Laser-Ion Acceleration using Gold Nanorods

For plasmonic enhancement of Fusion reactions

Dr. Chris Grayson

FULBRIGHT Hungary

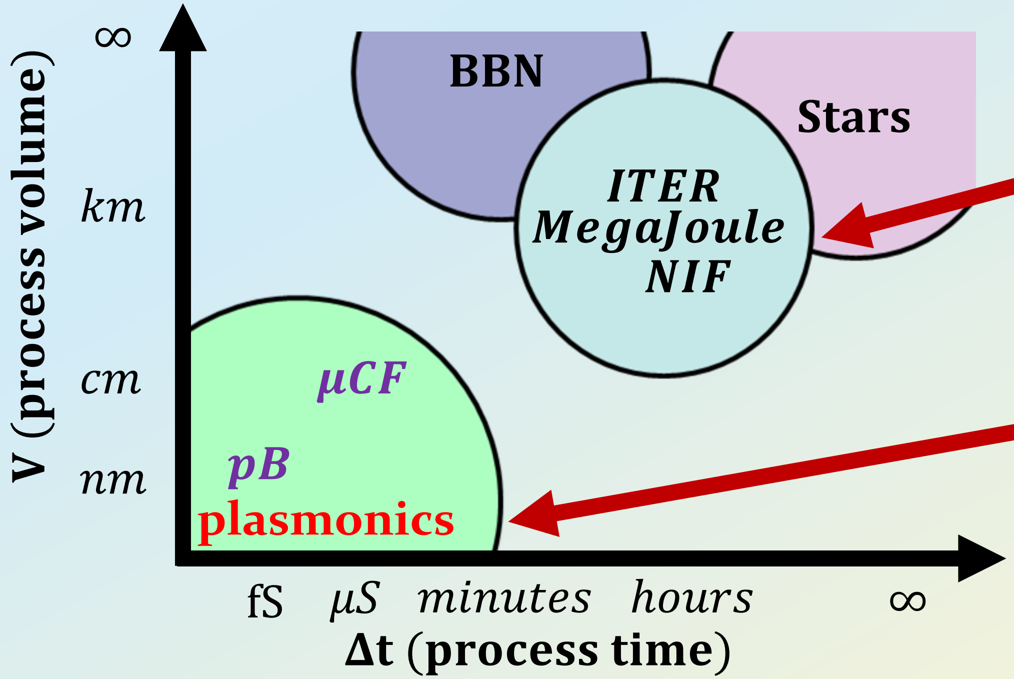
THE UNIVERSITY OF ARIZONA
Physics

WIGNER

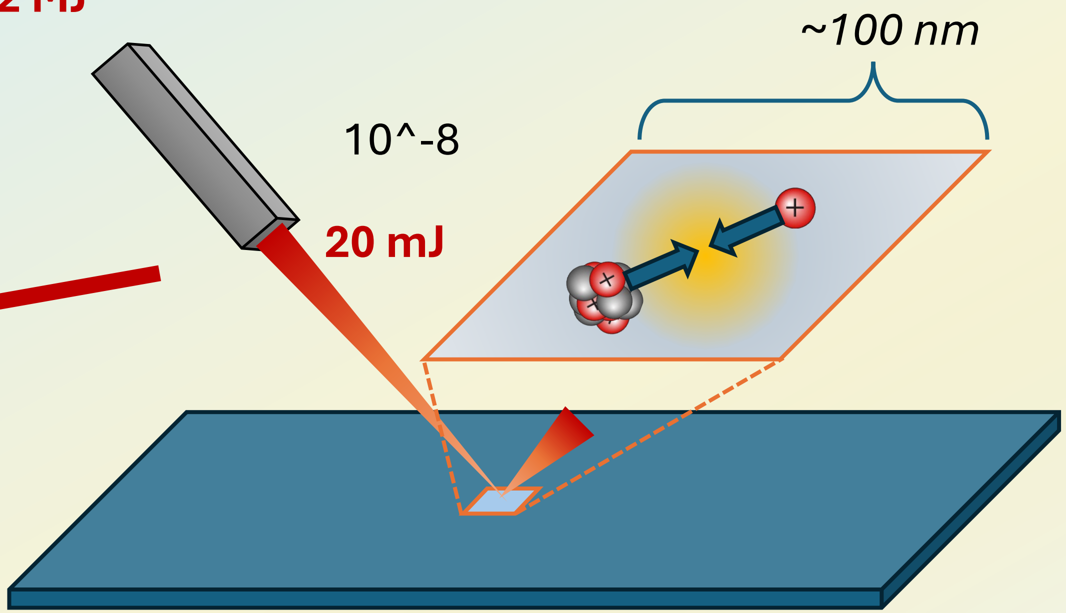
The NanoPlasmonic Laser Induced Fusion Energy (NAPLIFE)

Laser Fusion

- Our goal is to use low energy **lasers** to **accelerate ions** to produce **nonequilbirum nano fusion**.



Equilibrium fusion requires enormous energies and densities
NIF – 2 MJ



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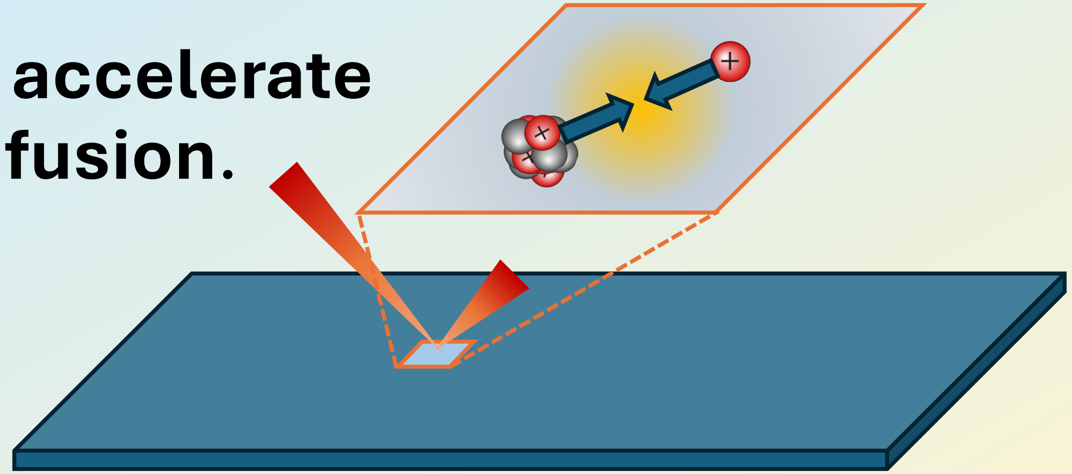

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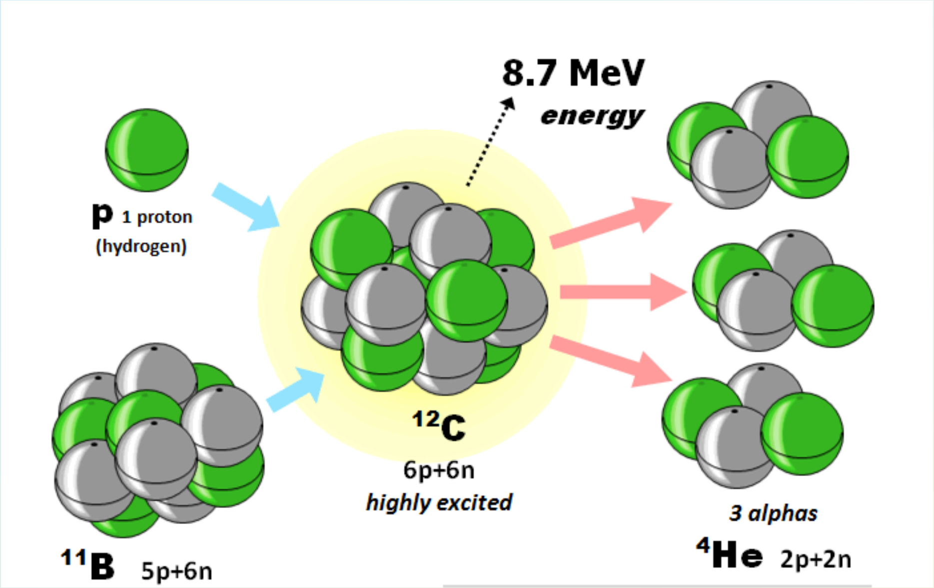

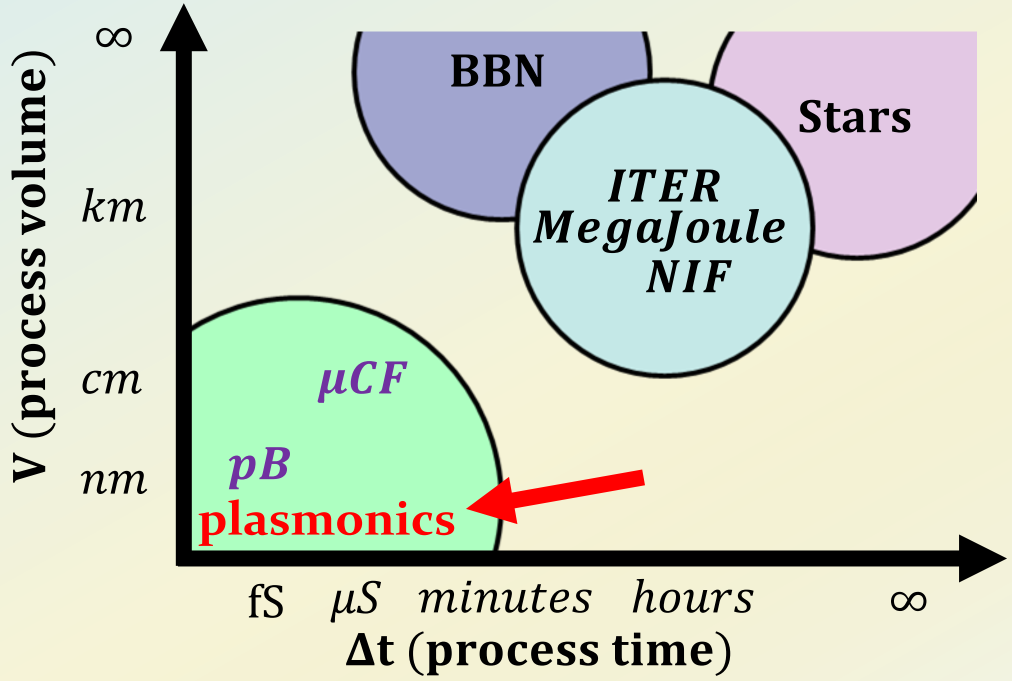

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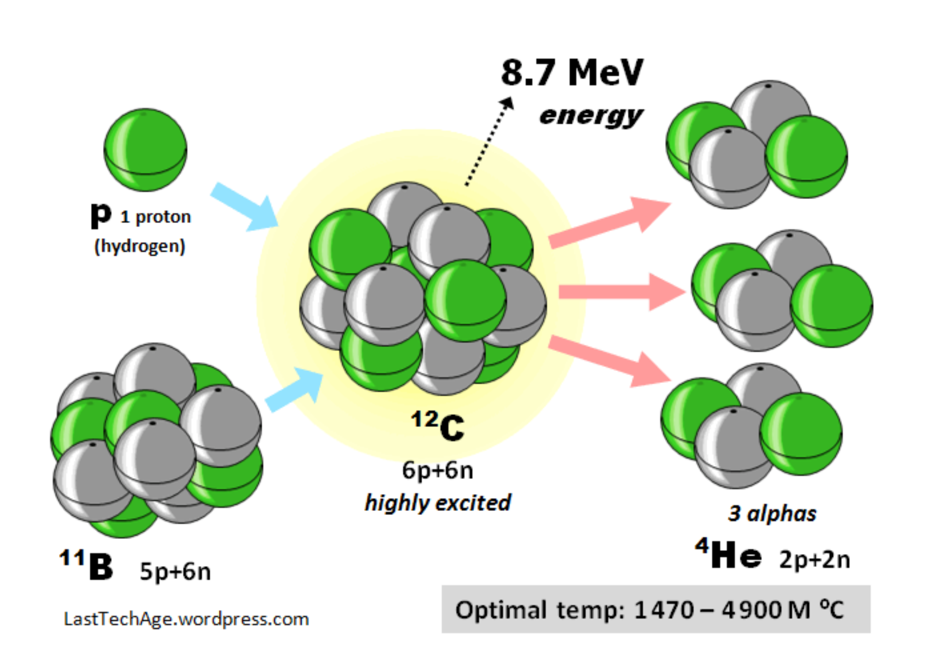

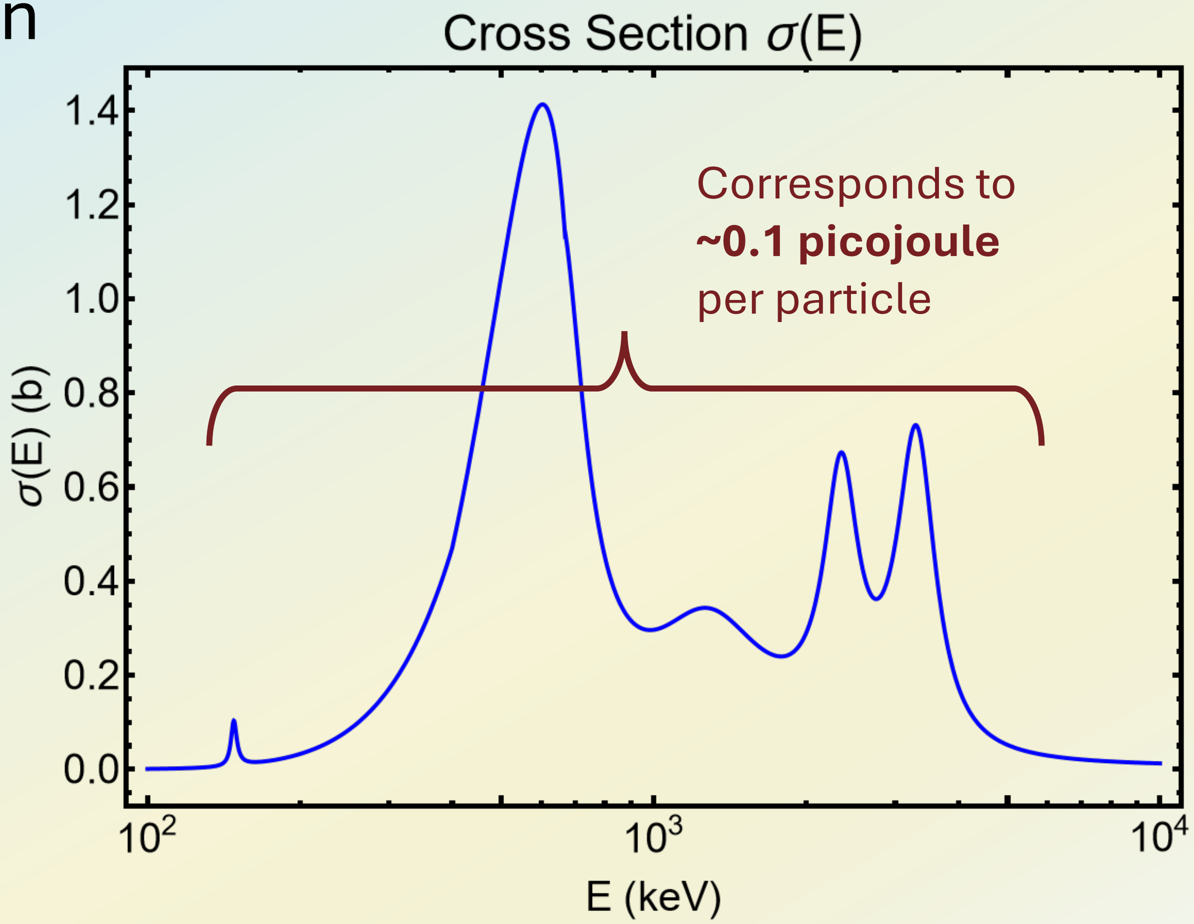

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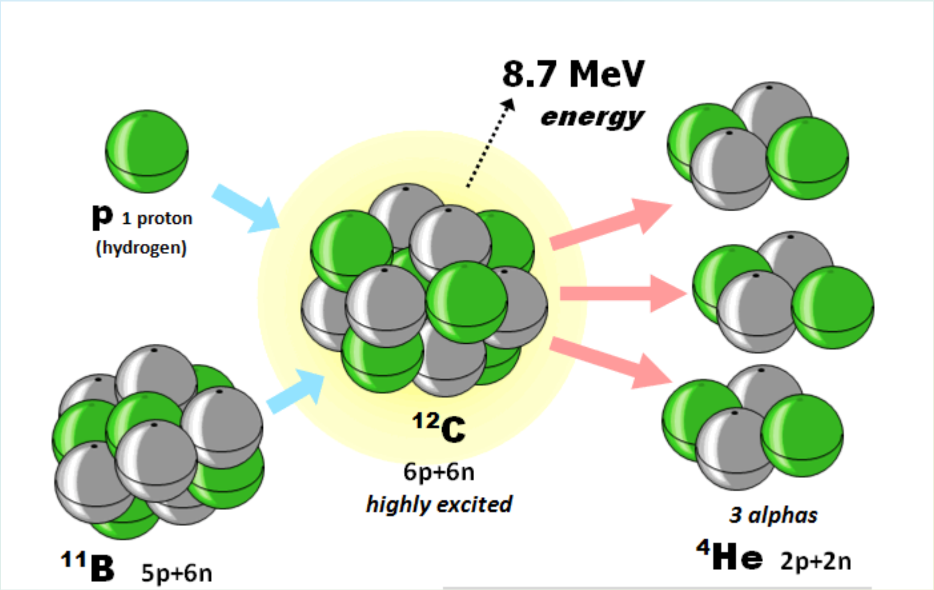

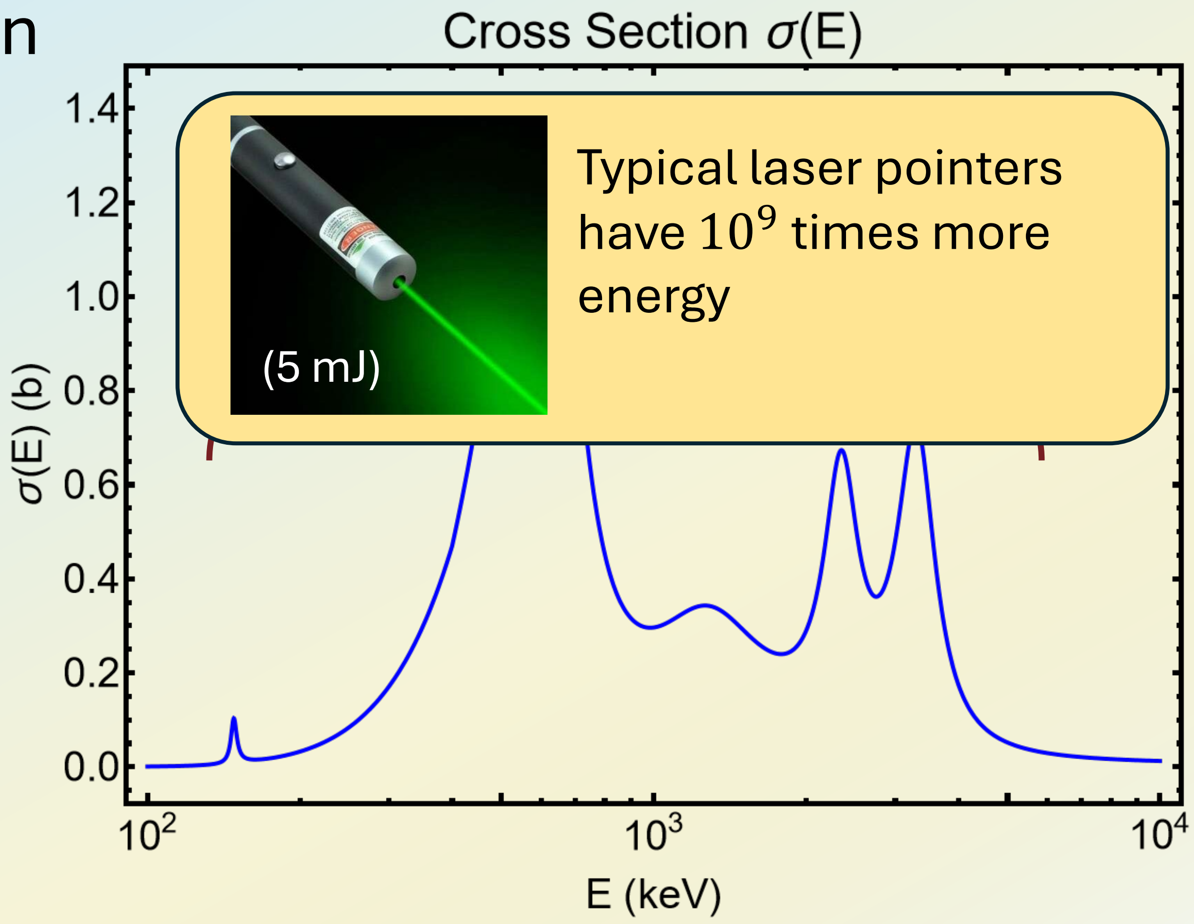

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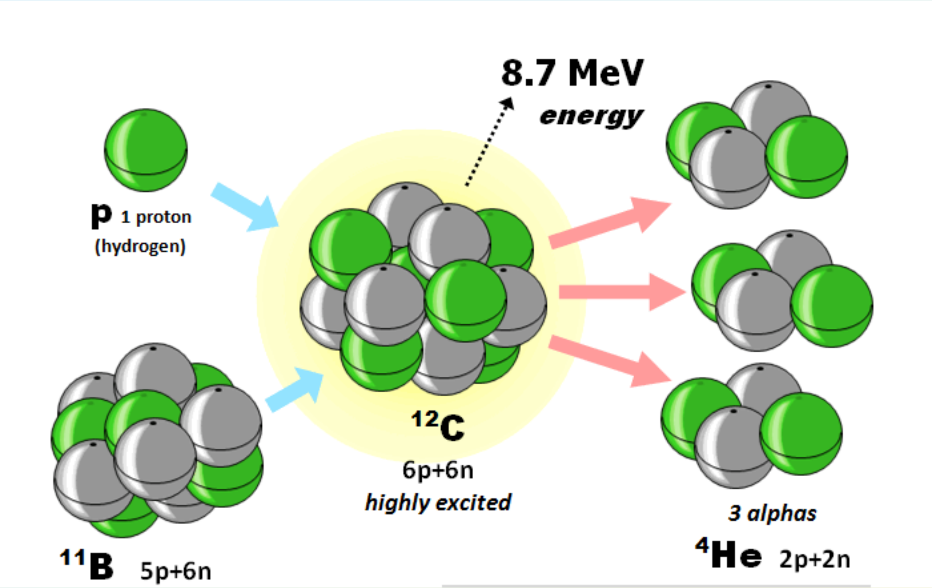

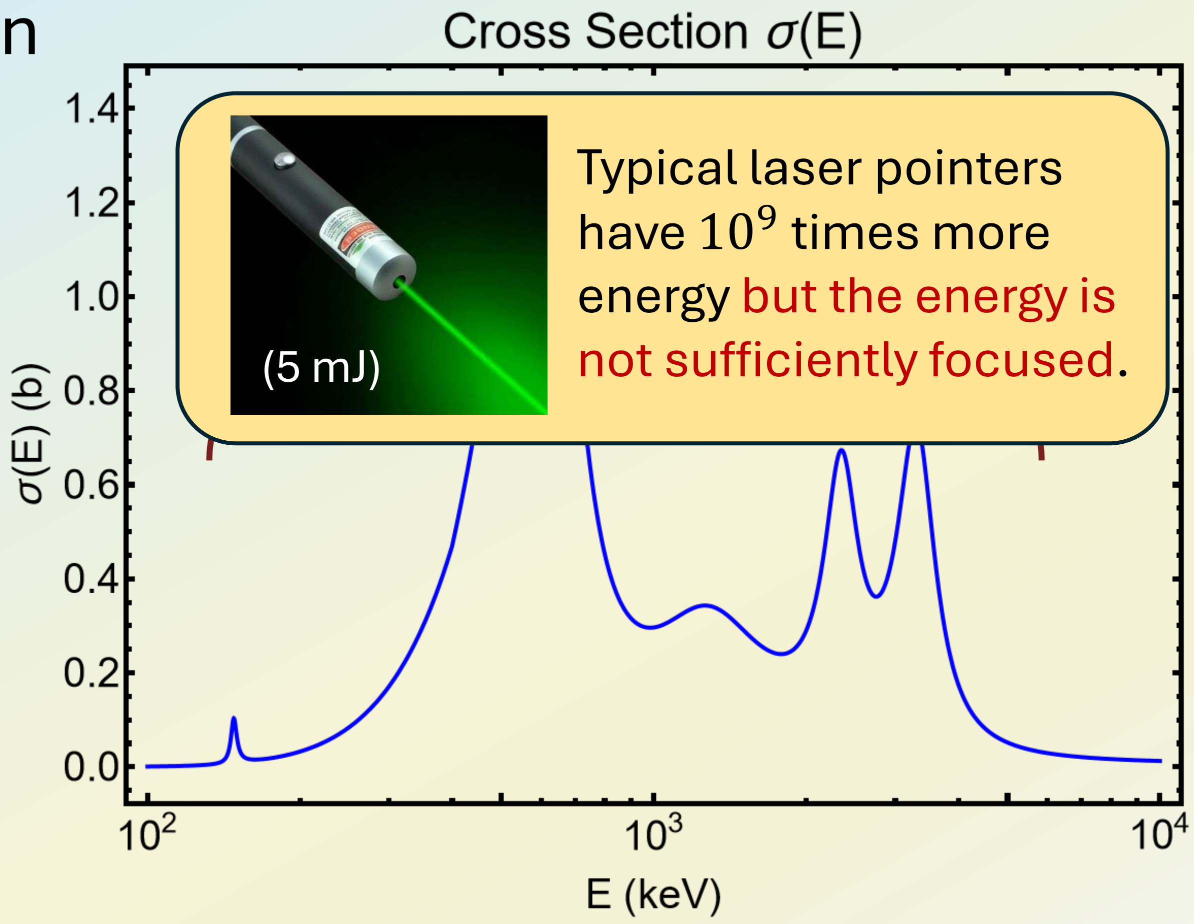

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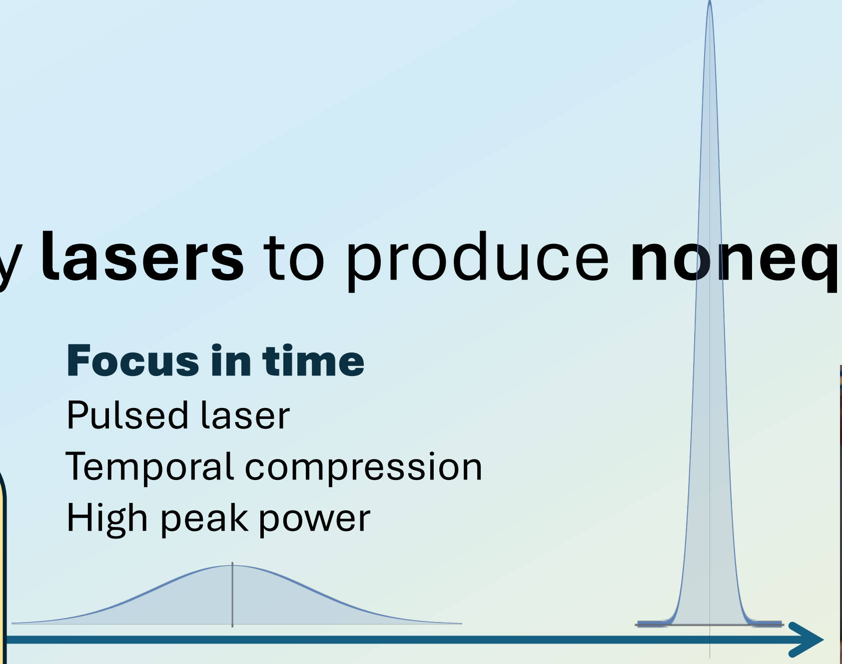
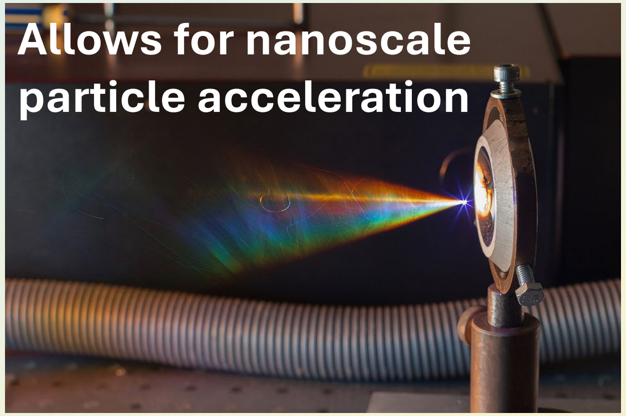
Focus in time
Pulsed laser
Temporal compression
High peak power



5mW → **50 GW**
(5×10^{10} W)



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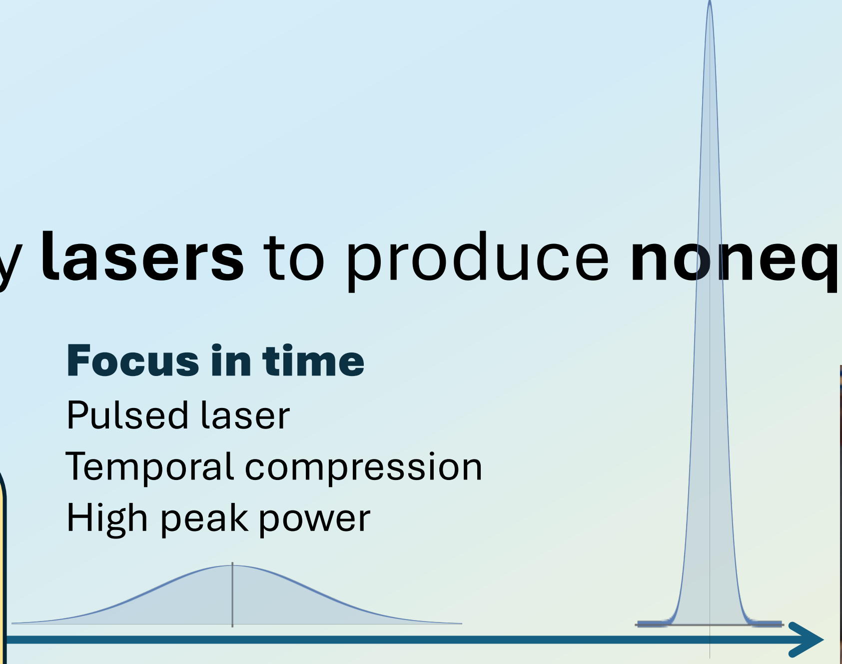

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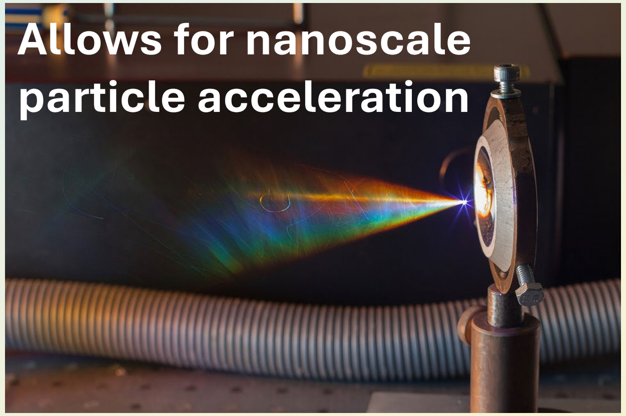

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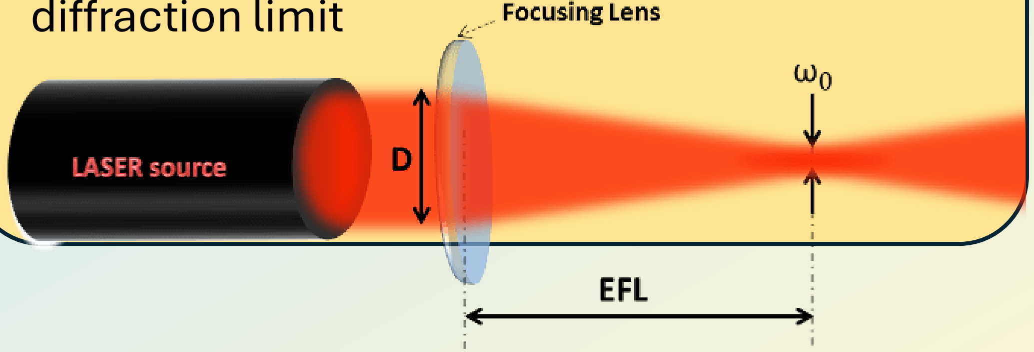
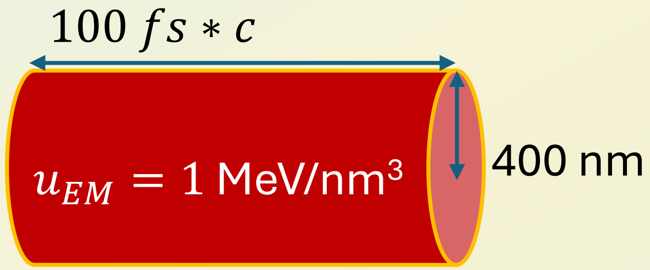
Light cannot be focused past the diffraction limit

$$d_{limit} \approx \frac{\lambda}{2} \approx 400\ nm$$





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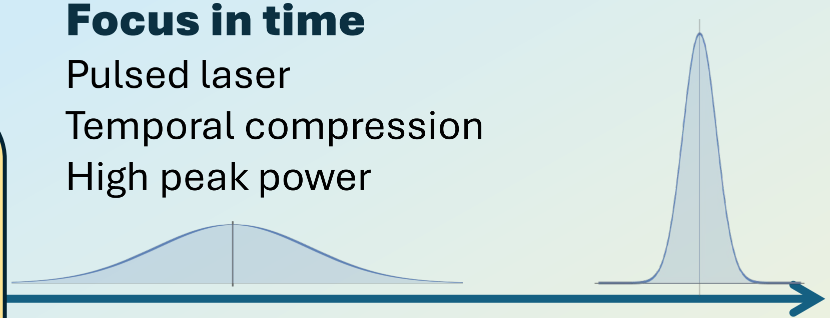

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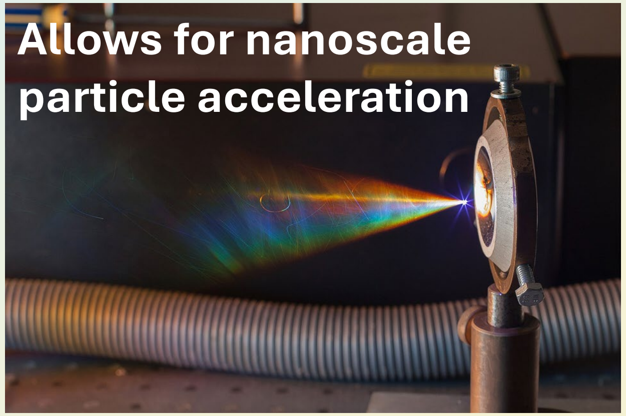

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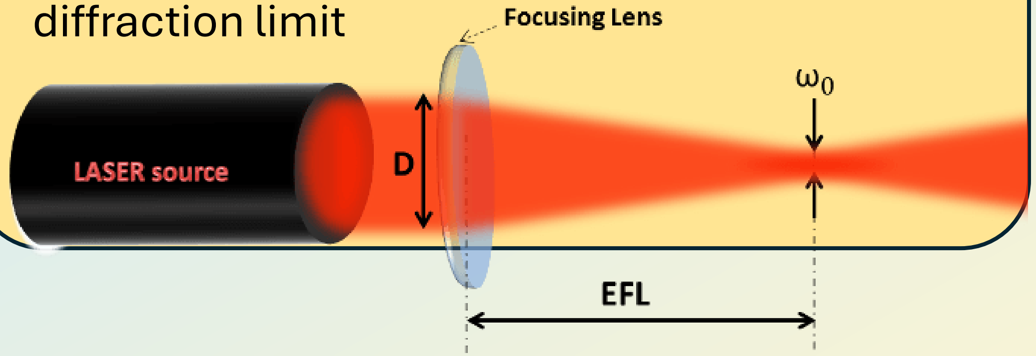
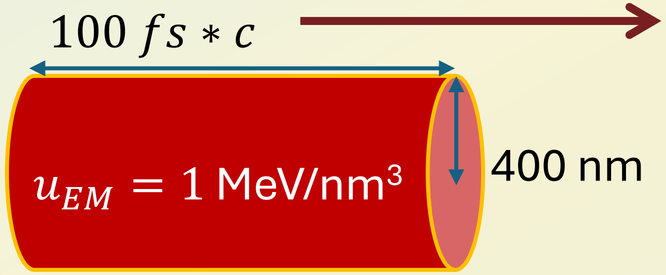
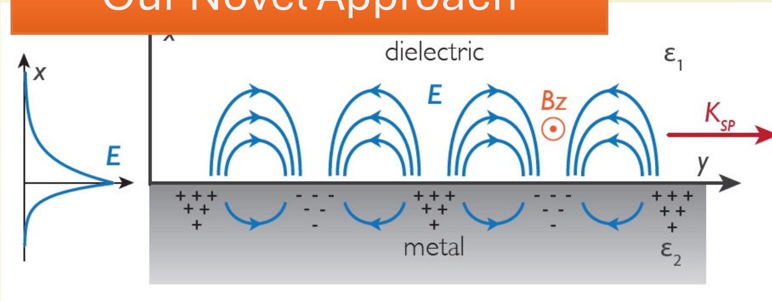


Focus in space

Couple to massive electron wave
Surface Plasmon Resonance (LSPR)



Our Novel Approach



Focuses light past the diffraction limit

$400\ nm \rightarrow 85\ nm$

The NanoPlasmonic Laser Induced Fusion Energy

How can **nano-structured** targets lead to **femtosecond** scale **ion acceleration**?

Huge Energy ***mismatch*** *between* ***laser*** *and* ***Fusion*** *threshold*

$E = \hbar\omega_{laser} = 2\ eV \ \rightarrow 100's\ keV$ *50,000 photons*

"Atomic" **"Nuclear"**

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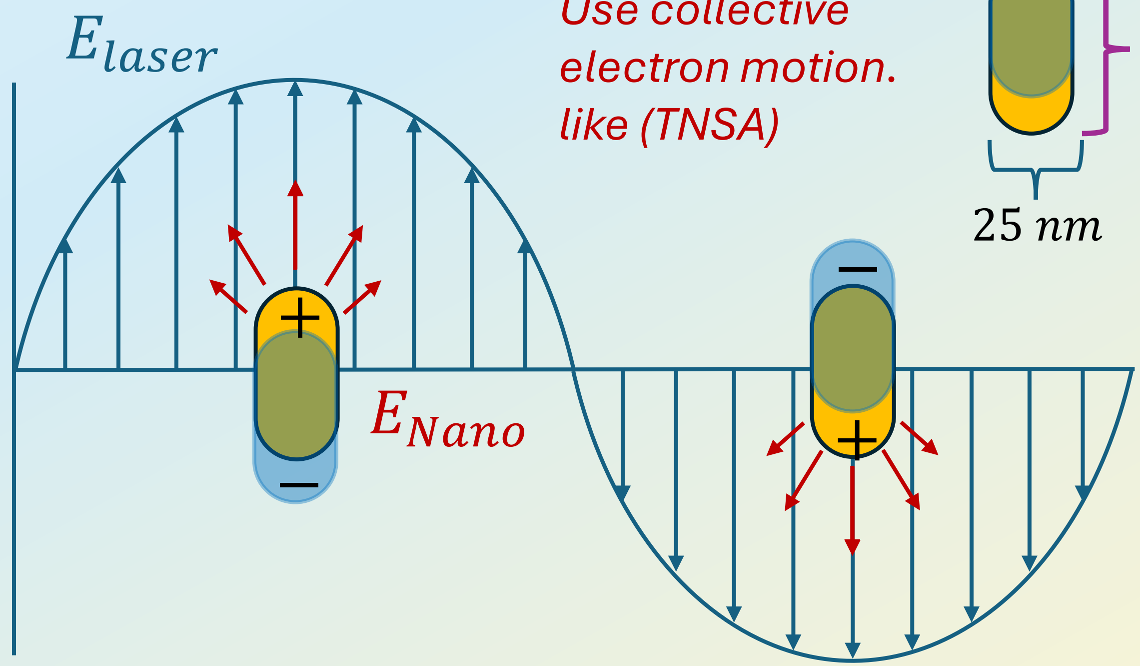
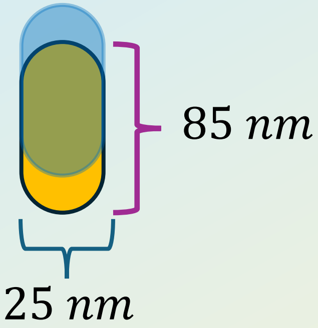
<u>*Solution*</u>

Use collective electron motion. like (TNSA)





*Locally **increased** Electric field **Strength** and Field **Gradient***

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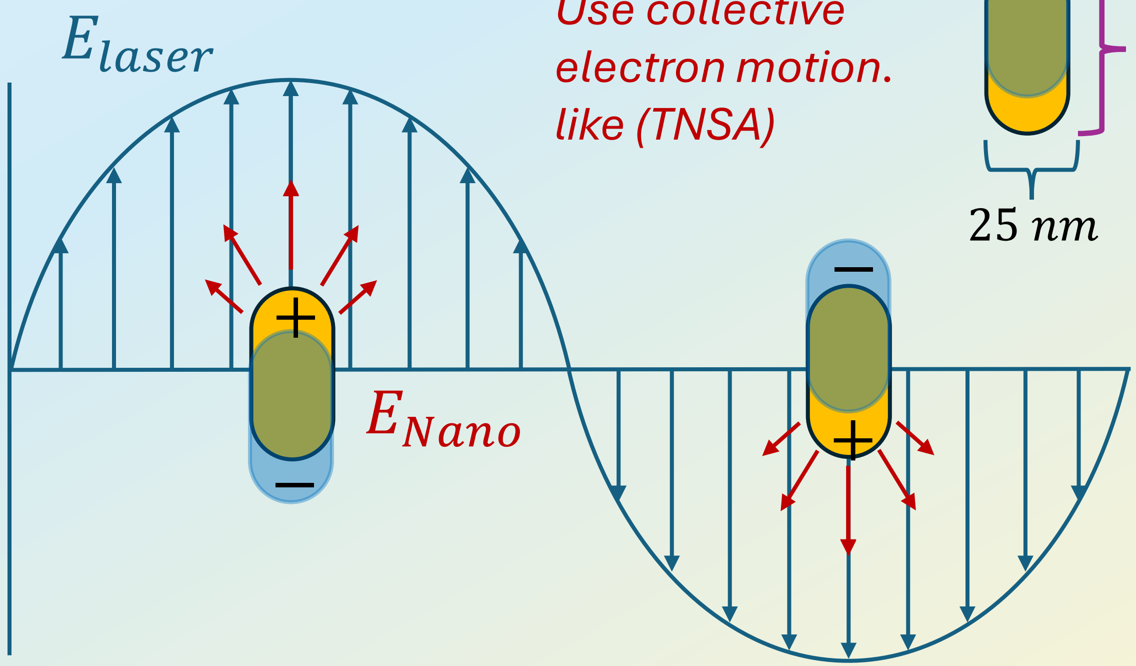

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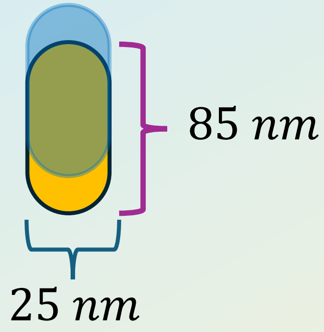
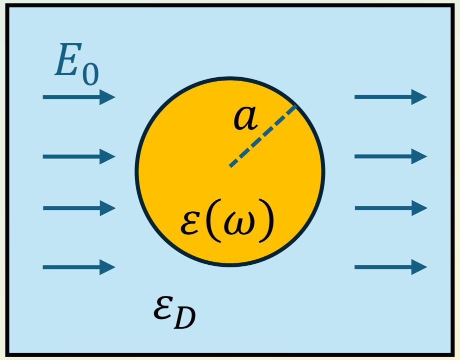


Antennas For Light

These nanorods **resonantly** absorb 800 nm light via **Localized Surface Plasmon resonance** (LSP)

$$\boldsymbol{p} = \varepsilon_0\, \varepsilon_D\, \alpha\, \boldsymbol{E_0} \qquad \alpha = 4\,\pi\, a^3 \frac{\varepsilon(\omega) - \varepsilon_D}{\varepsilon(\omega) + 2\,\varepsilon_D}$$



The classical **polarizability diverges** when

$$Re[\varepsilon(\omega)] = -2\,\varepsilon_D$$

Maier, Stefan A. Plasmonics: fundamentals and applications. Vol. 1. New York: springer, 2007.

LSP **resonance** can **respond** incredibly **fast.**

$\omega_{LSP} = 2.65\ fs$

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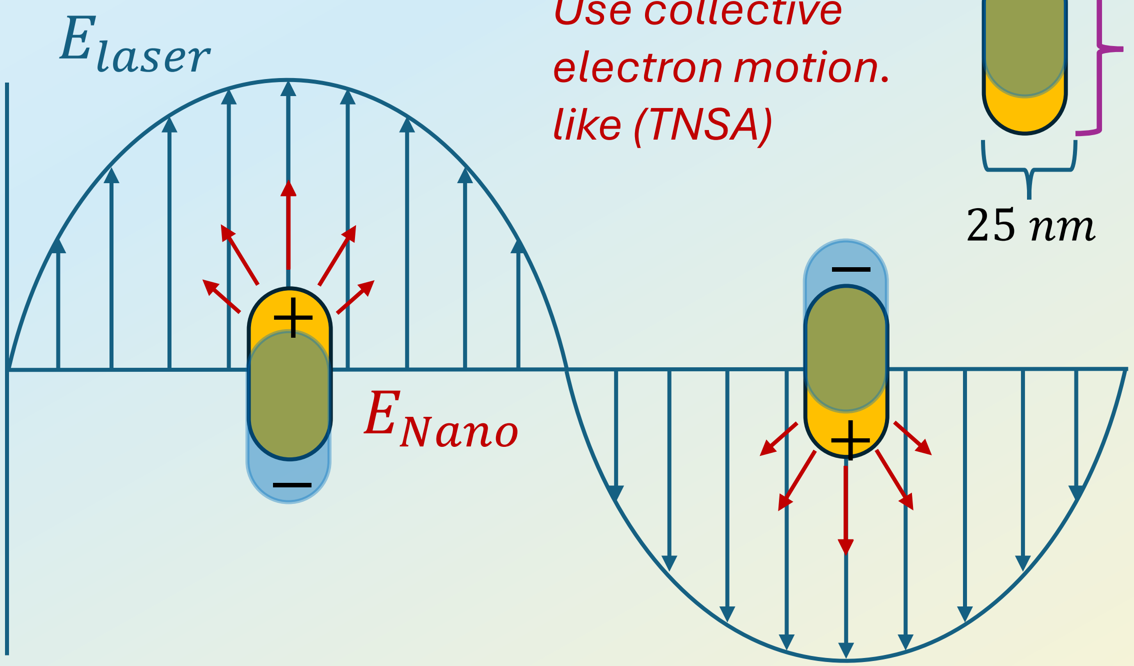

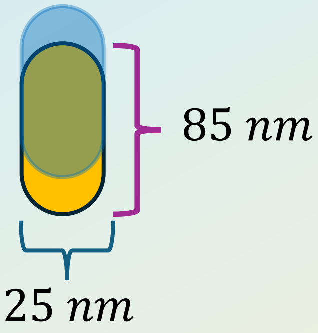

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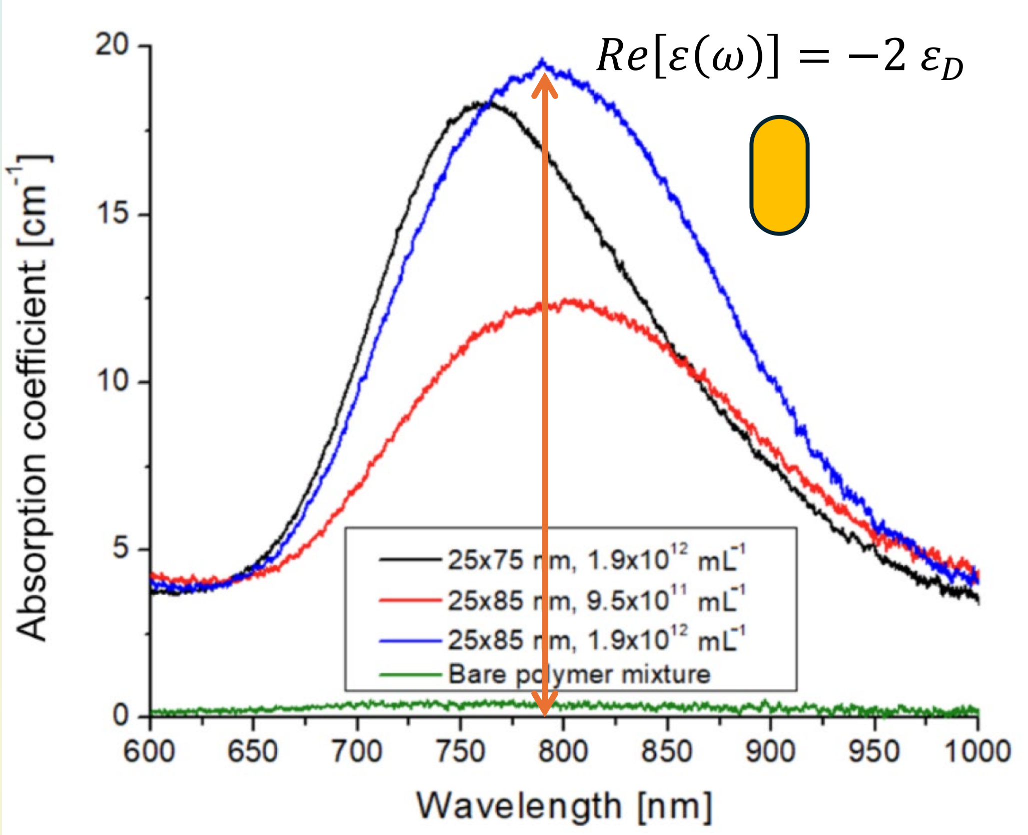

Bonyár, A. et al. Int. J. Mol. Sci. 2022, 23, 13575.

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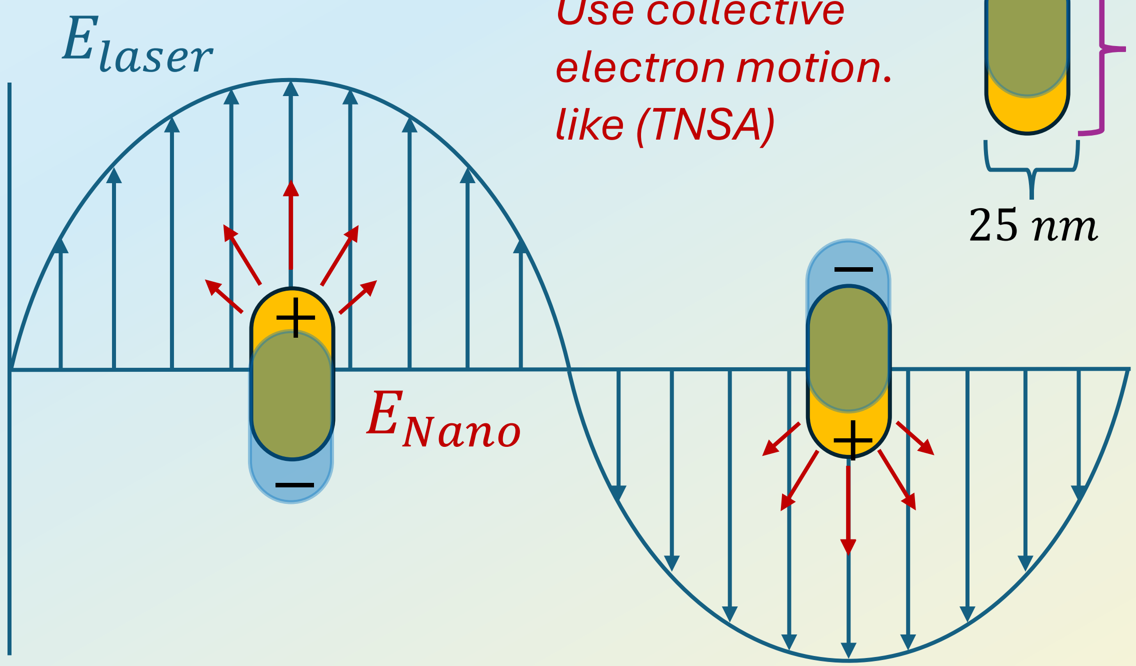

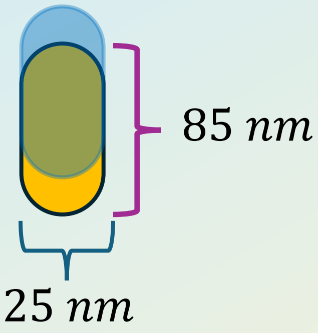

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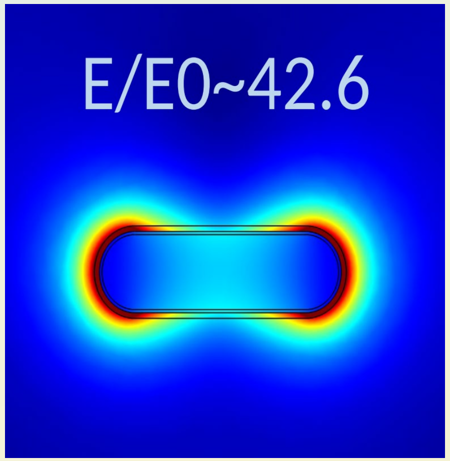
These nanorods **resonantly** absorb 800 nm light via **Localized Surface Plasmon resonance** (LSP)

Near Field Enhancement (NFE) of the electric field around the Nano rods causes **increased** ion **acceleration**



The **electric** field around the nanorod E is **enhanced** over the background **laser** field E_0 on the scale of 10's of nm

M. Csete Group: Dávid Vass, Emese Tóth, András Szenes, Balázs Bánhelyi

Field enhancement and absorption measured at low intensity: $I \sim 10^{12}$ W/cm^2

The NanoPlasmonic Laser Induced Fusion Energy (NAPLIFE)

$I \approx 10^{17} - 10^{18} \mathrm{W/cm^2}$

eli

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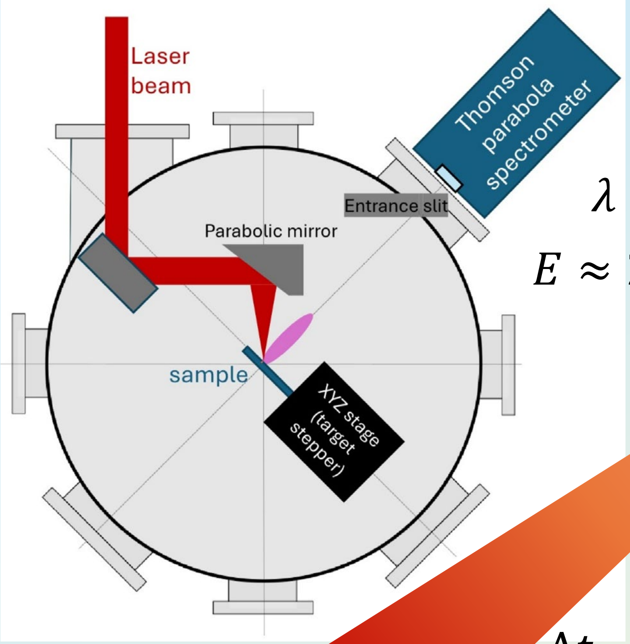
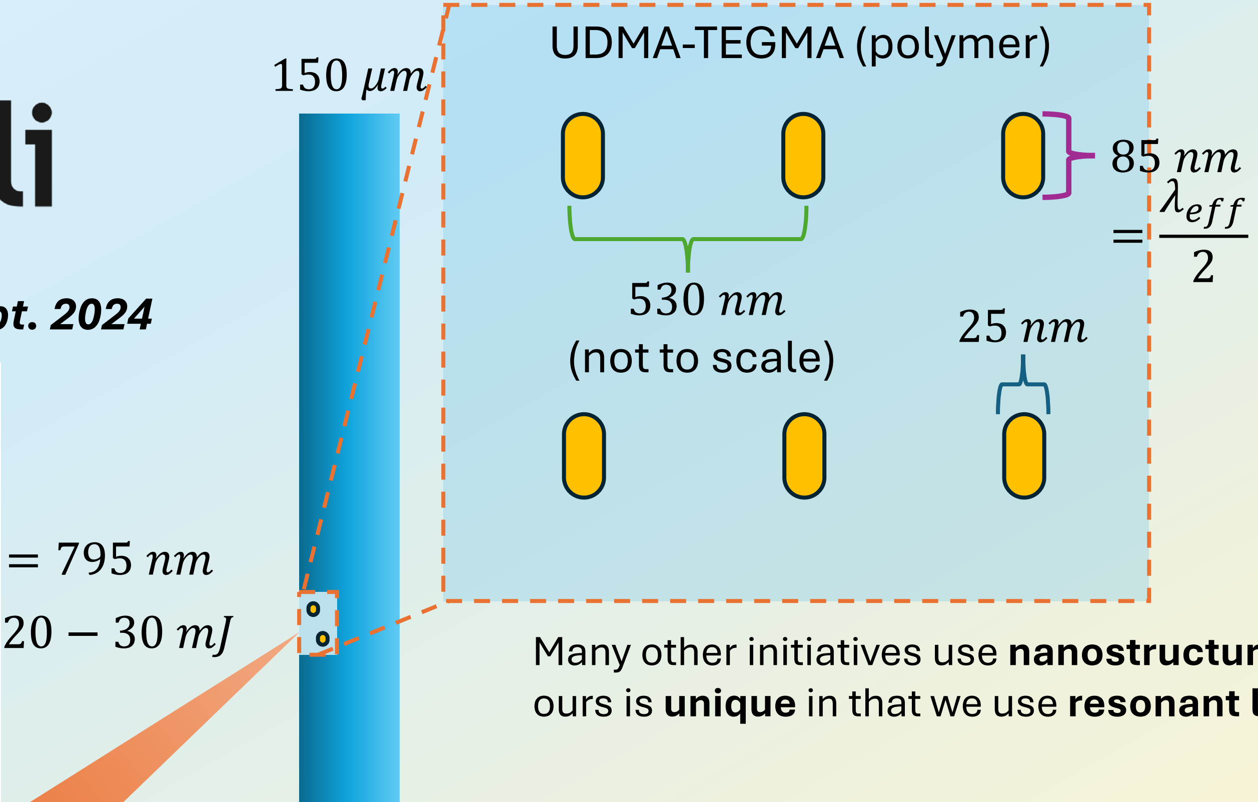
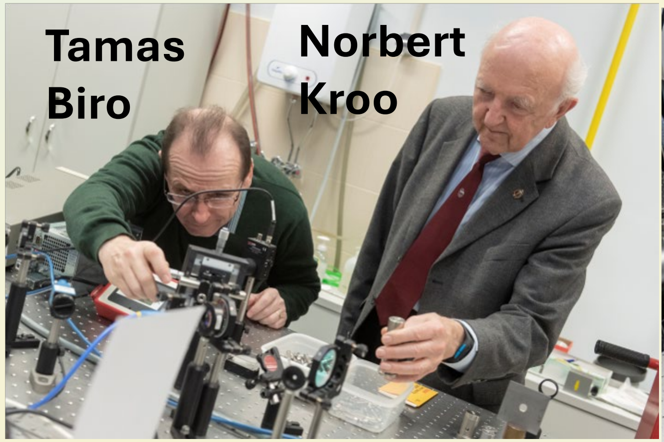
Joined Sept. 2024





The NAPLIFE project introduces **nanostructured laser targets** to generate more efficient **proton acceleration** for ultra-fast pulsed lasers.

Many other initiatives use **nanostructures** to **enhance** ion **acceleration** – ours is **unique** in that we use **resonant localized plasmons (LSPR).**



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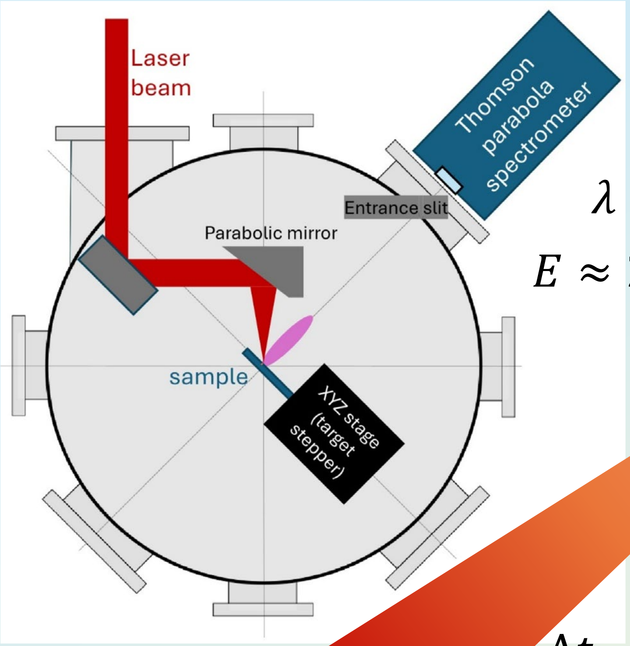

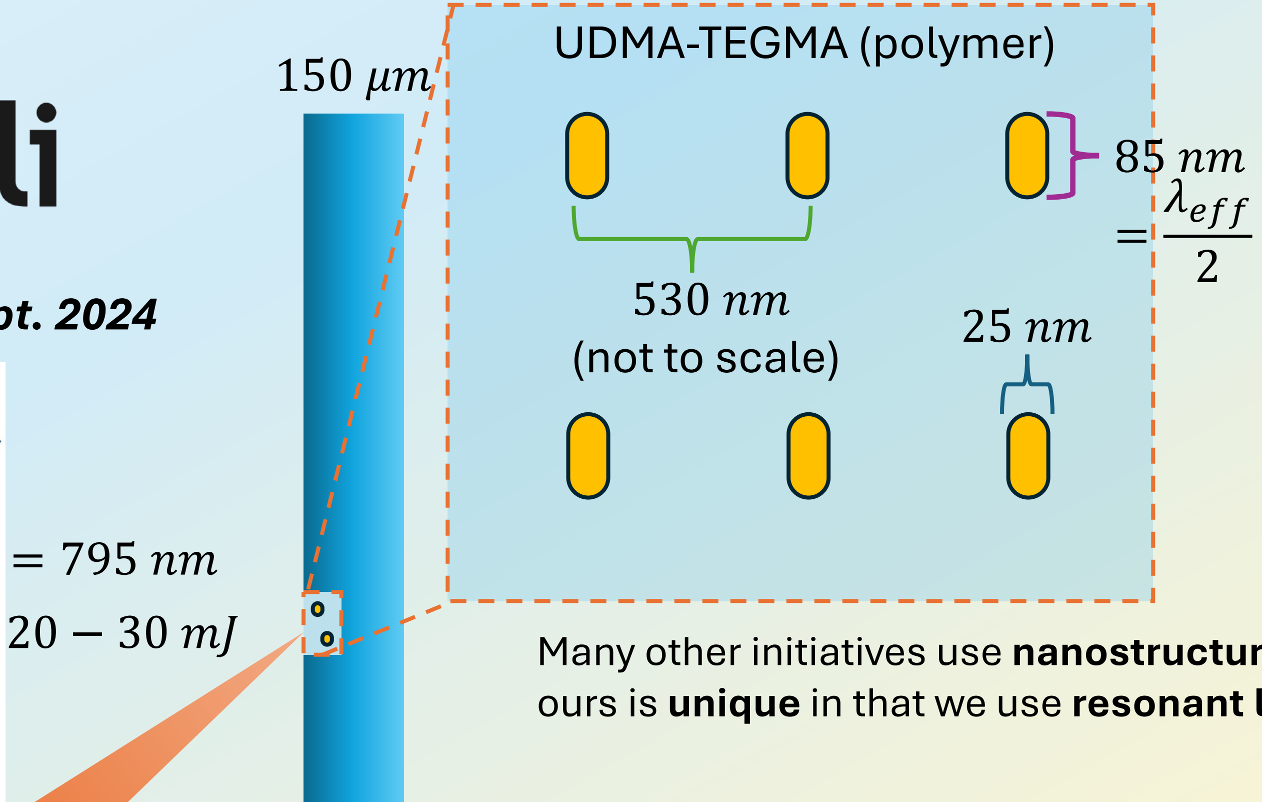

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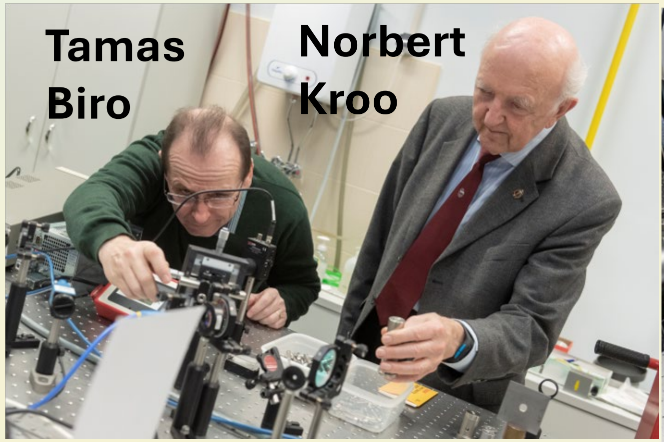

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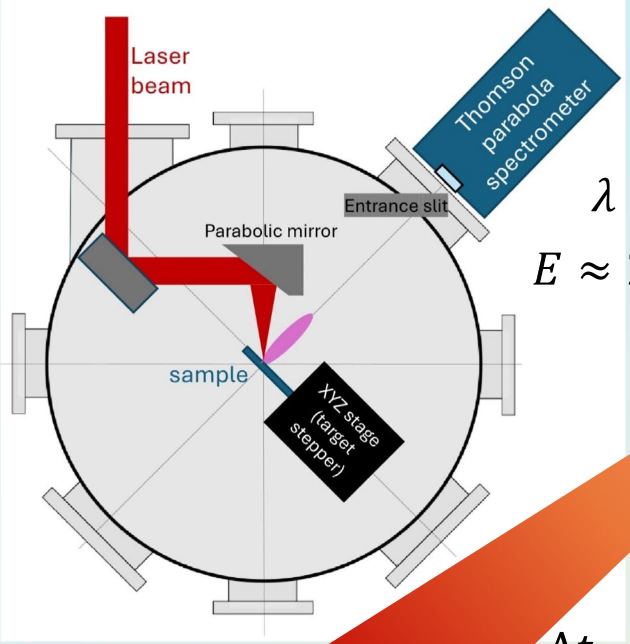

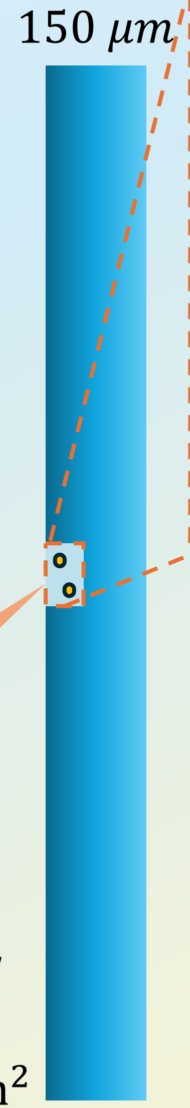

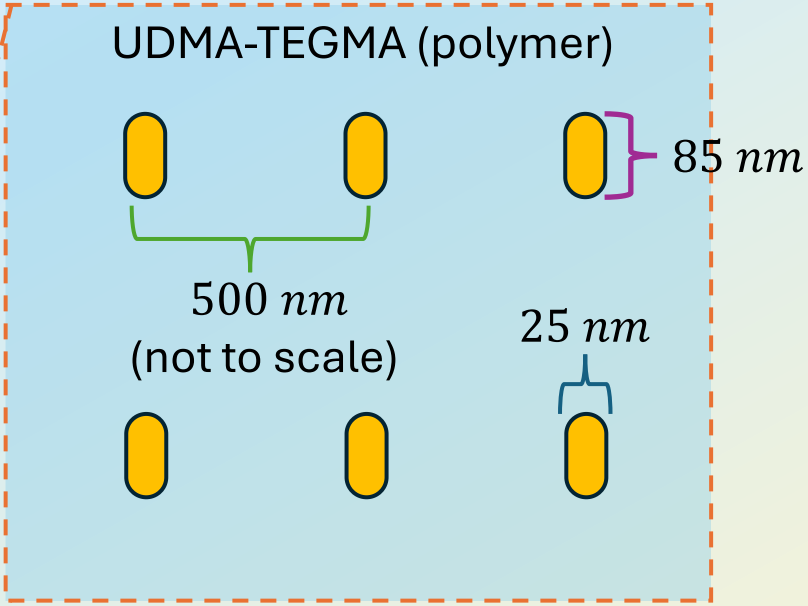

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Article | Open access | Published: 03 December 2024

Indication of $p + {}^{11}B$ reaction in Laser Induced Nanofusion experiment

N. Kroó, L. P. Csernai, I. Papp, M. A. Kedves, M. Aladi, A. Bonyár, M. Szalóki, K. Osvay, P. Varmazyar, T. S. Biró & (for the NAPLIFE Collaboration)

Scientific Reports **14**, Article number: 30087 (2024) | Cite this article

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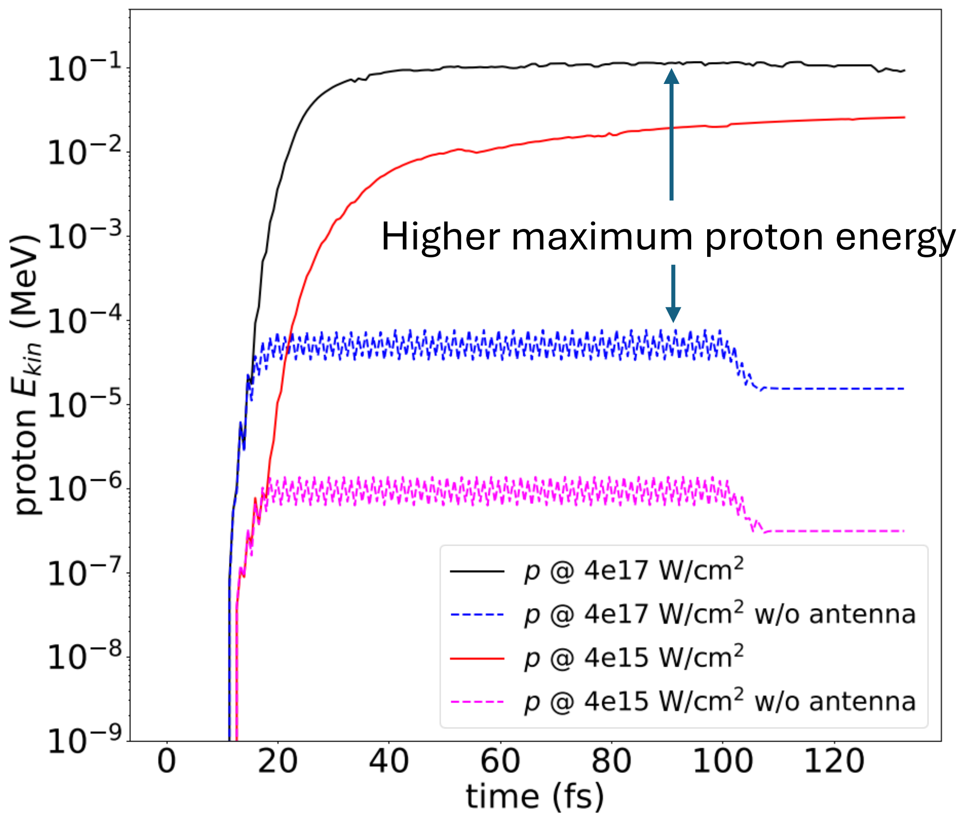
The NanoPlasmonic Laser Induced Fusion Energy

NAPLIFE Previous Results

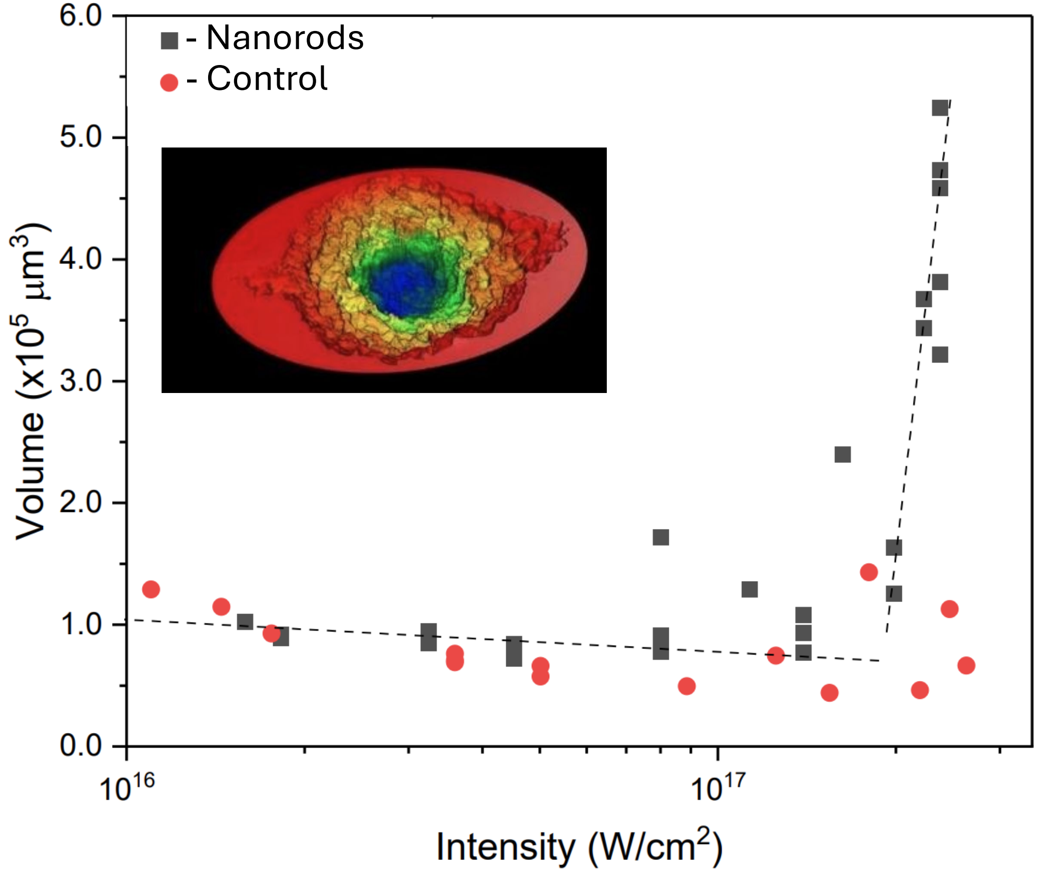
EPOCH 3D PIC Simulation of Surface plasma



Nanorods lead to increased **proton acceleration** in **EPOCH** Simulations

István Papp et al. 2023 arXiv:2306.13445v2

Wigner Experimental Results



Nanorod **doped** samples led to exponential **energy deposition** in the **target.**

Ágnes Szokol et al. arXiv:2402.18138

The NanoPlasmonic Laser Induced Fusion Energy

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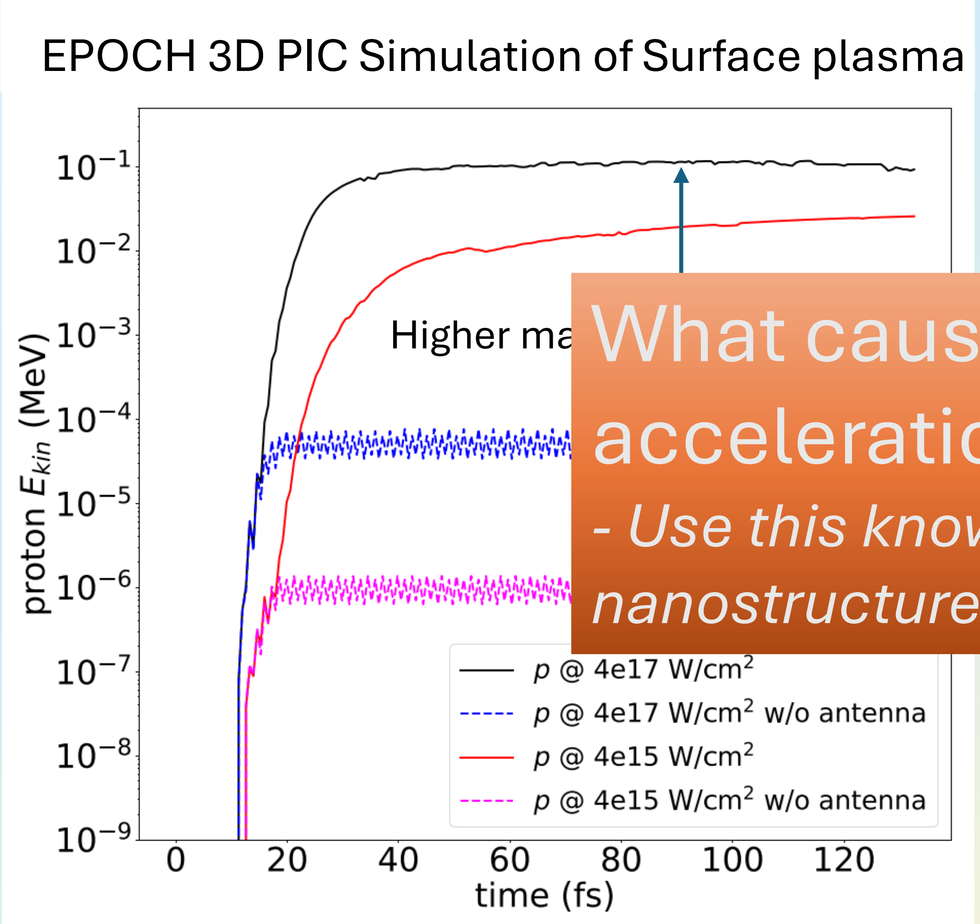

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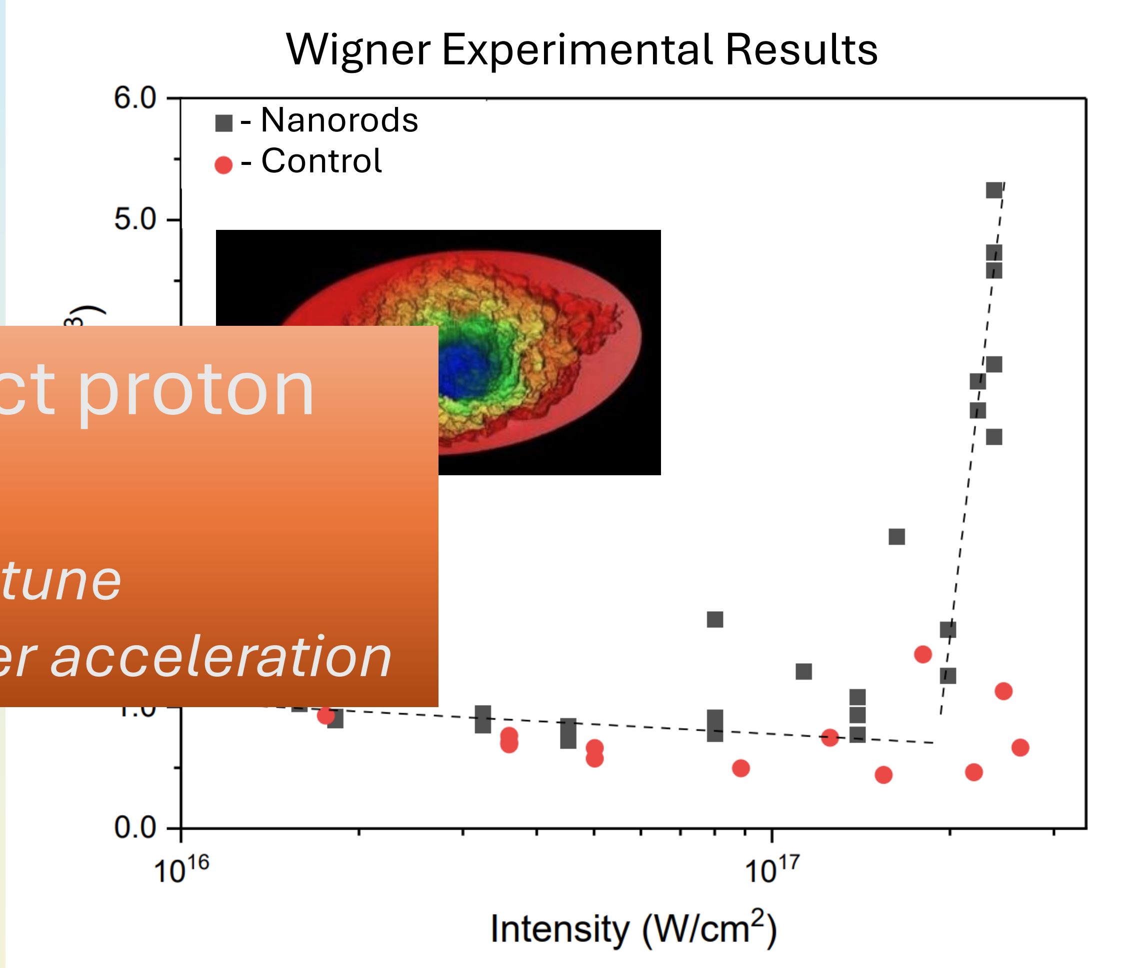

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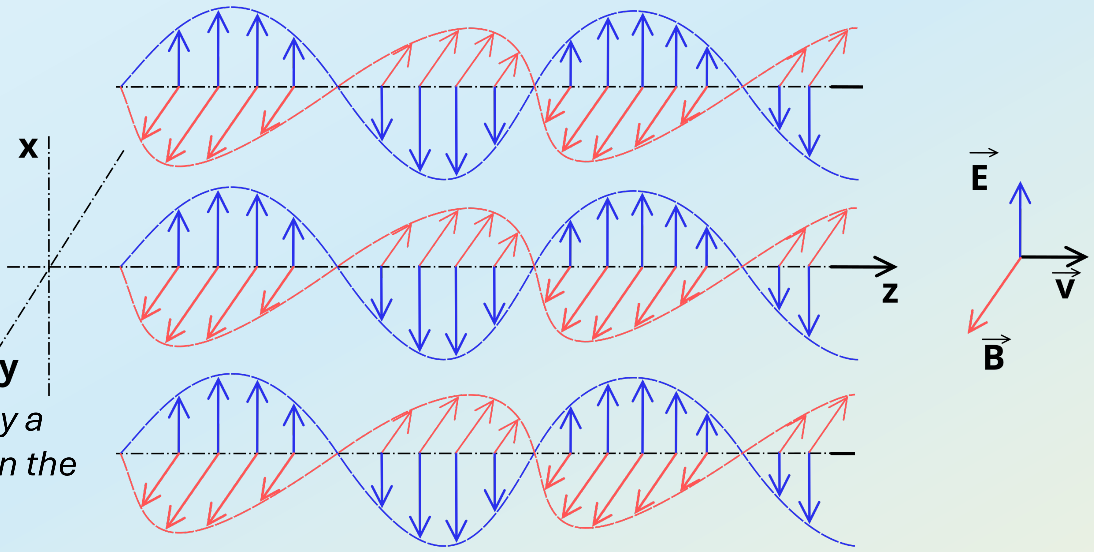
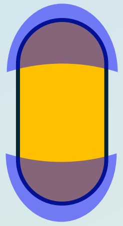
What causes direct proton acceleration?

- Use this knowledge to tune nanostructures for better acceleration

Proton Acceleration from Nanoplasmons

Laser is approximately a plane wave on the scale of the nanorod





Laser light (795 nm, I~$10^{18} W/cm^2$) impinges on Gold nanorod (~85 nm)

The acceleration of protons and ions in the **pre-plasma**, is simulated with the **EPOCH** (**Particle in Cell**) simulation software in **3D** for a single nanorod.

- *The output of **PIC** programs can tell you with accuracy what **proton energies** you will get but **not** the **mechanisms** by which they are **accelerated***

$$\boldsymbol{F}(\boldsymbol{E}, \boldsymbol{B}, Z, \lambda, I, \dots) = ?$$

Using PIC to determine this dependencies requires a huge amount of computation time and data storage.

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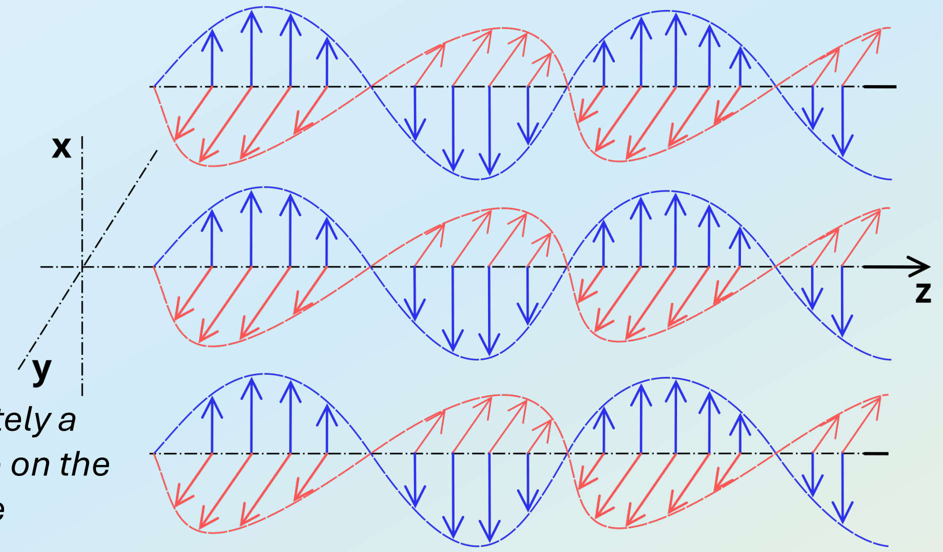
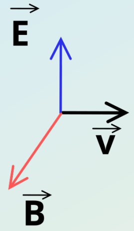
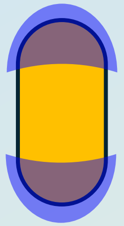


Laser is approximately a plane wave on the scale of the nanorod





Laser light (795 nm, I~$10^{18} W/cm^2$) impinges on Gold nanorod (~85 nm)

Simple theoretical calculations motivated by simulation can serve as efficient guiding principles to determine how to perform experiments and simulations.

$$\boldsymbol{F}(\boldsymbol{E},\boldsymbol{B},Z,\lambda,I,\dots) = \boxed{F_{CoulombExp} + F_{pond}} + F_{TNSA} + \cdots$$

Goal: make simplest model possible and compare to experiment

We will focus on forces that come directly from the nanorod the **ponderomotive force** and **Coulomb Explosion**

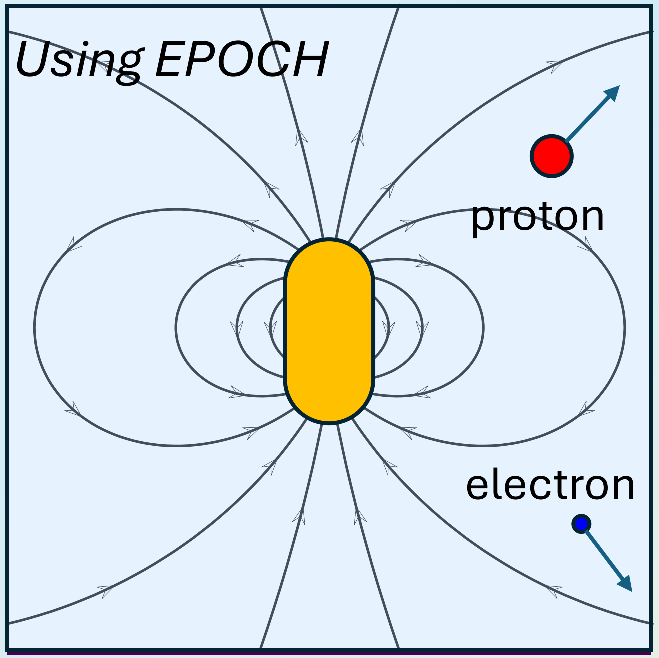
Plasmonic Ponderomotive Force

$\omega = 2.65\ fs$

$hc = 0.197$ keV nm

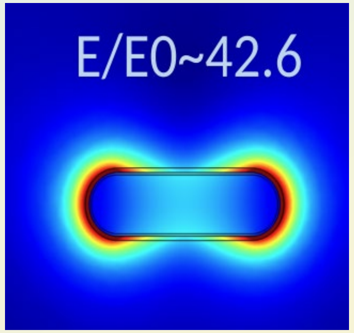
Drift force due to the **oscillating electron cloud** around the gold nanorod due to **LSP resonance** that could **directly** couple to **ions**.

3D Simulations of Single Nanorods



Typically, the **ponderomotive force** is far too **small** to cause **direct acceleration** to **protons** due to their large **mass**.

$$\boldsymbol{F_p} = -\frac{Z^2\,(hc)^2}{2\,m\,\omega^2}\nabla\langle(e\boldsymbol{E})^2\rangle_{2\,\pi/\omega}$$



However, the ponderomotive force due to the oscillating field around a **nanorod** can be much larger due to **field enhancement** and increase in the field **gradient**.

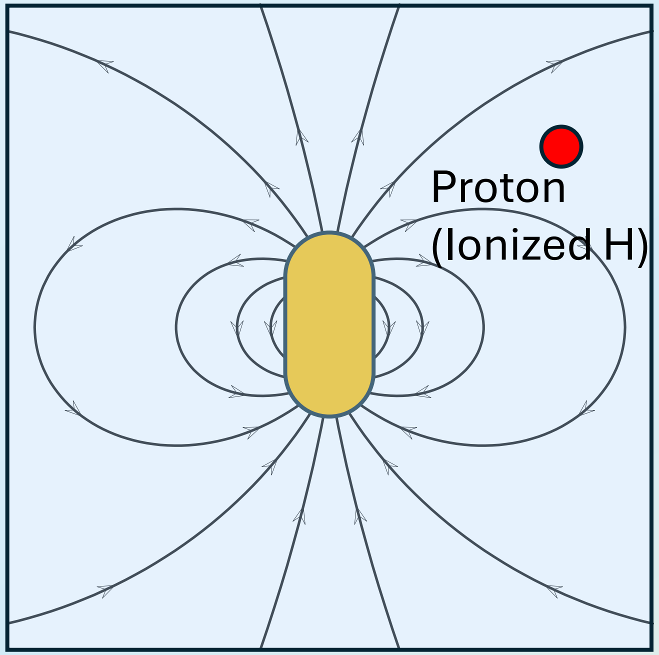
the $\boldsymbol{\nabla E^2}$ *term is ~**1600** times **larger** for the **nanorod** than the laser by field enhancement.*

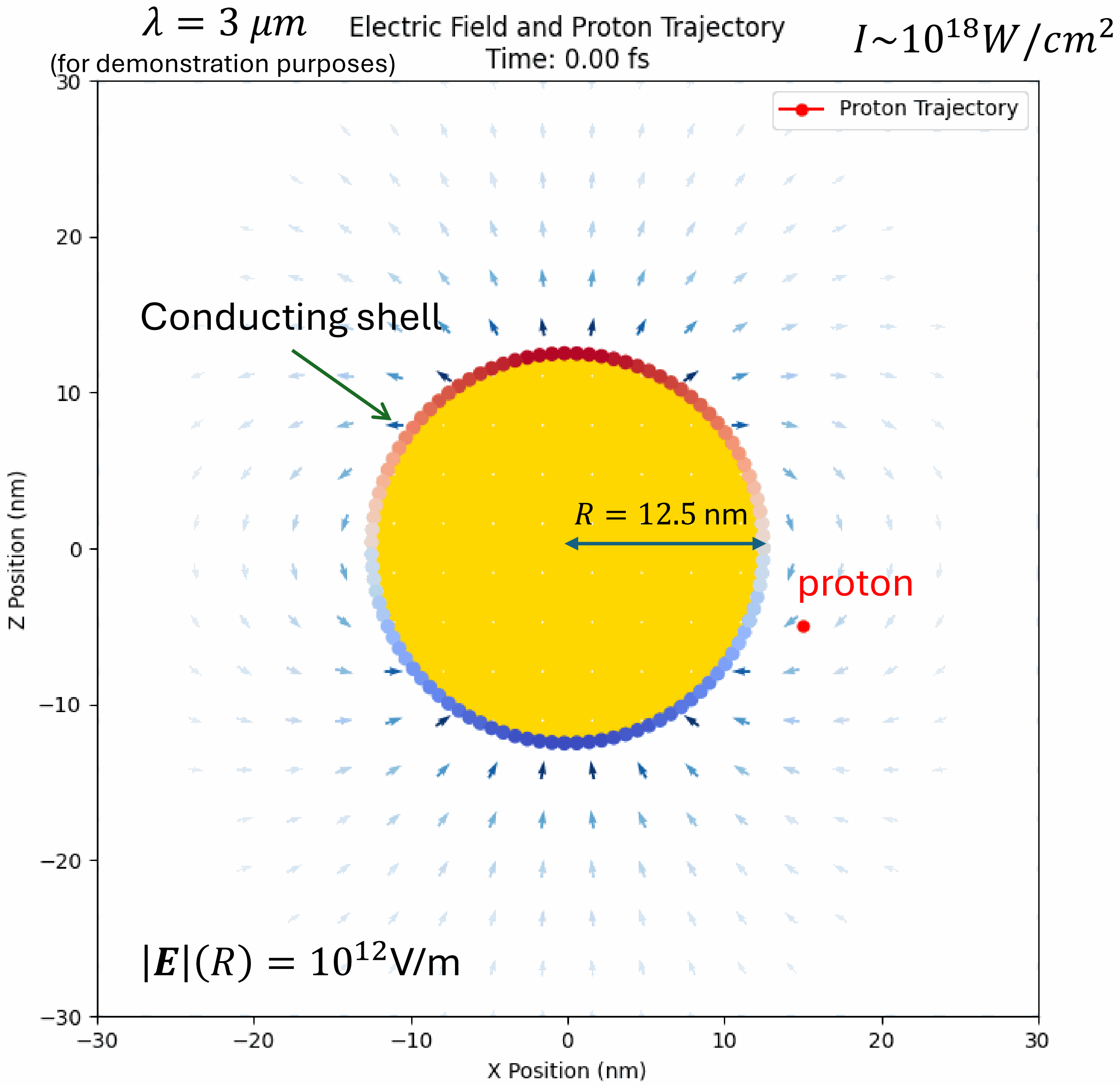
$$\mathbf{E} = \frac{1}{4\pi\varepsilon_0}\left\{\cancel{\frac{\omega^2}{c^2 r}}(\hat{\mathbf{r}}\times\mathbf{p})\times\hat{\mathbf{r}} + \left(\frac{1}{r^3} - \cancel{\frac{i\omega}{cr^2}}\right)(3\hat{\mathbf{r}}\,[\hat{\mathbf{r}}\cdot\mathbf{p}] - \mathbf{p})\right\} e^{\frac{i\omega r}{c}} e^{-i\omega t} \Rightarrow \mathrm{U_p} = \frac{Z^2\alpha^2 hc^4}{4m\omega^2}\left(\frac{p^2r^2 + 3(\mathbf{r}\cdot\mathbf{p})^2}{r^8}\right)$$

Near field approx.

Increase in the ponderomotive force leads to **more** efficient **electron** and **ion acceleration**

Toy Model – Conducting Sphere

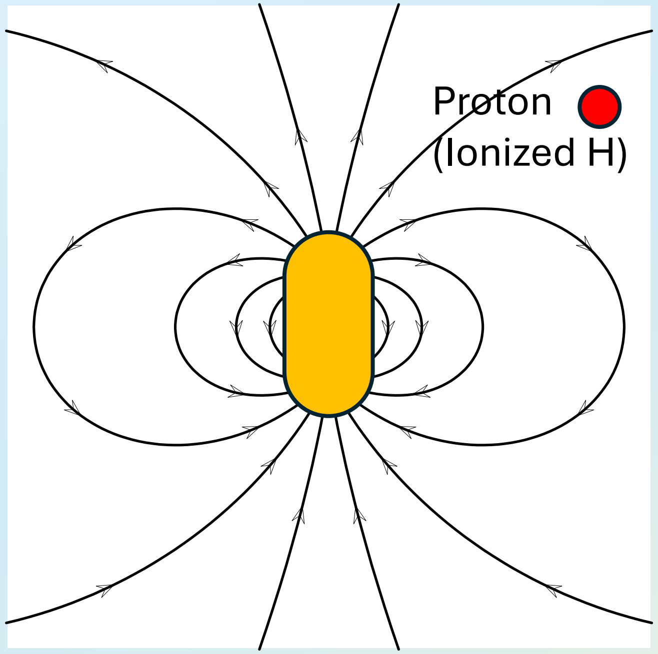


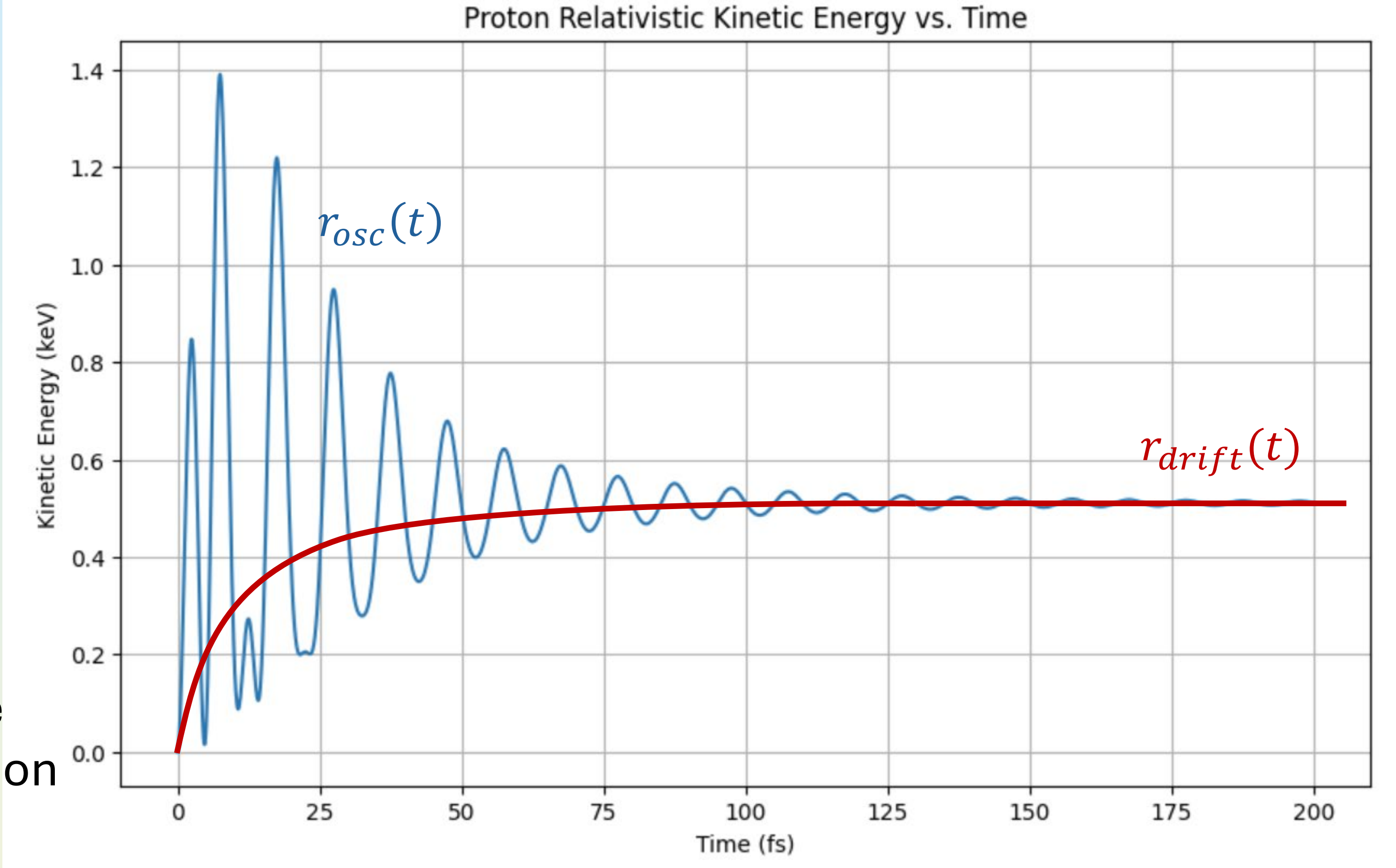


Dipole moment $p = Z_e\ R$ decide the field strength outside the nanorod

Z_e $- number\ of\ electrons \approx 10^6$

Toy Model – Conducting Sphere





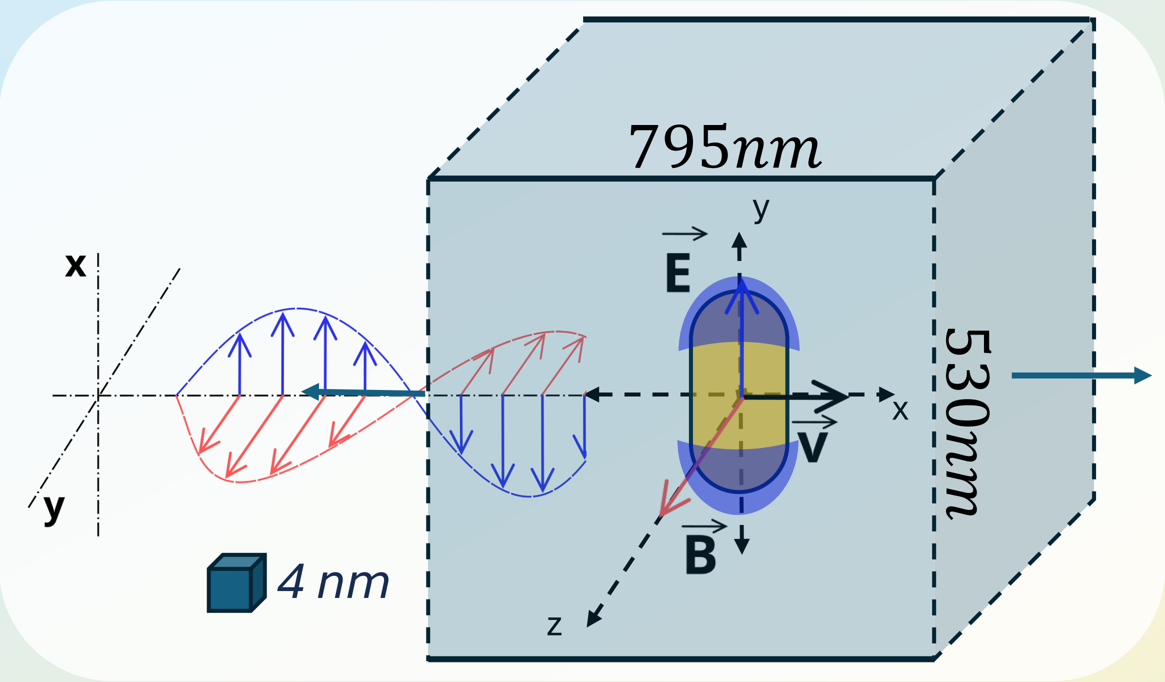
If the laser frequency is much larger than the characteristic timescale of proton acceleration

$$\omega \gg \frac{qE}{mc} \implies r(t) = r_{osc}(t) + r_{drift}(t)$$

EPOCH Simulations on GPU Cluster

The **3D nanorod** simulation study's the **plasmonic** effects of a **single** nanorod (*made partially by me and the previous postdoc István Papp*)

Simple model contains a **gold** cylinder with spherical end caps (85x25 nm) at solid density $5.9 \times 10^{22}\ cm^{-3}$ with charge state Au^{3+} covered with a neutralizing cloud of "**conducting**" electrons. The nanorod is surrounded by an **ionized** medium of **carbon** and **hydrogen**. The nanorod is then illuminated with $\lambda = 795\ nm$ light with some pulse duration Δt_{pulse} with open boundary conditions for fields and particles run for 200 fs.

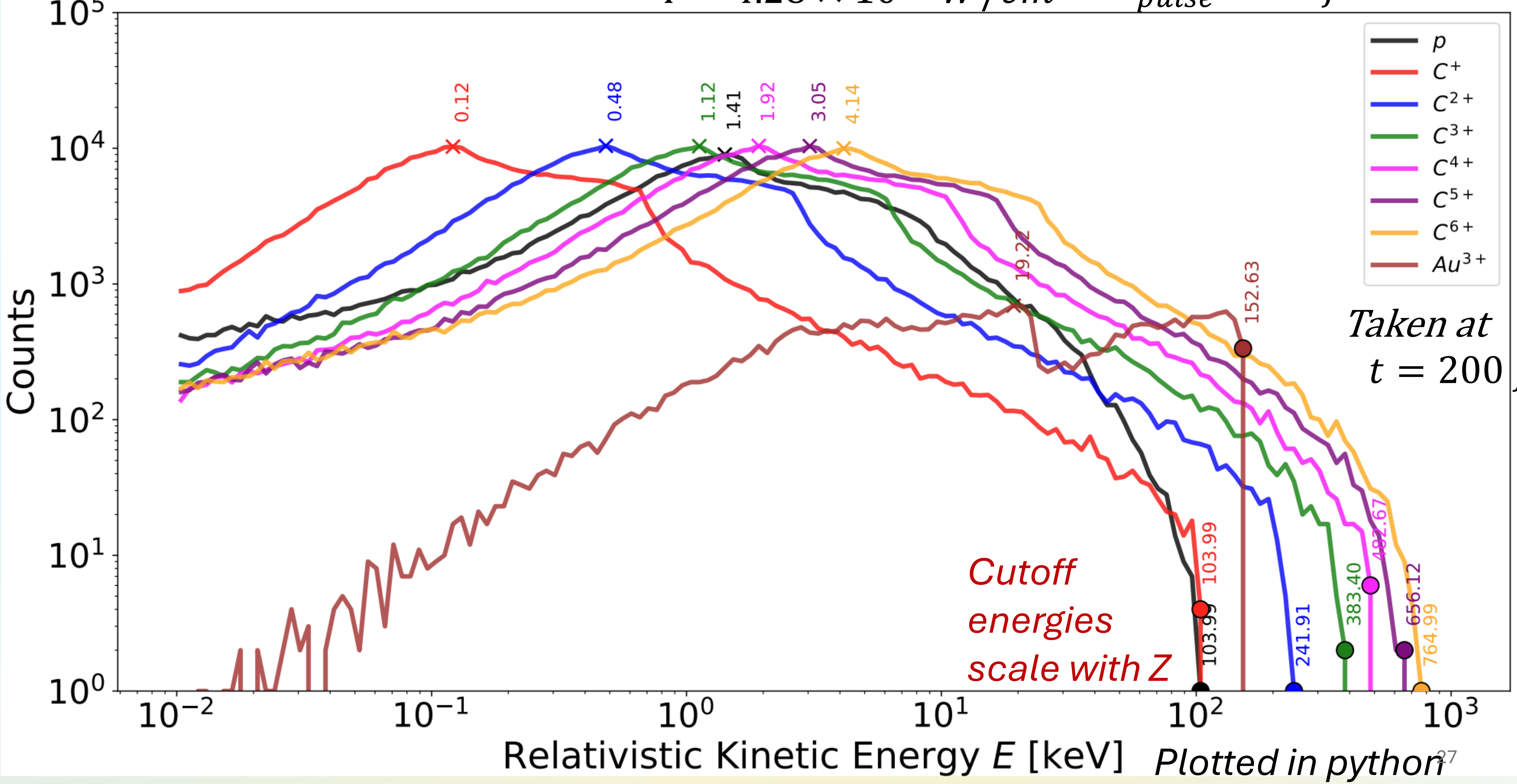


Focus on analyzing **evolution** of:

- Electric field
- Charge density
- Number density
- Particle energies
- <u>**Ion Energy spectra**</u> **–** observable via Thompson parabola in experiment.

Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\ W/cm^2 \quad \Delta t_{pulse} = 21\ fs$

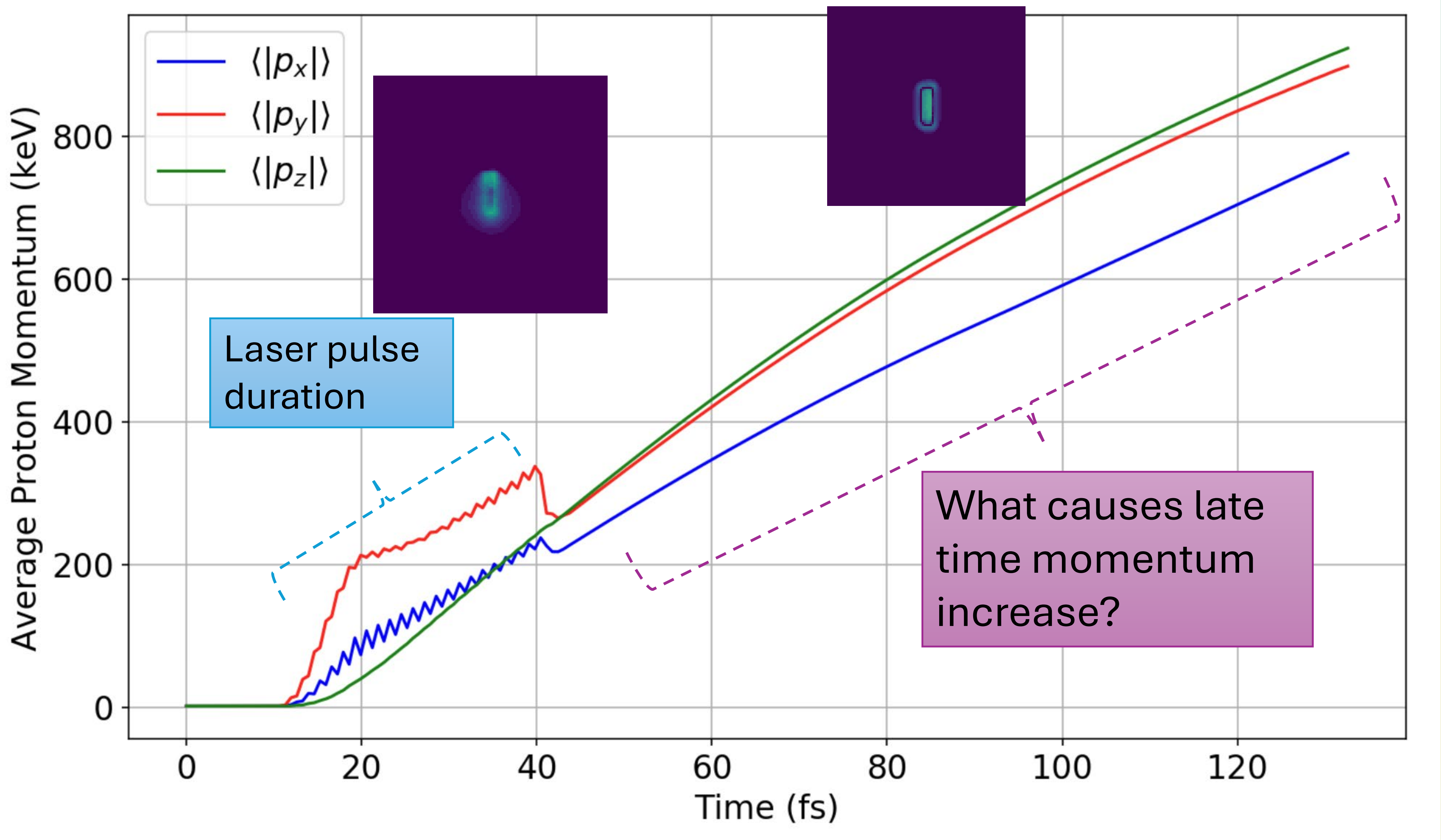


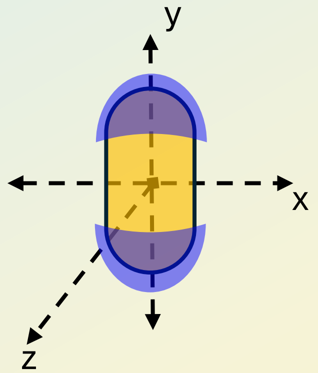
Taken at $t = 200\ fs$

Plotted in python

Proton Acceleration

$I = 4.28 \times 10^{18}\ W/cm^2 \quad \Delta t_{pulse} = 21\ fs$





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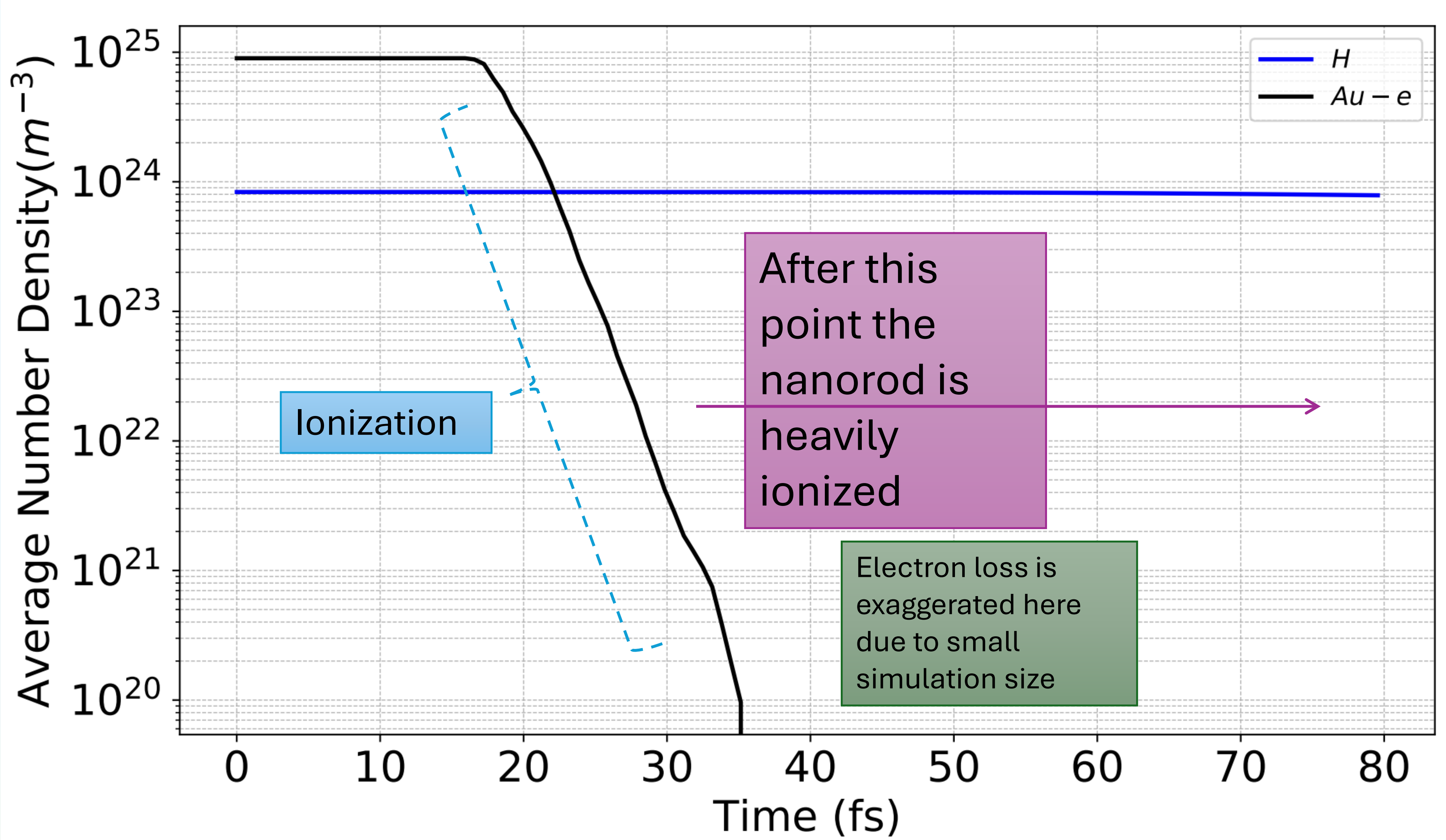

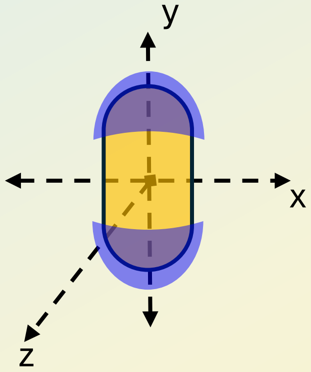

Ionization limit was set to Au^{3+}, but ionization can go up to Au^{20+}

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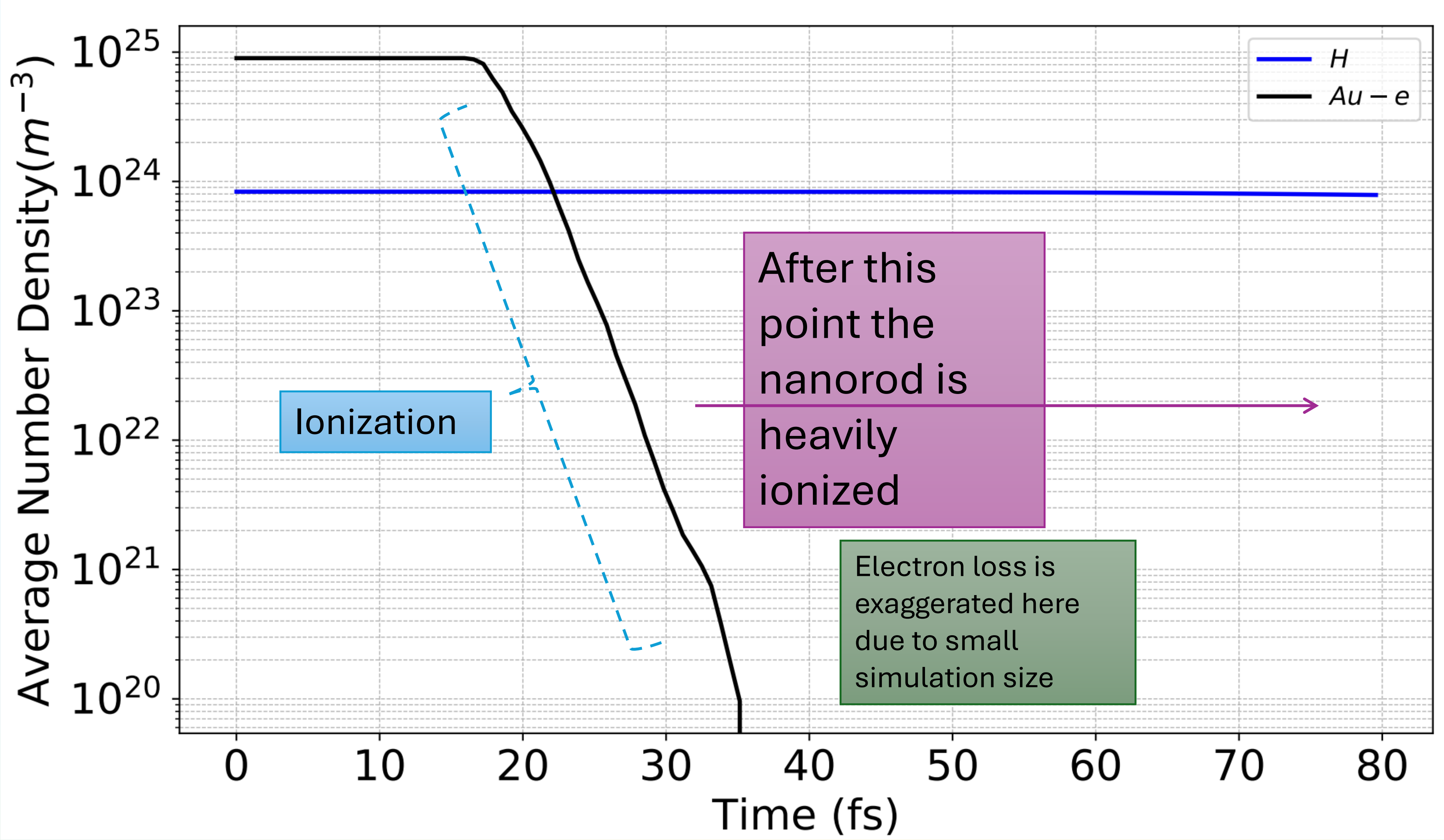
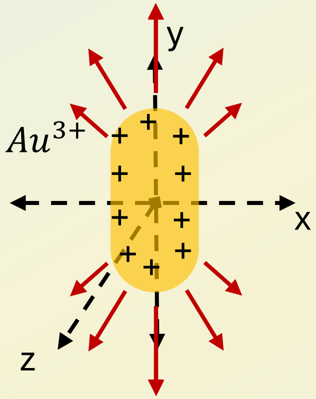


Highly **ionized** gold nanorod causes a **Coulomb explosion** in the local material.



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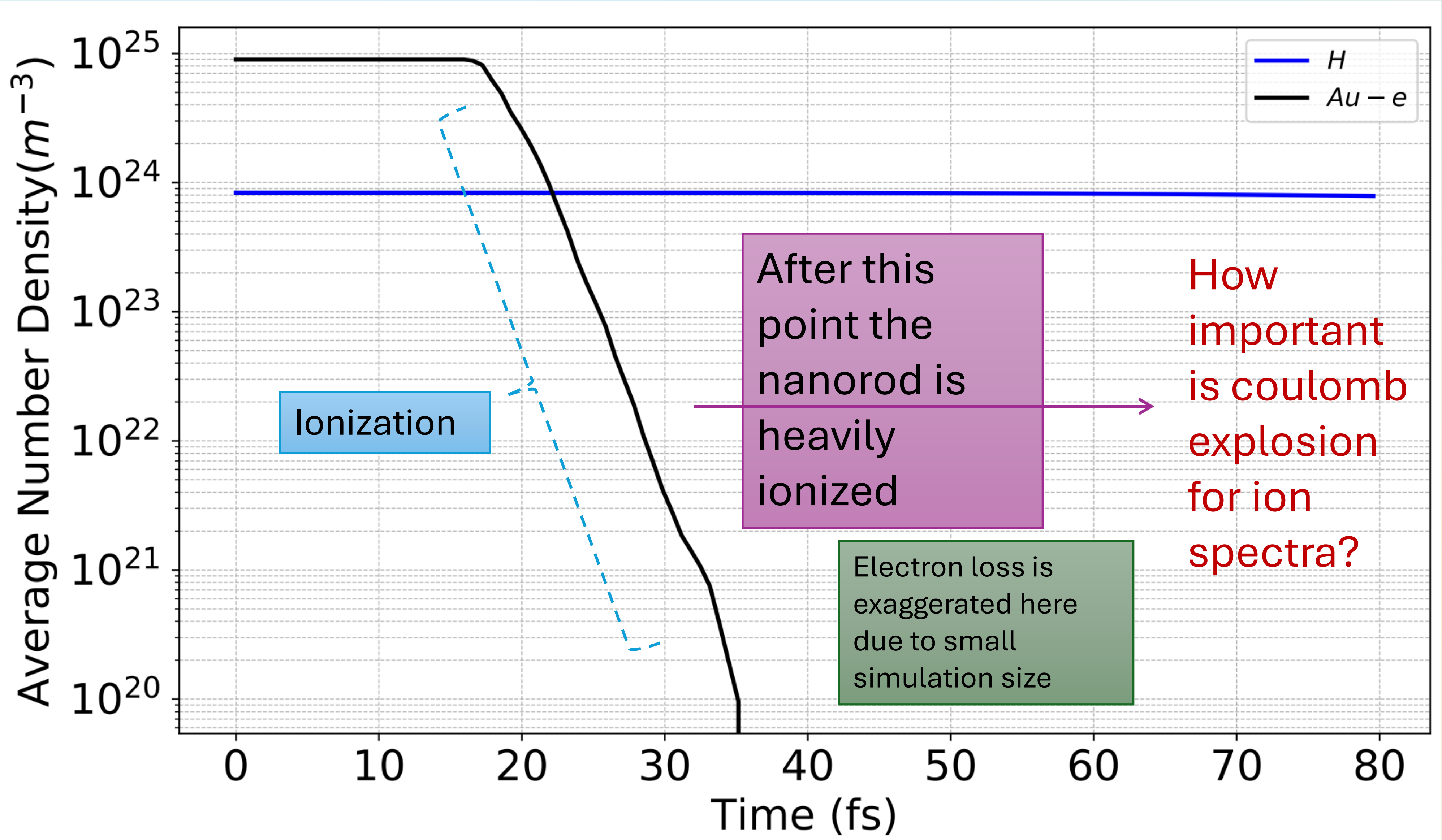

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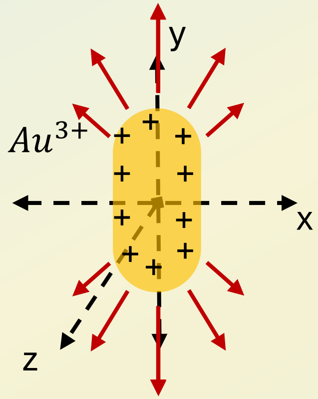

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Ion spectra –Coulomb Explosion

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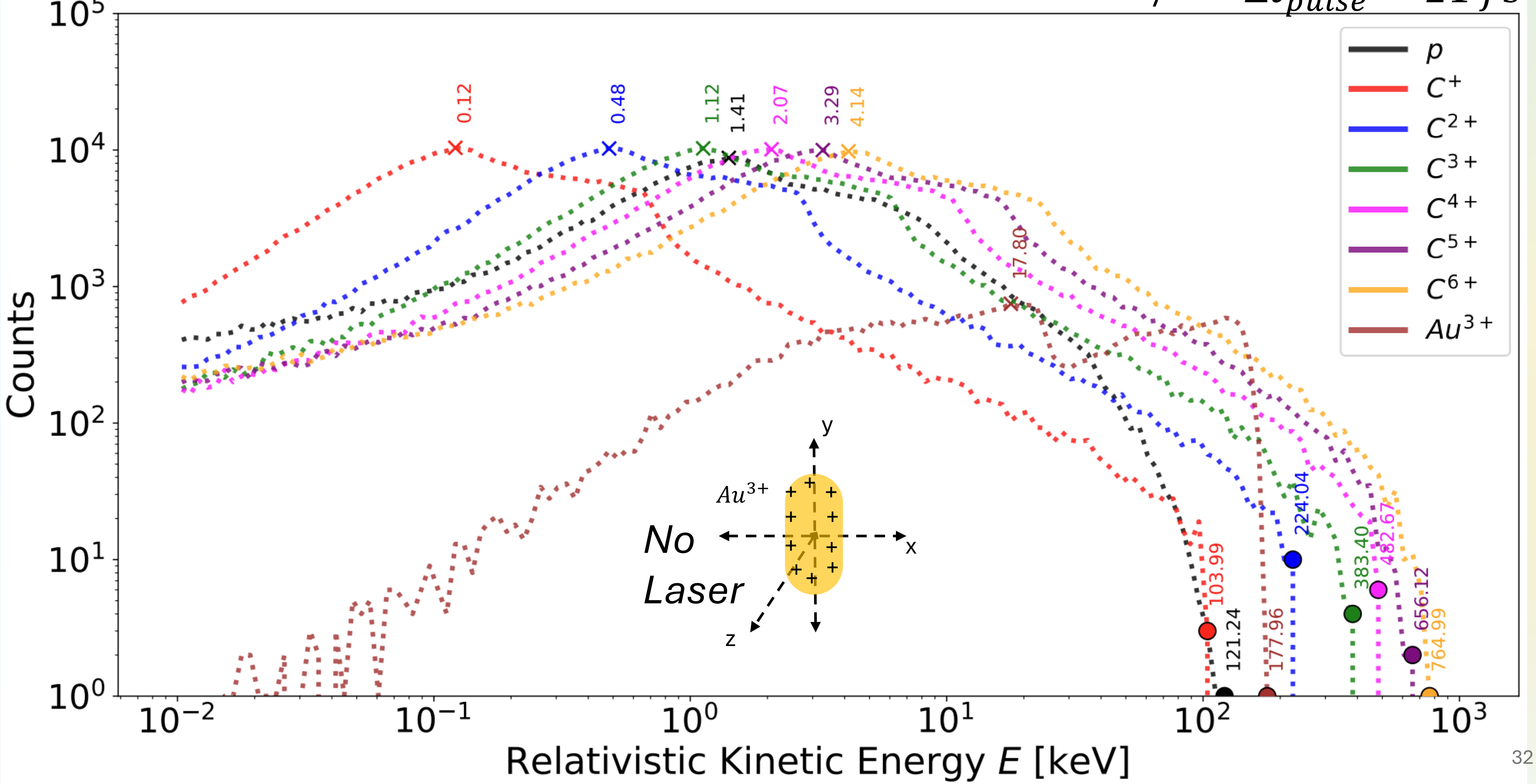
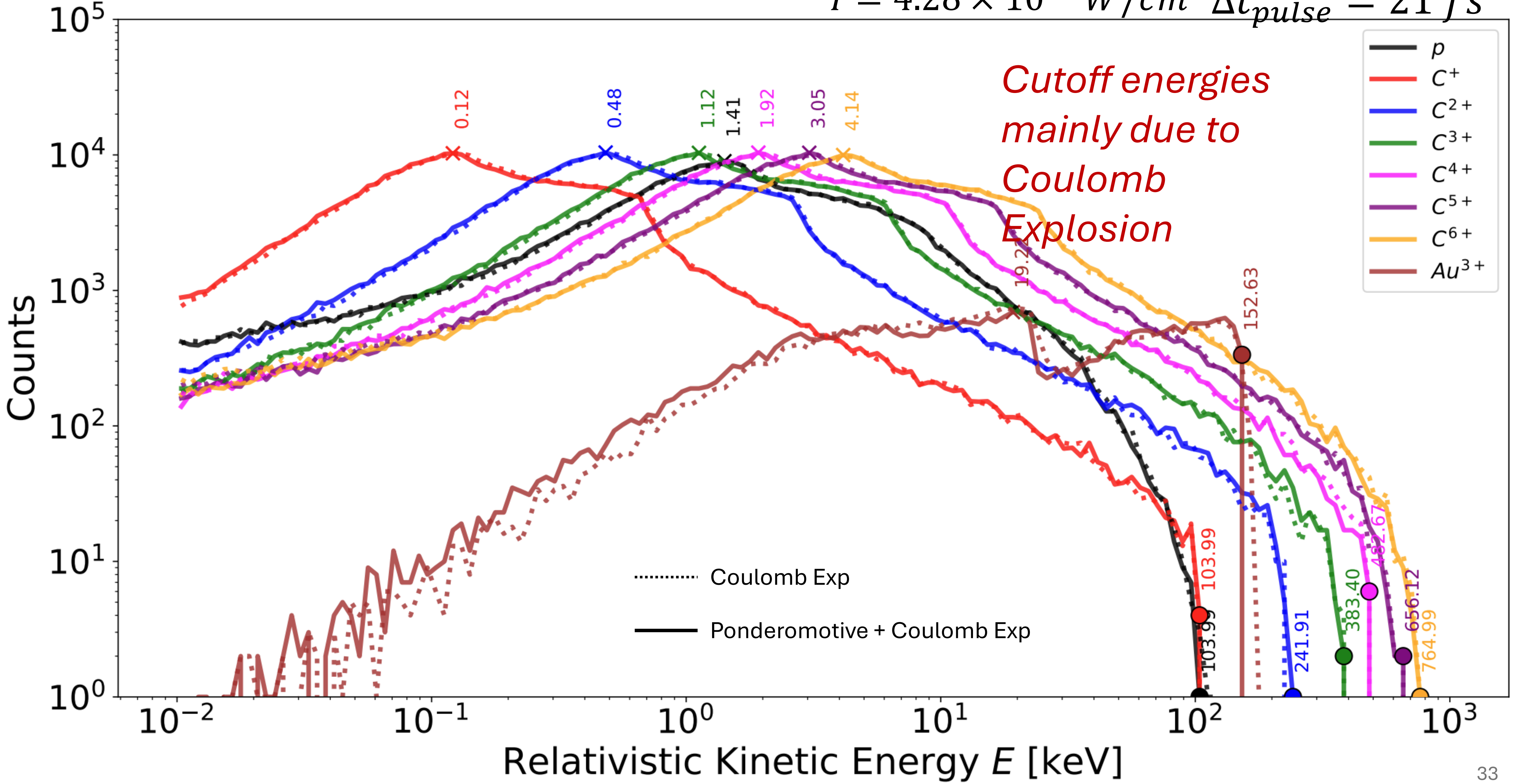


Ion spectra - Comparison

$I = 4.28 \times 10^{18}\ W/cm^2\ \Delta t_{pulse} = 21\ fs$



Cutoff energies mainly due to Coulomb Explosion

Ion spectra - Comparison

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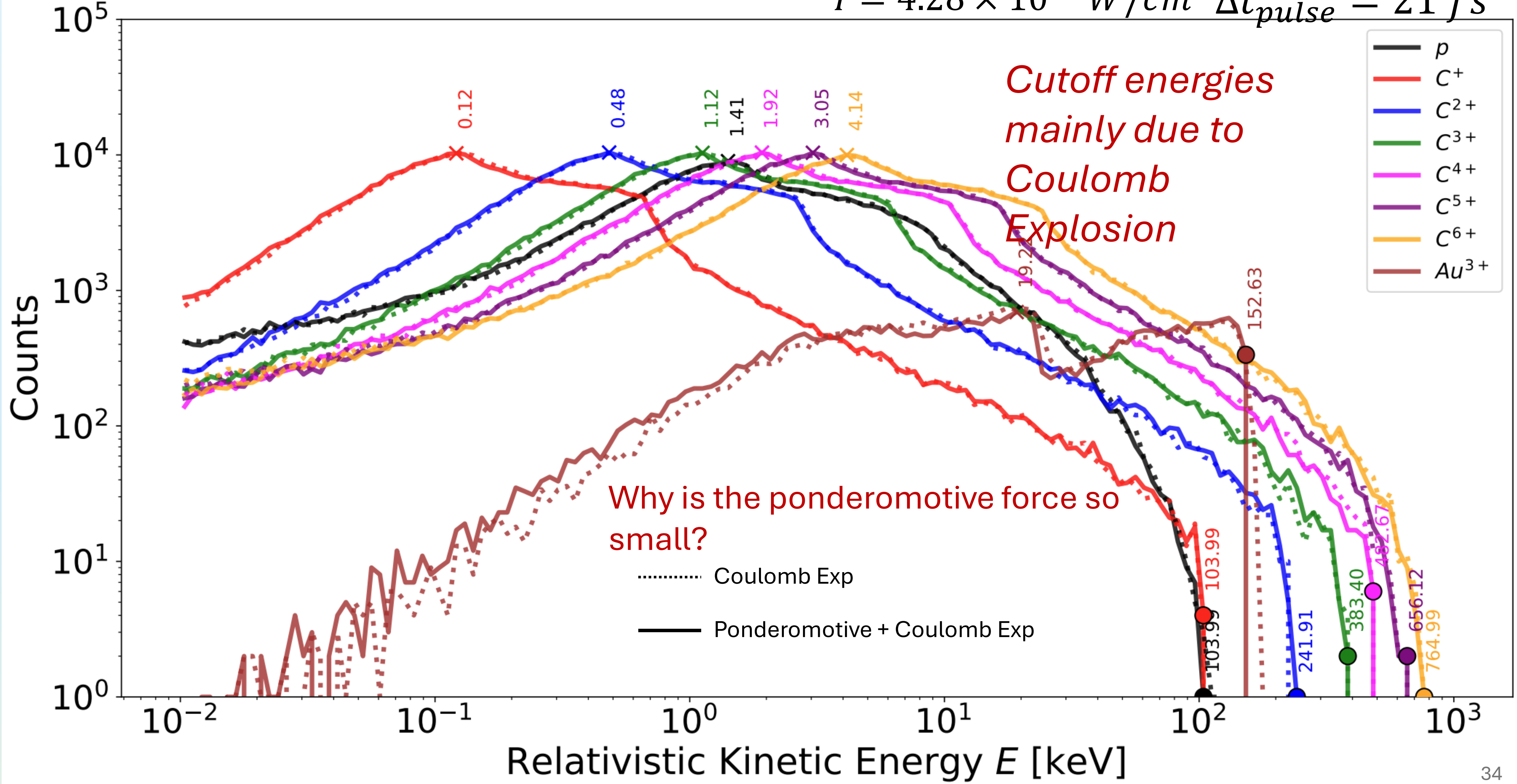
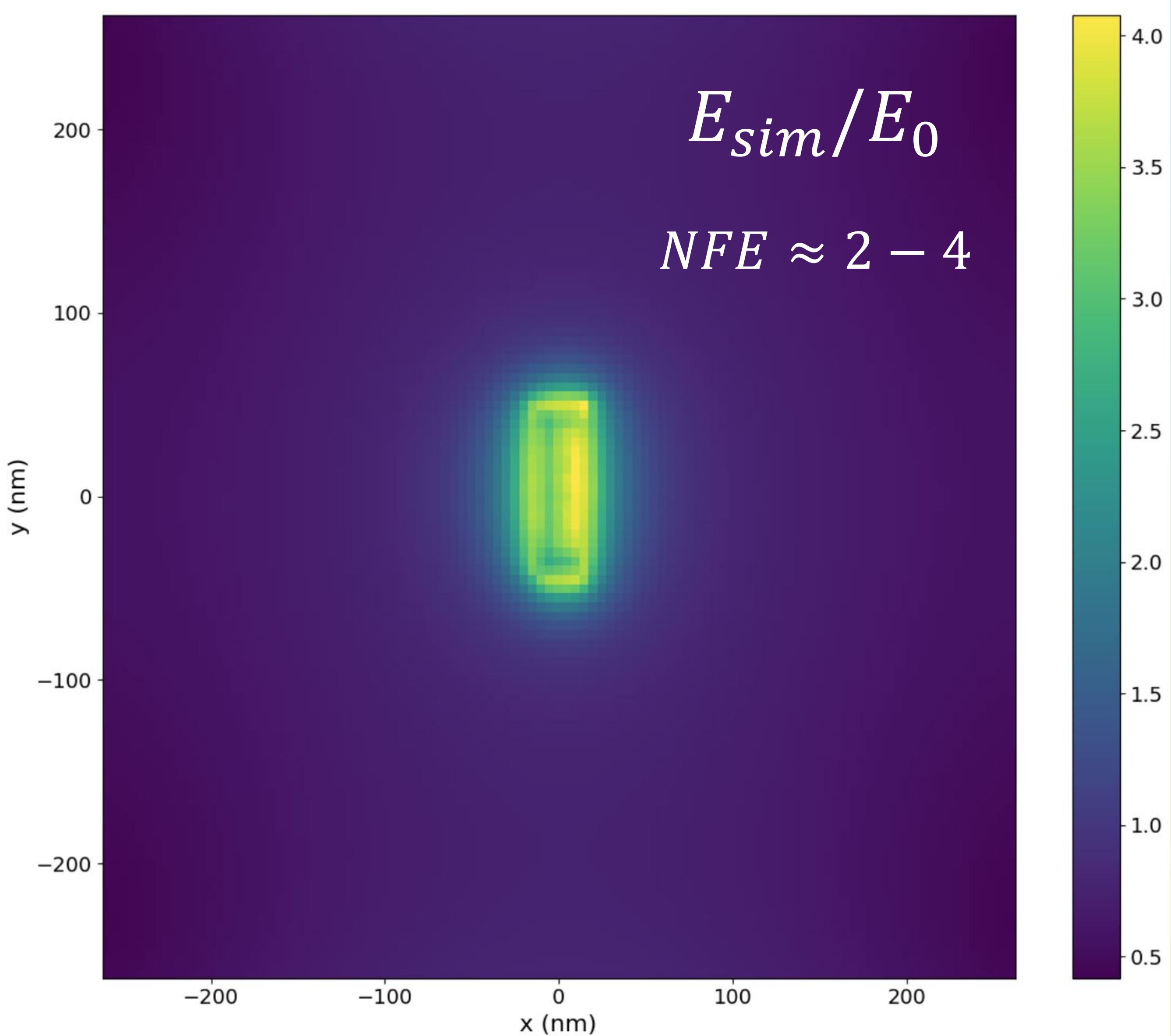


Cutoff energies mainly due to Coulomb Explosion

Why is the ponderomotive force so small?

Why is the Ponderomotive effect on ions so small?



$$E_0 = \sqrt{\frac{2\,I}{\varepsilon\,c}}$$

$$I = 4 \times 10^{18}\ W/cm^2$$

$$\varepsilon = \varepsilon_r \varepsilon_0 = 2.25\varepsilon_0$$

similar results found in a previous publication

$$I_x = 0.3\, I_p \frac{S_{\mathrm{CB}}}{S_{\mathrm{NR}}} = 25.9\, I_p$$

I. Papp (2022)

$$NFE = \sqrt{25.9} \approx 5$$

EPOCH predicts very **small near field** enhancement (NFE) in comparison to theory.

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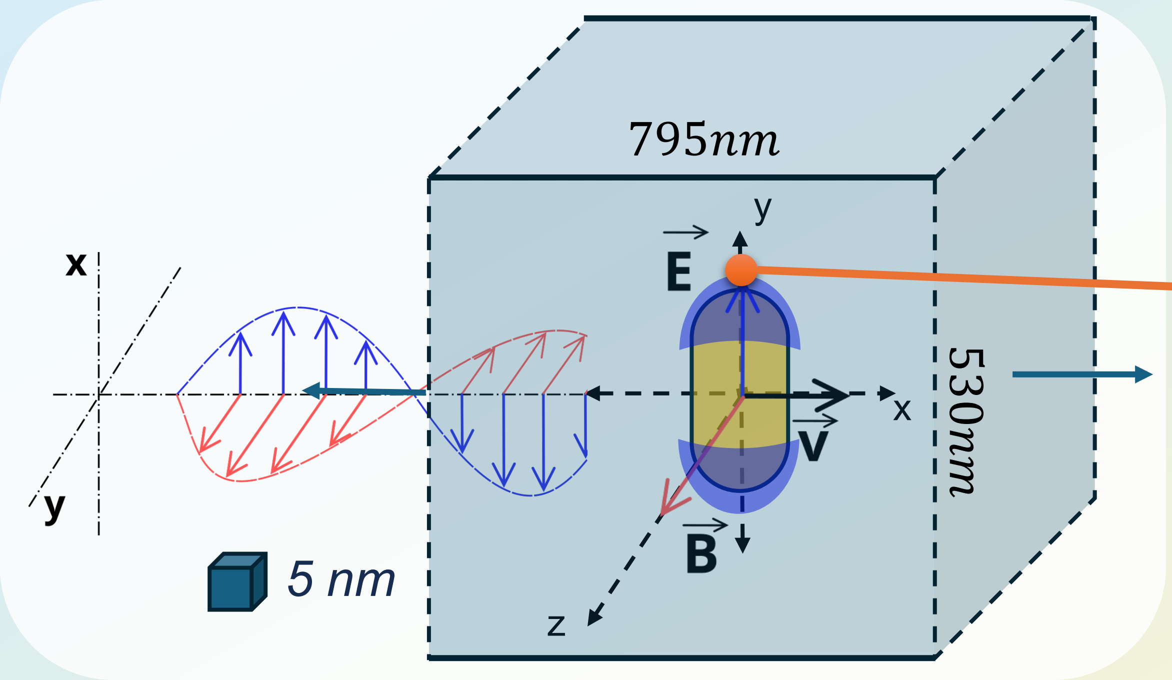
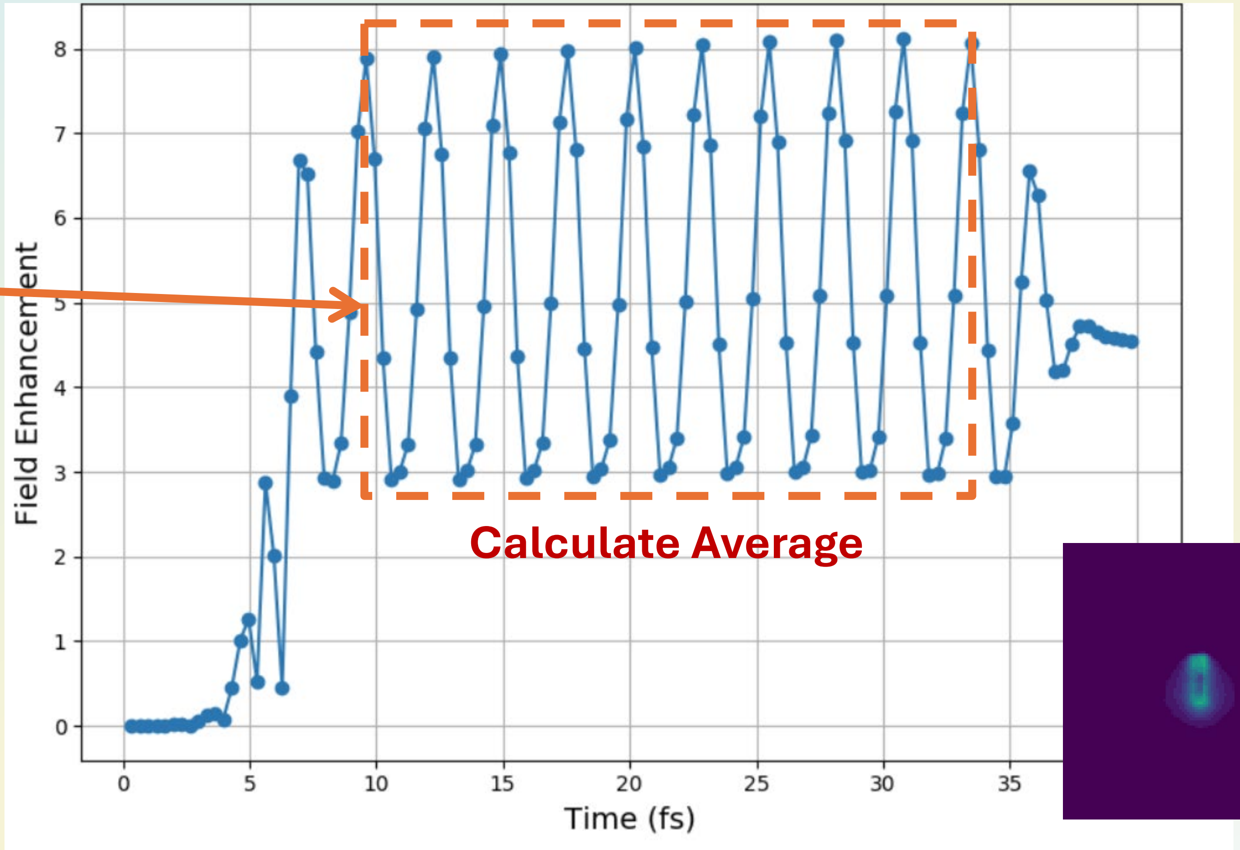
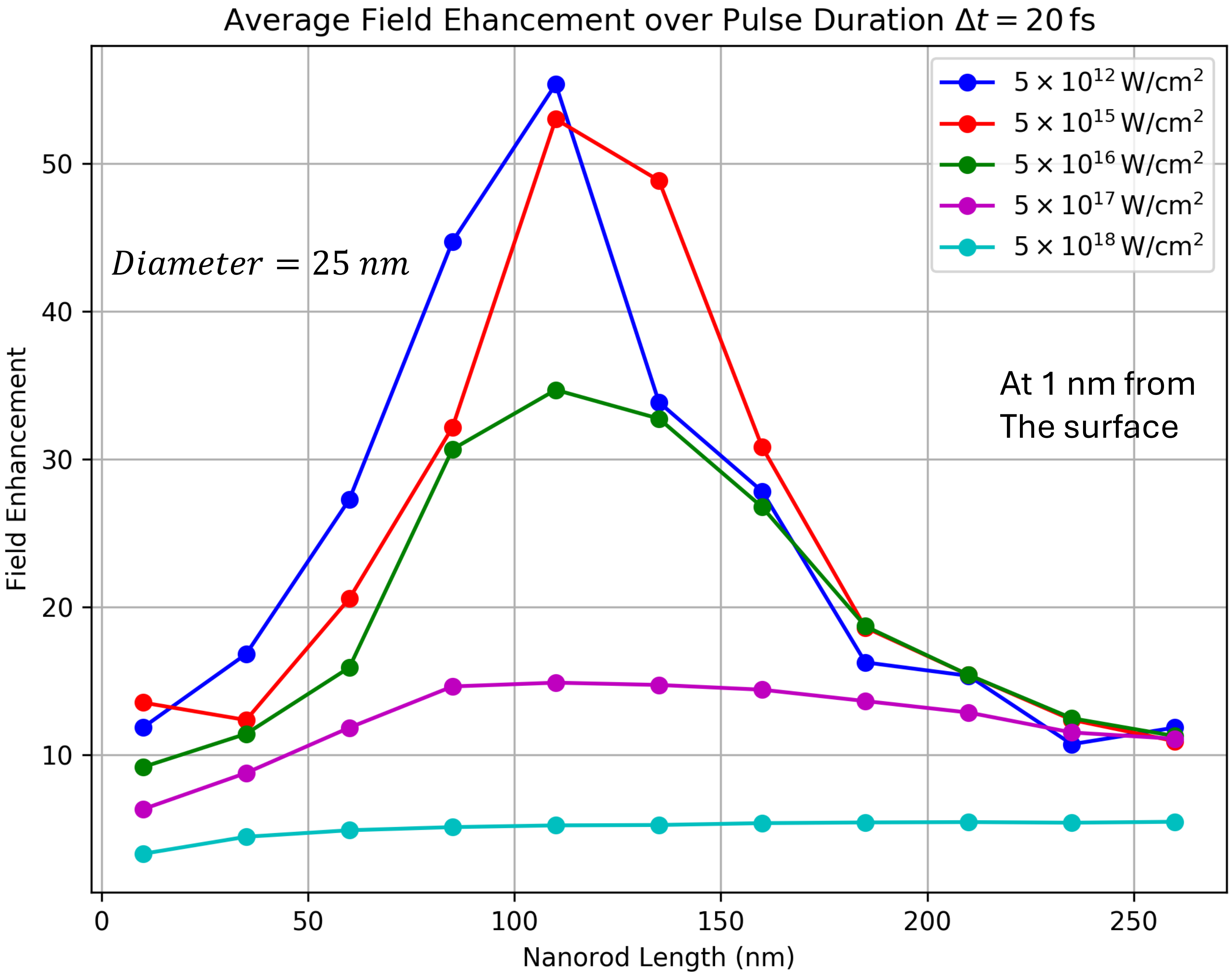
The **3D nanorod** simulation study's the **plasmonic** effects of a **single** nanorod

gold cylinder with spherical end caps (85x25 nm) at solid density $5.9 \times 10^{22}\ cm^{-3}$ with charge state Au^{3+} covered with a neutralizing cloud of "**conducting**" electrons. The nanorod is surrounded by an **ionized** medium of **hydrogen**. The nanorod is then illuminated with $\lambda = 795\ nm$ light with some pulse duration Δt_{pulse} with open boundary conditions for fields and particles run for 200 fs.

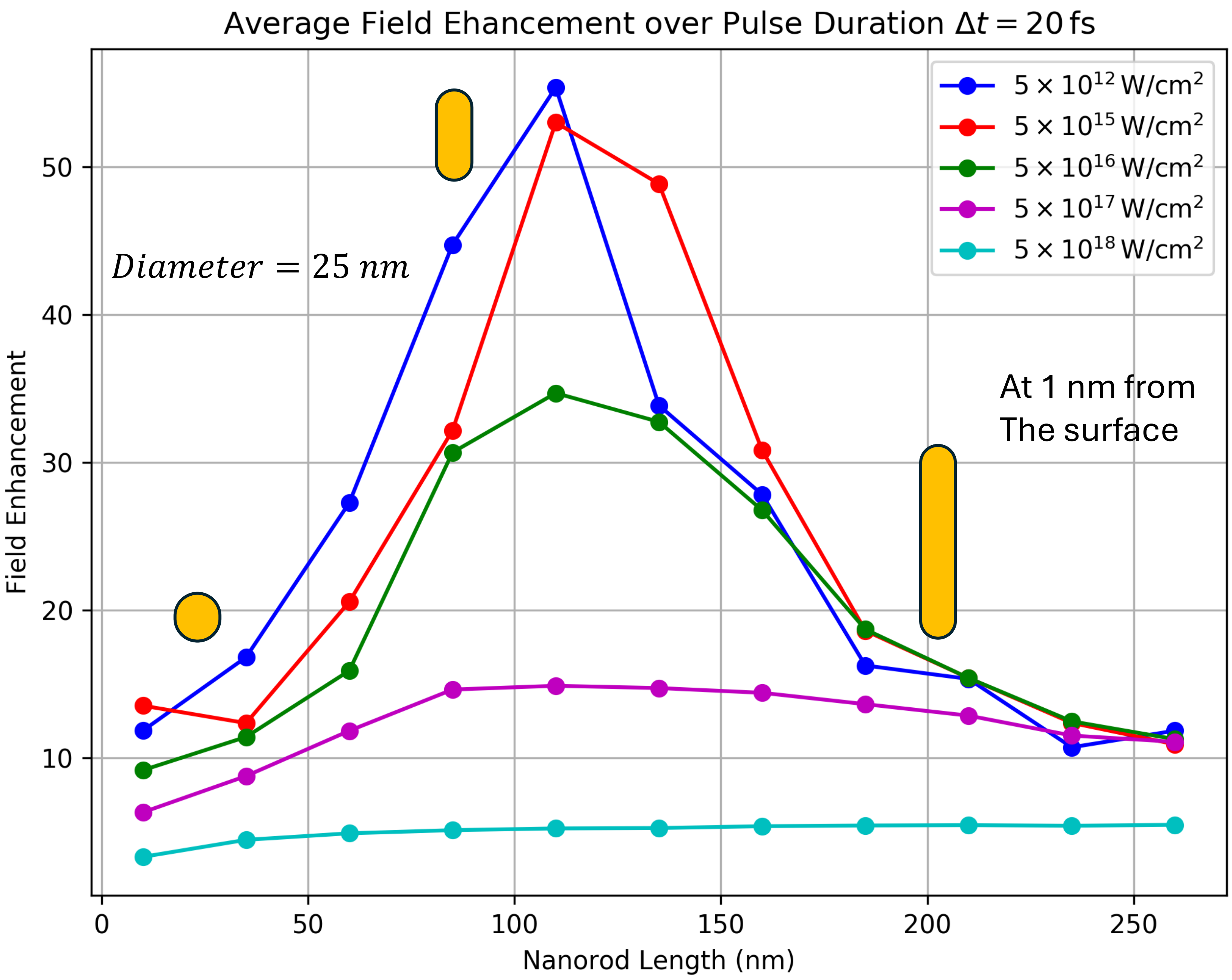


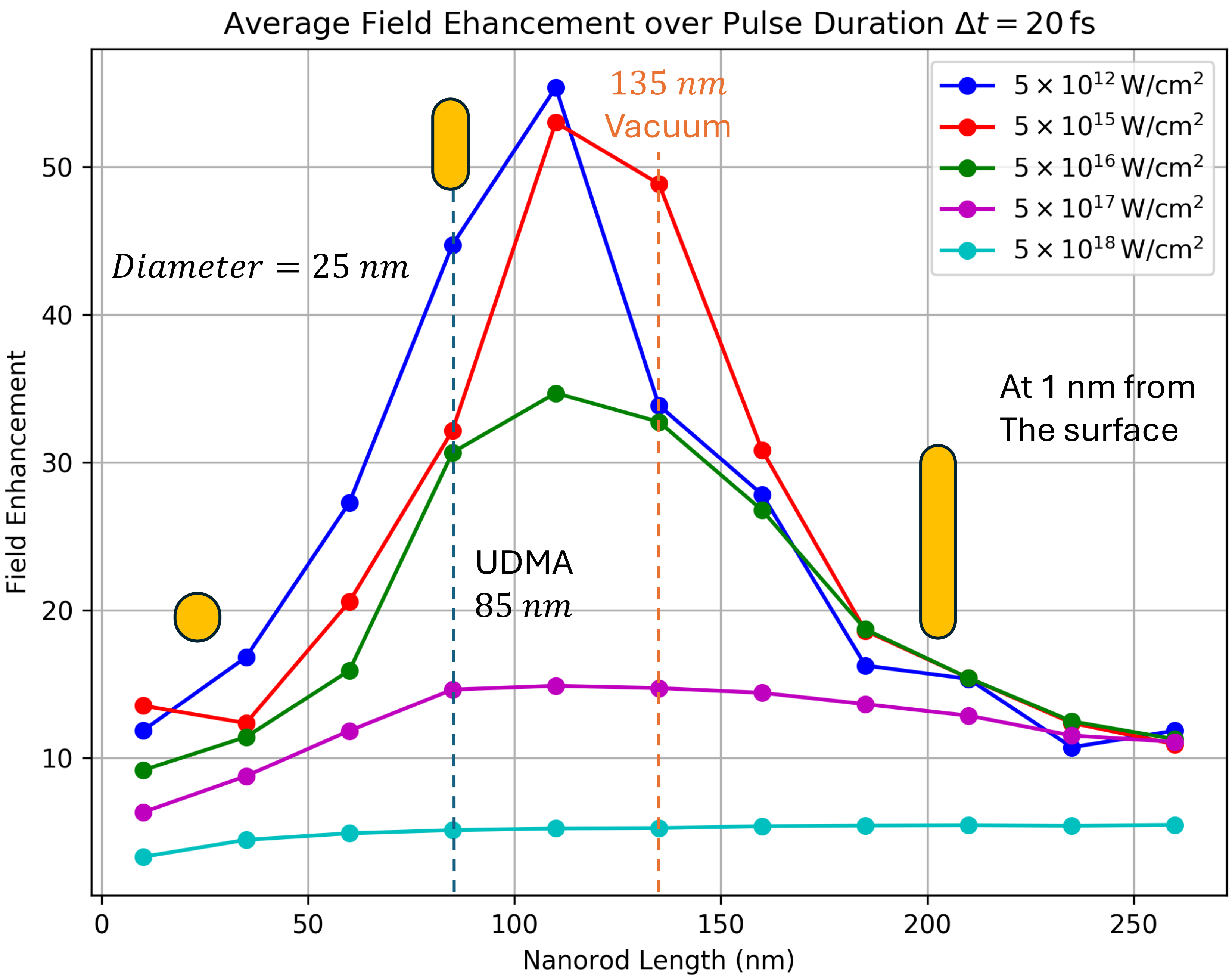
Measure the Near Field Enhancement Predicted by Epoch





55 simulations
2,200 fs
11hrs – Wall hours
704 – CPU hours


Average Field Ehancement over Pulse Duration $\Delta t = 20\,\mathrm{fs}$
$5 \times 10^{12}\,\mathrm{W/cm^2}$
$5 \times 10^{15}\,\mathrm{W/cm^2}$
$5 \times 10^{16}\,\mathrm{W/cm^2}$
$5 \times 10^{17}\,\mathrm{W/cm^2}$
$5 \times 10^{18}\,\mathrm{W/cm^2}$
$Diameter = 25\ nm$
At 1 nm from
The surface
Field Enhancement
Nanorod Length (nm)
0
50
100
150
200
250
10
20
30
40
50



Average Field Ehancement over Pulse Duration $\Delta t = 20\,\mathrm{fs}$
$135\ nm$
Vacuum
$Diameter = 25\ nm$
UDMA
$85\ nm$
At 1 nm from
The surface
$5 \times 10^{12}\,\mathrm{W/cm^2}$
$5 \times 10^{15}\,\mathrm{W/cm^2}$
$5 \times 10^{16}\,\mathrm{W/cm^2}$
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Field Enhancement
Nanorod Length (nm)
10
20
30
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50
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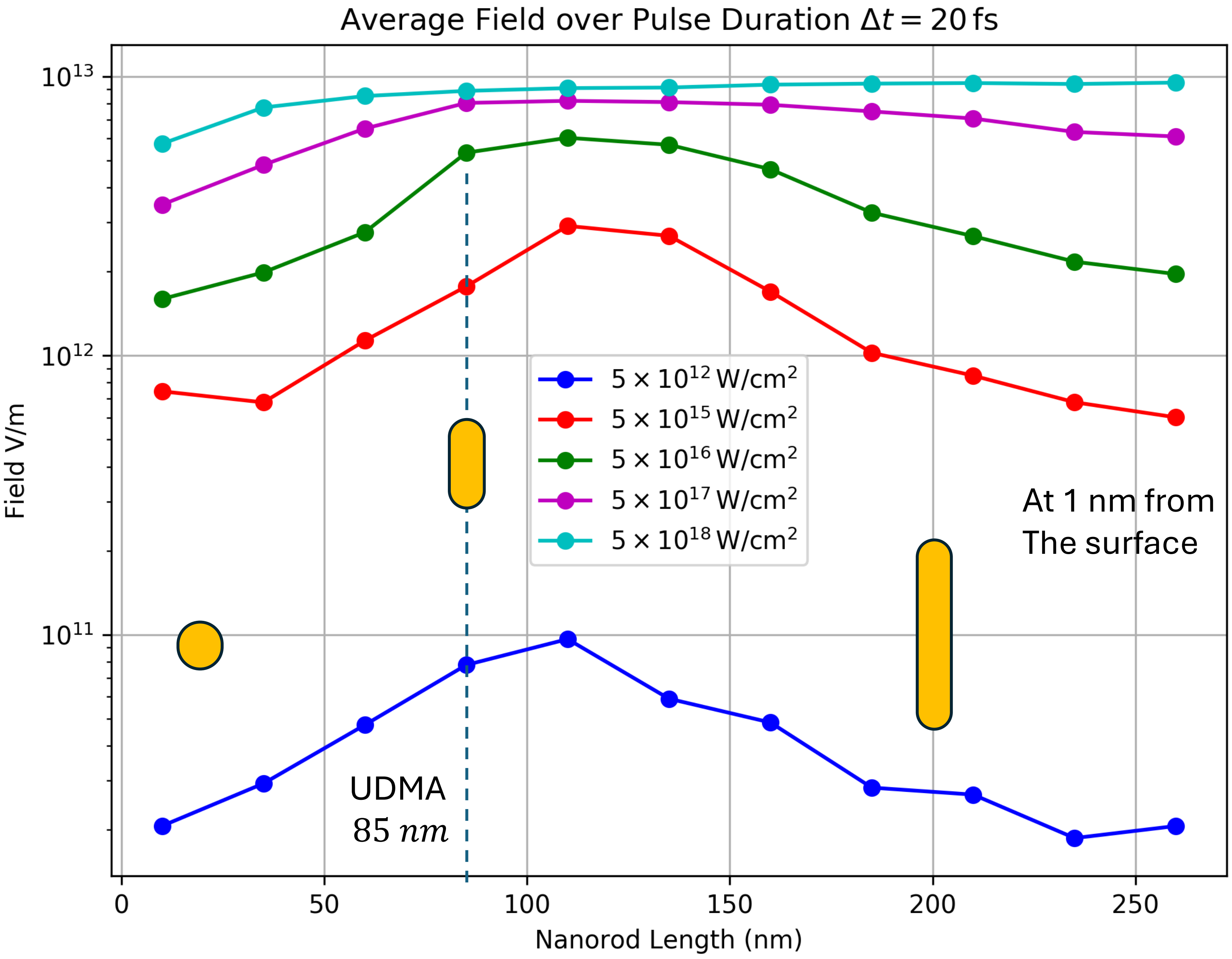
Average Field over Pulse Duration Δt = 20 fs
10^{13}
10^{12}
10^{11}
Field V/m
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50
100
150
200
250
Nanorod Length (nm)
5×10^{12} W/cm^2
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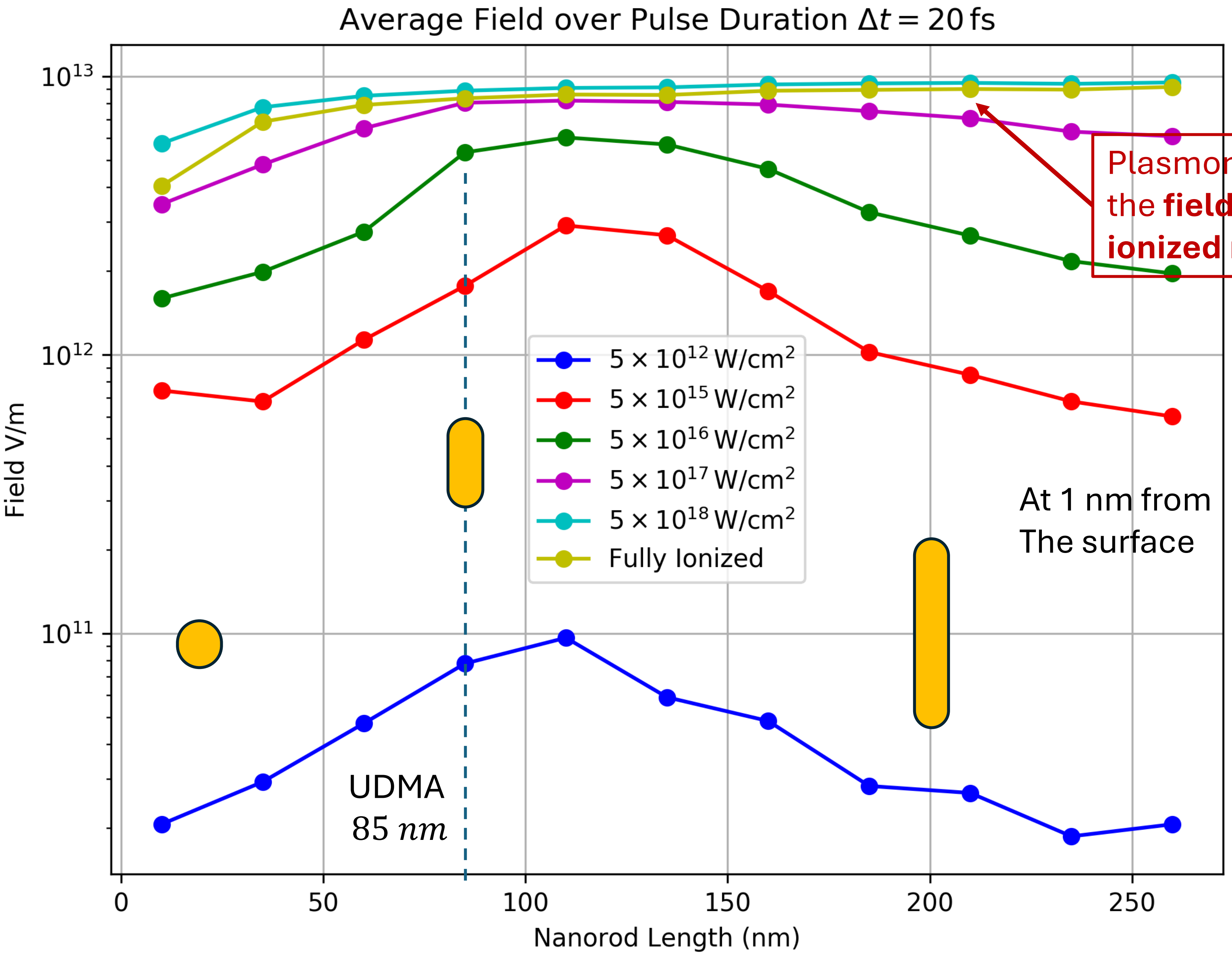
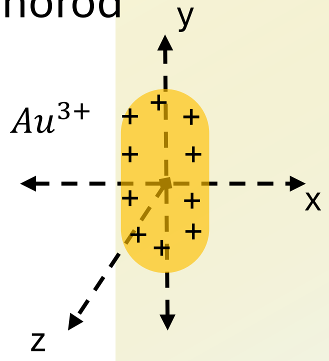


Plasmonic field **saturates** at the **field** strength of a fully **ionized nanorod**

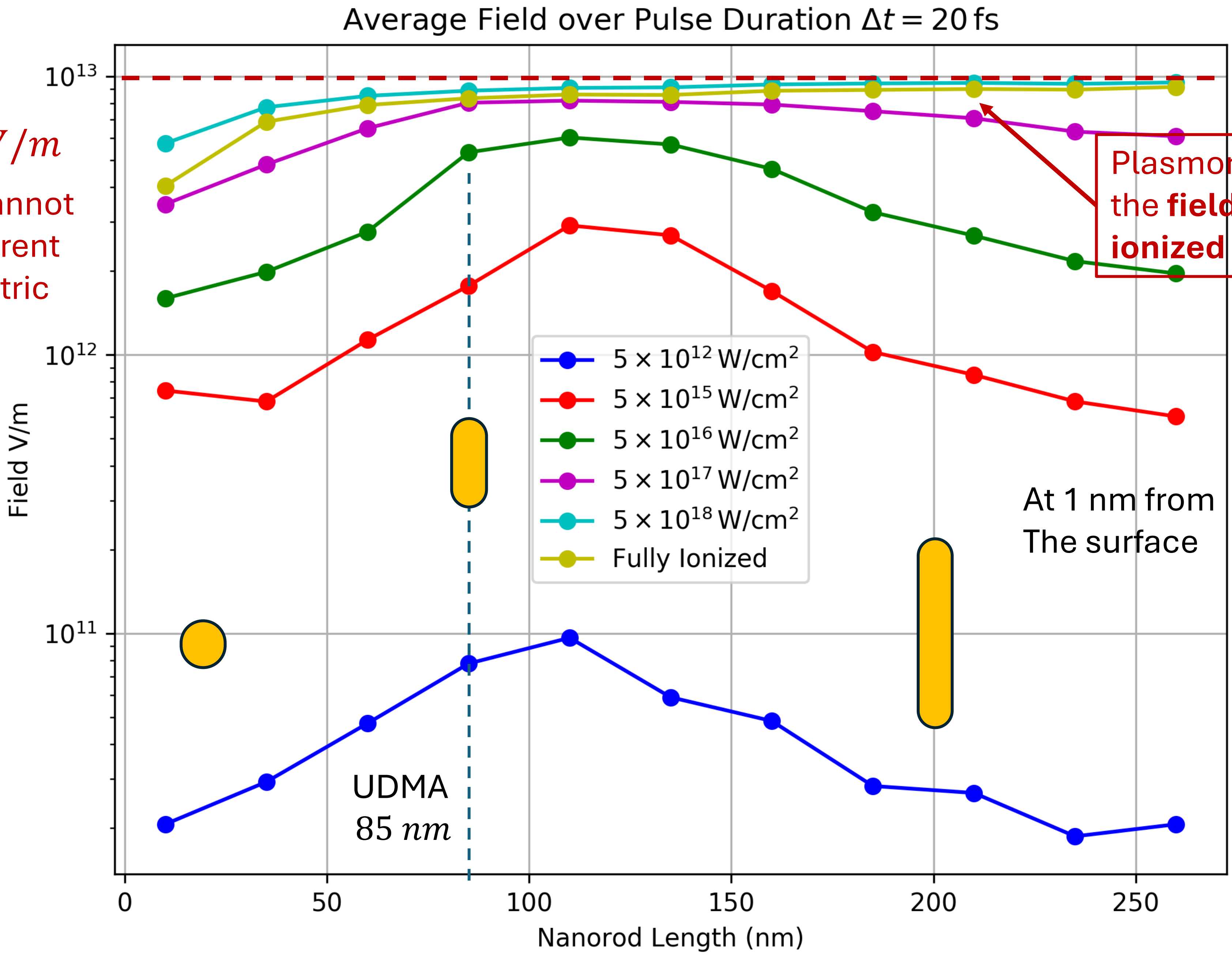
The near field enhancement cannot go above the ionization field of the nanorod



$E_{WB} \sim n_e^{1/2}$

$E_{WB} \approx 10^{13} V/m$

The nanorod cannot support a coherent oscillating electric field above this limit.



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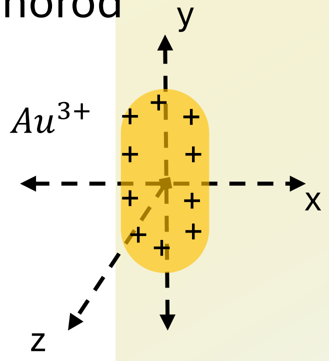
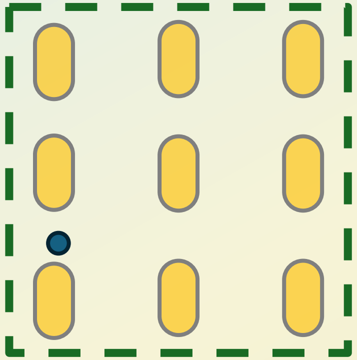


Conclusion

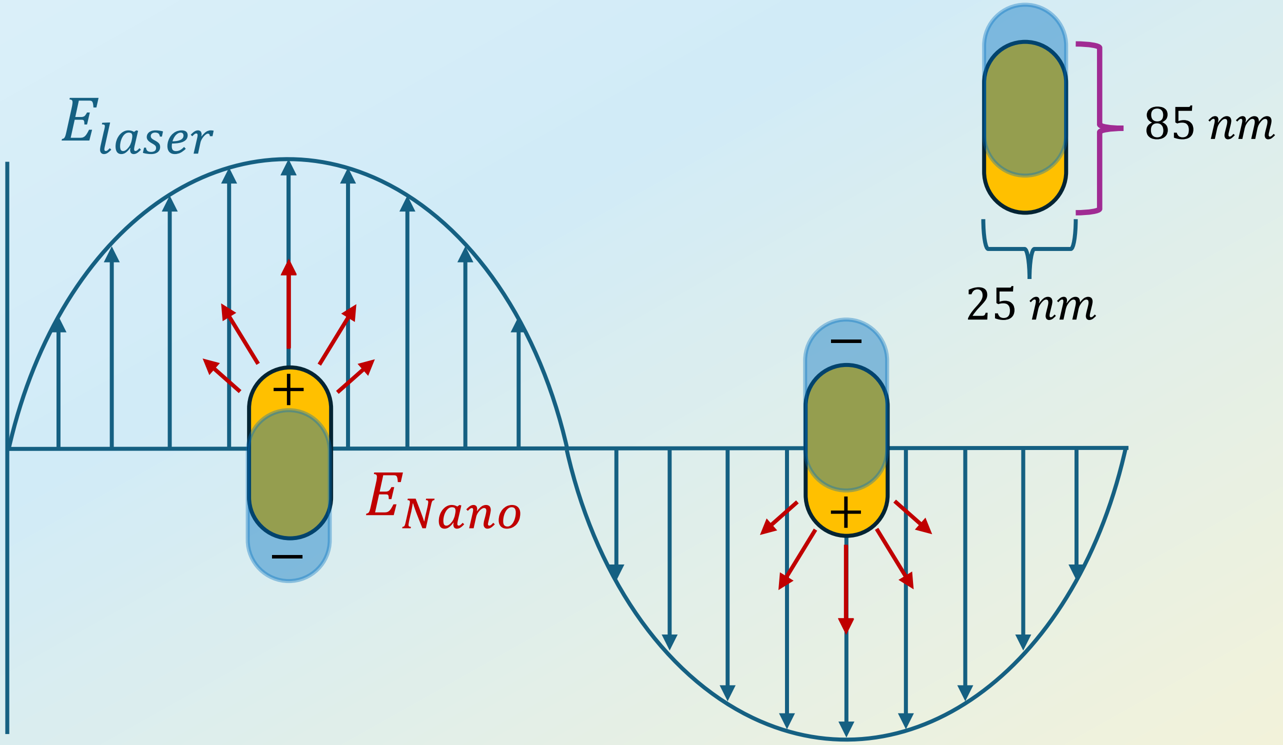
- For **low** density of nanorods **unoriented**, **short** pulse duration and **high** intensity ($I > 10^{17} W/cm^2$). The **dominant** acceleration process is **coulomb explosion.**
- In the future we will use nanorod **arrays** which can reduce **electron spillage** by sharing electrons.
  - Preliminary results show that **arrays** lead to **larger** peak energies for **ions** and **increase** in **electron temperature** T_e.
  - Previously shown for non-resonant nanorods Vallières et al. (2021)



- For ponderomotive nanorod acceleration one must use **low intensity** $< 10^{15}\ \mathrm{W/cm^2}$ and **long pulse duration**, and **structured nano targets**, such that protons will continue to acquire energy from multiple nano rods.
- Next week we go to **ELI** in Szeged for more measurements!

The NanoPlasmonic Laser Induced Fusion Energy

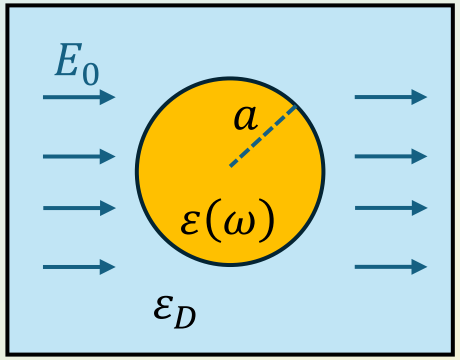
Resonance condition



*Locally **increased** field **Strength** and Field **Gradient***

These nanorods **resonantly** absorb 800 nm light via **Localized Surface Plasmon resonance** (LSP)

$$\boldsymbol{p} = \varepsilon_0\, \varepsilon_D\, \alpha\, \boldsymbol{E_0} \qquad \alpha = 4\,\pi\, a^3 \frac{\varepsilon(\omega) - \varepsilon_D}{\varepsilon(\omega) + 2\,\varepsilon_D}$$



The classical **polarizability diverges** when

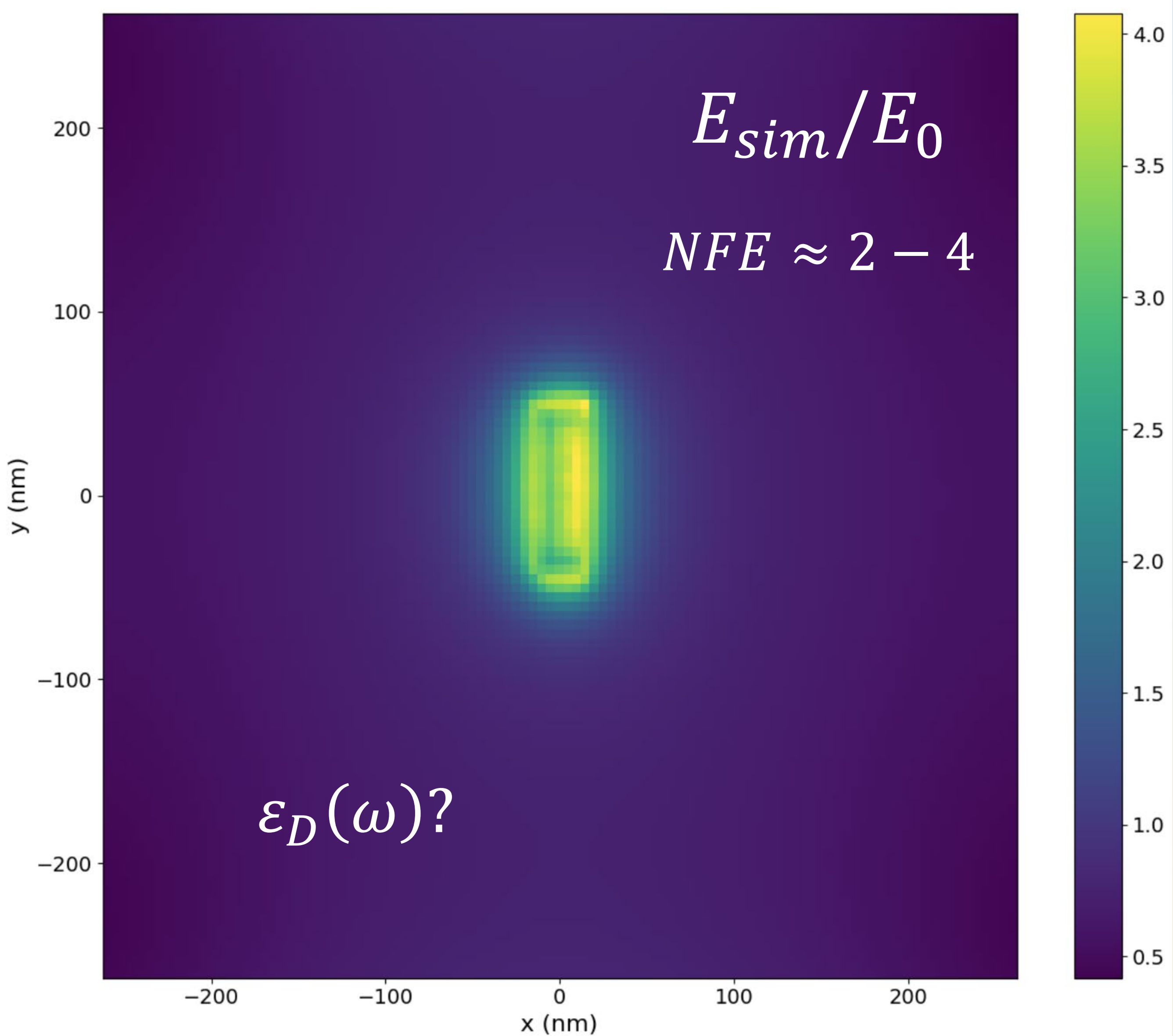
$$Re[\varepsilon(\omega)] = -2\,\varepsilon_D$$

Maier, Stefan A. Plasmonics: fundamentals and applications. Vol. 1. New York: springer, 2007.

This leads to quantum effects such as **focusing** past the **diffraction limit, resonant absorption** of light, and electric **field enhancement.**

LSP **resonance** can **respond** incredibly **fast.**
$\omega_{LSP} = 2.65\ fs$

Why is the NFE effect on ions so small?



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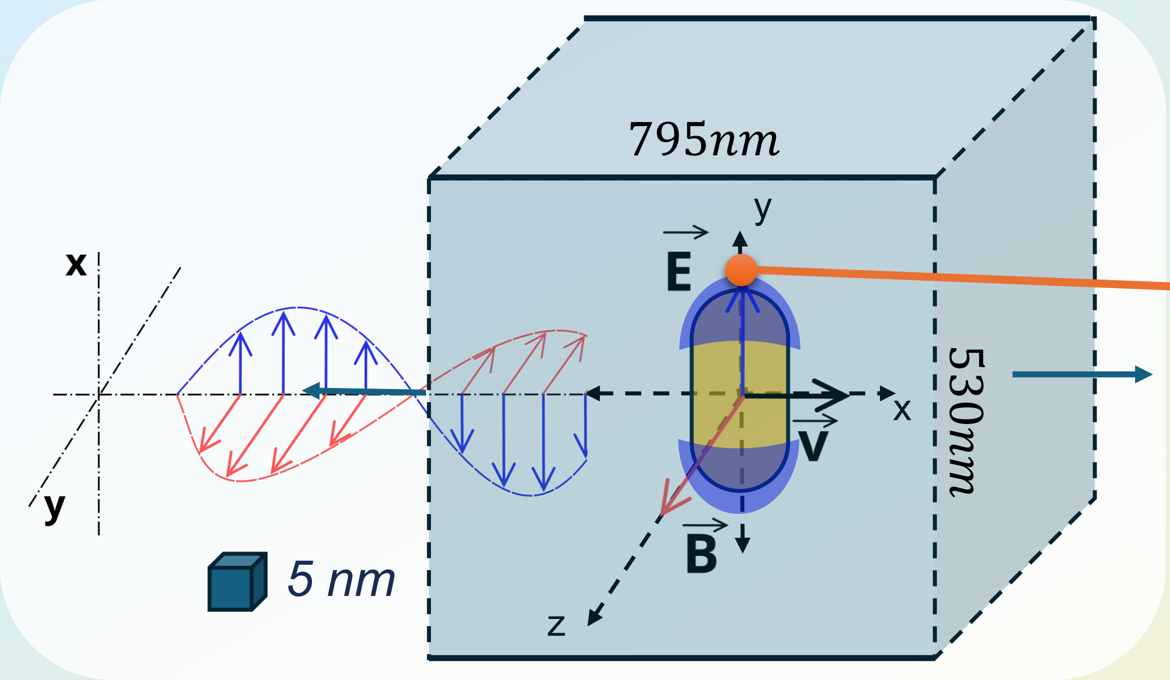

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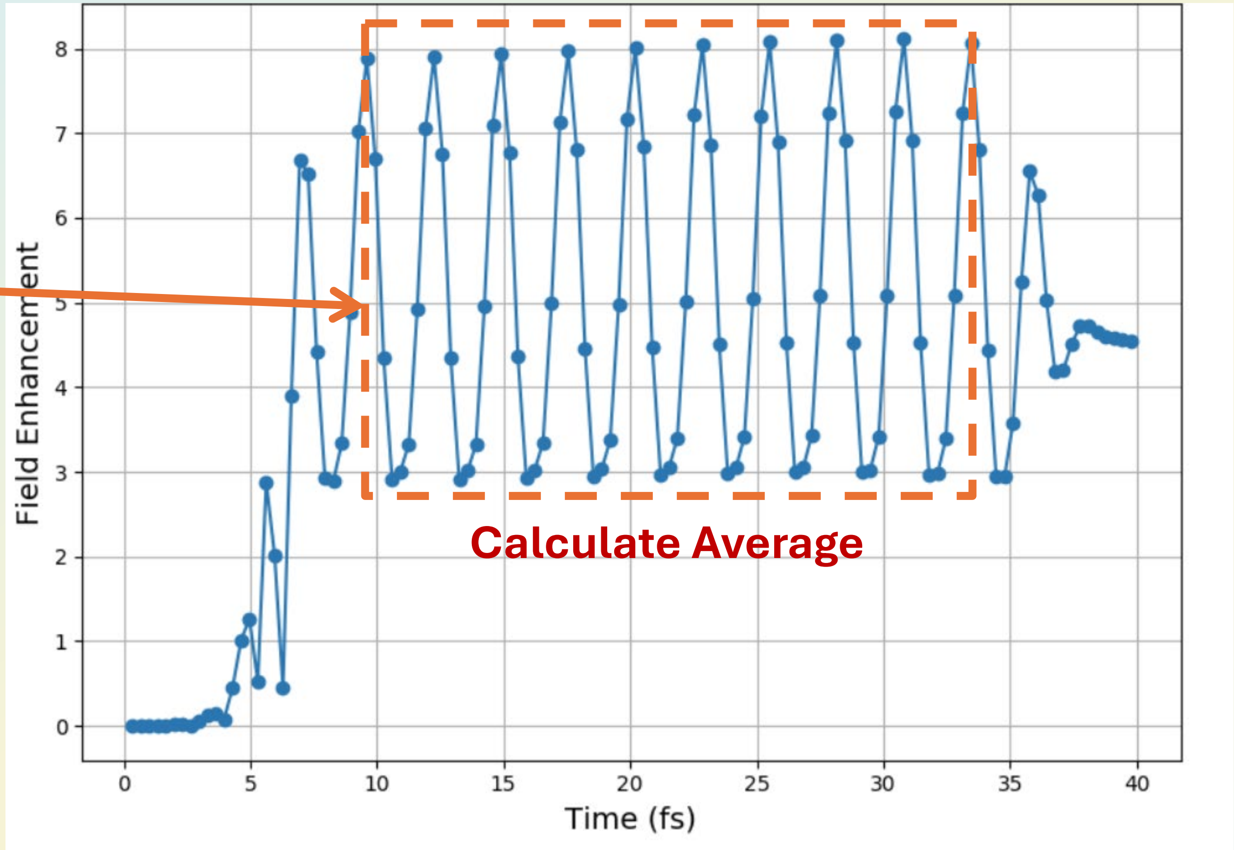
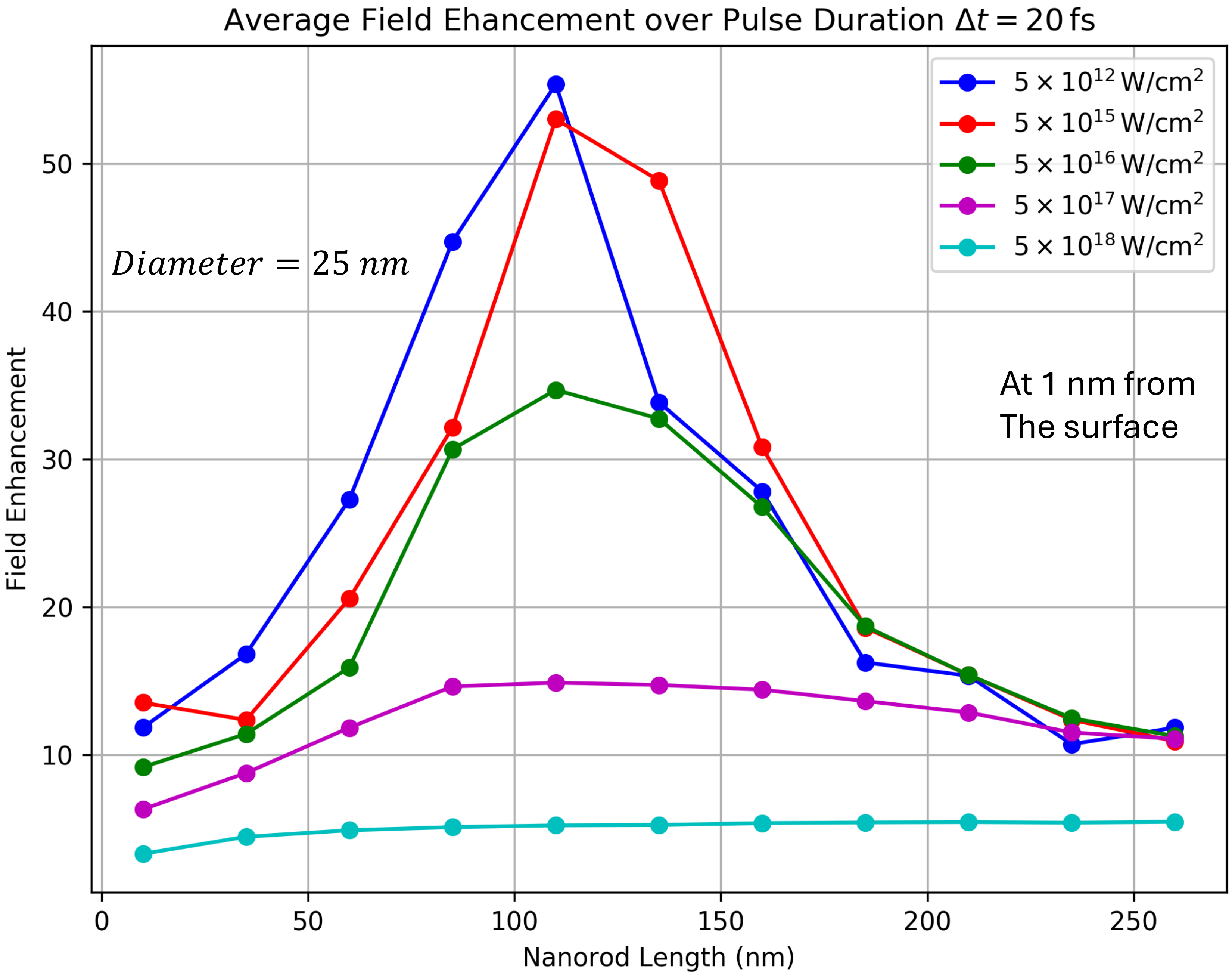
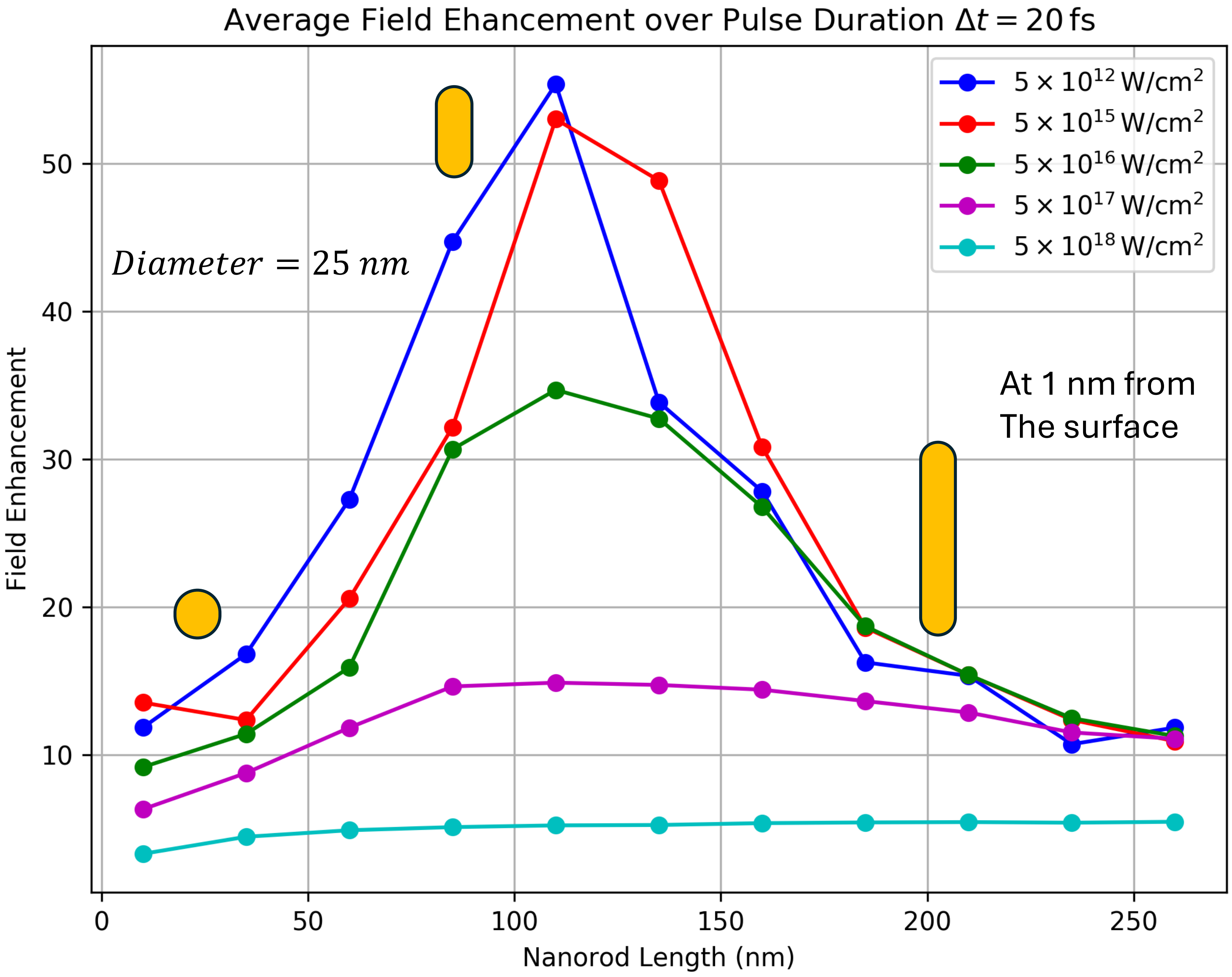




55 simulations
2,200 fs
11hrs – Wall hours
704 – CPU hours
-fairly cheap simulation



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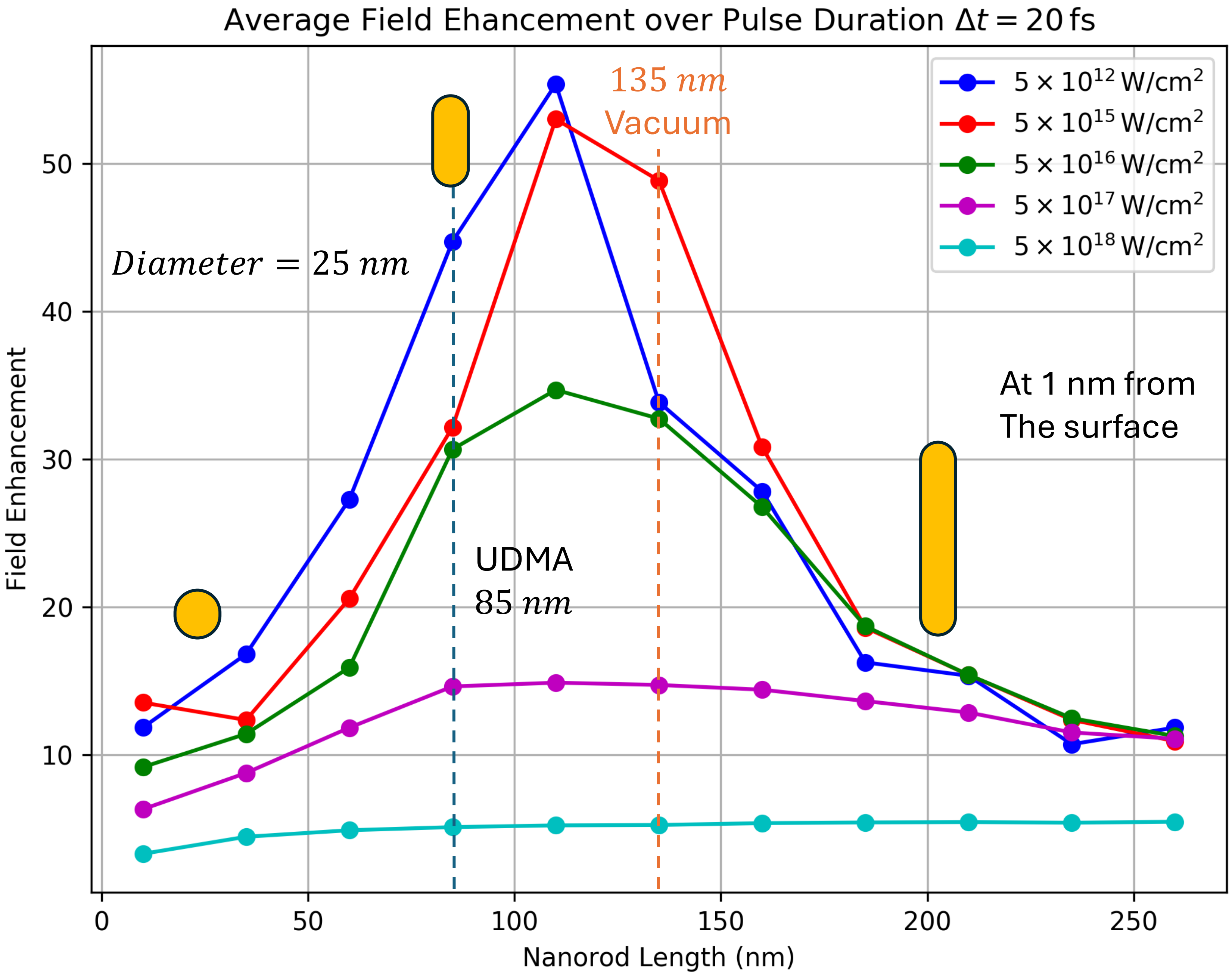

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135 nm
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Field Enhancement
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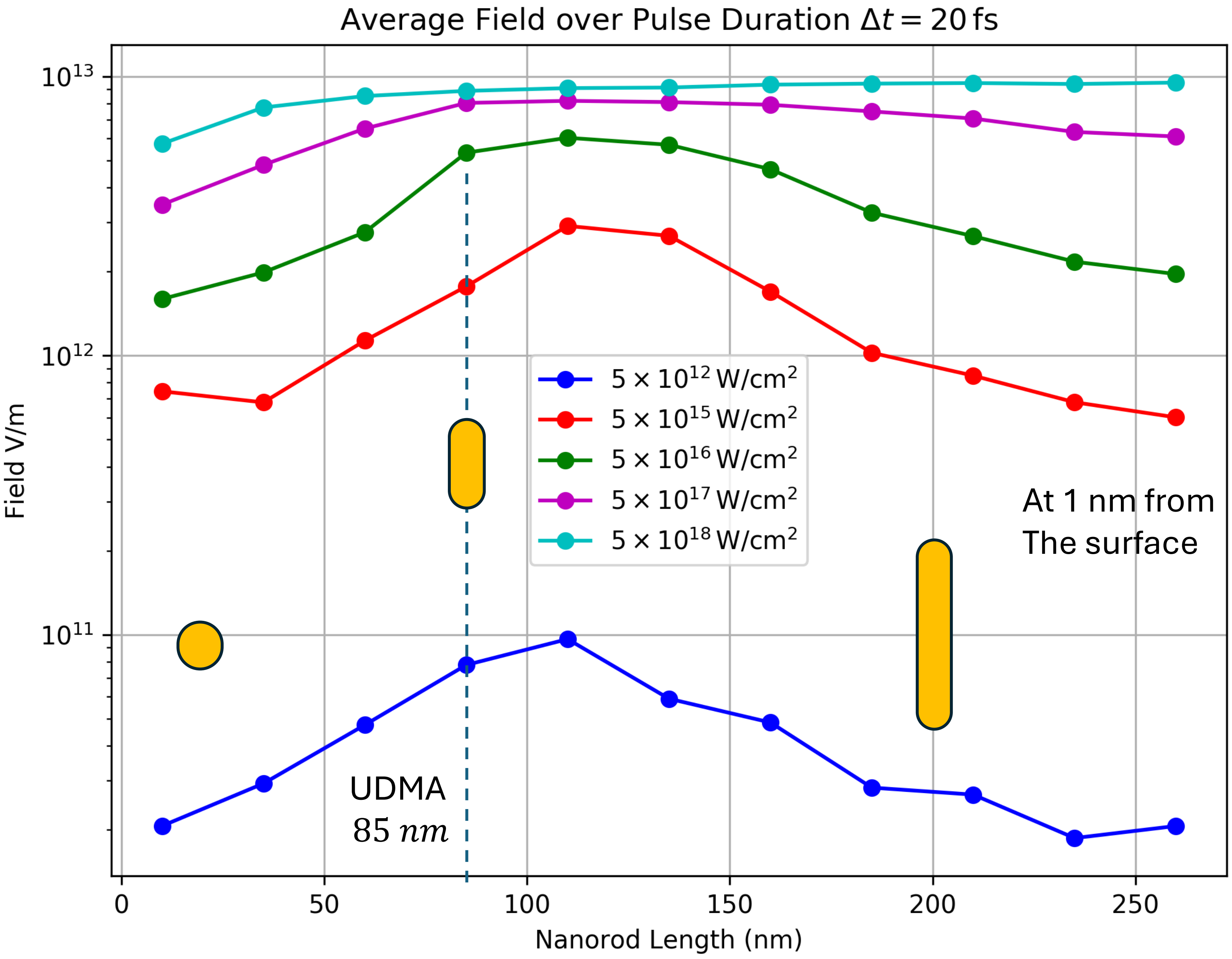
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Field V/m
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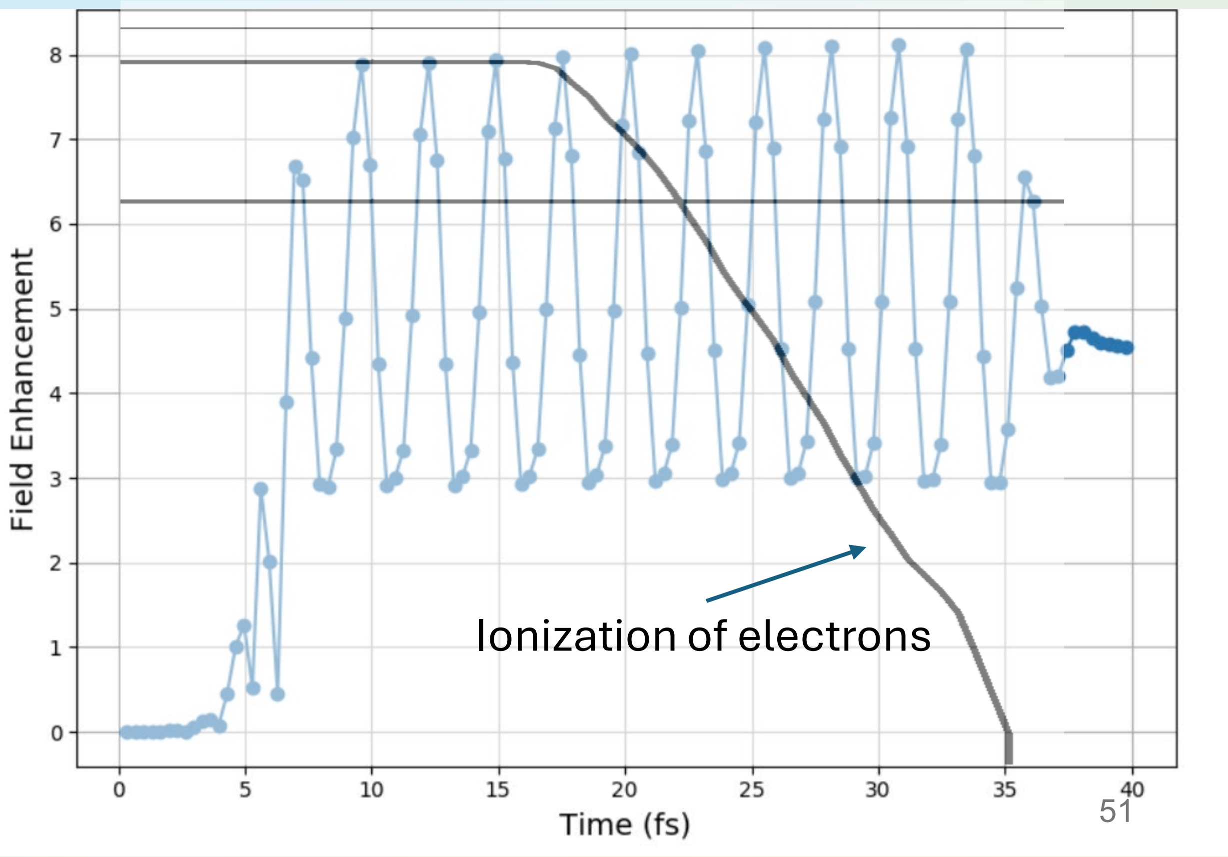
10^{11}
0
50
100
150
200
250
5×10^{12} W/cm^2
5×10^{15} W/cm^2
5×10^{16} W/cm^2
5×10^{17} W/cm^2
5×10^{18} W/cm^2
At 1 nm from
The surface
UDMA
85 nm

Conclusion

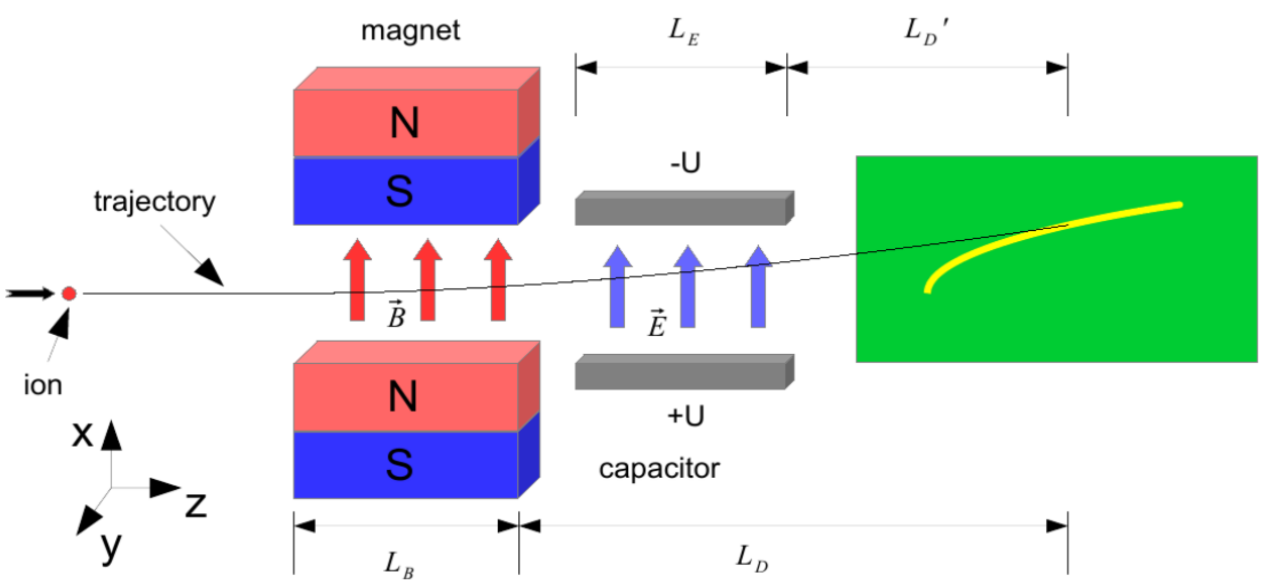
Open Questions and Improvements:

- Tailor **medium** to better **represent UDMA** $\varepsilon_D(\omega)$.
- Why does the resonance **decrease** at **high intensity**?
- **NFE** does **not decrease** due to **ionization….**
  - Why is there a **late time** NFE?
  - Can the **phenomena** responsible for this be used to create more **resonance** at **higher intensities**?

Measure the Near Field Enhancement Predicted by Epoch



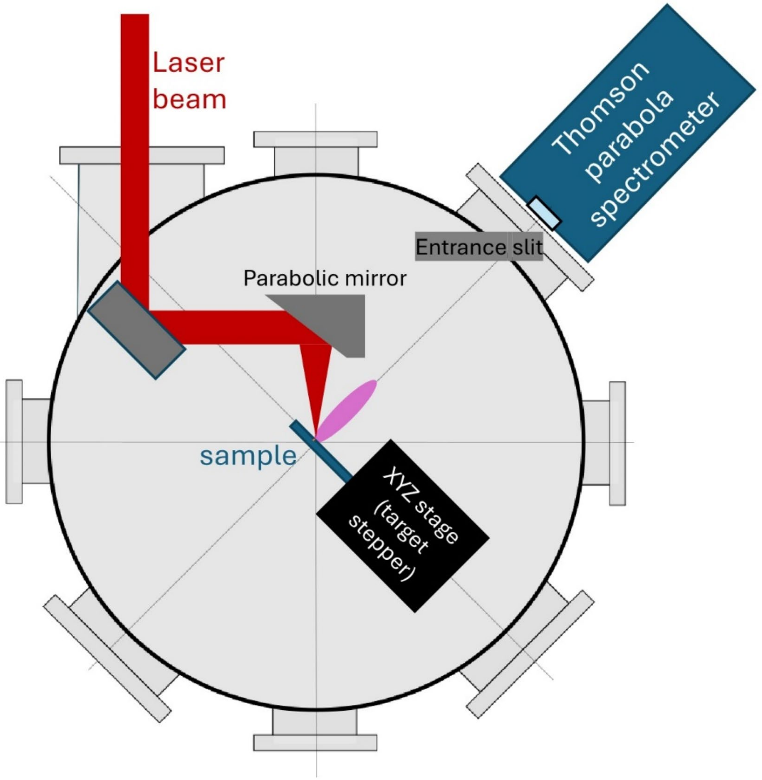
Thompson parabola spectra



(Master Thesis of Nils Schroeter, 2017)

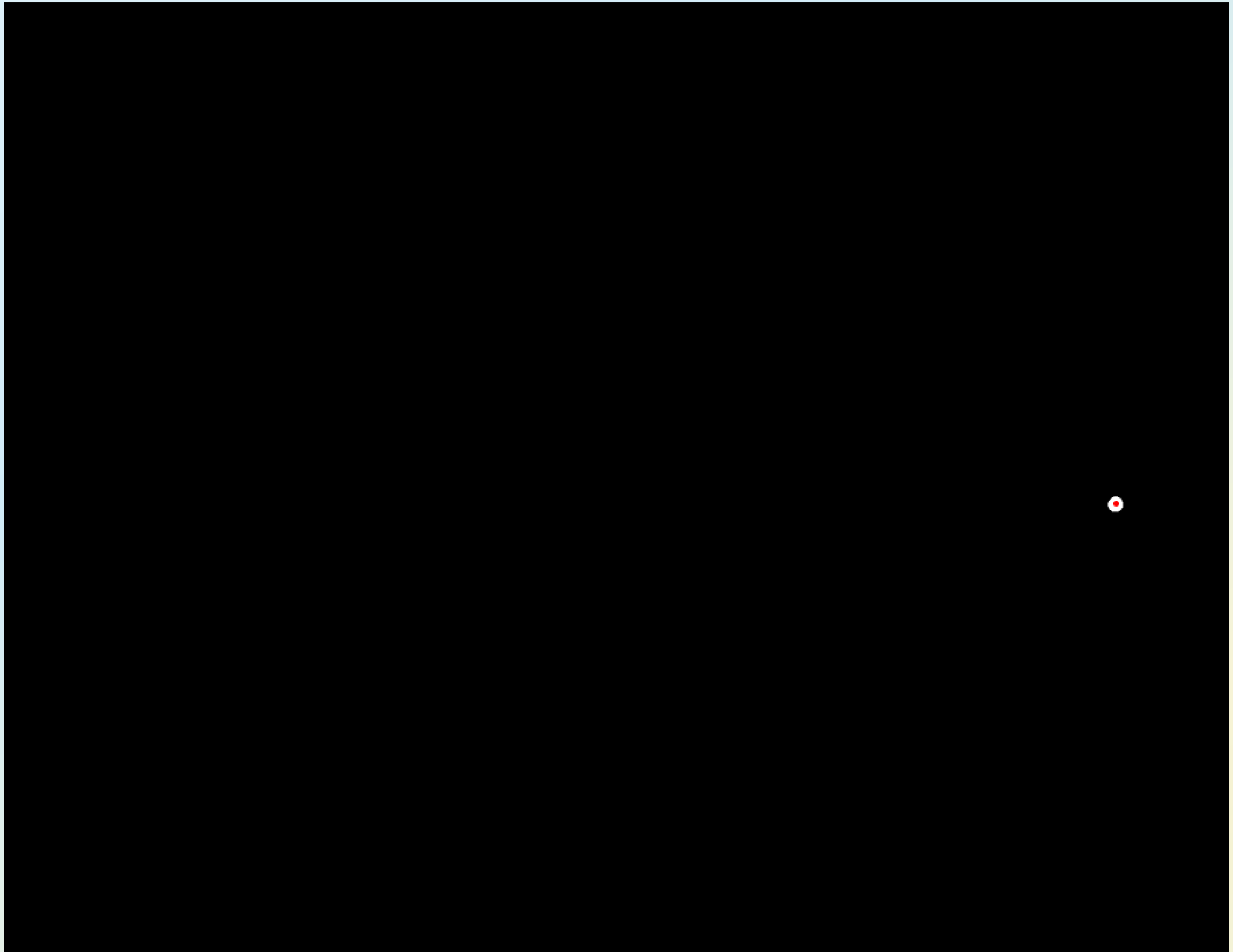
$$x = \frac{q|\vec{B}|}{\sqrt{2m_i E_{kin}}}\left(\frac{L_B^2}{2} + L_B\, L_D\right)$$

$$y = \frac{q|\vec{E}|}{2E_{kin}}\left(\frac{L_E^2}{2} + L_E L_D'\right)$$

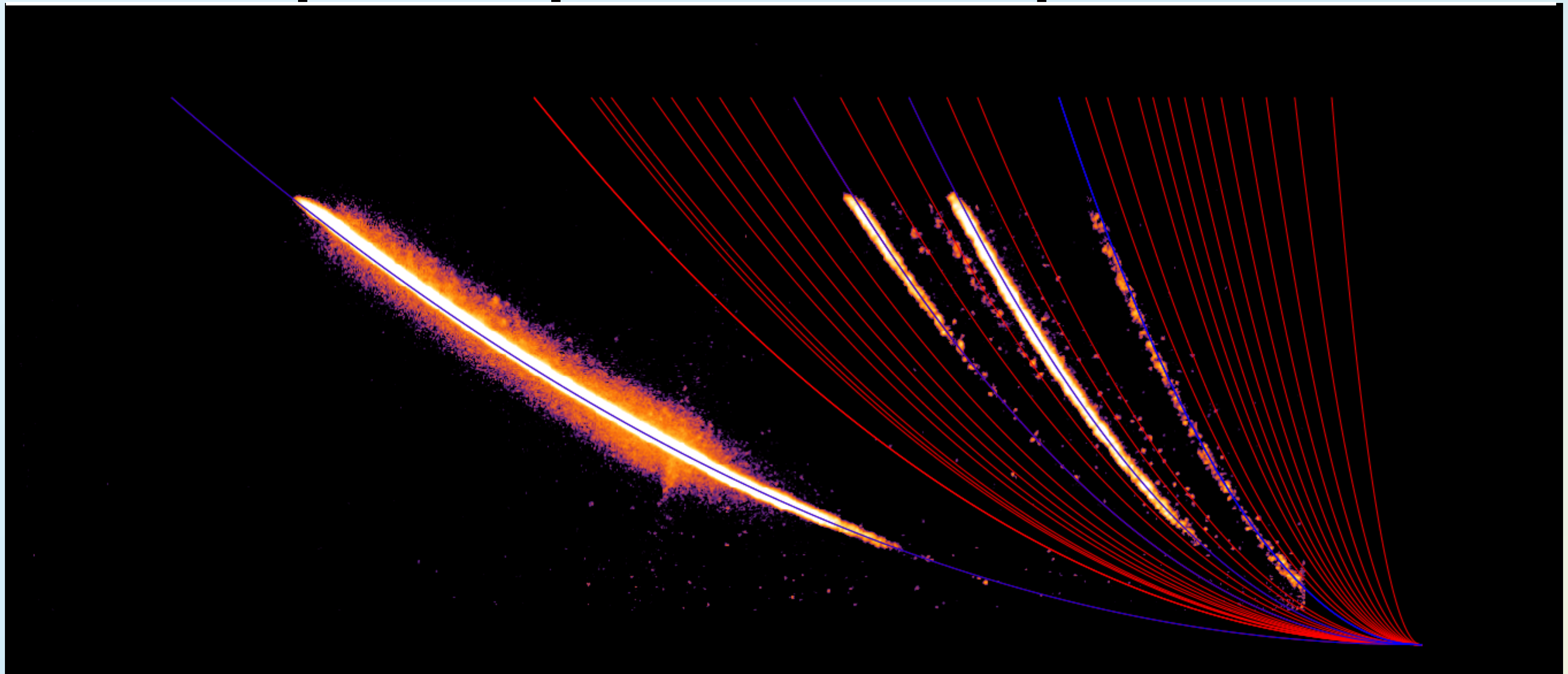


Thompson parabola spectra

Thompson parabola spectra

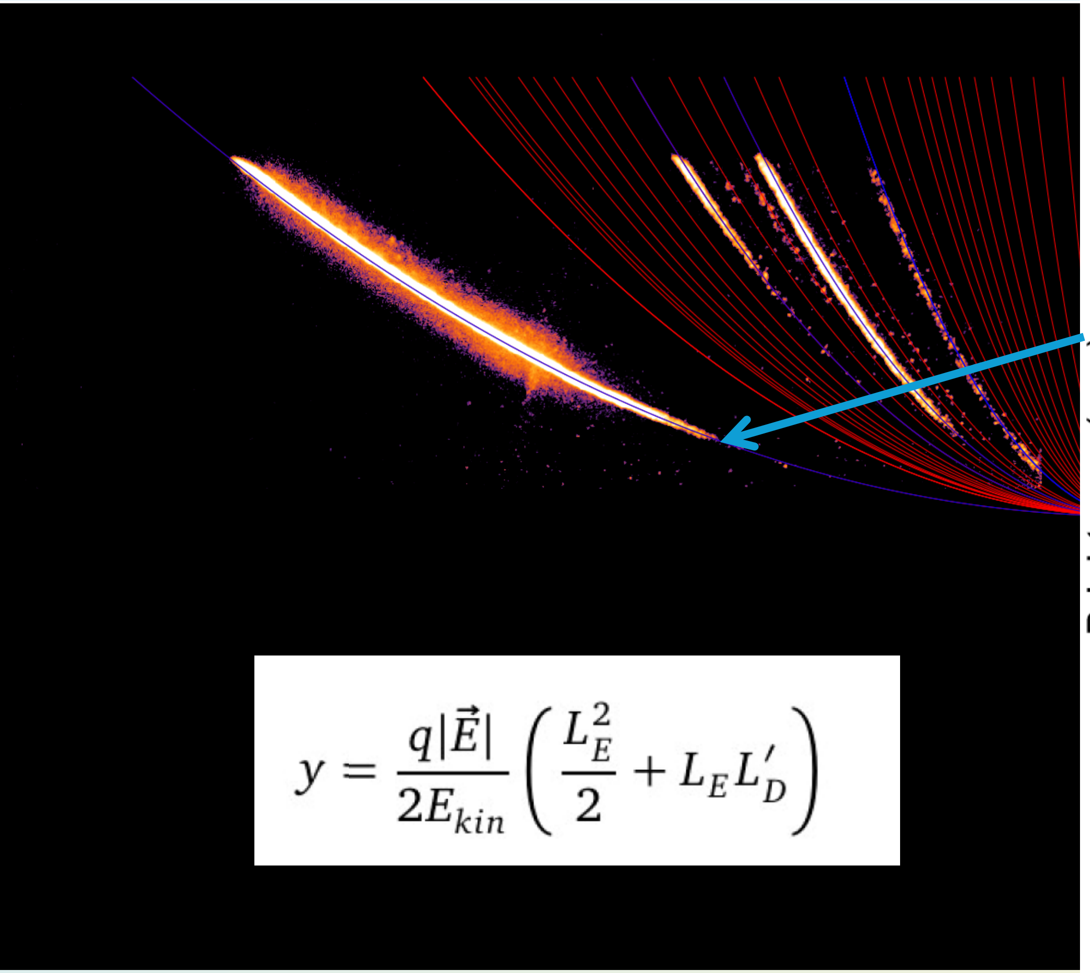


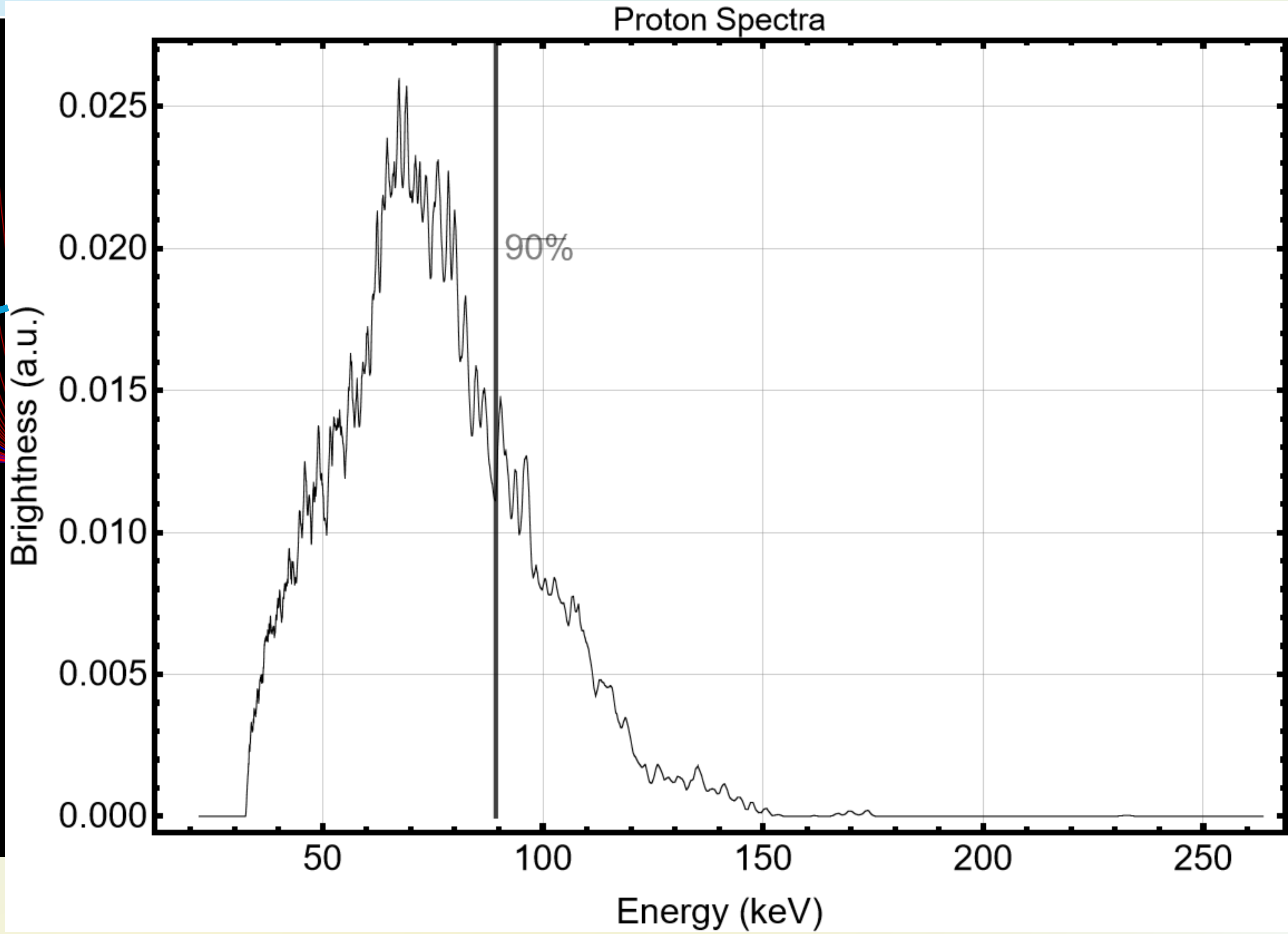
Thompson parabola spectra



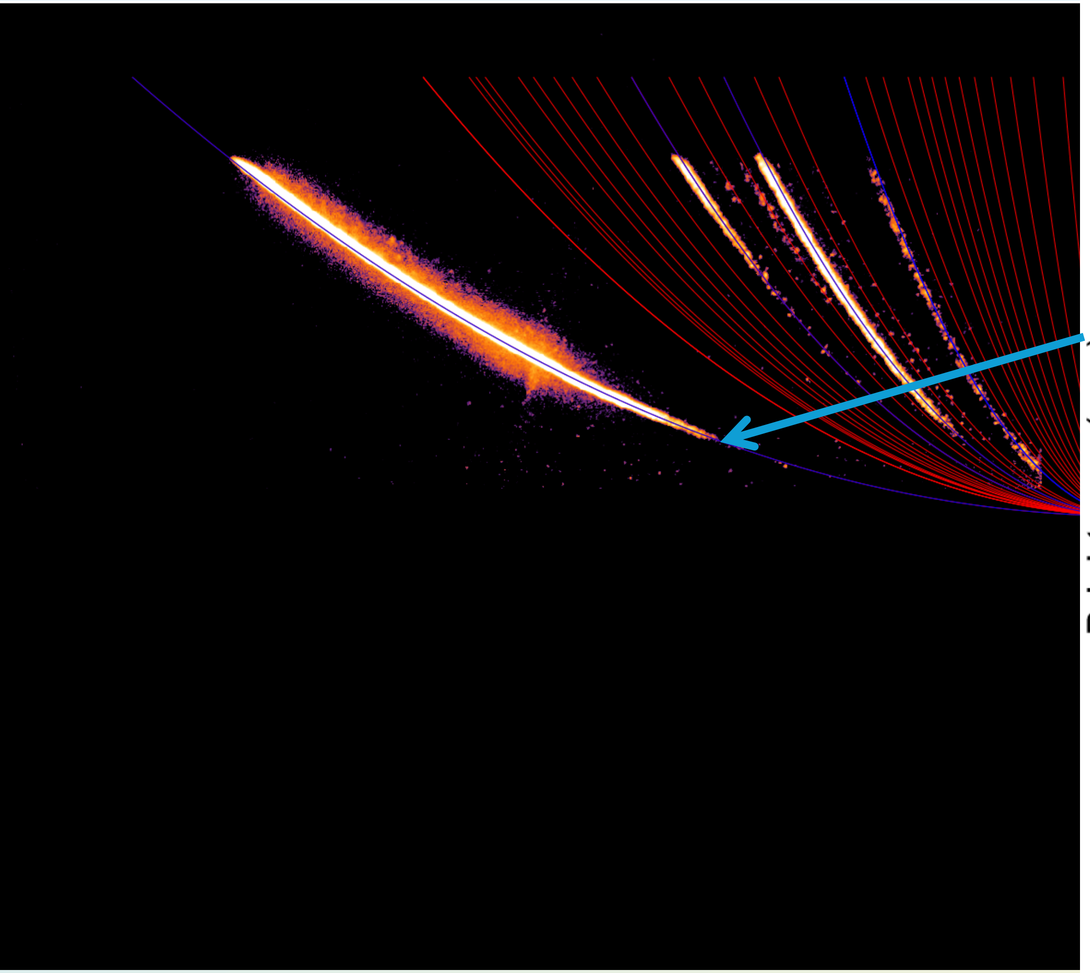
Measured	Species	Charge	Theoretical q/m	Absolute Error	Percent Error (%)
0.167201	C	2	0.166667	0.000533845	0.319284
0.251389	O	4	0.250079	0.00130935	0.520845
0.987769	H	1	0.992236	0.00446716	0.452247
0.18821	O	3	0.18756	0.000650176	0.345453
0.0847248	C	1	0.0833333	0.00139142	1.64228
0.0846572	C	1	0.0833333	0.00132385	1.56378
0.249623	C	3	0.25	0.000376823	0.150957
0.0843078	C	1	0.0833333	0.000974506	1.15589
0.0834283	C	1	0.0833333	0.0000949199	0.113774
0.0837136	C	1	0.0833333	0.000380273	0.454255
0.0838404	C	1	0.0833333	0.000507066	0.604799

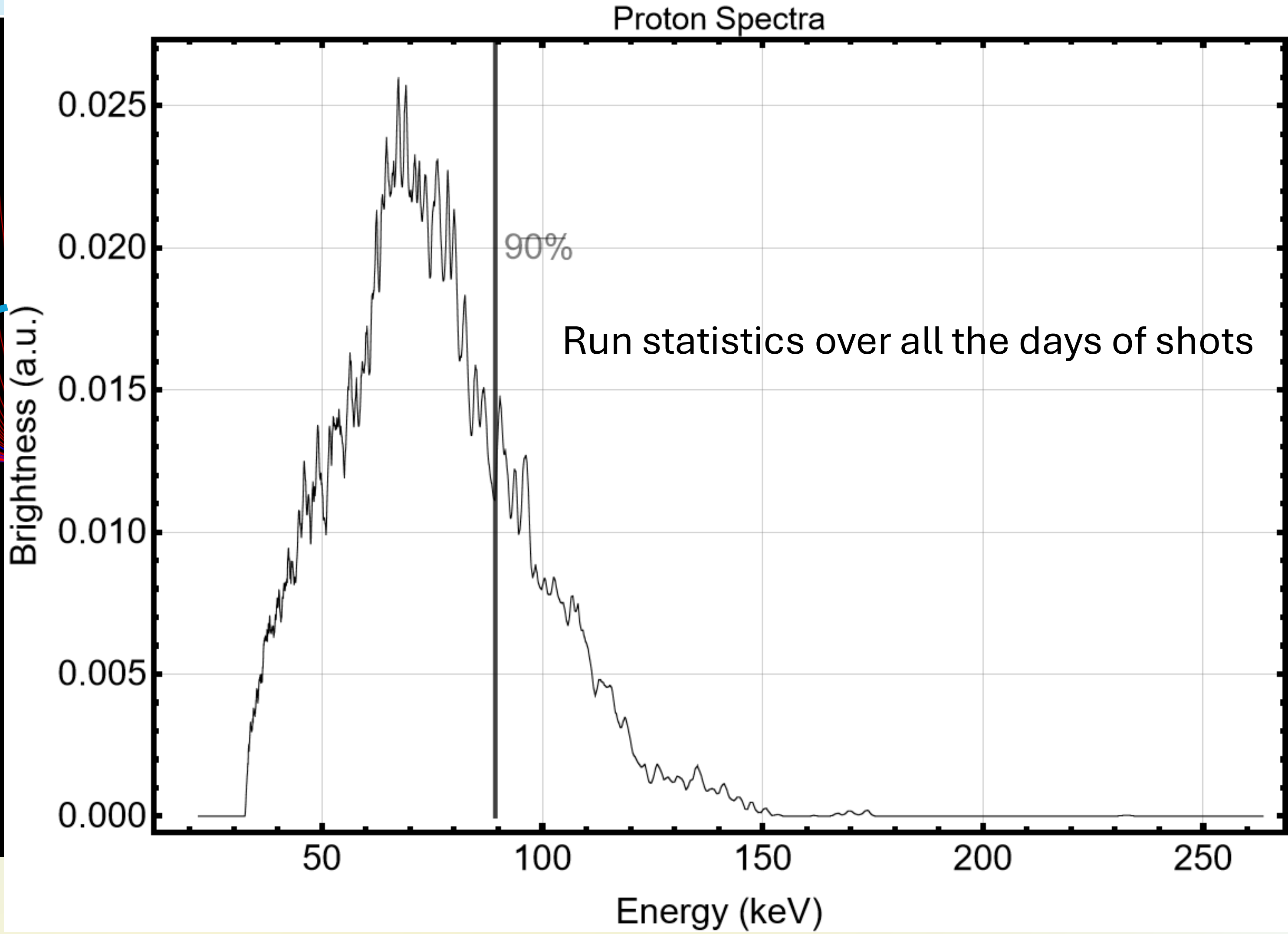
Thompson parabola spectra



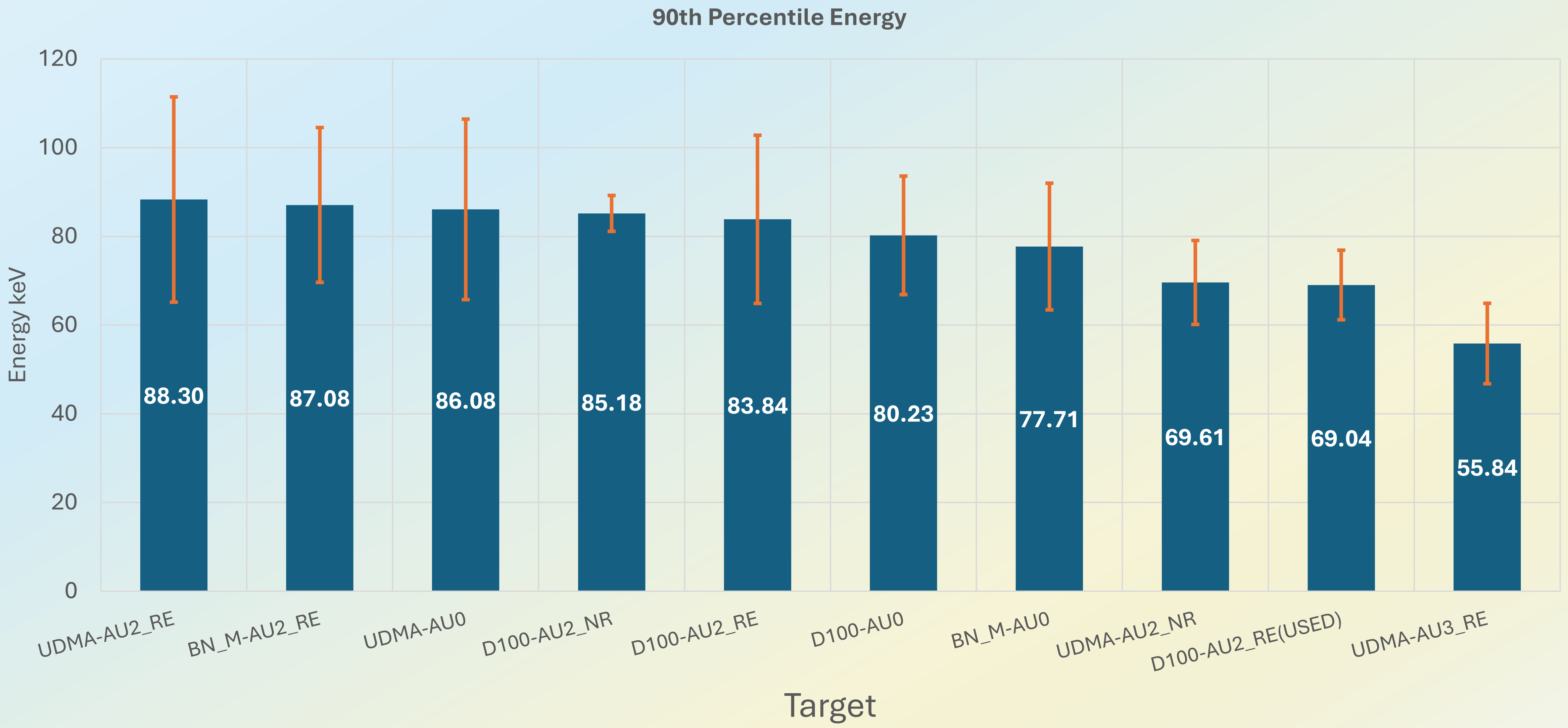


Thompson parabola spectra





Thompson parabola spectra



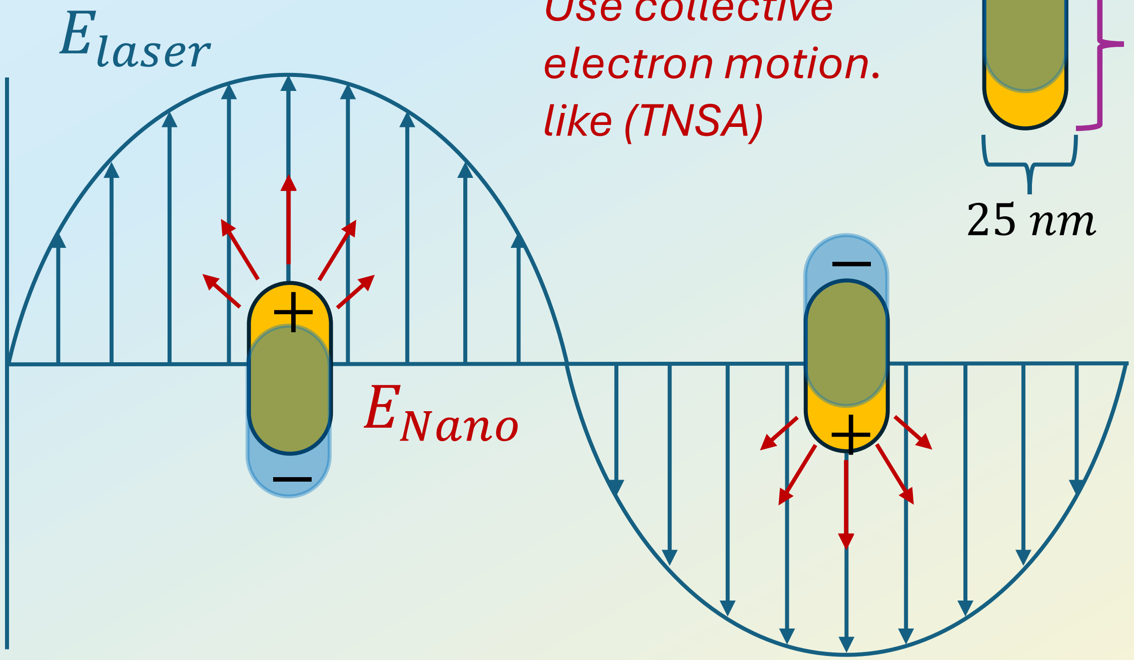
The NanoPlasmonic Laser Induced Fusion Energy

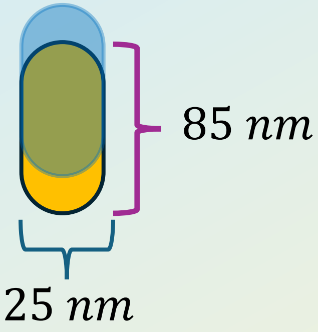
Can **nano-structured** targets lead to **femtosecond** scale **ion acceleration**?

Huge factor to overcome → $\frac{a_{0p}}{a_{0e}} = \frac{m_e}{m_p} \approx 5 \times 10^{-4}$

Solution

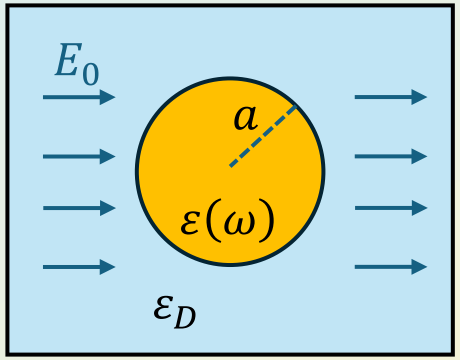
Use collective electron motion. like (TNSA)





These nanorods **resonantly** absorb 800 nm light via **Localized Surface Plasmon resonance** (LSP)

$$\boldsymbol{p} = \varepsilon_0\, \varepsilon_D\, \alpha\, \boldsymbol{E_0} \qquad \alpha = 4\,\pi\, a^3 \frac{\varepsilon(\omega) - \varepsilon_D}{\varepsilon(\omega) + 2\,\varepsilon_D}$$



The classical **polarizability diverges** when

$$Re[\varepsilon(\omega)] = -2\,\varepsilon_D$$

Maier, Stefan A. Plasmonics: fundamentals and applications. Vol. 1. New York: springer, 2007.

This leads to quantum effects such as **focusing** past the **diffraction limit, resonant absorption** of light, and electric **field enhancement.**

LSP **resonance** can **respond** incredibly **fast.**
$\omega_{LSP} = 2.65\ fs$

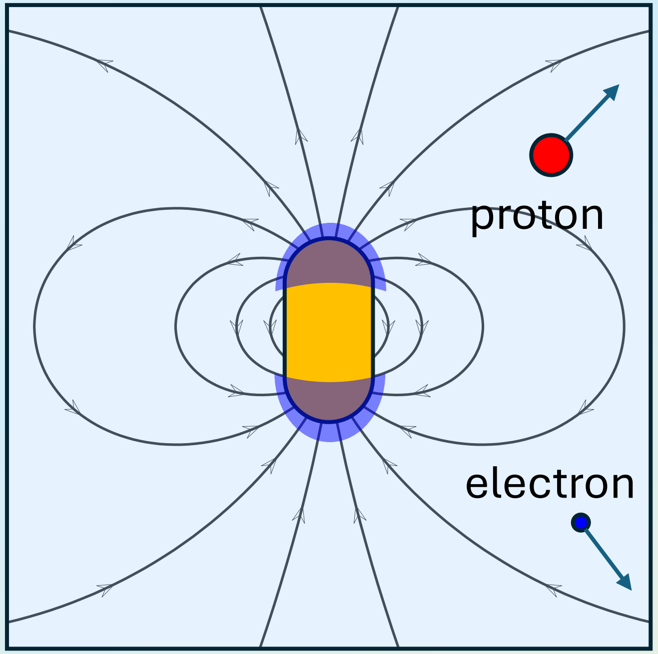
*Locally **increased** field **Strength** and Field **Gradient***

Plasmonic Ponderomotive Force

$\omega = 2.65\ fs$

$hc = 0.197$ keV nm

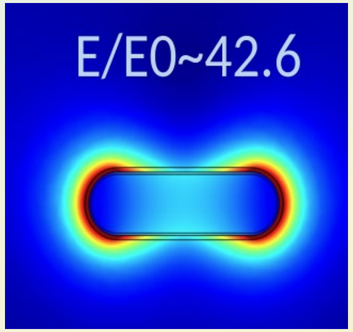
Our group was mainly interested in forces due to the **oscillating electron cloud** around the gold nanorod due to **LSP** resonance that could directly couple to ions.



Typically, the **ponderomotive force** is far too **small** to cause **direct acceleration** to **protons** due to their large **mass**.

$$\mathbf{F}_\mathrm{p} = -\frac{Z^2 hc^2}{4m\omega^2}\nabla\left\langle (e\mathbf{E})^2\right\rangle_{2\pi/\omega}$$

However, the ponderomotive force due to the oscillating field around a **nanorod** can be much larger due to **field enhancement** and increase in the field **gradient**.



*the ∇E^2 term is ~**1600** times **larger** for the **nanorod** than the laser by field enhancement.*

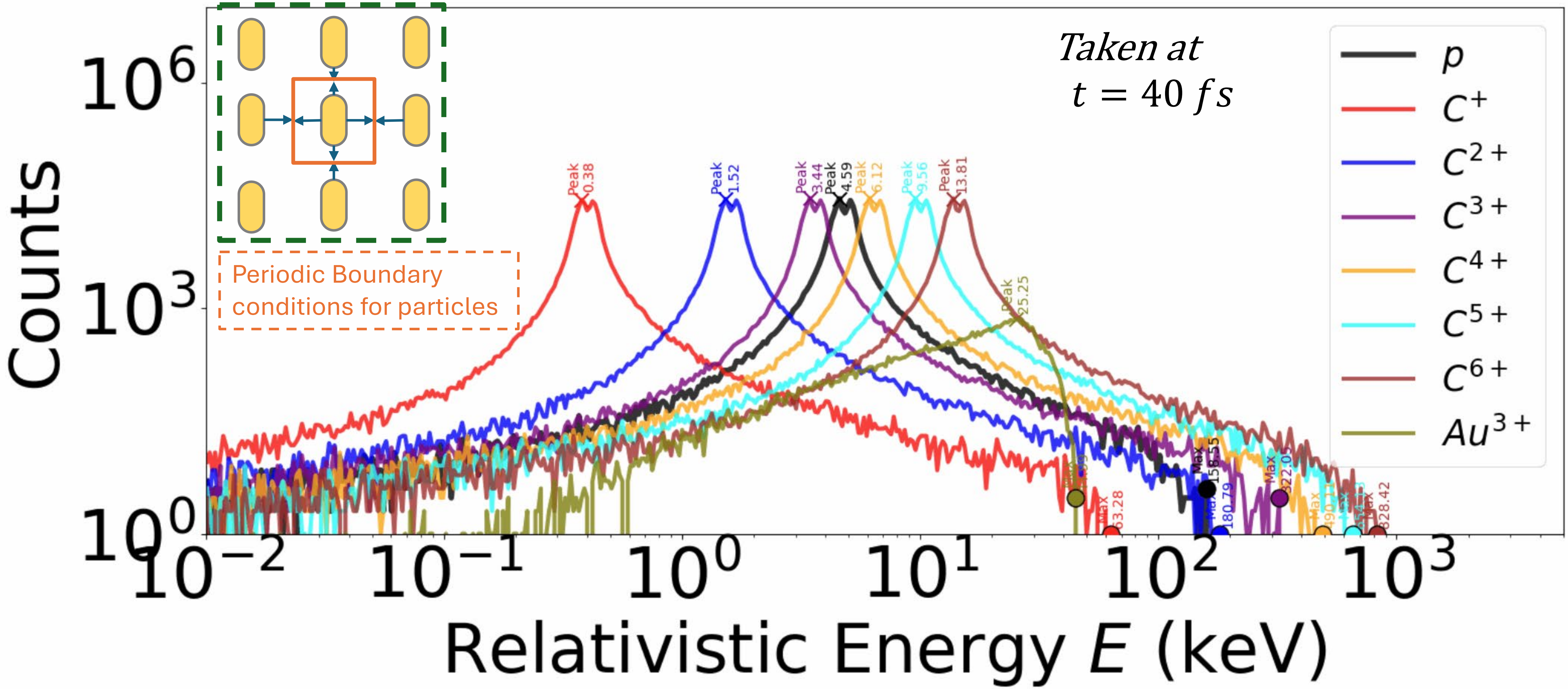
$$\mathbf{U}_\mathrm{p} = \frac{Z^2\alpha^2 hc^4}{4m\omega^2}\left(\frac{p^2r^2 + 3(\mathbf{r}\cdot\mathbf{p})^2}{r^8}\right)$$

$U_p(nanorod) = \mathbf{4\ MeV}$ (for a proton)

$$\boldsymbol{p} = N_e e\boldsymbol{d} \approx 10^6 e\ (85\ nm)\hat{\boldsymbol{z}}$$

Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\, W/cm^2 \quad \Delta t_{pulse} = 21\, fs$

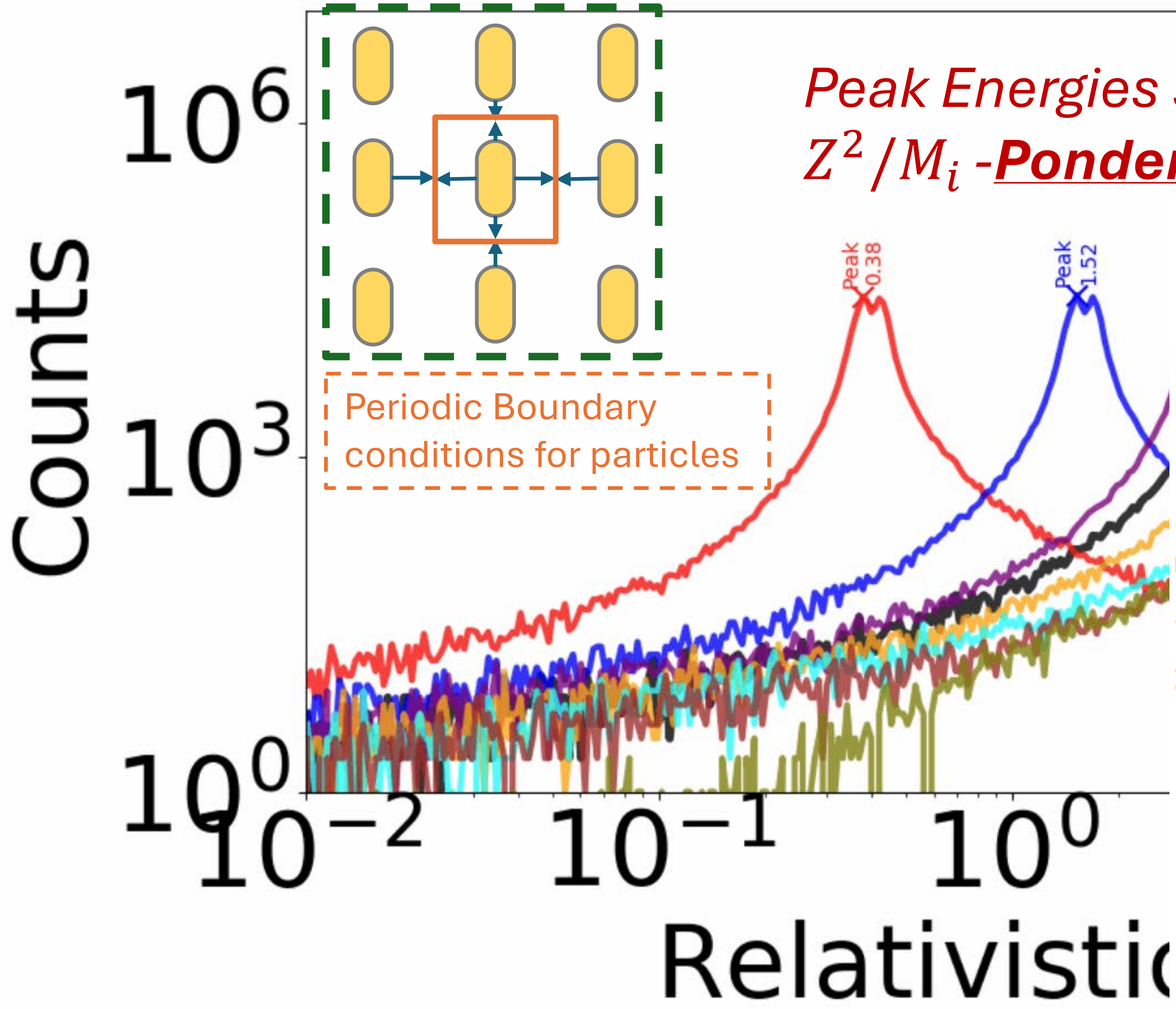


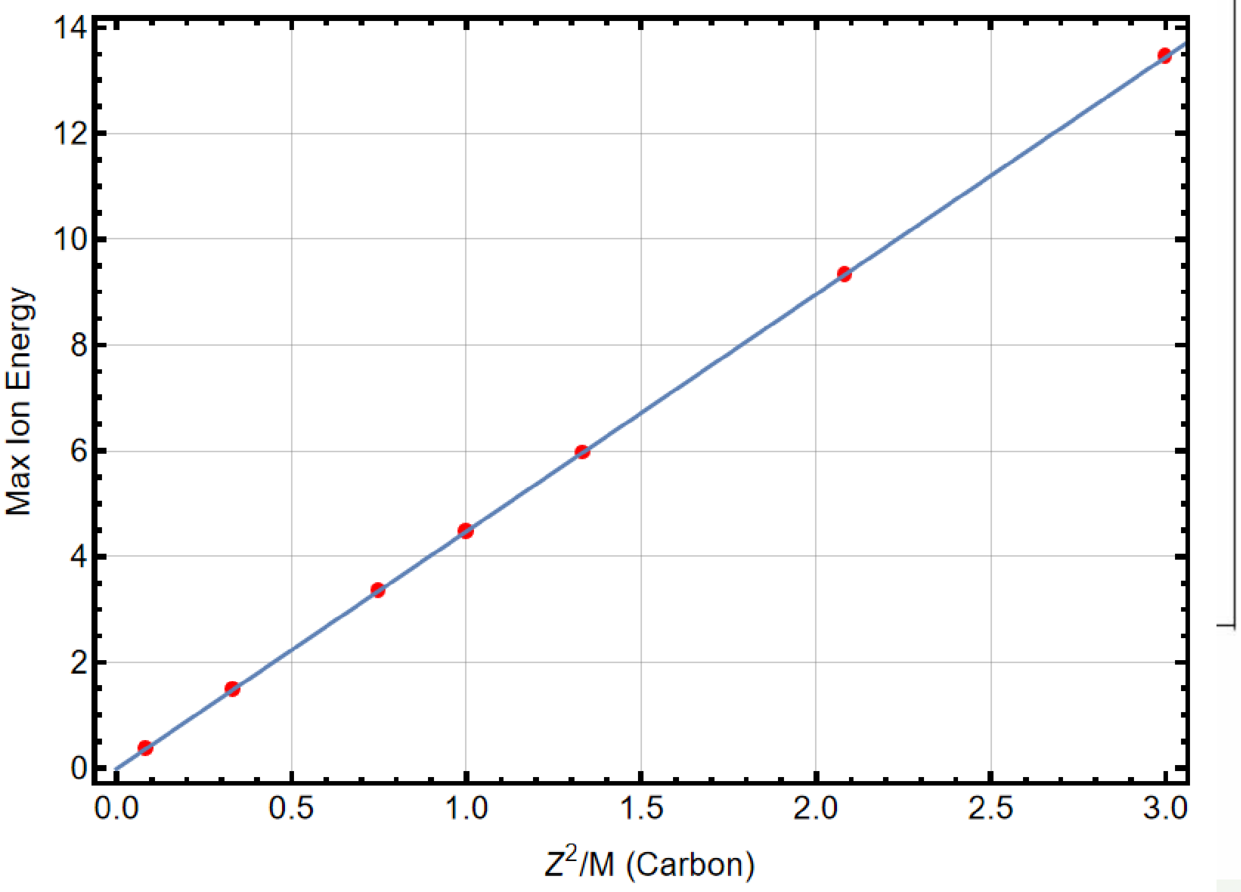
Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\ W/cm^2 \quad \Delta t_{pulse} = 21\ fs$

*Peak Energies scale with Z^2/M_i -**Ponderomotive***

Taken at $t = 40\ fs$





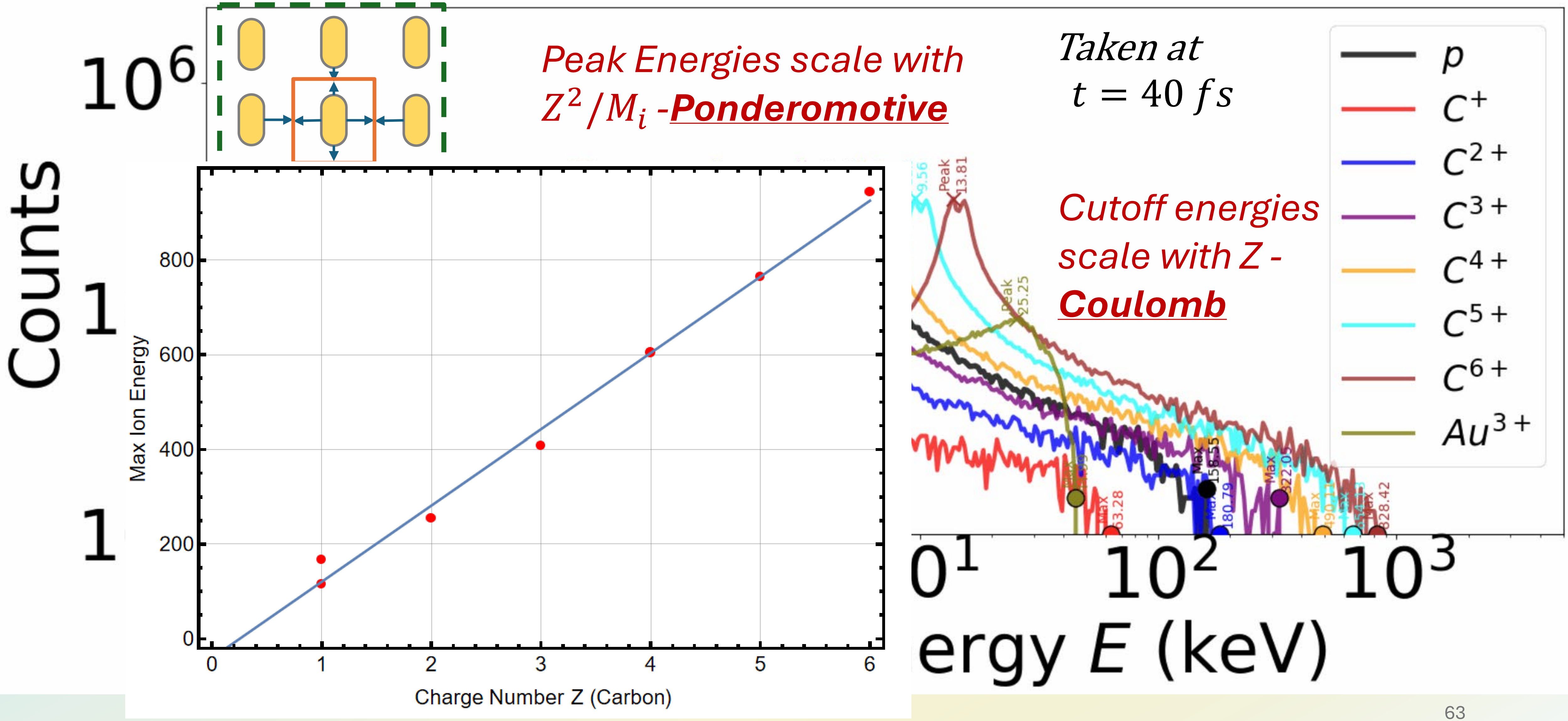
Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\, W/cm^2 \quad \Delta t_{pulse} = 21\, fs$

*Peak Energies scale with Z^2/M_i -**Ponderomotive***

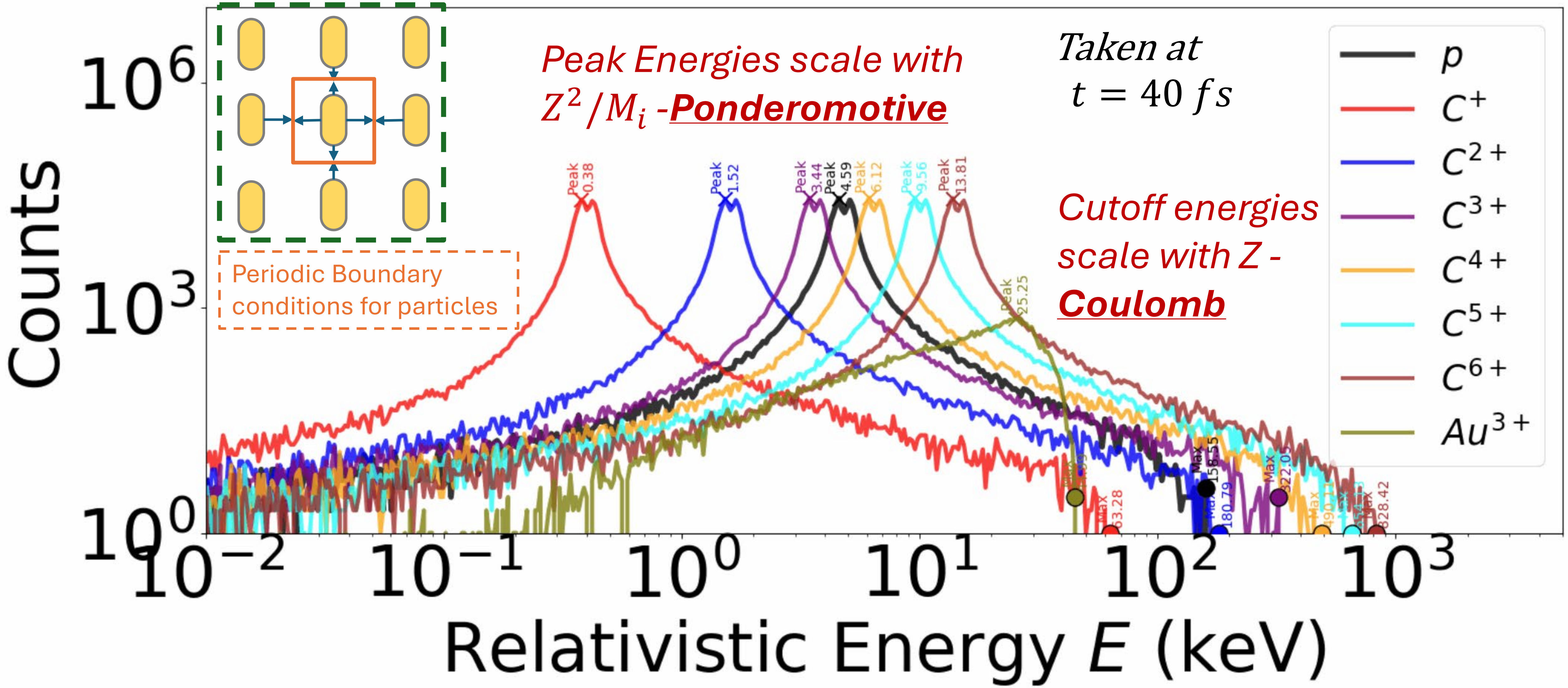
Taken at $t = 40\, fs$

*Cutoff energies scale with Z - **Coulomb***



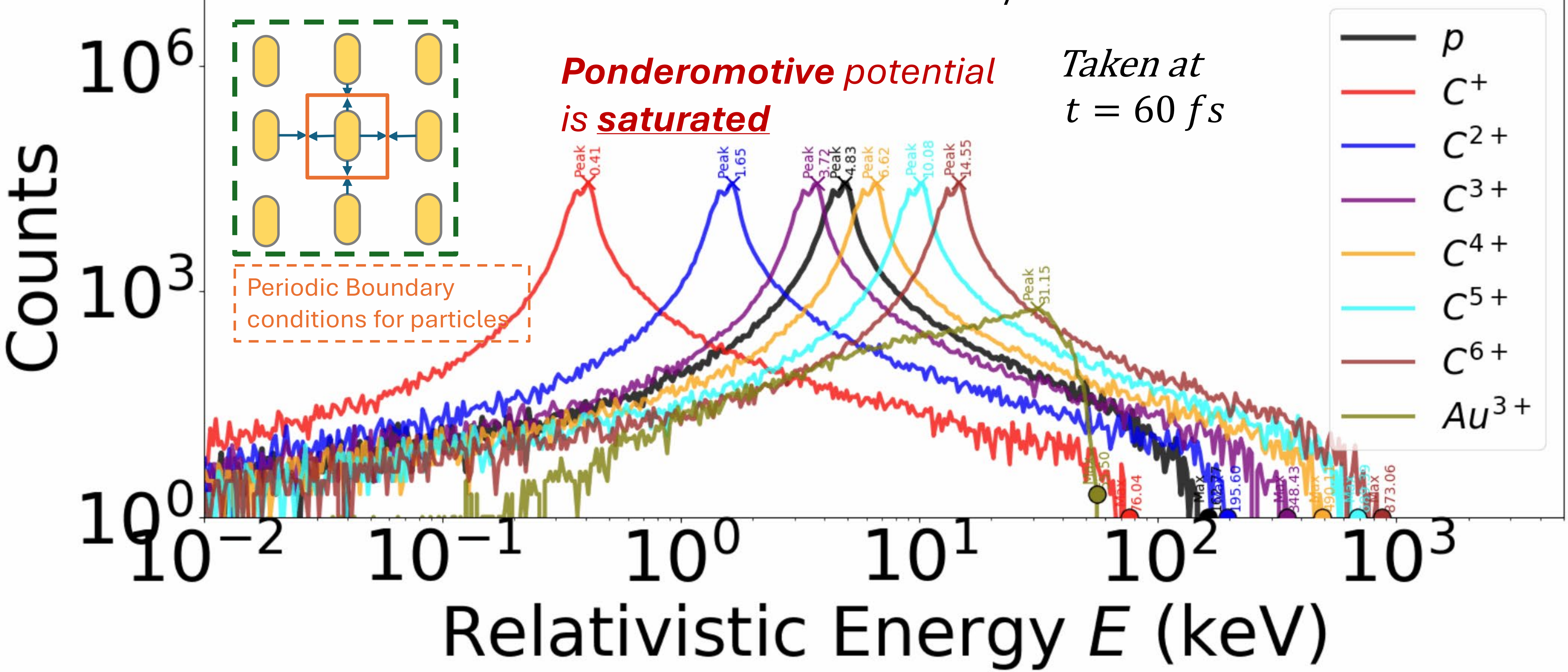
Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\ W/cm^2 \quad \Delta t_{pulse} = 21\ fs$



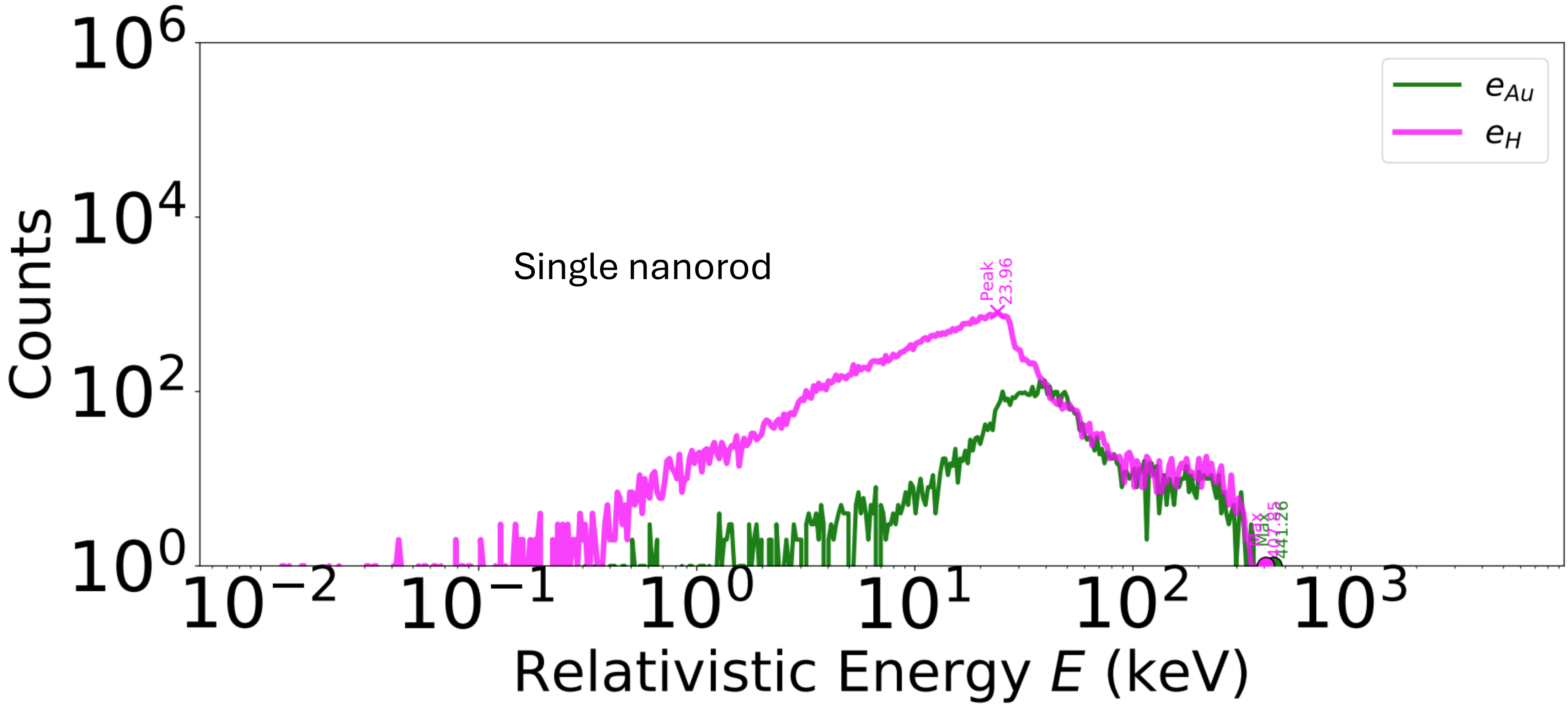
Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\ W/cm^2$ $\Delta t_{pulse} = 60\ fs$



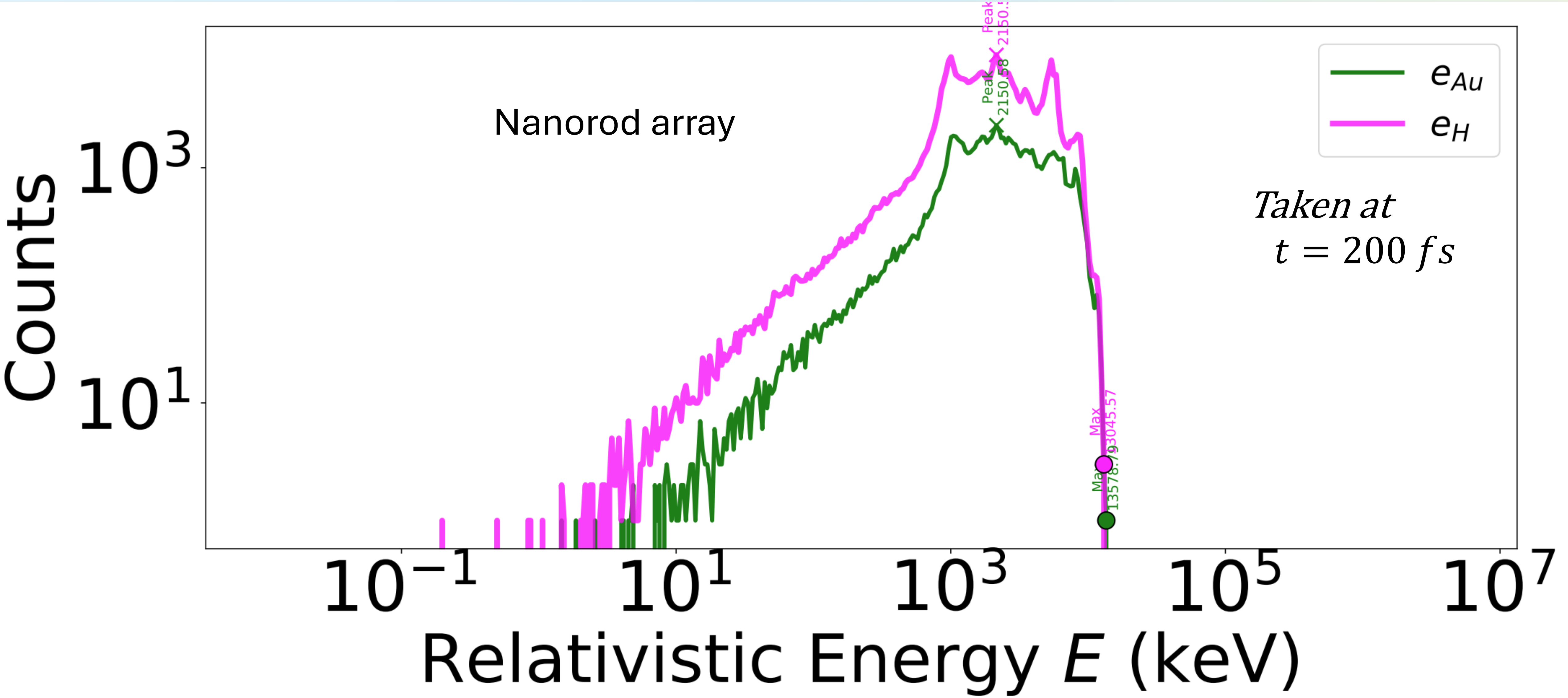
Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\, W/cm^2 \quad \Delta t_{pulse} = 21\, fs$



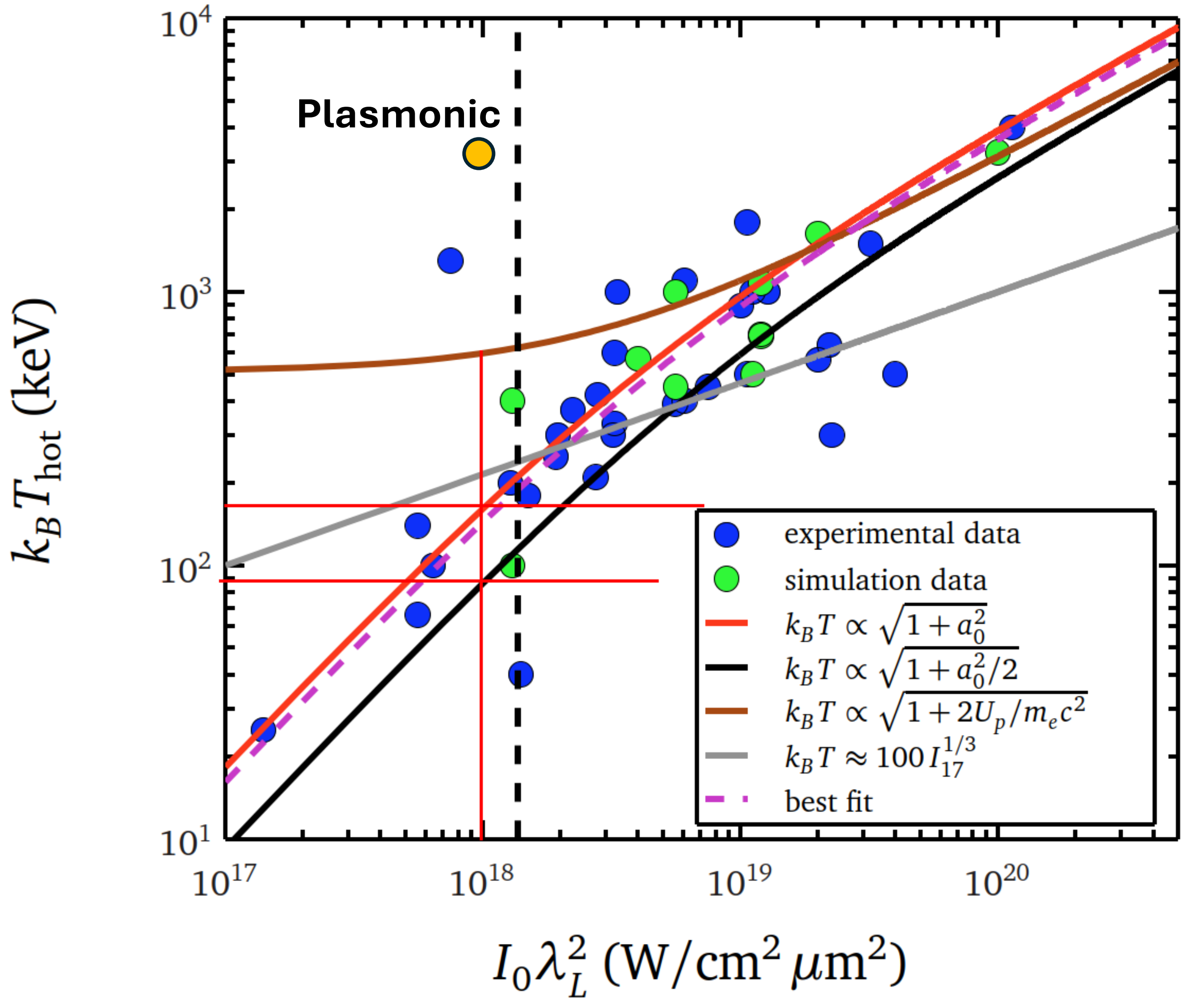
Ion spectra -EPOCH

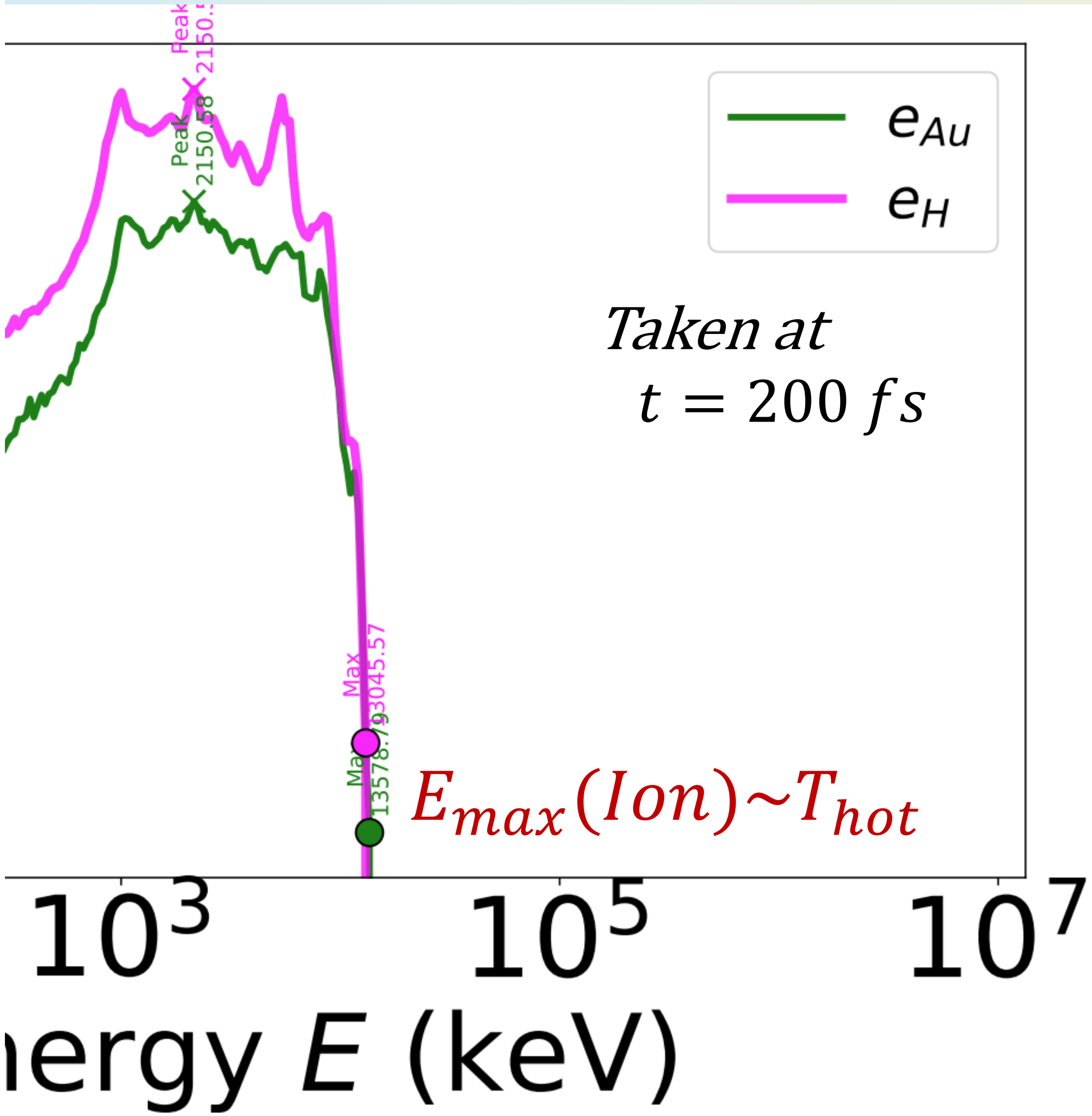
$I = 4.28 \times 10^{18}\ W/cm^2 \quad \Delta t_{pulse} = 21\ fs$



Ion spectra -EPOCH

$I = 4.28 \times 10^{18}\ W/cm^2 \quad \Delta t_{pulse} = 21\ fs$





Taken at
$t = 200\ fs$

$E_{max}(Ion) \sim T_{hot}$