Xenon Clusters in Intense VUV Laser Fields

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letters to nature

Multiple ionization of atom clusters by intense soft X-rays from a free-electron laser

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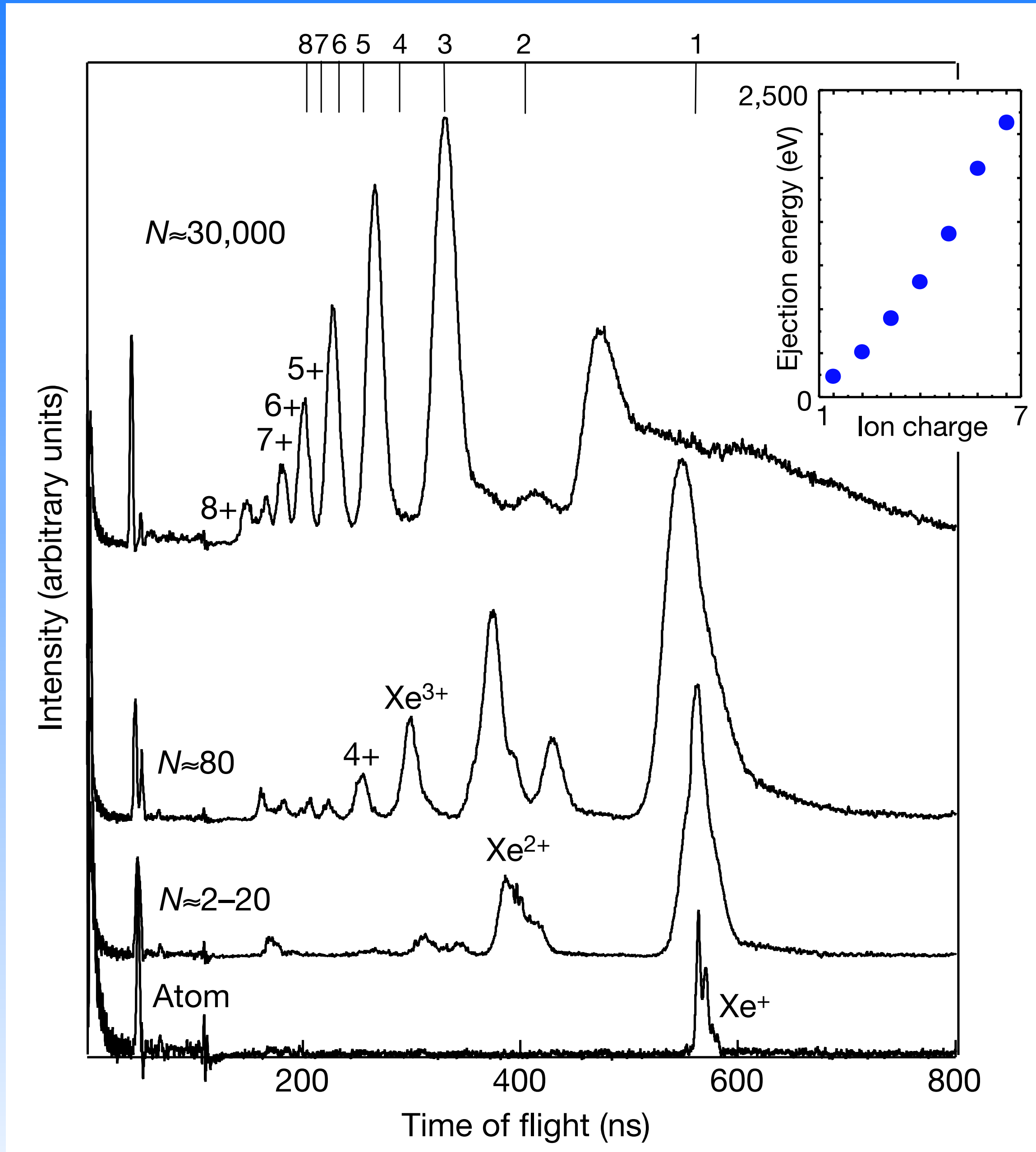
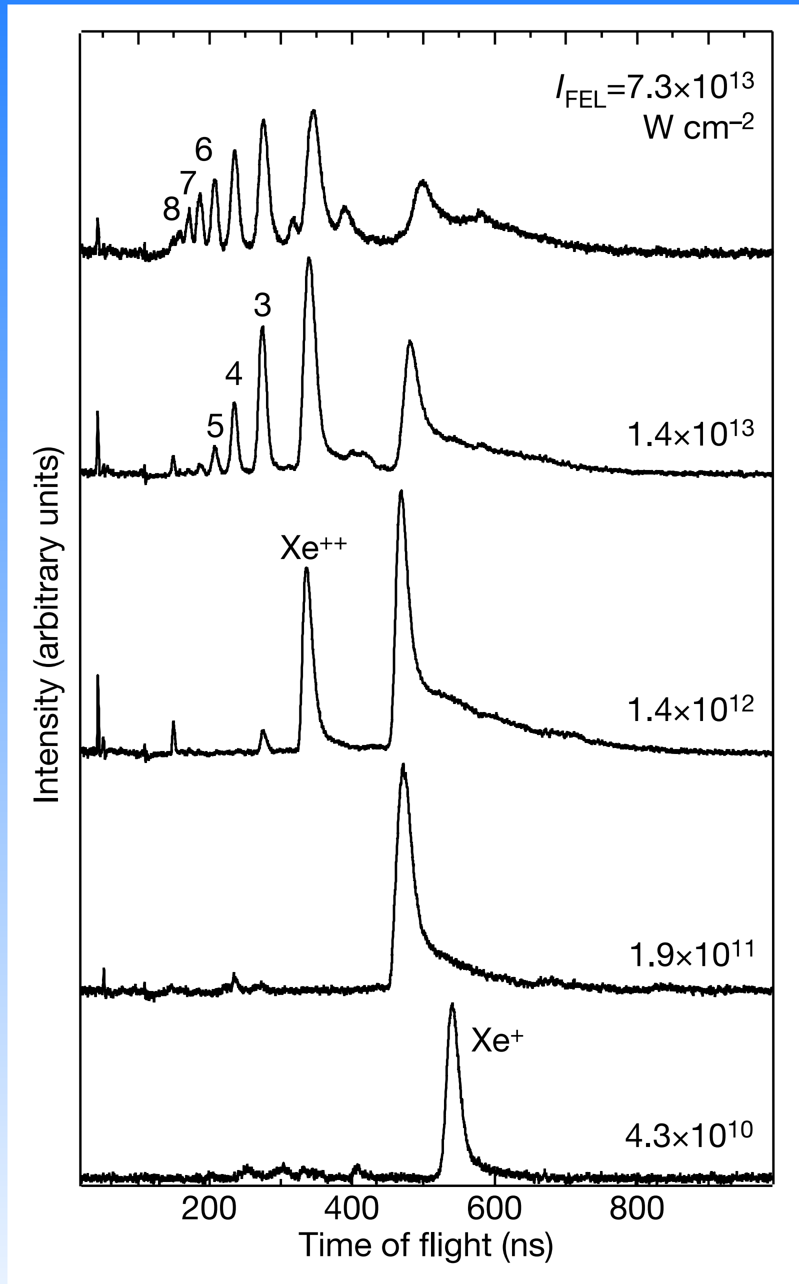
"We find that, whereas Xe atoms become only singly ionized by the absorption of single photons, absorption in clusters is strongly enhanced. On average, each atom in large clusters absorbs up to 400 eV, corresponding to 30 photons."

Nature **420**, 482 (2002).


8 7 6 5 4 3 2 1
N≈30,000
5+
6+
7+
8+
N≈80
4+
Xe3+
N≈2–20
Xe2+
Atom
Xe+
Intensity (arbitrary units)
200
400
600
800
Time of flight (ns)
2,500
0
Ejection energy (eV)
1
7
Ion charge


I_{FEL}=7.3×10[13] W cm[−2]
8
7
6
3
4
5
1.4×10[13]
Xe[++]
1.4×10[12]
1.9×10[11]
Xe[+]
4.3×10[10]
Intensity (arbitrary units)
200
400
600
800
Time of flight (ns)

back

1 – Why is the experimental result surprising?

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- the atomic unit of intensity is about 10^{16} W/cm^2

  $\Rightarrow$ cluster–laser interaction is still in the perturbative regime
- in classical simulations, Wabnitz *et al.* found that each atom absorbs only about one photon (not 30 photons, as observed experimentally)

2 – Proposed mechanism

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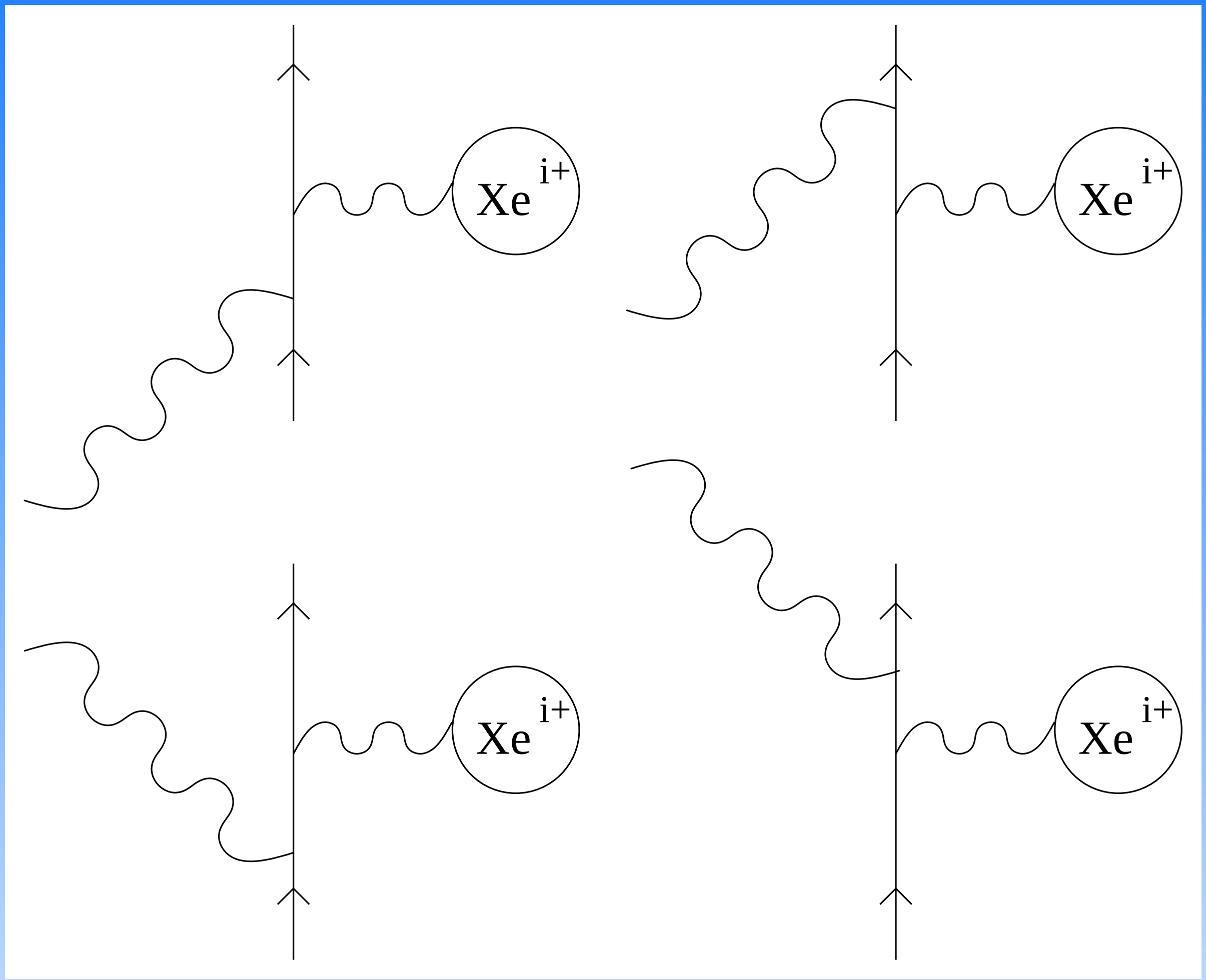
  - thermal equilibrium of electron gas is maintained at all times during the laser pulse
  - heating via *inverse bremsstrahlung*



- both absorption and emission processes can take place
- in thermal equilibrium, there are more cold than warm electrons
- this leads to effective heating ⇒ inverse bremsstrahlung

3 – Our treatment of inverse bremsstrahlung

- second-order perturbation theory: first order in electron–ion and electron–photon coupling, respectively; classical limit $\hbar \longrightarrow 0$ is avoided

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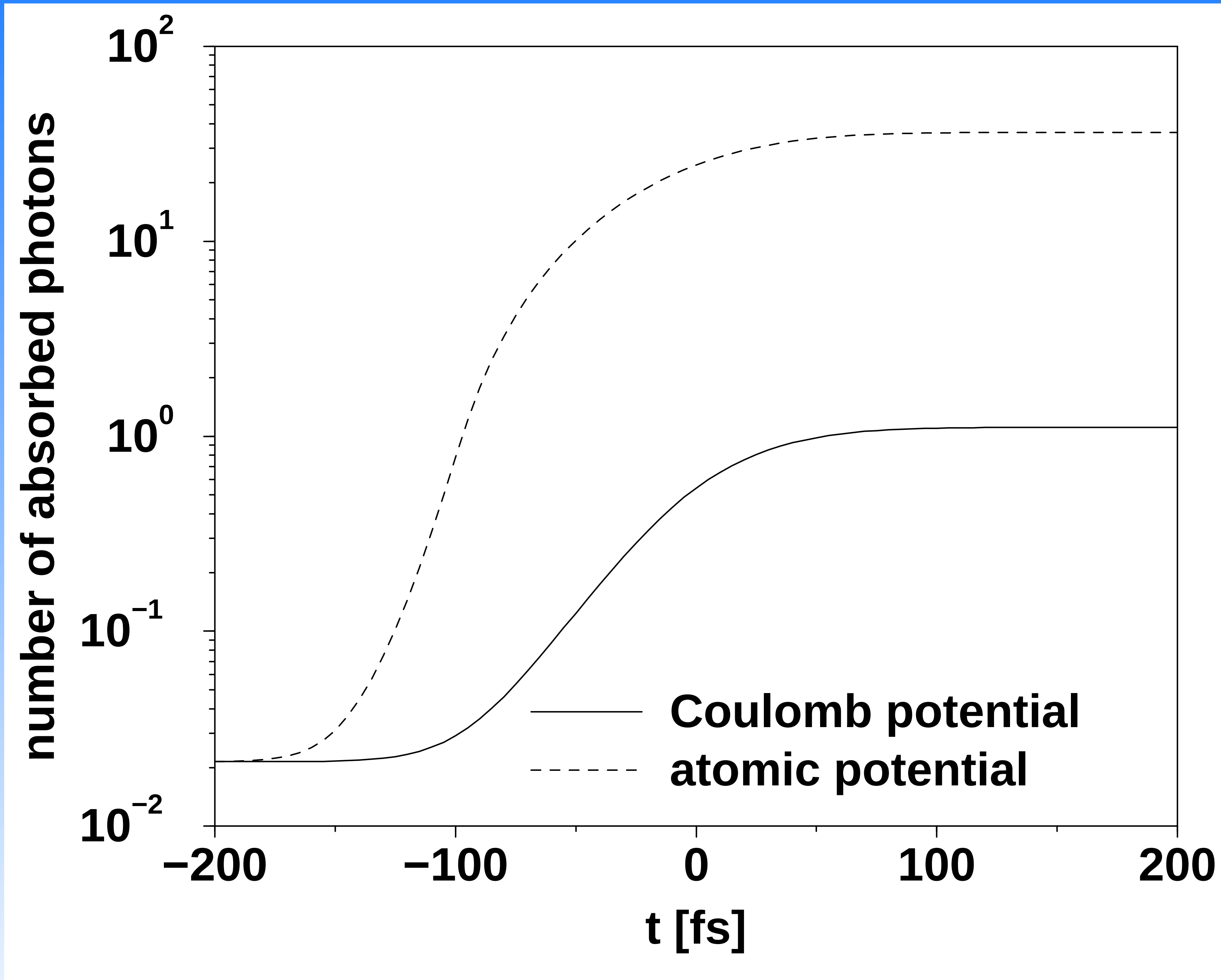
- we use ionic potential of the form

$$V_i(r) = -\frac{1}{r}\left\{i + [Z - i]\exp\left(-\alpha_i r\right)\right\}\exp\left(-r/\lambda_D\right)$$

i: ionic charge state; Z: nuclear charge

$$\lambda_D = \sqrt{\frac{T}{4\pi n}}$$

T: temperature in units of energy; n: plasma-electron density



10^2
10^1
10^0
10^{-1}
10^{-2}
number of absorbed photons
−200
−100
0
100
200
t [fs]
Coulomb potential
atomic potential

4 – Coupled rate equations

$$\begin{aligned}
\dot{n}_0(t) &= -\sigma_1(t) j_{\mathrm{ph}}(t) n_0(t) \\
\dot{n}_1(t) &= \sigma_1(t) j_{\mathrm{ph}}(t) n_0(t) - \sigma_2(t) j_{\mathrm{ph}}(t) n_1(t) \\
&\vdots
\end{aligned}$$

$n_i(t)$: probability of finding Xe^{i+} in the cluster; $\sigma_{i+1}(t)$: photoionization cross section of Xe^{i+}; $j_{\mathrm{ph}}(t)$: photon flux

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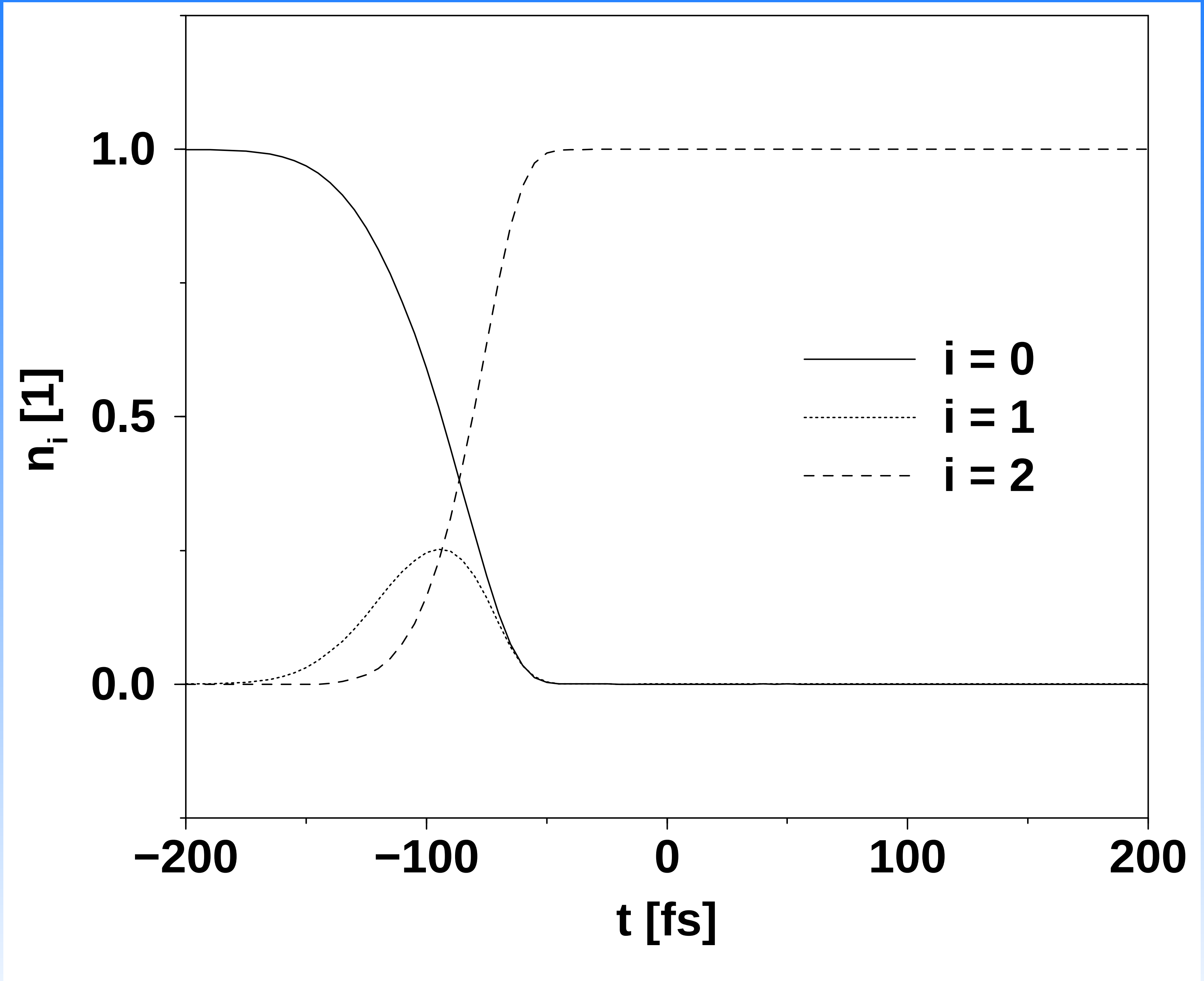
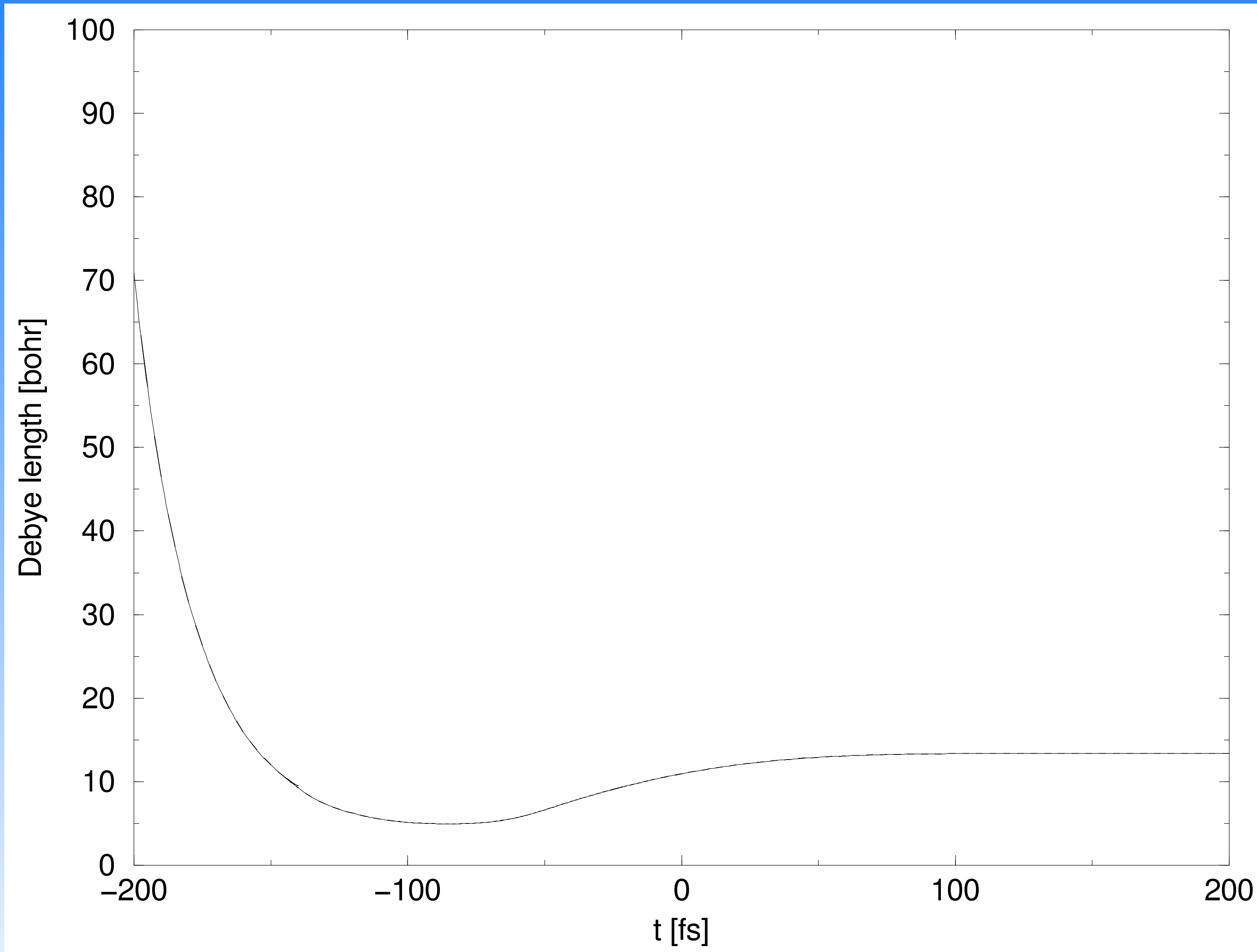
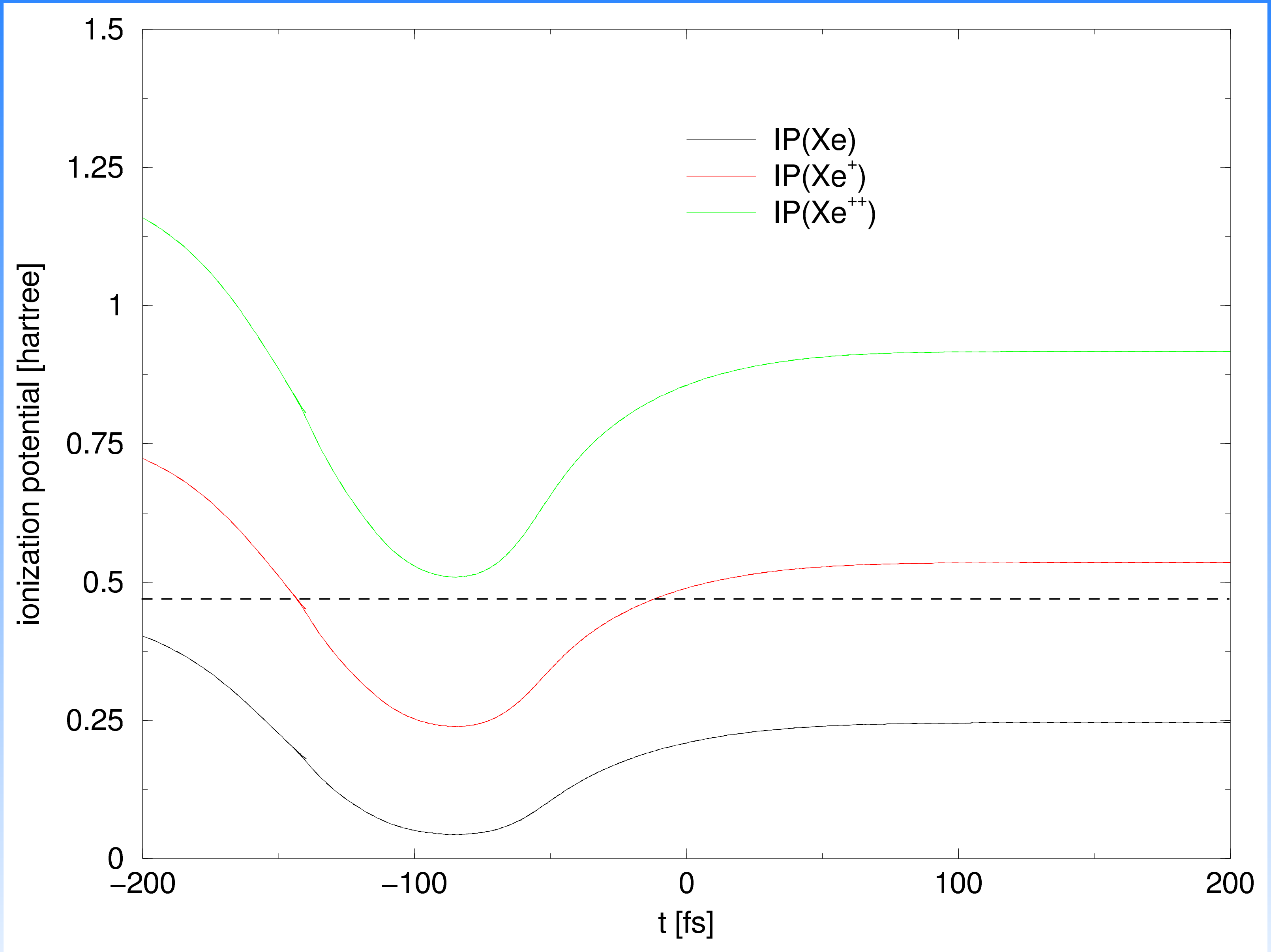
$$\dot{\mathcal{E}}_{\mathrm{kin}}(t) = \frac{3}{2} q(t) \dot{T}(t) + \sum_i \varepsilon_i(t) \sigma_i(t) j_{\mathrm{ph}}(t) n_{i-1}(t)$$

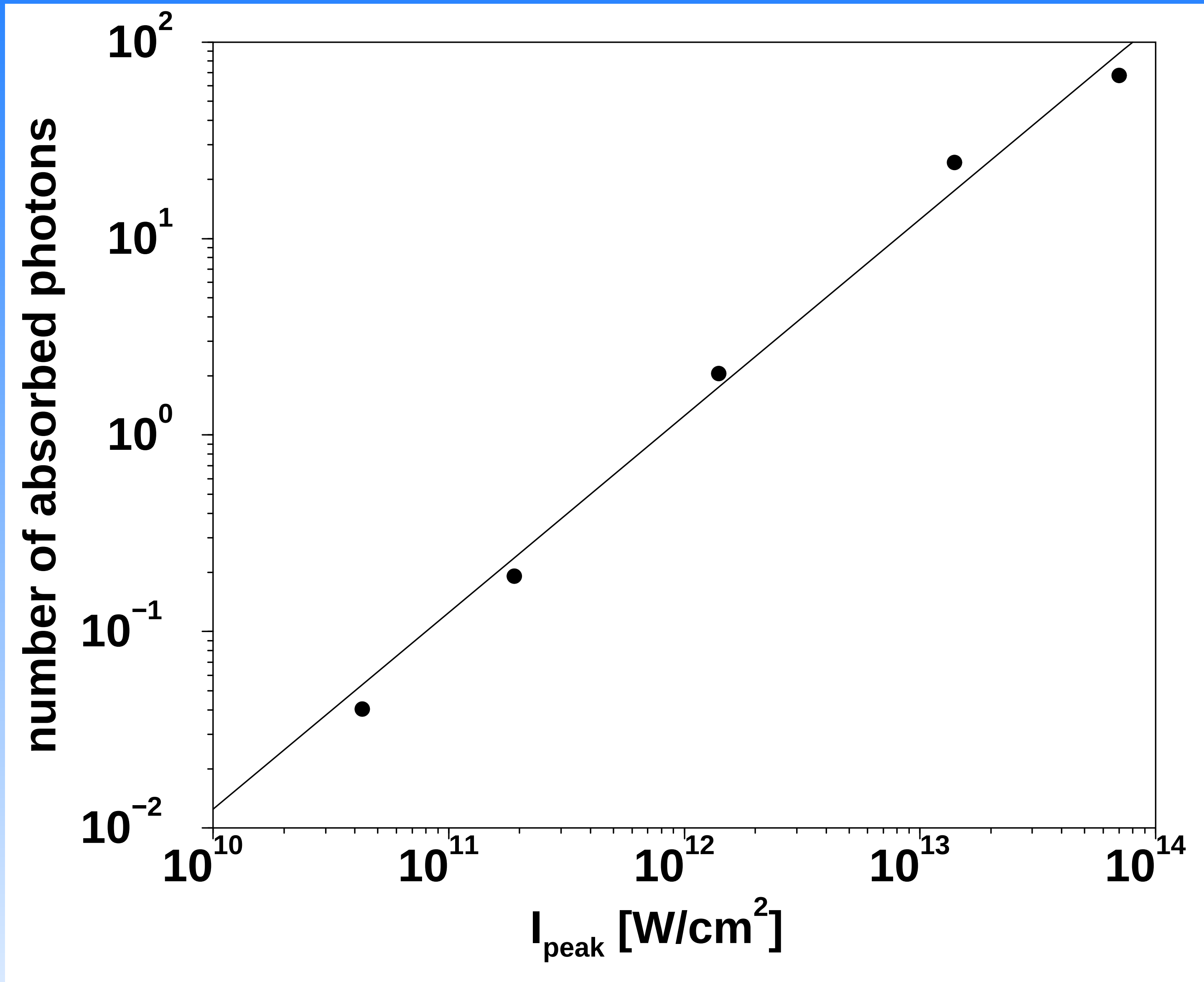
$$\mathcal{E}_{\mathrm{kin}}(t) = \frac{3}{2} q(t) T(t)$$

$q(t)$: average number of plasma electrons per atom; $\varepsilon_i(t)$: kinetic energy of photoelectron leaving Xe^{i+} behind


1.0
0.5
0.0
n_i [1]
−200
−100
0
100
200
t [fs]
i = 0
i = 1
i = 2


Debye length [bohr]
t [fs]
100
90
80
70
60
50
40
30
20
10
0
–200
–100
0
100
200


1.5
1.25
1
0.75
0.5
0.25
0
–200
–100
0
100
200
t [fs]
ionization potential [hartree]
IP(Xe)
IP(Xe+)
IP(Xe++)



mass spectra

5 – Thermalization model

$$\sum_{i=0}^{8} n_i(t \to \infty) I_0^{(N-i)} + \frac{3}{2} q(t \to \infty) T(t \to \infty) = E_{\text{tot}}$$

$$= \sum_{i=0}^{8} g_i \frac{\exp\left\{-I_0^{(N-i)}/T_{\text{eq}}\right\}}{Z_{\text{eq}}} I_0^{(N-i)} + \frac{3}{2} q_{\text{eq}} T_{\text{eq}} \left(+\frac{3}{2} T_{\text{eq}}\right)$$

g_i: statistical weight of Xe^{i+}; Z_{eq}: canonical partition function

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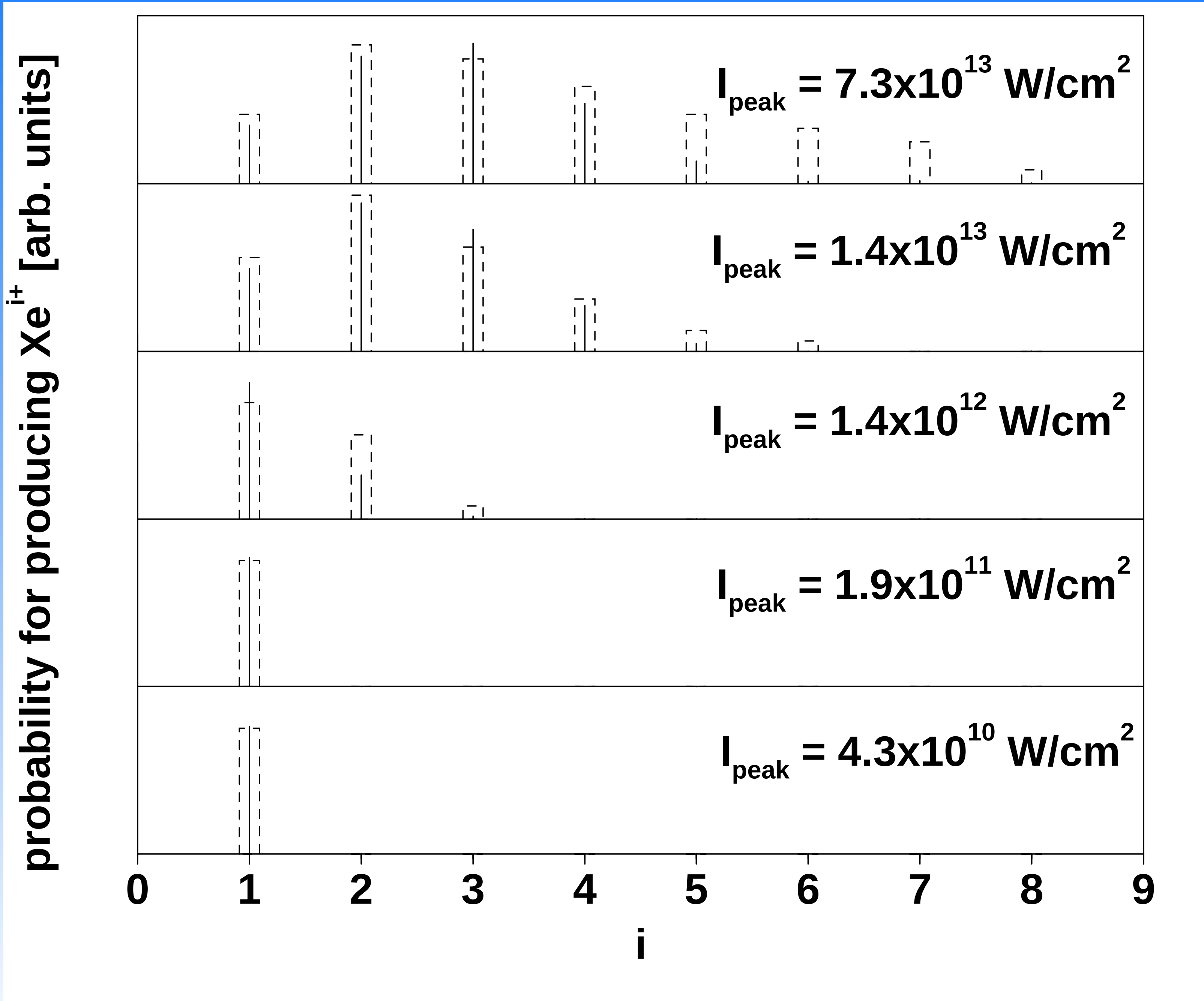
ionization potentials $I_0^{(N-i)}$ of atomic xenon, in eV

i	1	2	3	4	5	6	7	8
ab initio	11.7	31.7	60.7	103	156	220	310	414
experiment	12.1	33.1	66.2	108				


probability for producing Xe^{i+} [arb. units]
I_{peak} = 7.3x10^{13} W/cm^2
I_{peak} = 1.4x10^{13} W/cm^2
I_{peak} = 1.4x10^{12} W/cm^2
I_{peak} = 1.9x10^{11} W/cm^2
I_{peak} = 4.3x10^{10} W/cm^2
0
1
2
3
4
5
6
7
8
9
i

6 – Reference

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