Selective Strong-Field Excitation and Ionization with Short Laser Pulses

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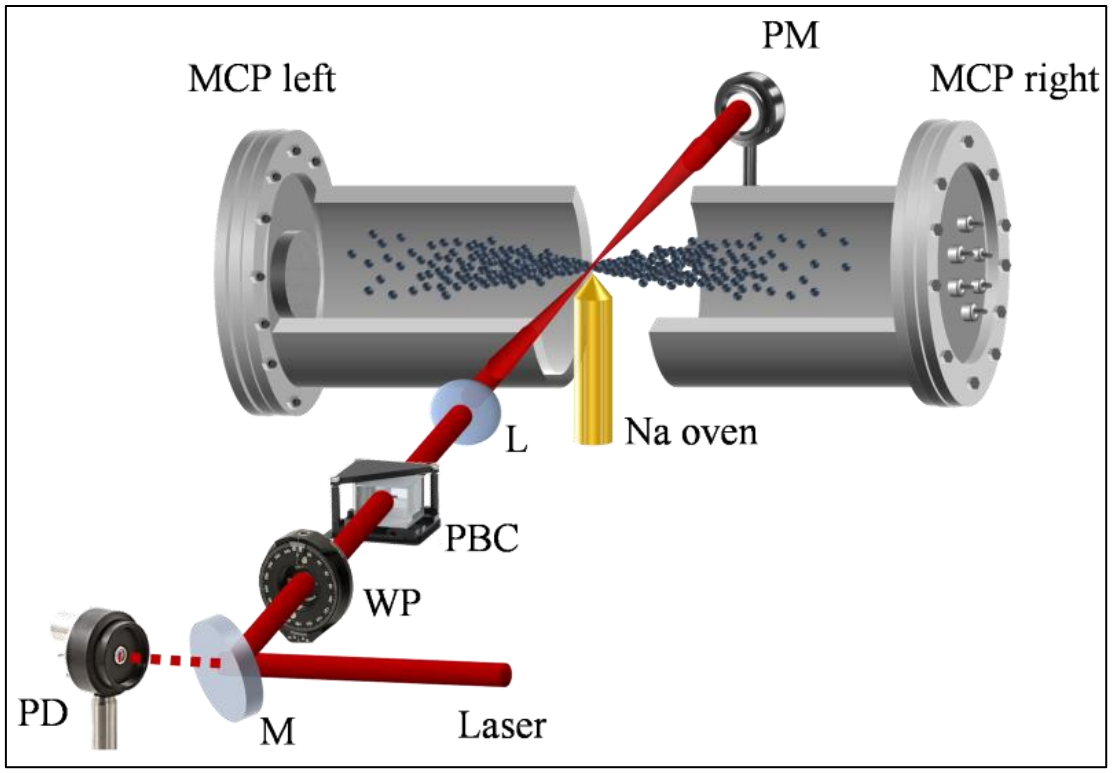
Introduction

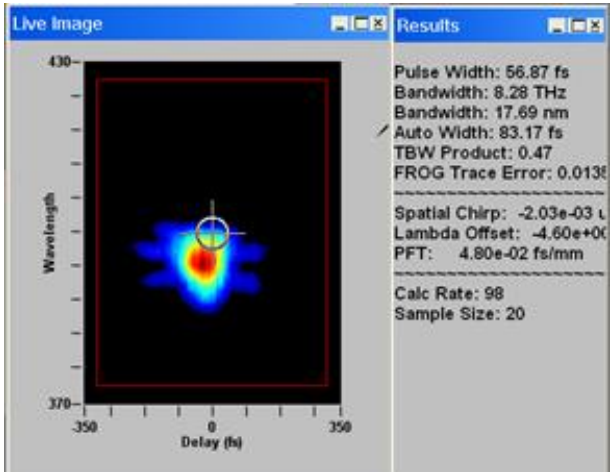
… **In** this research, we:

1. Experimentally demonstrate coherent control of atomic energy level population in sodium vapor.
2. Selectively excite and ionize a sodium Rydberg level with 83% efficiency using high intensity laser pulses.
3. Theoretically describe atomic excitation in high intensity laser fields ($> 10^{12}$ W/cm^2) with a model referred to here as Resonance Sampling.
4. Directly measure the ponderomotive shift of the continuum.
5. Identify Freeman resonances as the dominant means of atomic excitation in high intensity laser fields

Experimental Apparatus

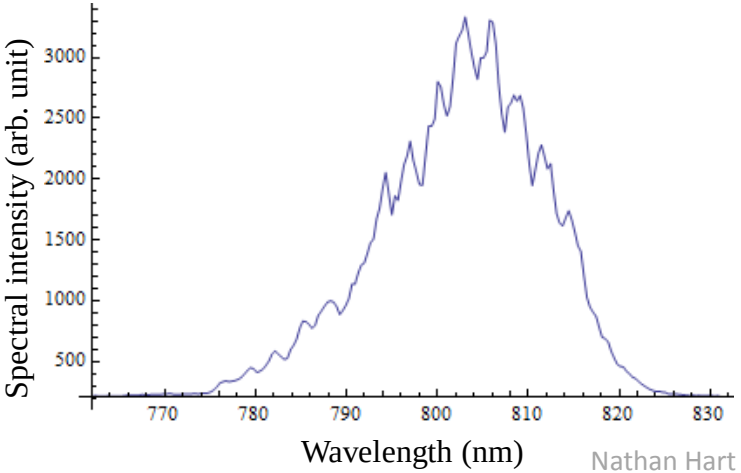


- Pulses from a Ti:Sapphire oscillator are amplified by a regenerative amplifier.
- 0.9 mJ per pulse
- Pulse spectrum centered at ~800nm and a transform limit duration of 57 fs.

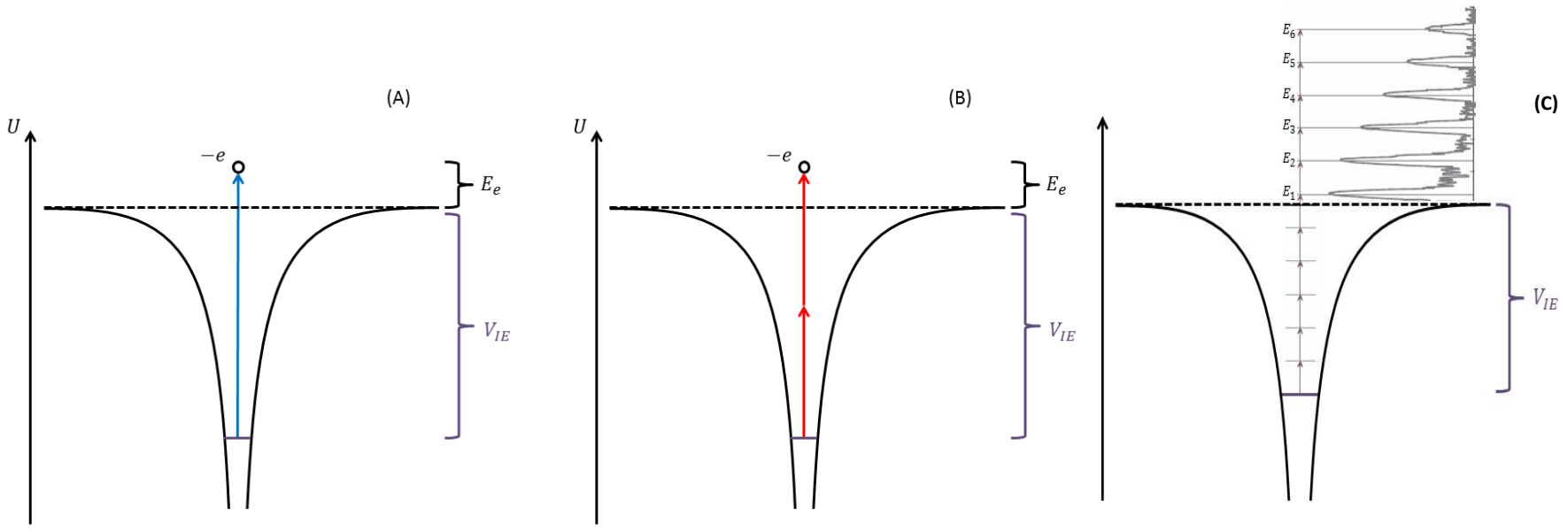


Laser Pulse Spectrum

L = lens
M = mirror
MCP = microchannel plate detector
Na oven = sodium oven
PBC = polarizing cube
PD = photodiode
PM = power meter
WP = half-wave plate



Multiphoton Ionization



Photoelectric effect

Multiphoton Ionization

Above Threshold Ionization (**ATI**)

n photons ionize an atom:

$$n\hbar\omega + A \rightarrow e^- + A^+$$

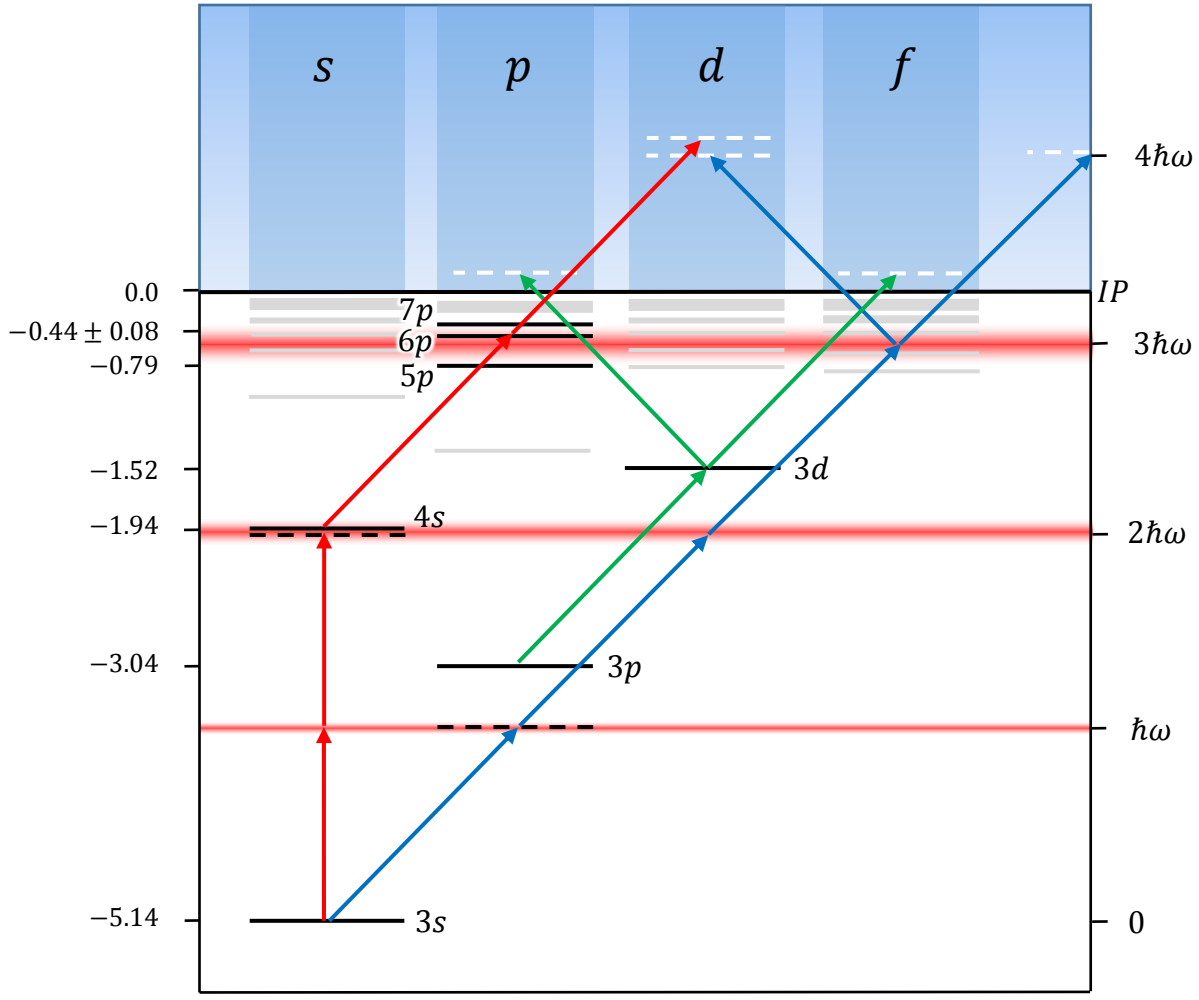
Kinetic energy of the electron:

$$KE = n\hbar\omega - V_{IE}$$

Multiphoton condition (from Keldysh theory): $\gamma \gg 1$

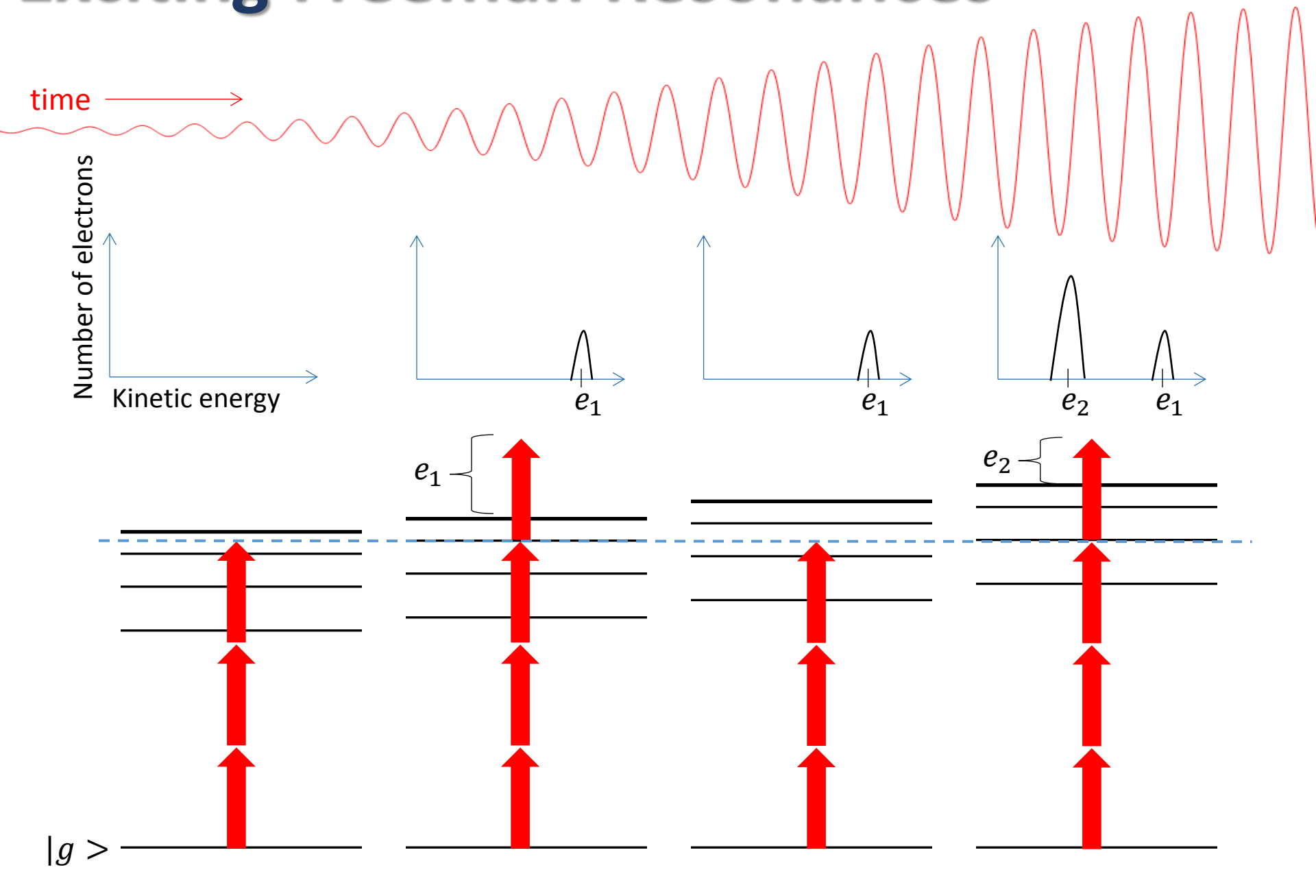
Ionization probability from perturbation theory: $P(I) \propto I^n$

Sodium (Na) Energy Level Diagram



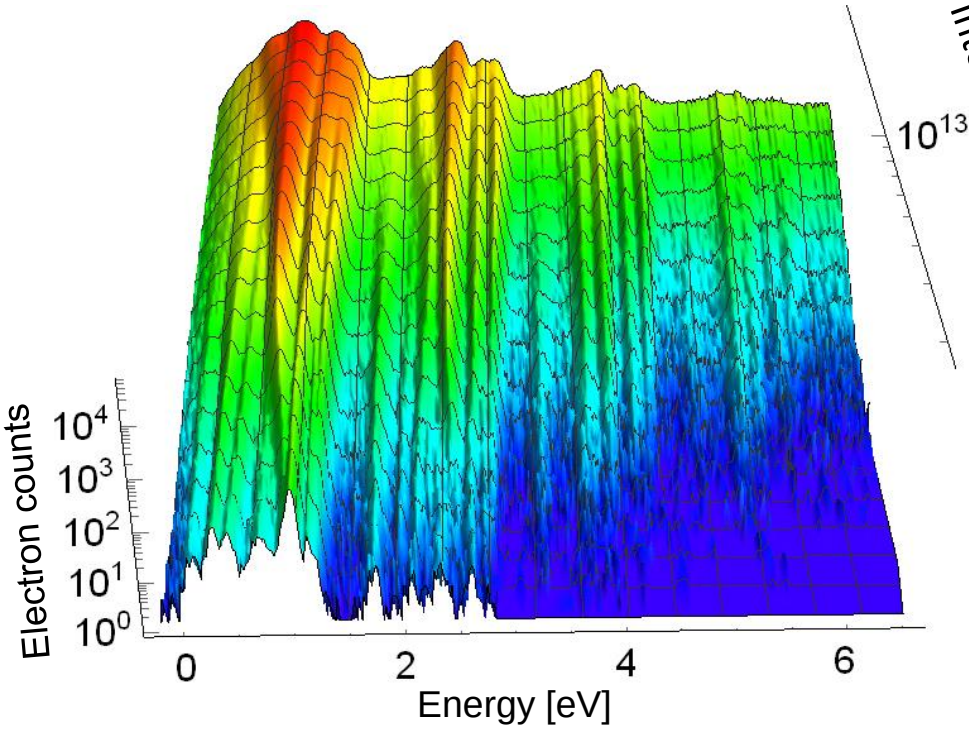
The red path (dominant):
$3s \rightarrow\rightarrow 4s \rightarrow (5p, 6p, 7p)$
The blue path:
$3s \rightarrow\rightarrow\rightarrow (4f, 5f, 6f)$
The green path:
$3s \rightarrow 3p \rightarrow 3d$

Exciting Freeman Resonances

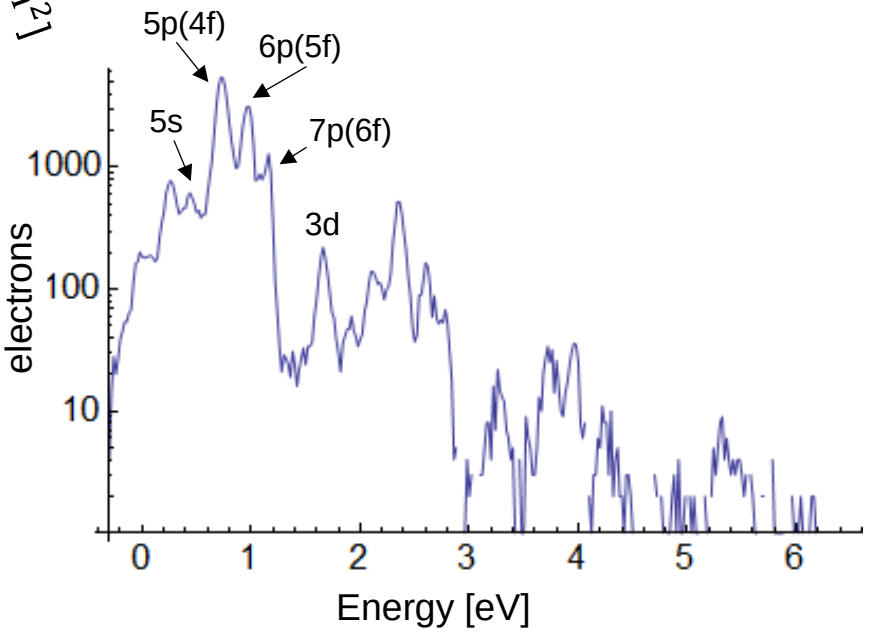


ATI Spectrum of Sodium Vapor

~ 150 fs negatively chirped pulses (Na pressure $\sim 10^{-3}$ mbar, $\varphi' = -2800\ \mathrm{fs}^2$)

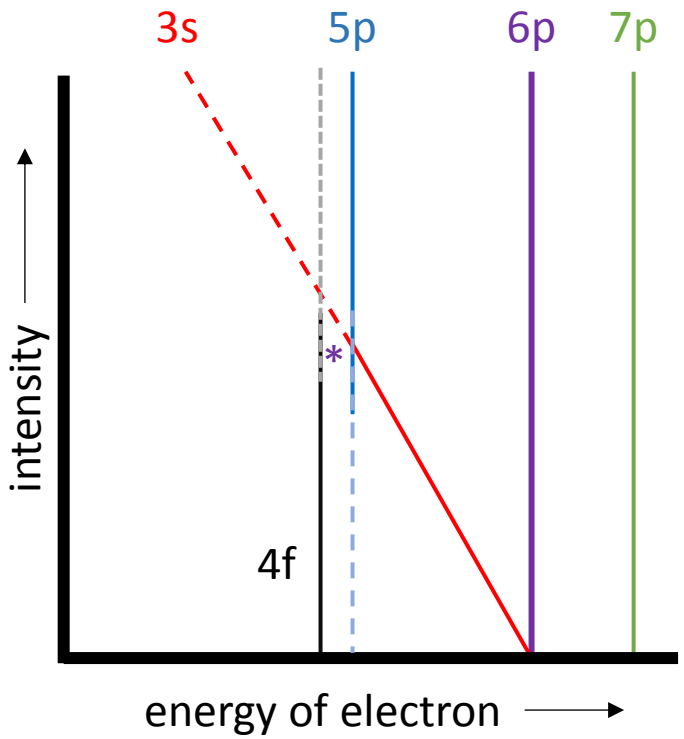


$I_0 = 4.5 \times 10^{12}\ \mathrm{W/cm^2}$

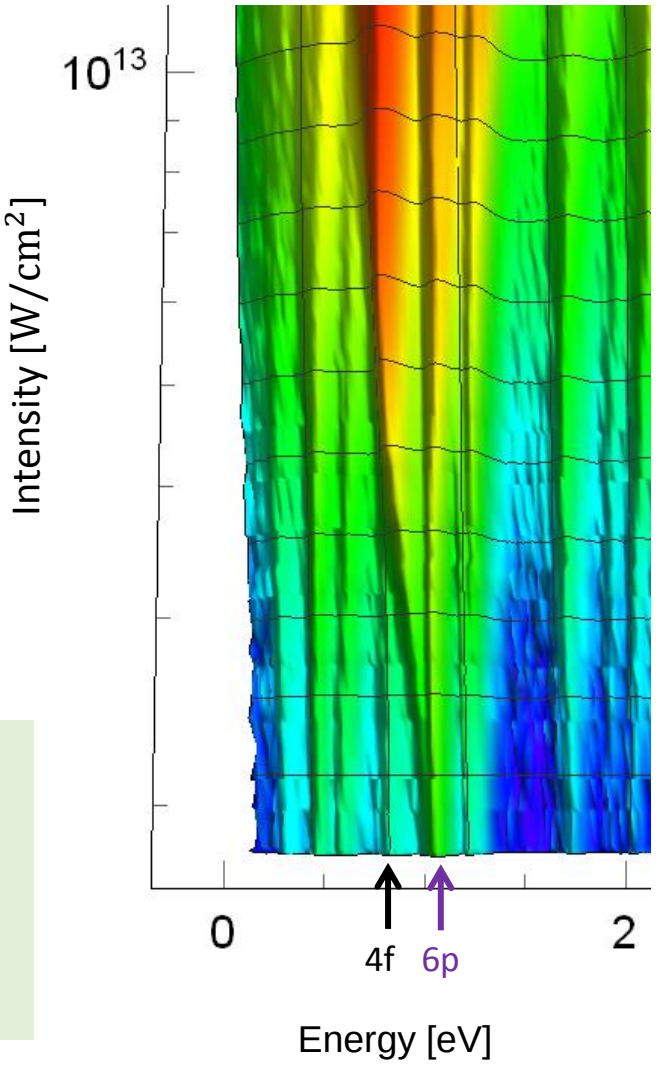


- At low intensities, the peak representing ionization from the 3s ground state is degenerate with the 6p(5f) peak.
- As the intensity increases, this 3s peak shifts to smaller energies because of the upward ponderomotive shift of the continuum.

Reading an Intensity Scan



Zoom In of the First ATI Peak

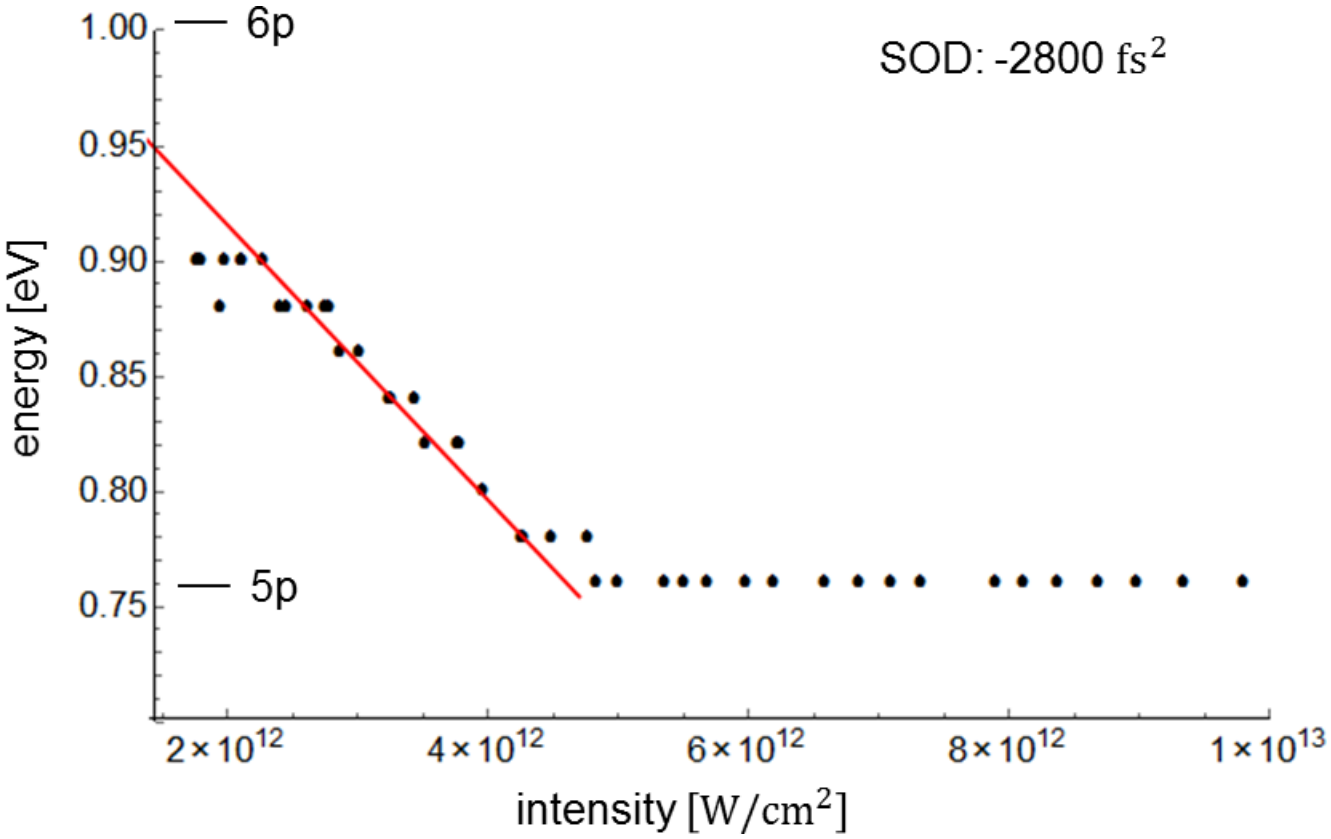


Relative to the continuum and Rydberg states (i.e. 4f, 5p, 6p and 7p), the 3s ground state shifts to lower energies by an amount equal to the ponderomotive energy U_p.

$$E_{6p} - E_{3s} = U_p$$

Shift of the 3s Peak

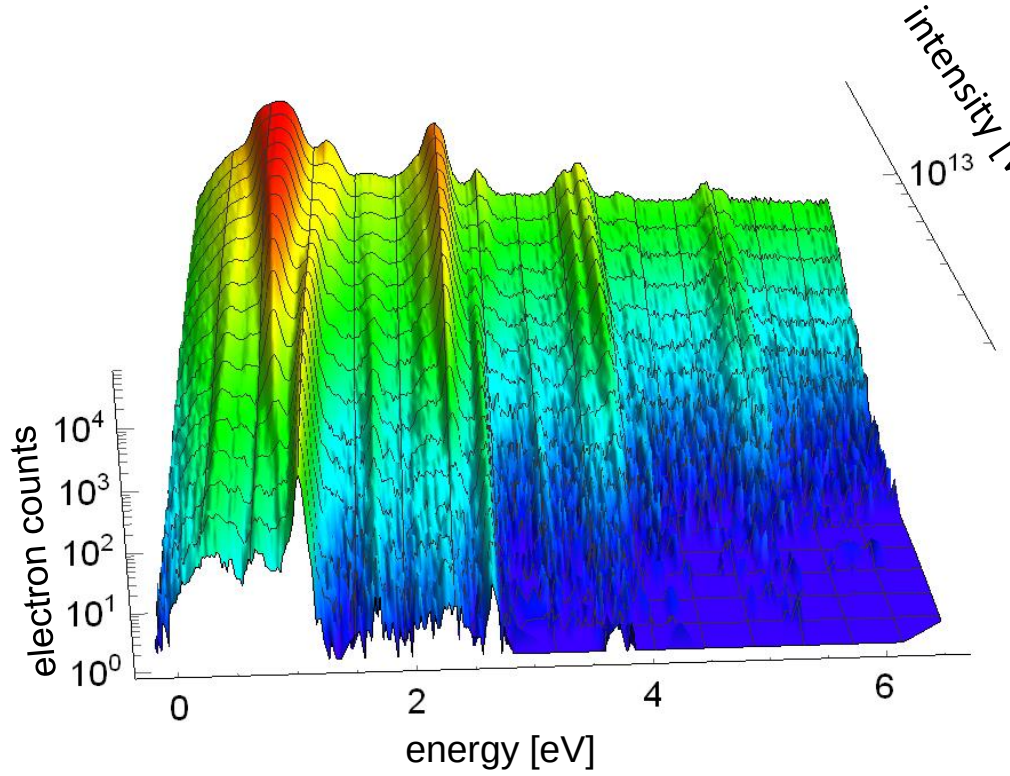
Energy of 3s ionization peak vs. intensity



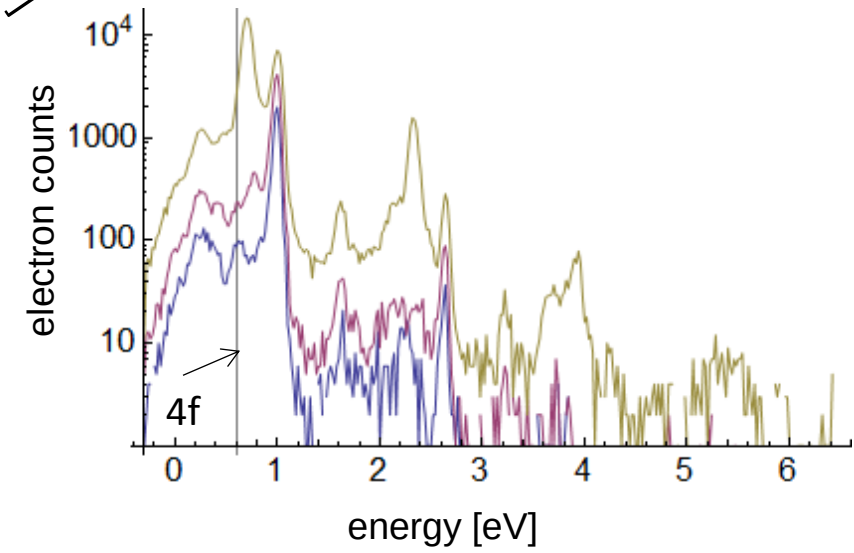
- The 3s peak position decreases linearly with increasing intensity and is used to calibrate the laser peak intensity in the focus.
- At 4.68×10^{12} W/cm^2 (theoretically predicted), the 3s peak merges with the 5p peak.

ATI Spectrum of Sodium Vapor

~ 150 fs positively chirped pulses (Na pressure $\sim 10^{-3}$ mbar)

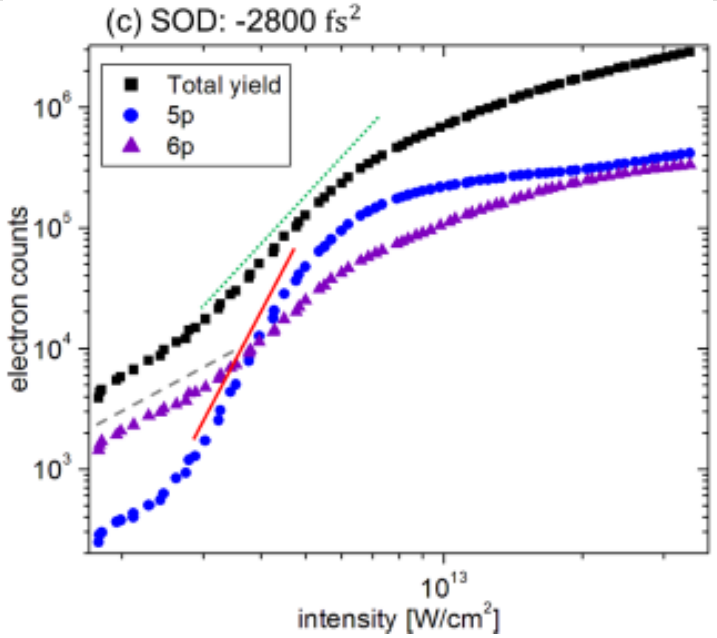


gold: $6.7\times10^{12}\ W/cm^2$
purple: $3.5\times10^{12}\ W/cm^2$
blue: $2.4\times10^{12}\ W/cm^2$



- The shift of 3s peak is also present for positive chirp as well.
- Positive chirp ATI is missing the 7p(6f) peak.

What About Chirp Dependence?



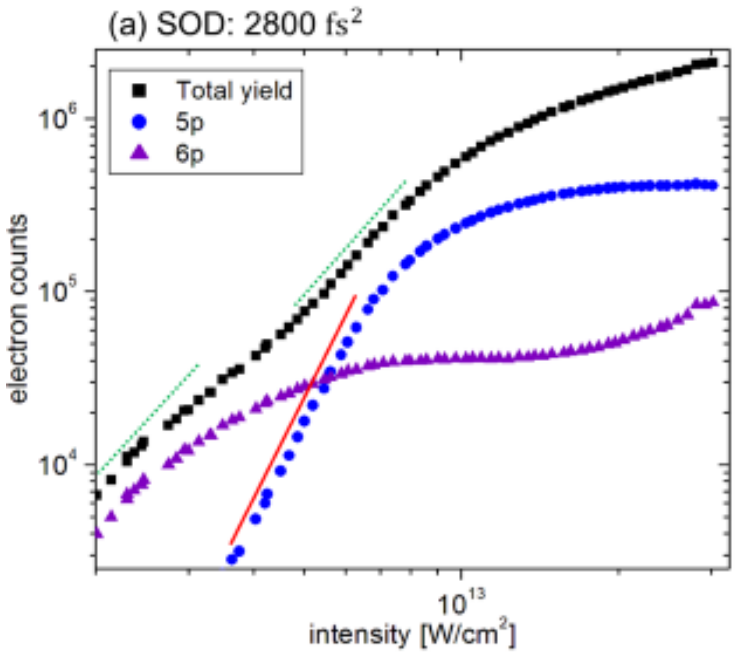
Intensity scans for three chirp values

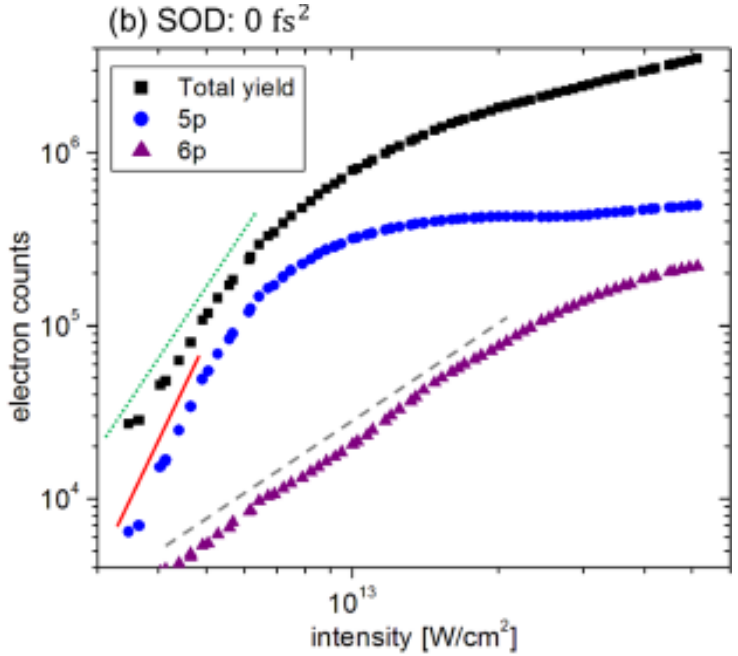
·····Dotted green line has a slope of 4

—Solid red line has a slope of 6

- -Dashed gray line has a slope of 2

- The 6 slope process is independent of chirp (occurring for positive, negative and zero chirp).
- Moreover, the slope-6 process is hidden within the total yield.





Collective Action?

Let's consider collisional mechanisms for increased electron yields. Collective (multi-particle) effects also show enhanced (N_i^2) absorption/ionization dependence, where N_i is the number of atoms in some initial state i.

Collective effects include (non-exhaustive):

1. Penning Ionization
2. Electron-Atom collision

Let N_{4s} be the total number of atoms in state 5p.

Diagonal Density Matrix Element:

$$\rho_{4s} = W_{3s\to 4s} \cdot \tau$$
$$\approx \left| \frac{1}{\hbar^2} \sum_n \frac{\mu_{4s,n}\mu_{n,3s}E^2}{(\omega_{n,3s} - \omega)} \right|^2 2\pi\delta_L(2\omega = \omega_{4s,3s}) \cdot \tau$$
$$= \sigma_{3s\to 4s} I^2$$

At least 2 photons are needed to transfer population from state $4s$ to the continuum

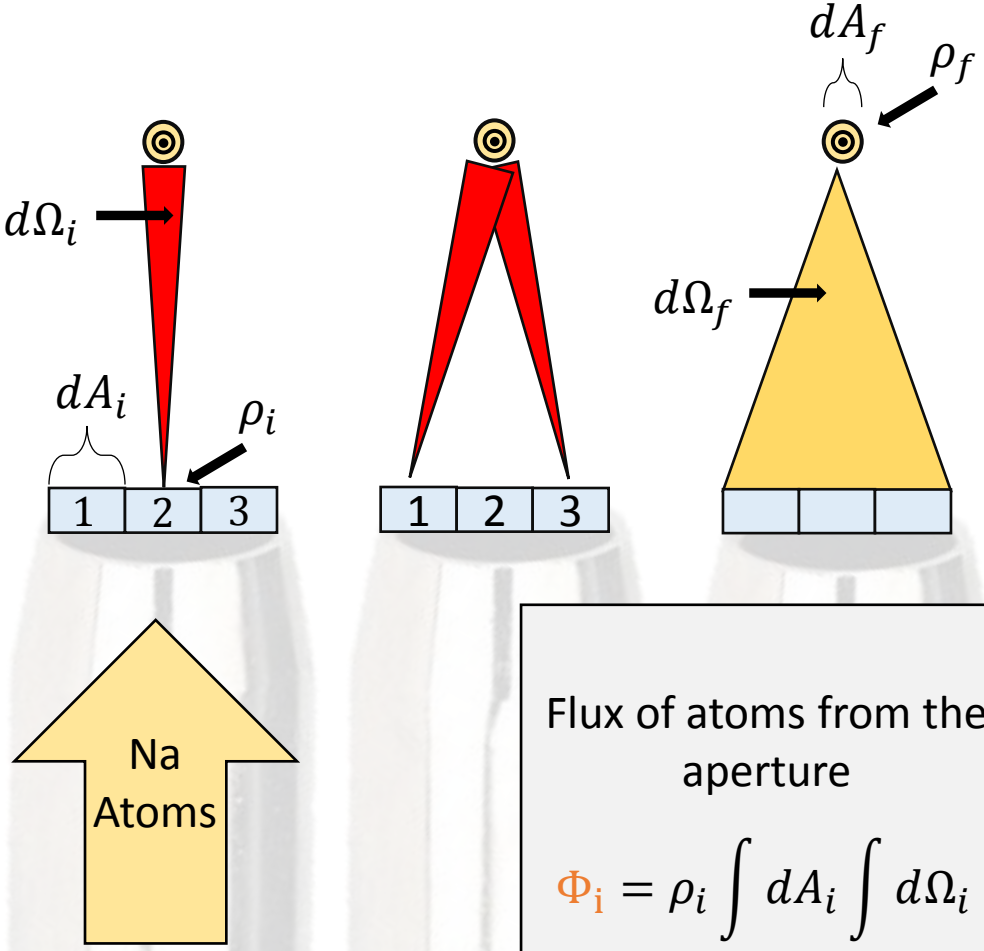
$$N_{4s} = N_{total} \cdot \rho_{4s}$$
$$= N_{total} \cdot \sigma_{3s\to 4s} I^2$$

Penning Ionization

$$N_{ion} \propto N_{4s}^2 I^2$$
$$\propto N_{total}^2 \cdot \sigma_{3s\to 4s}^2 I^6$$

Appendix A

Calculating the sodium density using Lambert's Cosine Law



ρ_i = initial density
ρ_f = initial density
$d\Omega$ = solid angle = $\sin(\theta)d\theta\, d\phi$
dA = differential area of aperture
$\Phi_i = \Phi_f$ = atomic flux (assuming constant velocity)

Flux of atoms from the aperture

$$\Phi_i = \rho_i \int dA_i \int d\Omega_i$$

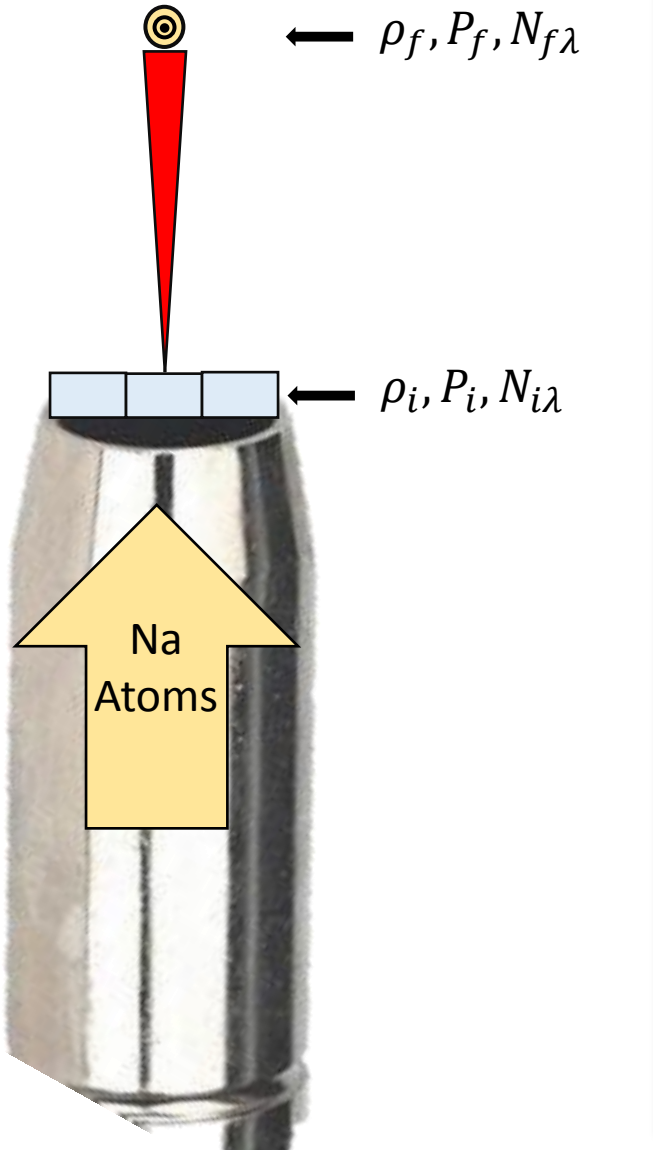
$$= \rho_i A_i\, 2\pi\left(1 - cos(\theta_i)\right)$$

Density of atoms reaching the target

$$\rho_f = \frac{\Phi_i}{\int dA_f \int d\Omega_f}$$

$$= \frac{\Phi_i}{\left(A_f\, 2\pi\left(1 - cos(\theta_f)\right)\right)}$$

Nozzle Characterization for Oven Temperature T=256°C



Ideal Gas Law: $PV = NkT$

ρ_i = initial density = 1.98×10^{14} cm^{-3}
ρ_f = target density = 5.17×10^{13} cm^{-3}

$P_i = 1.45 \times 10^{-2}$ mbar
$P_f = 3.79 \times 10^{-3}$ mbar

At the target:

Mean free path of Na (atom-atom) = 2.3mm
Beam waist = 10μm
Beam Rayleigh range = 0.79mm

Thermal velocity of Na atoms: 349.3 m/s
Time between collisions: 72.2 µs
Energy Level Lifetimes: $\tau_{4s} = 37.9$ ns
$\tau_{5p} = 0.37$ µs
$\tau_{6p} = 1.56$ µs

- Atomic or electron collisions not likely.

Appendix B

Sodium parameters

Width of 6th order peak:
$\Delta E = 0.127$ eV

Ionization width of 6th peak:
$\Delta t = 2.5$ fs

Na crossection:
$\sigma_{5p} = 2.8 \times 10^{-19}\ \mathrm{m}^2$

Na Dimer percentage at $\mathrm{T} = 256$ ℃:
$\mathrm{Na}_2 \sim 2\%$

Ionization potential:
$\mathrm{E_I} = 5.139$ eV

Over the Barrier (OTB) Intensity:
$I_{OTB} = 2.78 \times 10^{12}\ \mathrm{W/cm^2}$

Keldysh tunneling parameter for OTB Intensity:
$\gamma = 3.94$

The Absence of Tunneling Ionization

Bohr model for hydrogen-like atoms

Solve for the orbital velocity

$$\vec{F} = m\vec{a}$$
$$= -\frac{ke^2}{r^2} = -\frac{mv^2}{r} \longrightarrow v = e\sqrt{\frac{k}{mr}}$$

Solve for the radius r

$$U(r) = -\frac{ke^2}{r} = e\mathrm{E}_{5\mathrm{p}} \longrightarrow r = \frac{ke}{|\mathrm{E}_{5\mathrm{p}}|}$$

Orbital period of level 5p

$$T_{5p} = \frac{2\pi r}{v} = \frac{2\pi\sqrt{kmr}}{|\mathrm{E}_{5\mathrm{p}}|} = 2\pi k\sqrt{\frac{me}{|\mathrm{E}_{5\mathrm{p}}|^3}}$$
$$= 4.07 \text{ fs}$$

Energy levels:
$\mathrm{E}_{3\mathrm{s}} = -5.139$ eV
$\mathrm{E}_{5\mathrm{p}} = -0.794$ eV

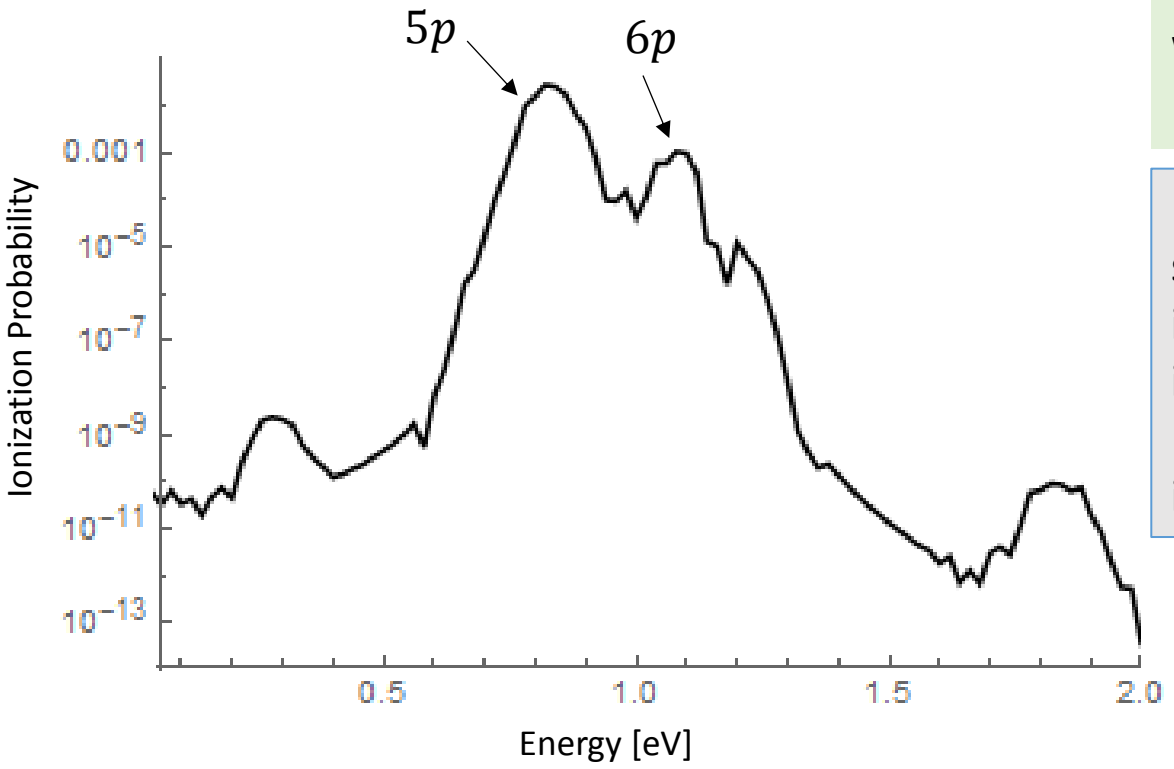
Over the Barrier (OTB) Intensity:
$I_{OTB} = 2.78 \times 10^{12}$ W/cm^2

Keldysh tunneling parameter for OTB Intensity:
$\gamma = 3.94$

Laser period:
$T_L = 2.7$ fs

- The Keldysh parameter remains in the multiphoton regime.
- The long orbital periods may indicate that the tunneling time is longer than the laser period.
- The laser-field induced ionization of sodium atoms remains a multiphoton process up to the over-the-barrier intensity.

1D Na TDSE Simulation



Intensity = 1.7×10^{12} W/cm^2
Wavelength = 800 nm
Pulse duration = 53 fs

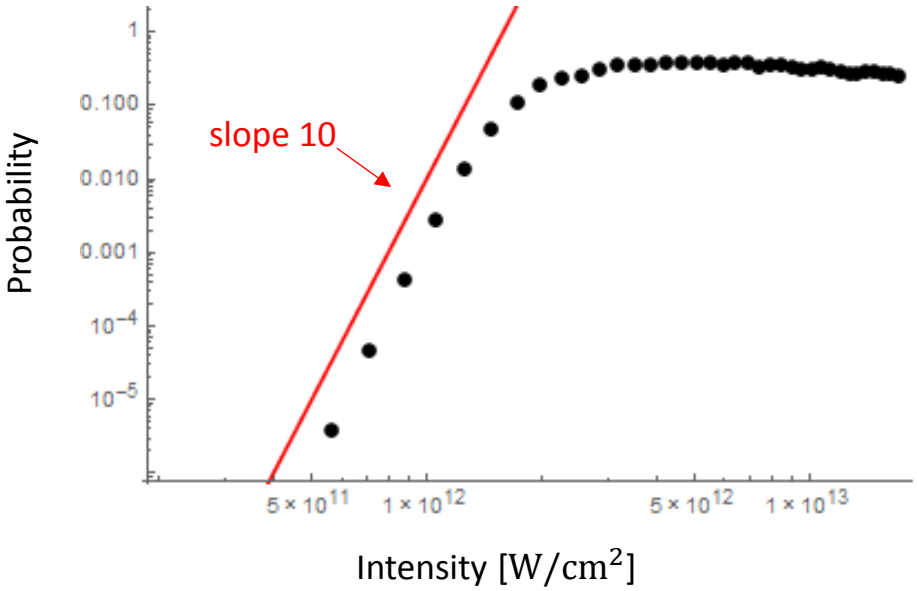
1D TDSE produces even and odd states without (S,P,D,F,G) information. Even states are interpreted as S and odd states as P. Higher angular momentums are ignored.

- Program solves the 1D time dependent Schrodinger equation in the length gauge.
- Atom is represent by 1D soft-core Coulomb potential.

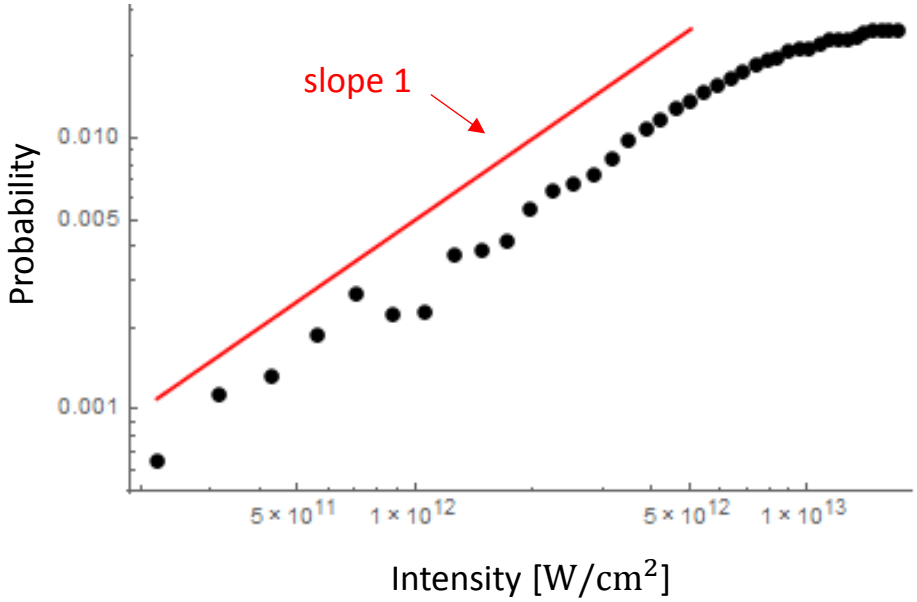
1D Na TDSE Simulation

Intensity Scan (log-log plots)

5p integrated



6p integrated



- Fermi's golden rule predicts that both slopes (5p and 6p) should be of order 4.
- PPT theory predicts an overall decrease in the ion yield slope to roughly order 2.
- Linewidth Sampling selectively excites 5p and attenuates all other higher P states.

Multiphoton Excitation

1. Cross section for n photon absorption:

$$\sigma_n = \left(\frac{2}{c\epsilon_0\hbar}\right)^n \left| \sum_{a_1 \cdots a_n} \frac{\langle f|x|a_{n-1}\rangle \cdots \langle a_1|x|g\rangle}{(\omega_{n-1} - (n-1)\nu) \cdots (\omega_1 - \nu)} \right|^2 2\pi\rho(\omega_f - n\nu)$$

2. Absorption rate:

$$W_n(t) = \sigma_n I(t)^n$$

3. Rate integration :

$$s = \int_{-\infty}^{\infty} W_n(t)\, dt$$

The Lorenztian Density of States

- Defines linewidth for energy level ω_f
- γ_f = decay rate of level f
- ν = instantaneous laser frequency

$$\rho(\omega_f - n\nu) = \frac{1}{\pi} \frac{\left(\frac{\gamma_f}{2}\right)}{(\omega_f - n\nu)^2 + \left(\frac{\gamma_f}{2}\right)^2}$$

4. Absorption probability (see slide Appendix C):

$$P_n = \frac{1 - e^{-2s}}{2}$$

Appendix C

Integrating Rate equations

1. The classical rate equations:

$$\frac{dN_a}{dt} = -W_n N_a + W_n N_b = -W_n(N_a - N_b)$$

$$\frac{dN_b}{dt} = -W_n N_b + W_n N_a = W_n(N_a - N_b)$$

Notation:

$$n\ photon\ rate \rightarrow W_n \equiv W_n(t)$$
$$Population\ in\ state\ a \rightarrow N_a \equiv N_a(t)$$
$$Population\ in\ state\ a \rightarrow N_b \equiv N_b(t)$$

2. Subtract the rates:

$$\frac{d}{dt}(N_a - N_b) = -2W_n(N_a - N_b) \longrightarrow (N_a - N_b) = (N_{a0} - N_{b0})e^{-2W_n t}$$

Identity relations:

$$N_a = 1 - N_b$$

$$N_{a0} = 1$$
$$N_{b0} = 0$$

$$(1 - 2N_b) = e^{-2W_n t}$$

$$N_b = \frac{1 - e^{-2W_n t}}{2}$$

Average over the bandwidth

1. Modify the multiphoton absorption by:

- Setting: $\overline{\omega_f} = \omega_f + U_p$
- Replacing: $\omega_f \rightarrow \overline{\omega_f}$

2. Define the new density function

$$\rho\left(\overline{\omega_f} - n\nu\right) = \left(\frac{1}{\pi}\right)\frac{\left(\frac{\gamma_f}{2}\right)}{\left(\overline{\omega_f} - n\nu\right)^2 + \left(\frac{\gamma_f}{2}\right)^2}$$

3. Define the normalized Bandwidth function

$$a = \frac{nLog(2)}{\left(\frac{\tau}{2}\right)^2}$$

$$f(\nu) = \frac{1}{\sqrt{4a\pi}} e^{-n^2(\nu-\nu_0)^2/(4a)}$$

4. Average over the bandwidth

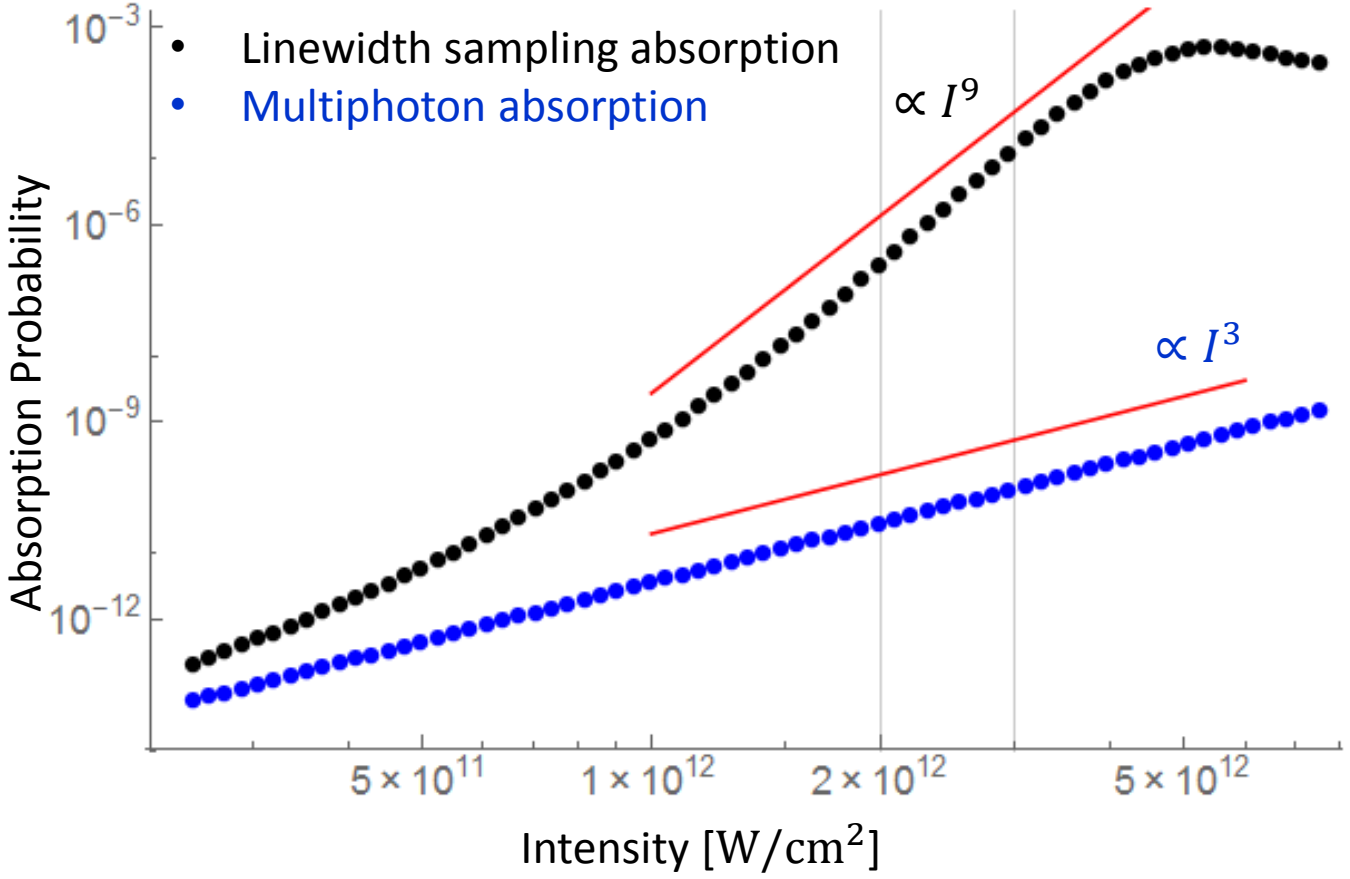
$$\tilde{\rho}(\overline{\omega_f}) = \int_0^\infty \rho\left(\overline{\omega_f} - n\nu\right) f(\nu) d\nu$$

5. New cross section for n photon absorption:

$$\sigma_n = \left(\frac{2}{c\epsilon_0\hbar}\right)^n \left| \sum_{a_1 \cdots a_n} \frac{\langle f|x|a_{n-1}\rangle \cdots \langle a_1|x|g\rangle}{(\omega_{n-1} - (n-1)\nu) \cdots (\omega_1 - \nu)} \right|^2 2\pi\tilde{\rho}(\overline{\omega_f})$$

Resonance Sampling Model

Multiphoton excitation of 5p (shows enhancement)



Modify the multiphoton absorption by:

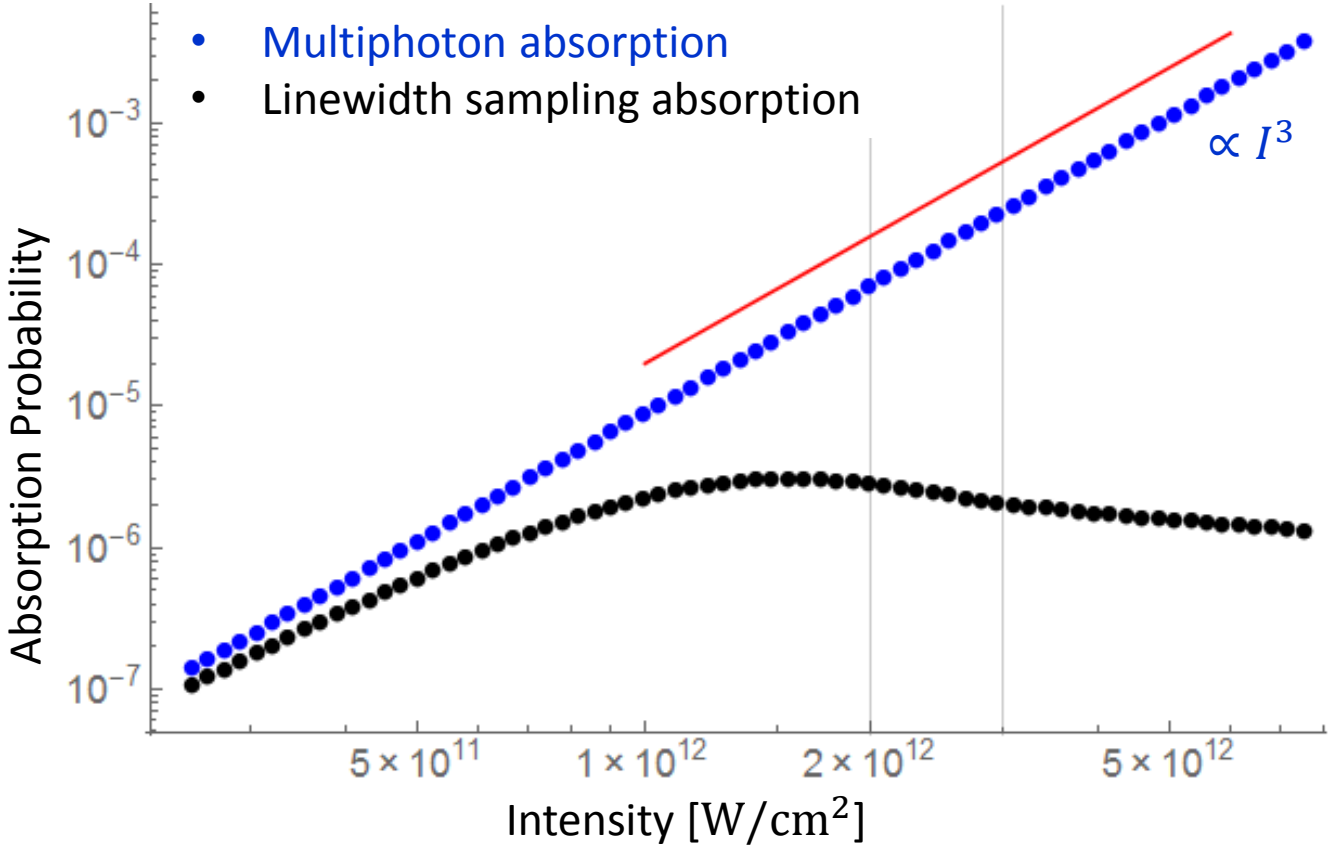
- Setting: $\overline{\omega_f} = \omega_f + U_p$
- Replacing: $\omega \rightarrow \overline{\omega_f}$

New Lorenztian

$$\Gamma\left(\overline{\omega_f} - n\omega\right) = \frac{\gamma_f}{\left(\overline{\omega_f} - n\omega\right)^2 + \left(\frac{\gamma_f}{2}\right)^2}$$

Resonance Sampling Model (Part 2)

Multiphoton excitation of 6p (shows attenuation)



- The saturation intensity of 6p ($1 \times 10^{12}\ W/cm^2$) occurs before that of 5p ($4.68 \times 10^{12}\ W/cm^2$)

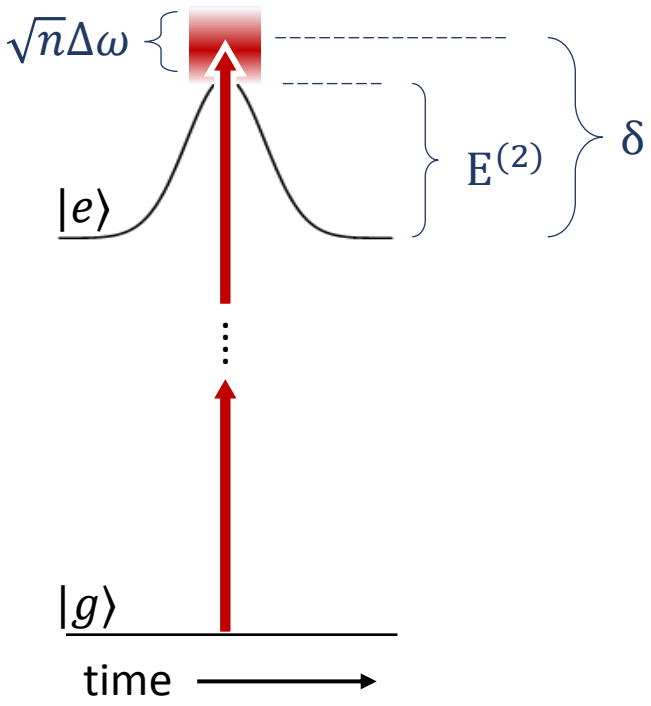
Resonance Sampling Regimes

Definition of parameters:

$\mathrm{E}^{(2)}$ = AC Stark shift (second order perturbation theory)
n = multiphoton order
δ = multiphoton detuning
γ = decay rate of excited state (sum of Einstein A coefficients)
$\Delta\omega$ = laser bandwidth

Note: The downward Stark shift of the initial (ground) state is small compared to $\mathrm{E}^{(2)}$ and is neglected.

Enhanced Excitation

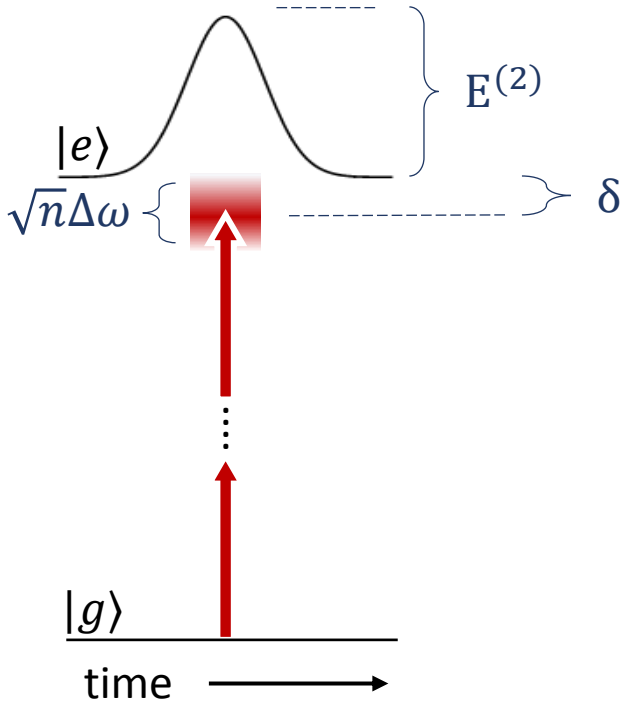


Sampling Condition

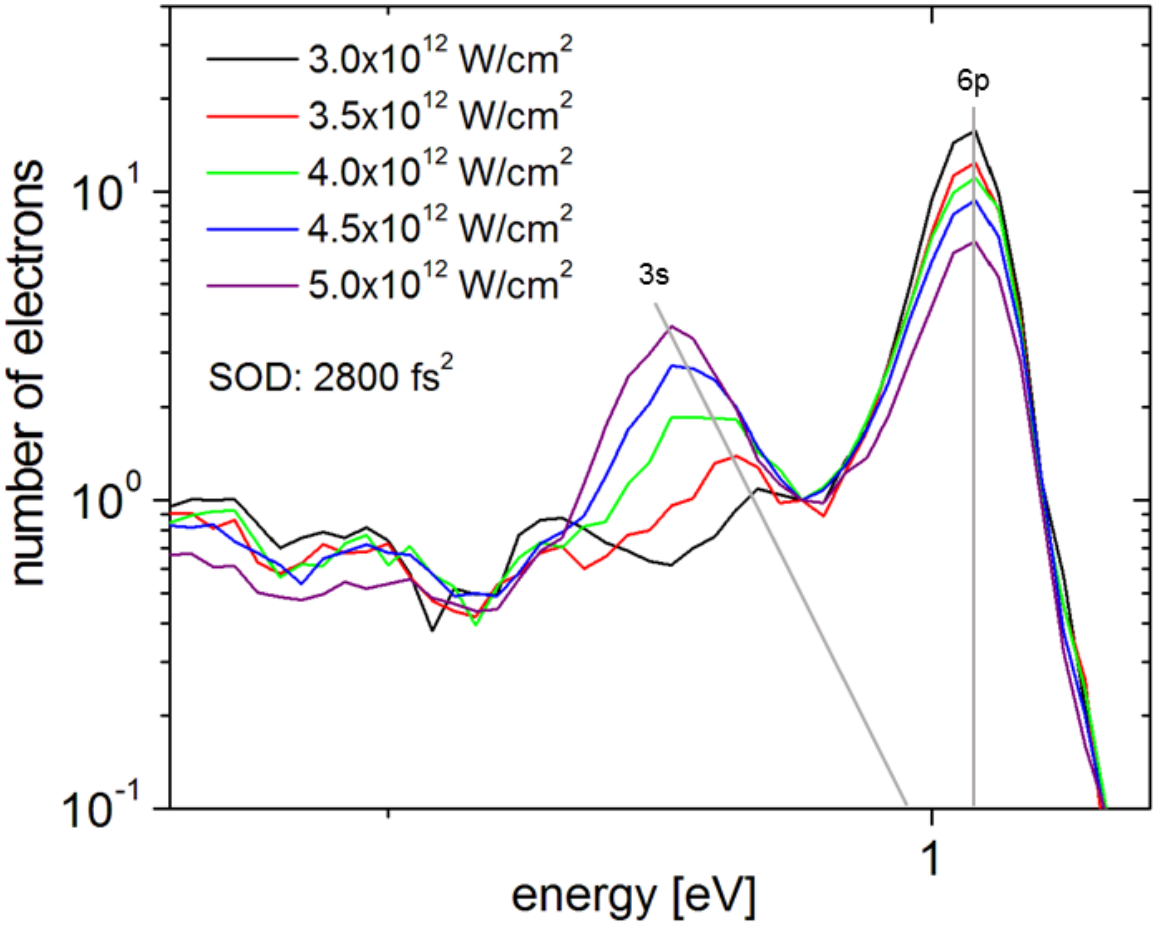
$$\mathrm{E}^{(2)} > \left(\delta - \frac{\gamma}{2}\right) - \frac{\sqrt{n}\Delta\omega}{2}$$

Selectivity of the excitation can be improved by decreasing the bandwidth ($\Delta\omega$) and **increasing the detuning** (δ) from the enhanced level.

Attenuated Excitation



Resonance Sampling Lifetime



The spectrum for each intensity is normalized at 0.82 eV, the common local minimum.

Near the saturation intensity (not plotted hear), the total width of the 5p peak $\Delta E = 0.127$ eV.

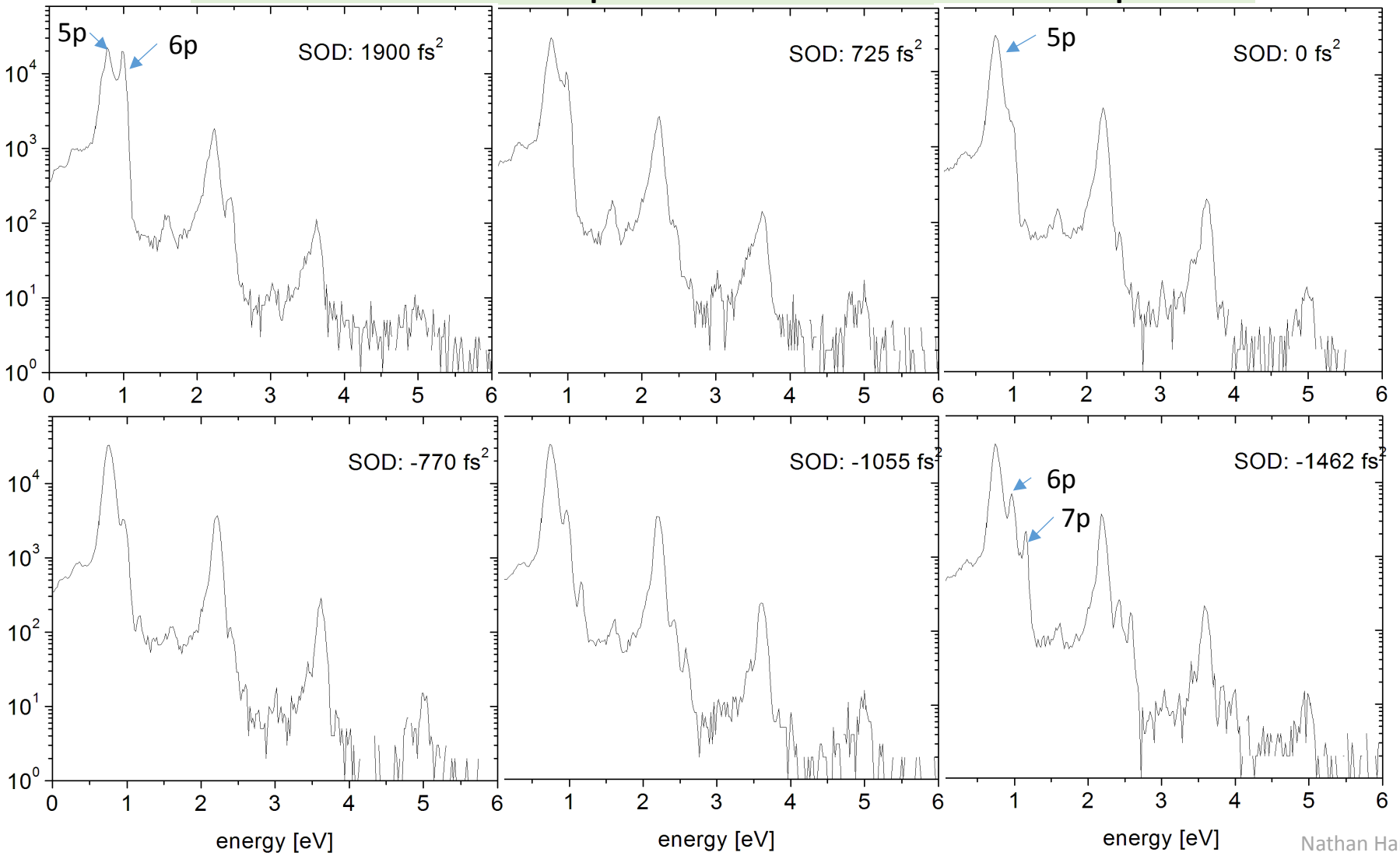
$$\hbar/(\Delta E) = 5 \text{ fs}$$

RS ionization lifetime ***cannot*** be obtained from the uncertainty principle. ($\Delta E \cdot \Delta t \gg \frac{1}{2}\hbar$)
During the Stark shift, the Freeman resonance broadens only towards lower energies.

Chirp scan at constant intensity ($I_0 \sim 2.5 \times 10^{12} \mathrm{W/cm^2}$)

Note: *Pulse energy increases with magnitude of chirp.*

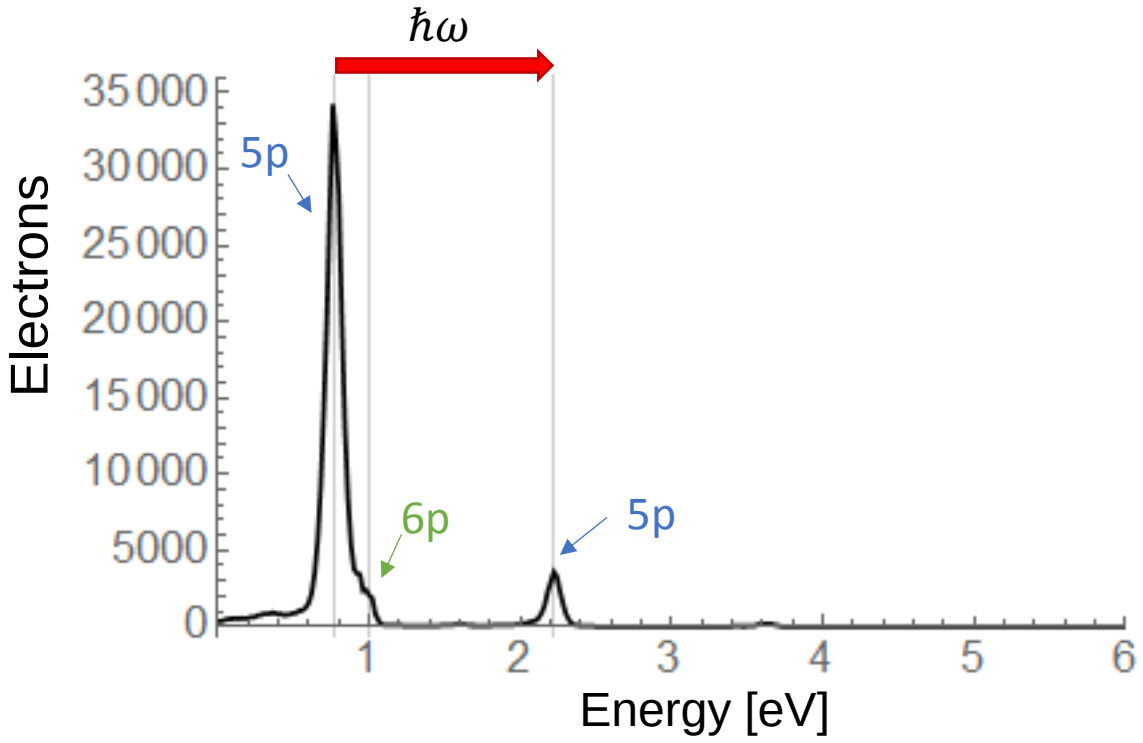
- 5p shows minimal chirp dependence (linewidth sampling)
- 6p disappears for zero chirp
- 7p only appears for negative chirp
- **Selective excitation for 5p is achieved with a transform limited pulse!**



Selective excitation of energy level 5p

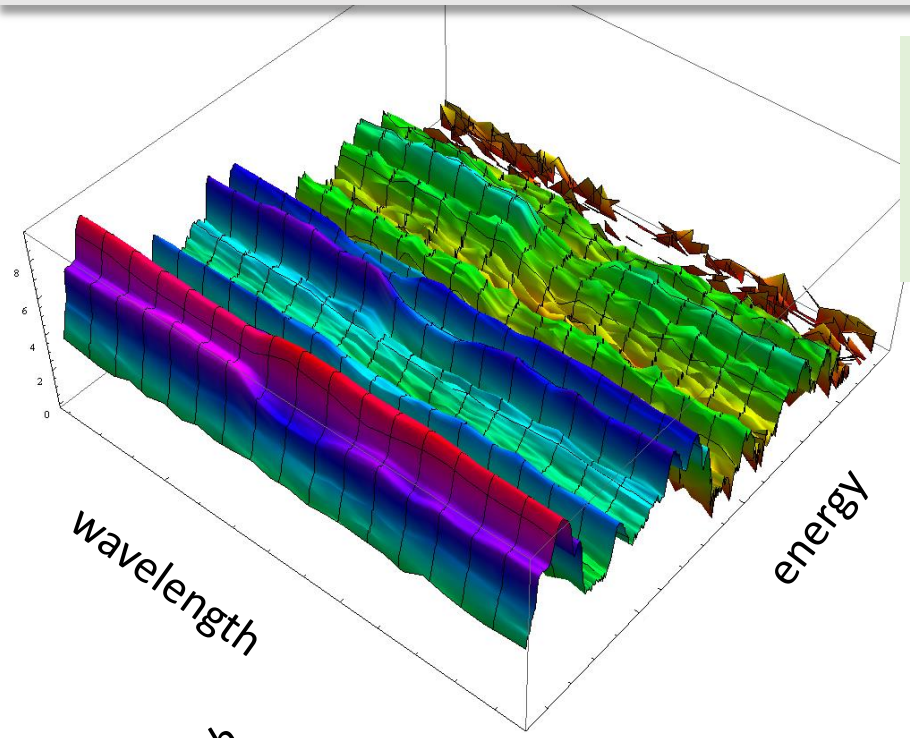
ATI spectra of transform limited pulse at

$I_0 \sim 2.5 \times 10^{12} W/cm^2$

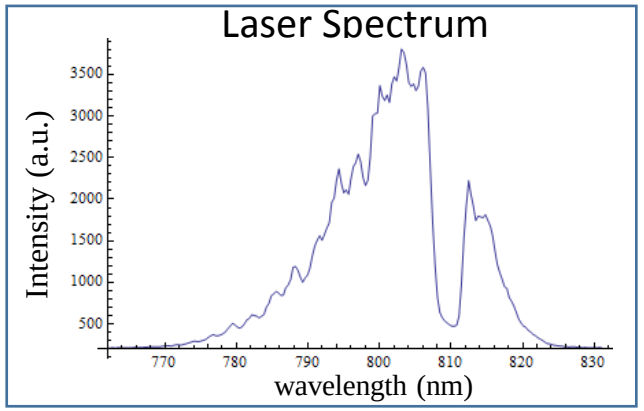


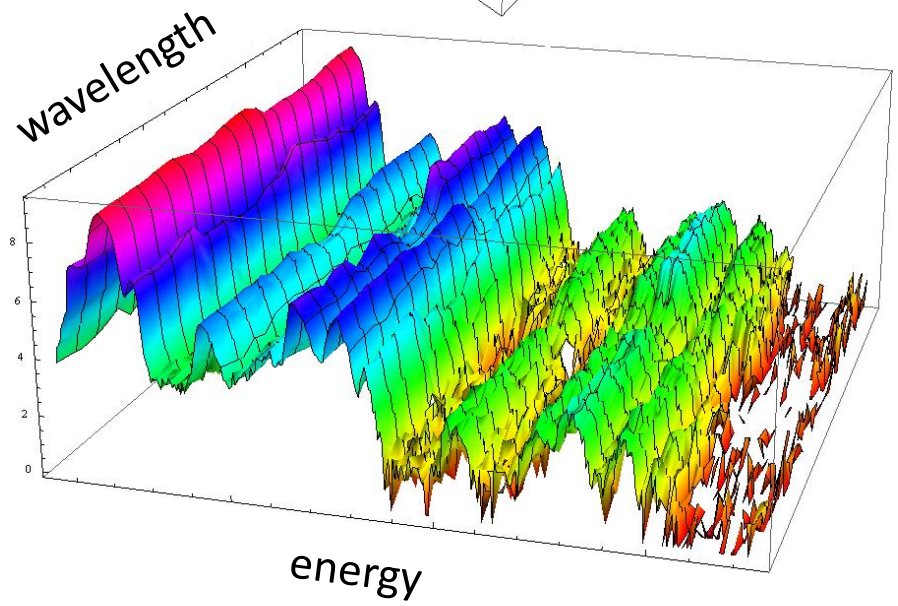
- Ionization from 5p is seen a energies ~ 0.75 and ~ 2.25 eV.
- 6p ionization has an energy of ~ 1 eV

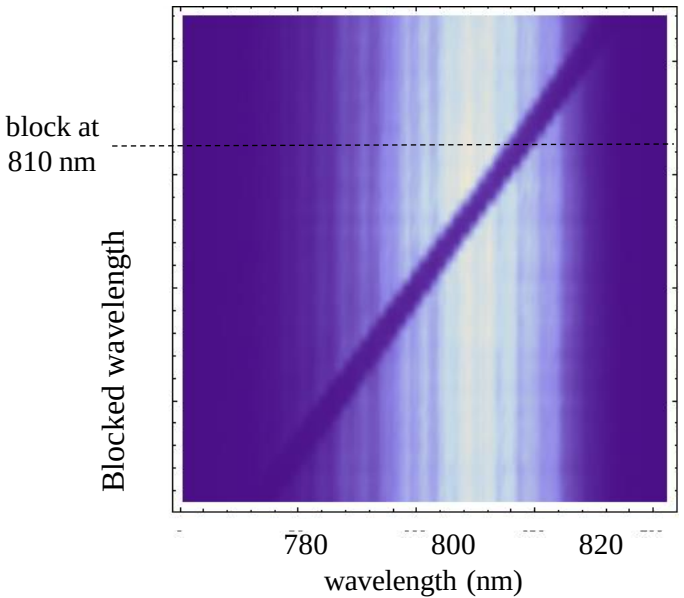
Wavelength block experiment: $\varphi_2 = -2800\ \mathrm{fs}^2$ (~156 fs)



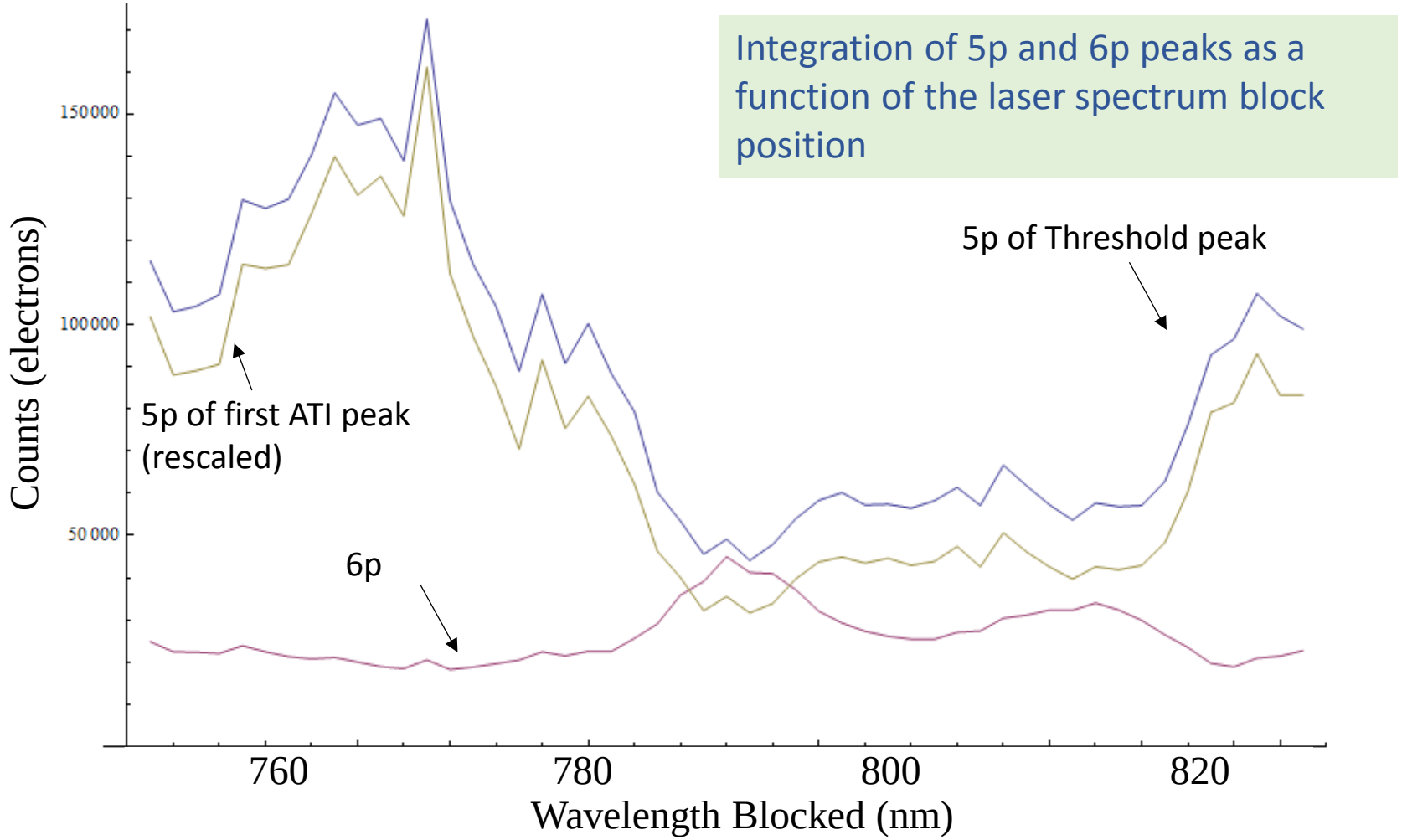
- 4 nm of the spectrum is attenuated, and an ATI measurement is taken.
- This process is repeated at a new spectral position until the attenuated position is scanned across the entire spectrum.
- Pulse energy is kept constant.





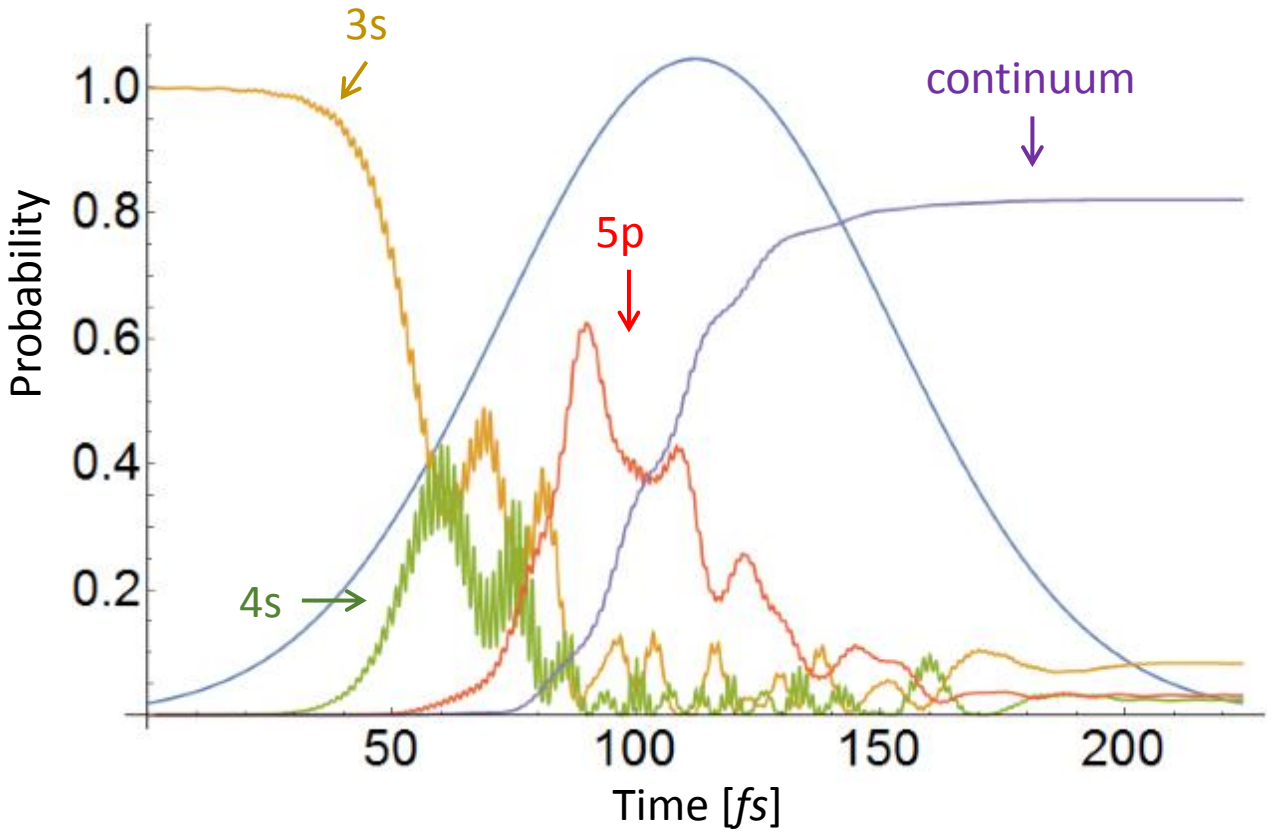


The Effect of Spectral Components



- 5p and 6p represent competitive ionization pathways.
- Enhancement in 5p at 765 nm is not accounted for by attenuation in 6p.
- Blocking higher energy photons enhances 5p excitation by reducing the average detuning for the pulse spectrum.

Density Matrix Simulation of Sodium Atom and Laser Pulse Interaction



$\tau = 56\ fs$
$I_0 = 6 \times 10^{12}\ W/cm^2$
$\lambda = 800\ nm$
Linear polarization

- Transient population inversion of ground state 3*s* and the excited state 5*p* at sufficiently high intensities.
- Possible applications for new laser mediums

Conclusion

Pros: *Within the high-field regime* ($> 10^{12}$ W/cm^2), excitation due to Freeman resonances is both efficient and stable with respect to changes in chirp.

Con: Energy state selectivity requires a compressed pulse.

Potential applications:

- Phase-independence make it practical for remote detection over a large range of distances (i.e. Radar REMPI).
- Population inversion using this mechanism could lead to new laser mediums.

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