

SOME POSSIBLE(?) EXPERIMENTS FOR ALFF

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- Non-sequential double ionization and inner-valence holes (**low field**)
- Photoionization dynamics of aligned molecules

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Non-sequential double ionization

- Much current interest in very high fields, tunneling ionization, the rescattering model, etc.
- Most work has been done in the IR and visible region. What happens in the high-frequency regime?
- Tunneling ionization or multiphoton ionization?

MULTIPHOTON OR TUNNELING?

The ionization mechanism depends on the adiabaticity, or Keldysh, parameter, which is given by:

$$\gamma = \left(\frac{I_p}{2U_p} \right)^{1/2}$$

where I_p is the ionization potential and U_p is the ponderomotive potential. This is essentially the ratio of the tunneling time to the optical period.

For the parameters of ALFF (100 nm, 200 μ J, 300 fs, 10 μ m diameter spot), γ is ~ 2.5 , and thus clearly in the multiphoton regime.

This corresponds to a relatively low-field regime.

WHAT REGIME IS ALFF?

OPTIMISTICALLY(?)

For the parameters of ALFF (100 nm, 200 μJ , 300 fs, 10 μm diameter focal spot), γ is ~ 2.5 , and thus clearly in the multiphoton regime.

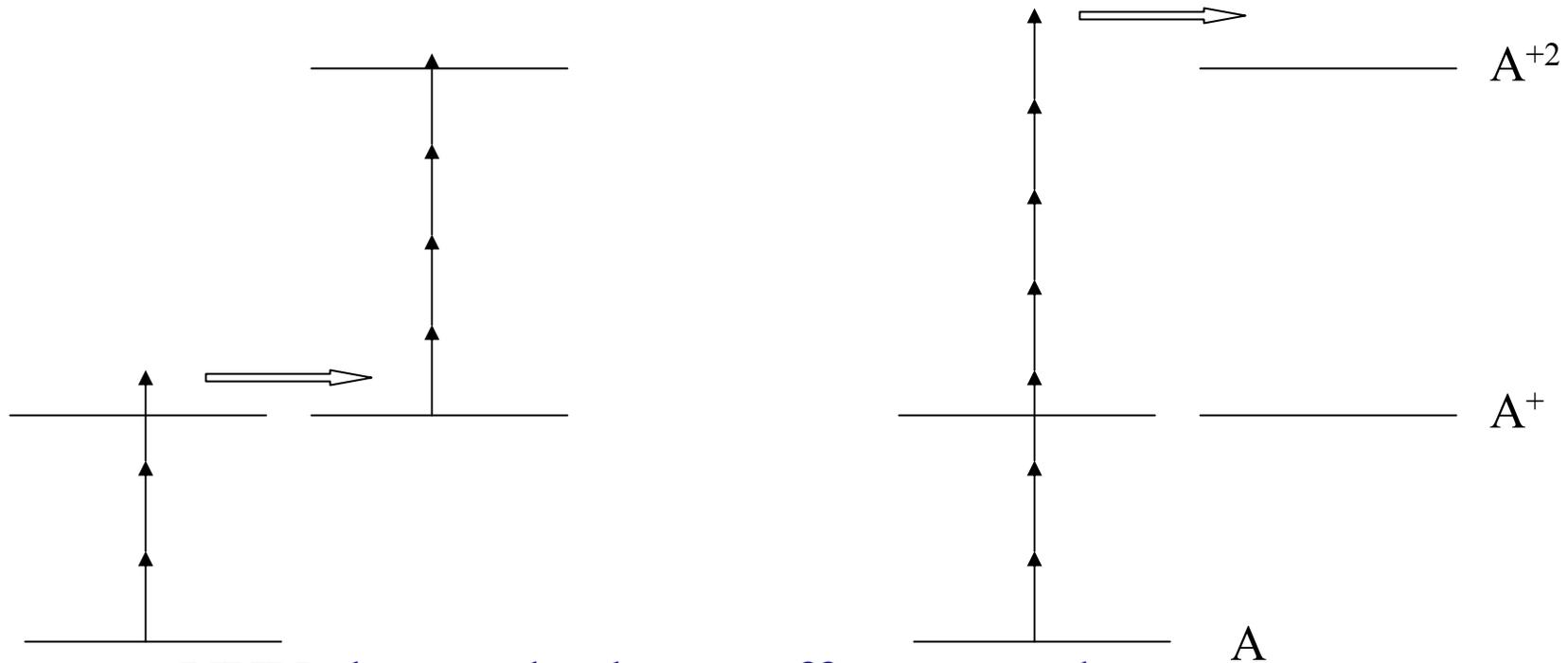
This corresponds to a relatively low-field regime.

In this case, resonances should be important, but of course the ability to see them will depend on their widths and the VUV bandwidth.

Non-Sequential Double Ionization and Multiphoton Production of Inner-Valence Holes

- Enhance the likelihood of non-sequential double ionization?
- Create inner-shell holes through a multistep process?
- Enhance the likelihood of Auger processes and final state interactions?

The problem with non-sequential double ionization in the multiphoton regime is that sequential processes generally dominate.



VUV short-pulse lasers offer some advantages

- Fewer photons involved
- Fast enough to compete with ionization
- High peak intensities

Kr is an interesting system for non-sequential ionization at relatively low pulse energies (i.e., in the multiphoton regime)

- Resonant excitation minimizes the need for high peak intensities
- Potential for isolated core excitation allows production of promising doubly excited states
- Ionization thresholds and resonances in the right places

Resonant Three-Photon Ionization of Kr at 112830.0 cm^{-1} (88.6 nm)

One photon at a time...

Resonant Three-Photon Ionization of Kr at 112830.0 cm^{-1} (88.6 nm)

Photon 1: $4s^2 4p^6 \rightarrow 4s^2 4p^5 (\text{Kr}^+ \text{}^2\text{P}_{3/2}) 35s/d$

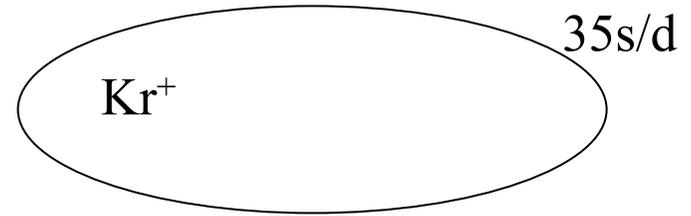
Kr^+

35s/d

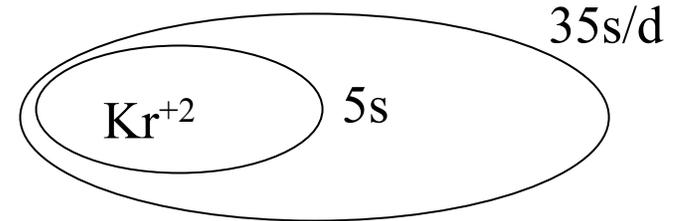
The ALFF pulse will actually prepare a wavepacket
resulting from the superposition of Rydberg
states with a range of n values

Resonant Three-Photon Ionization of Kr at 112830.0 cm^{-1} (88.6 nm)

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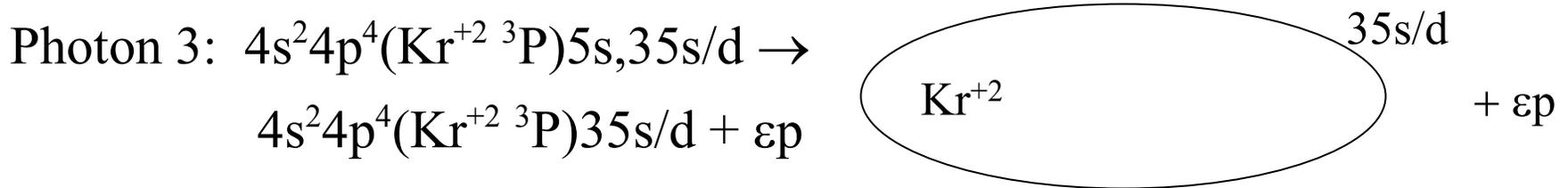
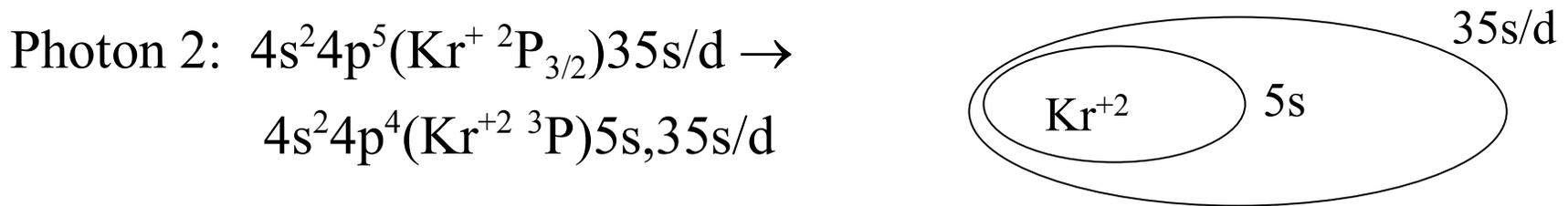
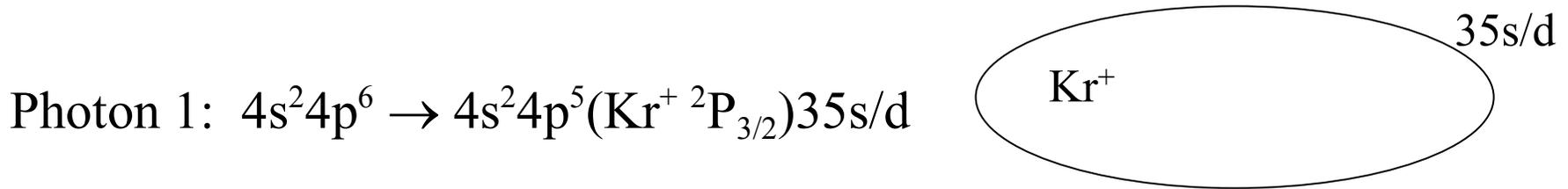


Photon 2: $4s^2 4p^5 (\text{Kr}^+ \text{}^2\text{P}_{3/2}) 35s/d \rightarrow$
 $4s^2 4p^4 (\text{Kr}^{+2} \text{}^3\text{P}) 5s, 35s/d$

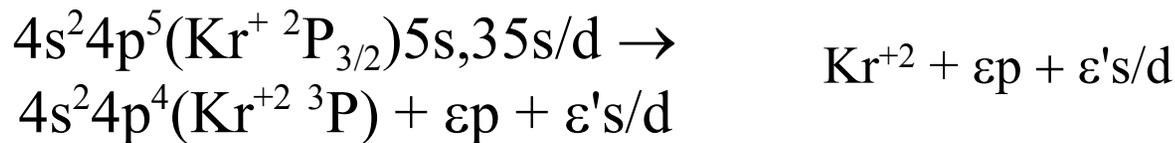


The photoionization cross section for the $n = 35$ state is very small at 88.6 nm, and it is much more likely that the core will absorb the second photon. The wavelength was chosen to match the core transition.

Resonant Three-Photon Ionization of Kr at 112830.0 cm^{-1} (88.6 nm)



and, as a satellite



Photoelectron Spectroscopy

- Direct ionization leaving Kr^+ in an $n \sim 35$ Rydberg state
- Shake up and final state interactions to give $\text{Kr}^{+2} + \epsilon_s + \epsilon_p$
- Imaging would give angular distributions
- Selective Field Ionization to characterize Kr^+ state distribution

Total three-photon energy is ~ 3.4 eV above the double ionization threshold

Disclaimers

- The single-photon cross section to excite the $(^2P_{3/2})35s/d$ Rydberg states is not huge
- The transition at 112830.0 cm^{-1} is $^2P \rightarrow ^4P$, but it is quite strong in the emission spectrum of Kr^+
- It would be nice if the bandwidth was somewhat smaller (seeding?)
- Better shot-to-shot regularity of spectral envelope would also be nice

With a second tunable laser...
(not necessarily ultrafast or vuv)

- More flexibility in first excitation step
- Possible extensions to inner-valence shell
- Converting valence shell excitation into inner-valence excitation

The $\text{Kr}^+ 4s^2 4p^5 \rightarrow 4s^1 4p^6$ transition is at $109002.06 \text{ cm}^{-1}$ (91.7 nm)

1. First laser excites $4s^2 4p^5 n l$ via a two-photon transition
2. First ALFF photon excites $4s^2 4p^5 n l \rightarrow 4s^1 4p^6 n l$
3. Second ALFF photon puts system above the double ionization threshold, but what actually happens?

Continuations

- Shake up and shake off as a function of the initial excited state quantum numbers (n, l , etc.)
- PCI when the slow electron is bound
- Extensions to double ionization and inner-valence shell excitation in molecules
- New flexibility to probe selected high-energy doubly excited states