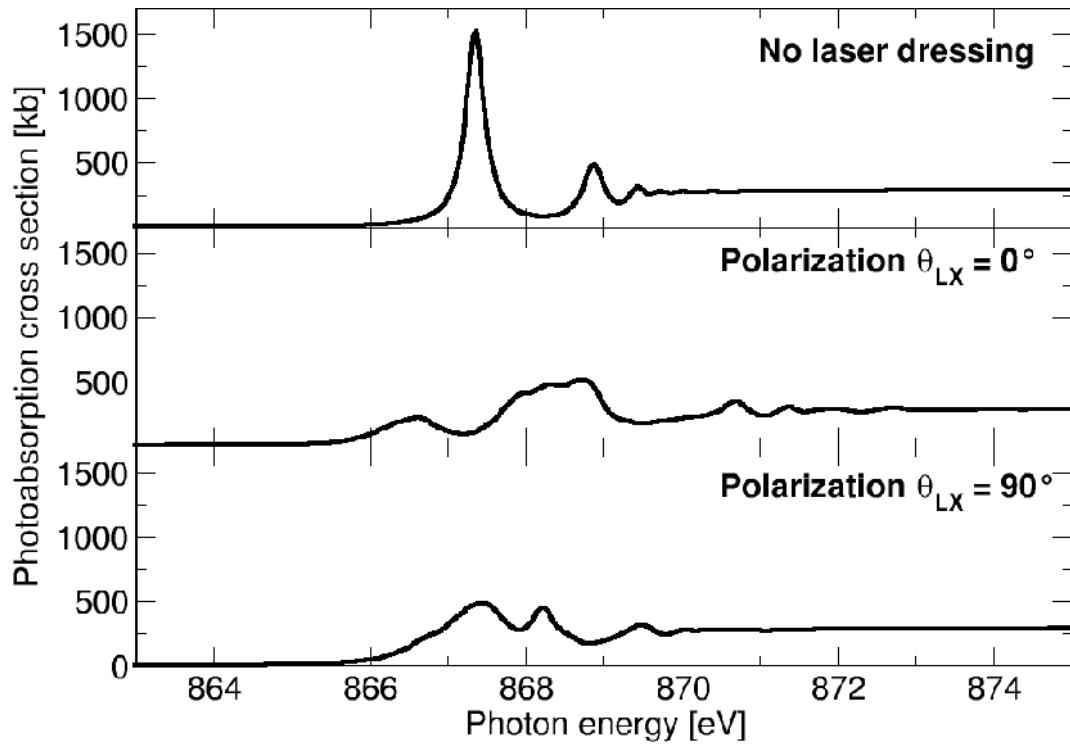


[Buth, Santra, Phys. Rev. A 75, 033412 (2007)]

Neon K-edge

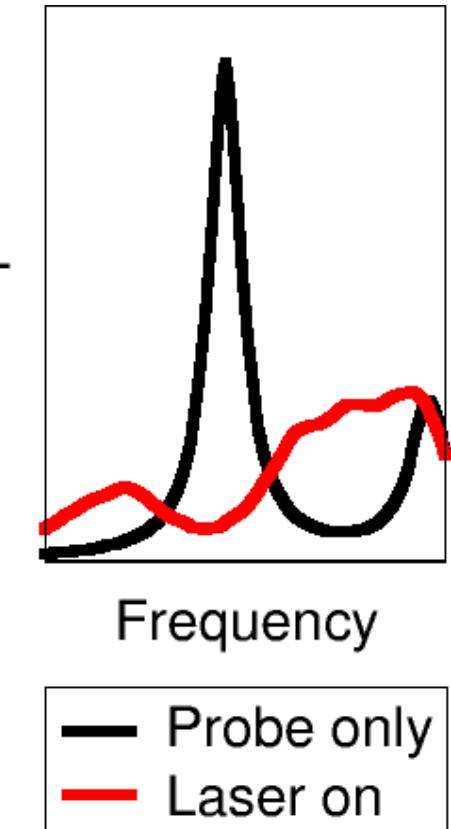
- Rydberg series clearly resolved due to a **lower line width** $\Gamma_{1s} = 0.27$ eV
- Big effect of laser dressing
- For **parallel polarizations** transparency at the $1s \rightarrow 3p$ transition
- Suppression but no transparency for **perpendicular polarizations**
- Absorption and emission of up to **20 laser photons** to converge calculations

[Buth, Santra, Young, Phys. Rev. Lett. 98, 253001 (2007)]



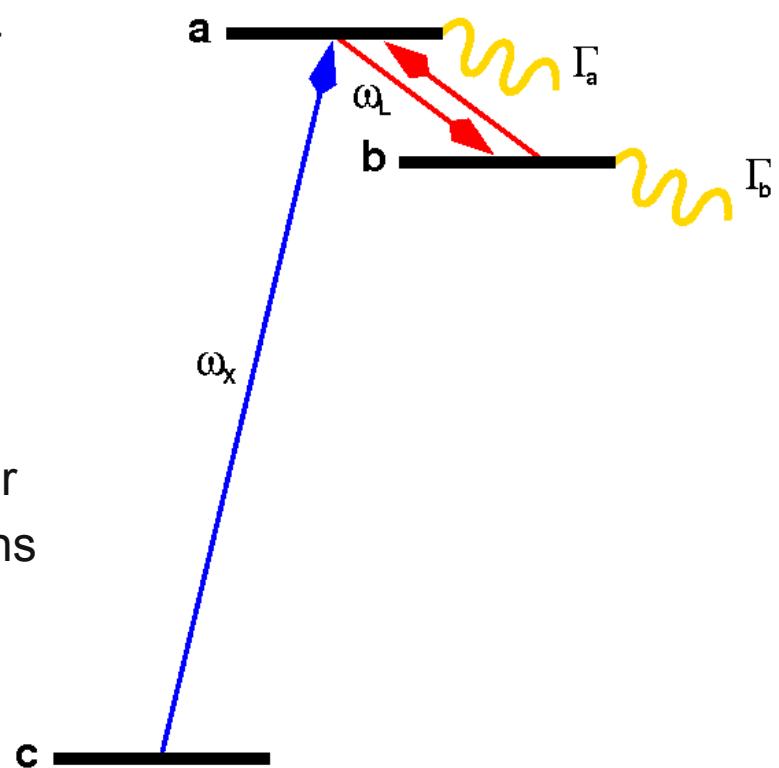
Electromagnetically induced transparency (EIT)

- Gas is **opaque** for light at an intraatomic transition
- Gas becomes **transparent** for this light by laser-dressing
- So far EIT for **optical** wavelengths has been studied
- We investigate EIT for **x-rays**
- **Review:** Fleischhauer, Imamoğlu, Marangos, **Rev. Mod. Phys. 77, 633 (2005)**
- Extremely versatile tool in quantum optics
 - Nonlinear optics
 - Atomic clocks
 - Quantum computer



Λ -type three-level model for EIT

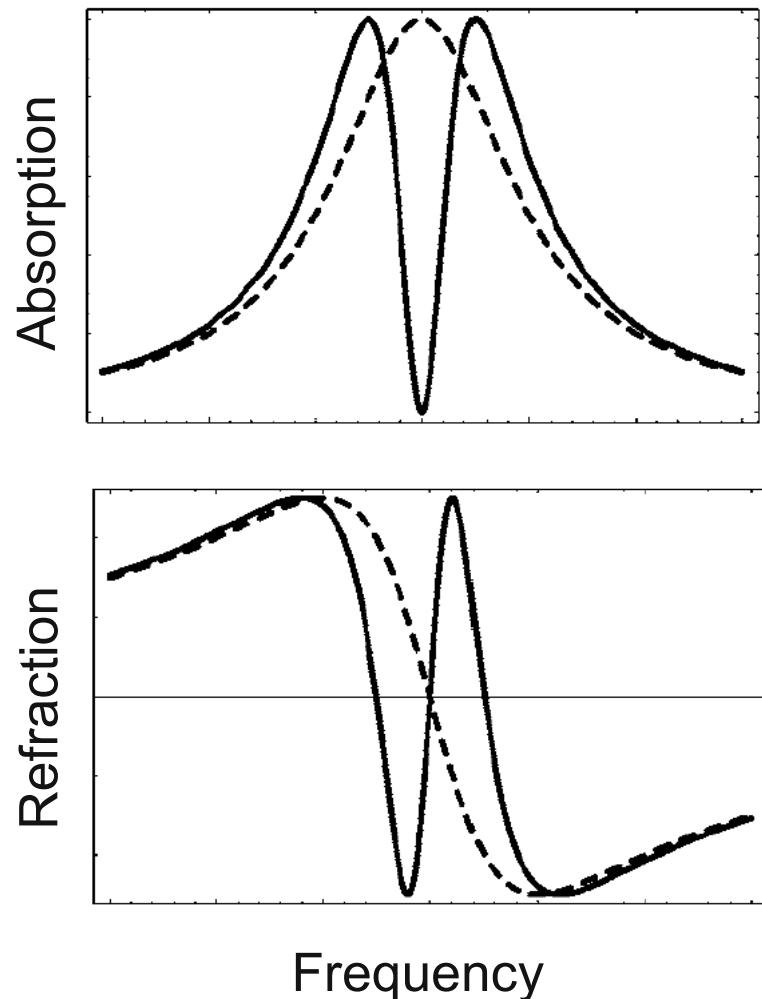
- EIT is understood in terms of a Λ -type three-level model
- Two-color light
 - Probe transition $c \rightarrow a$
 - Laser coupling $a \rightarrow b$
- In resonance transition **without** coupling laser
- Laser resonant with transition within line widths
- Coupling laser dresses levels a, b
 - Rabi flopping
 - Autler-Townes doublet
- Destructive interference



[Buth, Santra, Young, Phys. Rev. Lett. 98, 253001 (2007)]

Optical properties of EIT media

- The EIT dramatically changes the **refraction** of the medium
- Absorption vanishes on resonance => ideal for optics
- **Phase velocity** speed of light in vacuum
- **Group velocity** substantially reduced => **slow light** 17 m/s [Hau *et al.*, *Nature* **397**, 594 (1999)] in a Bose-Einstein Condensate of sodium atoms
- Few photons; light stored in excitations

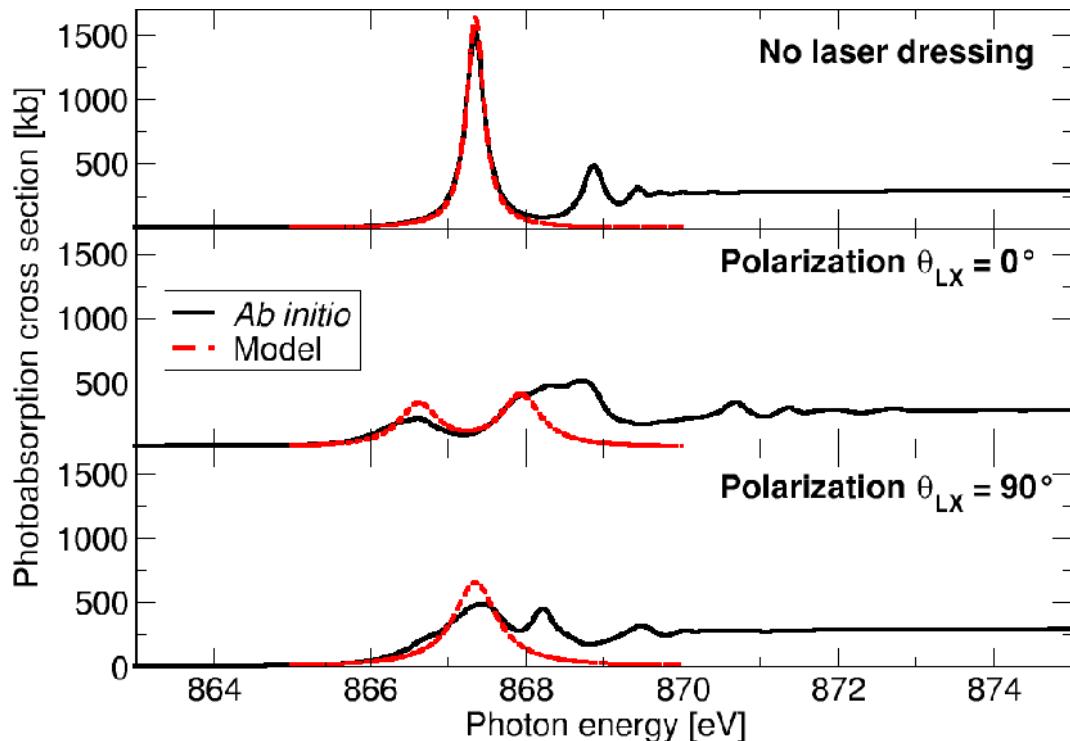


EIT in strontium

- First observation by Boller, Imamoglu, Harris, **Phys. Rev. Lett. 66, 2593 (1991)** in strontium
- Probe laser with intensity 10^4 W cm^{-2}
- Laser coupling with intensity $1.5 \times 10^7 \text{ W cm}^{-2}$
- Upper level is autoionizing with lifetime 4.4 ps larger by more than **three orders of magnitude** than lifetime of neon core hole
- Transmission change from $\exp(-1)$ to $\exp(-20)$ by coupling laser

Λ -type EIT model for neon

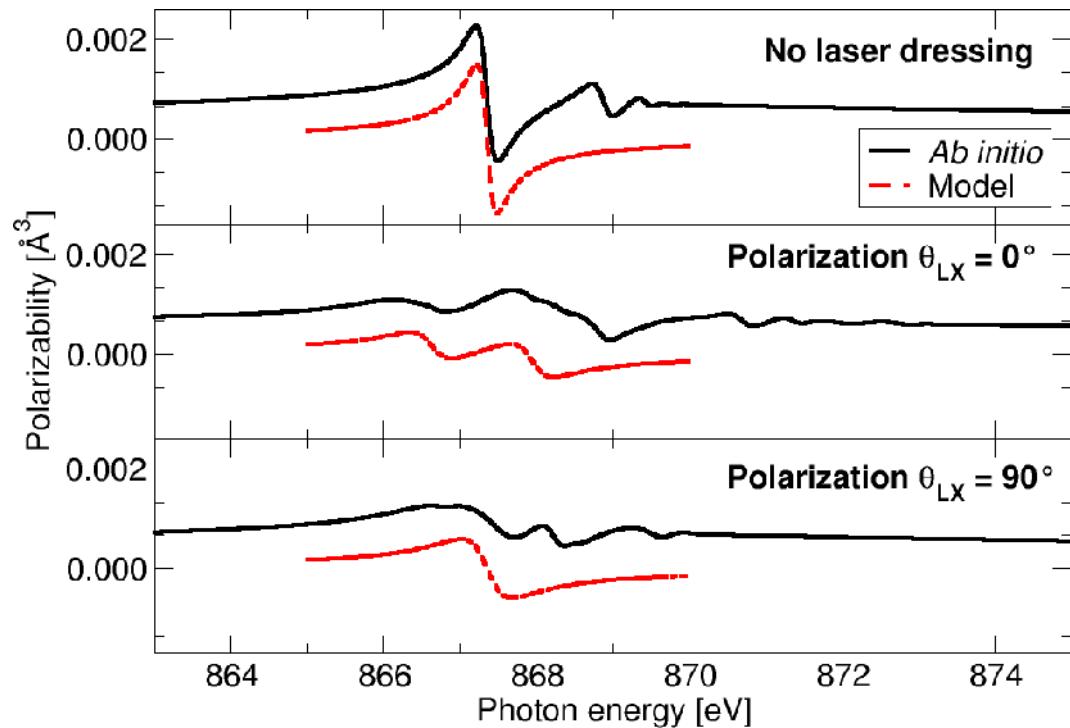
- Assume dominant physics results from three levels of neon: $1s$, $3s$, and $3p$.
- Parameters are level energies and widths; dipole moments between $3p$ and $1s$, $3s$.
- Other levels contribute
- Multiphoton processes



[Buth, Santra, Young, Phys. Rev. Lett. 98, 253001 (2007)]

Polarizability of laser-dressed neon atoms

- Atomic polarizability proportional to real part of the **index of refraction**
- EIT at optical frequencies leads to a large change in the atomic polarizability
- Instead for x-rays the atomic polarizability with laser dressing is lower than without



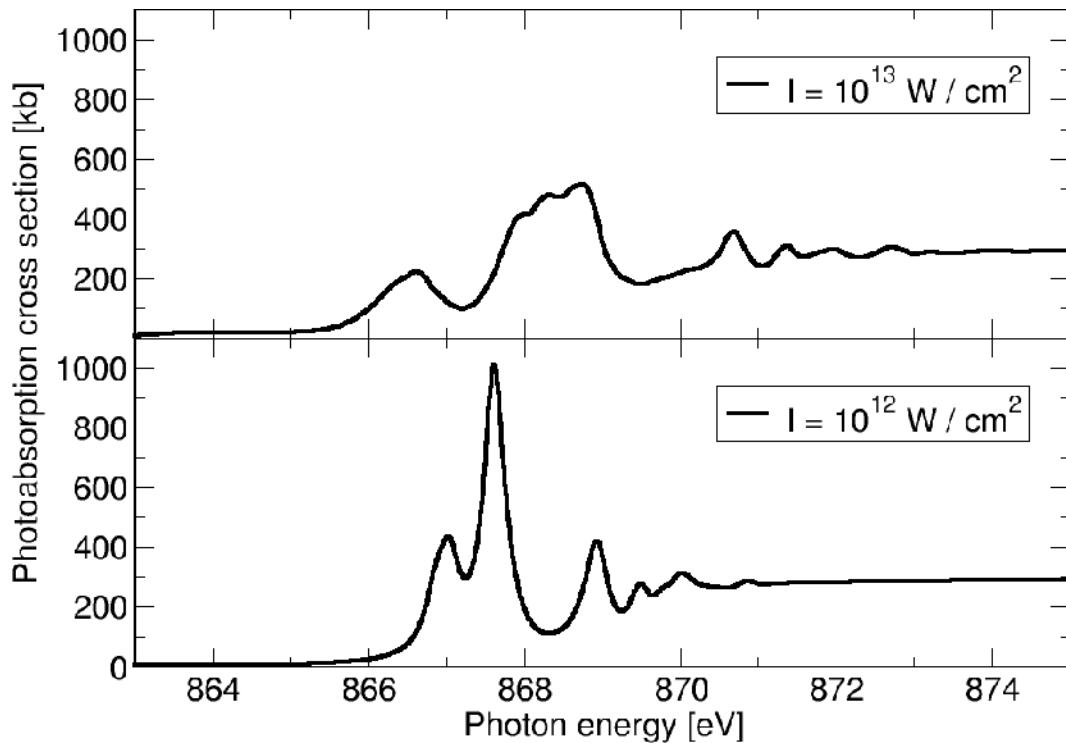
[Buth, Santra, Young, Phys. Rev. Lett. 98, 253001 (2007)]

Laser-intensity dependence for parallel polarizations

- Crude estimate for the laser intensity $|\Omega_{ab}| > \Gamma_{1s}$ to see EIT yields:

$$I_L > 4.3 \times 10^{11} \frac{\text{W}}{\text{cm}^2}$$

- The cross section depends crucially on the laser intensity
- Main features persist even if the intensity is lowered by a factor of 10



[Buth, Santra, Young, Phys. Rev. Lett. 98, 253001 (2007)]

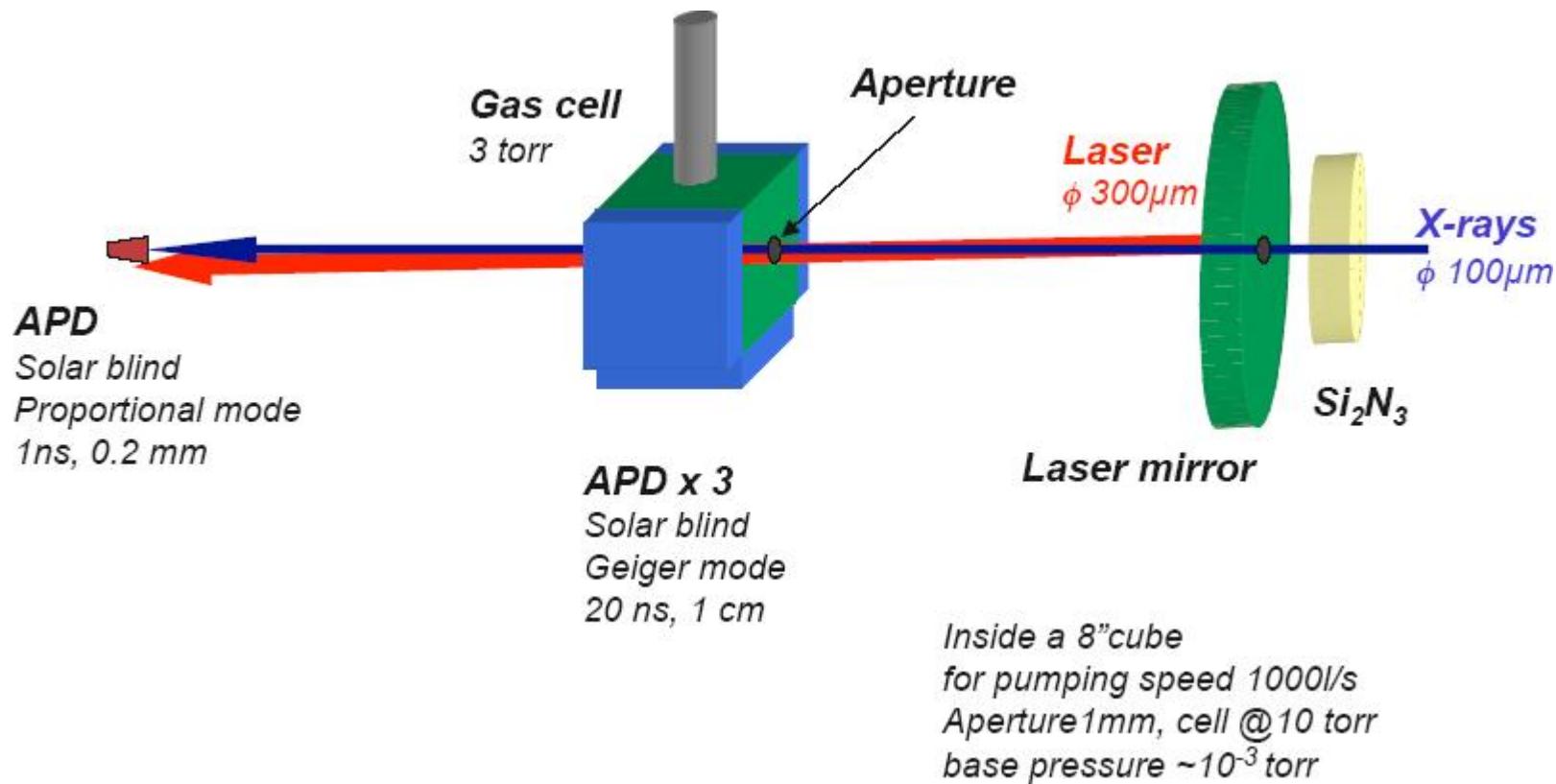
Experimental feasibility

- Number density 10^{17} atoms cm^{-3} for characteristic absorption length of 1 cm
- EIT for x-rays for **pulse shaping** of the intensity envelope
- Dispersion of 2π after 1 cm requires 10^{19} atoms cm^{-3}
- Hard to use as an experimental tool
- X-ray pulses must be **shorter** than laser pulse to probe laser-dressed atom
- Ti:Sapphire laser system produces pulses with energy, 1 mJ waist $300 \mu \text{m}$ and **duration** 140 fs at the intensity $10^{13} \text{ W cm}^{-2}$
- Need **ultrafast slicing source** for neon experiment
- Weak dependence on dressing-laser wavelength
[for a variation of 30% there is still EIT]

[Buth, Santra, Young, Phys. Rev. Lett. 98, 253001 (2007)]

Schematic experimental setup of two-color neon experiment

- Overlap x-ray and laser beams both in **space** and **time**



Conclusion

- X-ray probe of laser-dressed atom is described using **Hartree-Fock-Slater** approximation and nonrelativistic **quantum electrodynamics**
- Laser dressing causes **strong field multiphoton physics**
- X-ray probe is treated as a **one-photon** process
- Formalism suitable to investigate multiphoton x-ray processes for x-ray FELs

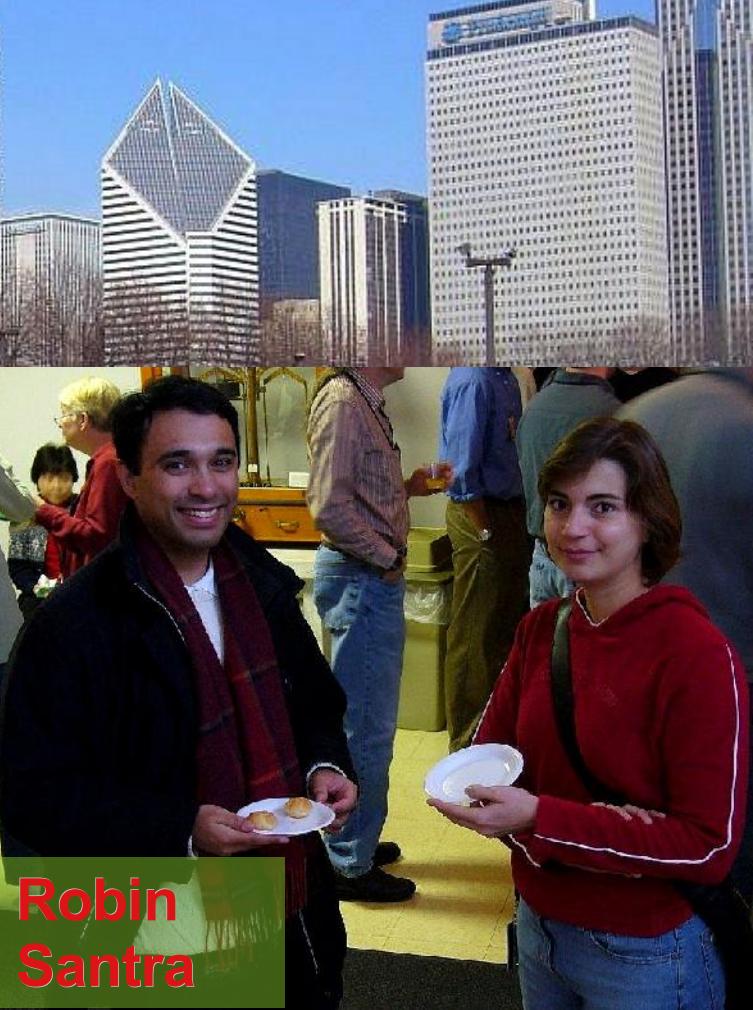
- Laser influences the photoabsorption cross section of krypton and neon
- Large width of core-excited states yields a moderate effect in krypton
- Find **electromagnetically induced transparency** effect in neon

- EIT effect can be measured in neon
- **High intensity laser**
- Need **ultrafast** x-rays due to ultrashort laser pulses

Acknowledgment



Linda Young



Robin Santra