Atomic Physics with Intense X-rays at LCLS

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Dual Motivation to Perform Atomic Physics Studies

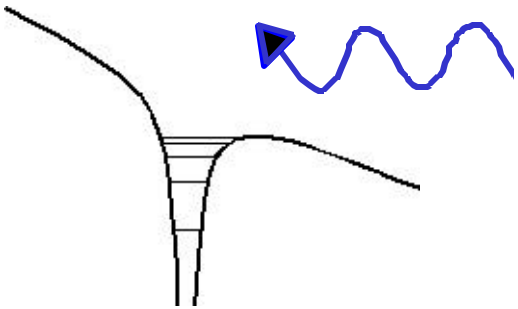
Fundamental Science

- **The LCLS, as a high-intensity high-energy photon source, provides a unique opportunity to study fundamental aspects of x-rays interacting with atoms, ions, molecules, and clusters**

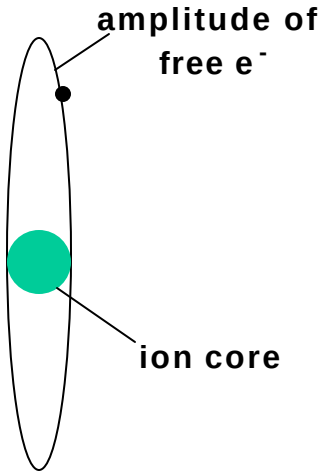
Foundation for all experimental planning

- **The understanding of x-ray–atomic physics interactions is central to experimental designs at the LCLS, as well as all next generation x-ray sources.**

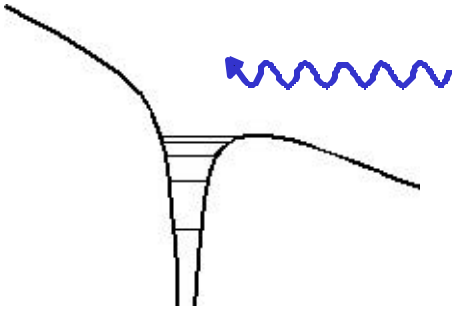
- **Current laser-atom process at I 10^{14} W/cm^2**



- Field modulates the atomic potential at visible laser frequency
- Outer e$^-$ has time to tunnel free:
  - $2U_p > I_p$ where $U_p \propto (I\lambda^2)^2$
- Strong interaction between free e$^-$ and ion core is of interest

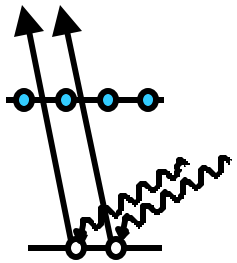


- **LCLS-atom process at I 10^{14} W/cm^2**

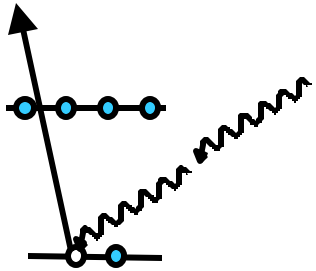


- Field modulates the atomic potential at x-ray laser frequency
- e$^-$ do not have time to tunnel free
- Important processes are with deeply bound core e$^-$

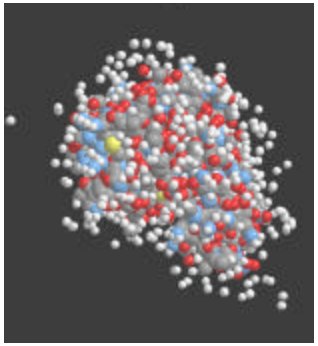
Experiment 1: Multiple ionization sufficiently rapid to form hollow atoms

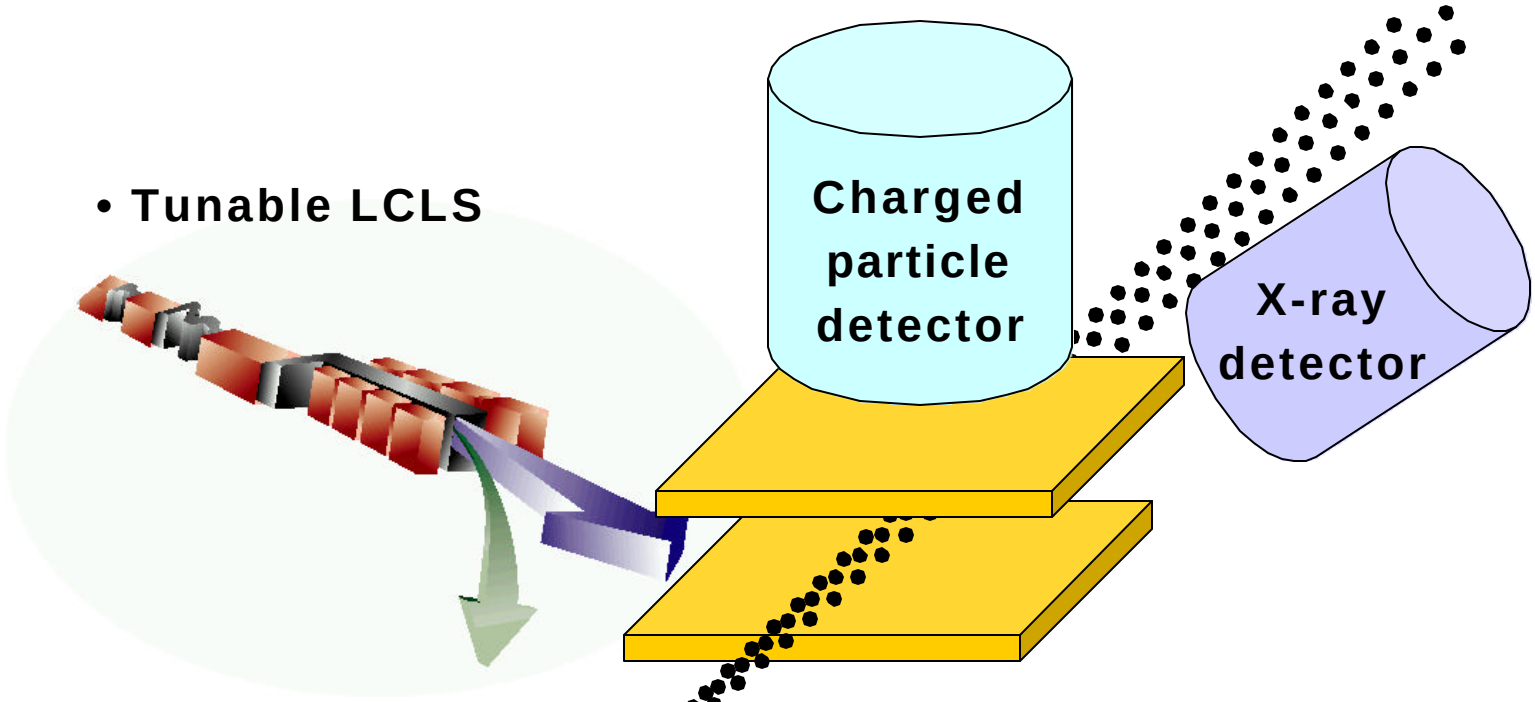


Experiment 2: Multiphoton ionization yielding absorption below the edge

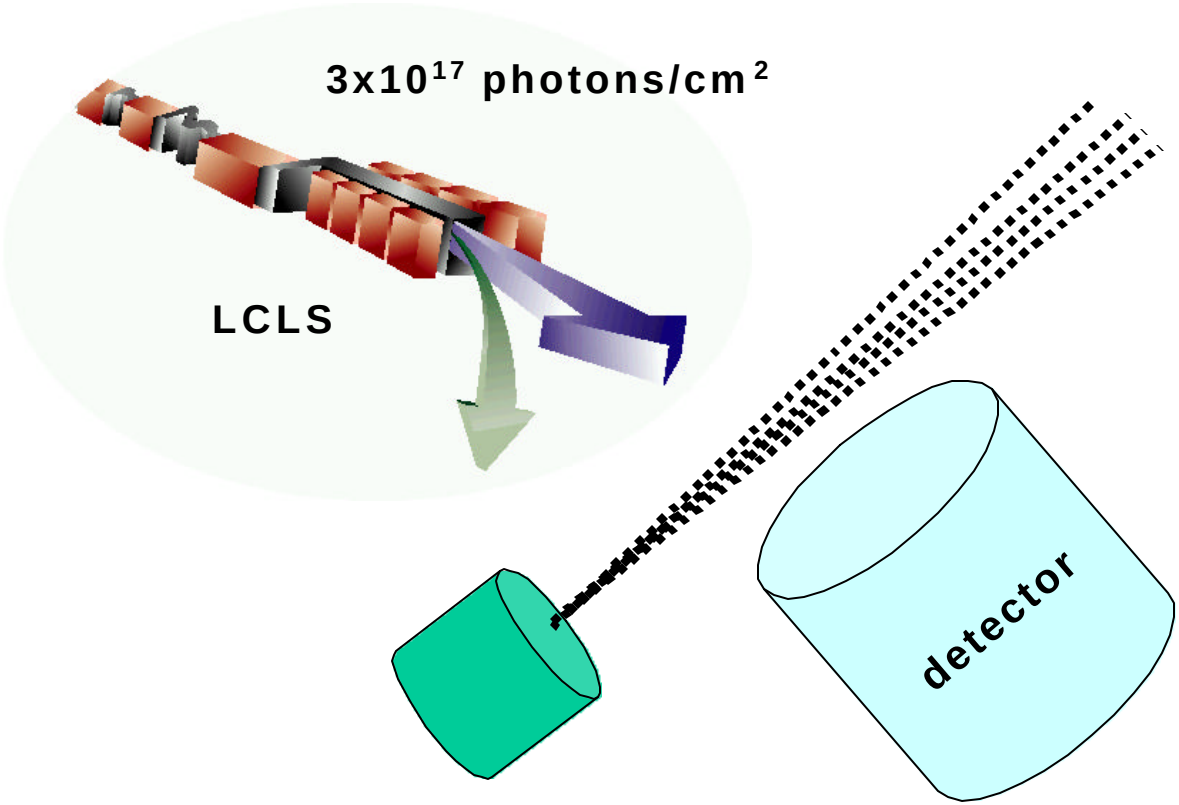


Experiment 3: Giant Coulomb explosions of clusters




• Tunable LCLS
Charged particle detector
X-ray detector
• Atom or cluster source
• Detectors
• Charge state spectrometer
• Electron energy spectrometer
• Ion recoil detector
• X-ray fluorescence detector

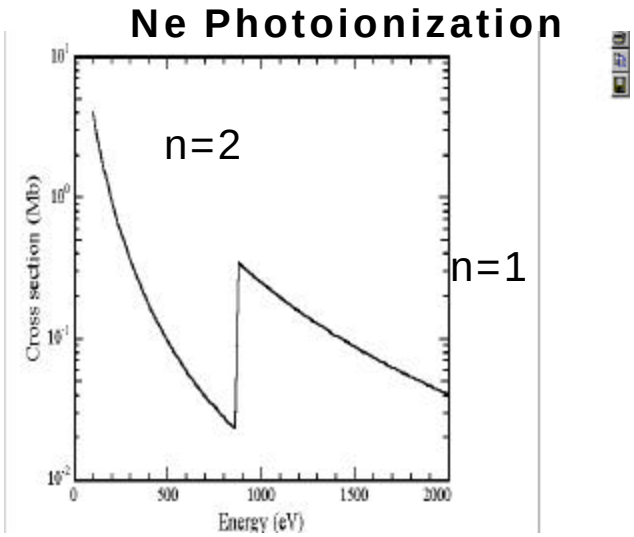
Experiment 1: Multiple ionization forming hollow atoms



Ionization: $G = 10^{12} s^{-1}$

Auger: $G = 4 \times 10^{14} s^{-1}$

- **Neon will display the effect well**
- **Relatively high photoionizations**
- **Non-corrosive monatomic sample**
- **Simple, well understood spectrum**
- **Relatively long Auger decay rate (2.5 fs)**
- **Auger relaxation > 100 x radiative fluorescence**

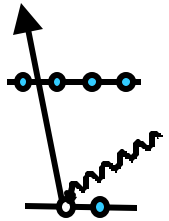


Possible Ionization Processes for LCLS Interacting with Ne

LCLS

- **Photoionization:**

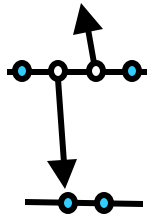
  $Ne + h\nu_{>870eV} \rightarrow Ne^{+*}(K) + e$



- **Auger Decay:**

  $Ne + h\nu_{>870eV} \rightarrow Ne^{+*}(K) + e \rightarrow Ne^{2+*}(LL) + e$

  $\rightarrow Ne^{3+*} + 2e$



LCLS only

- **Sequential multiphoton ionization:**

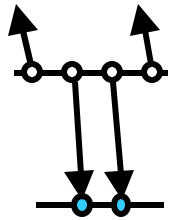
  $Ne + h\nu_{>870eV} \rightarrow Ne^{+*}(K) + e + h\nu_{>993eV} \rightarrow Ne^{2+*}(KK) + e \rightarrow Ne^{3+} + e$

  $\rightarrow Ne^{4+} + 2e$

  $\rightarrow Ne^{5+} + 3e$

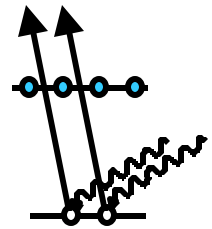
  $\rightarrow \ldots$

  $Ne + h\nu_{>870eV} \rightarrow Ne^{+*}(K) + e + h\nu_{>993eV} \rightarrow Ne^{3+*}(KLL) + e$



- **Direct multiphoton ionization:**

  $Ne + 2\,h\nu_{>932eV} \rightarrow Ne^{2+*}(KK) + 2e$

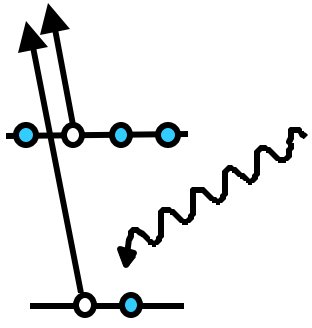


One Photon or Two? An Extremely Difficult Question for Multielectron/multiphoton Systems

LCLS

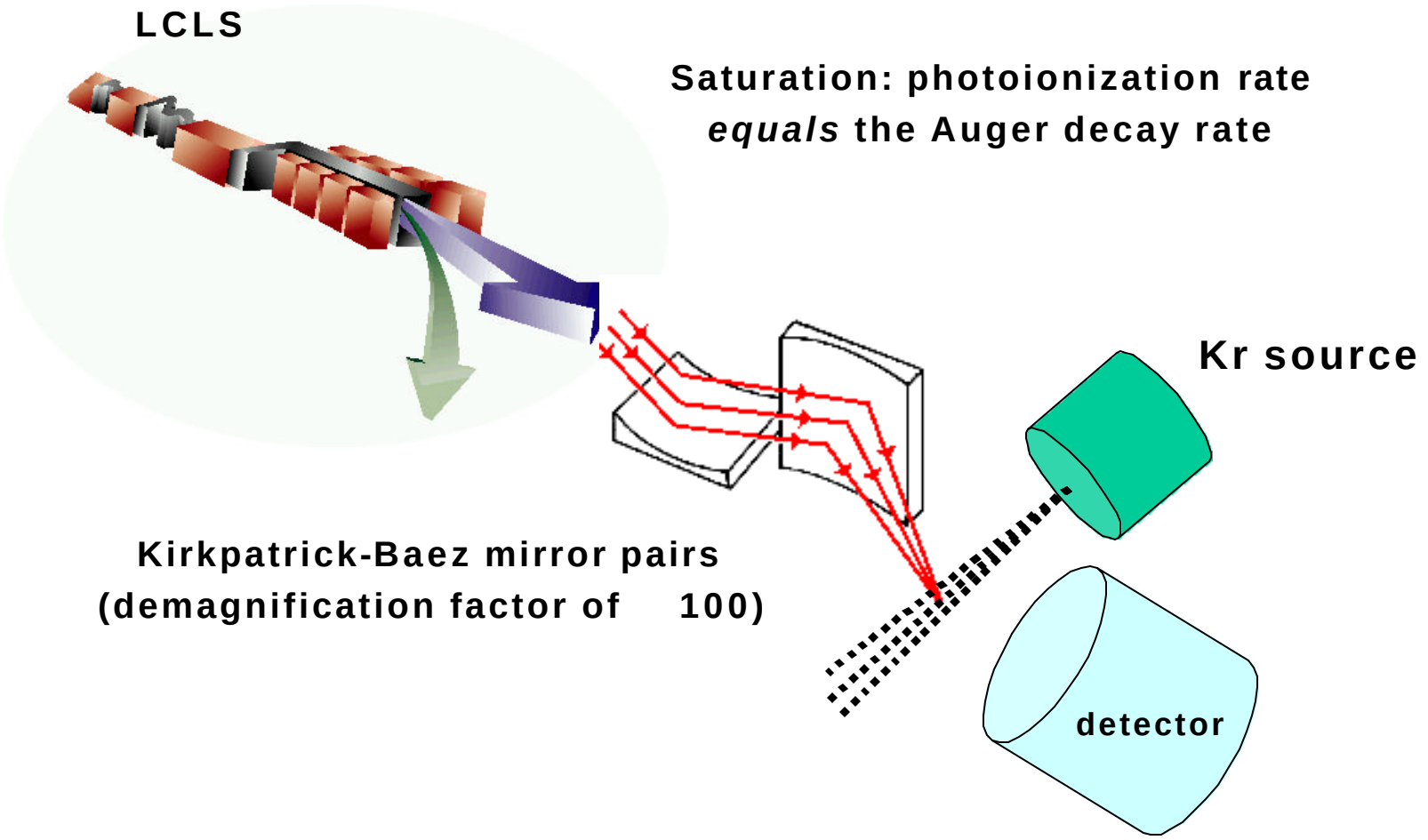
- **The intensity of the LCLS makes numerous processes possible/probable**
  - **For example: (KL) double vacancies are possible:**

$$Ne + h\nu_{>910ev} \rightarrow Ne^{2+*}(KL) + 2e\ (10\%)$$
$$\rightarrow Ne^{3+} + e\ (9.5\%)$$
$$\rightarrow Ne^{4+} + e\ (0.5\%)$$



- **Experimentally background signals of this type can be rejected by electron spectroscopy**
- **Calculationally simulations of the LCLS atom interactions and the core relaxations are necessary**

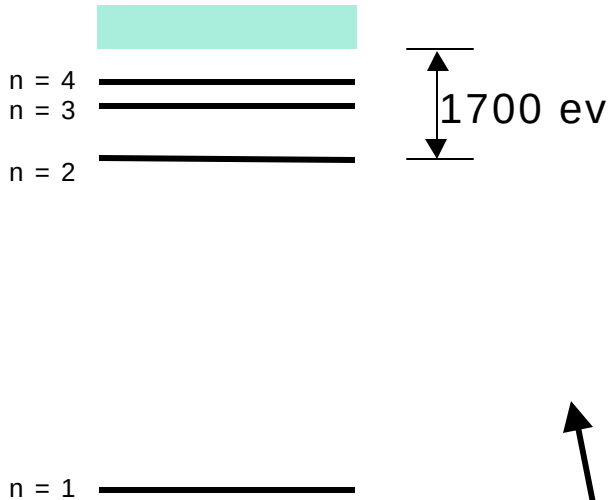
- ***LCLS will allow the study of detailed multiphoton atomic core processes***



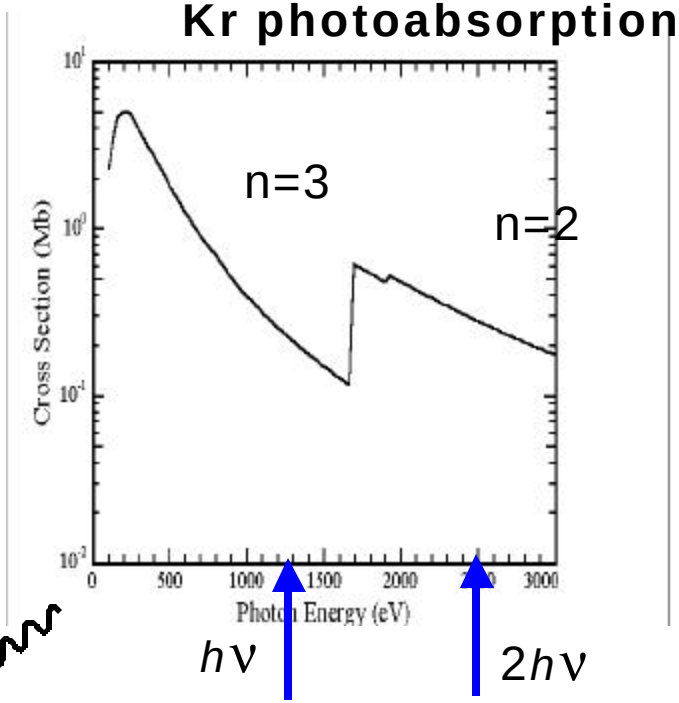
- **Focusing permits observation of two-photon photoabsorption**

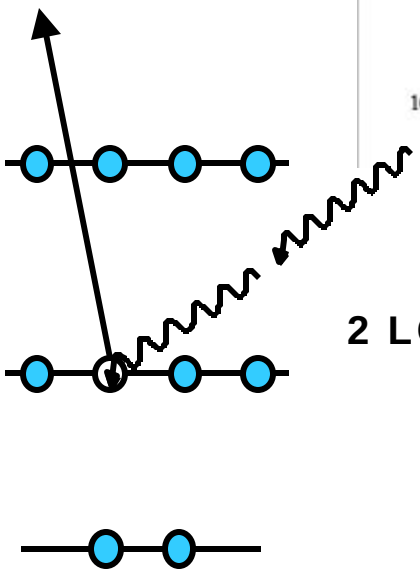
2-Photon Absorption in Kr

Kr energy levels



Kr photoabsorption



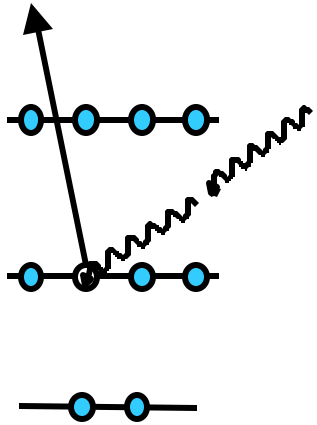


2 LCLS photons with $h\nu_{>850eV}$

Schematic of Kr ionization process

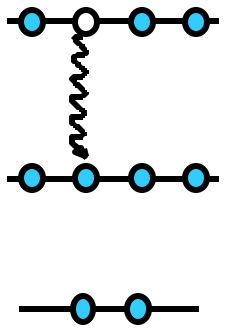
- **Excitation mechanism**

$Kr + 2h\nu_{>850eV} \rightarrow Kr^{+*}(L) + e$



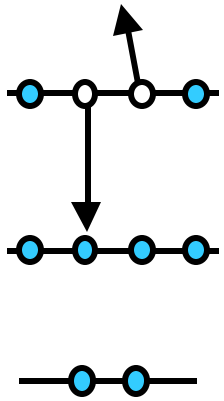
- **Detection signatures:** radiation *and* 2×10^6 1.5 keV e^-/pulse

- $Kr^{+*}(L) + e \rightarrow Kr^{+*}(M) + h\nu_{1.5keV}$



radiation

- $Kr^{+*}(L) + e \rightarrow Kr^{+*}(MM) + e(1.5keV)$



particles

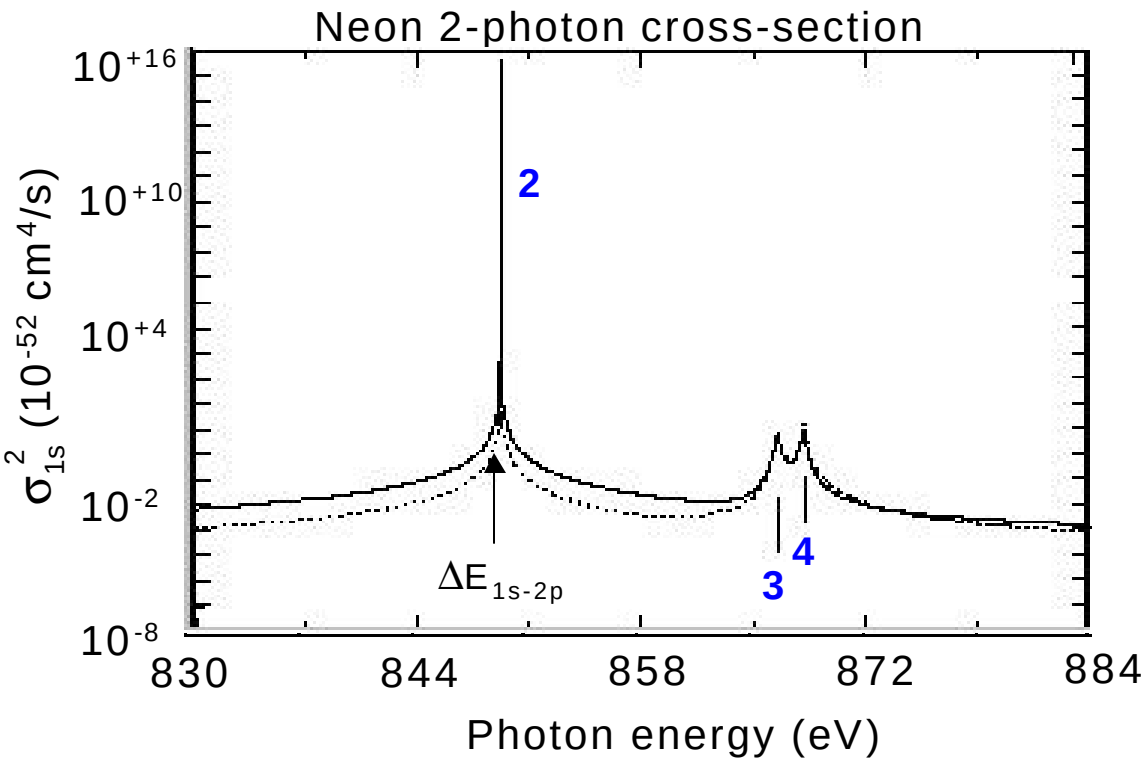
Theory of Resonant 2-Photon Processes Requires Data Only LCLS Can Provide

- Huge enhancements associated with single photon resonances
  S. A. Novikov, J. Phys. B. 33 (2000)
- 2-photon rate exceeds 1-photon rate!
- Rate can be affected by coherence and enhancement due to correlation

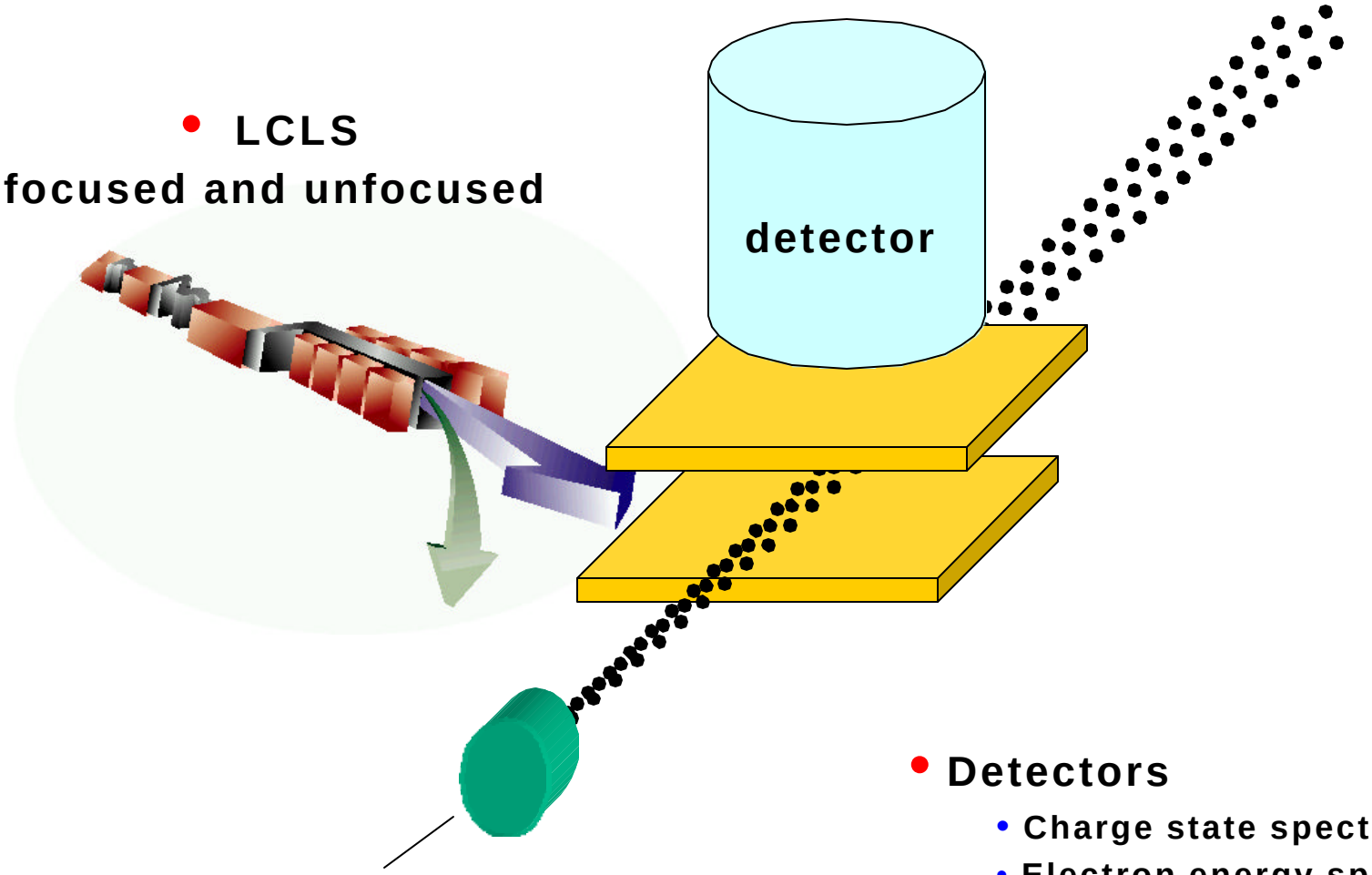
- 2-photon ionization couples to an intermediate state
- $1s^2 2s^2 2p^6$

  $1s2s^2 2p^6 np$

  $1s2s^2 2p^6 + e^-$



- **LCLS**
  focused and unfocused



- **Detectors**
  - Charge state spectrometer
  - Electron energy spectrometer
  - Ion recoil detector

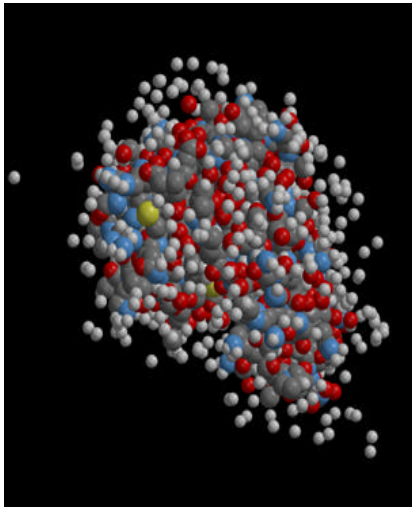
- **cluster source**

- **Xe clusters (10^9 atoms)**
- **Each atom exposed to the unfocused beam will undergo:**
  - **~1 ionization event (10^{31} photons/cm^2/s x10^{-19} cm^2 x10^{-13} s)**
  - **If the ionization will saturate**
- **The dominant relaxation mechanism is Auger decay**
  - **Therefore, each ionized atom creates 2 or more electrons**
- **The cluster becomes a ball of charge with ~ 10^9 ions**
- **Yields fast electrons, fast ions, and x-rays**
- ***Due to x-ray penetration the Coulomb explosion >> conventional lasers***

- **Focusing the LCLS beam to 0.01 mm**
  - **Each atom in the cluster will be classically-ionized nearly 10^4 times over**
- **The atom will continue to ionize, as the ~0.1 fs Auger rates are ~ 1000 times faster than the ionization rate**

- **Thus, each atom will ionize until it strips down to the core level of the initial ionization event**

- **Understanding these processes in detail is central to the imaging of bio-molecular samples**



Lysozyme molecule irradiated by LCLS

Summary: Dual Payoff From Atom Studies

- **Fundamental Science**
  - **LCLS is a unique opportunity to study new fundamental multiple photon x-ray phenomena.**
- **Foundation for all experimental planning**
  - **Ionization and cluster dynamics are central to experimental designs at the LCLS, as well as all next generation x-ray sources.**