Rydberg atoms

Tobias Thiele

References

- T. Gallagher: Rydberg atoms

Content

- Part 1: Rydberg atoms

- Part 2: A typical beam experiment

Introduction – What is „Rydberg“?

- Rydberg atoms are (any) atoms in state with high principal quantum number *n*.

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- Rydberg atoms are (any) atoms with exaggerated properties

Introduction – What is „Rydberg“?

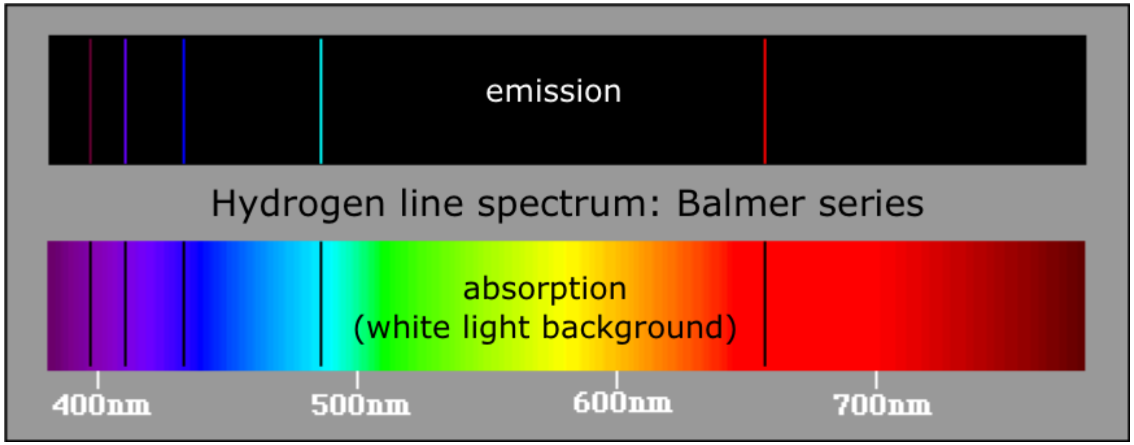
- Rydberg atoms are (any) atoms in state with high principal quantum number *n.*

- Rydberg atoms are (any) atoms with exaggerated properties

➡ *equivalent!*

Introduction – How was it found?

- In 1885: Balmer series:
  - Visible absorption wavelengths of H:



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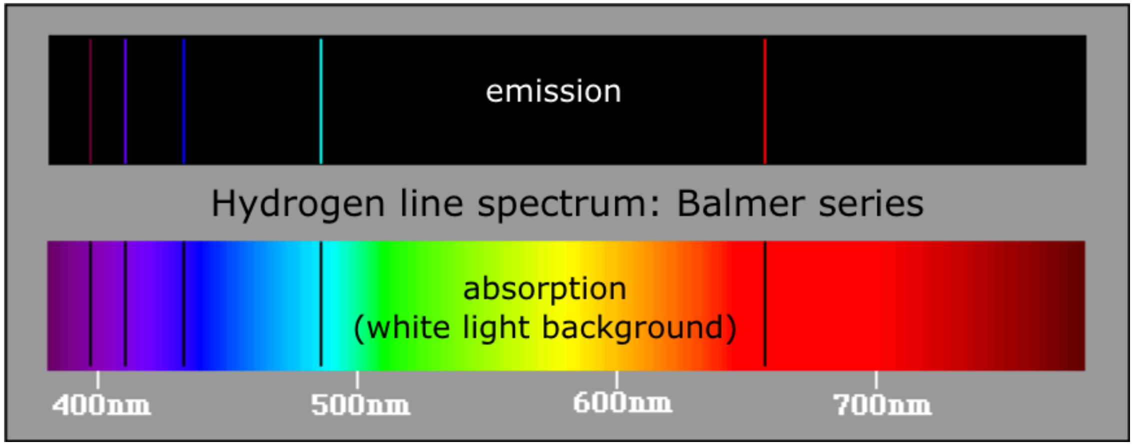

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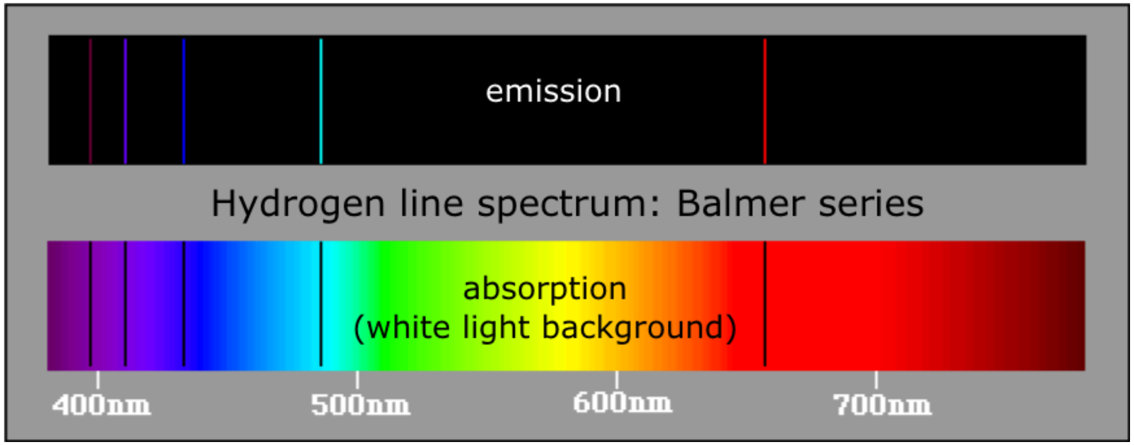
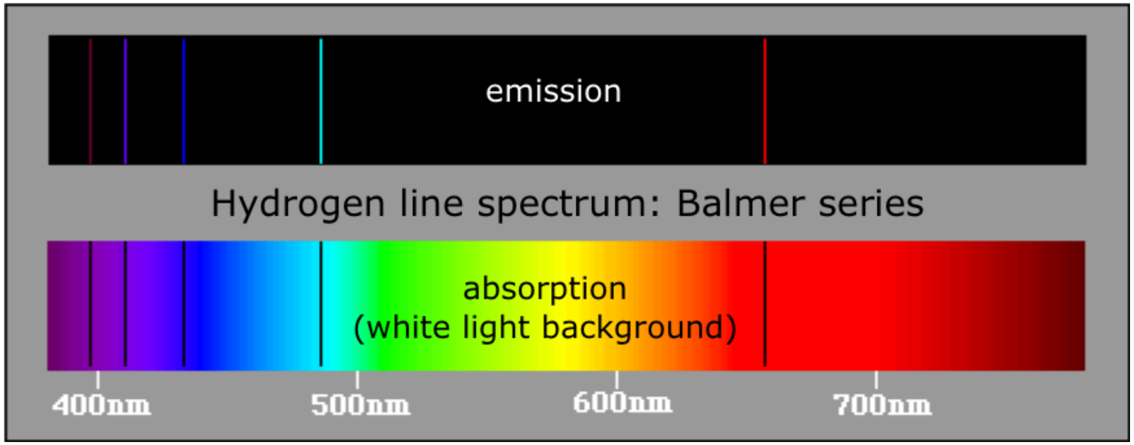


$$\lambda = \frac{bn^2}{n^2 - 4}$$

  - Other series discovered by Lyman, Brackett, Paschen, ...
  - Summarized by Johannes Rydberg: $\tilde{\nu} = \tilde{\nu}_\infty - \frac{Ry}{n^2}$

Introduction – Generalization

- In 1885: Balmer series:
  - Visible absorption wavelengths of H:



$$\lambda = \frac{bn^2}{n^2 - 4}$$

  - Other series discovered by Lyman, Brackett, Paschen, ...
  - Quantum Defect was found for other atoms: $\tilde{\nu} = \tilde{\nu}_\infty - \frac{Ry}{(n-\delta_l)^2}$

Introduction – Rydberg formula?

- Energy follows Rydberg formula:

$$E = E_\infty - \frac{hRy}{(n - \delta_l)^2}$$

Introduction – Rydberg formula?

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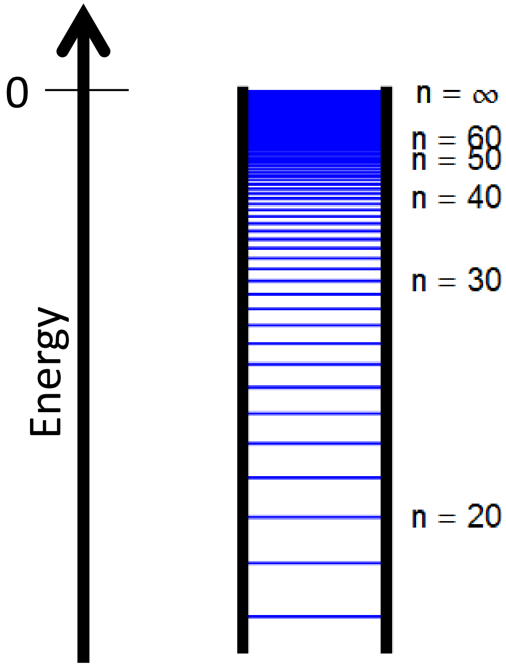
$$E = E_{\infty} - \frac{hRy}{(n-\delta_l)^2}$$

$hRy \approx 13.6$ eV

Introduction – Hydrogen?

- Energy follows Rydberg formula:

$$E = E_\infty - \frac{hRy}{(n-\delta_l)^2} = -\frac{hRy}{n^2}$$



Quantum Defect?

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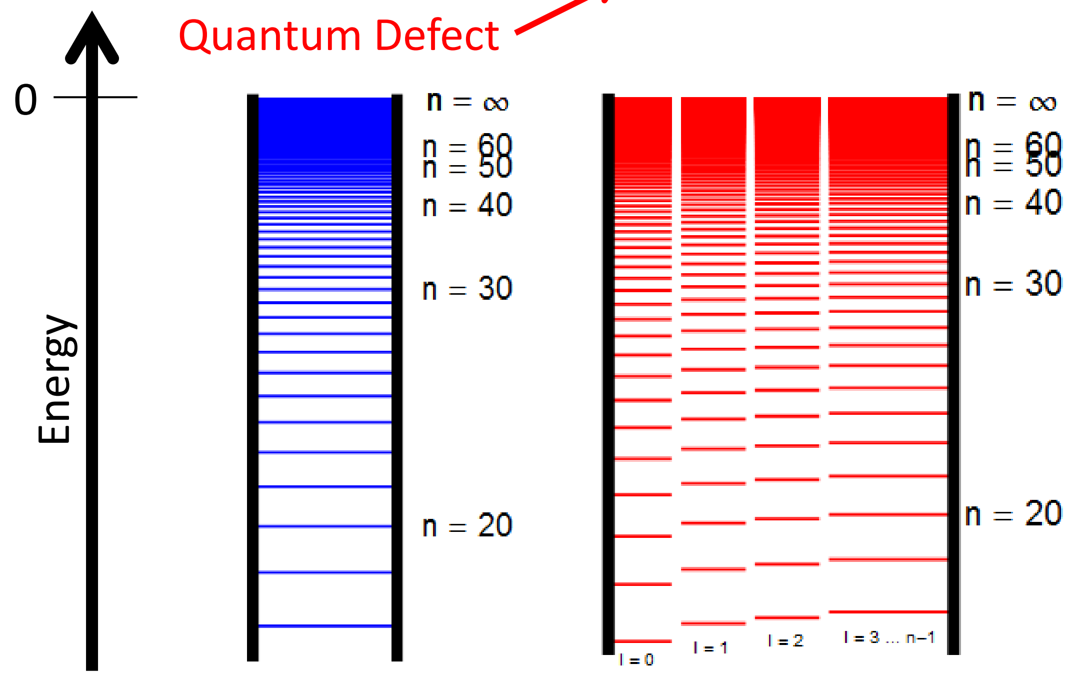
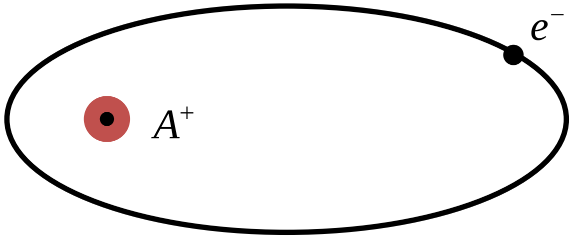
Quantum Defect



Rydberg Atom Theory

- Rydberg Atom



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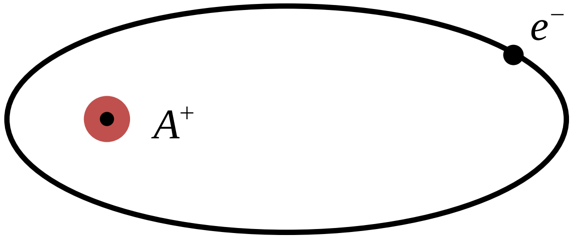

- Almost like Hydrogen
  - Core with one positive charge
  - One electron

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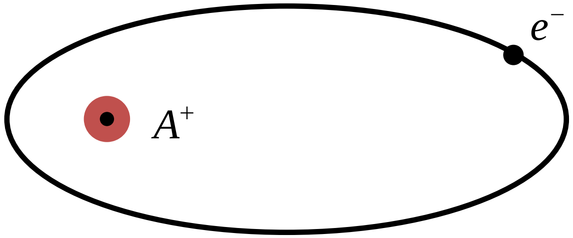

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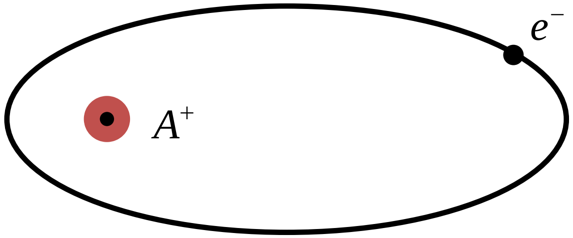
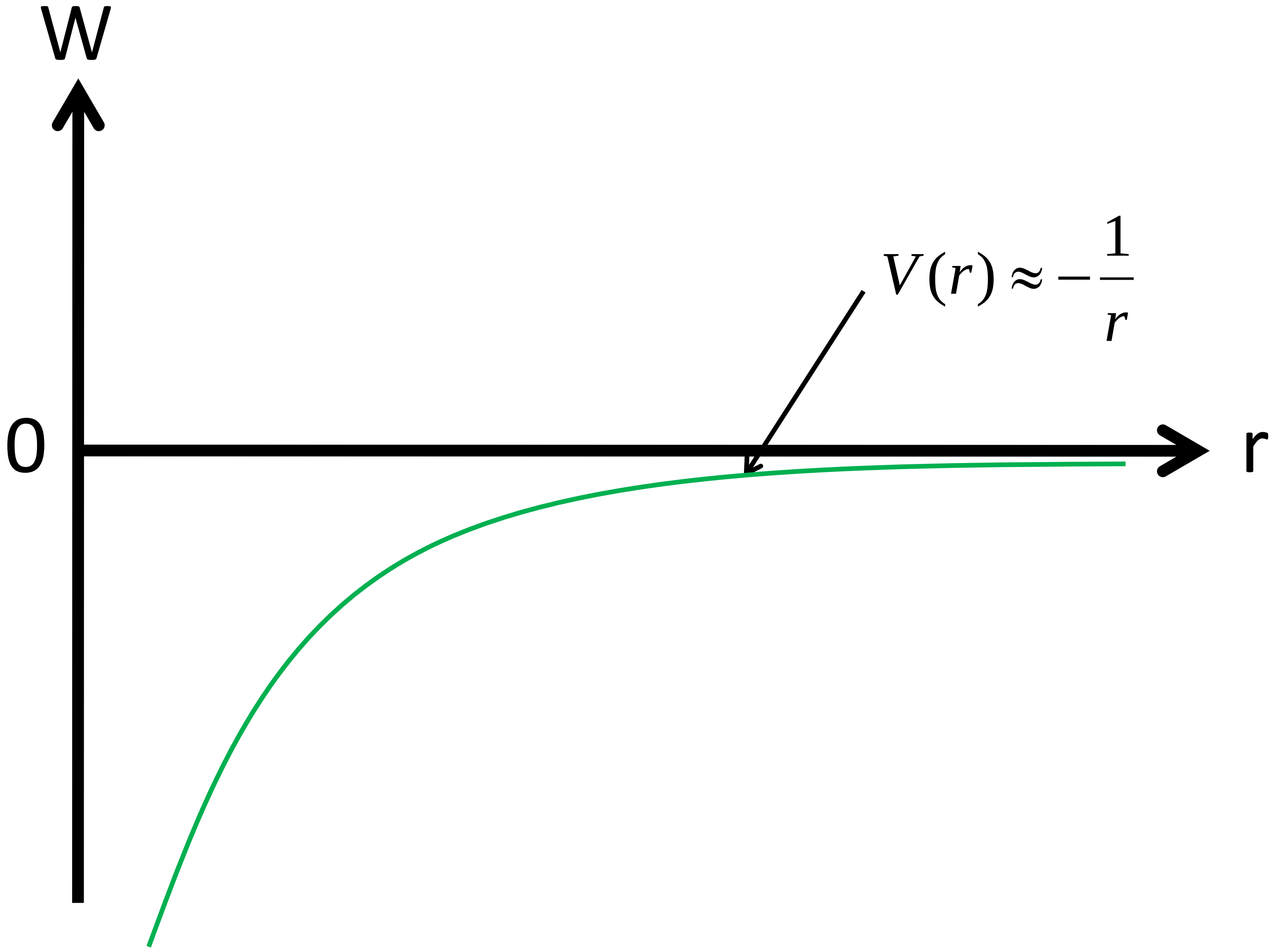


- Almost like Hydrogen
  - Core with one positive charge
  - One electron
- What is the difference?
  - No difference in angular momentum states

Radial parts-Interesting regions



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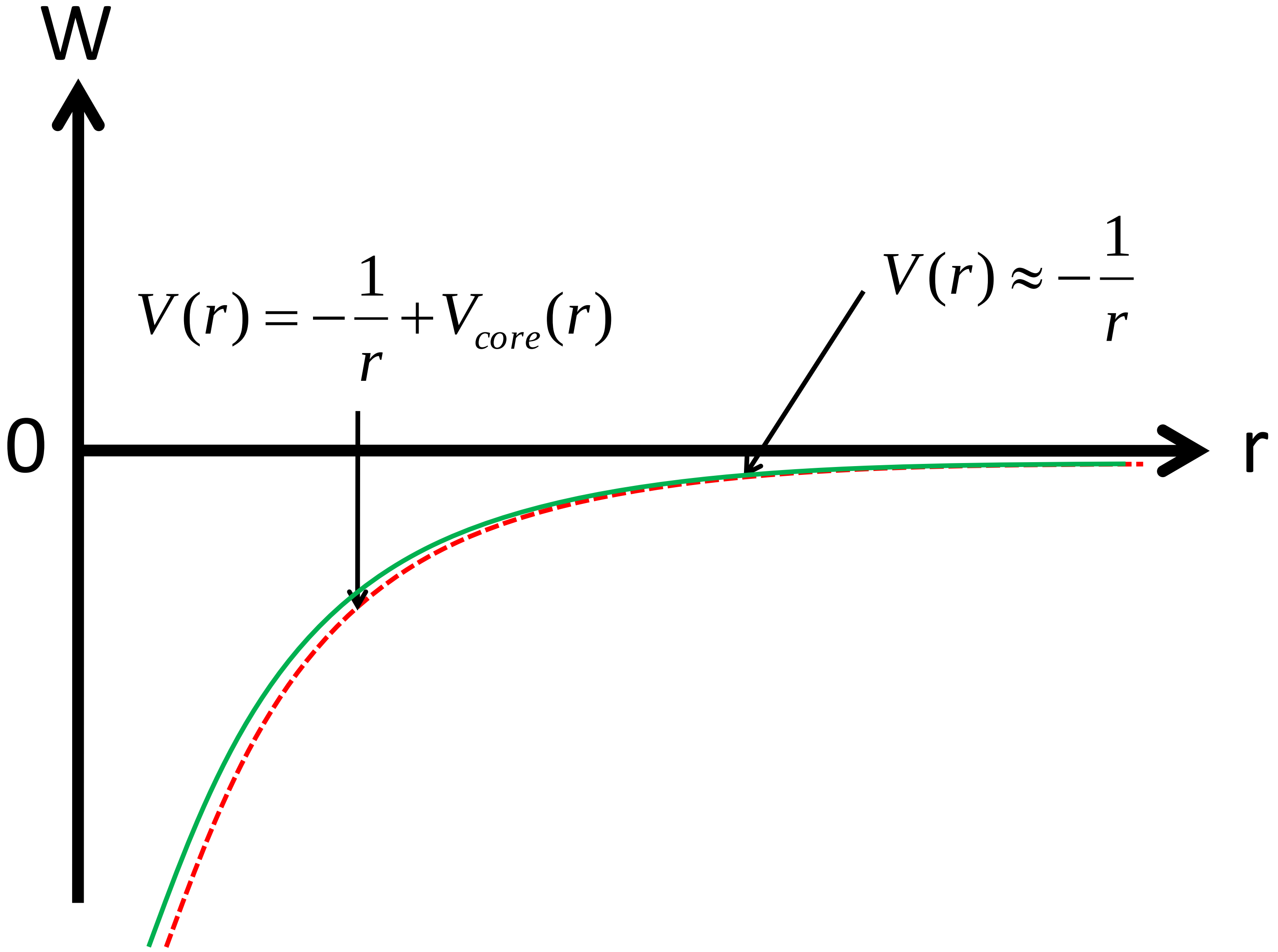

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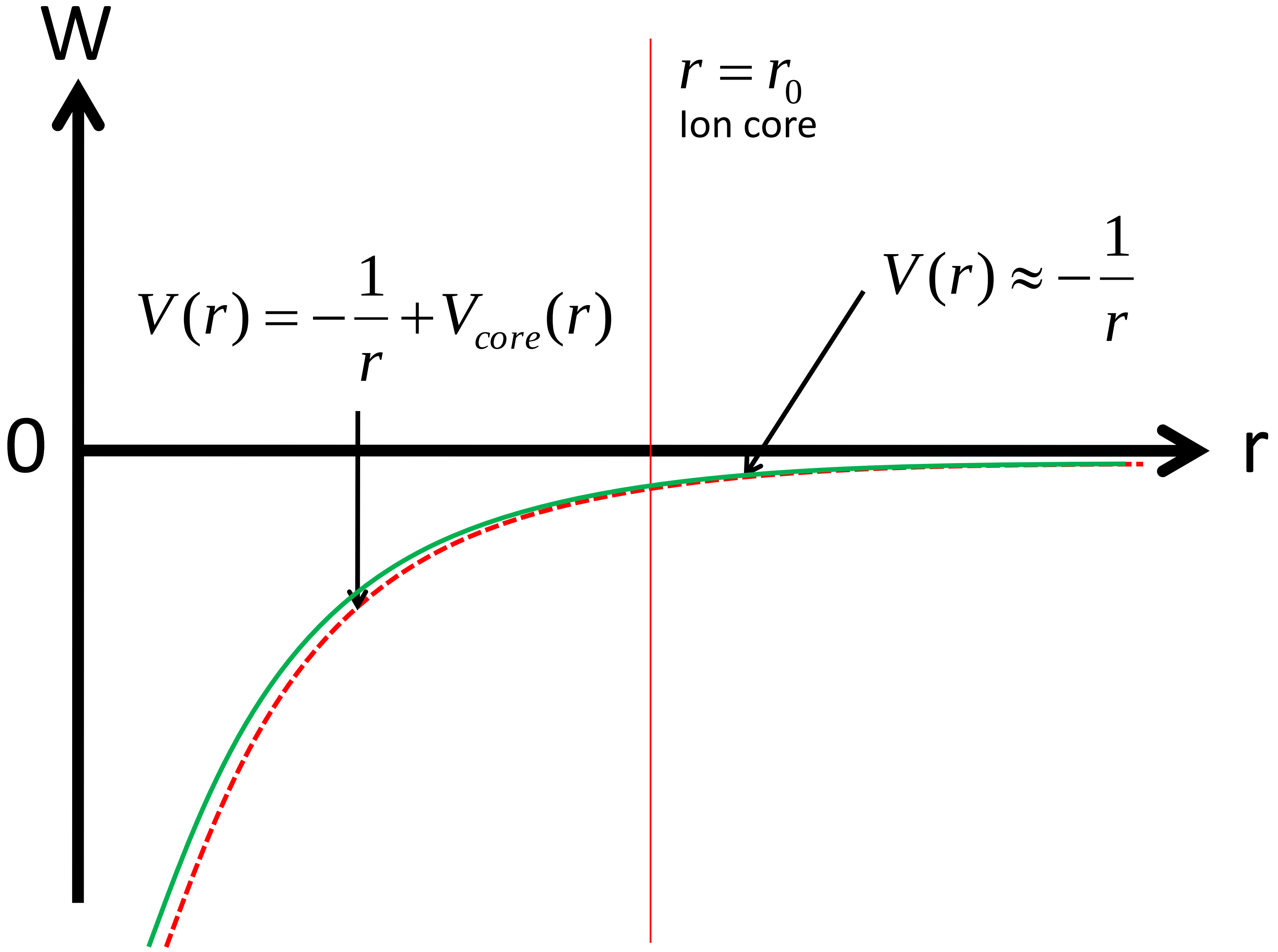

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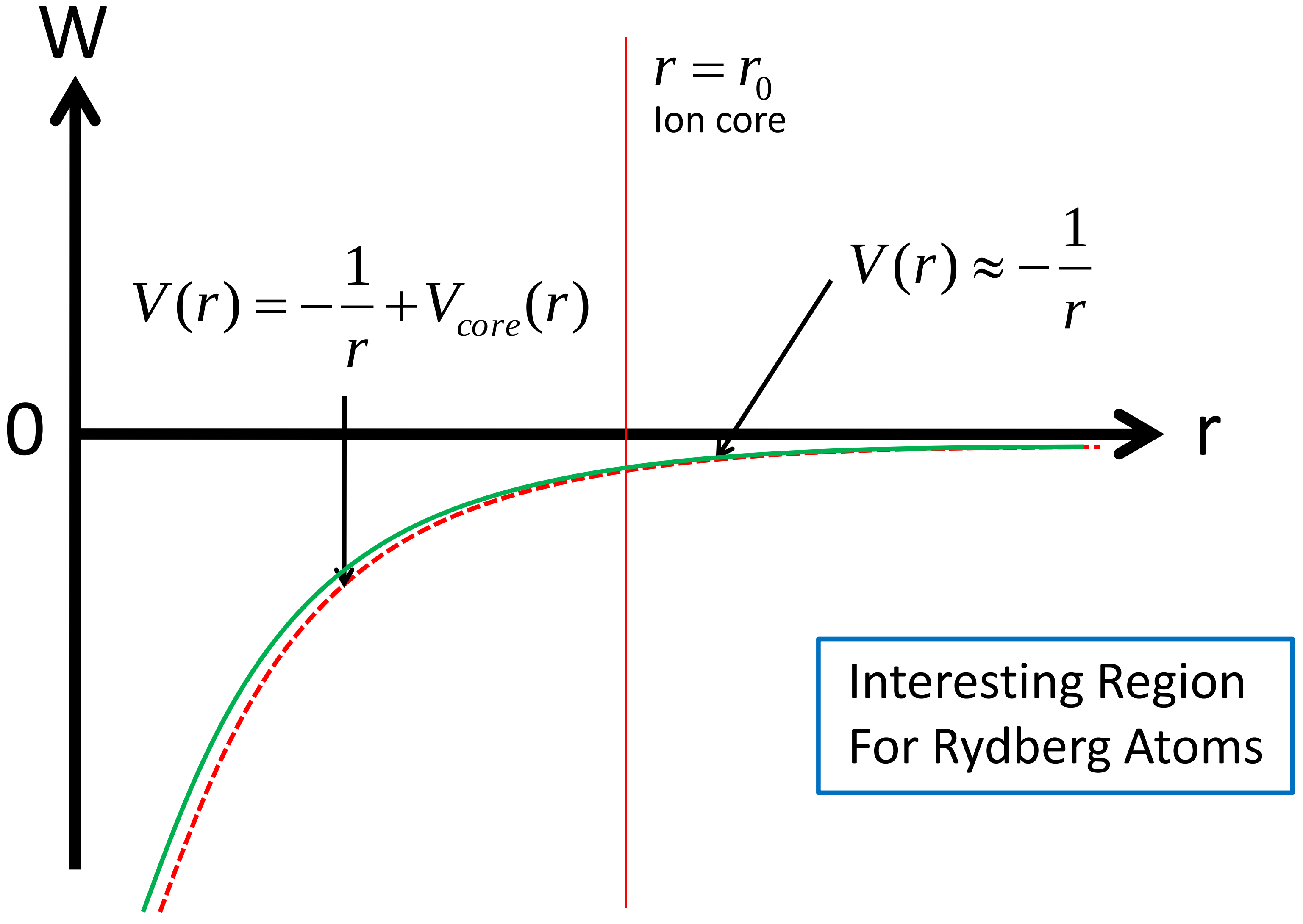

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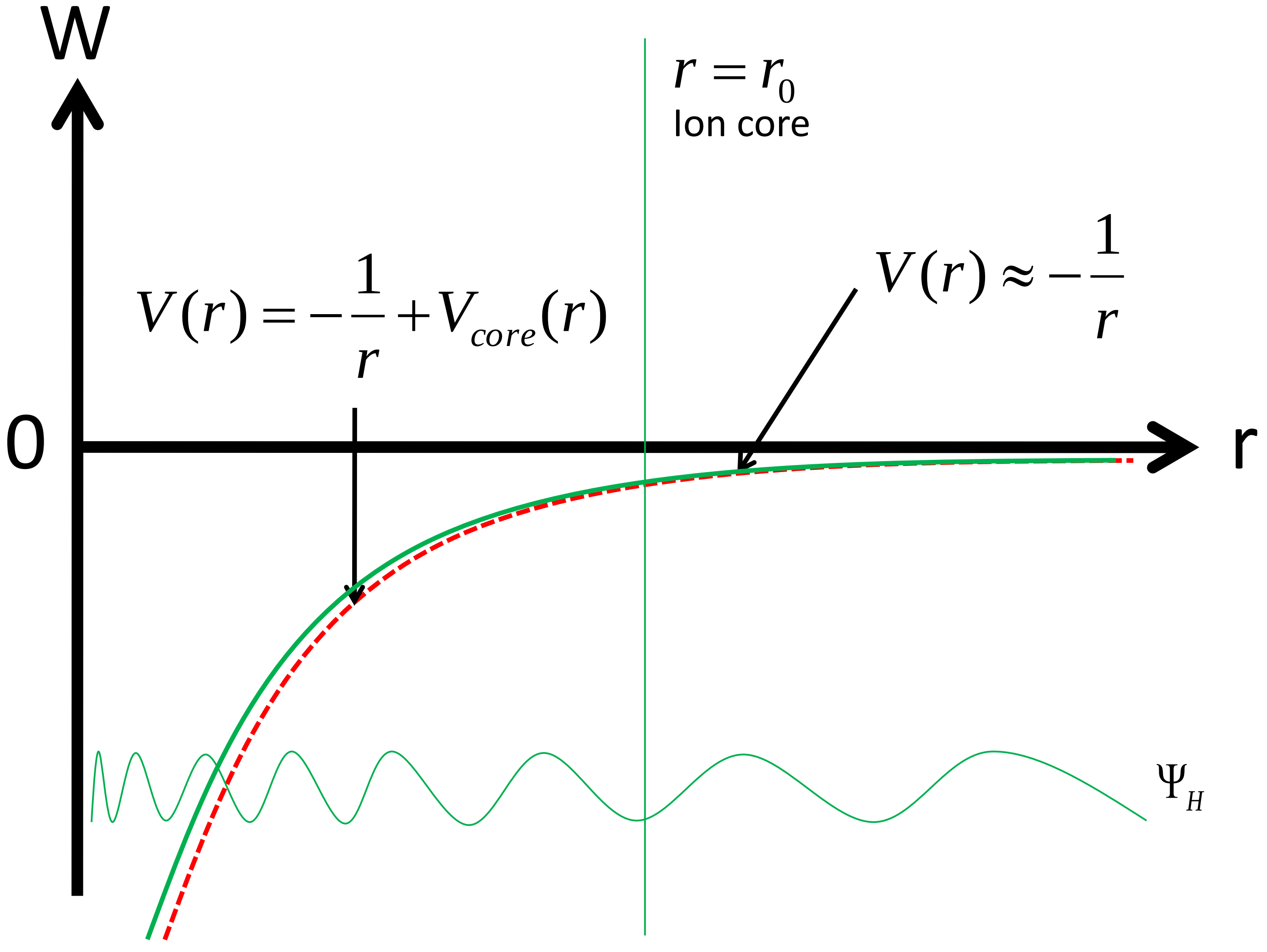

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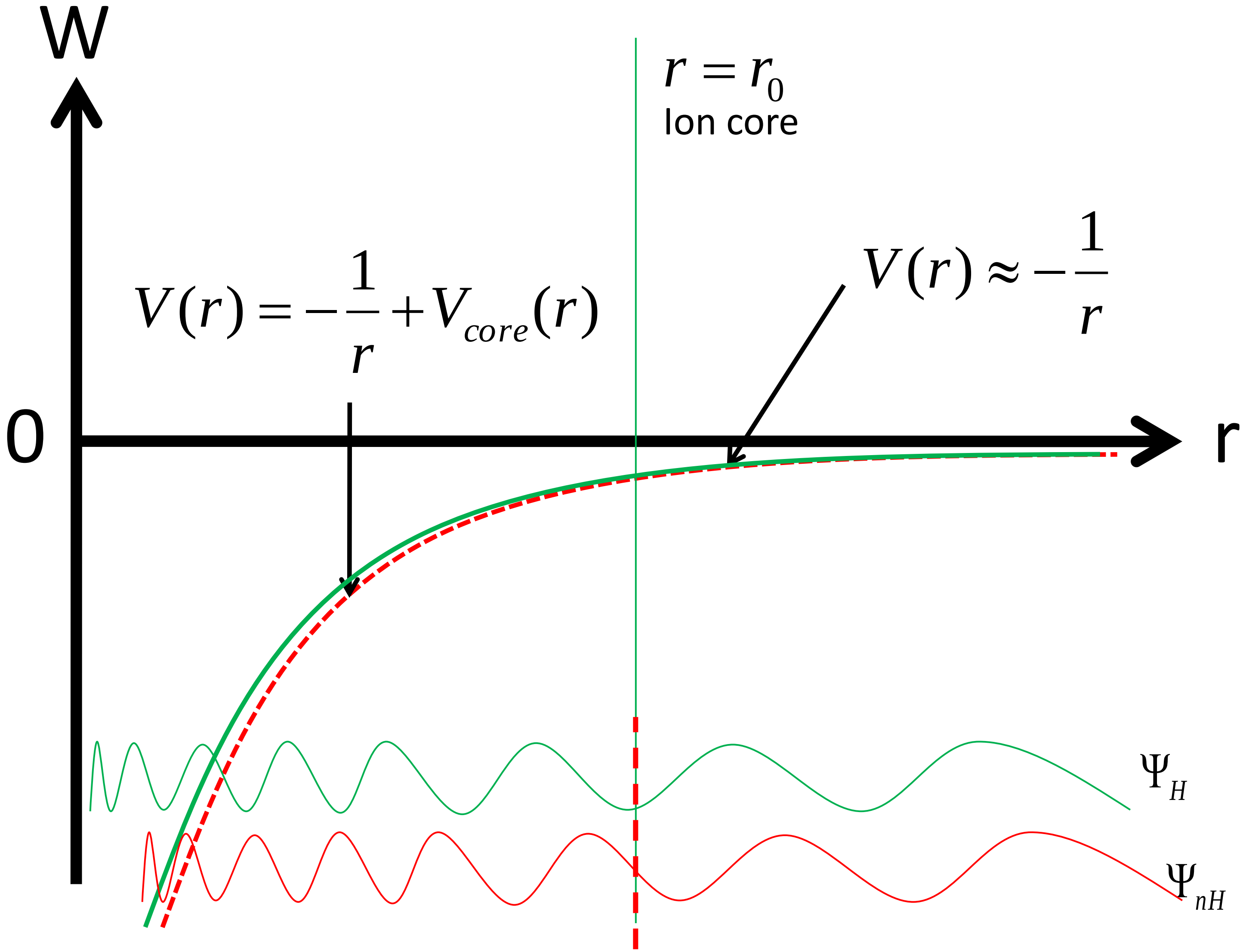

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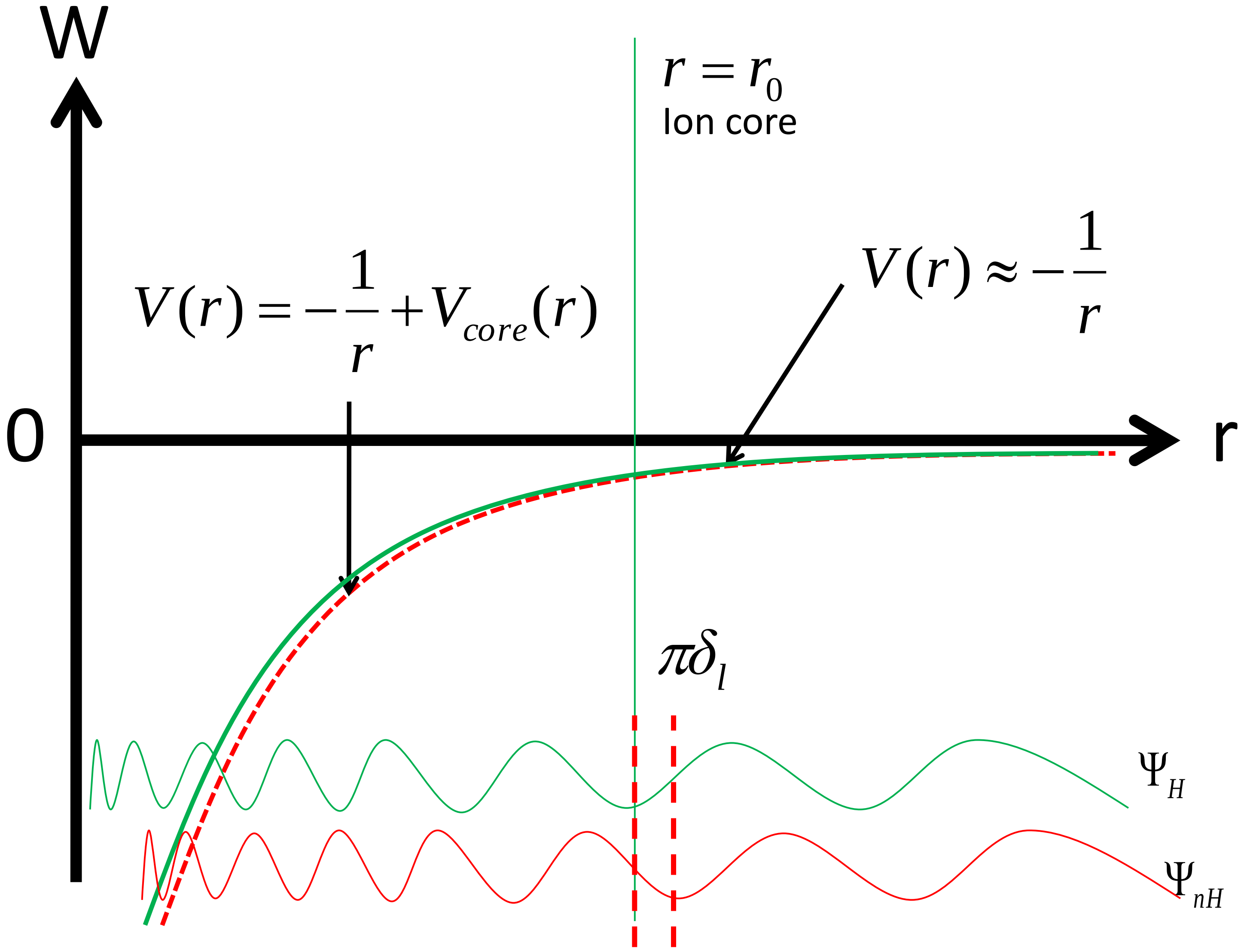

(Helium) Energy Structure

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

- δ_l usually measured
  - Only large for low l (s,p,d,f)

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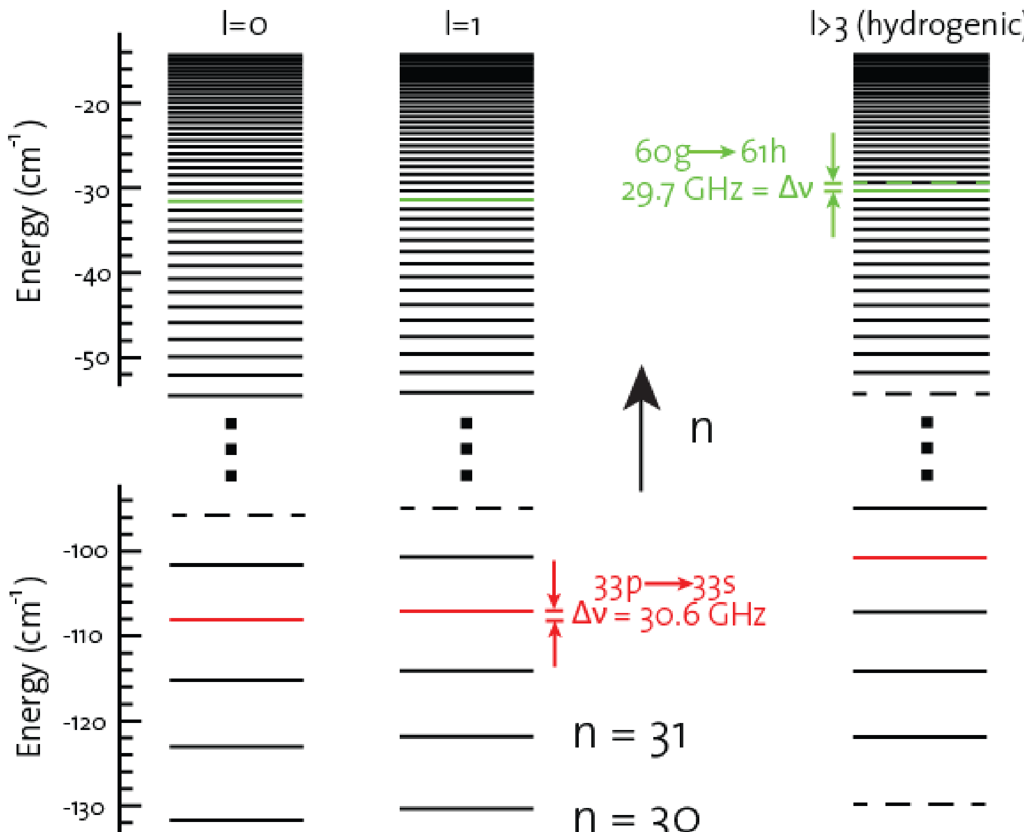

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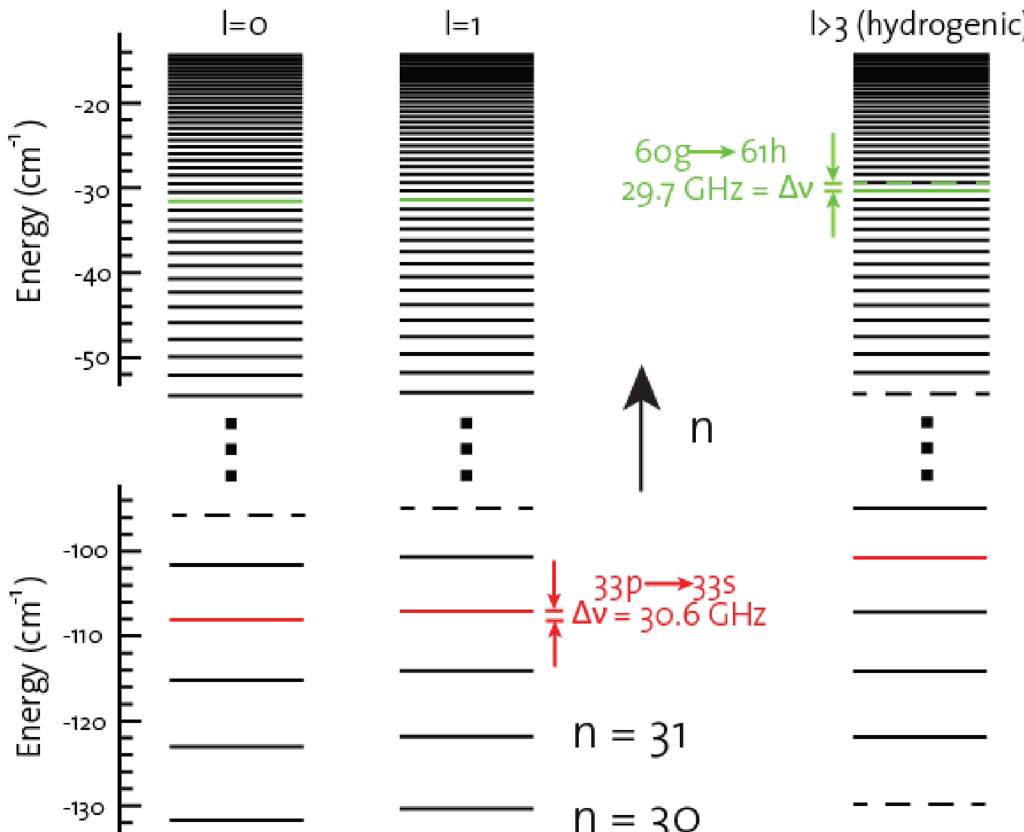

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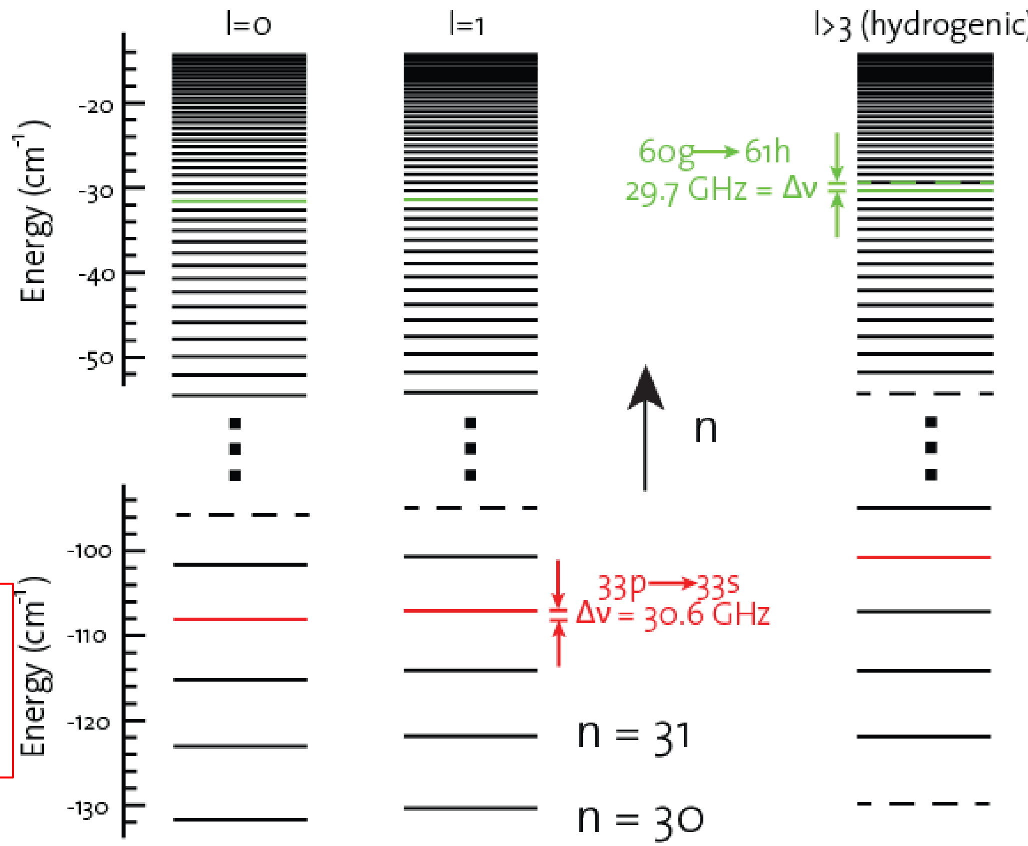
Excentric orbits penetrate into core.
Large deviation from Coulomb.
Large phase shift-> large quantum defect



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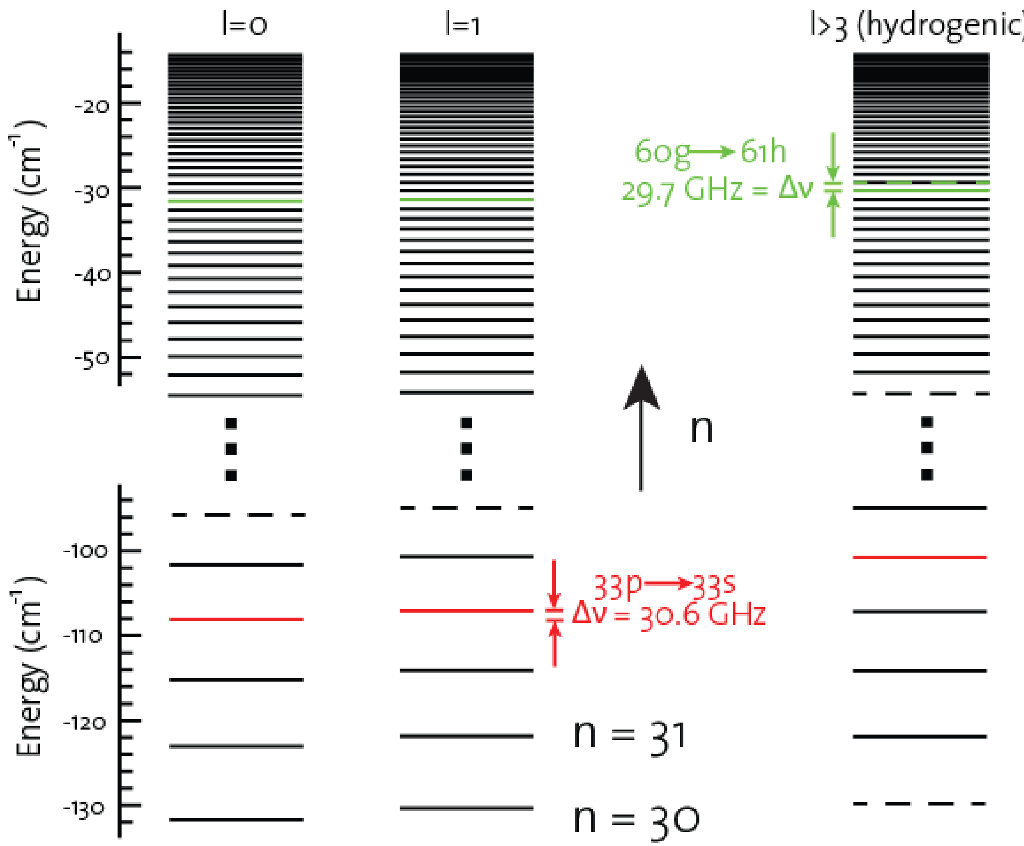

- δ_l is big for s,p
- $$\frac{dW}{dn} = \frac{1}{(n-\delta_l)^3}$$

Electric Dipole Moment

- Electron most of the time far away from core

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  - Strong electric dipole: $\vec{d} = e\vec{r}$

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  - Proportional to transition matrix element $\langle \Psi_f | \vec{d} | \Psi_i \rangle = e\langle \Psi_f | \vec{r} | \Psi_i \rangle = e\langle \Psi_f | r\cos(\theta) | \Psi_i \rangle$

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- We find electric Dipole Moment
  - $\langle \Psi_f | \vec{d} | \Psi_i \rangle \propto \langle r \rangle \langle l \pm 1 | \cos(\theta) | l \rangle \propto n^2$

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Electric Dipole Moment

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- Cross Section: $\sigma \propto \left\langle r \right\rangle^2 \propto n^4$

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Stark Effect $H\Psi = \left(H_0 + \vec{d}\vec{F}\right)\Psi = E\Psi$

- For non-Hydrogenic Atom (e.g. Helium)
  - „Exact“ solution by numeric diagonalization of

$$\left\langle \Psi_f \middle| H \middle| \Psi_i \right\rangle = \left\langle \Psi_f \middle| H_0 \middle| \Psi_i \right\rangle + \left\langle \Psi_f \middle| \vec{d} \middle| \Psi_i \right\rangle \vec{F}$$

in undisturbed (standard) basis ($\tilde{n}$,l,m)

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Numerov

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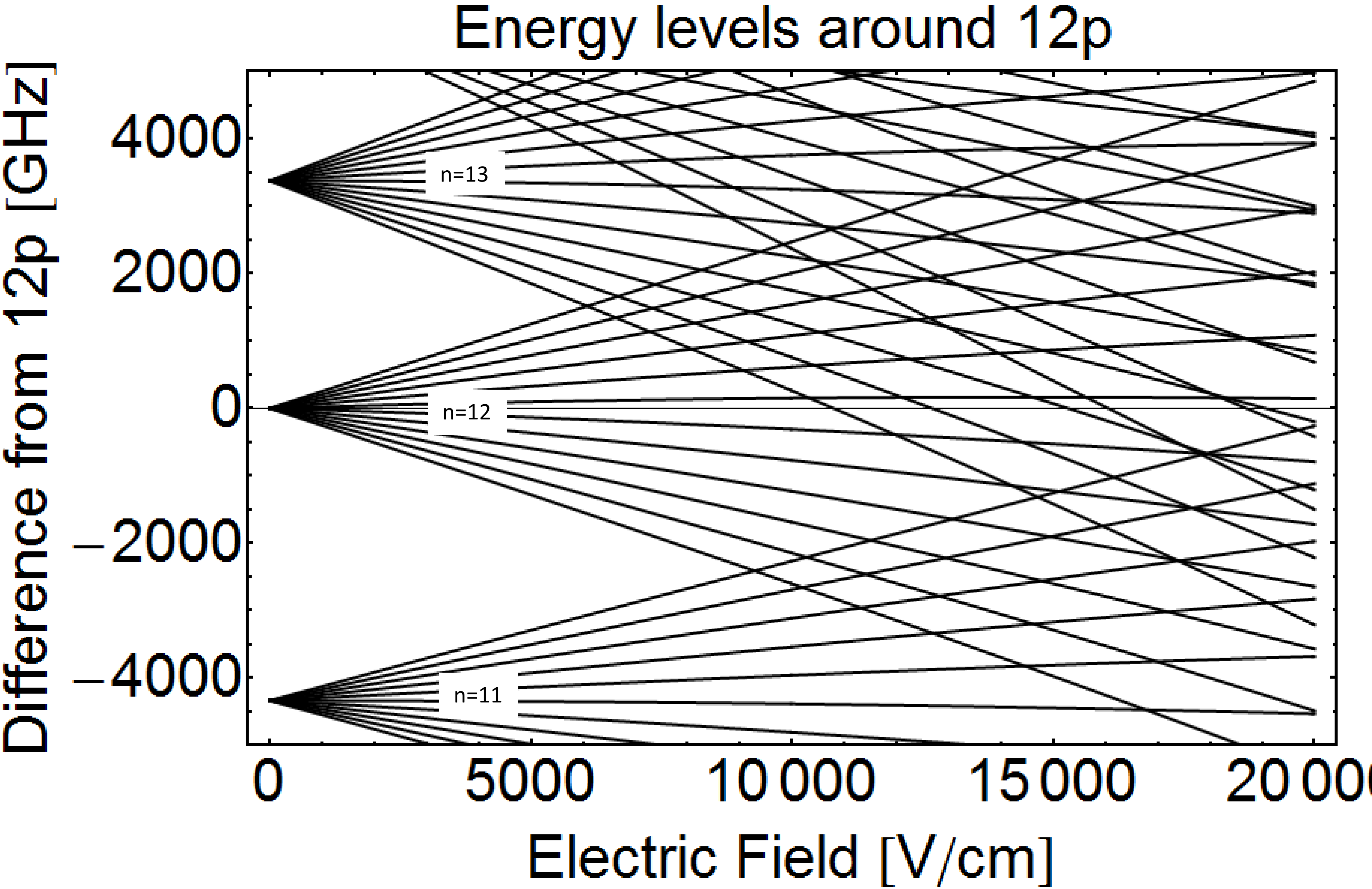
Stark Map Hydrogen



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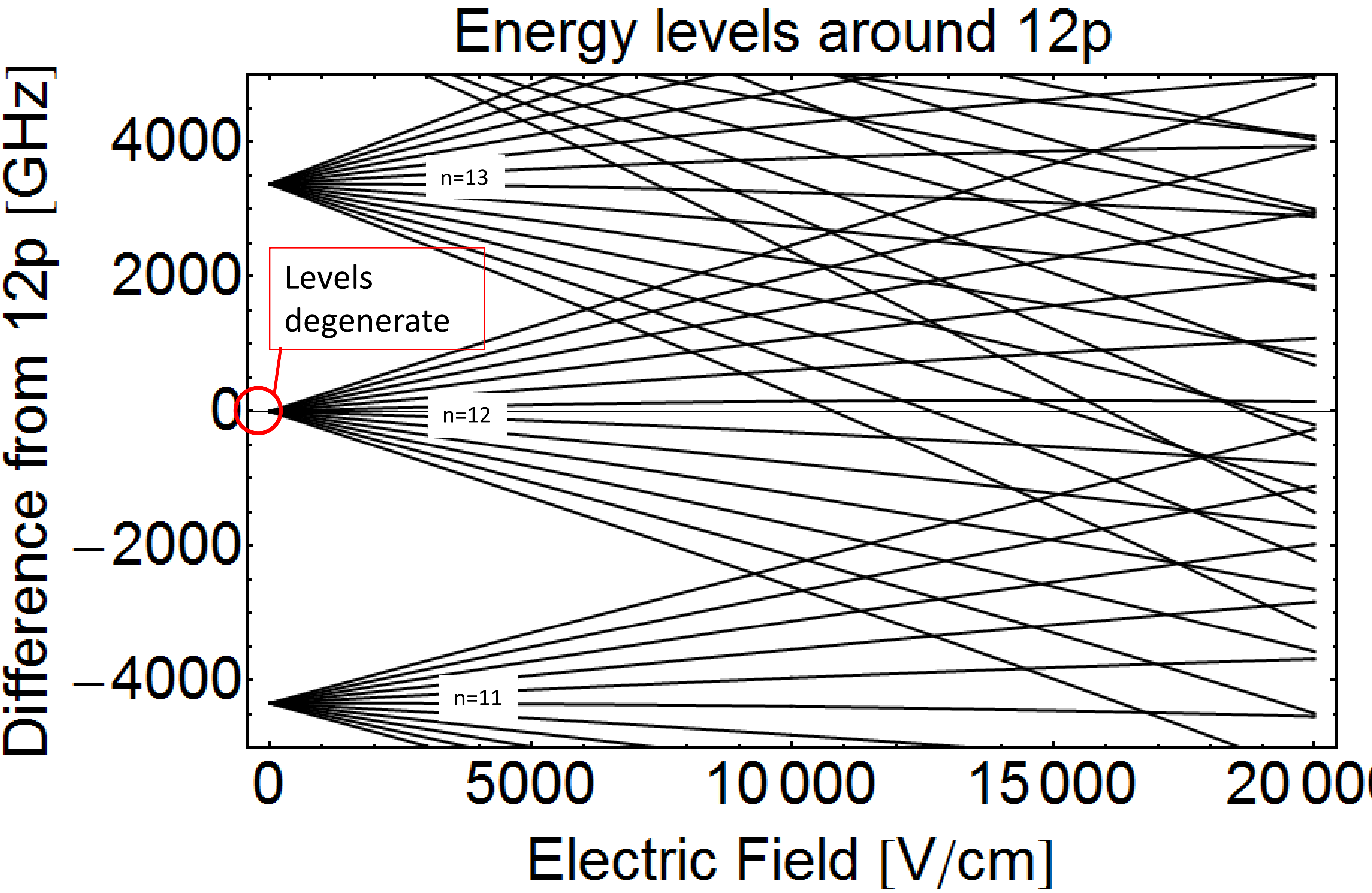

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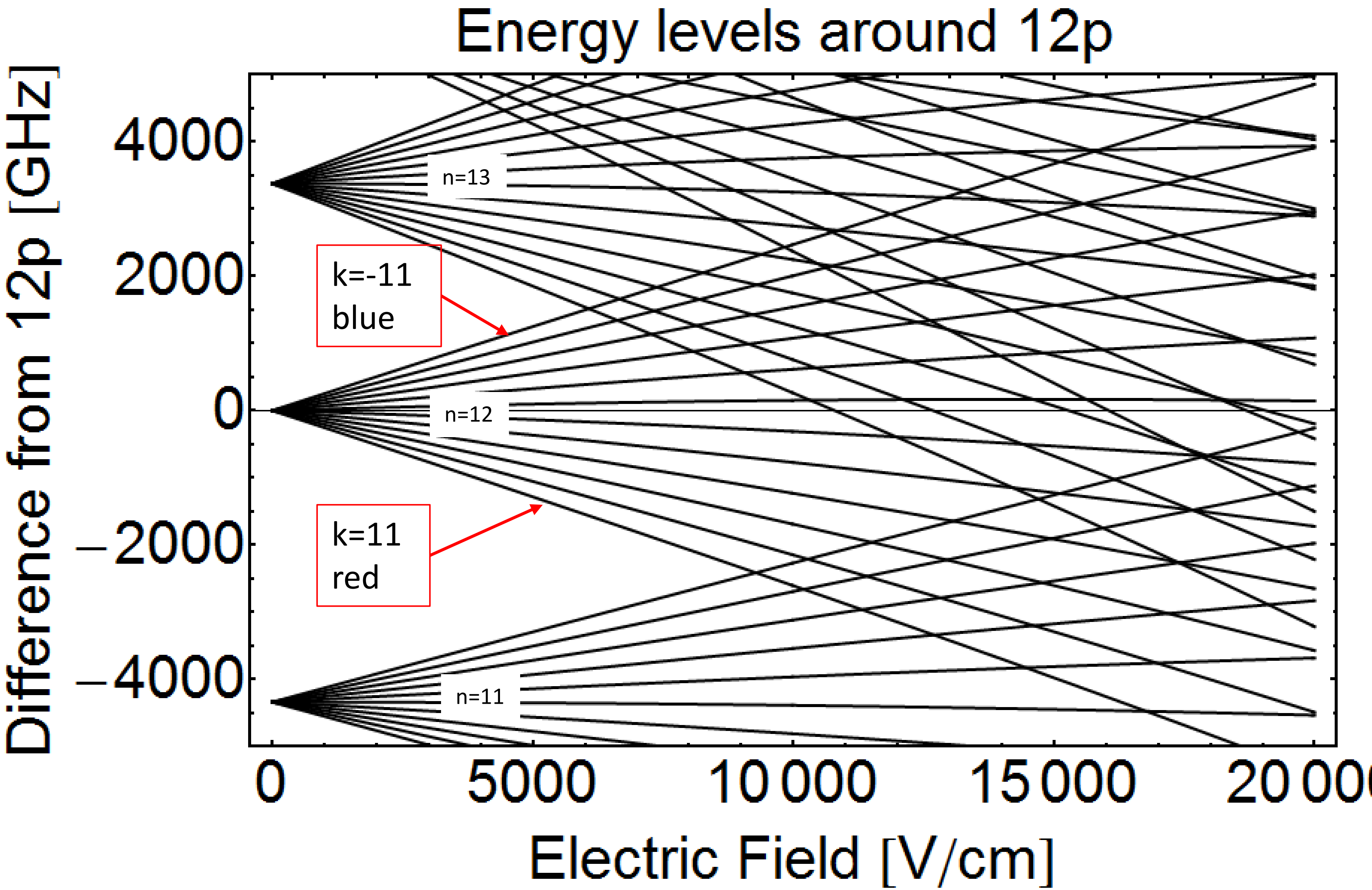

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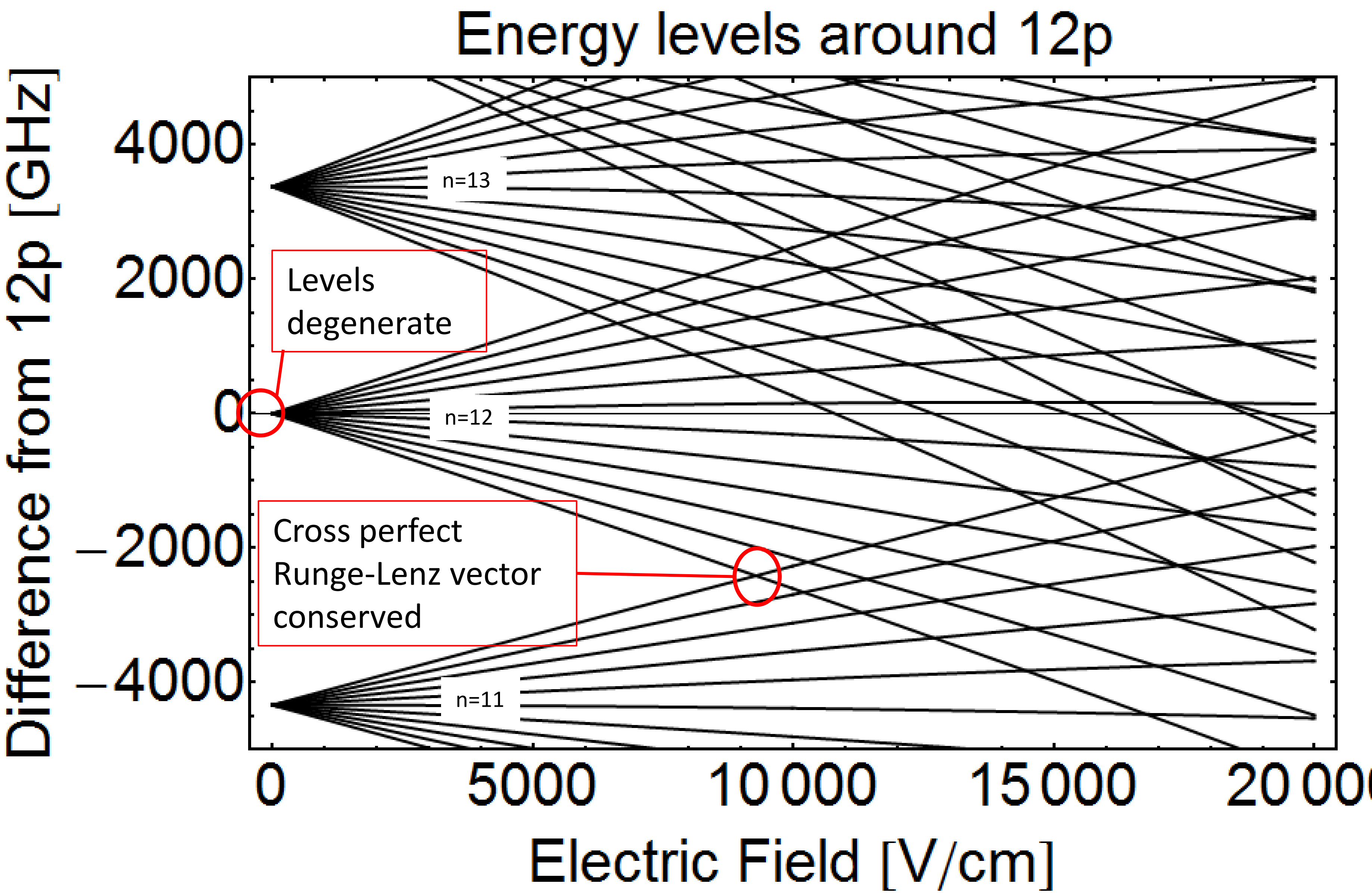

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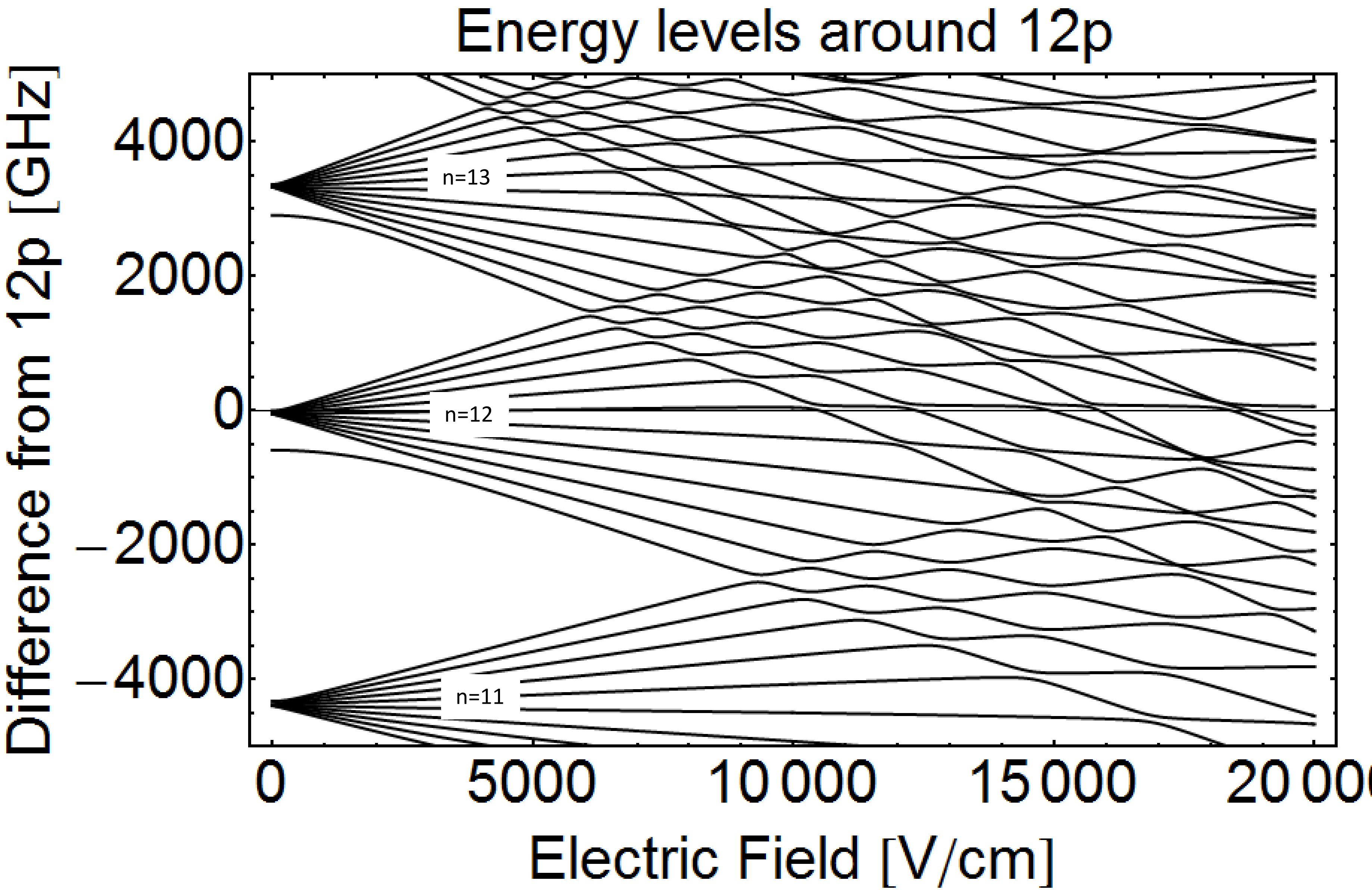

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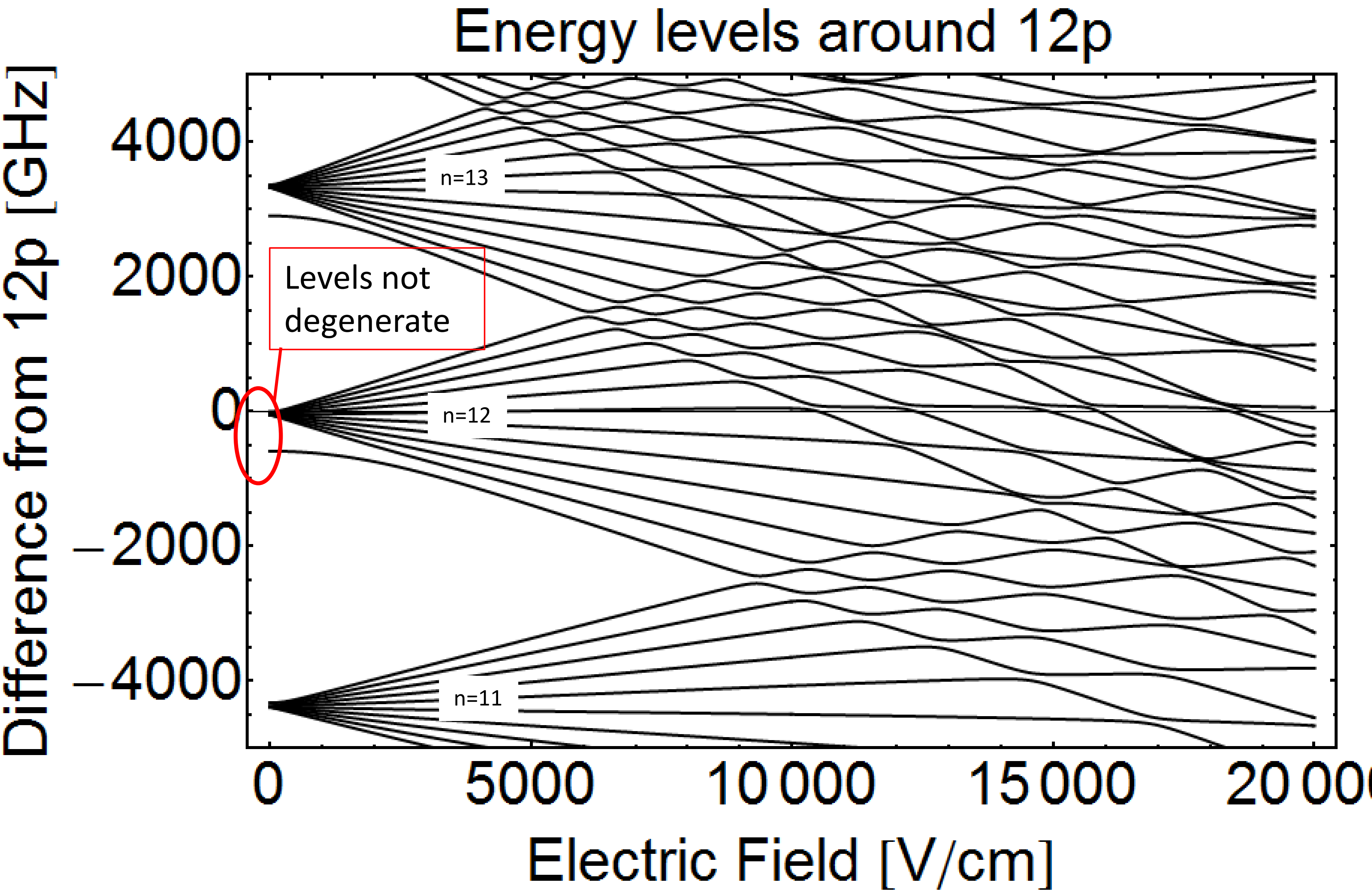

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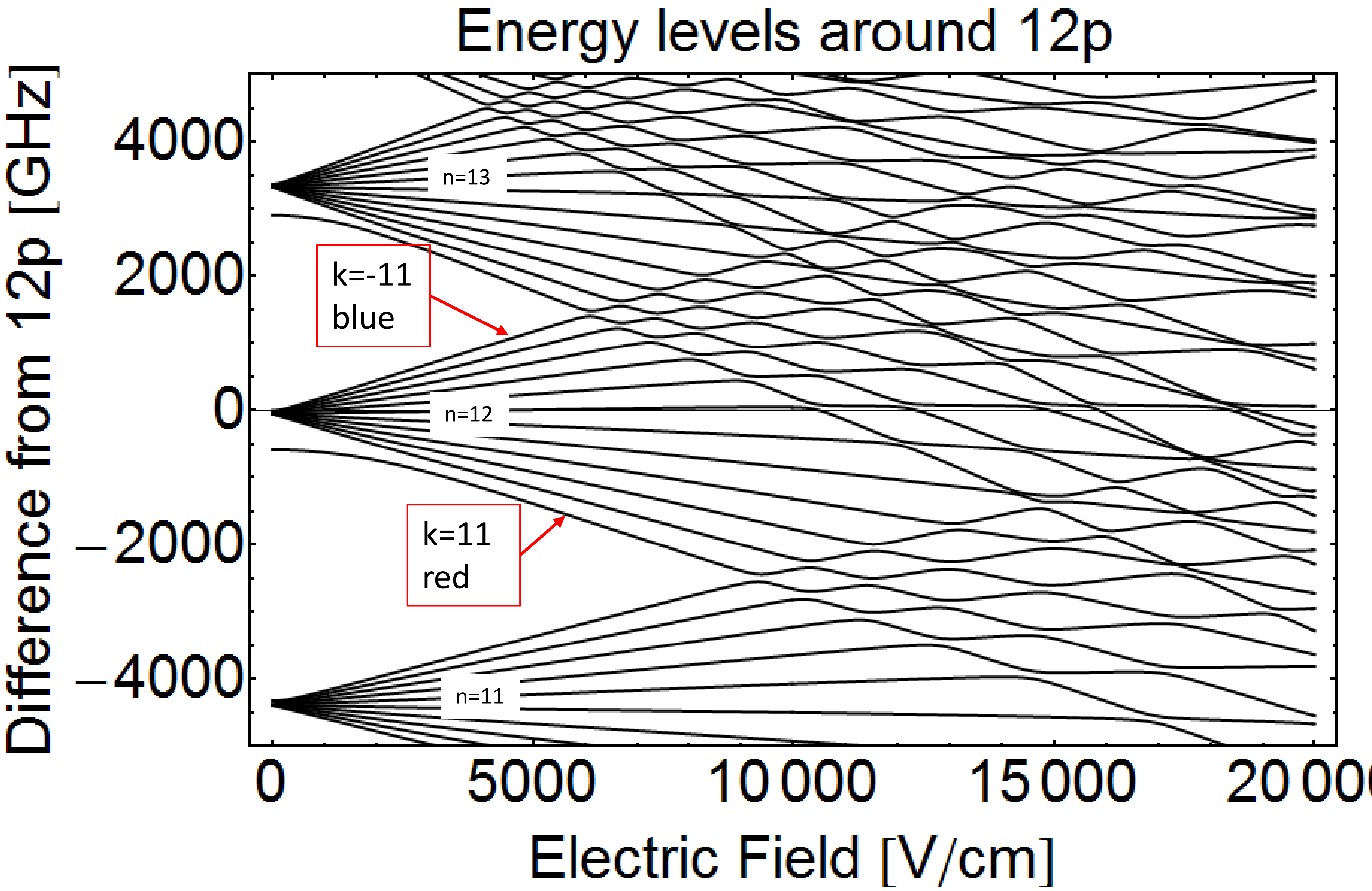

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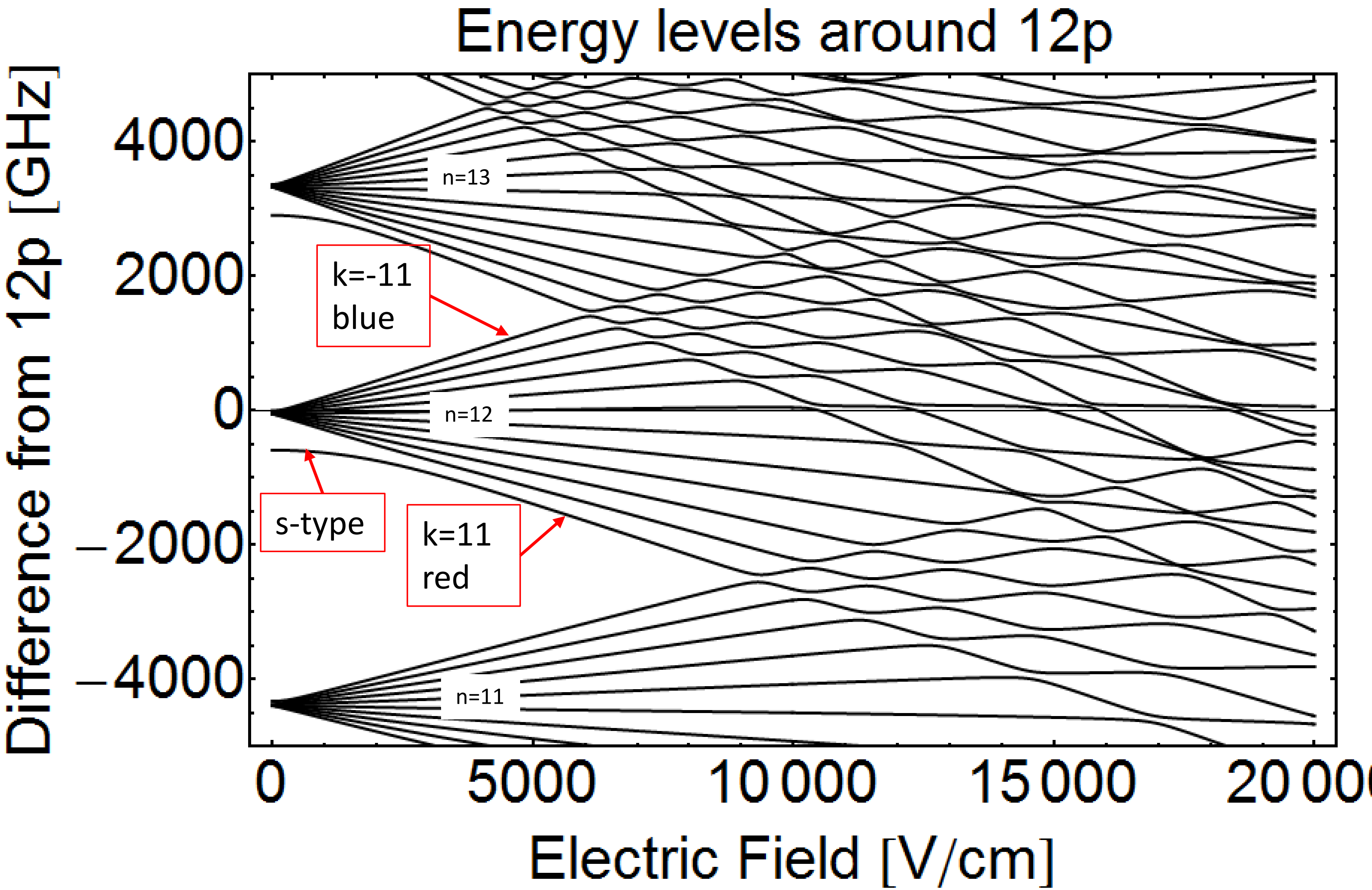

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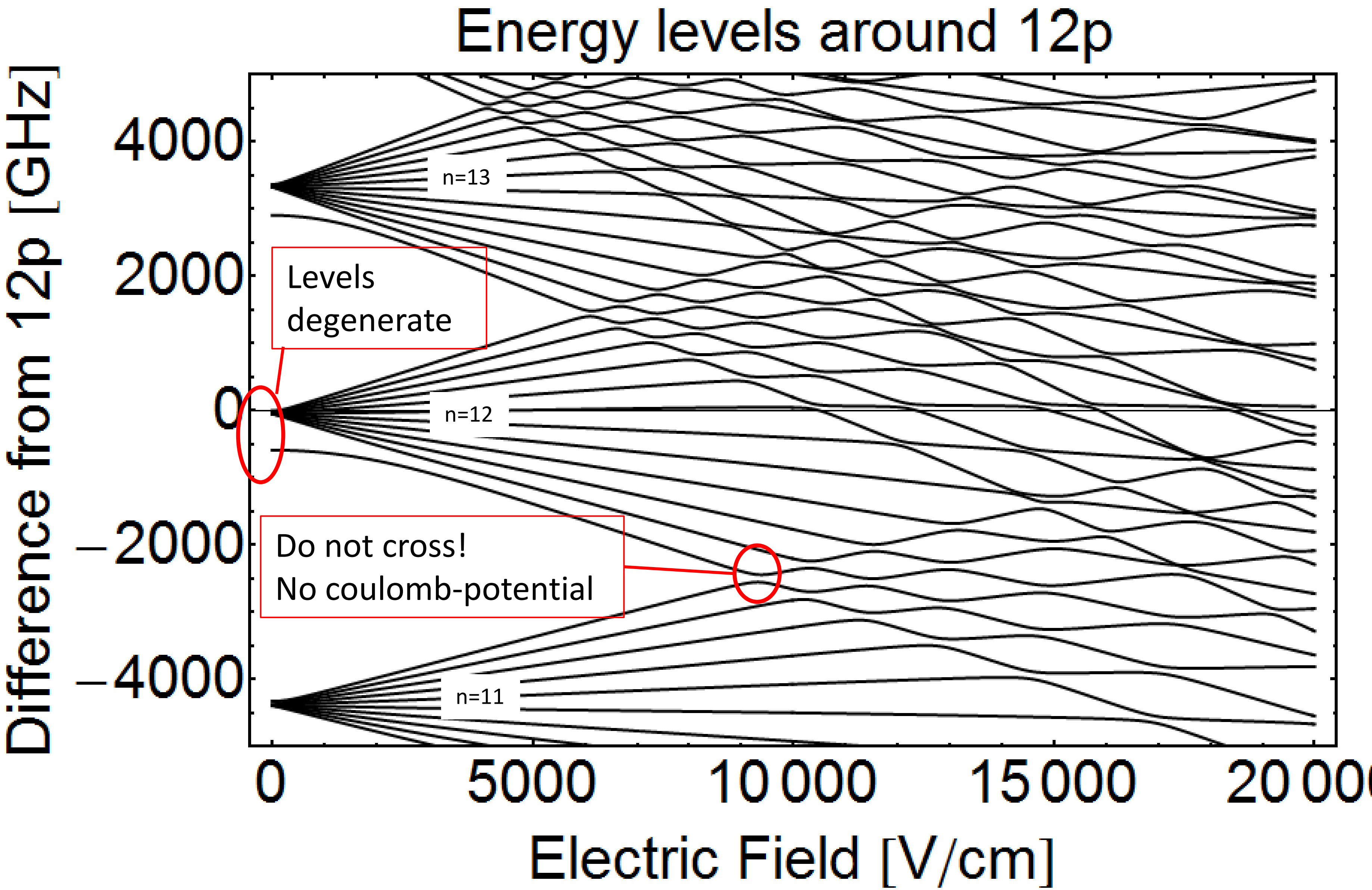
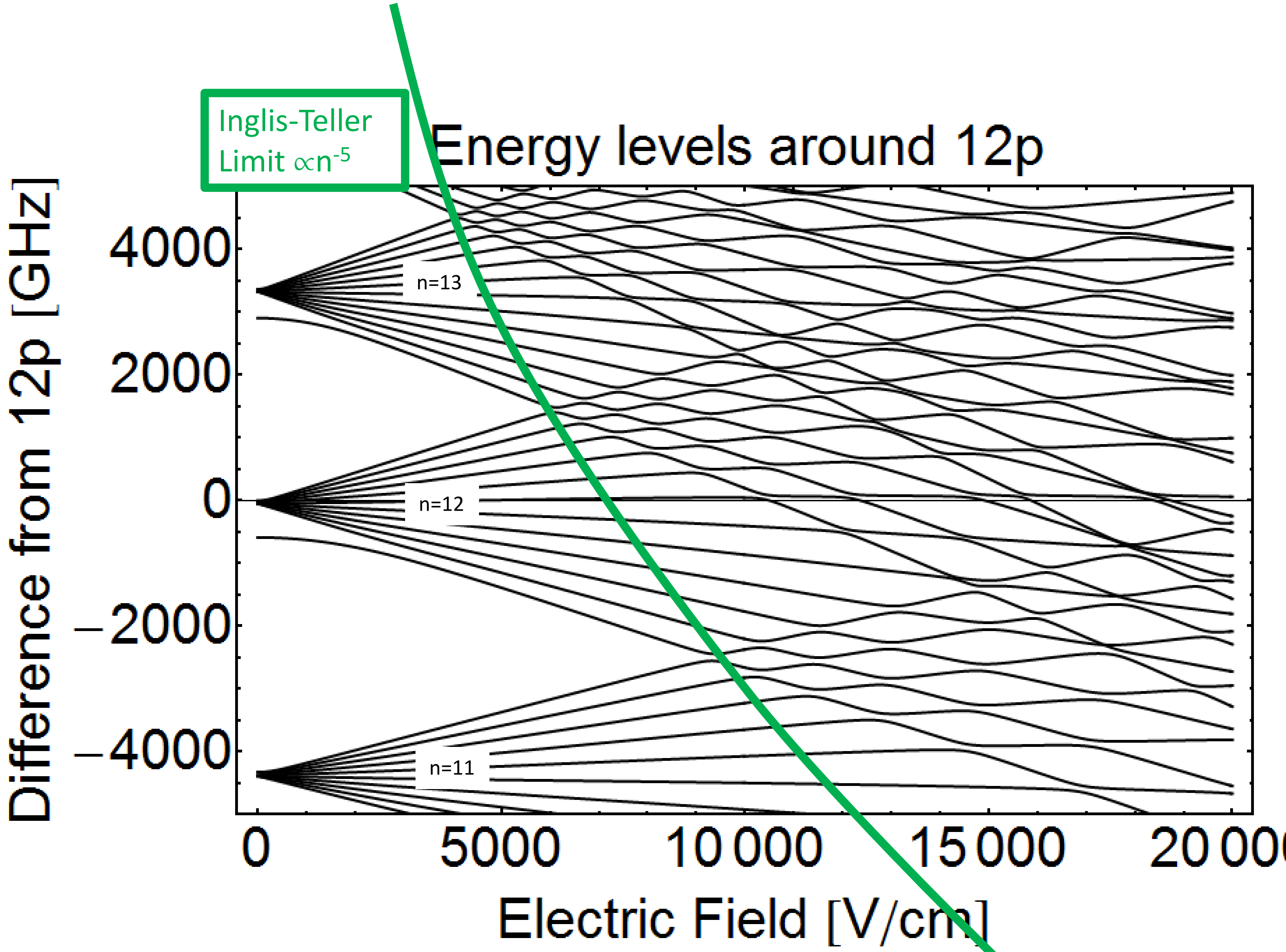




Inglis-Teller
Limit ∝n-5
Energy levels around 12p
Difference from 12p [GHz]
4000
2000
0
−2000
−4000
n=13
n=12
n=11
0
5000
10000
15000
20000
Electric Field [V/cm]

Hydrogen Atom in an electric Field

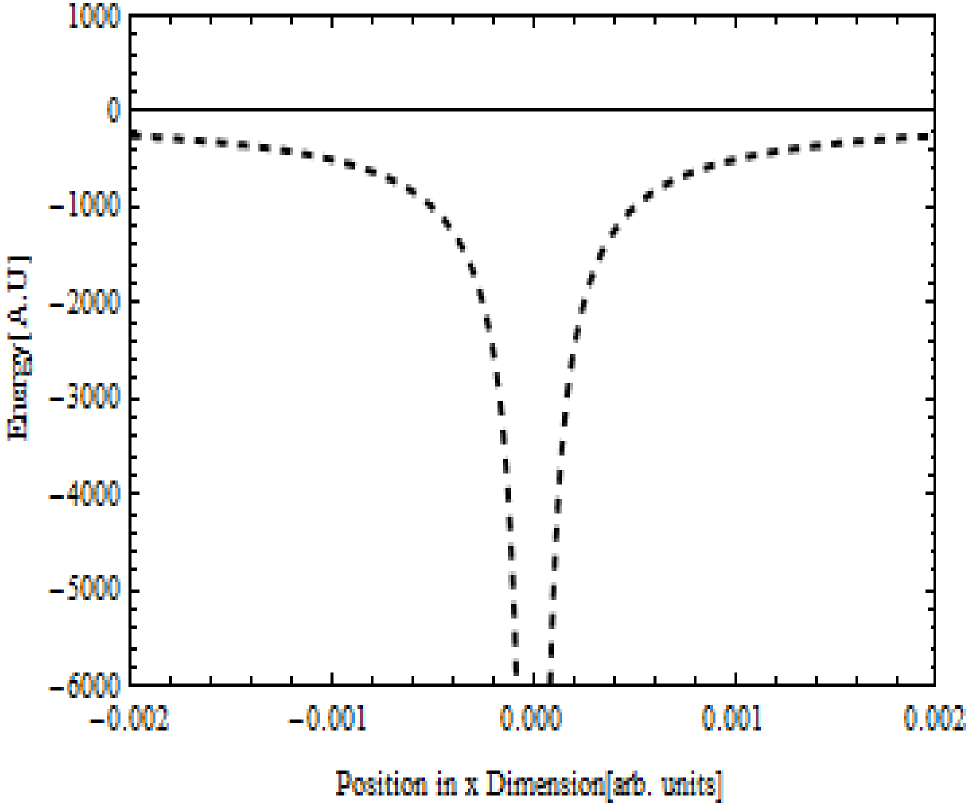
- Rydberg Atoms very sensitive to electric fields
  - Solve: $H\Psi = \left(H_0 + \vec{d}\vec{F}\right)\Psi = E\Psi$ in parabolic coordinates

- Energy-Field dependence: Perturbation-Theory

$$W = -\frac{1}{2n^2} - \frac{3}{2}F\underbrace{(n_1 - n_2)}_{k}n + \frac{F}{16}n^4\left(17n^2 - 3\underbrace{(n_1 - n_2)}_{k}^2 - 9m^2 + 19\right) + O(n^5)$$

Hydrogen Atom in an electric Field

- When do Rydberg atoms ionize?
  - No field applied



$W=-\frac{1}{2(n-\delta_l)^2}$	$\frac{dW}{dn}=\frac{1}{(n-\delta_l)^3}$	$\langle \vec{d} \rangle \approx a_0 n^2$	$\sigma \propto n^4$	$F_{IT} \propto n^{-5}$

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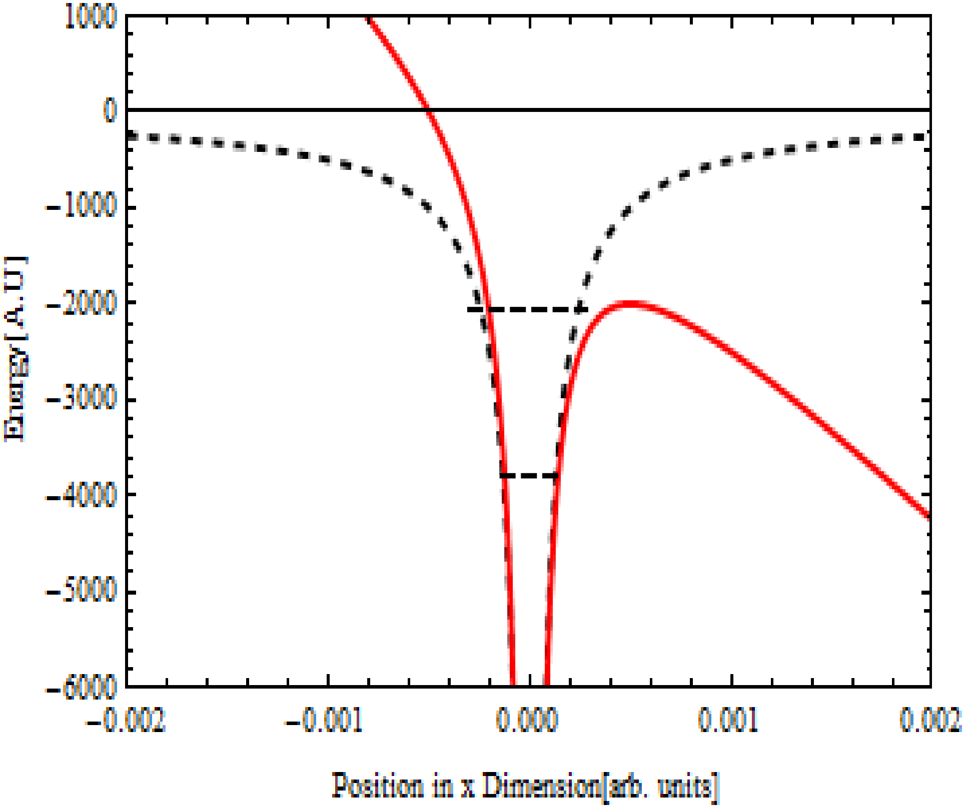

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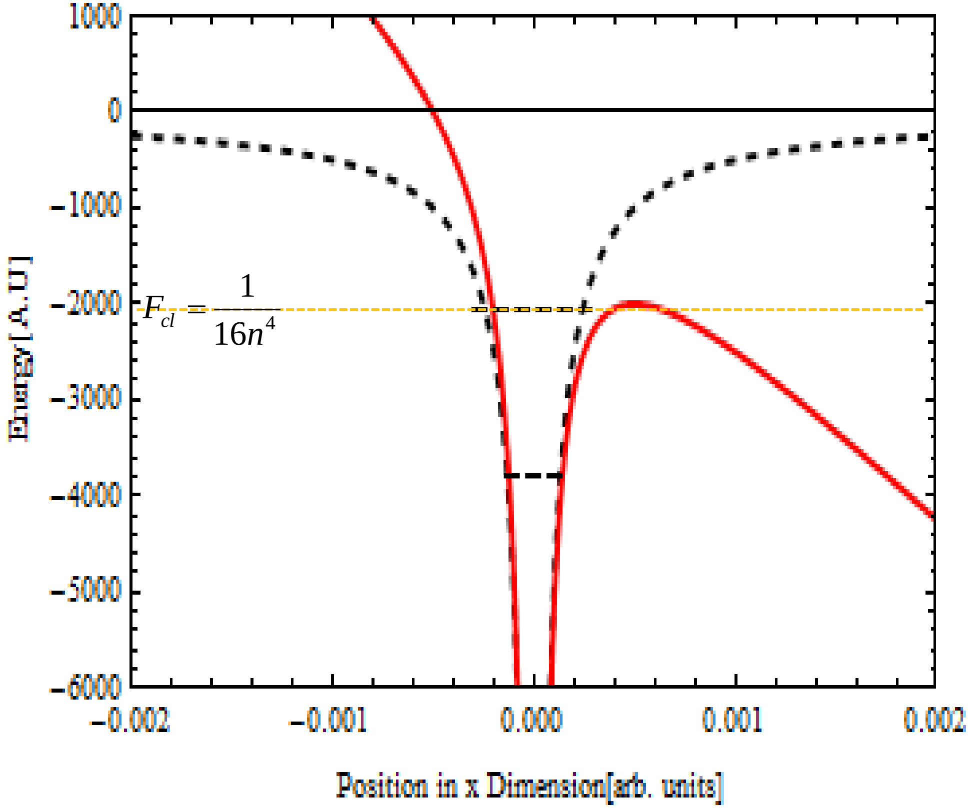
  - Classical ionization:

$$V = -\frac{1}{r} + Fz$$

$$\Rightarrow F_{cl} = \frac{W^2}{4} = \frac{1}{16n^4}$$



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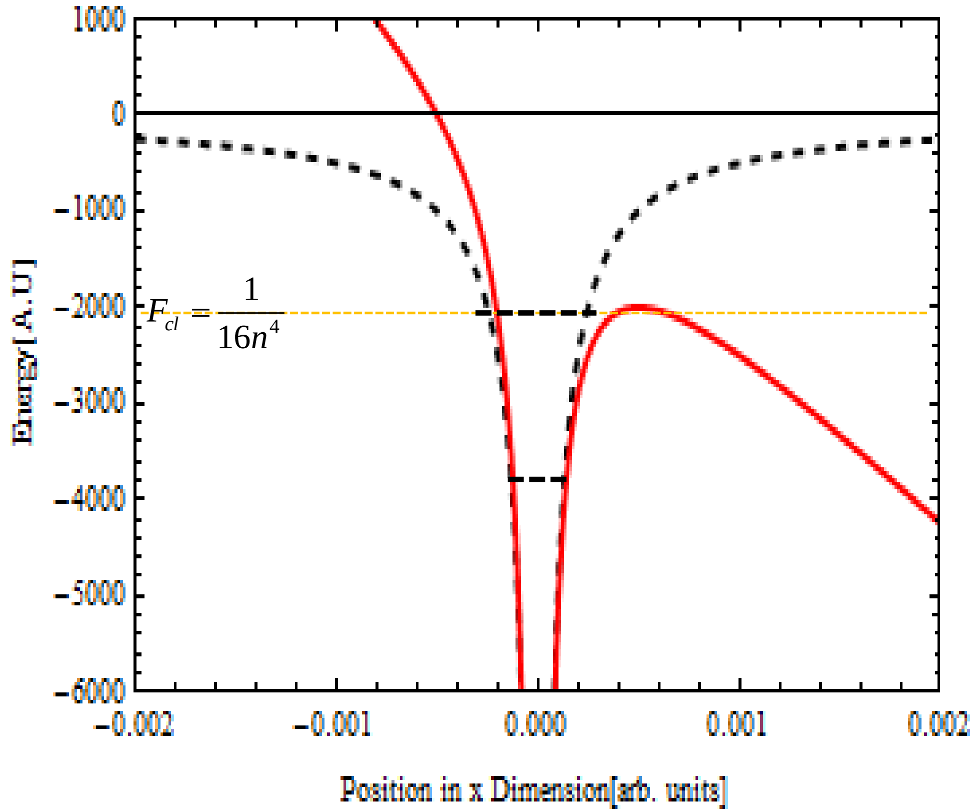
  - Valid only for
    - Non-H atoms if F is Increased slowly



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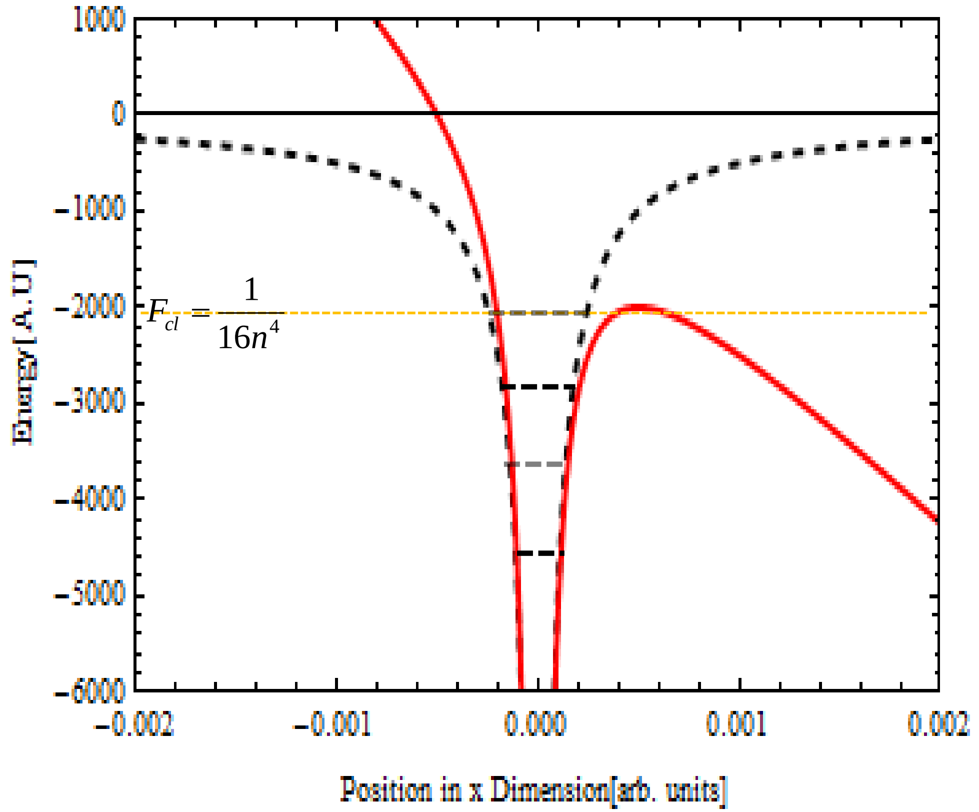
  - Quasi-Classical ioniz.:



$$V(\eta) = 2\left(-\frac{Z_2}{\eta} + \frac{m^2-1}{4\eta^2} - \frac{F\eta}{4}\right)$$

$$\overset{m=0}{\Rightarrow} F = \frac{W^2}{4Z_2}\bigg|$$

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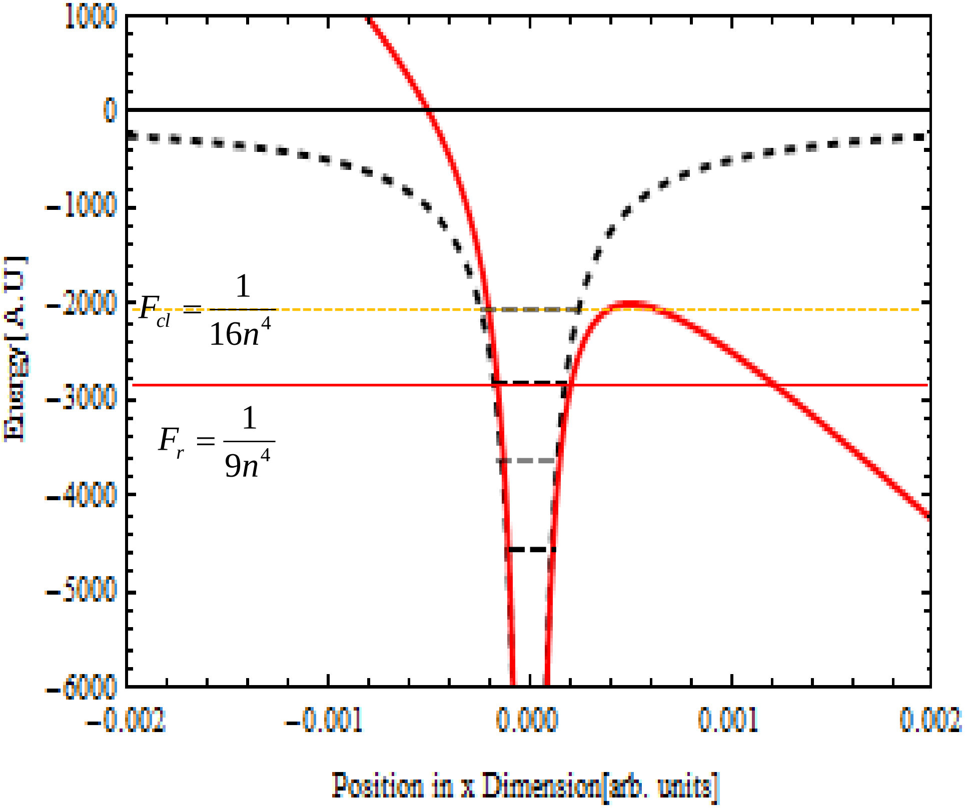

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$$\overset{m=0}{\Rightarrow} F = \frac{W^2}{4Z_2}\Bigg| = \frac{1}{9n^4}$$ red

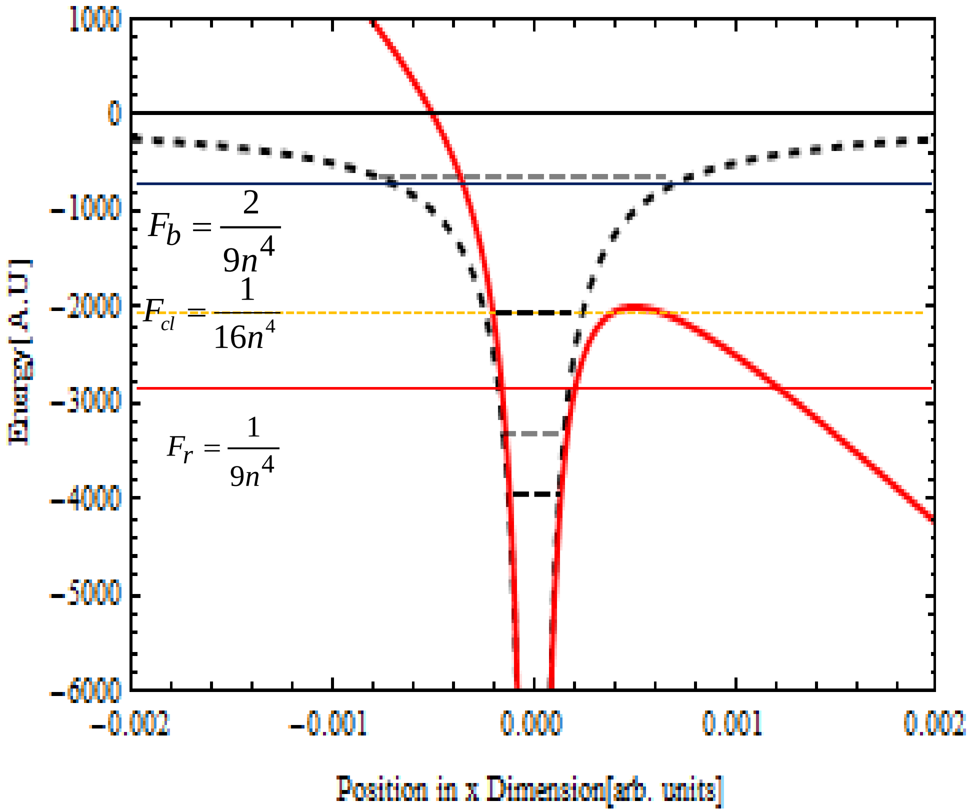
$W = -\frac{1}{2(n-\delta_l)^2}$	$\frac{dW}{dn} = \frac{1}{(n-\delta_l)^3}$	$\langle \vec{d} \rangle \approx a_0 n^2$	$\sigma \propto n^4$	$F_{IT} \propto n^{-5}$	$F_{cl} \propto \frac{1}{16} n^{-4}$

Hydrogen Atom in an electric Field

- When do Rydberg atoms ionize?
  - No field applied
  - Electric Field applied
  - Quasi-Classical ioniz.:

$$V(\eta) = 2\left(-\frac{Z_2}{\eta} + \frac{m^2 - 1}{4\eta^2} - \frac{F\eta}{4}\right)$$

$$\Rightarrow F = \frac{W^2}{4Z_2}\Bigg|\begin{array}{ll} = \frac{1}{9n^4} & \text{red} \\ \approx \frac{2}{9n^4} & \text{blue} \end{array}$$



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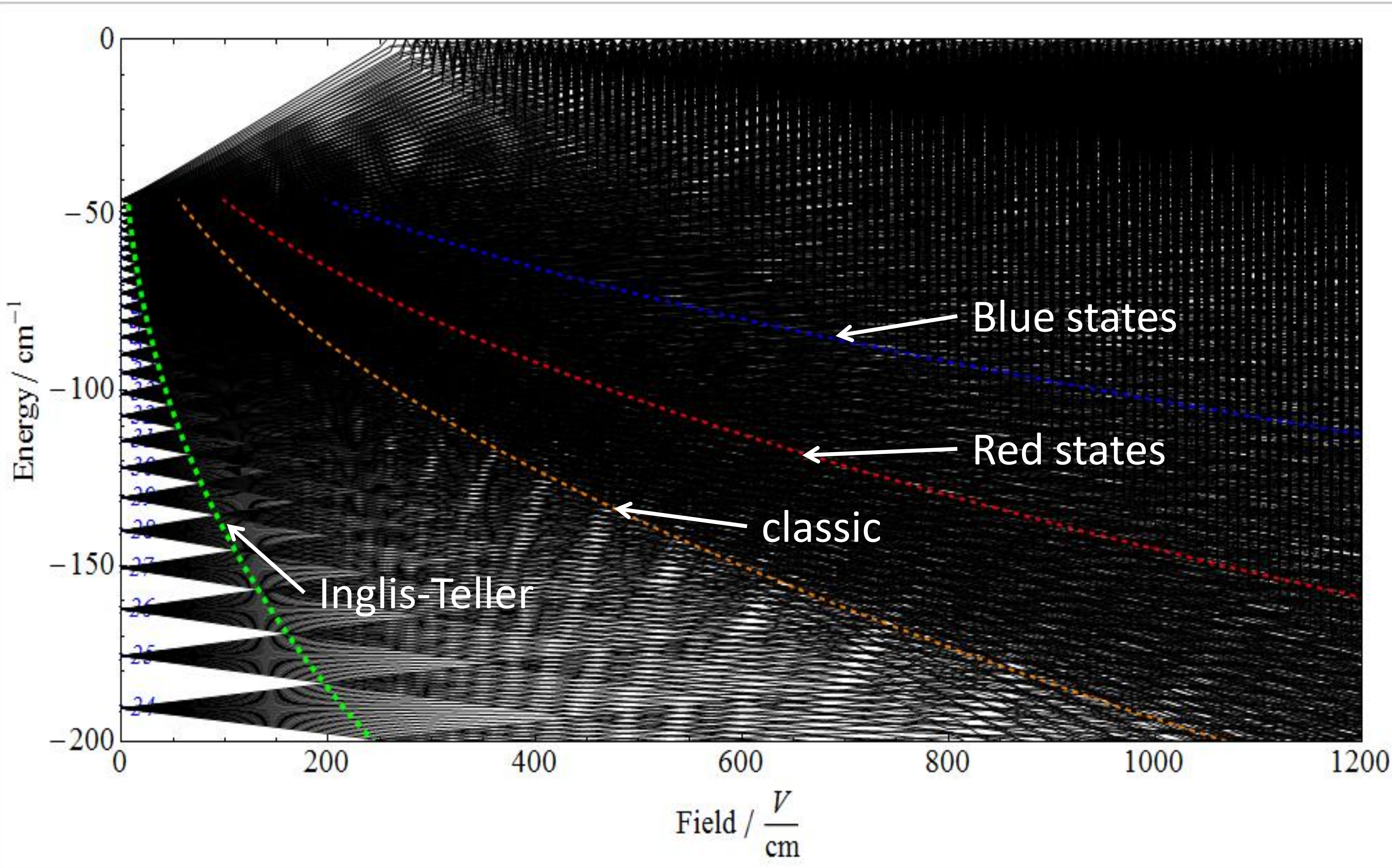
Hydrogen Atom in an electric Field



Lifetime

- From Fermis golden rule
  - Einstein A coefficient for two states $n,l \rightarrow n',l'$

$$A_{n',l',n,l} = \frac{4e^2\omega^3_{n',l',n,l}}{3\hbar c^3}\frac{\max(l,l')}{2l+1}\left|\langle n'l'|r|nl\rangle\right|^2$$

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For l≈0: $\propto n^{-3/2}$
Overlap of WF

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$W = -\frac{1}{2(n-\delta_l)^2}$	$\frac{dW}{dn} = \frac{1}{(n-\delta_l)^3}$	$\langle \vec{d} \rangle \approx a_0 n^2$	$\sigma \propto n^4$	$F_{IT} \propto n^{-5}$	$F_{cl} \propto 1/16\, n^{-4}$	$\tau_{n,0} \propto n^3$

Lifetime

- From Fermis golden rule
  - Einstein A coefficient for two states $n,l \rightarrow n',l'$

$$A_{n',l',n,l} = \frac{4e^2\omega^3_{n',l',n,l}}{3\hbar c^3}\frac{\max(l,l')}{2l+1}\left|\langle n'l'|r|nl\rangle\right|^2$$

  - Lifetime $\tau_{n,l} = \left(\sum_{n',l'<n,l} A_{n',l',n,l}\right)^{-1}$

For l≈n: $\propto n^2$
Overlap of WF

For l≈n: $\propto n^{-3}$
Overlap of WF

$W = -\frac{1}{2(n-\delta_l)^2}$	$\frac{dW}{dn} = \frac{1}{(n-\delta_l)^3}$	$\langle \vec{d} \rangle \approx a_0 n^2$	$\sigma \propto n^4$	$F_{IT} \propto n^{-5}$	$F_{cl} \propto \frac{1}{16} n^{-4}$	$\tau_{n,l} \propto n^3, n^5$

Lifetime

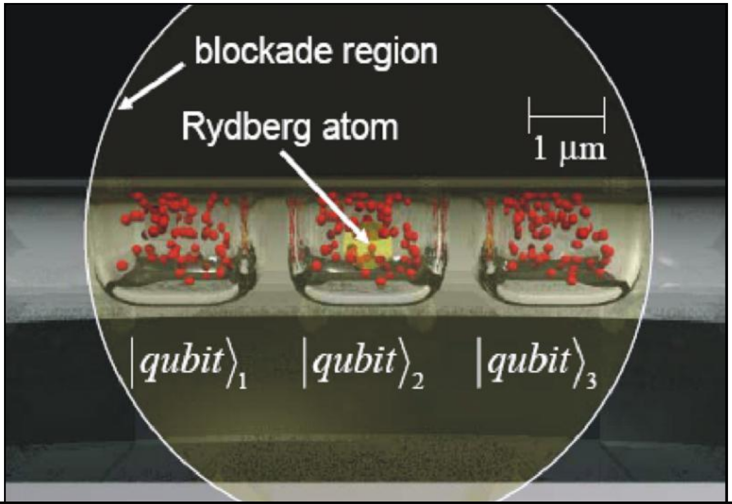
$$A_{n',l',n,l} = \frac{4e^2\omega^3_{n',l',n,l}}{3\hbar c^3}\frac{\max(l,l')}{2l+1}\left|\langle n'l'|r|nl\rangle\right|^2 \qquad \tau_{n,l} = \left(\sum_{n',l'<n,l} A_{n',l',n,l}\right)^{-1}$$

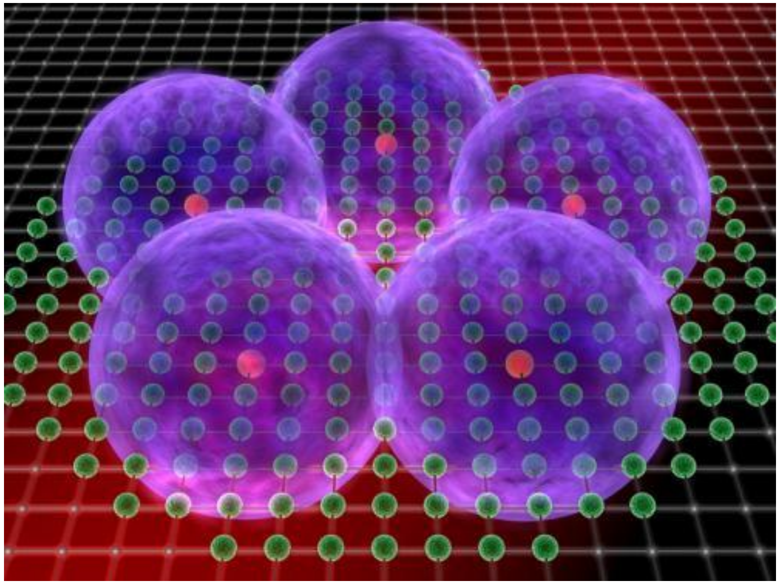
State	Stark State 60 p (n',l') small	Circular state 60 l=59 m=59 $(n',l') \approx (n\pm1, l\pm1)$	Statistical mixture
Scaling	n^3 (overlap of $\psi \propto n^{-3/2}$)	n^5 $\langle r\rangle \propto n^2$	$n^{4.5}$
Lifetime	7.2 μs	70 ms	$\approx$ms

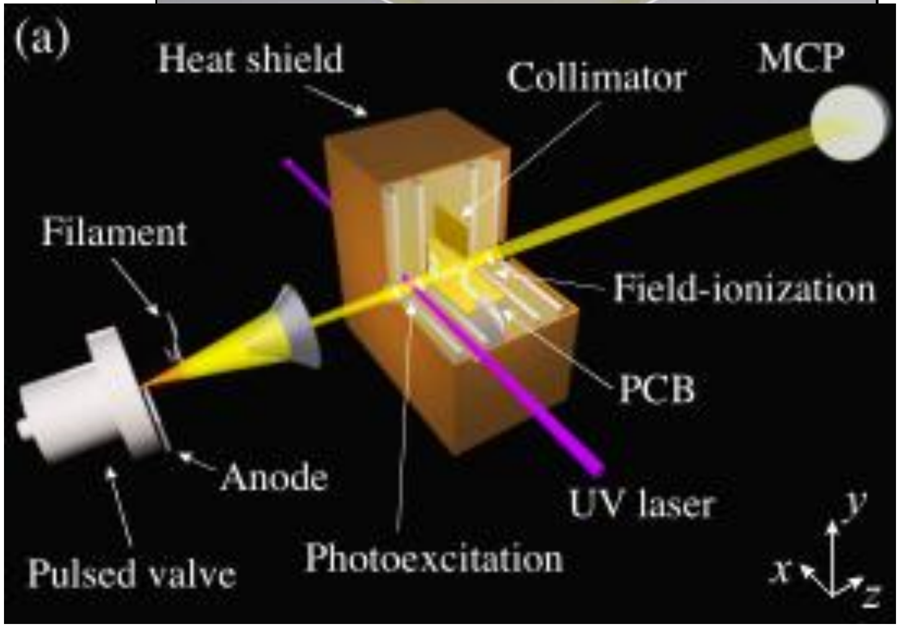
$W = -\frac{1}{2(n-\delta_l)^2}$	$\frac{dW}{dn} = \frac{1}{(n-\delta_l)^3}$	$\langle \vec{d}\rangle \approx a_0 n^2$	$\sigma \propto n^4$	$F_{IT} \propto n^{-5}$	$F_{cl} \propto \frac{1}{16} n^{-4}$	$\tau_{n,l} \propto n^{-3}, n^{-5}$

Part 2- Generation of Rydberg atoms

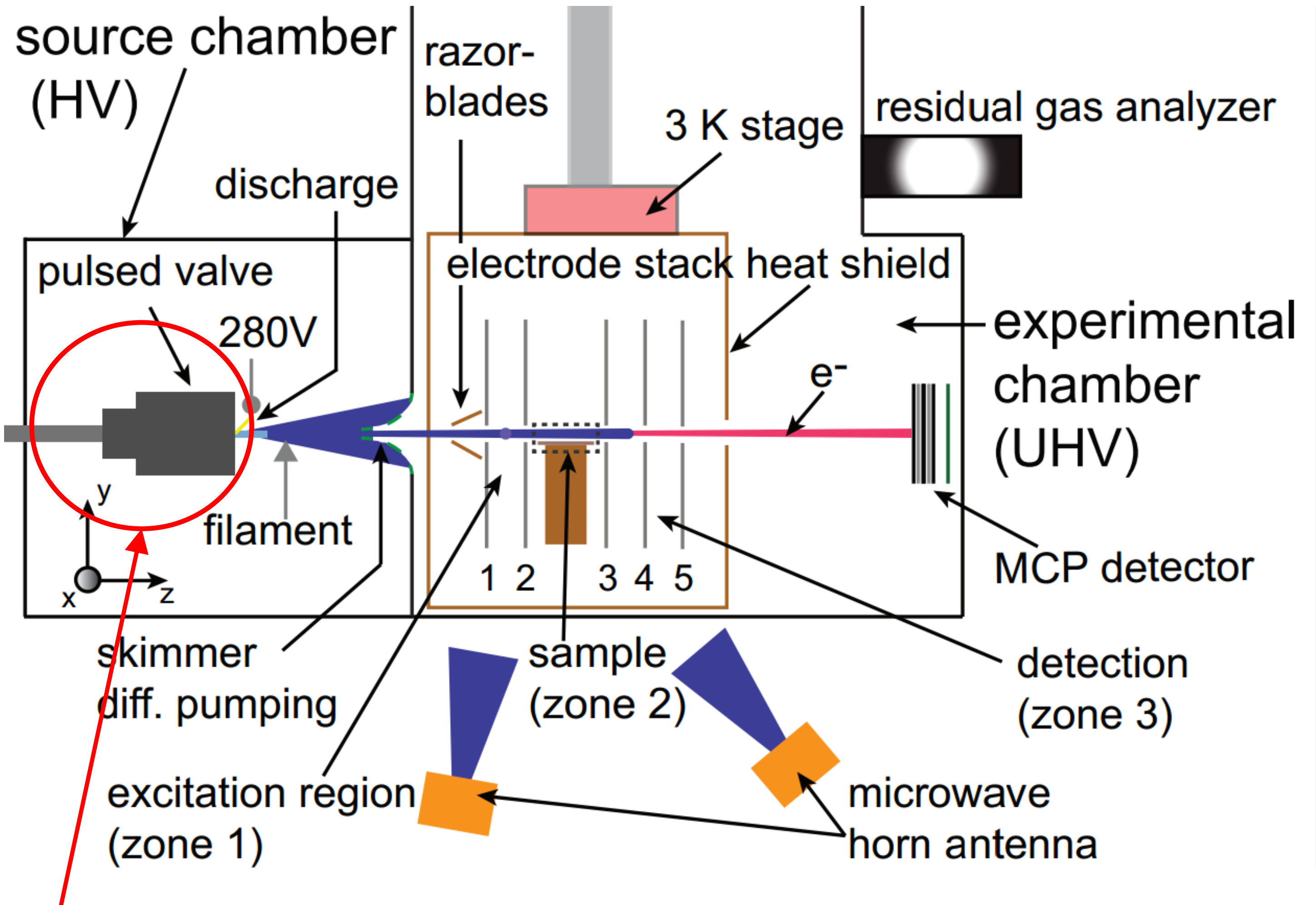
- Typical Experiments:
  - Beam experiments
  - (ultra) cold atoms
  - Vapor cells





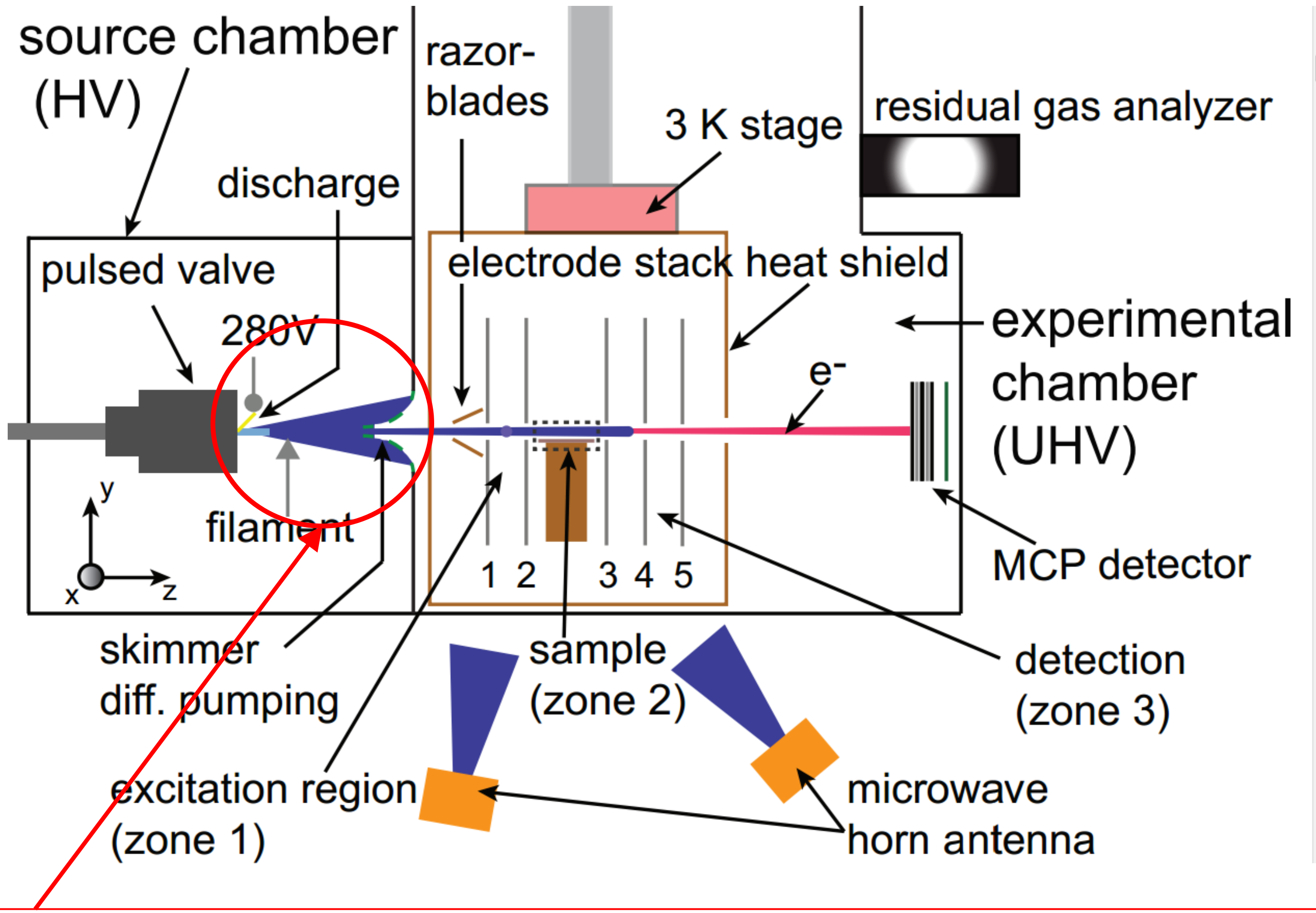


ETH physics Rydberg experiment



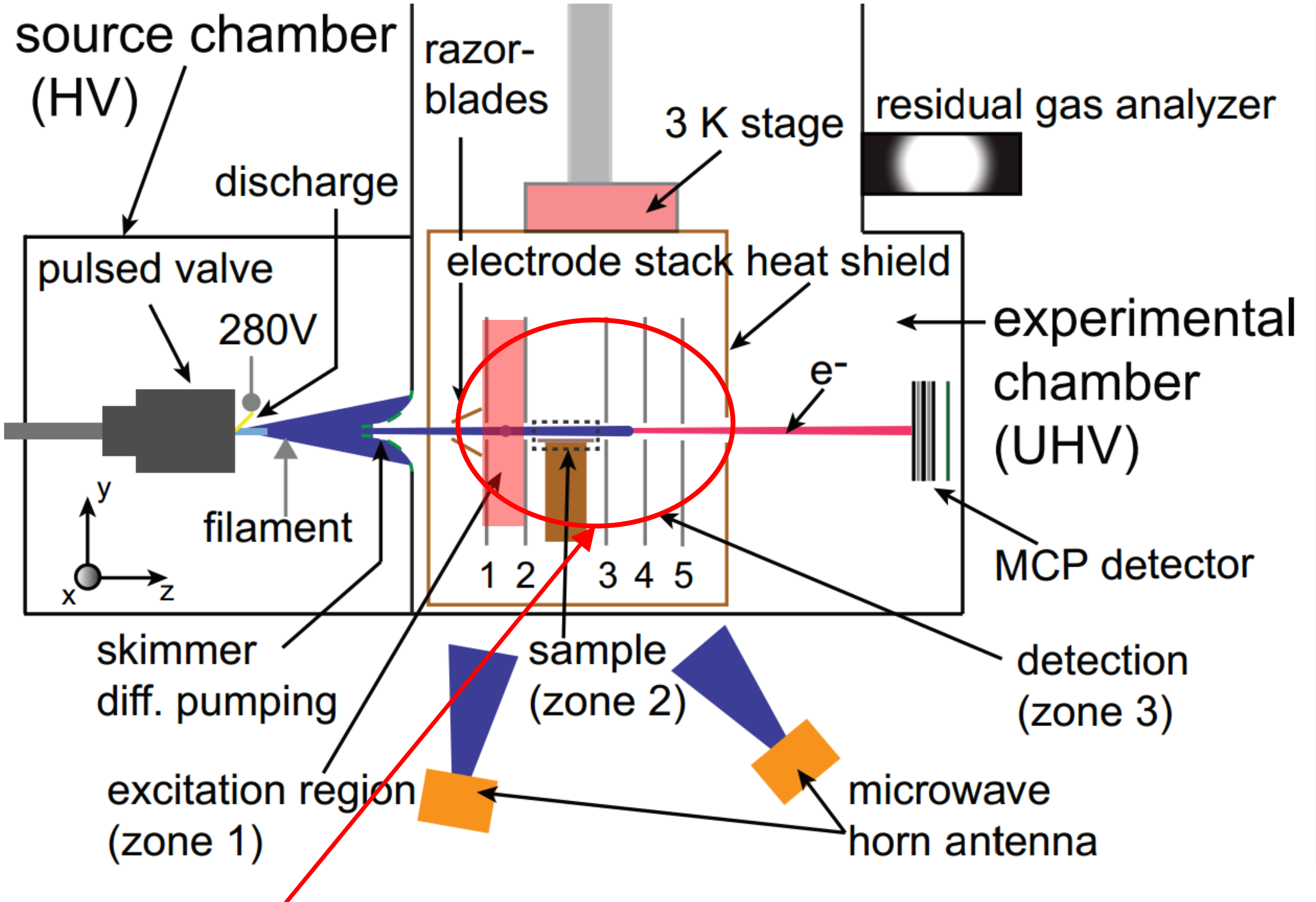
Creation of a cold supersonic beam of Helium.
Speed: 1700m/s, pulsed: 25Hz, temperature atoms=100mK

ETH physics Rydberg experiment



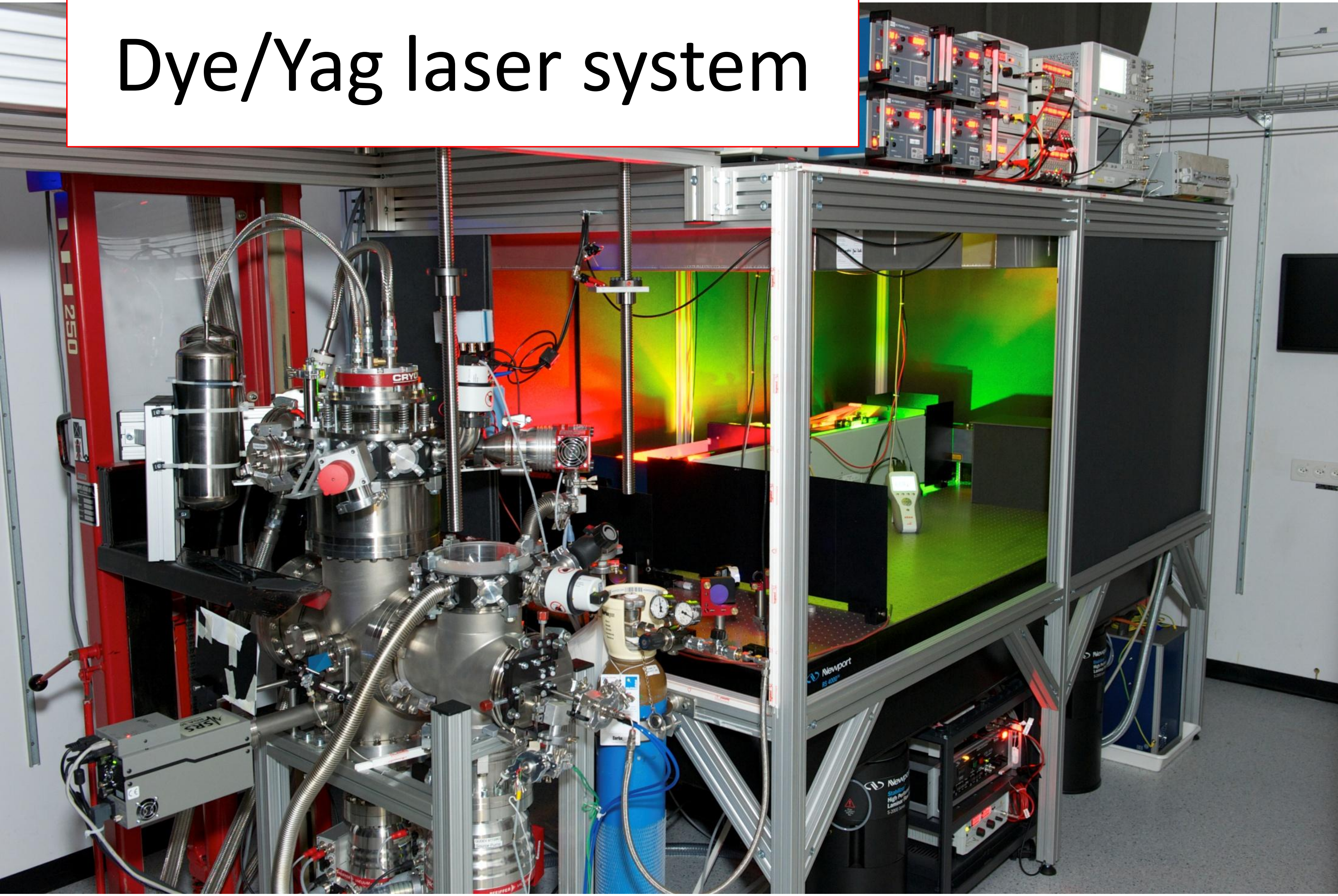
Excite electrons to the 2s-state, (to overcome very strong binding energy in the xuv range) by means of a discharge – like a lightning.

ETH physics Rydberg experiment

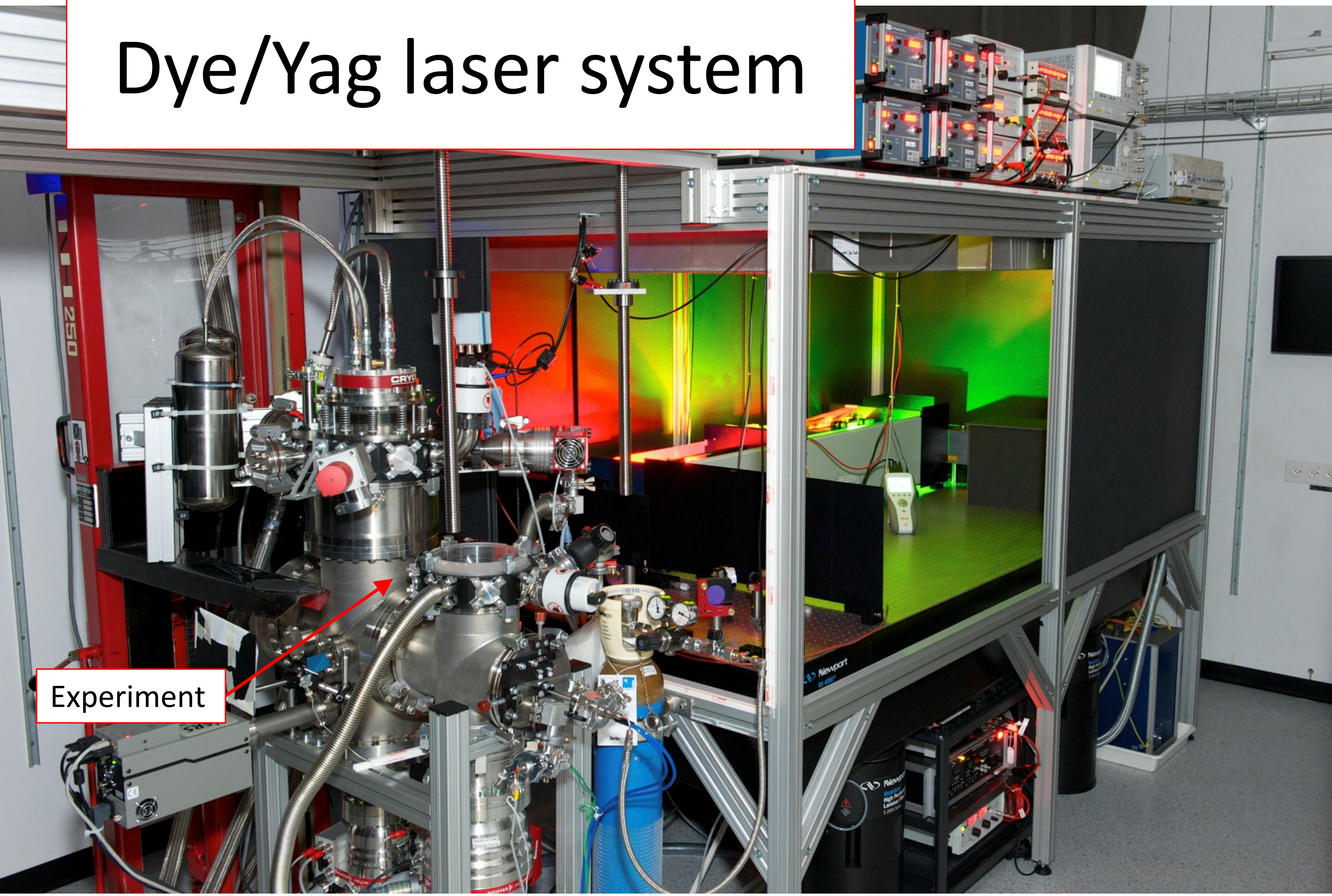


Actual experiment consists of 5 electrodes. Between the first 2 the atoms get excited to Rydberg states up to n=inf with a dye laser.

Dye/Yag laser system



Dye/Yag laser system



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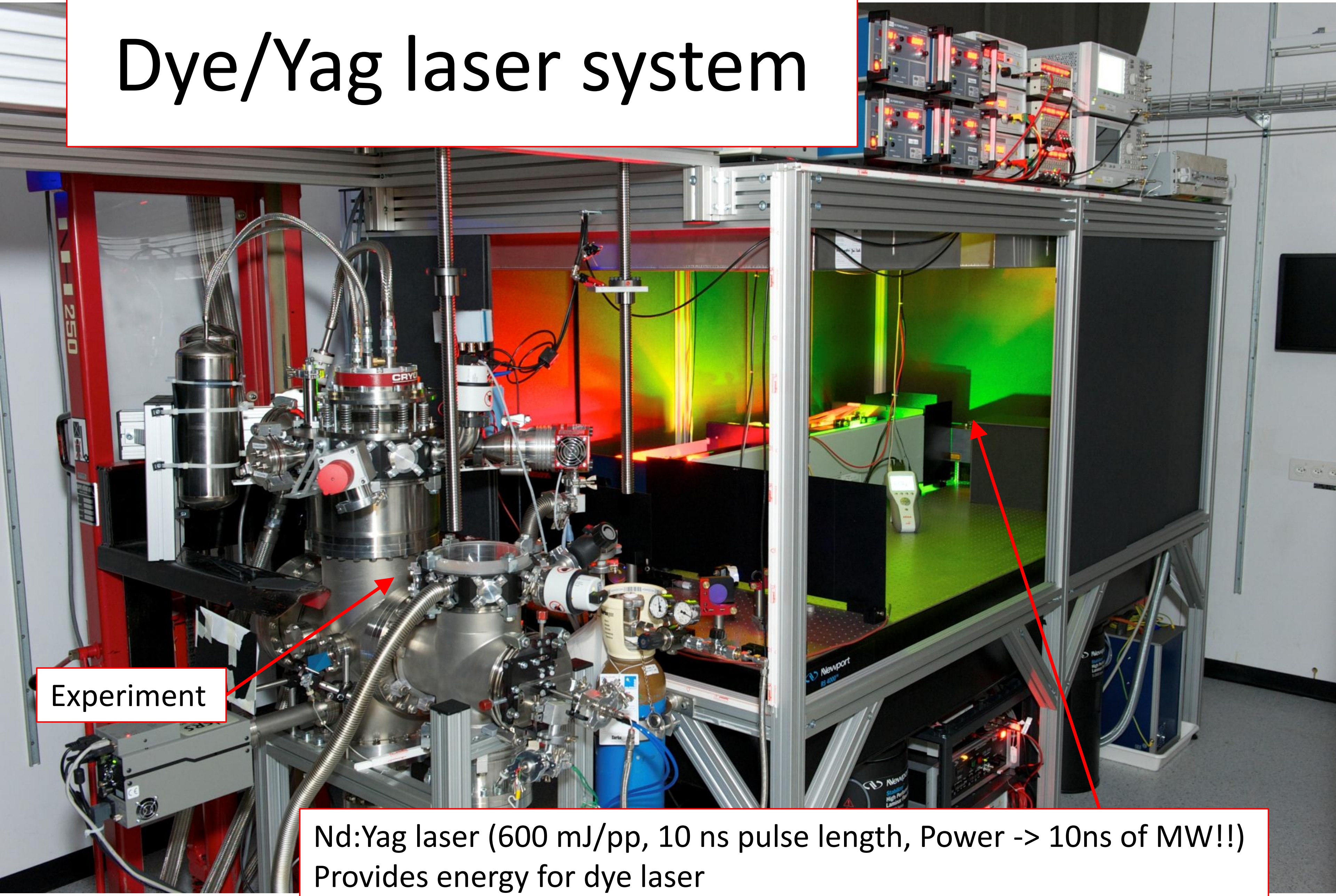

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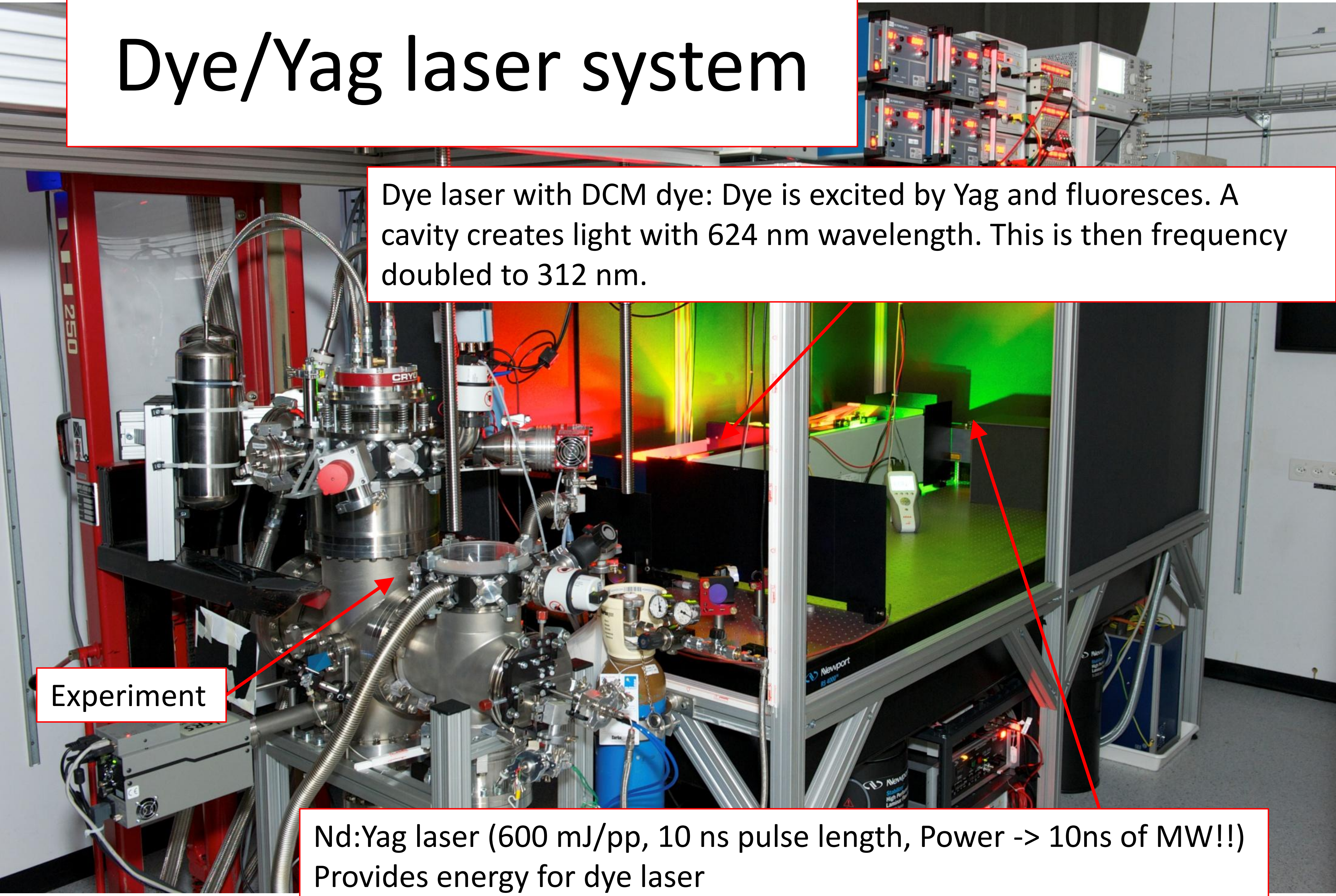
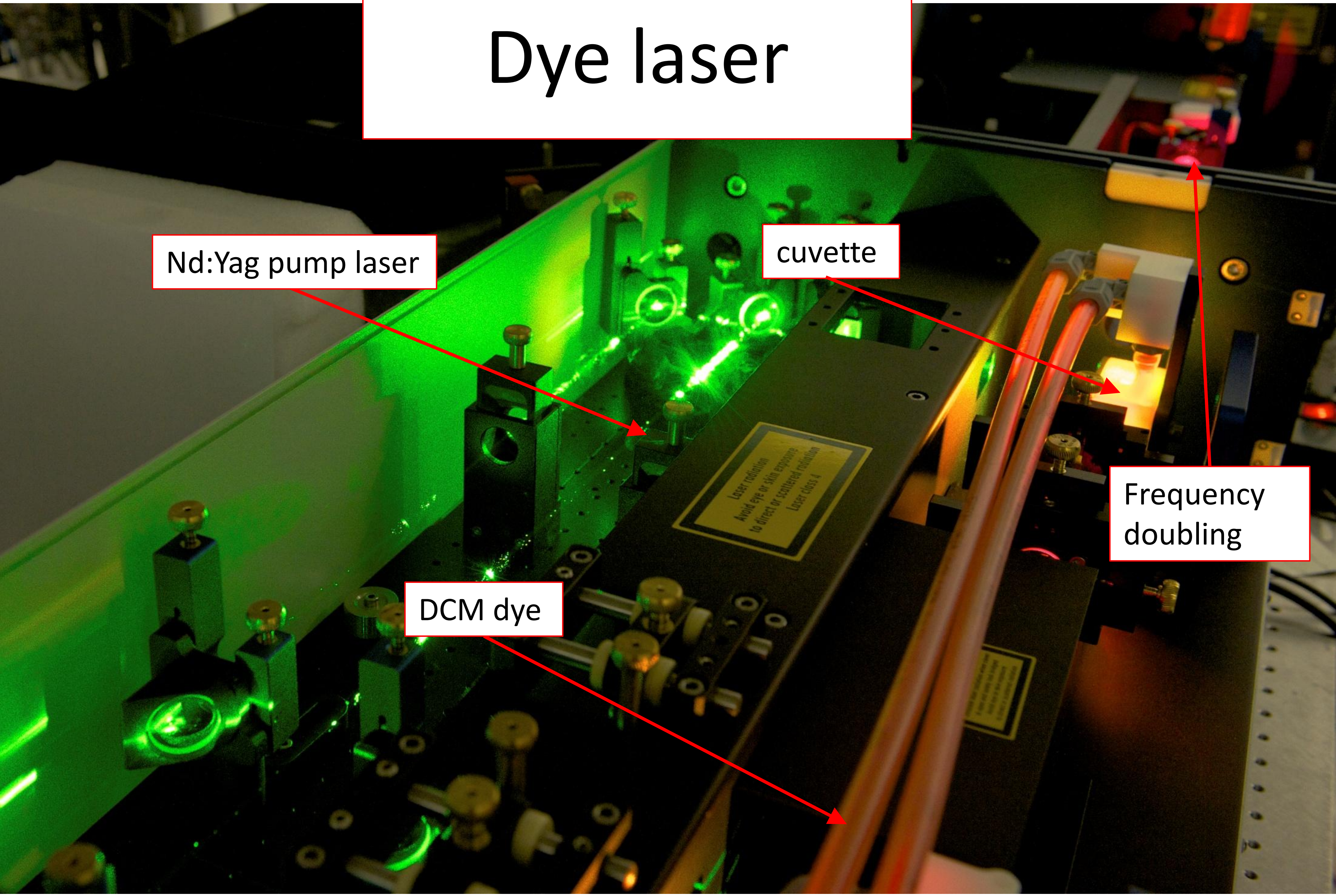


Dye laser with DCM dye: Dye is excited by Yag and fluoresces. A cavity creates light with 624 nm wavelength. This is then frequency doubled to 312 nm.

Experiment

Nd:Yag laser (600 mJ/pp, 10 ns pulse length, Power -> 10ns of MW!!) Provides energy for dye laser

Dye laser



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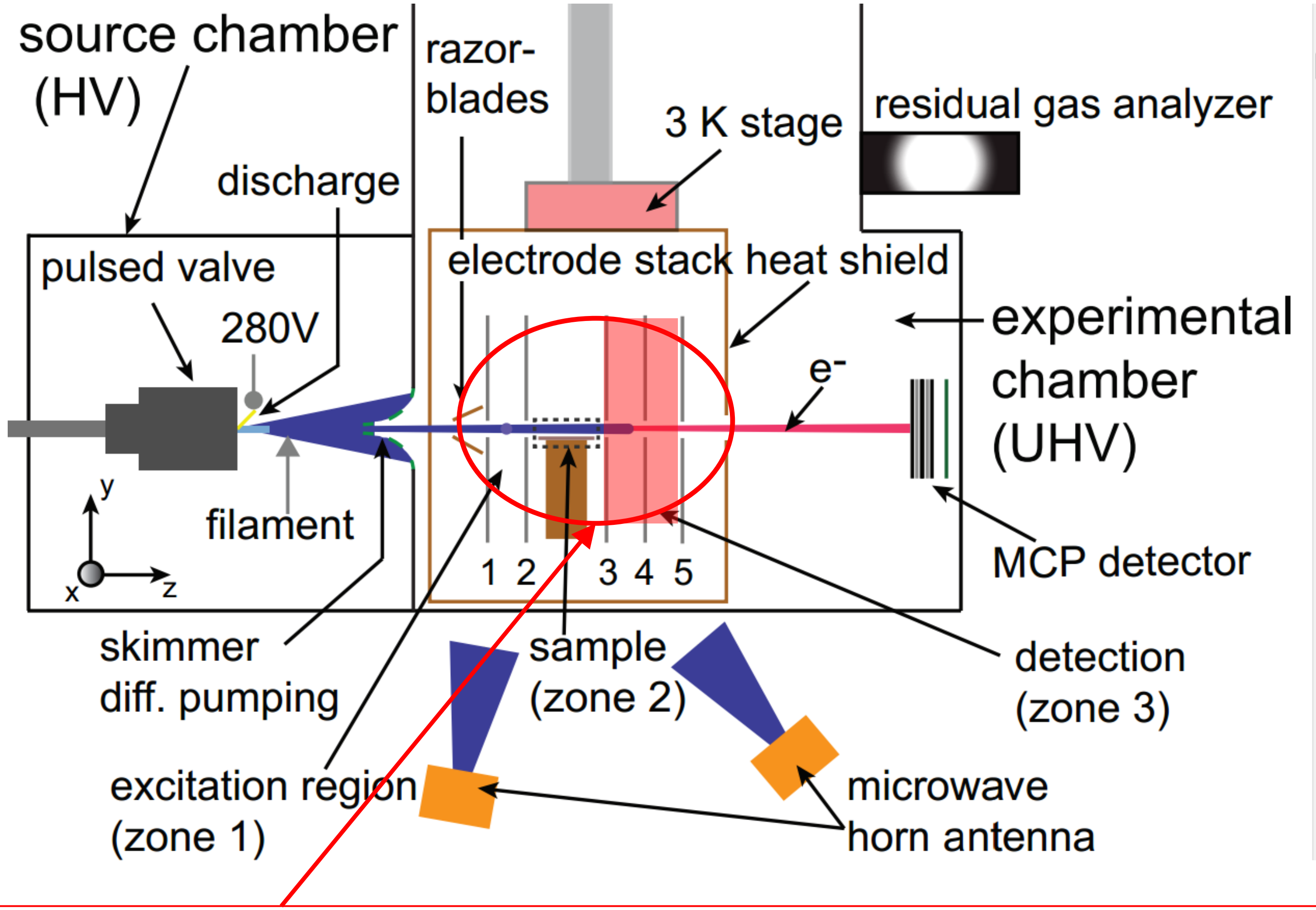
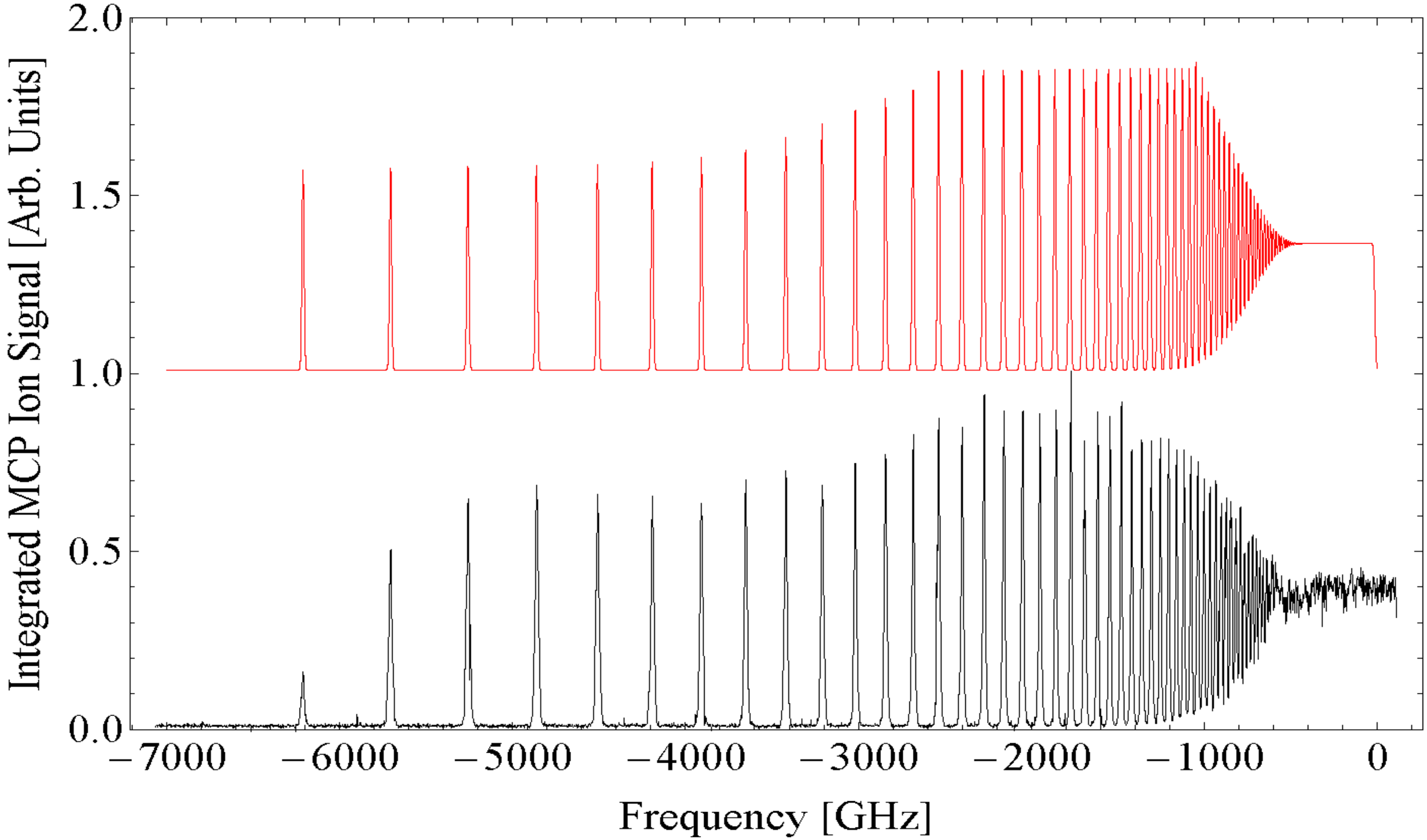


Detection: 1.2 kV/cm electric field applied in 10 ns. Rydberg atoms ionize and electrons are Detected at the MCP detector (single particle multiplier) .

Results TOF 100ns



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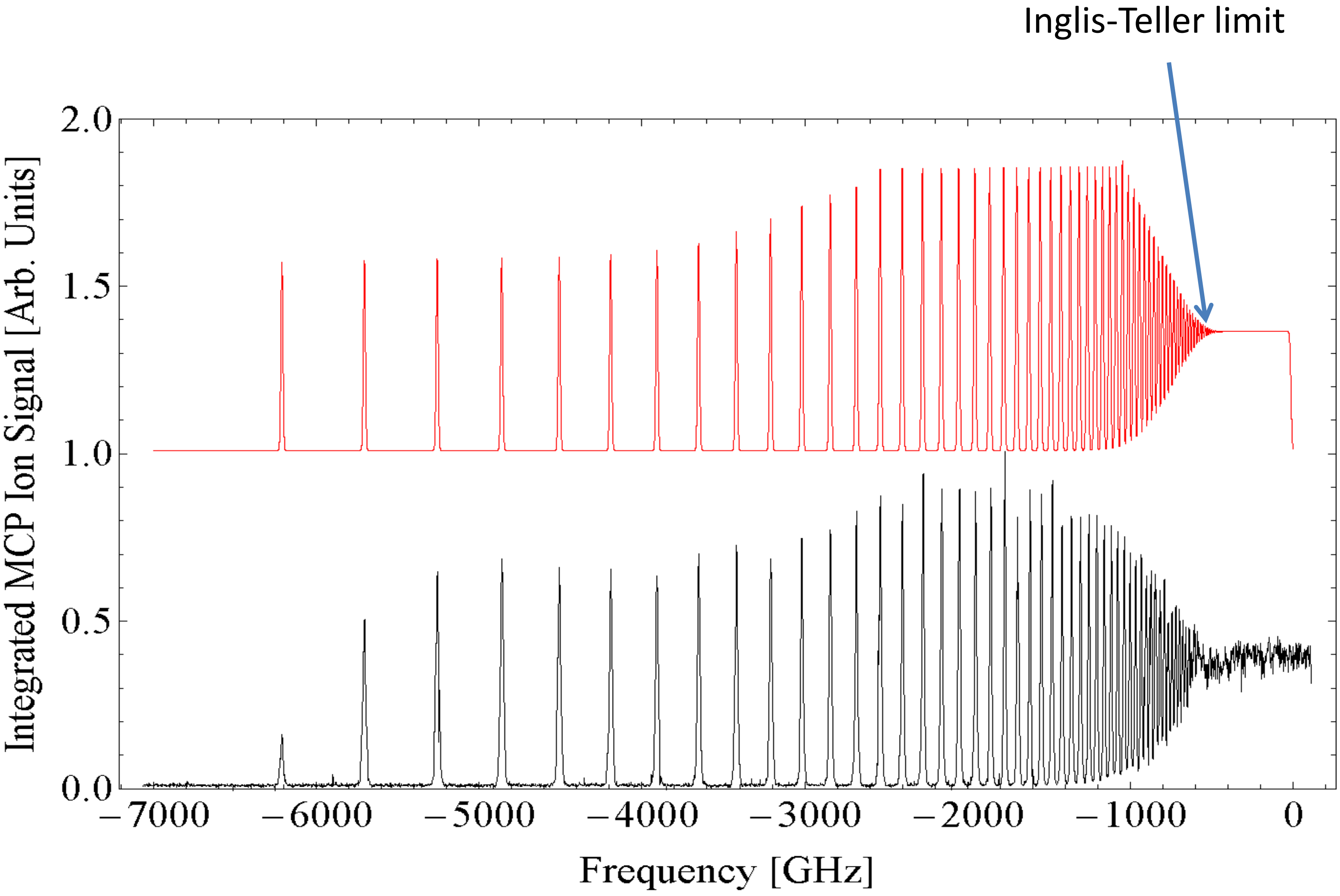
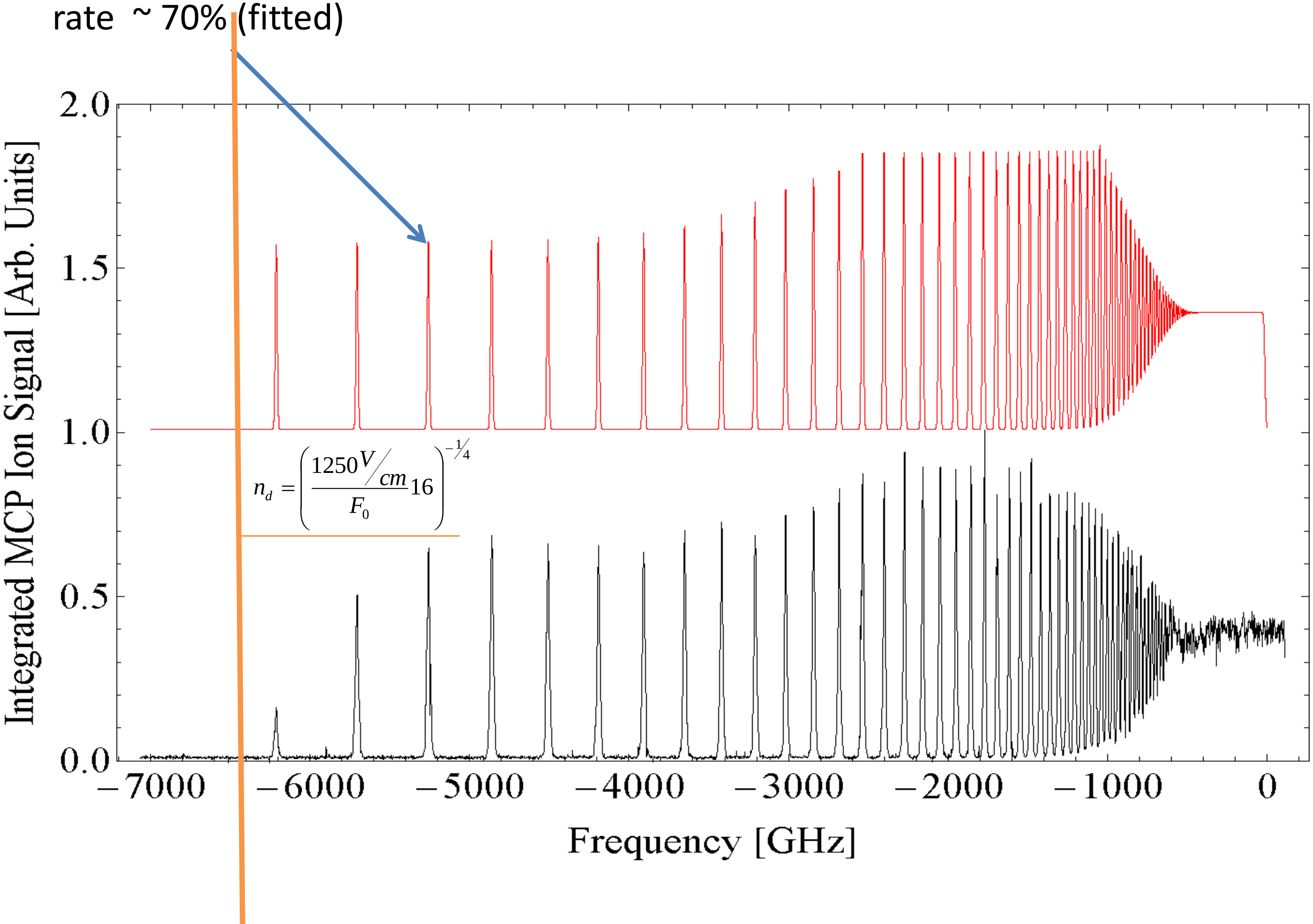


Results TOF 100ns

Adiabatic Ionization-rate ~ 70% (fitted)



Results TOF 100ns

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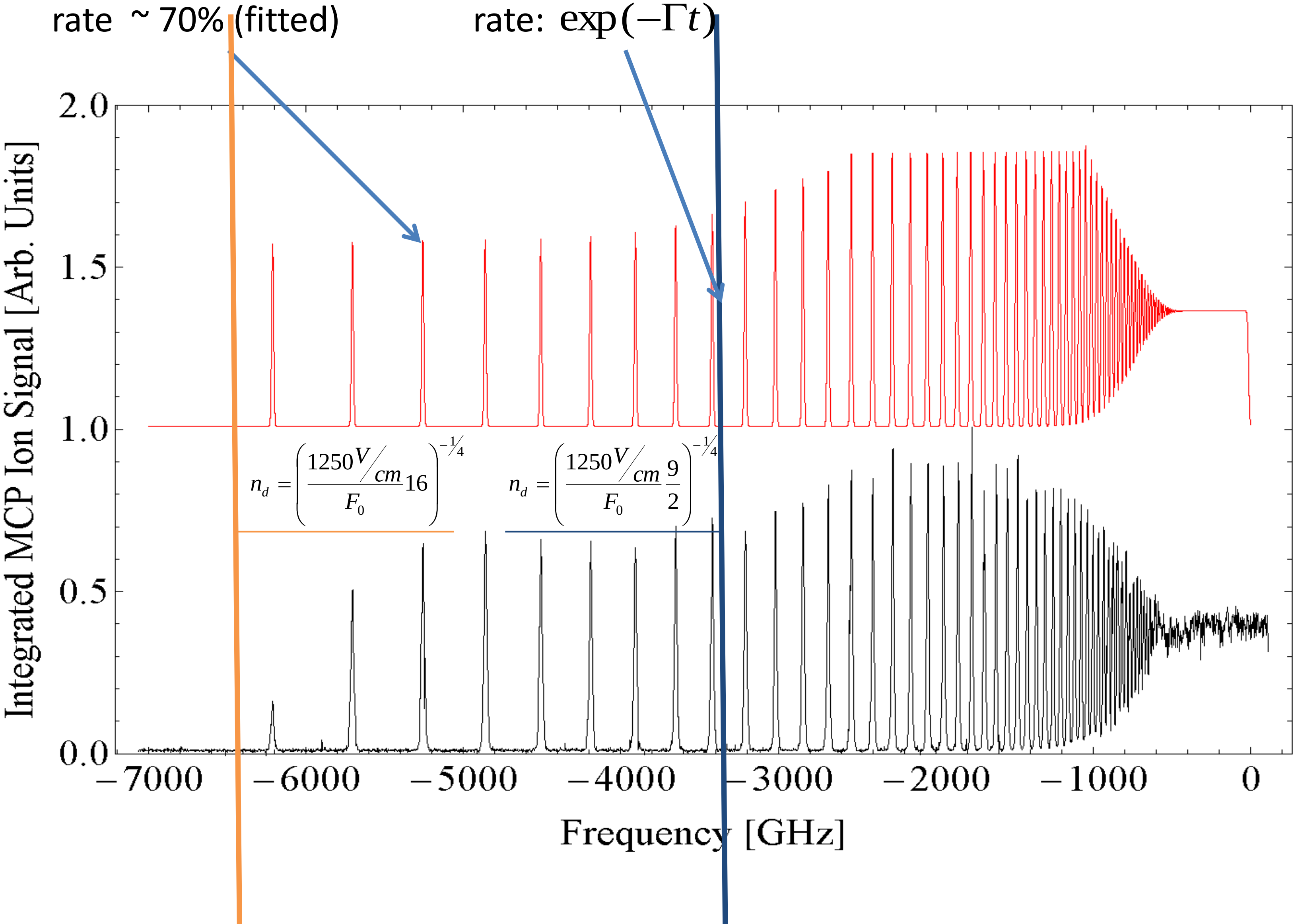
Pure diabatic Ionization-rate: $\exp(-\Gamma t)$



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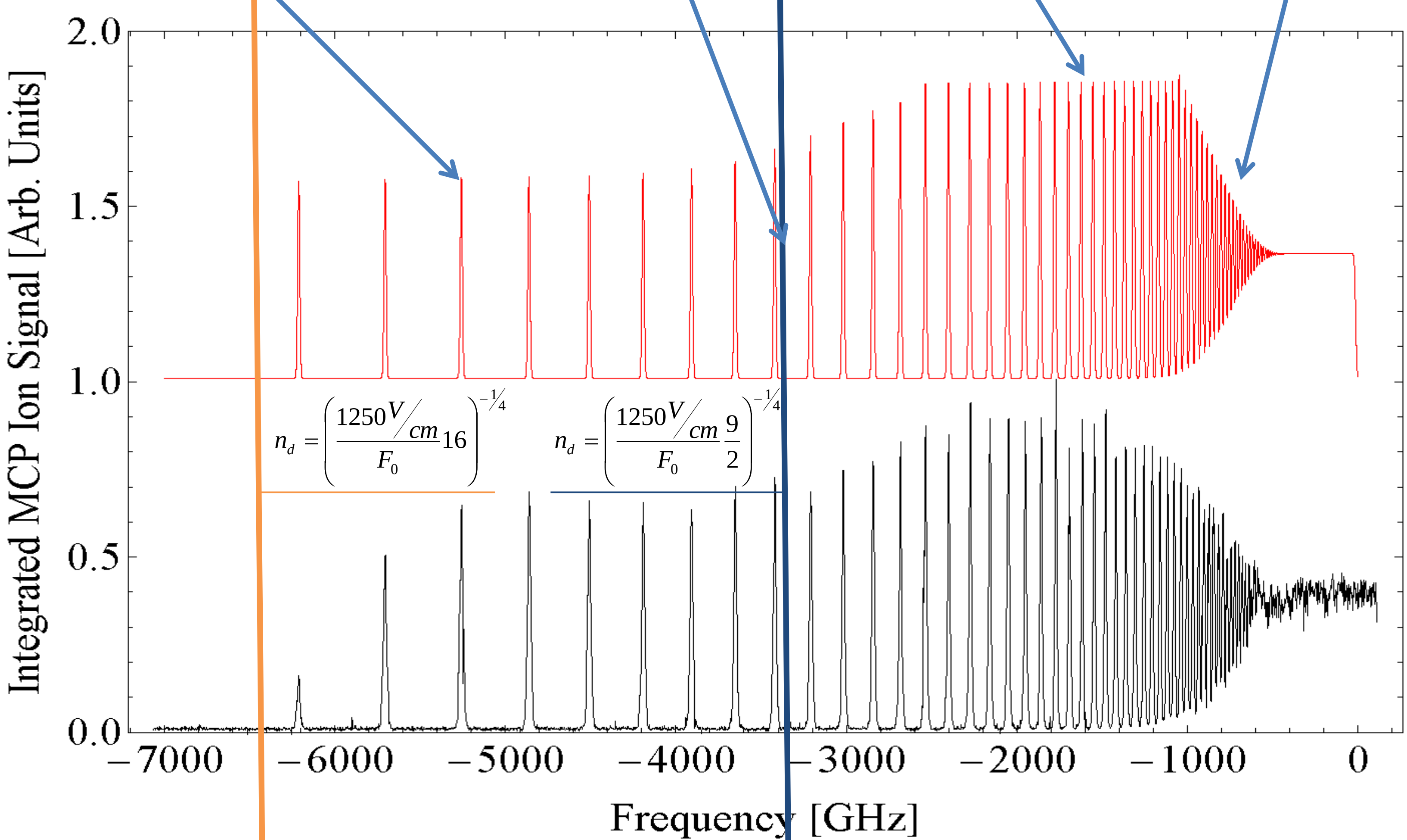
saturation

$\sigma \propto \langle r \rangle^2 \propto n^{-3}$

From ground state



Results TOF 15μs

$\sigma \propto \langle r \rangle^2 \propto n^{-3}$

From ground state