STUDY OF HIGH HARMONIC GENERATION IN HIGH REPETITION RATE SYSTEMS

ULTRAFAST SPECTROSCOPY LABORATORY

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Supervisor
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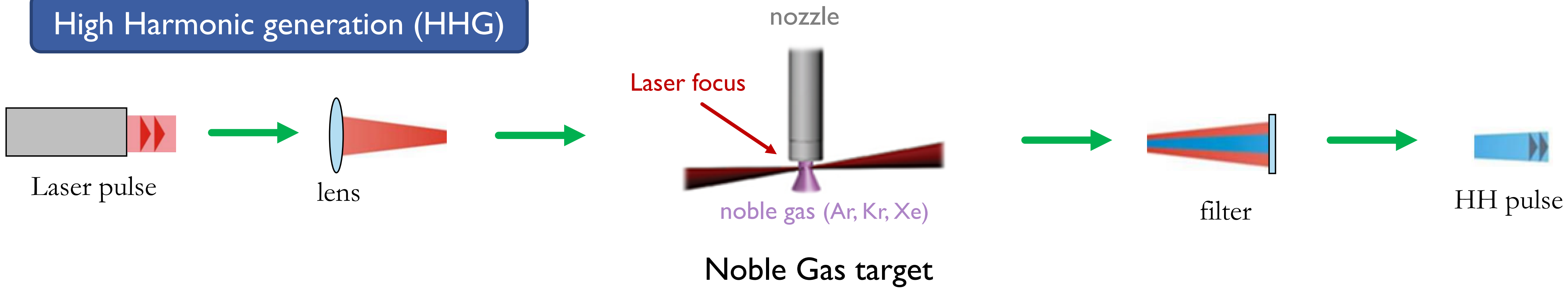
Monday September 4, 2017

INTRODUCTION

High Harmonic generation (HHG)



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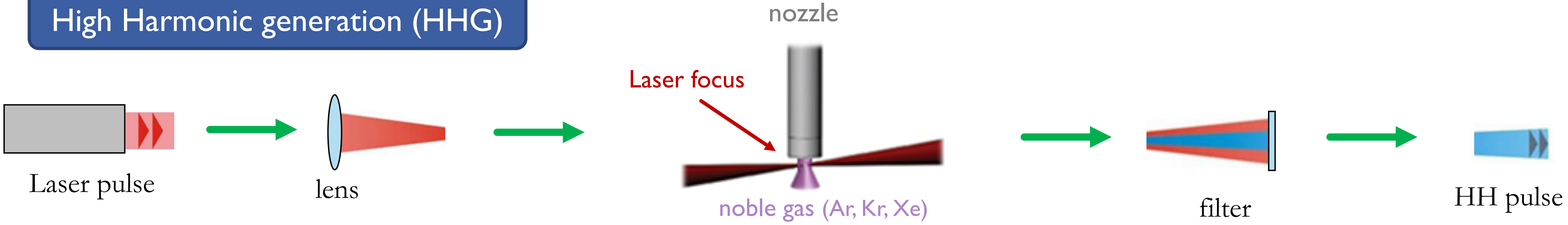

Noble Gas target

- ✓ Create coherent XUV light
- ✓ Use a noble gas target
- ✓ Realization on a table-top
- × Intense laser field required (~ 10^{14} W/cm^2)
- × Highly nonlinear process

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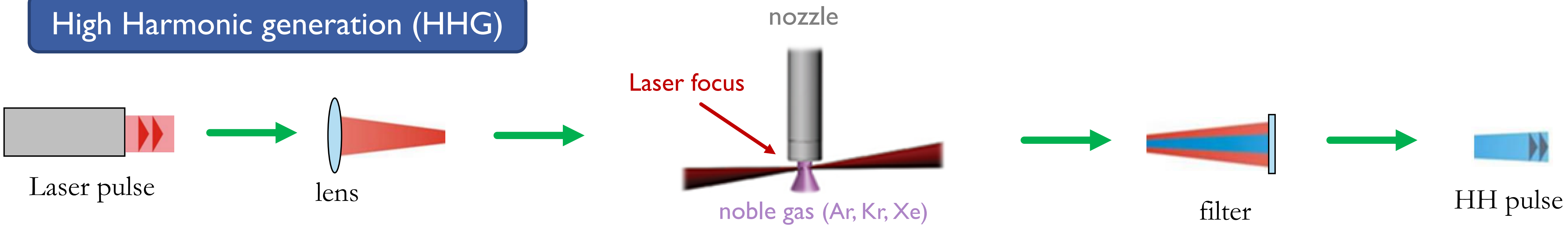

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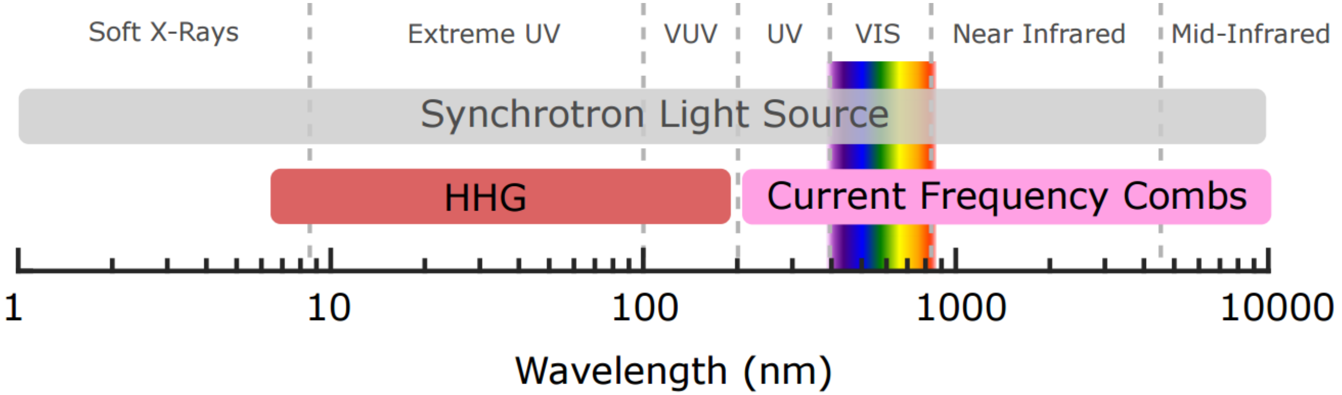
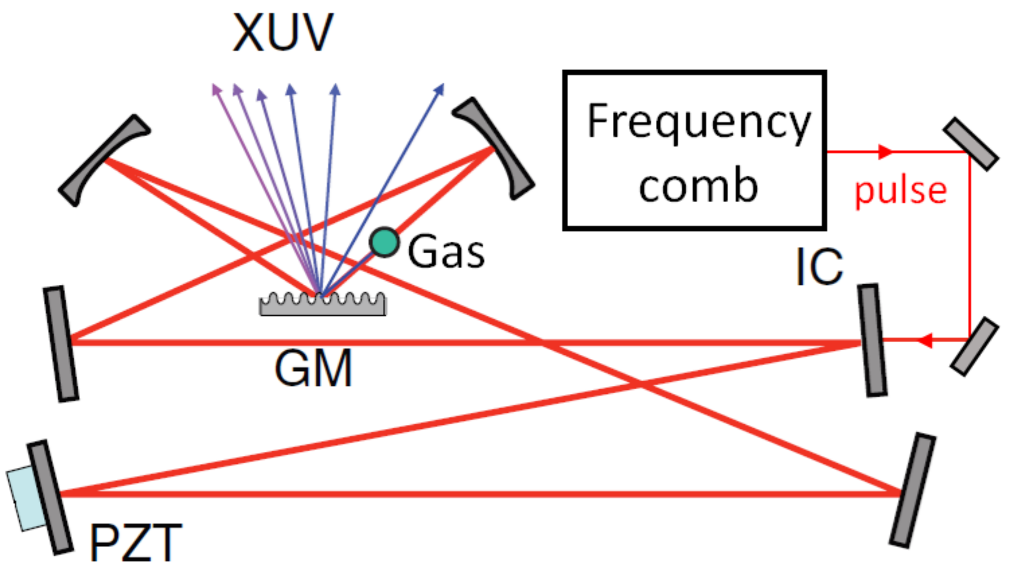


INTRODUCTION

High repetition rate systems

Traditional single pass HHG
1 - 500 kHz
✓ Loose focus
× Low data rates
× High peak power

High repetition rate HHG
10 -100 MHz
✓ High data rates
× Higher plasma density
× Tight focus (less efficient)

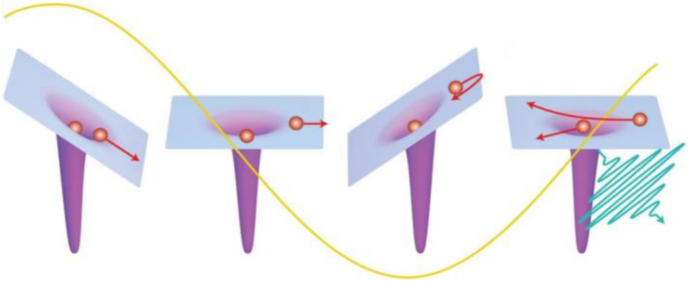


Femtosecond Enhancement Cavity (fsEC)

OUTLINE

I. Theory

Semi-classical model



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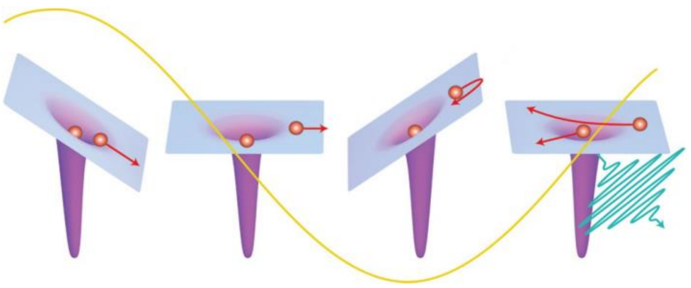
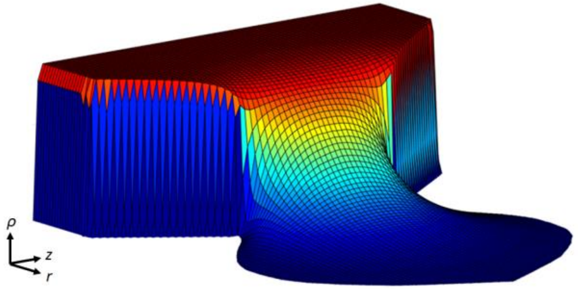


II. Atoms and ions dynamic

Supersonic flow physic

Plasma physic



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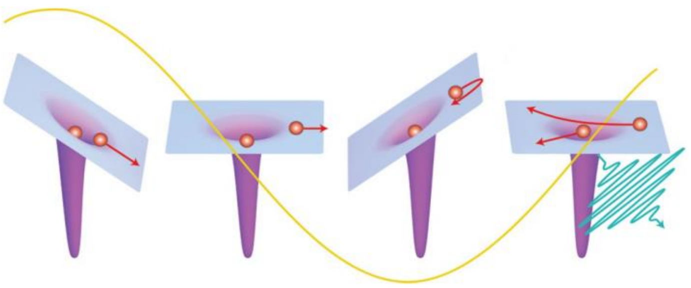
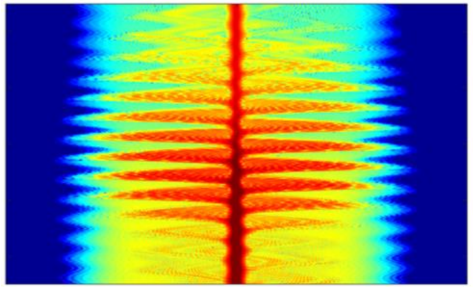


III. Dipole response
    Quantum mechanic



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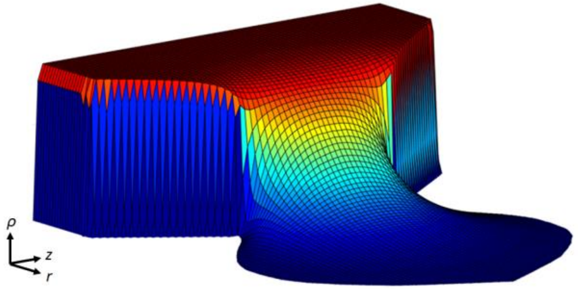

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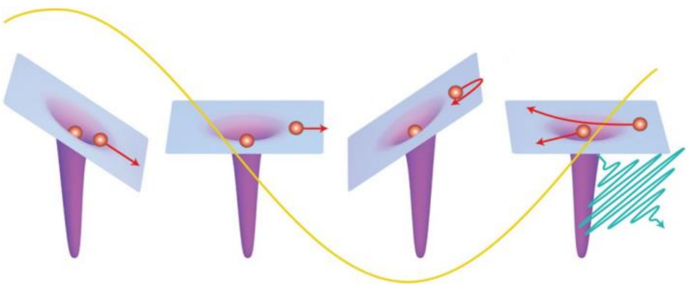

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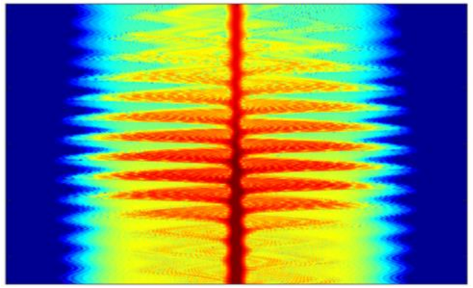

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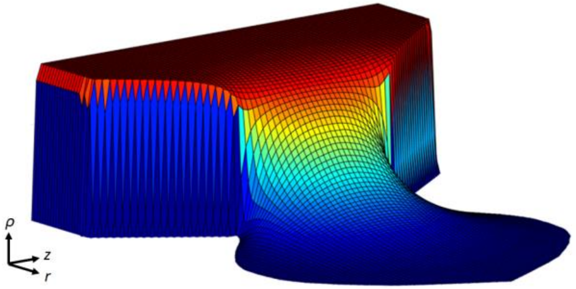
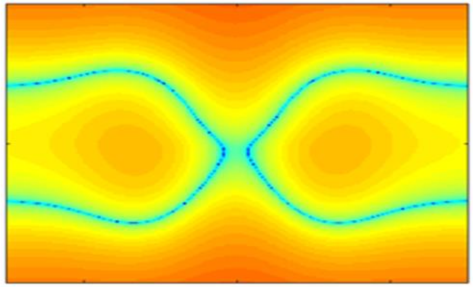


IV. Phase matching

Nonlinear optic



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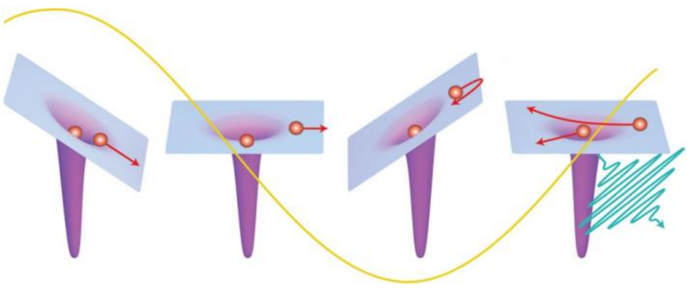

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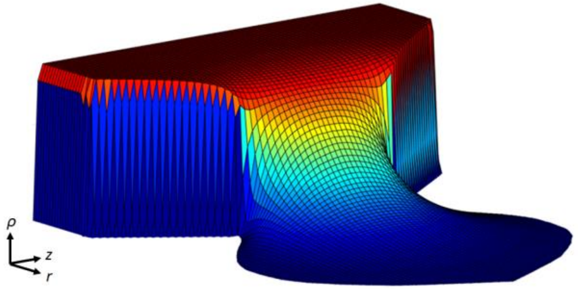

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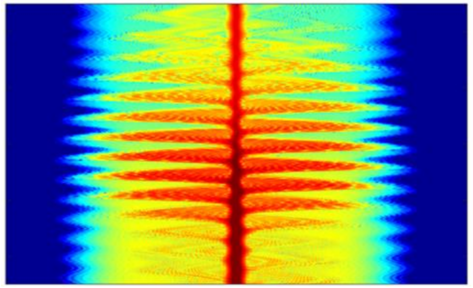

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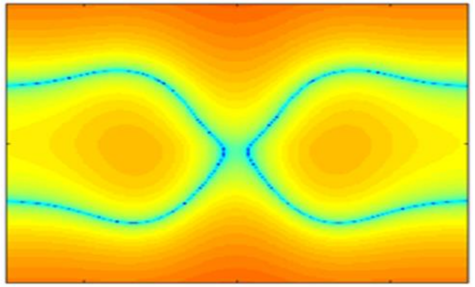
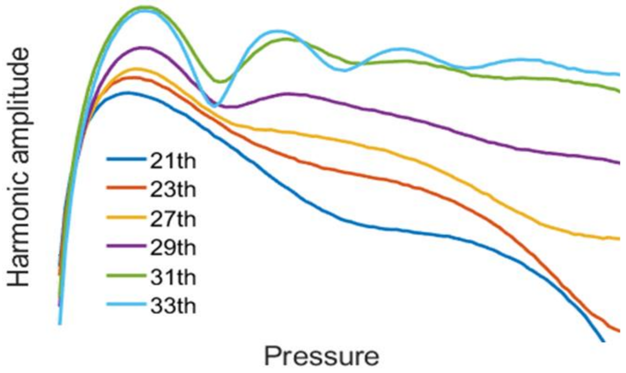


V. Harmonic calculation

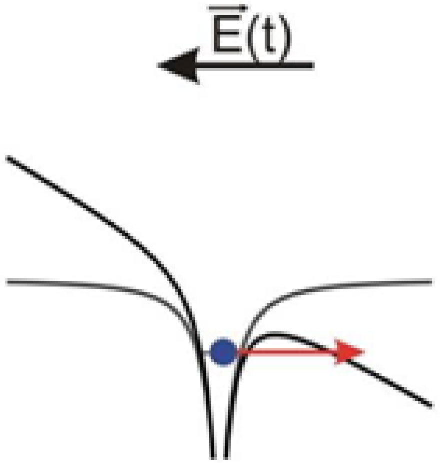
VI. Optimization



I. THEORY

The three step model

Ionization



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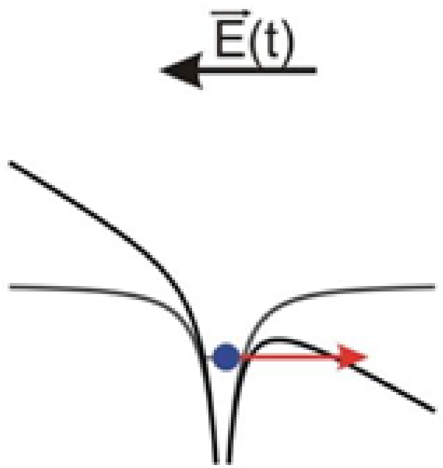
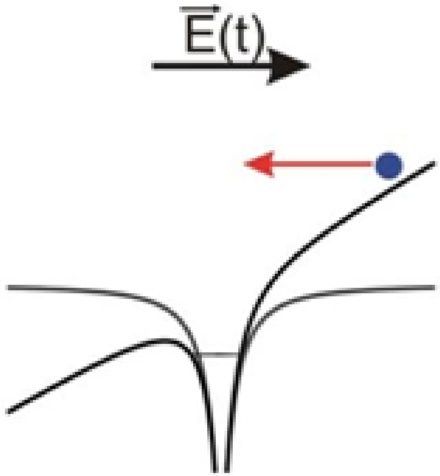


Acceleration



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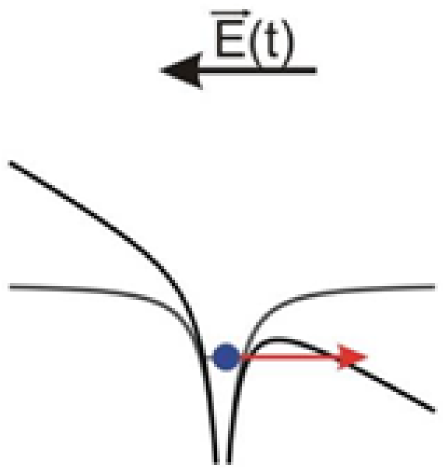

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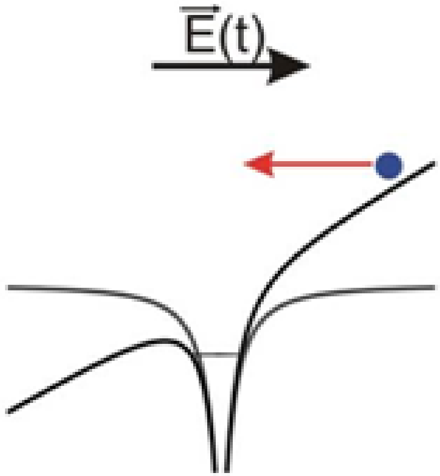

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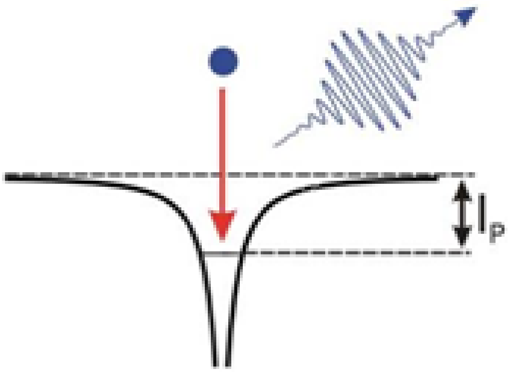

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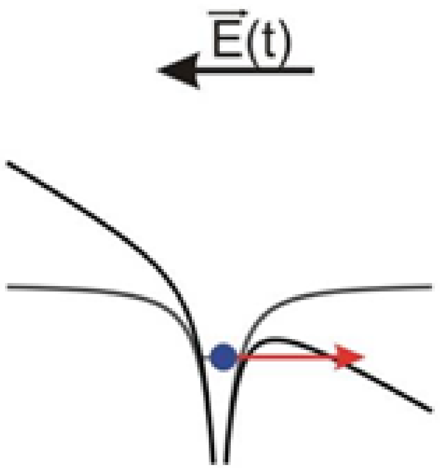

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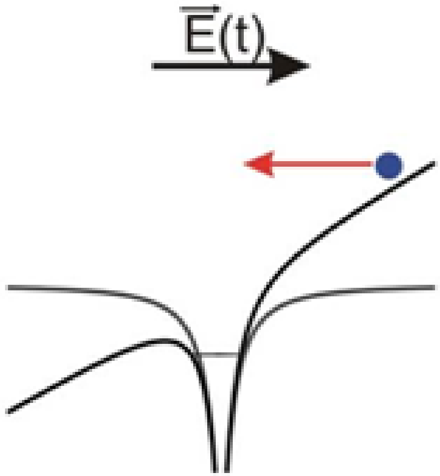

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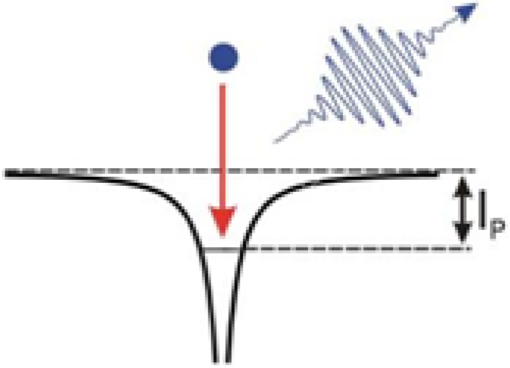


$\omega_q = q\omega_1$

q is an odd number

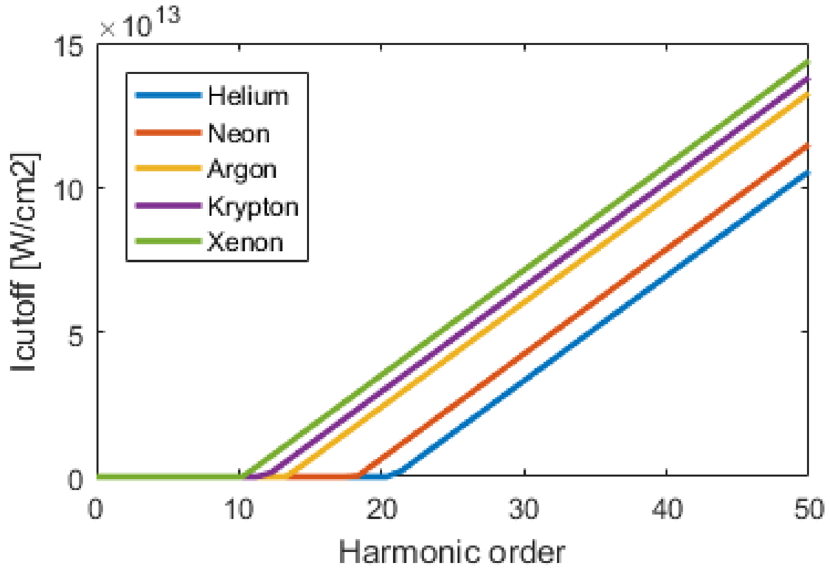
$$E_{q,max} = 3.17U_p + I_p$$

$$U_p \propto I$$

I. THEORY

Differences between gases

Gas	He	Ne	Ar	Kr	Xe
I_p [eV]	24.6	21.6	15.8	14.0	12.1



Required intensity to generate harmonics

I. THEORY

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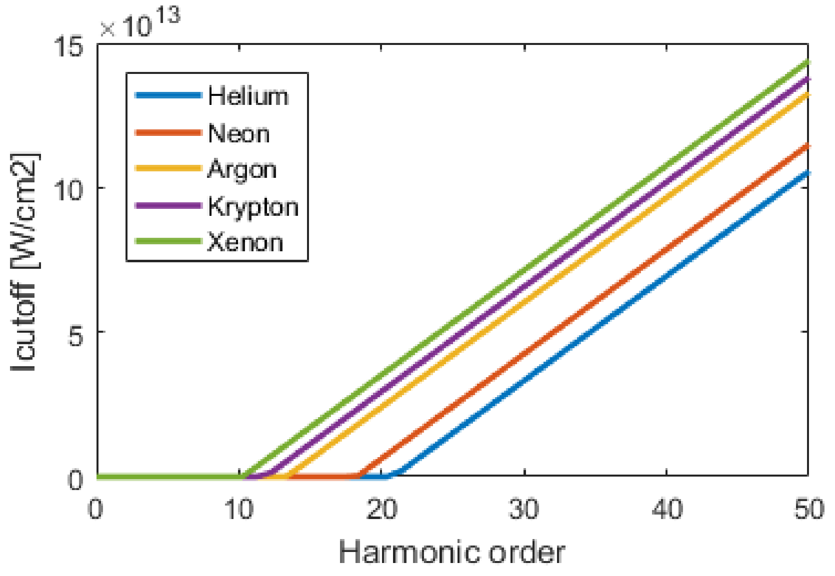
I_p [eV]	24.6	21.6	15.8	14.0	12.1



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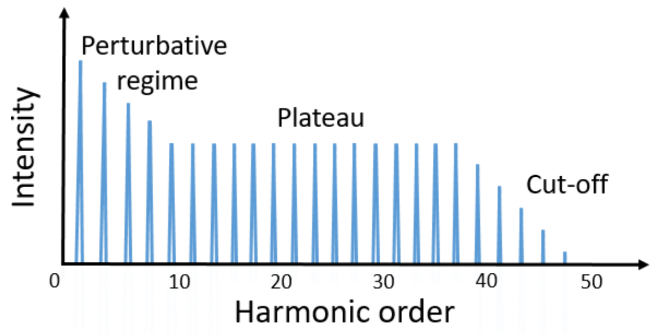
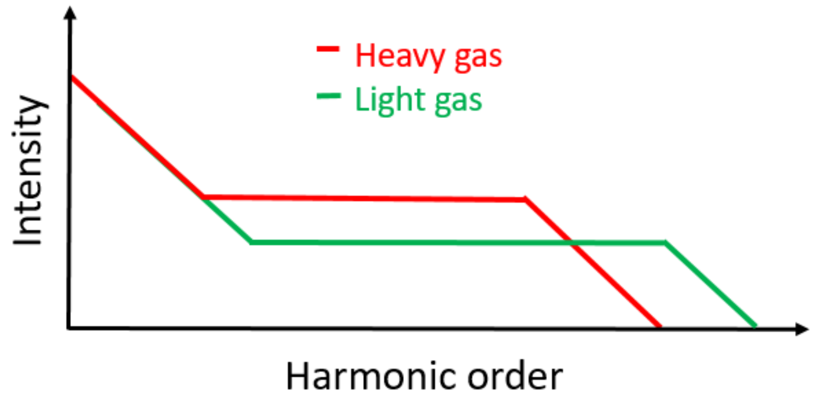
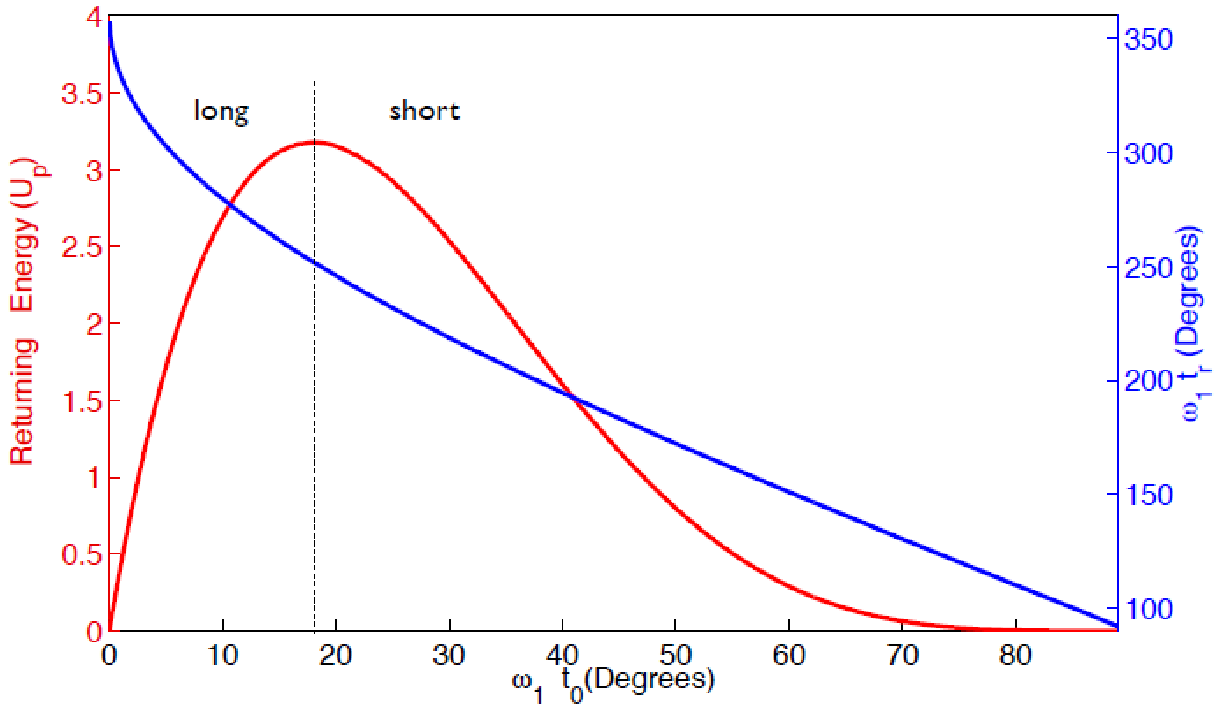


Typical Harmonic spectra



I. THEORY

Long and short trajectories



returning time t_r such that $x(t_r) = 0$

2nd Newton law:

$$m_e \ddot{x}(t) = -eE_o \cos(\omega_1 t)$$

Energy:

$$E = p^2(t_r)/2m_e$$

2 emission times t_0 that give the same impact energy

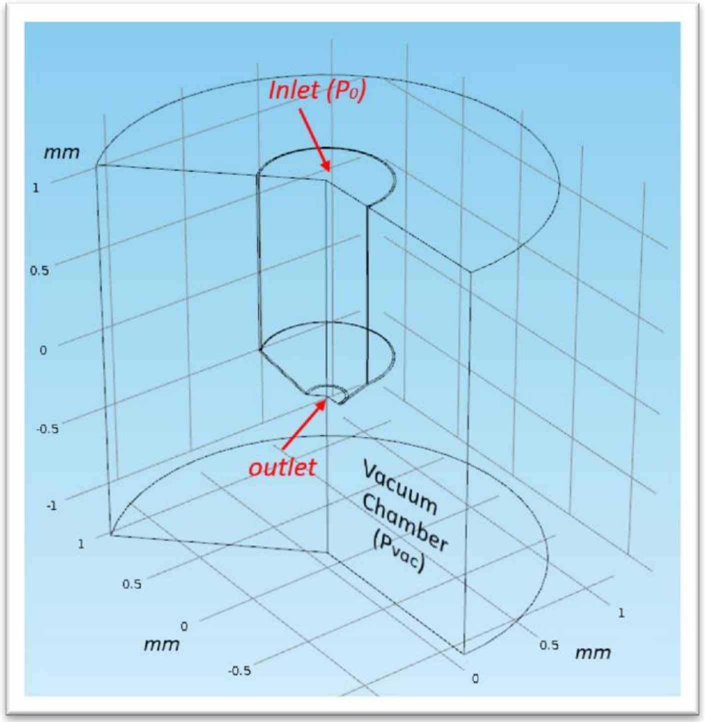
Short trajectory if $t_0 > 18/\omega_1$

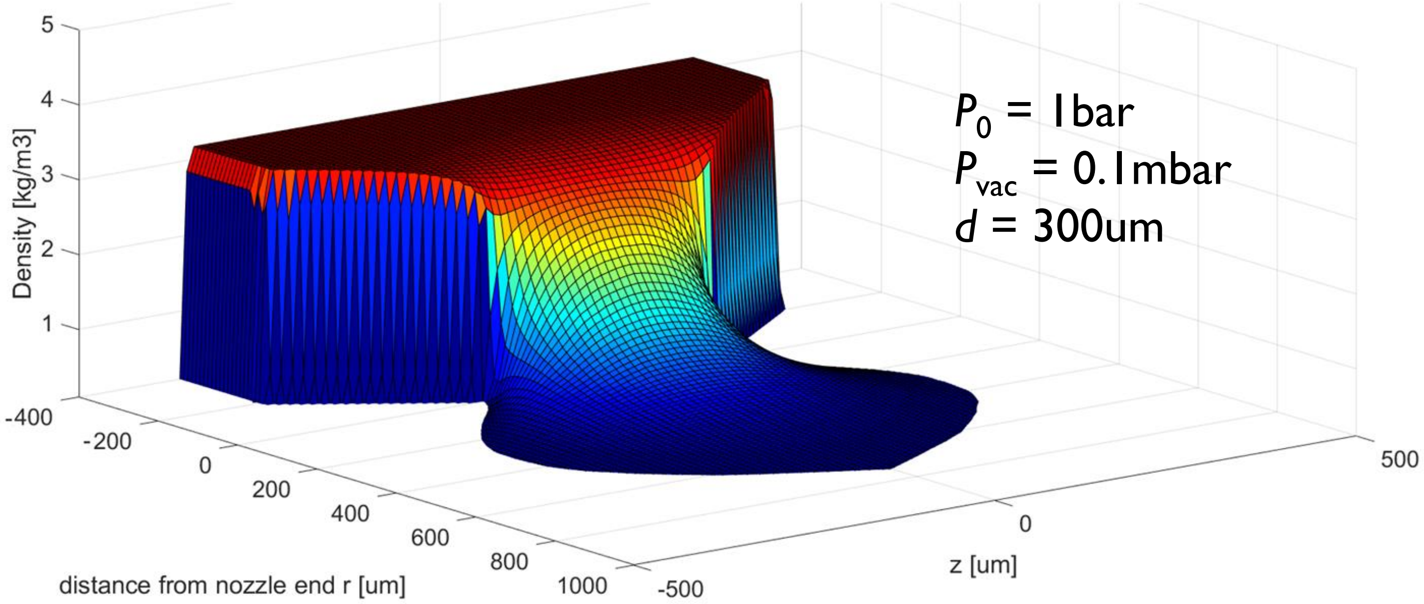
Long trajectory if $t_0 < 18/\omega_1$

II. ATOMS AND IONS DYNAMIC

Density distribution

COMSOL simulations:



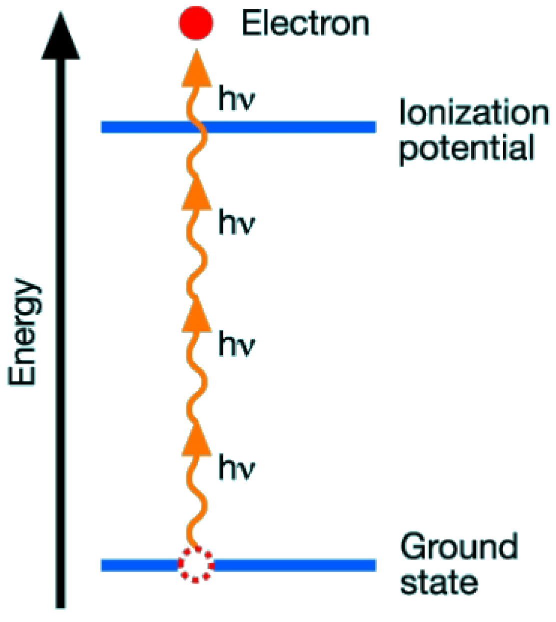


P_0 = 1bar
P_{vac} = 0.1mbar
d = 300um

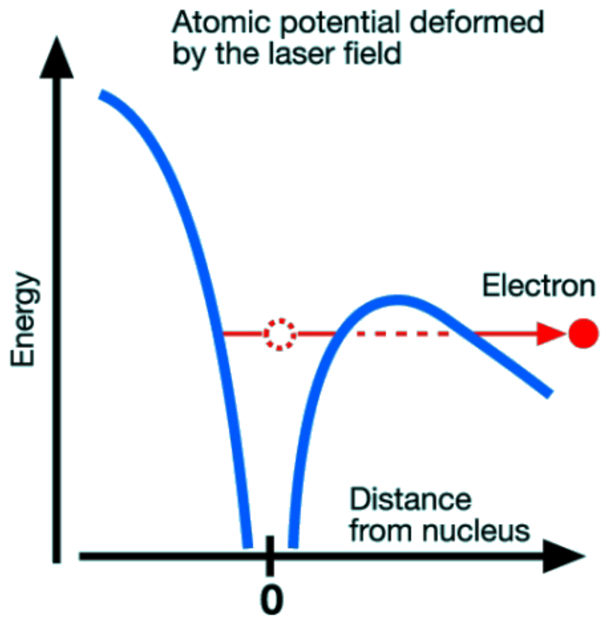
The density drop is very fast (80% drop at 400um), it is hard to focus at the right spot
→ uncertainty on the pressure

II. ATOMS AND IONS DYNAMIC

Ionization process



Classical ionization



Tunnel ionization

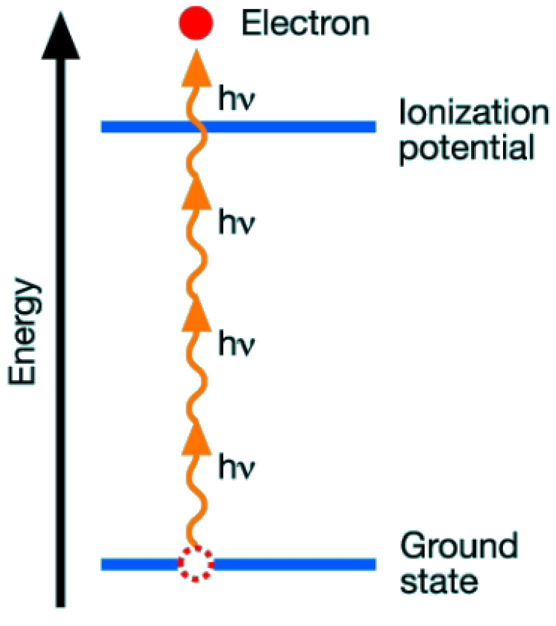
II. ATOMS AND IONS DYNAMIC

Ionization process

$$\gamma = \sqrt{\frac{I_p}{2U_p}}$$

$\gamma \gg 1$ Classical dominate

$\gamma \ll 1$ Tunnelling dominate



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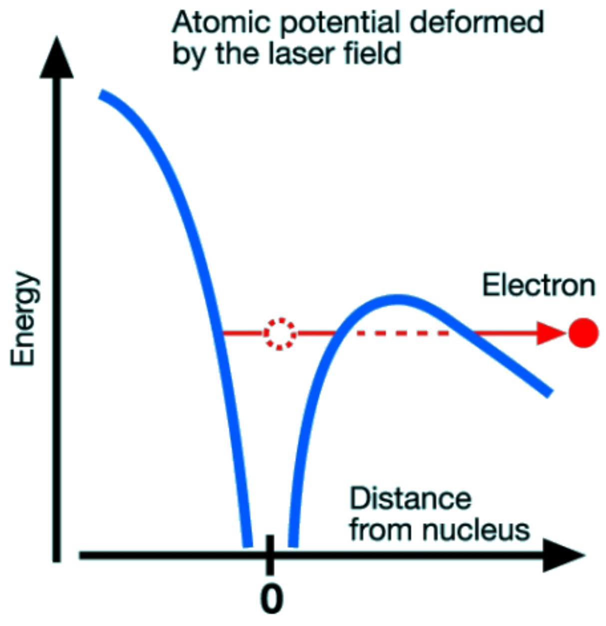

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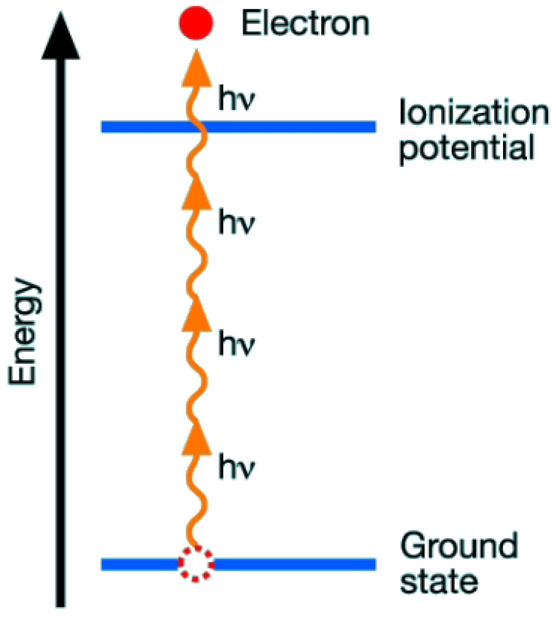

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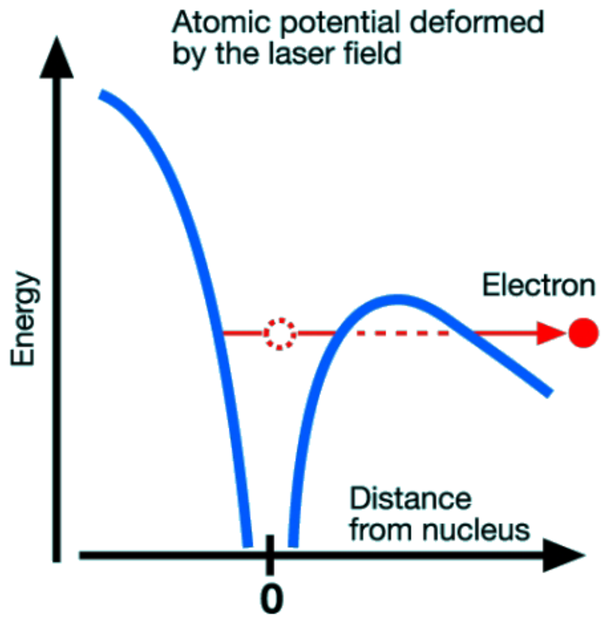

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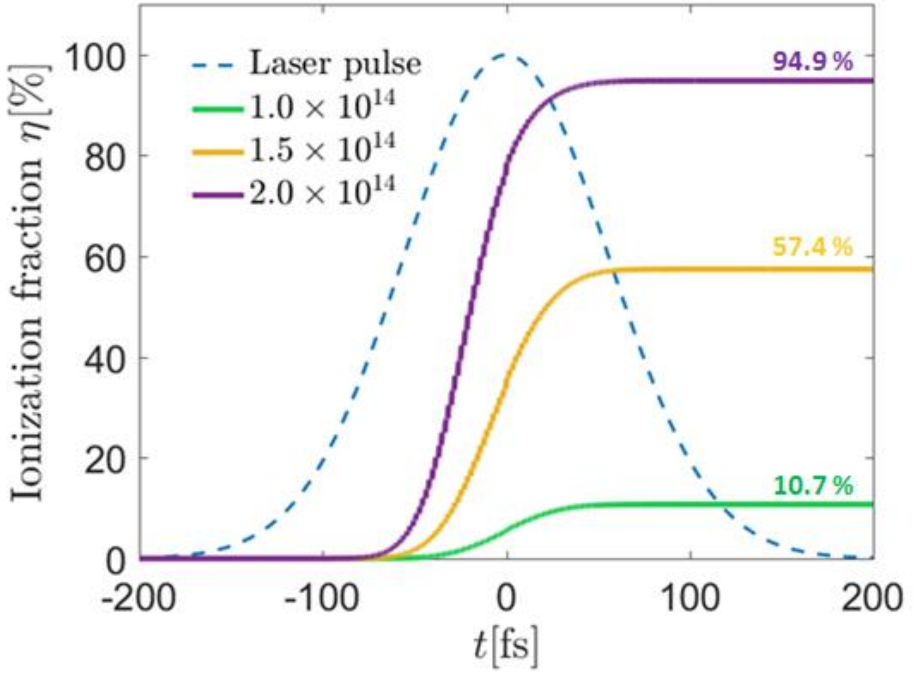
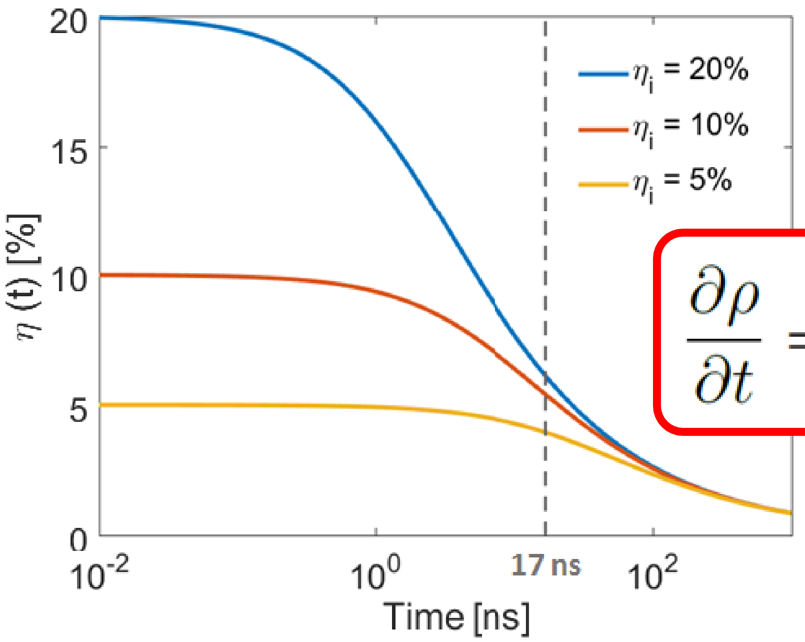


II. ATOMS AND IONS DYNAMIC

Ions recombination

$$A^+ + e + e \rightarrow A^* + e$$



$$\boxed{\frac{\partial \rho}{\partial t} = -\Gamma_o \rho^3}$$

Ions recombination for different initial ionization fraction

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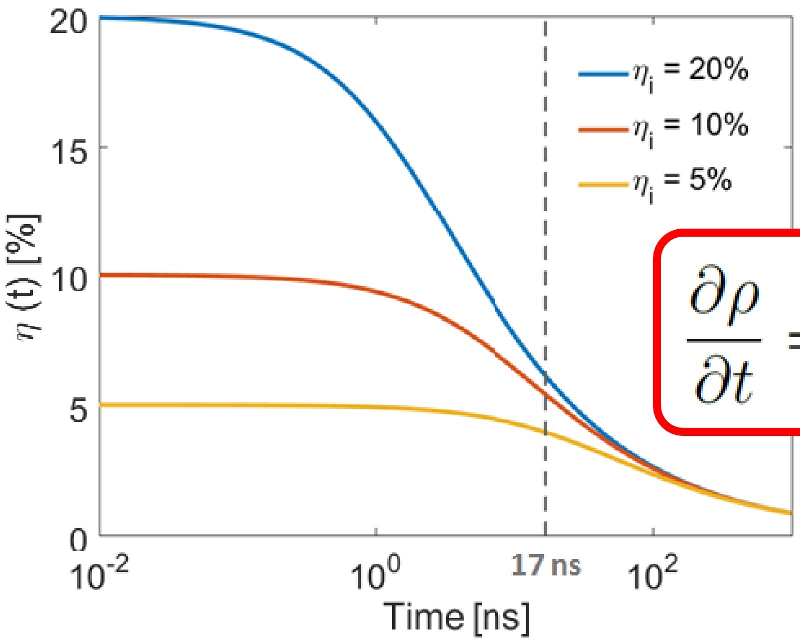

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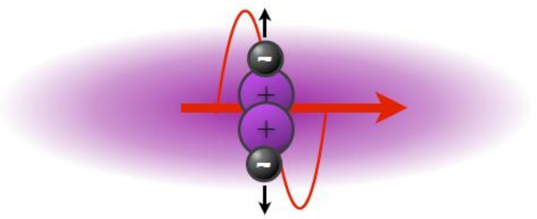
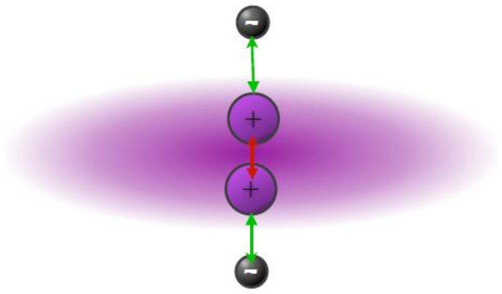
Ions recombination for different initial ionization fraction

Ambipolar diffusion



Freed electrons are moving away

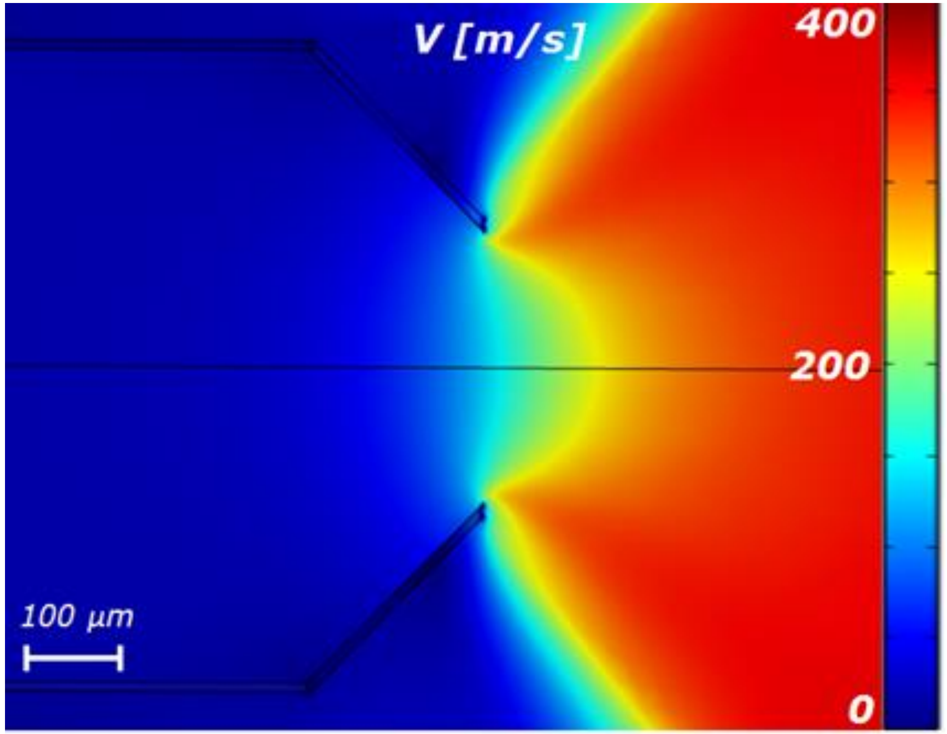


Ions repulsion and electron-ion attraction

$$\boxed{\frac{\partial \rho}{\partial t} = -D_\alpha \frac{\partial^2 \rho}{\partial r^2}} \qquad D_\alpha = D_i \left(1 + \frac{T_e}{T_i}\right)$$

II. IONS AND ATOMS DYNAMIC

Supersonic expansion



Velocity distribution at the nozzle outlet

II. IONS AND ATOMS DYNAMIC

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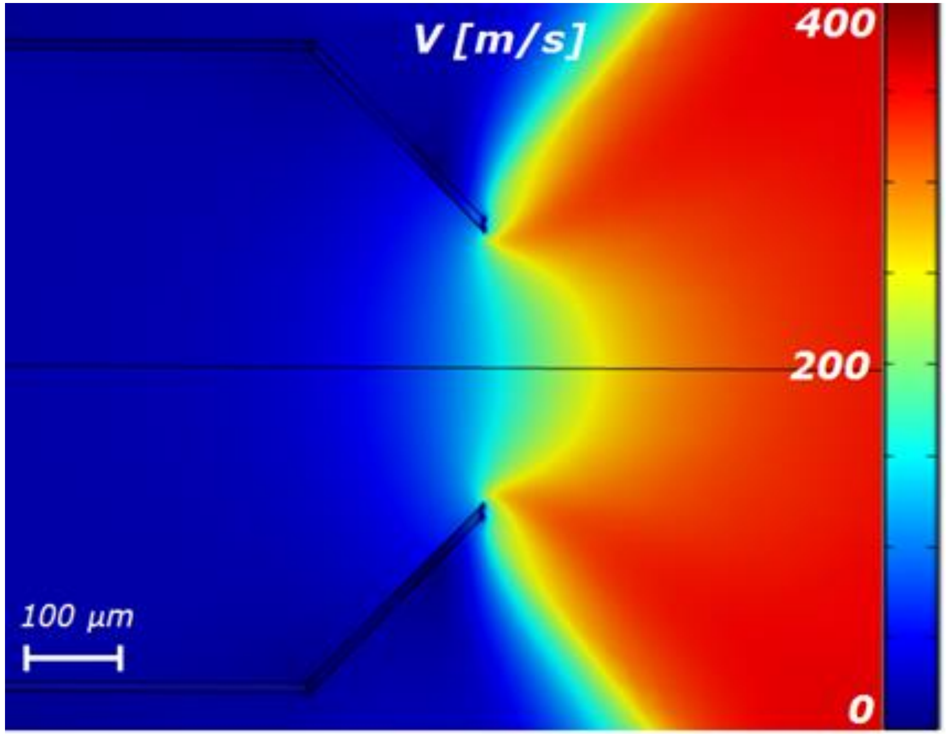

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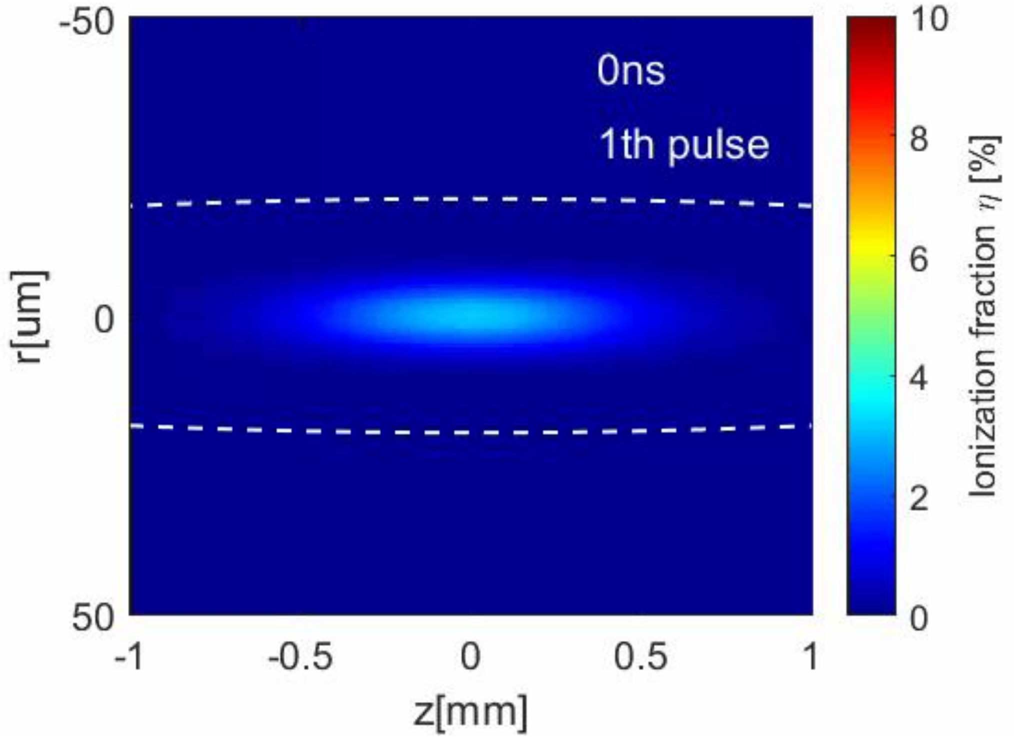
Simulation result

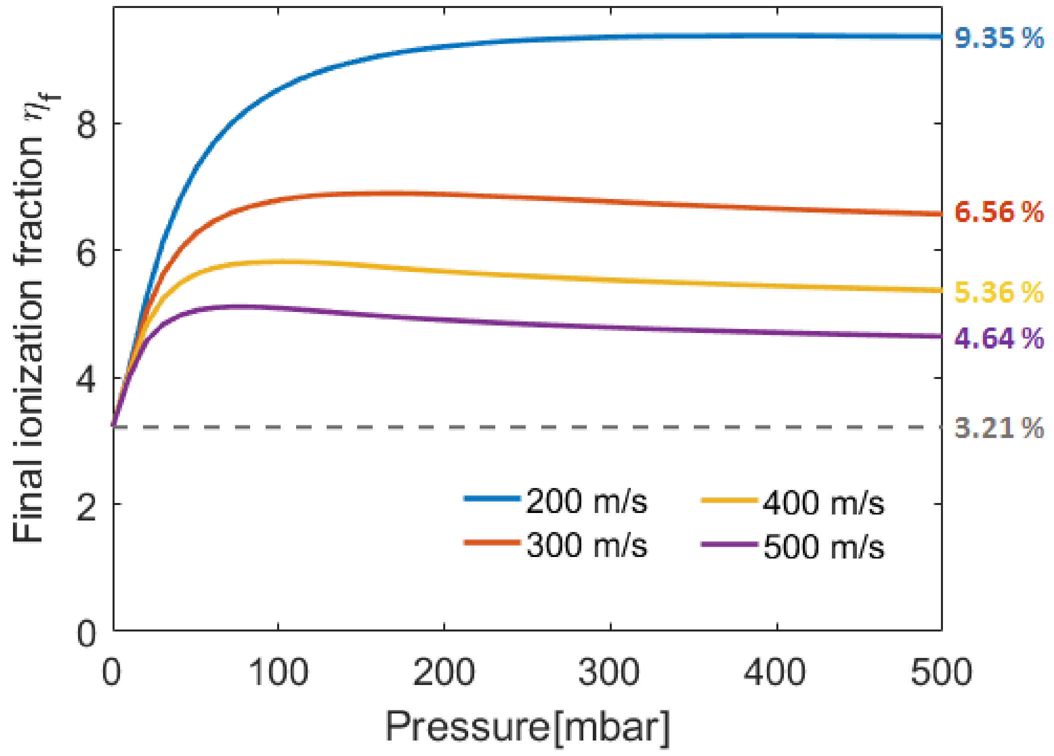


Ions recombination and diffusion through laser pulses

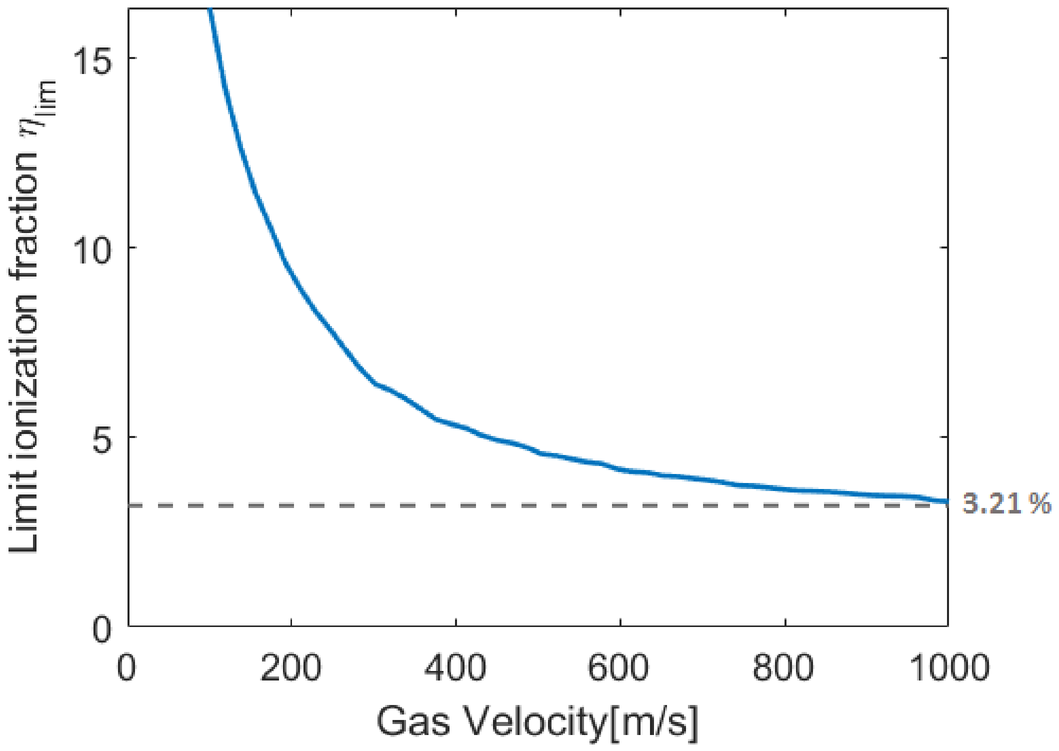
$I = 8 \times 10^{13}$ W/cm^2, P = 500 mbar, V = 250 m/s

II. IONS AND ATOMS DYNAMIC

The importance of gas velocity



Ionization fraction after 16 pulses as a function of pressure for different gas velocity



Ionization at 500 mbar as a function of gas velocity

III. DIPOLE RESPONSE

Ground state valence electron subjected to a potential $V = V_{atom} + V_{field}$

$$V_{\text{field}}(x,t) = -exE_o f(t)\cos(\omega_1 t)$$ *Oscillating potential of the driving field*

$$V_{\text{atom}}(x) = -\frac{1}{4\pi\varepsilon_o}\frac{e^2}{\sqrt{x^2+X_o^2}}$$ *Coulomb potential of the atom*

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Schrödinger equation:

$$i\hbar\frac{\partial\psi(x,t)}{\partial t} = \frac{p^2}{2m_e}\psi(x,t) + V(x,t)\psi(x,t)$$

Solving with split-step Fourier method :

$$\begin{cases} \phi(x,t+\mathrm{d}t) = & \mathcal{F}\left(\psi(k,t)e^{-\frac{i}{\hbar}V(x,t)dt}\right) \\ \psi(k,t+\mathrm{d}t) = & \mathcal{F}^{-1}\left(\phi(x,t+\mathrm{d}t)e^{-\frac{i\hbar}{2m_e}k^2 dt}\right) \end{cases}$$

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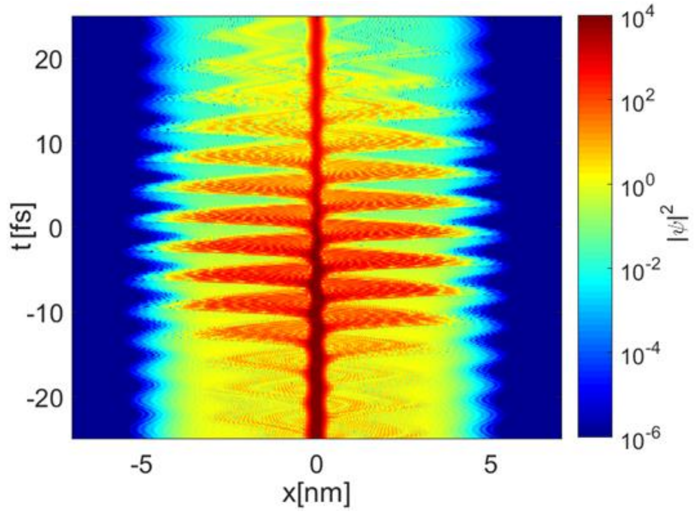


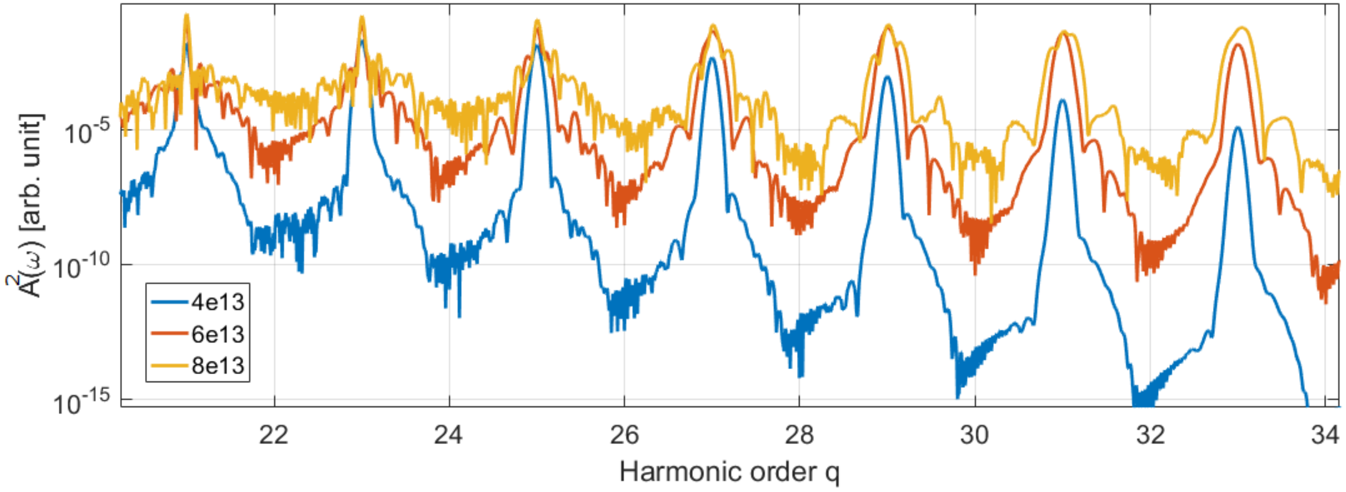
Time evolution of the electron wave-function in the space domain during the laser pulse.

III. DIPOLE RESPONSE

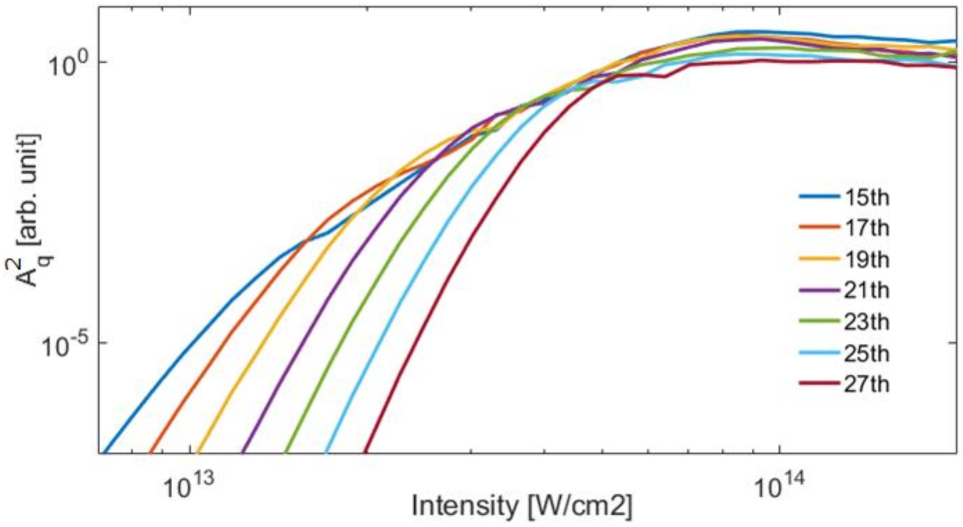
Dipole spectrum

$$d(t) \propto \langle \psi(x,t) \left| x \right| \psi(x,t) \rangle$$

$$A(\omega) = \left| \mathcal{F}(d)(\omega) \right|$$



Dipole spectrum for different peak intensities



Harmonics dipole amplitude as a function of intensity

IV. PHASE MATCHING

→ As the fundamental and harmonic field propagate, their phases can become mismatched.
→ The phase mismatch limits the efficiency of harmonic generation.

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Index modification by neutral atoms and plasma

$$n(z,r,t,\omega) = 1 + \boxed{\frac{P(z,r)}{P_{atm}}}\left(\left(1 - \boxed{\eta(z,r,t)}\right)\delta(\omega) - \boxed{\eta(z,r,t)}\frac{N_{atm}e^2}{2\omega^2 m_e \varepsilon_o}\right)$$

IV. PHASE MATCHING

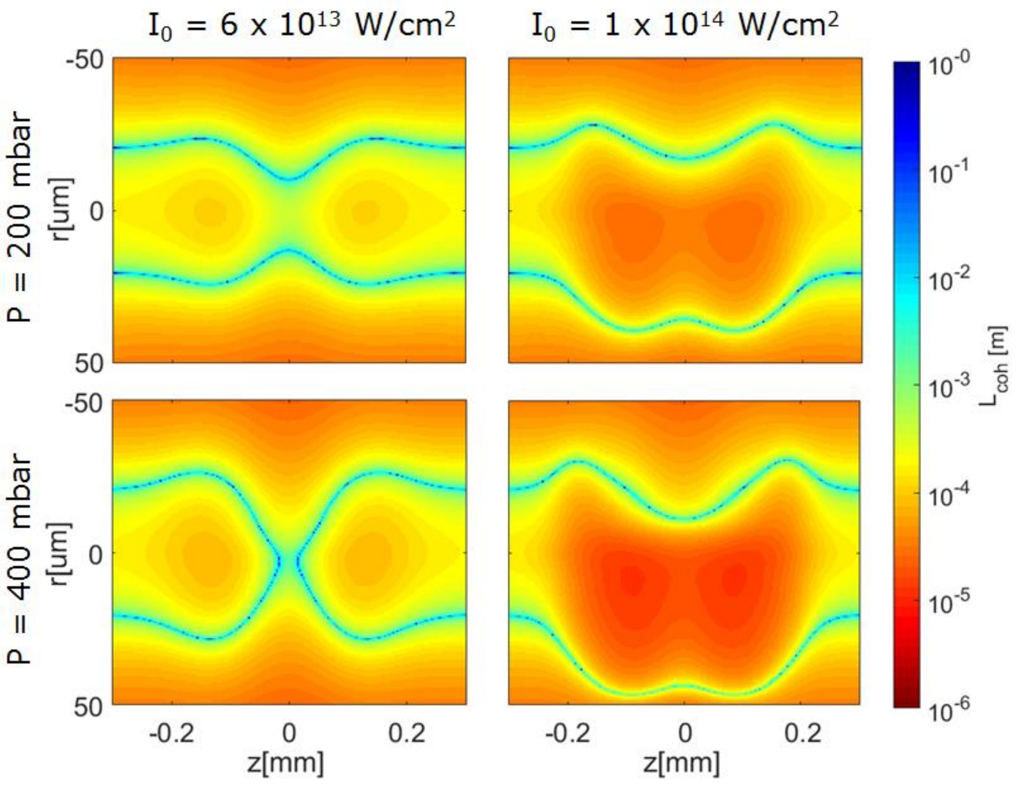
$$\Delta\phi = q\phi_1 - \phi_q$$

$$q\phi_1 = qk_1 n(\omega_1)z + q\phi_{\text{foc}}(\omega_1) + \Phi_{\text{dipole}} - q\omega_1 t$$

$$\phi_q = k_q n(\omega_q)z + \phi_{\text{foc}}(\omega_q) - \omega_q t$$

$\Delta k = |\vec{k}_q| - q|\vec{k}_1|$ with $\vec{k} = \vec{\nabla}\phi$

$\boxed{L_{\text{coh}} = \frac{\pi}{\Delta k}}$ *(a long coherence length correspond to a good phase matching)*



Coherence length for different pressure and intensities

IV. PHASE MATCHING

Phasematching at the focus :

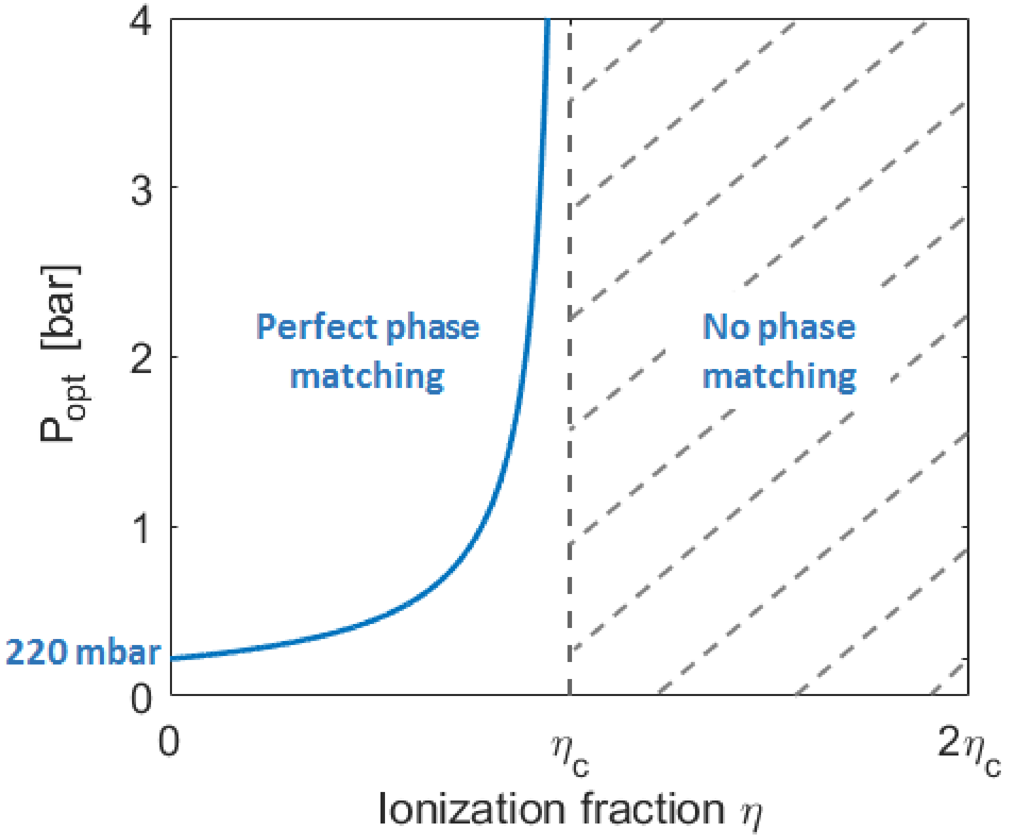
$$\Delta k(0,0) = k_q \frac{P}{P_{\mathrm{atm}}} \Delta\delta \left(1 - \frac{\eta}{\eta_c}\right) - \frac{q}{z_R}$$

Perfect phasematching ($\Delta k = 0$)
=> infinite coherence length

$$P_{\mathrm{opt}} = \frac{P_{\mathrm{atm}}}{k_1 z_R \Delta\delta \left(1 - \frac{\eta}{\eta_c}\right)}$$

Gas	Ar	Kr	Xe
η [%]	4,1	6,1	6,4

Critical ionization fraction for the 21rd harmonic



V. HARMONIC CALCULATION

$$\boxed{\frac{\partial E_o(z)}{\partial z} = -\frac{\rho(z)\sigma}{2}E_o(z) + i\frac{\mu_o c\omega_q}{2}A_q(z)\rho(z)[1-\eta(z)]e^{i\Delta\phi(z)}}$$

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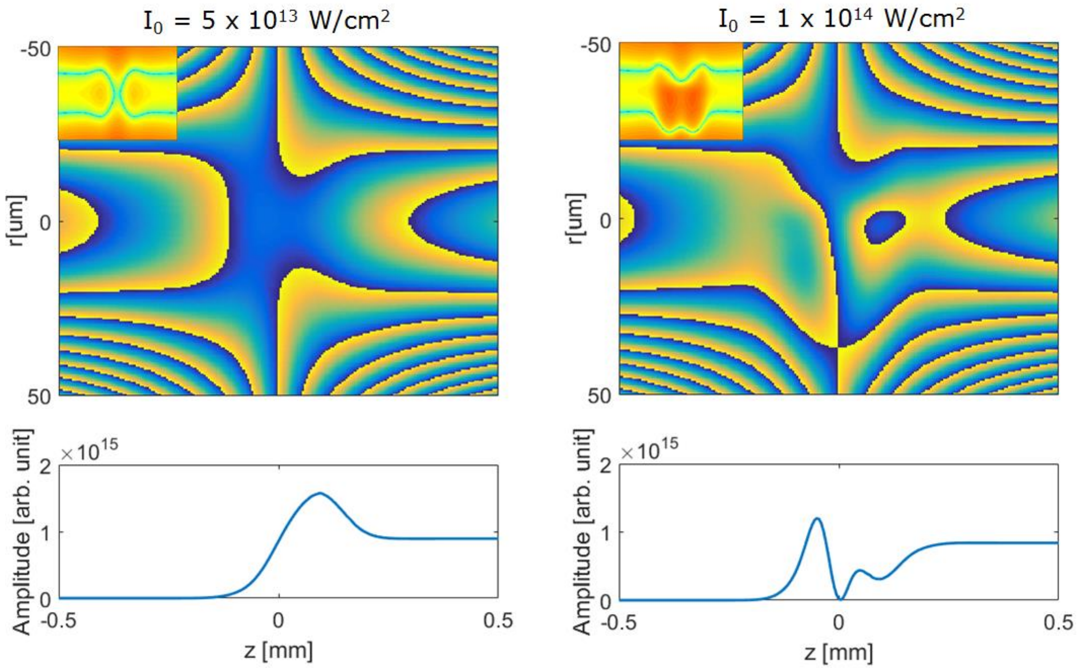
The importance of phase matching



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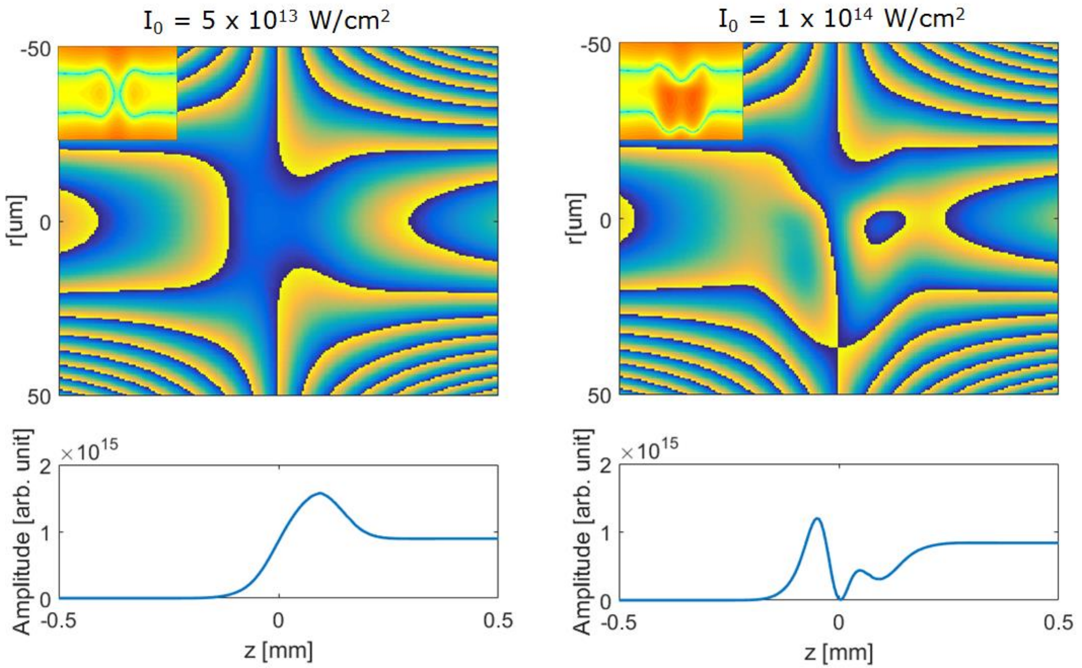
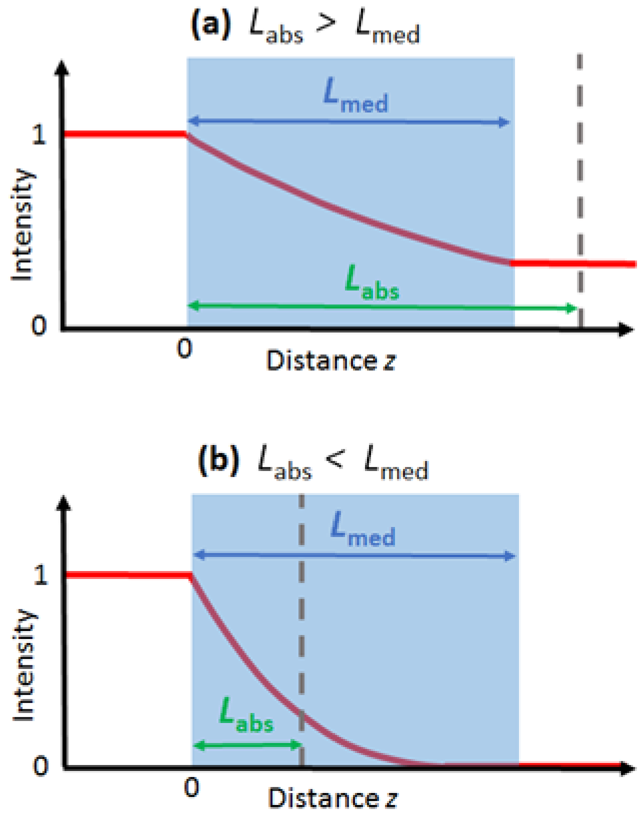
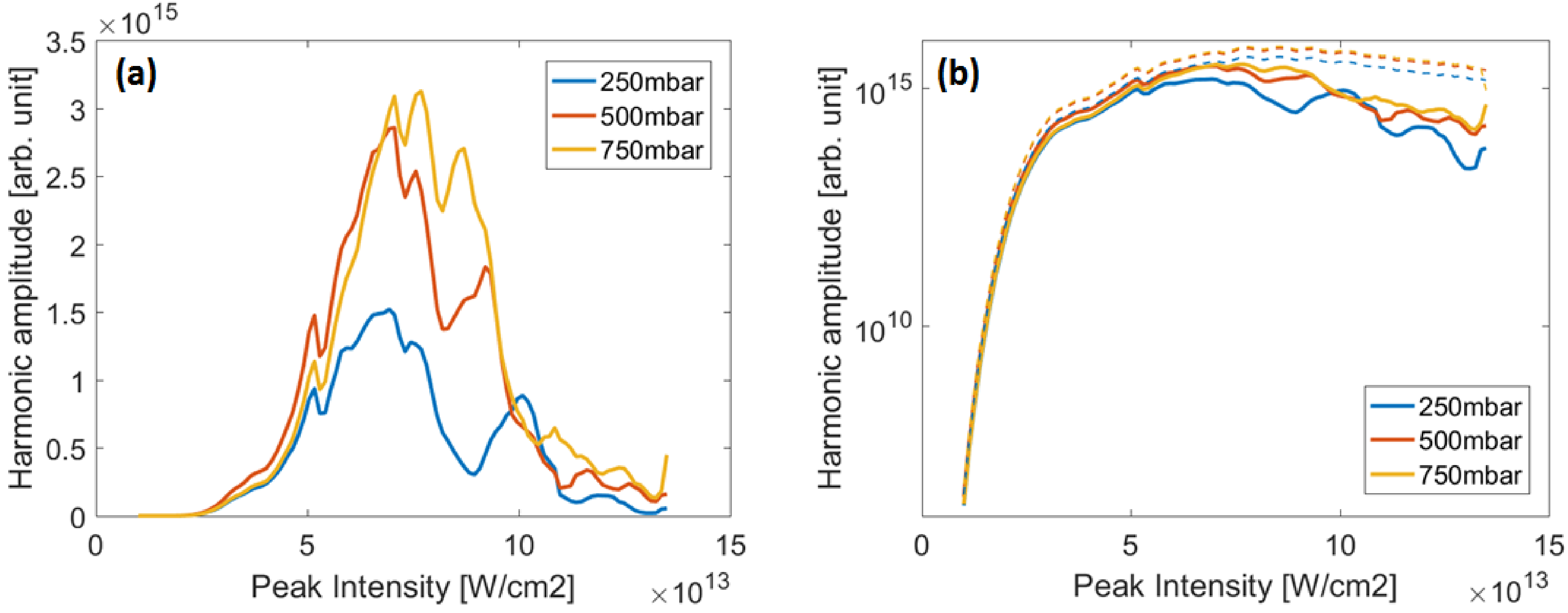


The importance of absorbtion



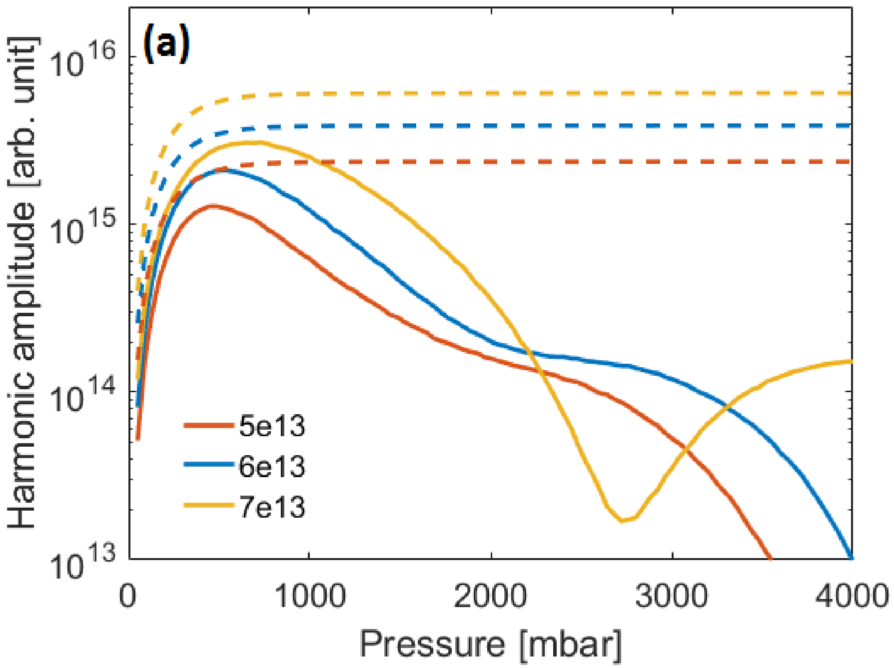
V. HARMONIC CALCULATION

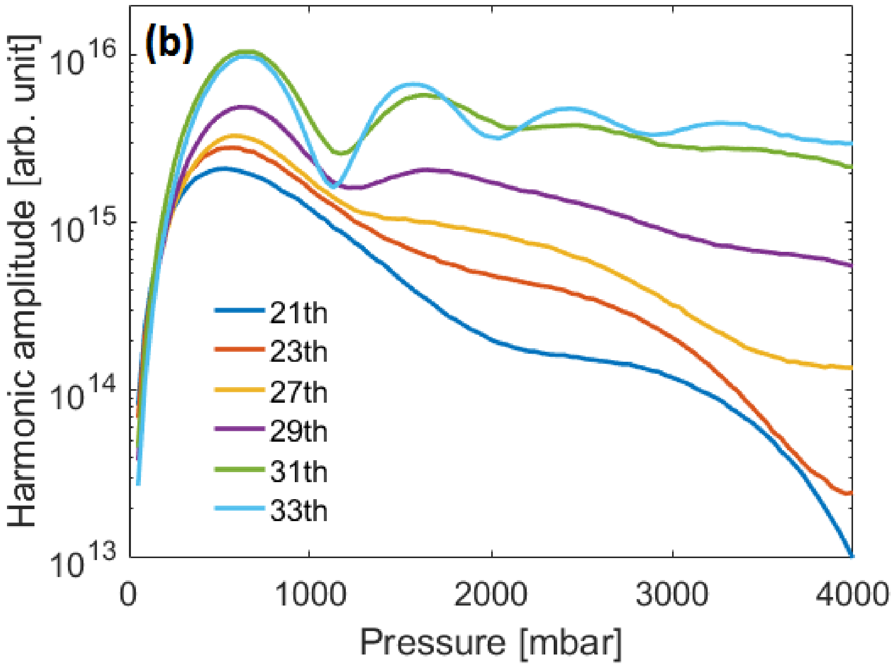
Scaling with the peak intensity



V. HARMONIC CALCULATION

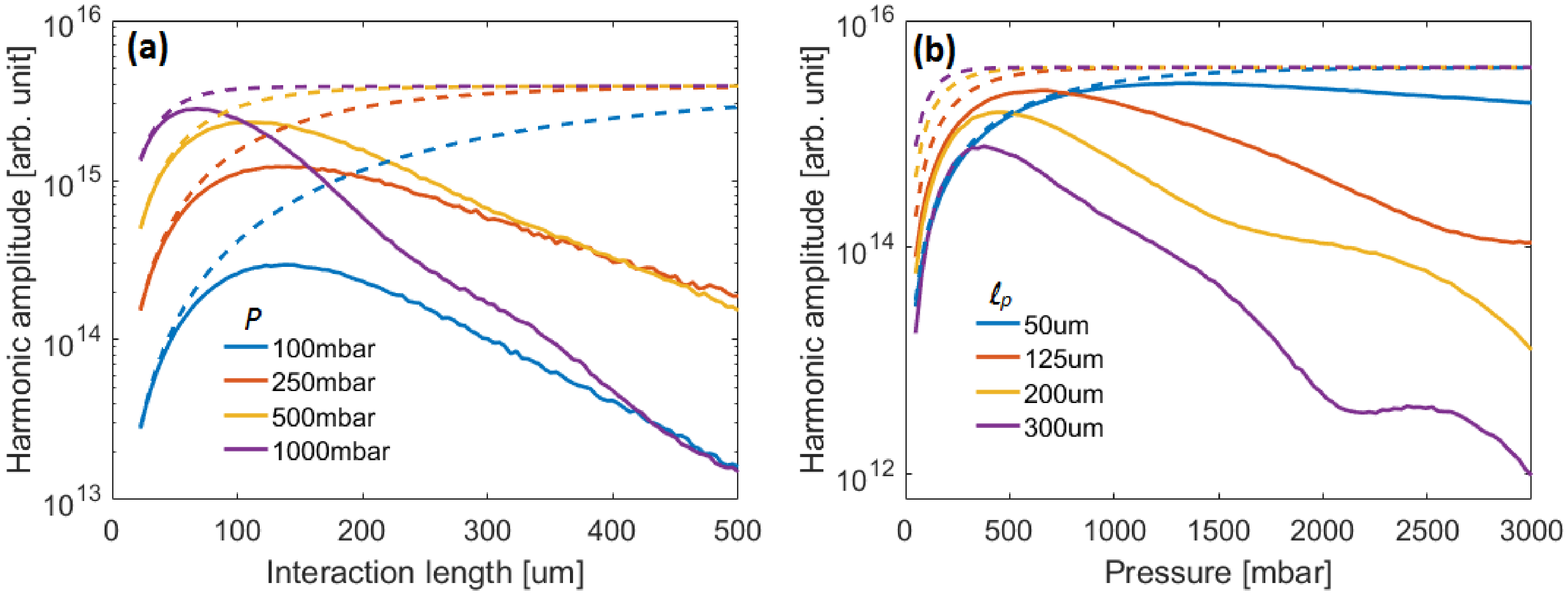
Scaling with the gas pressure





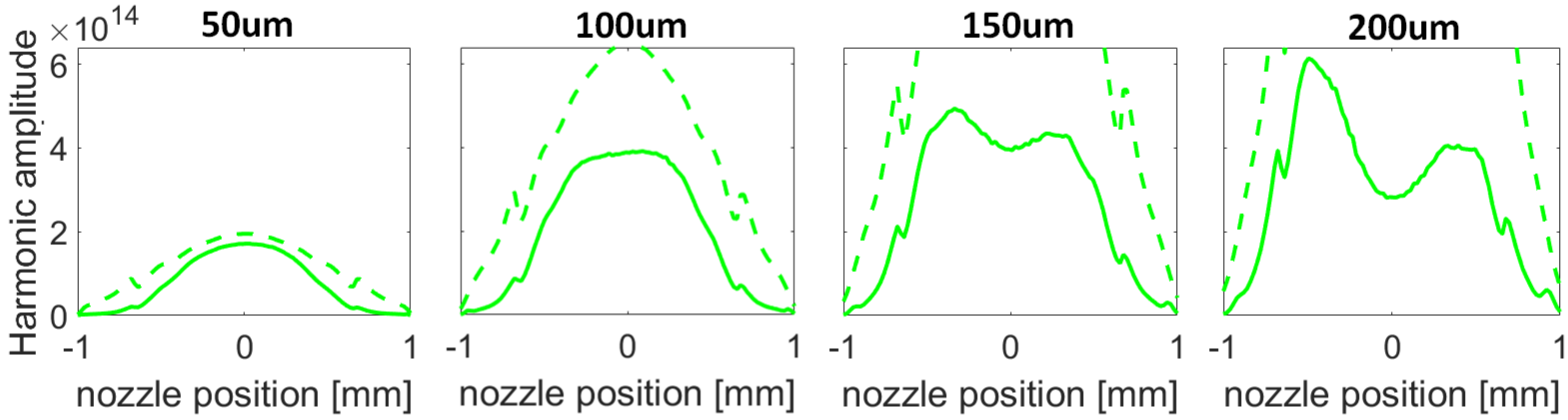
V. HARMONIC CALCULATION

Scaling with the interaction length



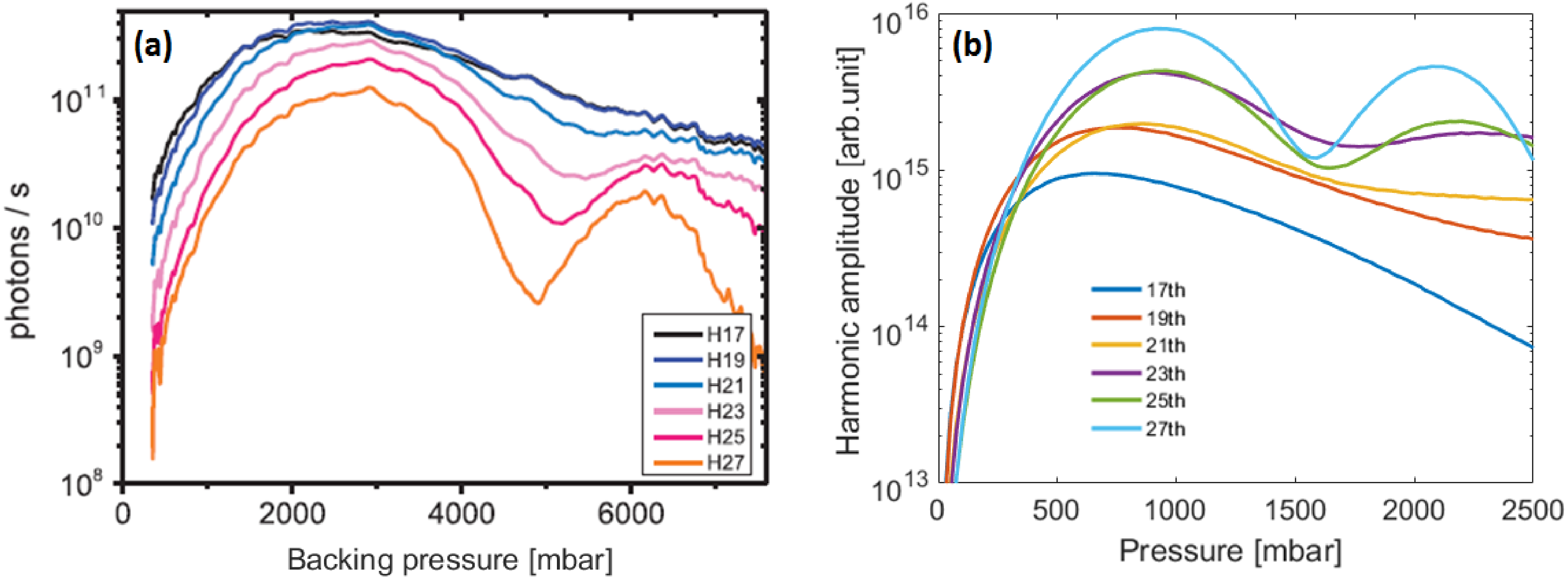
V. HARMONIC CALCULATION

Nozzle position dependance for different nozzle diameter



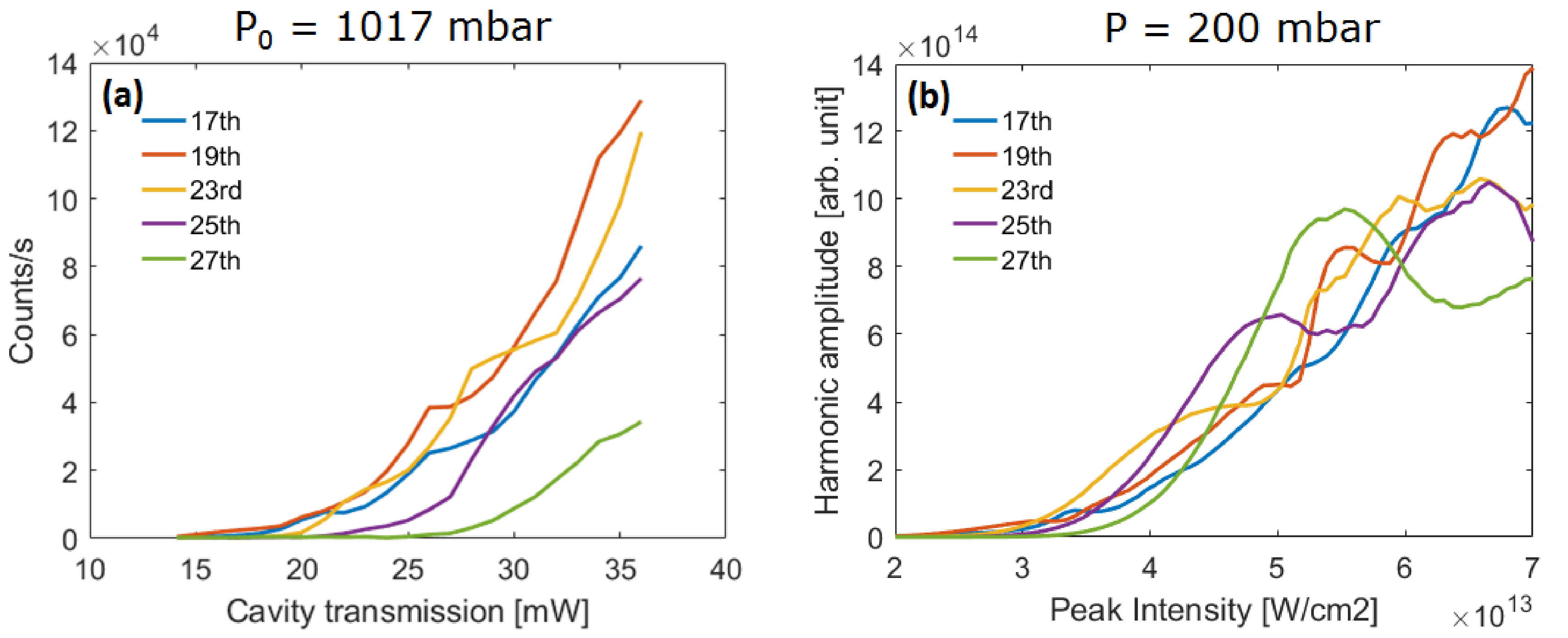
V. SIMULATION VS MEASUREMENTS

Gas pressure



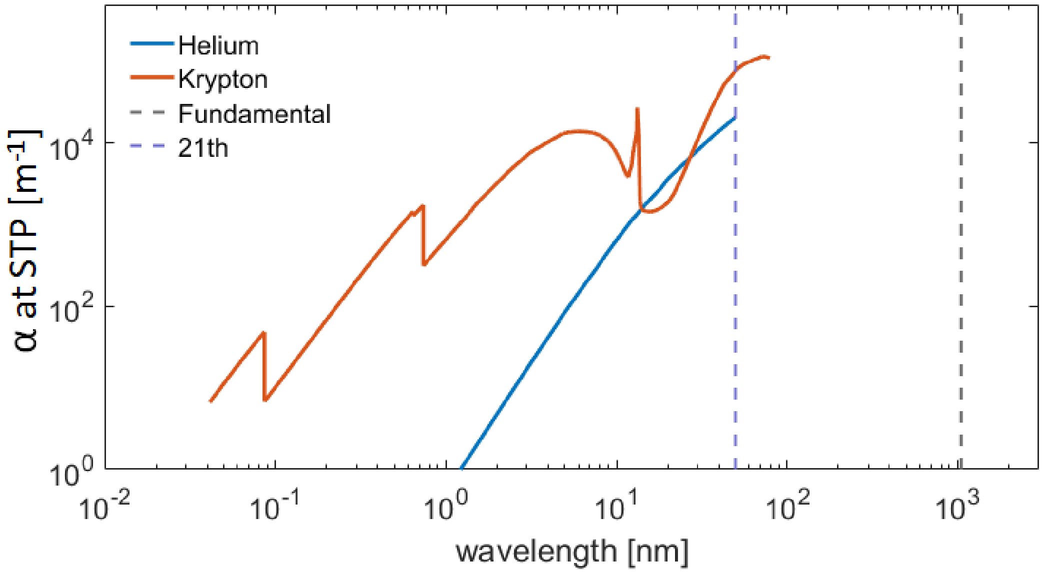
V. SIMULATION VS MEASUREMENTS

Peak Intensity



V.I OPTIMIZATION

Mixing with helium

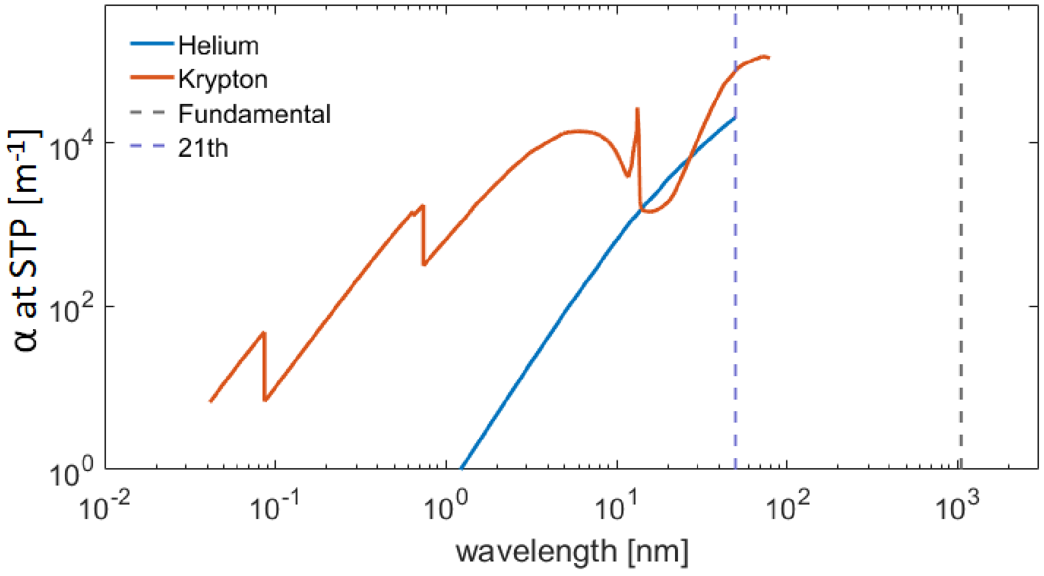


Absorption coefficient of Helium and Krypton at 1atm

V.I OPTIMIZATION

Mixing with helium

Does not work



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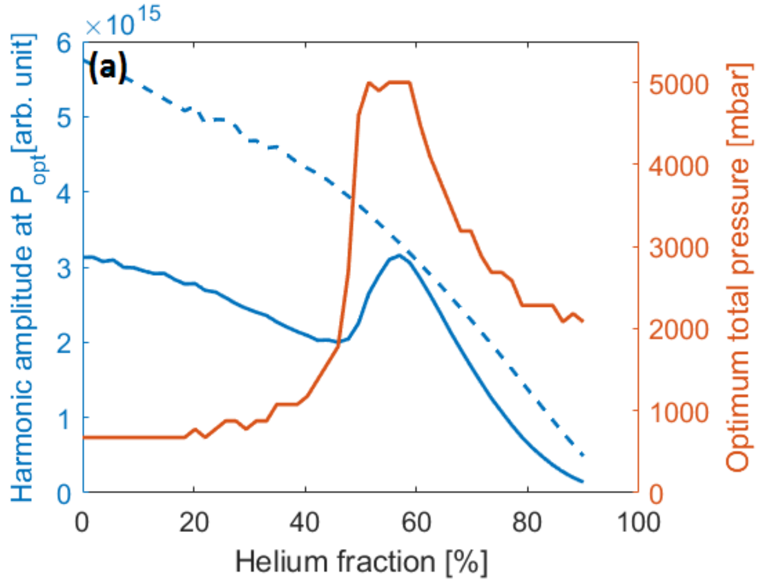
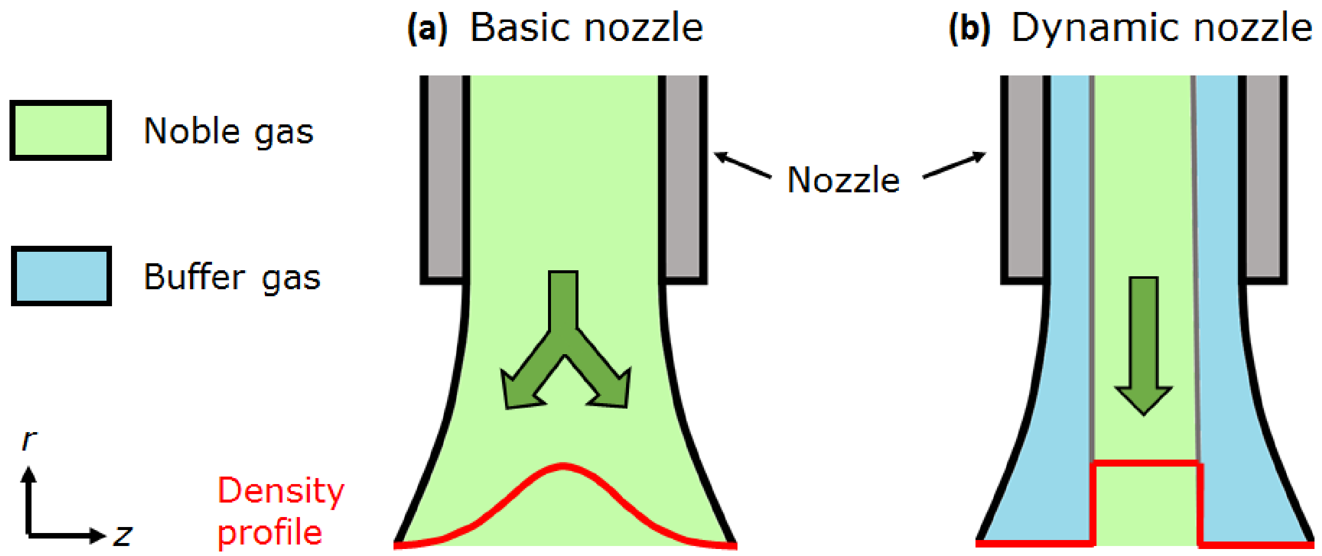


Optimum harmonic amplitude as a function of helium fraction (Io = 7 × 10^{13}W/cm^2)

V.I OPTIMIZATION

Changing the nozzle



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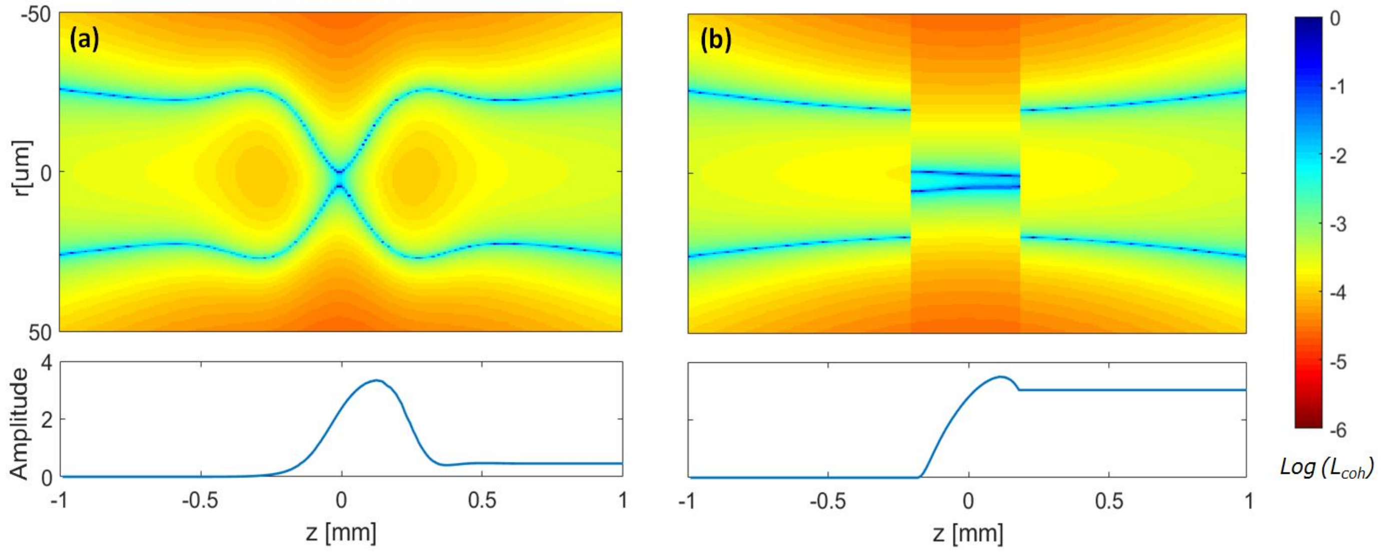
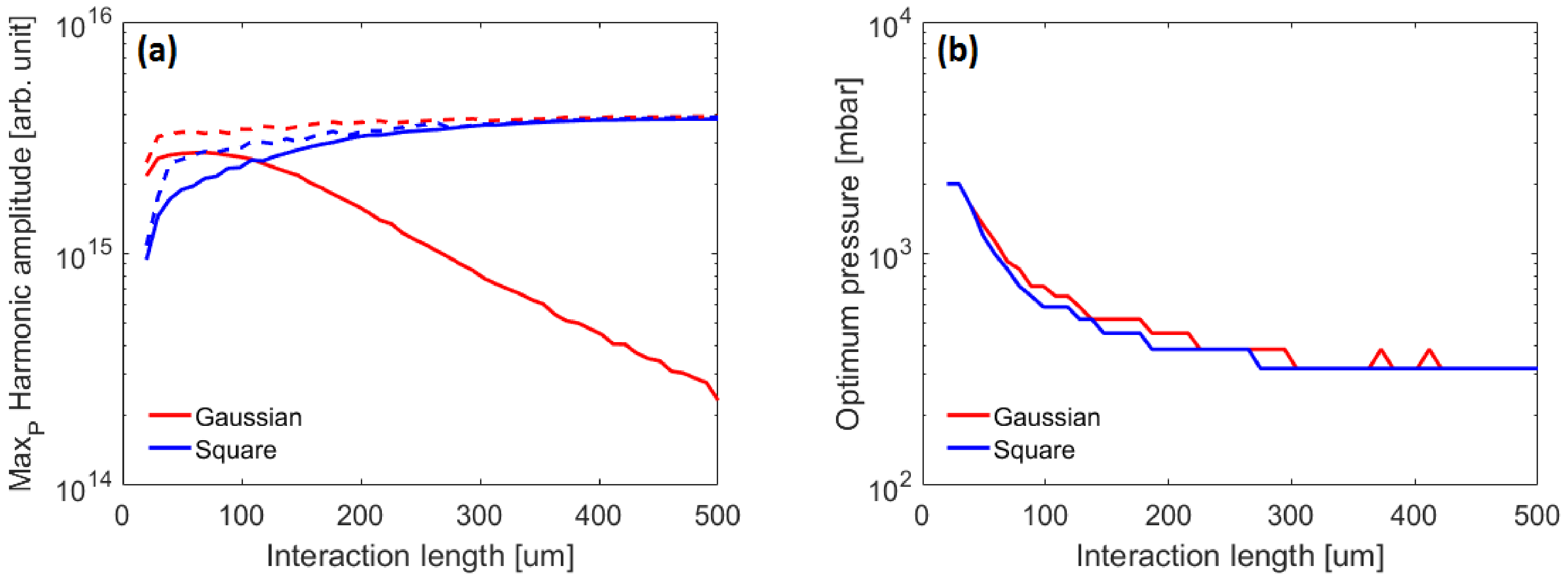


Gaussian density profile *Square density profile*

V.I OPTIMIZATION

Changing the nozzle → Does work



CONCLUSIONS ET PERSPECTIVE

- ✓ The plasma cloud behavior between each and the atomic response have been successfully simulated
- ✓ The consequences of the phase matching effects and absorption have been demonstrated
- ✓ The Harmonics amplitude has been calculated as a function of intensity, pressure, interaction length and nozzle position
- ✓ The optimum parameters have been highlighted considering absorption and phasematching limitation

Perspective:
- Construct a dynamic nozzle and do measurements
- Mixing with hydrogen because less absorption (but too dangerous)