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 \( (> 10^{12} \text{ W/cm}^2) \) 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.

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

![Laser Pulse Spectrum](image_url)
Multiphoton Ionization

\[ n \hbar \omega + A \rightarrow e^- + A^+ \]

Kinetic energy of the electron:

\[ KE = n \hbar \omega - V_{IE} \]

Ionization probability from perturbation theory:

\[ P(I) \propto I^n \]

\( n \) photons ionize an atom:

\( n \hbar \omega + A \rightarrow e^- + A^+ \)

Multiphoton condition (from Keldysh theory):

\[ \gamma \gg 1 \]
Sodium (Na) Energy Level Diagram

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

Kinetic energy

Number of electrons

\[ |g > \]

\[ e_1 \]

\[ e_2 \]

\[ e_1 \]

\[ e_1 \]

\[ e_1 \]

\[ e_2 \]

\[ e_1 \]

\[ e_1 \]

\[ e_1 \]

\[ e_1 \]

\[ e_1 \]

\[ e_1 \]
ATI Spectrum of Sodium Vapor

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

\[ I_0 = 4.5 \times 10^{12} \text{ W/cm}^2 \]

- 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.
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 $U_p$.

$$E_{6p} - E_{3s} = U_p$$
The 3s peak position decreases linearly with increasing intensity and is used to calibrate the laser peak intensity in the focus.

At $4.68 \times 10^{12}$ 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 ~10^{-3} mbar)

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

Gold: $6.7 \times 10^{12} \text{ W/cm}^2$

Purple: $3.5 \times 10^{12} \text{ W/cm}^2$

Blue: $2.4 \times 10^{12} \text{ W/cm}^2$
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.
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

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

**Diagonal Density Matrix Element:**

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

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

\[
N_{4s} = N_{\text{total}} \cdot \rho_{4s} \\
= N_{\text{total}} \cdot \sigma_{3s \rightarrow 4s} I^2
\]

**Penning Ionization**

\[
N_{\text{ion}} \propto N_{4s}^2 I^2 \\
\propto N_{\text{total}}^2 \cdot \sigma_{3s \rightarrow 4s}^2 I^6
\]
Appendix A

Calculating the sodium density using Lambert’s Cosine Law

Appendix A
Calculating the sodium density using Lambert’s Cosine Law

\[
\rho_i = \text{initial density}
\]
\[
\rho_f = \text{initial density}
\]
\[
d\Omega = \text{solid angle} = \sin(\theta)d\theta
d\phi
\]
\[
dA = \text{differential area of aperture}
\]
\[
\Phi_i = \Phi_f = \text{atomic flux (assuming constant velocity)}
\]

Flux of atoms from the aperture

\[
\Phi_i = \rho_i \int dA_i \int d\Omega_i
\]
\[
= \rho_i A_i 2\pi (1 - \cos(\theta_i))
\]

Density of atoms reaching the target

\[
\rho_f = \frac{\Phi_i}{\int dA_f \int d\Omega_f}
\]
\[
= \frac{\Phi_i}{\left( A_f 2\pi (1 - \cos(\theta_f)) \right)}
\]
Nozzle Characterization for Oven Temperature T=256°C

**Ideal Gas Law:** \(PV = NkT\)

- \(\rho_i\) = initial density = \(1.98 \times 10^{14}\) cm\(^{-3}\)
- \(\rho_f\) = target density = \(5.17 \times 10^{13}\) 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 = 10\(\mu\)m
- Beam Rayleigh range = 0.79mm

Thermal velocity of Na atoms: 349.3 m/s

Time between collisions: 72.2 \(\mu\)s

Energy Level Lifetimes:
- \(\tau_{4s} = 37.9\) ns
- \(\tau_{5p} = 0.37\) \(\mu\)s
- \(\tau_{6p} = 1.56\) \(\mu\)s

- Atomic or electron collisions not likely.
### Appendix B

**Sodium parameters**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Width of 6th order peak:</td>
<td>$\Delta E = 0.127$ eV</td>
</tr>
<tr>
<td>Ionization potential:</td>
<td>$E_I = 5.139$ eV</td>
</tr>
<tr>
<td>Over the Barrier (OTB) Intensity:</td>
<td>$I_{OTB} = 2.78 \times 10^{12}$ W/cm$^2$</td>
</tr>
<tr>
<td>Keldysh tunneling parameter for OTB Intensity:</td>
<td>$\gamma = 3.94$</td>
</tr>
<tr>
<td>Ionization width of 6th peak:</td>
<td>$\Delta t = 2.5$ fs</td>
</tr>
<tr>
<td>Na cross section:</td>
<td>$\sigma_{5p} = 2.8 \times 10^{-19}$ m$^2$</td>
</tr>
<tr>
<td>Na Dimer percentage at $T = 256$ °C:</td>
<td>Na$_2 \sim 2%$</td>
</tr>
</tbody>
</table>
The Absence of Tunneling Ionization

Bohr model for hydrogen-like atoms

Solve for the orbital velocity

\[ \vec{F} = m\vec{a} = -\frac{ke^2}{r^2} = -\frac{mv^2}{r} \quad \Rightarrow \quad v = e\sqrt{\frac{k}{mr}} \]

Solve for the radius \( r \)

\[ U(r) = -\frac{ke^2}{r} = eE_{5p} \quad \Rightarrow \quad r = \frac{ke}{|E_{5p}|} \]

Orbital period of level 5p

\[ T_{5p} = \frac{2\pi r}{v} = \frac{2\pi \sqrt{kmr}}{|E_{5p}|} = 2\pi k\sqrt{\frac{me}{|E_{5p}|^3}} \]

\[ = 4.07 \text{ fs} \]

Energy levels:

- \( E_{3s} = -5.139 \text{ eV} \)
- \( E_{5p} = -0.794 \text{ eV} \)

Over the Barrier (OTB) Intensity:

\[ I_{OTB} = 2.78 \times 10^{12} \text{ W/cm}^2 \]

Keldysh tunneling parameter for OTB Intensity:

\( \gamma = 3.94 \)

Laser period:

\( T_L = 2.7 \text{ fs} \)

- 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.
• Program solves the 1D time dependent Schrödinger equation in the length gauge.
• Atom is represent by 1D soft-core Coulomb potential.
- 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.
1. Cross section for n photon absorption:

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

2. Absorption rate:

\[W_n(t) = \sigma_n I(t)^n\]

3. Rate integration:

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

4. Absorption probability (see slide Appendix C):

\[p_n = \frac{1 - e^{-2s}}{2}\]

---

The Lorenztian Density of States

- Defines linewidth for energy level \(\omega_f\)
- \(\gamma_f\) = decay rate of level \(f\)
- \(\nu\) = instantaneous laser frequency

\[
\rho(\omega_f - n\nu) = \frac{1}{\pi} \frac{\left(\frac{\gamma_f}{2}\right)}{(\omega_f - n\nu)^2 + \left(\frac{\gamma_f}{2}\right)^2}
\]
Appendix C
Integrating Rate equations

1. The classical rate equations:
\[
\frac{dN_a}{dt} = -W_n N_a + W_n N_b = -W_n (N_a - N_b)
\]
\[
\frac{dN_b}{dt} = -W_n N_b + W_n N_a = W_n (N_a - N_b)
\]

2. Subtract the rates:
\[
\frac{d}{dt} (N_a - N_b) = -2W_n (N_a - N_b)
\]

Identity relations:
\[
N_a = 1 - N_b
\]
\[
N_{a0} = 1
\]
\[
N_{b0} = 0
\]

Notation:
\[
n \text{ photon rate } \rightarrow W_n \equiv W_n(t)
\]
\[
\text{Population in state } a \rightarrow N_a \equiv N_a(t)
\]
\[
\text{Population in state } a \rightarrow N_b \equiv N_b(t)
\]

\[
(N_a - N_b) = (N_{a0} - N_{b0}) e^{-2W_n t}
\]
\[
(1 - 2N_b) = e^{-2W_n t}
\]
\[
N_b = \frac{1 - e^{-2W_n t}}{2}
\]
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
   $$\rho(\overline{\omega_f} - n\nu) = \left(\frac{1}{\pi}\right) \frac{\left(\frac{\gamma_f}{2}\right)}{(\overline{\omega_f} - n\nu)^2 + \left(\frac{\gamma_f}{2}\right)^2}$$

3. Define the normalized Bandwidth function
   $$a = \frac{n \log(2)}{(\tau/2)^2}$$
   $$f(\nu) = \frac{1}{\sqrt{4a\pi}} e^{-n^2(\nu - \nu_0)^2/(4a)}$$

4. Average over the bandwidth
   $$\tilde{\rho}(\overline{\omega_f}) = \int_{0}^{\infty} \rho(\overline{\omega_f} - n\nu) f(\nu) d\nu$$

5. New cross section for n photon absorption:
   $$\sigma_n = \left(\frac{2}{c\varepsilon_0 \hbar}\right)^n \left| \sum_{a_1 \cdots a_n} \frac{\langle f | x | a_{n-1} \rangle \cdots \langle a_1 | x | g \rangle}{(\omega_{n-1} - (n-1)\nu) \cdots (\omega_1 - \nu)} \right|^2 2\pi \tilde{\rho}(\overline{\omega_f})$$
Resonance Sampling Model

Multiphoton excitation of $5p$ (shows enhancement)

- Linewidth sampling absorption
- Multiphoton absorption

$\frac{\gamma f}{(\omega_f - n\omega)^2 + \left(\frac{\gamma f}{2}\right)^2}$

Modify the multiphoton absorption by:

- Setting: $\bar{\omega}_f = \omega_f + U_p$
- Replacing: $\omega \rightarrow \bar{\omega}_f$
Resonance Sampling Model (Part 2)

Multiphoton excitation of 6p (shows attenuation)

- Multiphoton absorption
- Linewidth sampling absorption

\[
\alpha \propto I^3
\]

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

Definition of parameters:

- \( E^{(2)} = AC \) Stark shift (second order perturbation theory)
- \( n = \) multiphoton order
- \( \delta = \) multiphoton detuning
- \( \gamma = \) 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 \( E^{(2)} \) and is neglected.

**Sampling Condition**

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

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

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

Near the saturation intensity (not plotted here), the total width of the 5p peak \(\Delta E = 0.127 \text{ eV}\).

\[\frac{\hbar}{(\Delta E)} = 5 \text{ fs}\]
Chirp scan at constant intensity ($I_0 \sim 2.5 \times 10^{12}\text{ W/cm}^2$)

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

Ionization from 5p is seen at energies $\sim 0.75$ and $\sim 2.25$ eV.

6p ionization has an energy of $\sim 1$ eV.

ATI spectra of transform limited pulse at $I_0 \sim 2.5 \times 10^{12} \text{W/cm}^2$
Wavelength block experiment: $\varphi_2 = -2800 \text{ fs}^2 \ (\sim 156 \text{ fs})$

- 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.
• 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

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

\[ \tau = 56 \text{ fs} \]
\[ I_0 = 6 \times 10^{12} \text{ W/cm}^2 \]
\[ \lambda = 800 \text{ nm} \]
Linear polarization
**Pros:** Within the high-field regime ($> 10^{12} \text{ W/cm}^2$), 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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