

From One to Many Photons: Connecting field ionization to photoionization via GHz microwave ionization of Rydberg atoms

Joshua Gurian
T.F. Gallagher

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Rydberg Atoms and How They Ionize

Rydberg Atoms

Field Ionization

Photoionization

MW Ionization

Experimental Setup

Experimental Apparatus

kHz Laser

Experimental Results

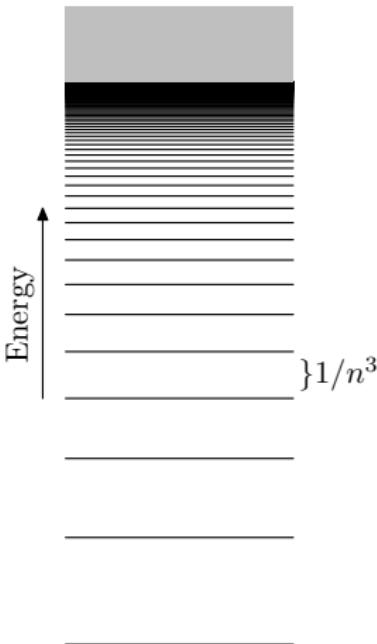
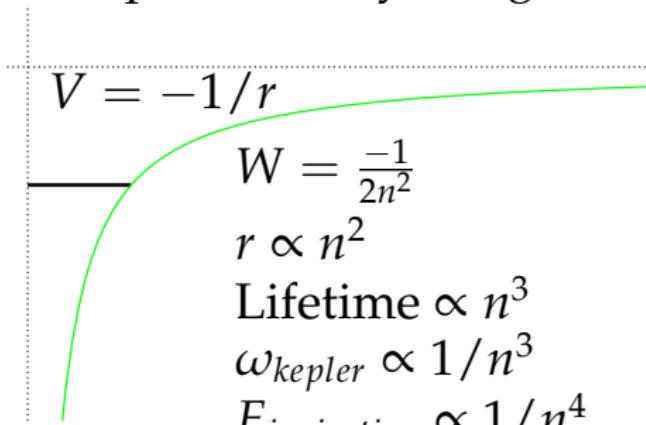
Multiphoton MW Ionization

Single Photon Ionization Rates

Above-Threshold Bound States

Introduction to Rydberg Atoms

Properties of Rydberg Atoms



Introduction to Rydberg Atoms

Properties of Rydberg Atoms

$$V = -1/r$$

$$W = \frac{-1}{2n^2}$$

$$r \propto n^2$$

$$\text{Lifetime} \propto n^3$$

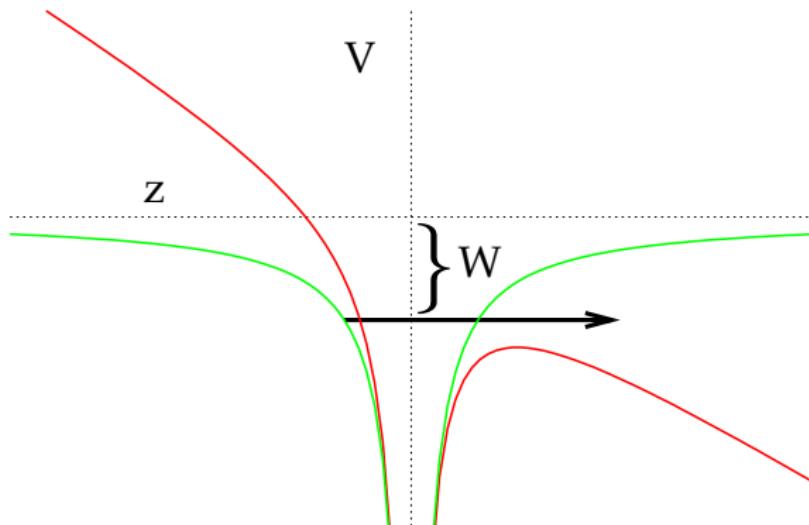
$$\omega_{\text{kepler}} \propto 1/n^3$$

$$E_{\text{ionization}} \propto 1/n^4$$

For n=100:

- ▶ $W = -1.4 \text{ meV}$
- ▶ $\langle r \rangle = 0.5 \mu\text{m}$
- ▶ $\tau = 1 \text{ ms}$
- ▶ $\omega_{\text{kepler}} = 2\pi \times 6.5 \text{ GHz}$
- ▶ $E_{\text{ionization}} = 5.7 \text{ V/cm}$

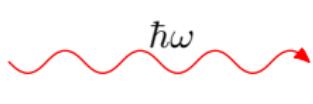
Field Ionization



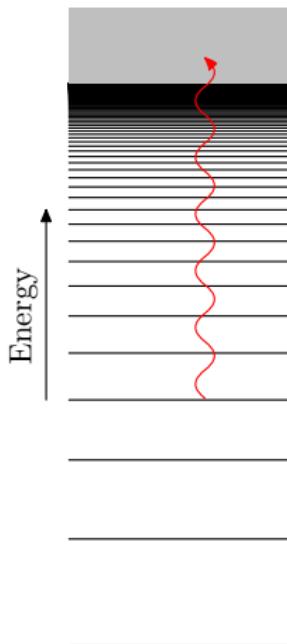
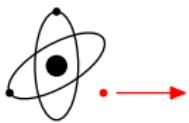
$$V(z) = \frac{-1}{|z|} - Ez$$

$$E = \frac{W^2}{4}$$

Photoionization



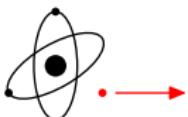
if $\hbar\omega > W$,



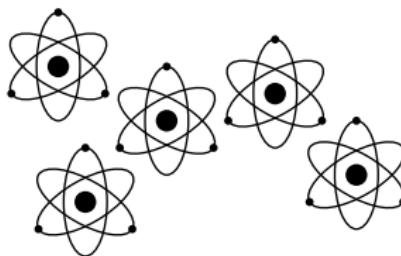
Photoionization



if $\hbar\omega > W$,

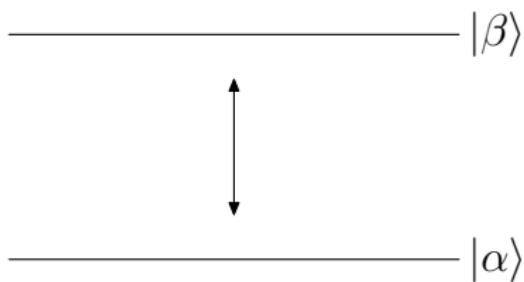


If we have some collection of atoms,



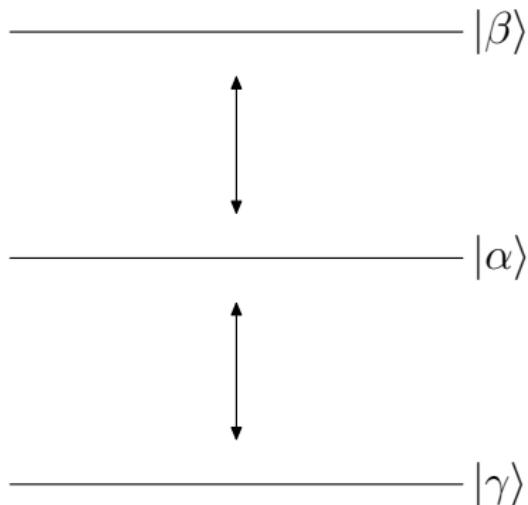
how do we calculate the ionization rate?

Fermi's Golden Rule



Fermi's Golden Rule:
 $\Gamma_1 = 2\pi|\langle\alpha|\mu E|\beta\rangle|^2\rho_f$

Fermi's Golden Rule

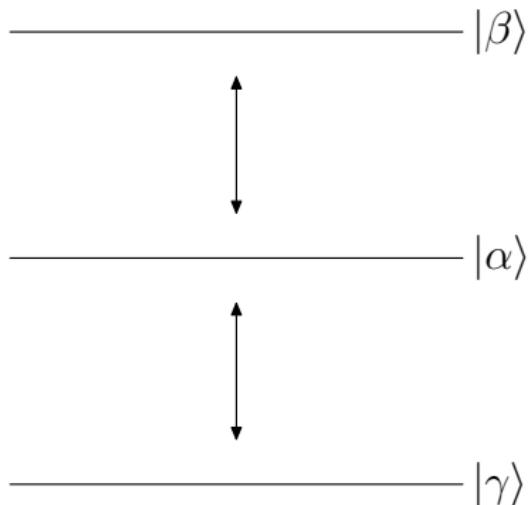


Fermi's Golden Rule:

$$\Gamma_1 = 2\pi |\langle \alpha | \mu E | \beta \rangle|^2 \rho_f$$

$$\Gamma_2 = 2\pi \left| \frac{\langle \gamma | \mu E | \alpha \rangle \langle \alpha | \mu E | \beta \rangle}{\Delta W} \right|^2$$

Fermi's Golden Rule



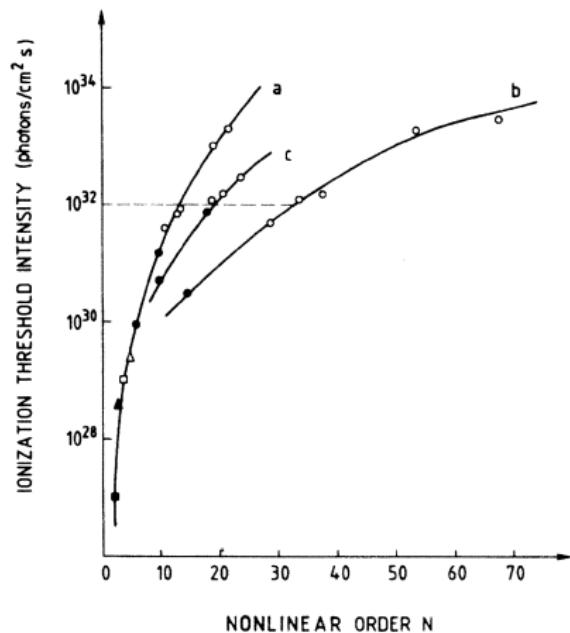
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$$\Gamma_N \propto E^{2N}$$

Fermi's Golden Rule



Fermi's Golden Rule:

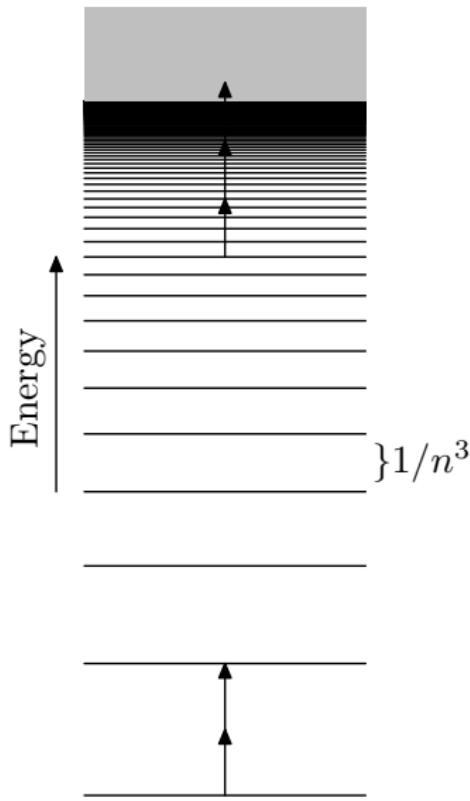
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$$\Gamma_N \propto E^{2N}$$

I'Huillier *et al.*, PRA 27 (1983).

Where does Microwave Ionization fit?



Scaled Microwave Units:

$$\Omega = \frac{\omega}{\omega_{Kepler}} = \omega n^3$$

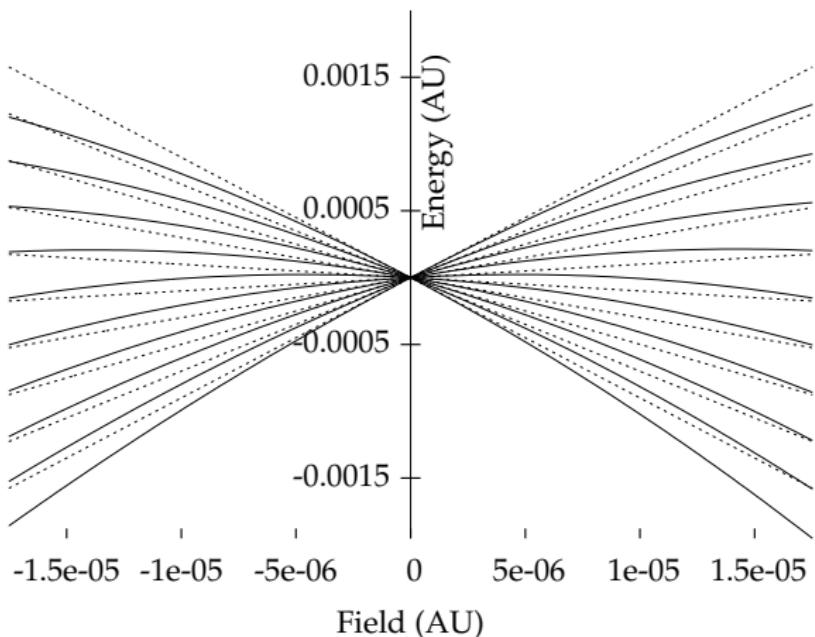
$$E_0 = \frac{E}{E_{Coulomb}} = En^4$$

Hydrogenic Microwave Ionization

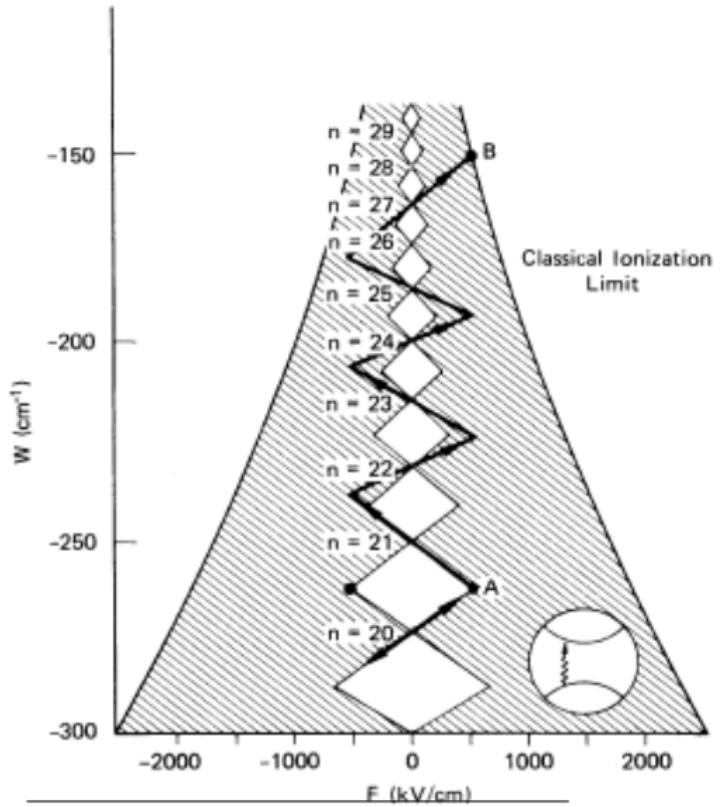
First experiments by Bayfield and Koch:

Non-constant $\frac{dW}{dE}$ projects (n, k) state onto Stark manifold

$$E = \frac{1}{9n^4}$$



Non-Hydrogenic Microwave Ionization

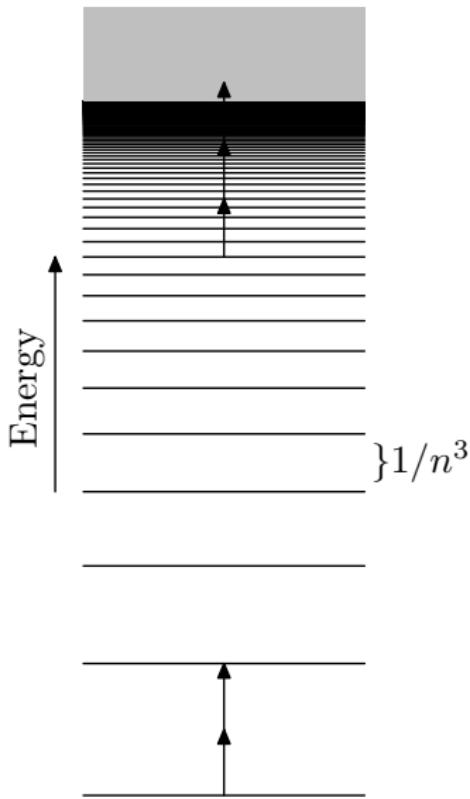


Ladder-climbing mechanism for ionization:

$$E = \frac{1}{3n^5}$$

Pillet et al., Phys. Rev. Lett. 50, 1983.

Where does Microwave Ionization fit?



Scaled Microwave Units:

$$\Omega = \frac{\omega}{\omega_{Kepler}} = \omega n^3$$

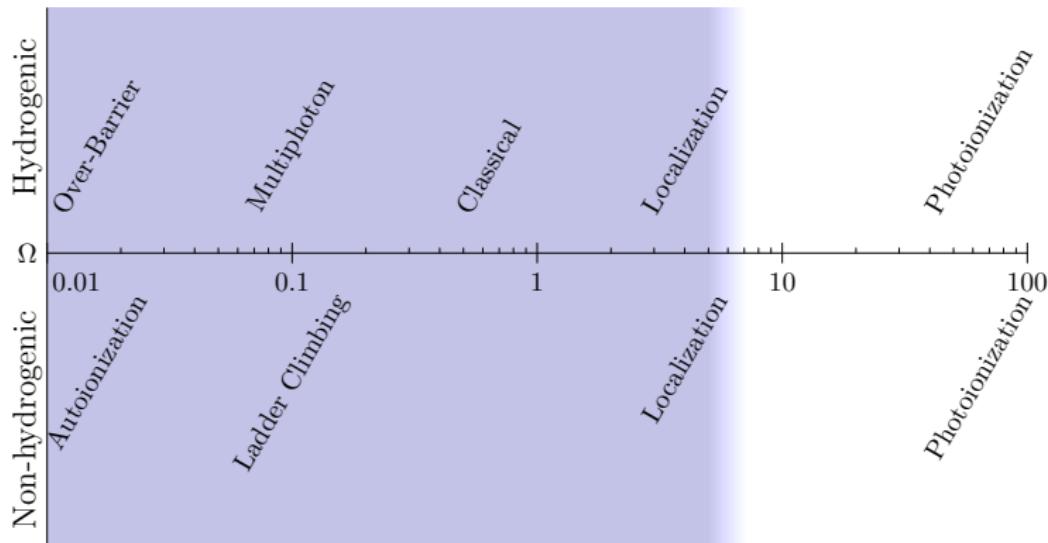
$$E_0 = \frac{E}{E_{Coulomb}} = En^4$$

Experimentally, what happens as we approach the photoionization limit?

Microwave Ionization

$$\Omega = \frac{\omega}{\omega_{Kepler}} = \omega n^3$$

$$E_0 = \frac{E}{E_{Coulomb}} = E n^4$$



What happens as we approach the photoionization limit?

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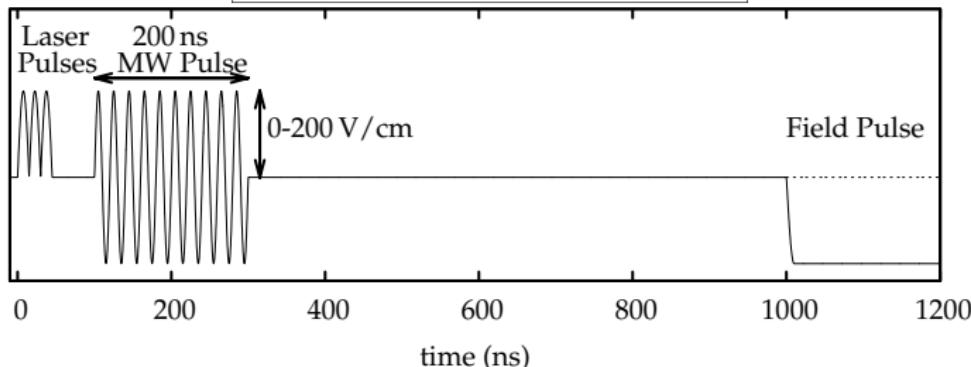
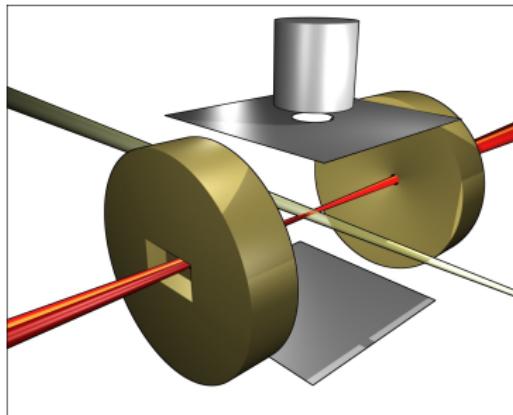
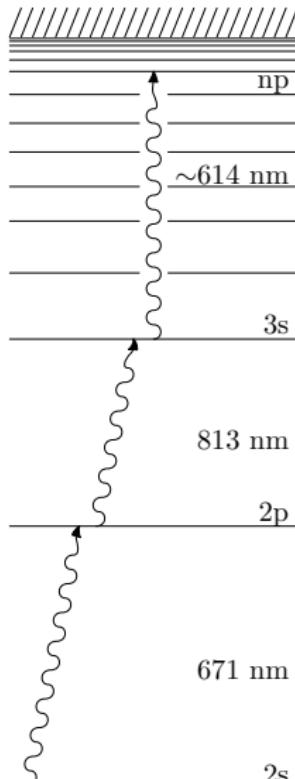
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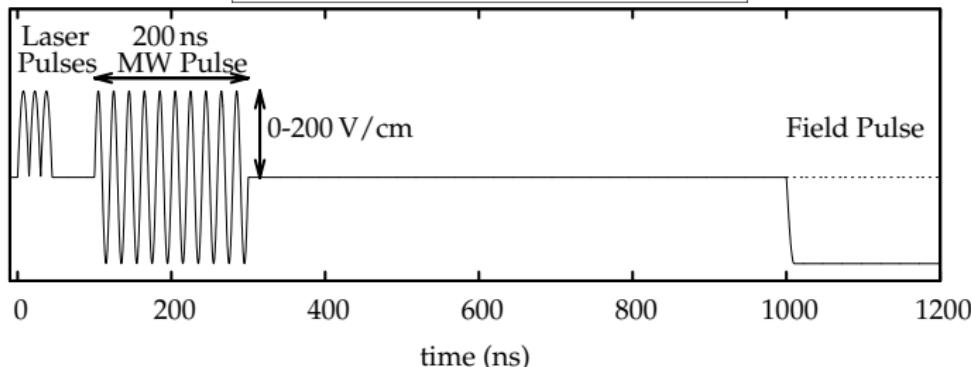
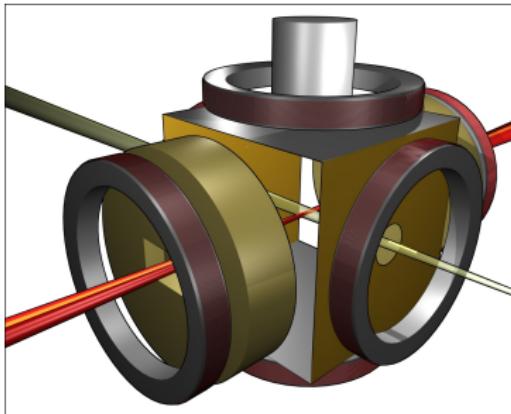
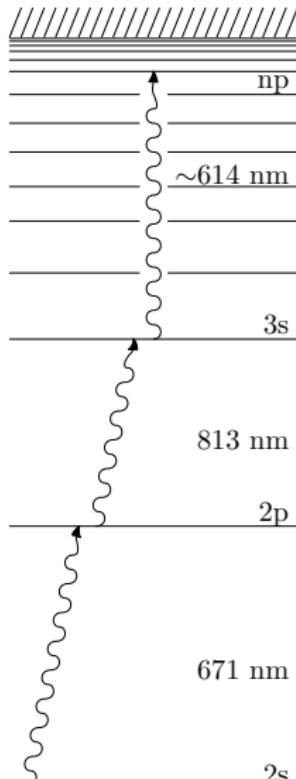
Single Photon Ionization Rates

Above-Threshold Bound States

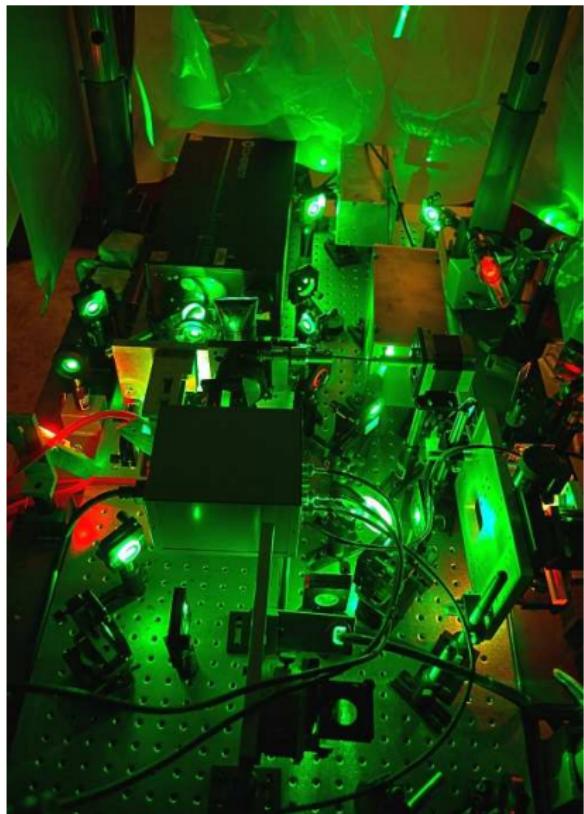
Experimental Setup



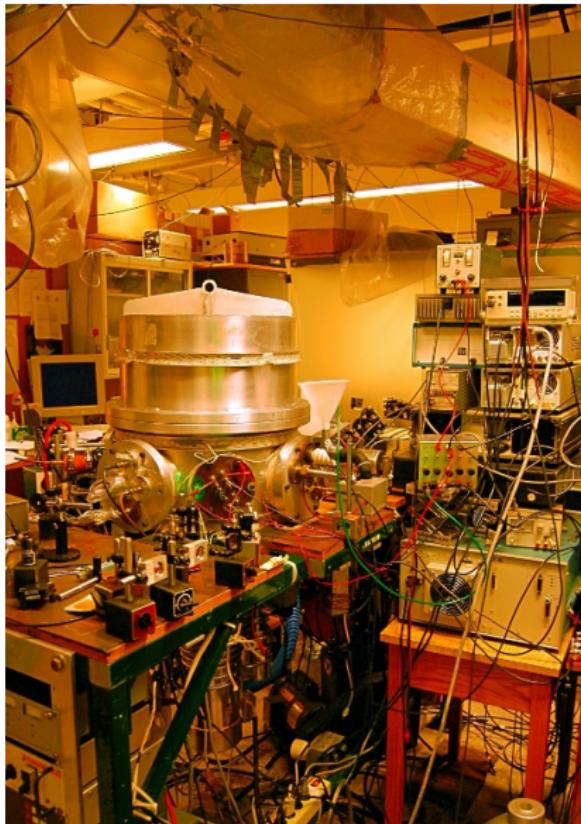
Experimental Setup



Pictures



J. Gurian

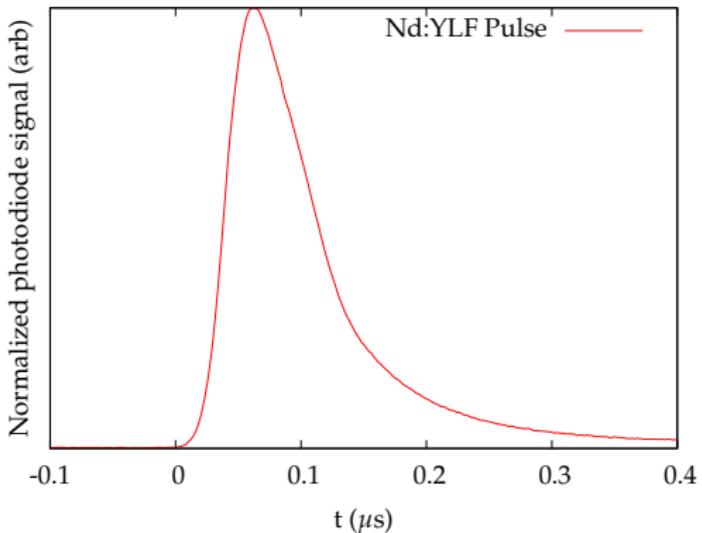


MW Ionization of Rydberg Atoms

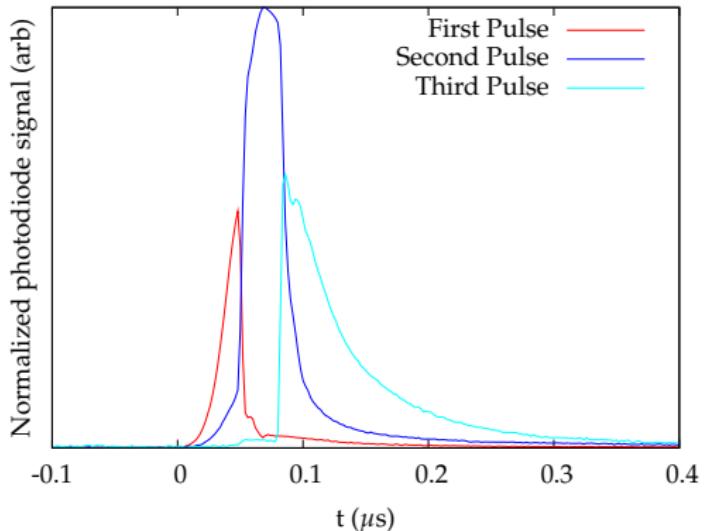
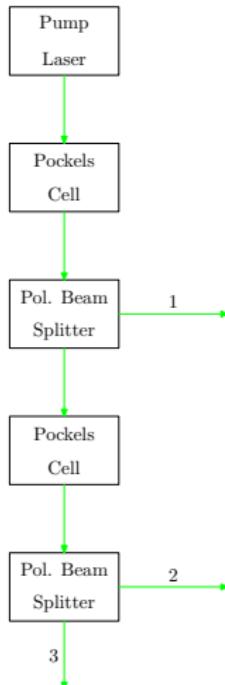
A New Laser System



- ▶ Coherent Evolution-30
- ▶ Nd:YLF @ 527 nm
- ▶ 20 mJ/pulse w/ 1 kHz Pulse Repetition Frequency



External Pulse Splitting



Dye Laser Output: $\approx 20 \mu\text{J}/\text{pulse}$

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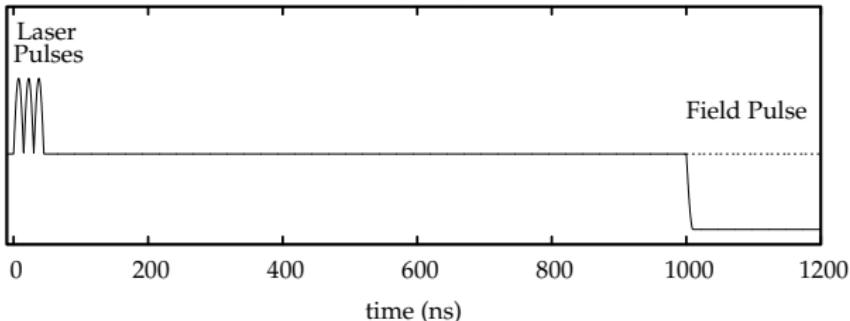
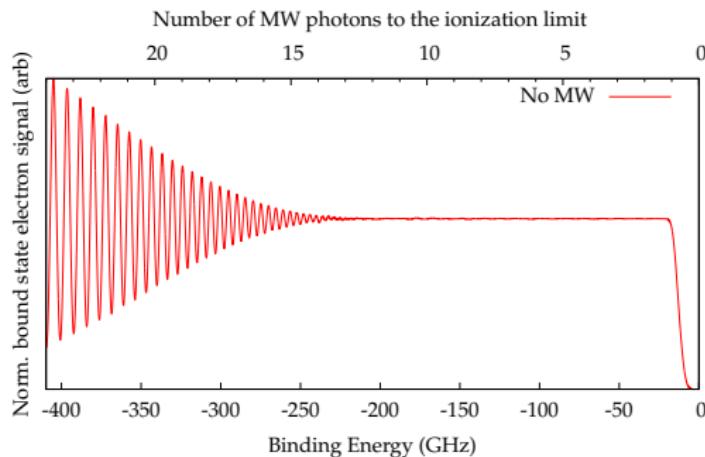
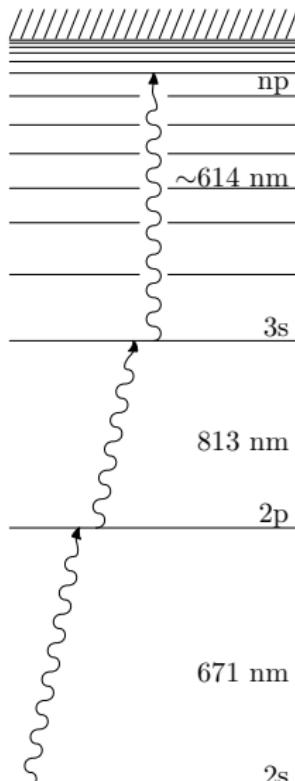
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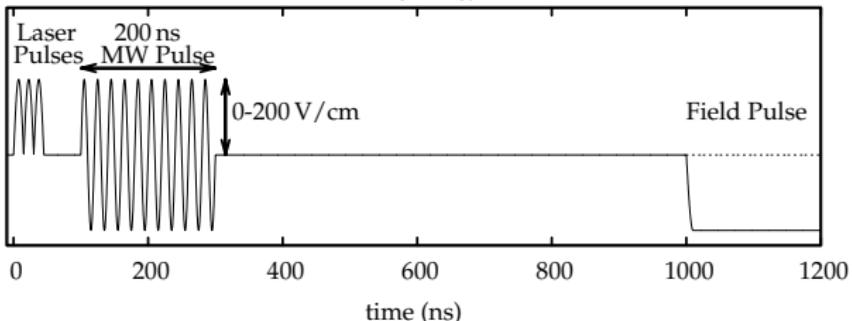
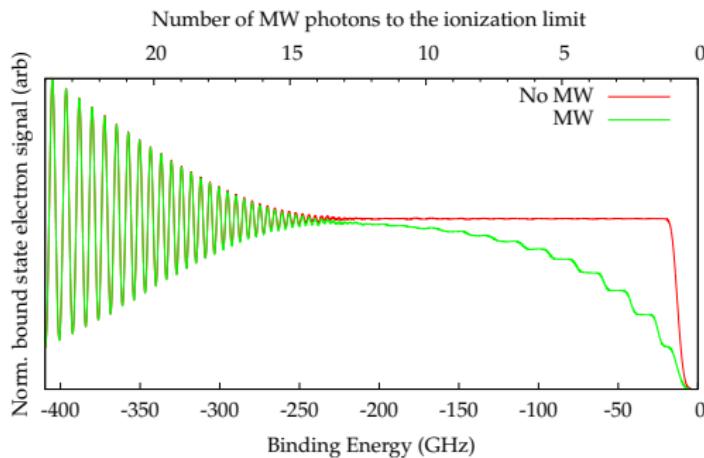
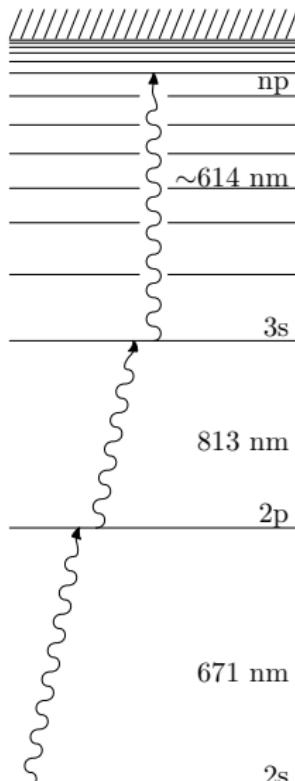
Single Photon Ionization Rates

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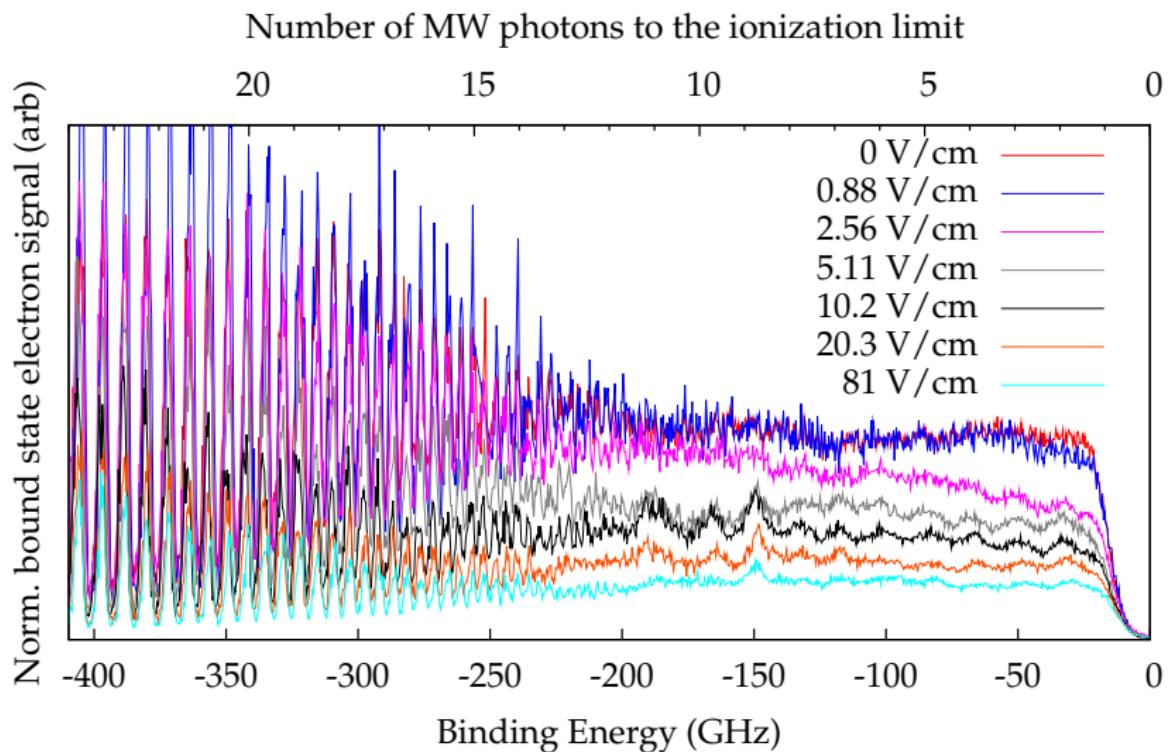
Expected Results



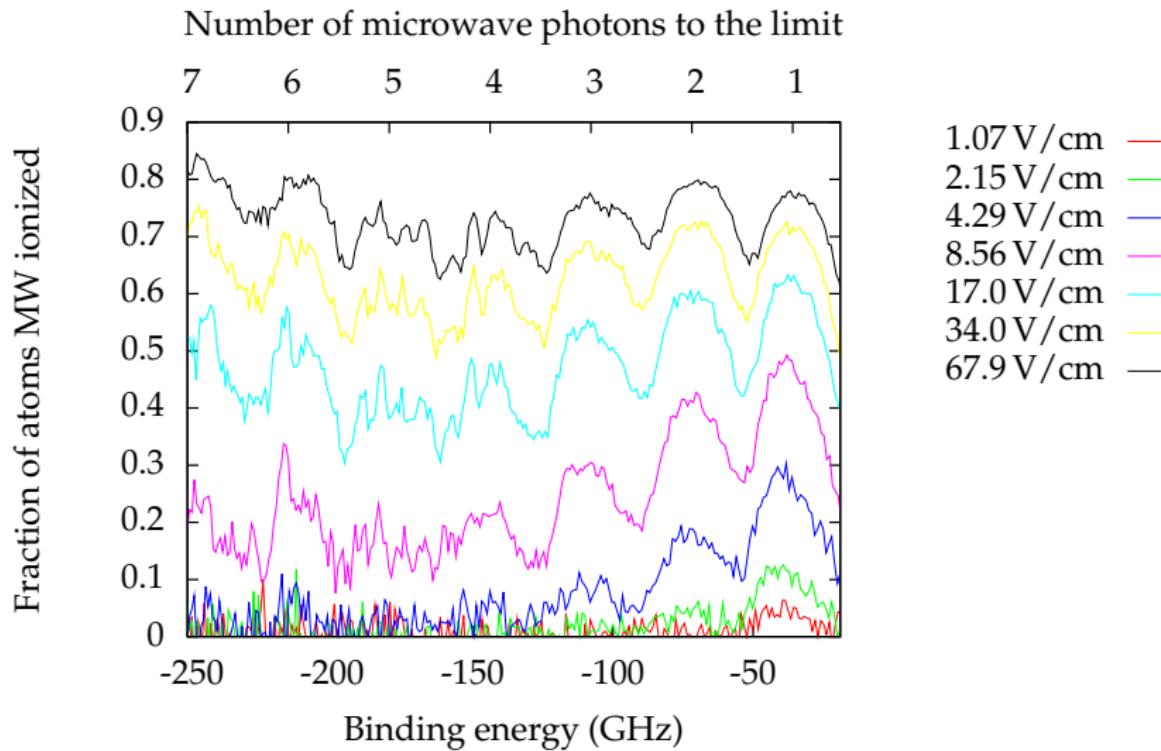
Expected Results



Microwave Ionization Steps - 17 GHz

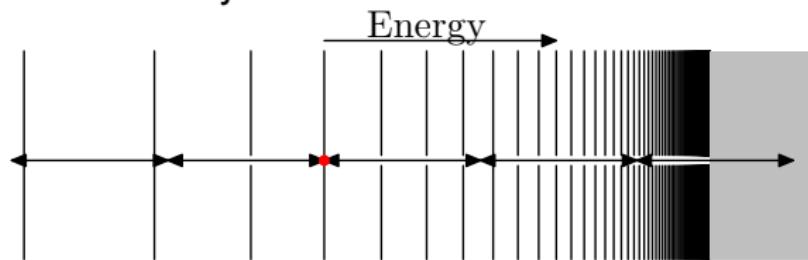


Microwave Ionization Steps - 36 GHz



Dynamical Anderson Localization

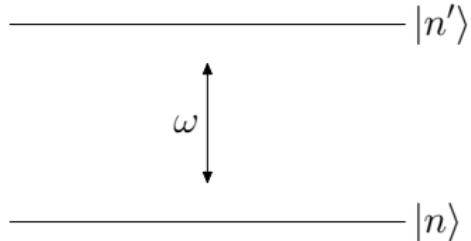
First work by Casati *et al.*



Anderson Localization - Destructive interference between many multiphoton paths localizes the electronic wave function, and ionization occurs when electron transport diffuses over the limit.
Schelle *et al.* - Anderson Localization crossing over to Fermi's Golden Rule

Schelle, Delande, and Buchleitner, *PRL* 102 (2009).
Casati *et al.*, *Phys. Rep.* 154 (1987).

Jensen *et al.* Model



$$\text{Rabi width} = \mu \cdot E = \frac{0.4108E}{\omega^{5/3} n^3}$$

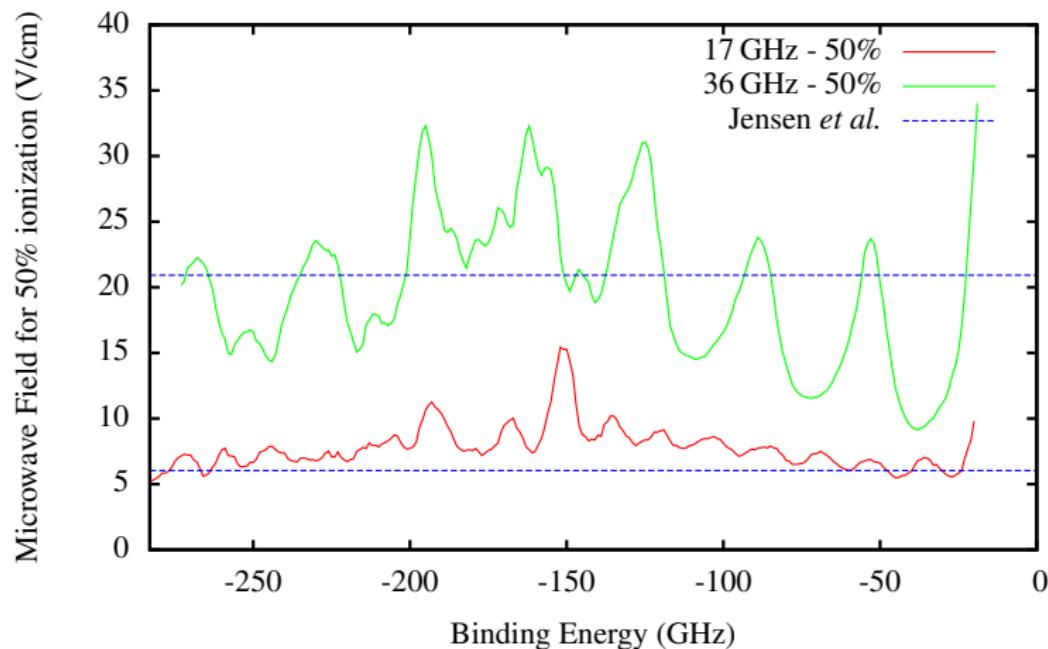
$$\text{State spacing} = \frac{1}{n^3}$$

MW ionization occurs when the Rabi width \geq state spacing

$$E = 2.4\omega^{5/3}$$

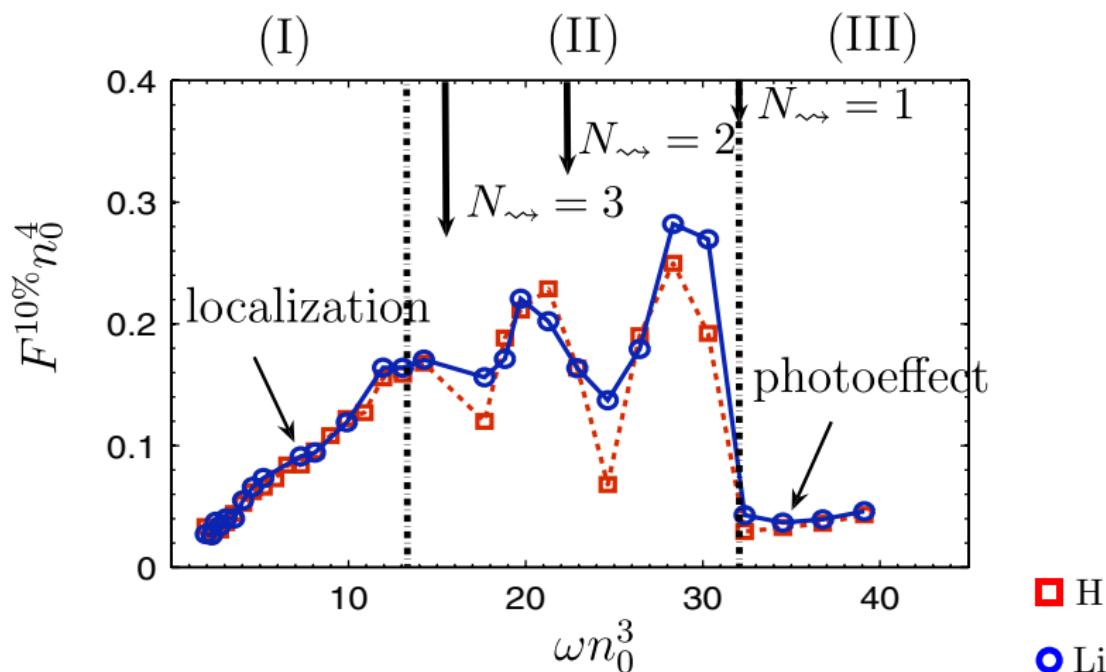
Jensen *et al.*, *Phys. Rev. Lett.* 62, (1989).

Jensen *et al.* Comparison



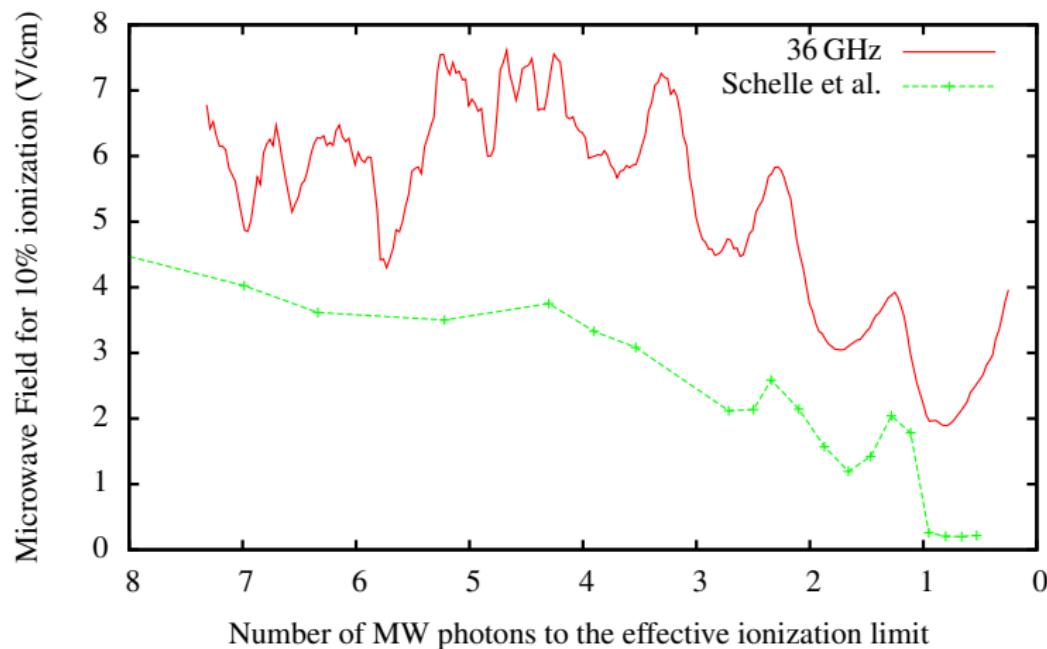
Jensen *et al.*, *Phys. Rev. Lett.* 62, (1989).

Schelle, Delande, and Buchleitner Model



Schelle, Delande, and Buchleitner, *Phys. Rev. Lett.* 102, (2009).

Schelle, Delande, and Buchleitner Comparison



Schelle, Delande, and Buchleitner, *Phys. Rev. Lett.* 102, (2009).

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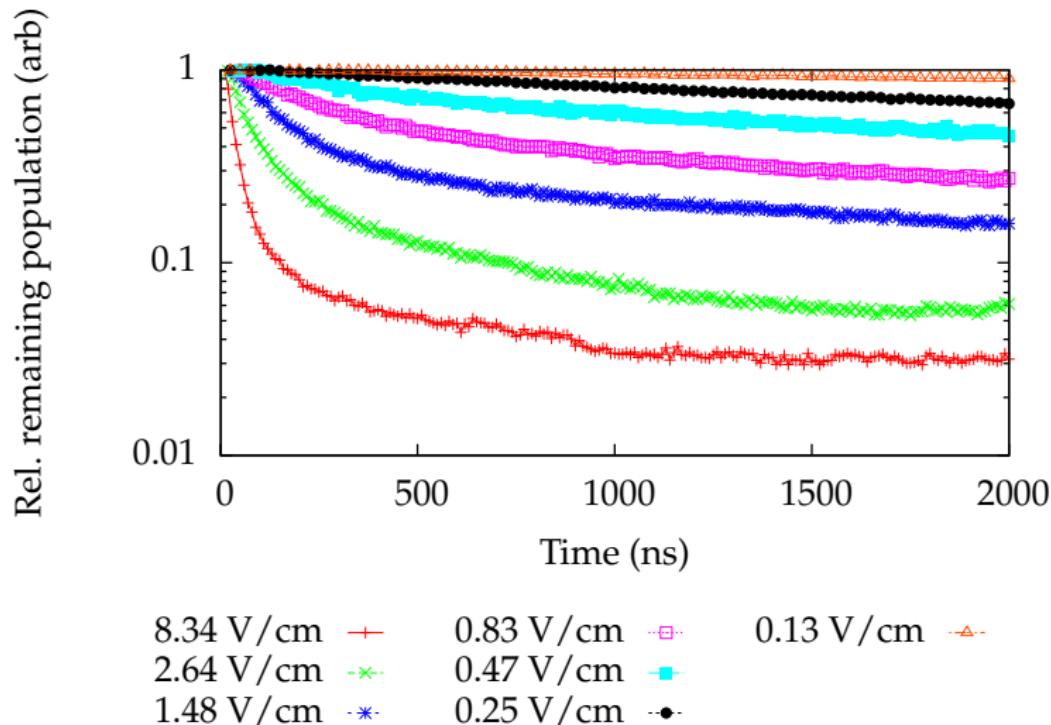
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Single Photon Ionization Rates

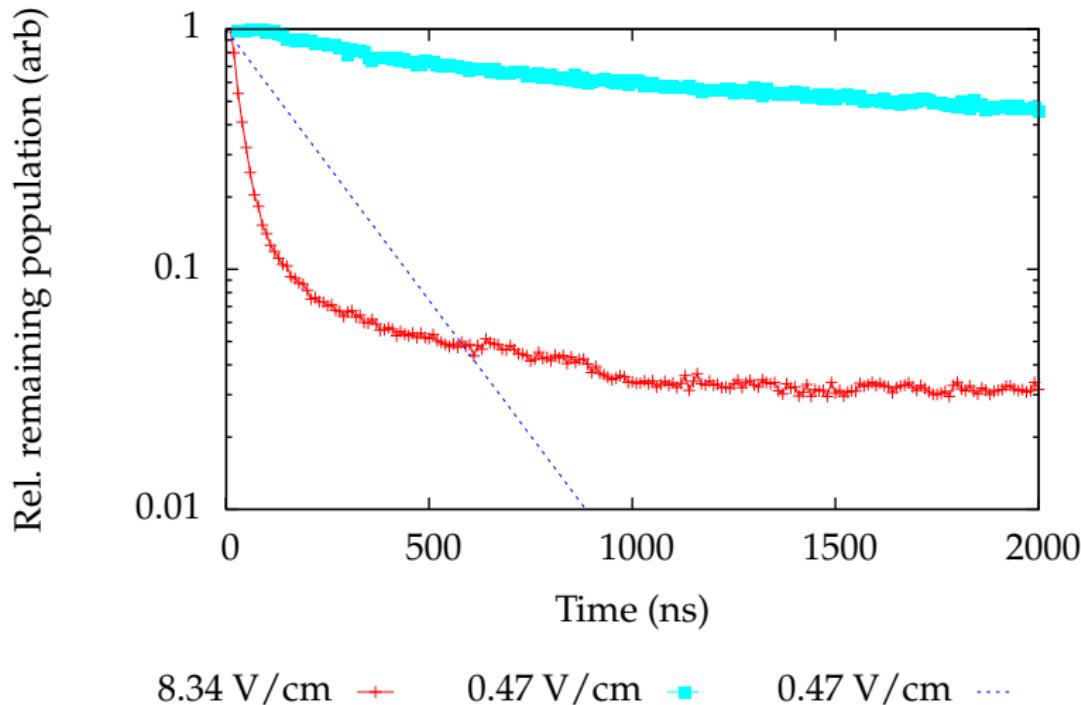
Above-Threshold Bound States

Photoionization - Timing

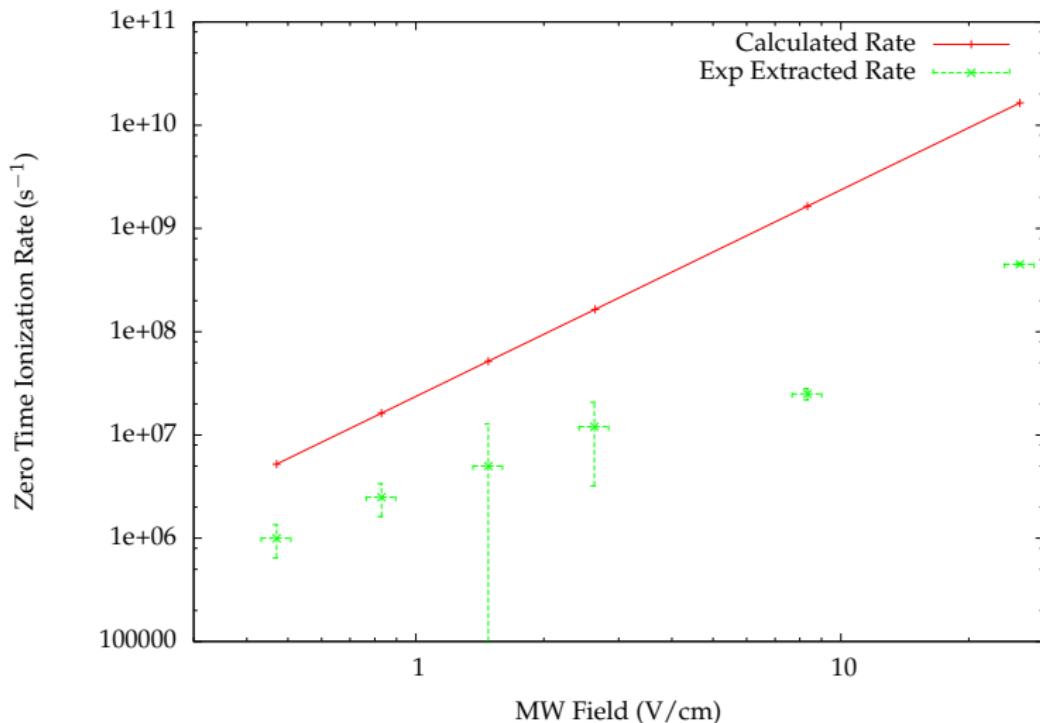
Single Photon Ionization



Single Photon Ionization



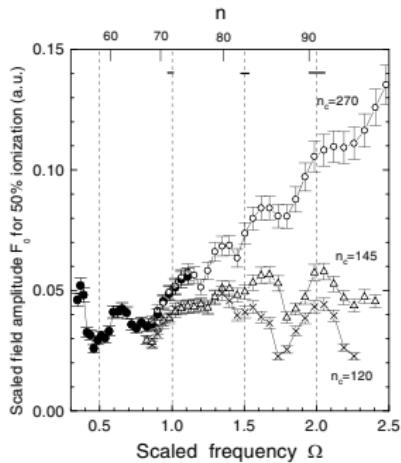
Fermi's Golden Rule Comparison



Influence of Stray Field

Stray fields limit the maximum n we can validly investigate

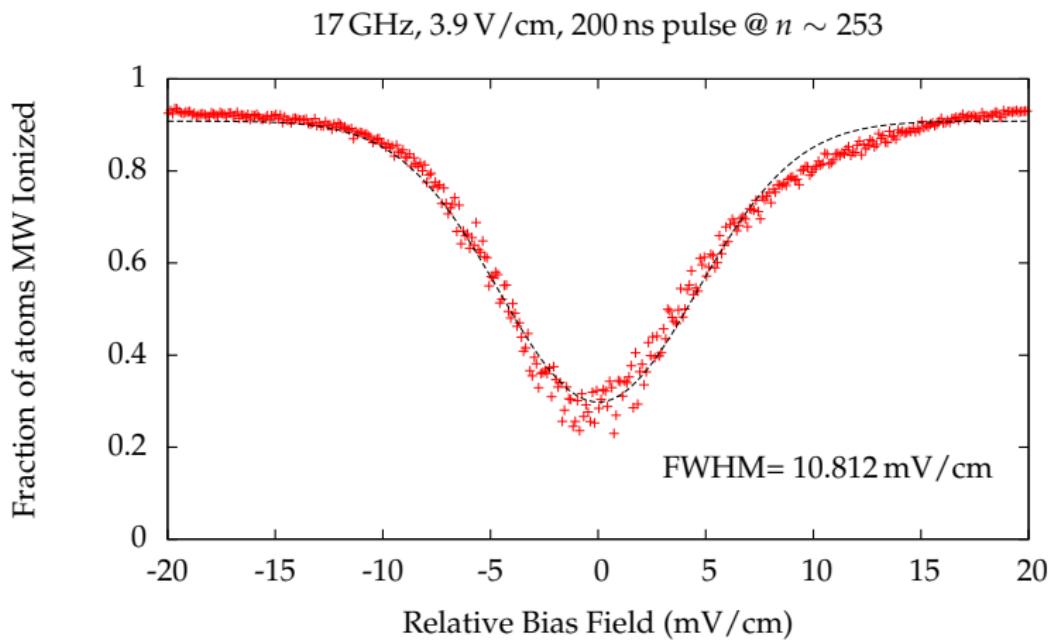
If we artificially depress n_c , we see that stray fields depress the microwave ionization threshold



New cut-off n has been increased from $n_c = 270$ to $n_c = 575$

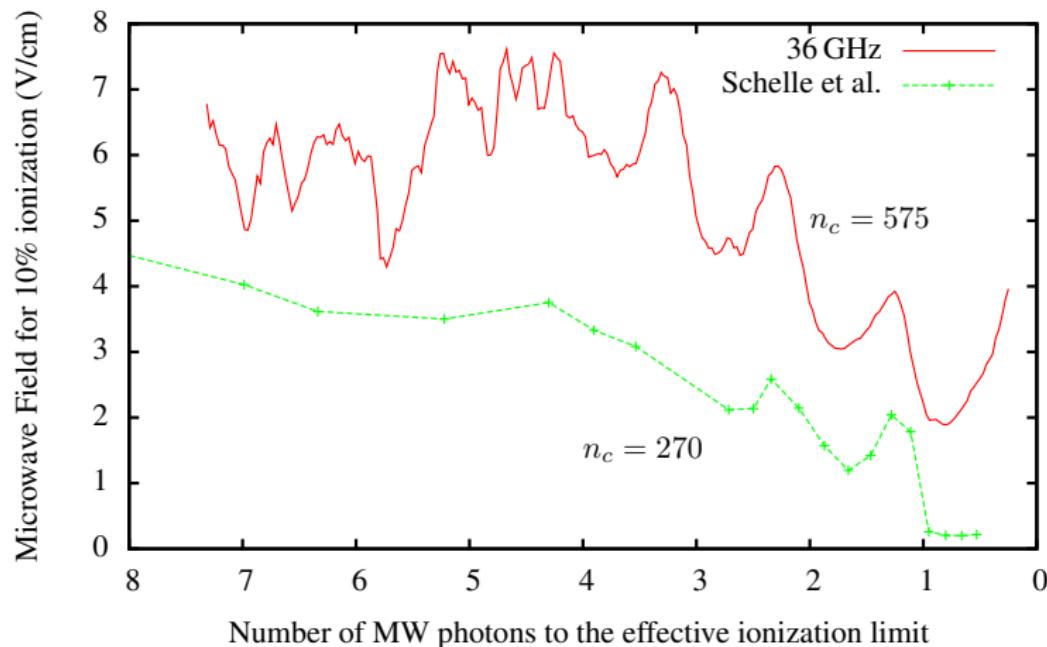
Maeda and Gallagher, *Phys. Rev. Lett.* 93, 2004.

Applied Bias Field



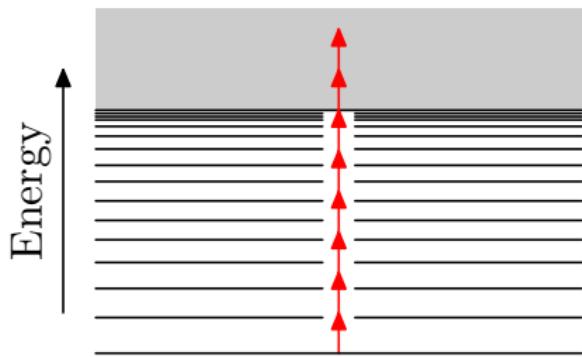
We can reduce stray field to below 3 mV/cm

Schelle, Delande, and Buchleitner Comparison



Schelle, Delande, and Buchleitner, *Phys. Rev. Lett.* 102, (2009).

Dressed Atom Picture



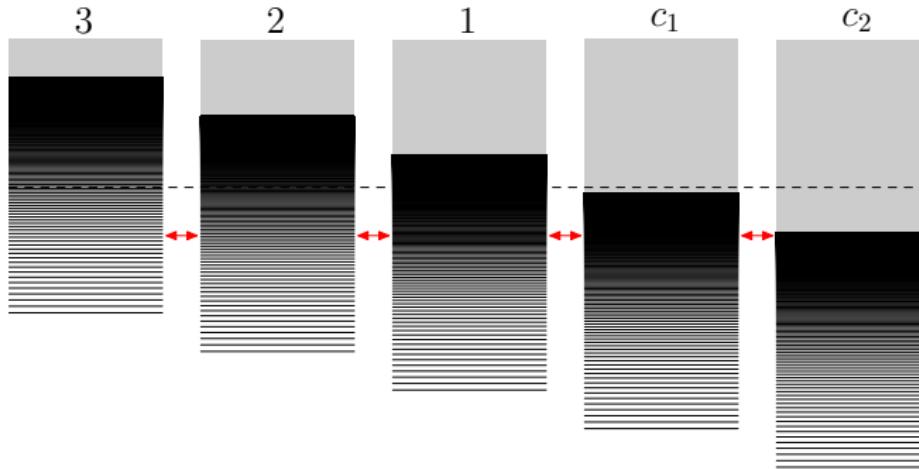
Can we simply use a Floquet picture to describe system?

How do we choose the correct levels to include?

How do we include the above-threshold continuum states?

MQDT-Floquet Model

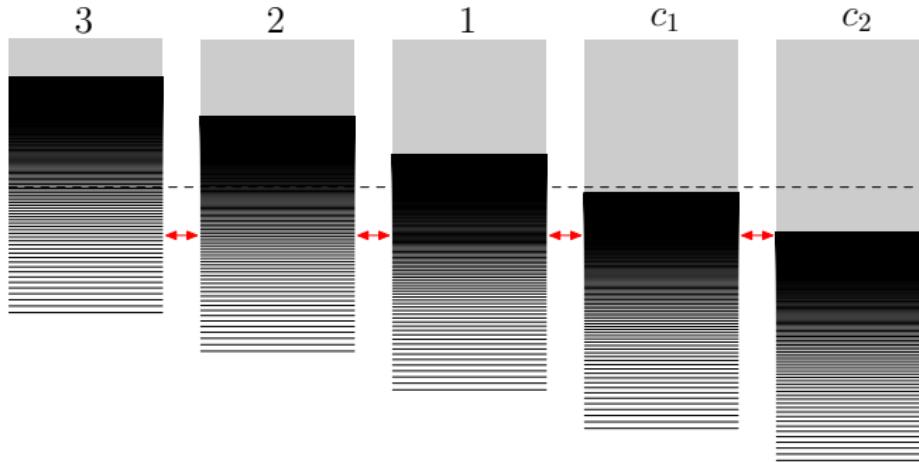
Giusti-Suzor and Zoller proposed a Floquet-MQDT model for Rydberg atoms in laser fields where the couplings between channels are radiative dipole couplings. The Rydberg electron is in a Coulomb potential, and can only scatter to other states near the core.



Giusti-Suzor and Zoller, *Phys. Rev. A* 36, (1987).

MQDT-Floquet Model

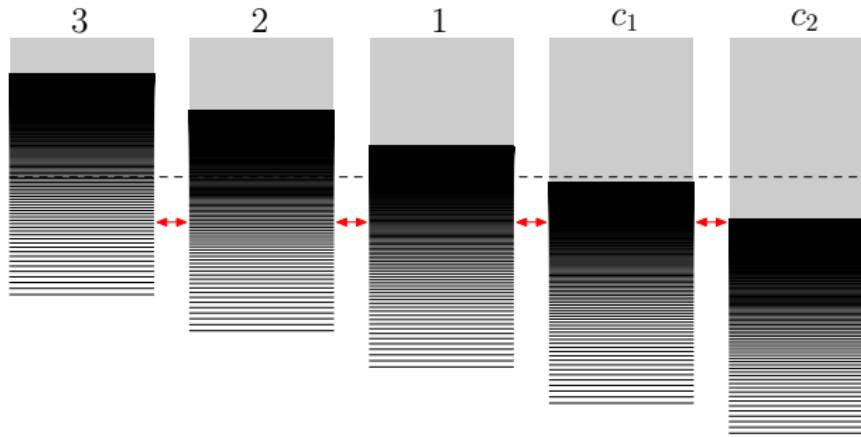
We can define a series of channels, each separated by one microwave photon.



The coupling between channels are radial dipole couplings - the same as the level coupling used by Jensen *et al.*

MQDT-Floquet Model

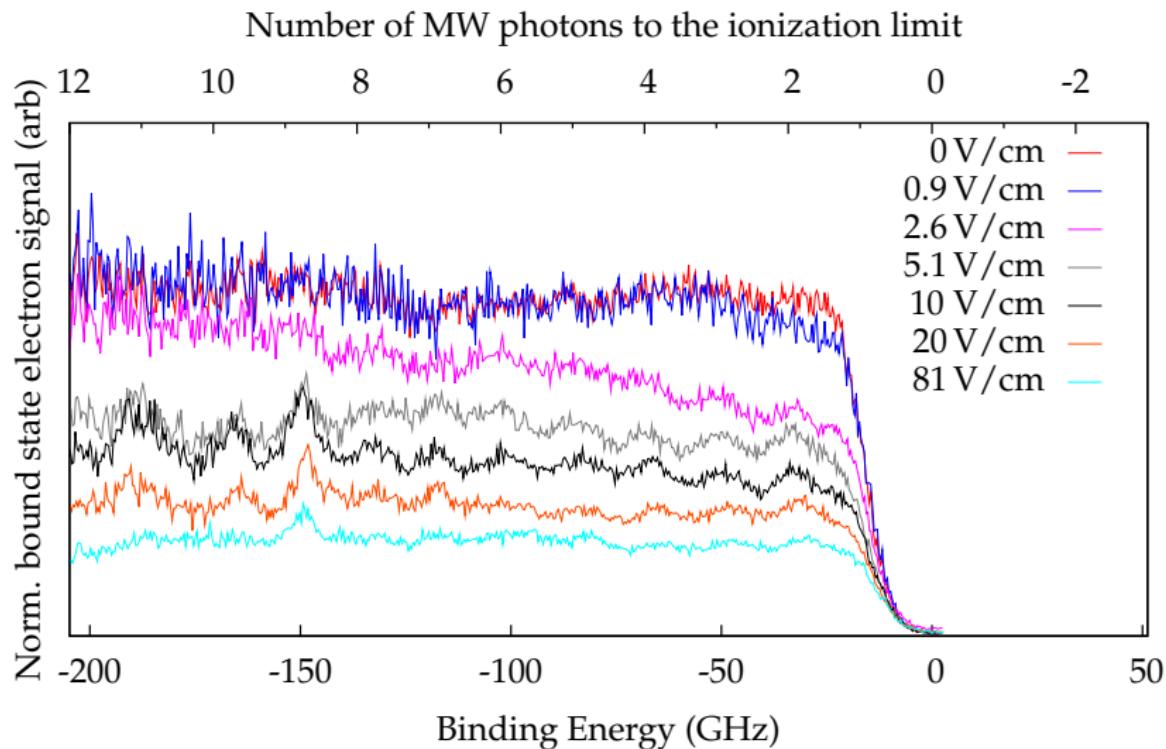
Following the method of Cooke and Cromer, we can easily calculate the time-dependent transfer of the bound channel populations to the continuum along the dashed line.



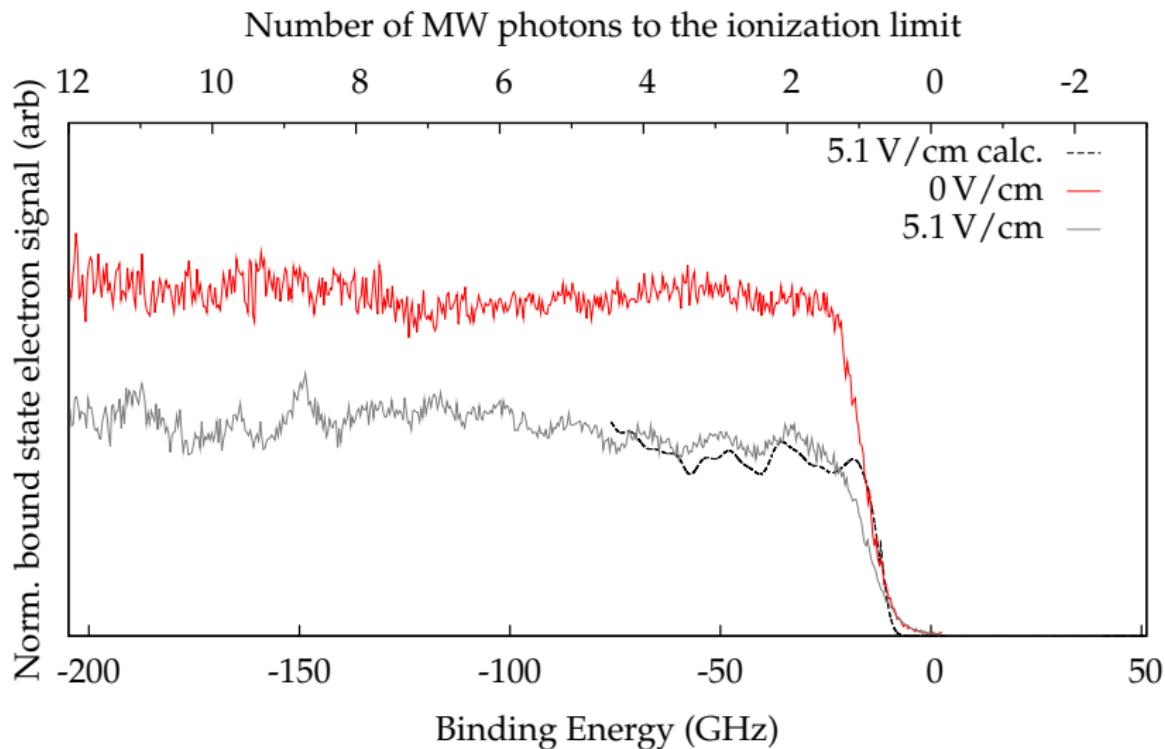
By iterating the binding energy (the dashed line) we can compute the ionization spectrum

Cooke and Cromer, *Phys. Rev. A* 32, (1985).

Model Comparison



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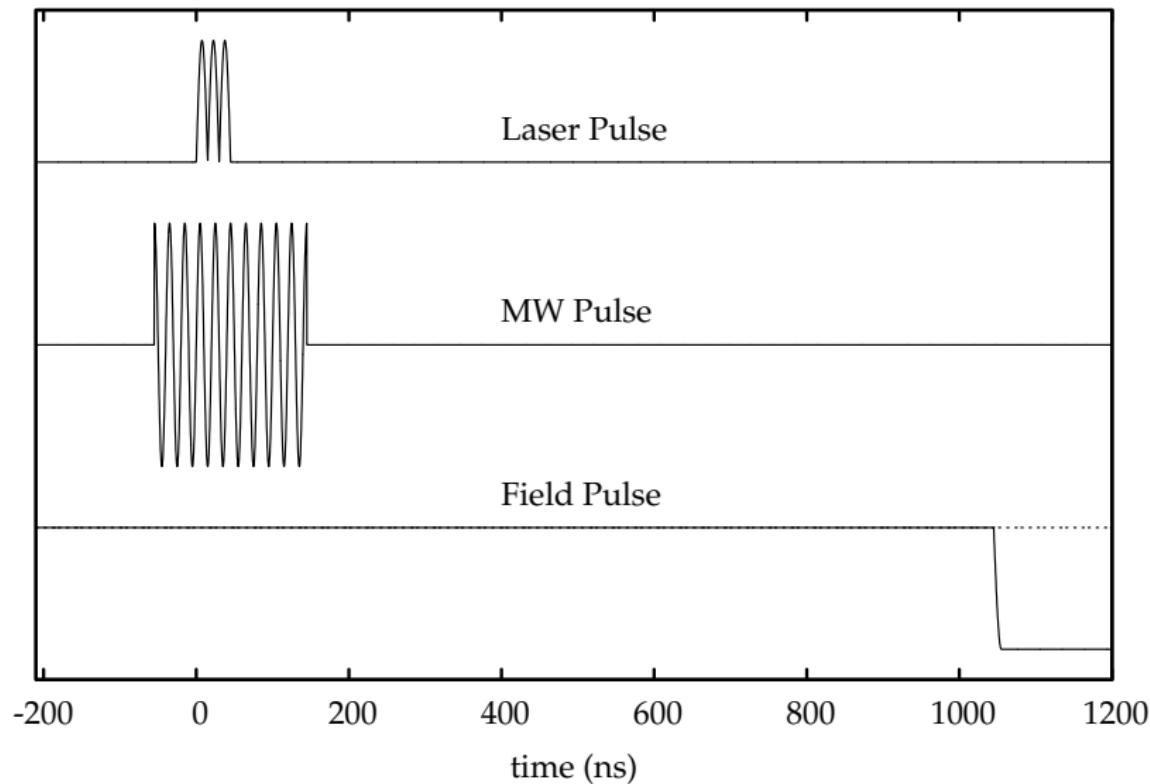
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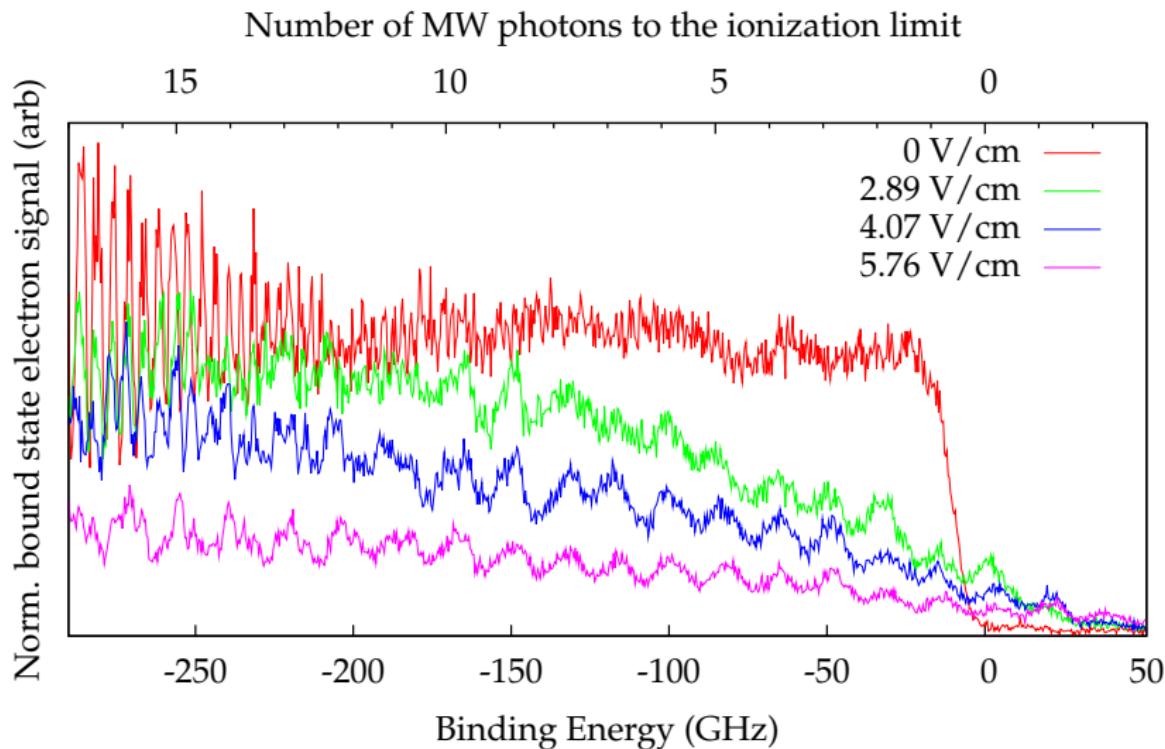
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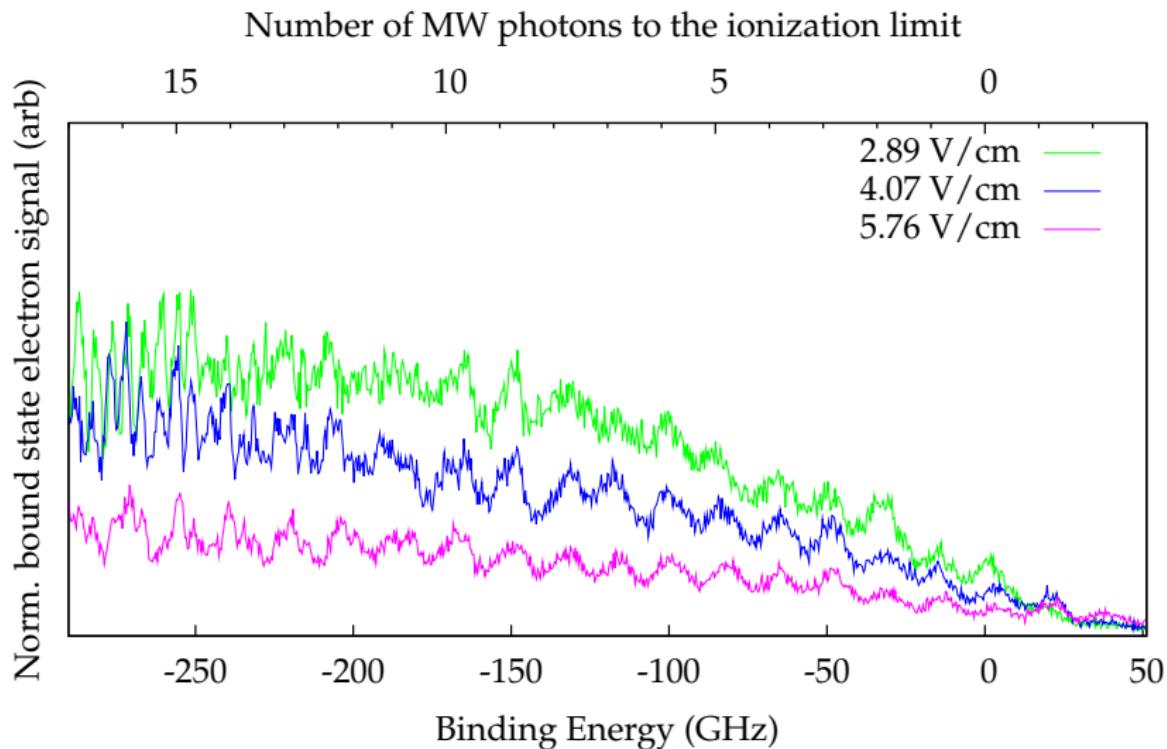
Above-Threshold Bound States - Timing



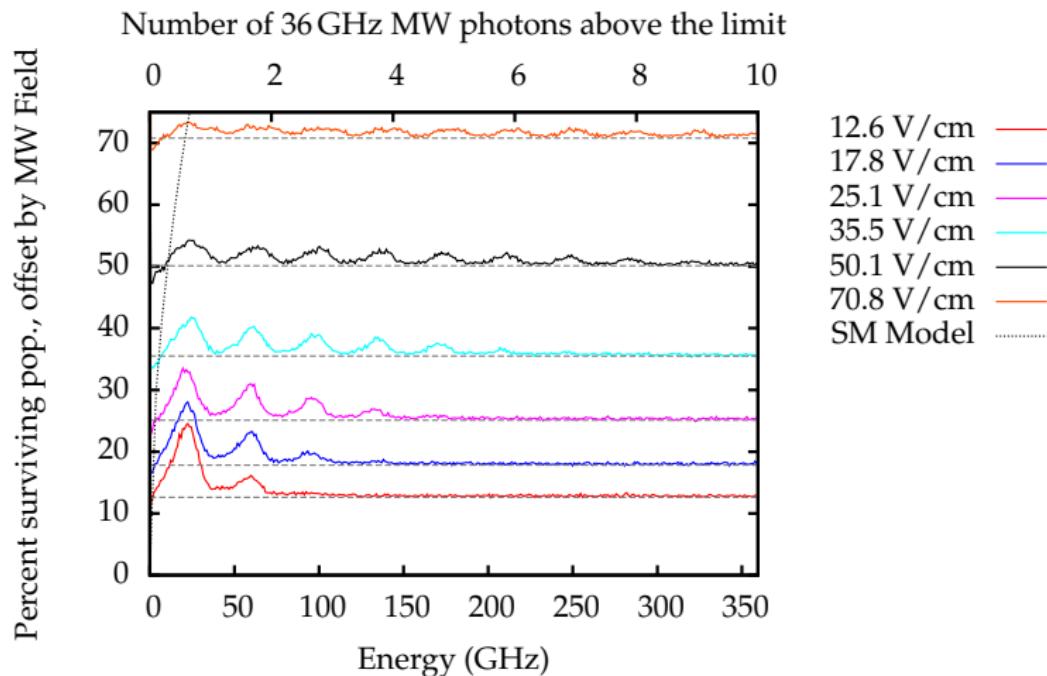
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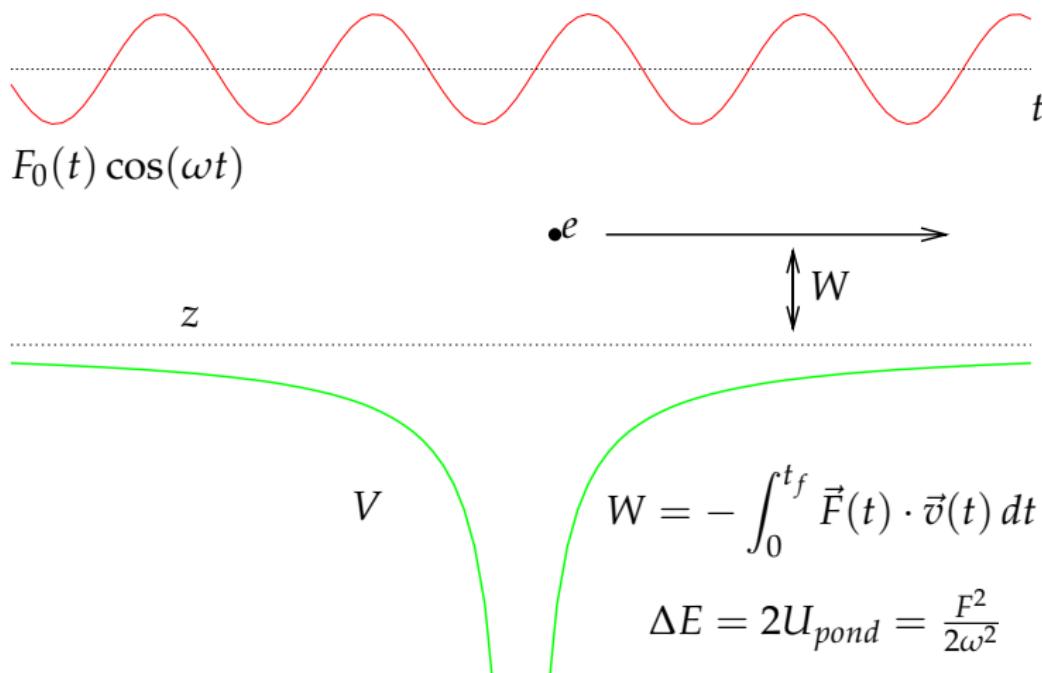
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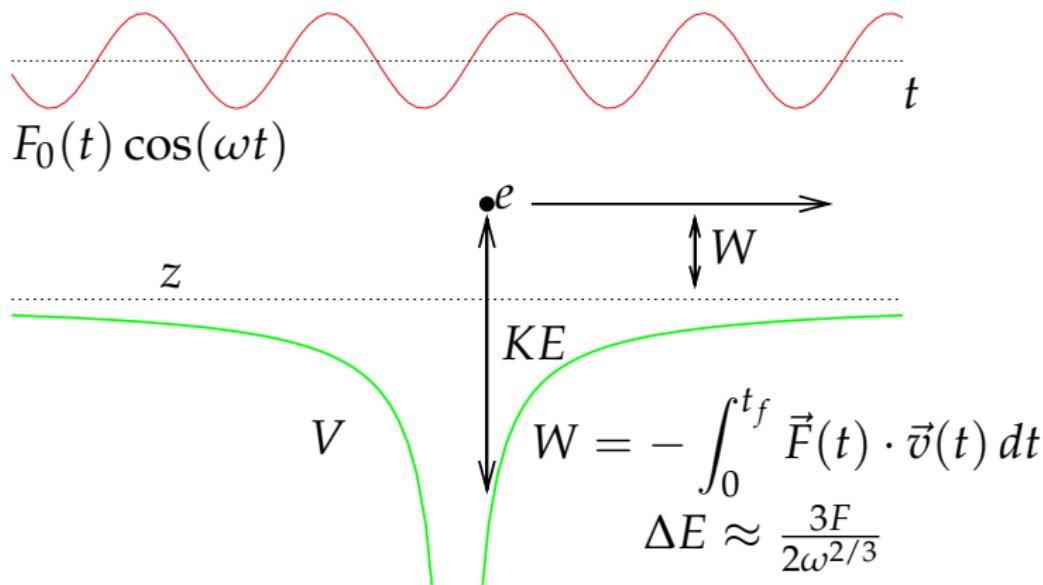
Above-Threshold Bound States



Simpleman's Model

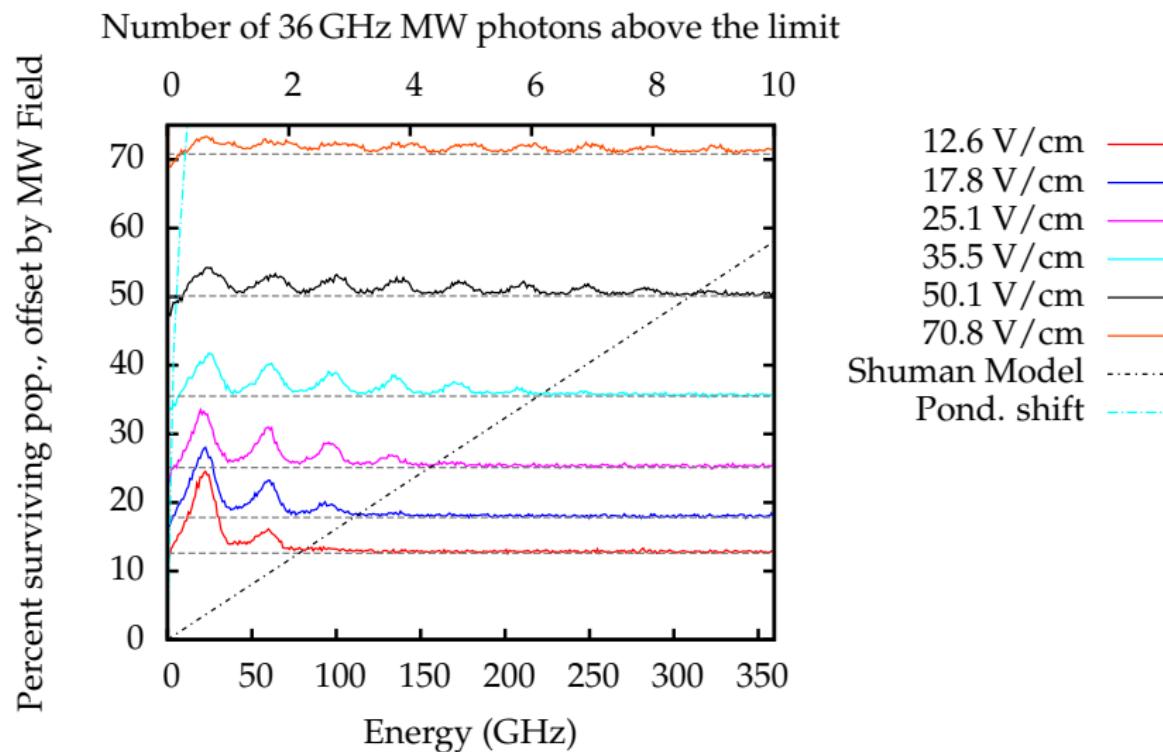


New Classical Model



Shuman et al., *Phys. Rev. Lett.* 101, (2009).

Above-Threshold Bound States



Conclusions

- ▶ Anderson Localization model fits experimental results
- ▶ There is no experimentally accessible regime where Fermi's Golden Rule applies
- ▶ The coherent coupling of levels both above and below the ionization limit describes high scaled frequency microwave ionization
- ▶ A simple classical model illustrates population transfer from above the limit to bound states