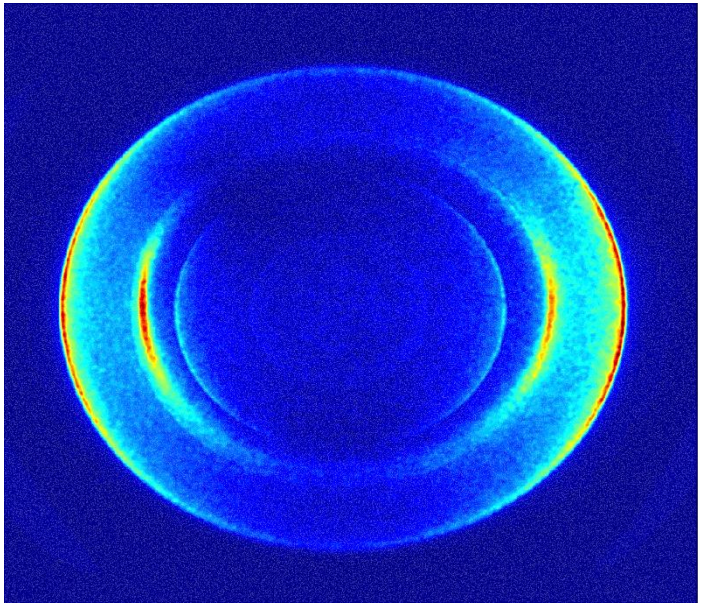
NONLINEAR PROCESSES IN THE EXTREME ULTRAVIOLET REGION USING THE FEL FERMI@ELETTRA

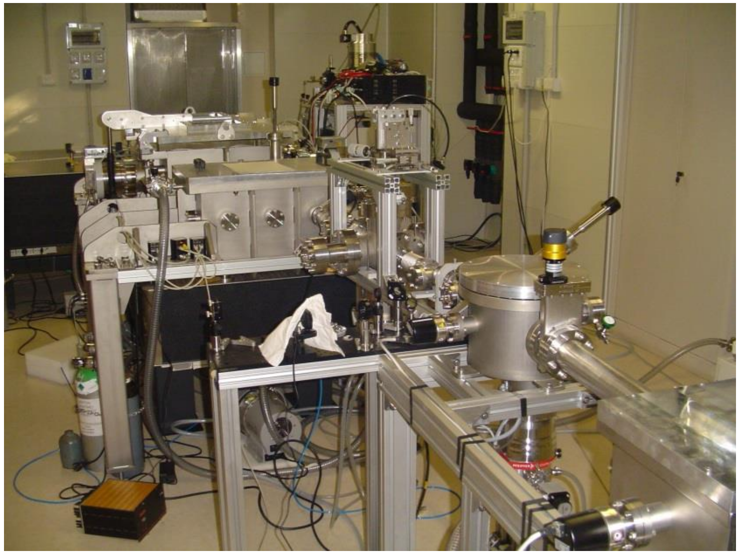
A. Dubrouil, M. Reduzzi, C. Feng, J. Hummert, P. Finetti, O. Plekan, C. Grazioli, M. Di Fraia, V. Lyamayev,A. LaForge, F. Stienkemeier, Y. Ovcharenko, M. Coreno, P. O'keeffe, M. Devetta, N. Berrah, K. Motomura, S. Mondal, P. Demekhin, A. Kuleff, L. Cederbaum, K.C. Prince, K. Ueda, C. Callegari, P. Carpeggiani, M. Negro, D. Faccialà, Klaus Bartschat, E. Gryzlova, A. Grum-Grzhimailo, G. Sansone

Moscow 28 October 2014



XUV light sources

High-harmonic generation



~ 1-10m

Attosecond time resolution

Low photon flux

Free Electron Lasers

~0.1-1 km

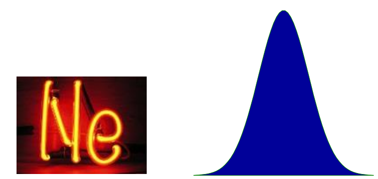
Femtosecond time resolution

High photon flux-> nonlinear effects

Experiments

1) XUV nonlinear excitation of neon dimers (FEL)

2) Two-photon double ionization of neon



Motivation:
Interatomic Coulombic Decay

Mechanism of electronic decay (after inner valence
Ionization) dominant in:

- *Hydrogen bonded clusters $(H_20)_n$, $(HF)_n$*
  Van der Waals clusters (Ne)n; Ne-Ar; He-He

- *Source of low energy electrons*

L.S. Cederbaum et al. Phys. Rev. Lett. **79**, 4778 (1997)
T. Jahnke et al. Phys. Rev. Lett. **93**, 163401 (2004)

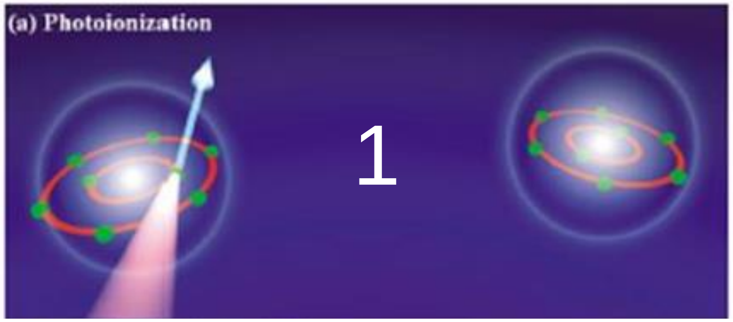
ICD: excitation

Mechanism of electronic decay (after inner valence Ionization) dominant in:

- *Hydrogen bonded clusters* $(H_20)_n$, $(HF)_n$
  Van der Waals clusters (Ne)n; Ne-Ar; He-He
- *Source of low energy electrons*

1) Ionization by XUV radiation
   of a 2s electron.

Neon Dimer



L.S. Cederbaum et al. Phys. Rev. Lett. **79**, 4778 (1997)
T. Jahnke et al. Phys. Rev. Lett. **93**, 163401 (2004)

ICD: electron correlation

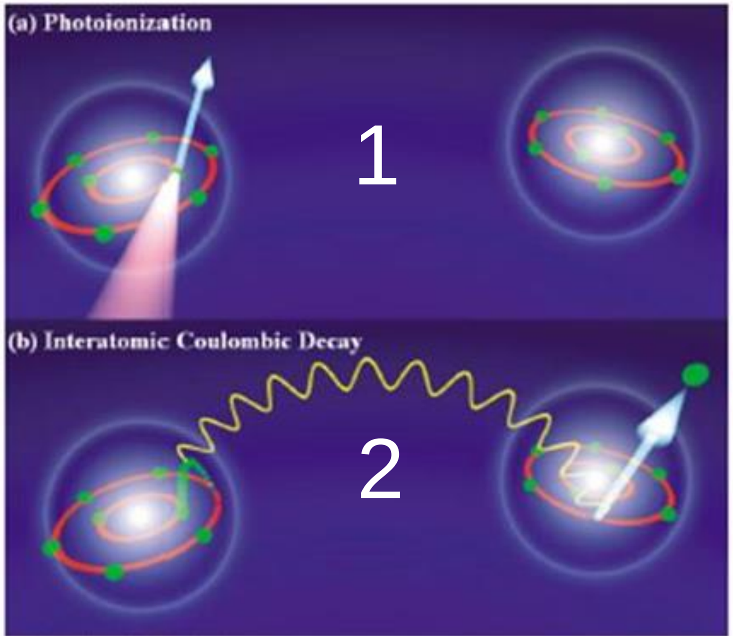
Mechanism of electronic decay (after inner valence Ionization) dominant in:

- *Hydrogen bonded clusters $(H_20)_n$, $(HF)_n$*
  Van der Waals clusters (Ne)n; Ne-Ar; He-He
- *Source of low energy electrons*

1) Ionization by XUV radiation of a 2s electron.

2) a 2p electron fills the 2s hole; emission of a 2p electron from the neighboring atom

Neon Dimer



L.S. Cederbaum et al. Phys. Rev. Lett. **79**, 4778 (1997)
T. Jahnke et al. Phys. Rev. Lett. **93**, 163401 (2004)

ICD: Coulomb explosion

Mechanism of electronic decay (after inner valence Ionization) dominant in:

- *Hydrogen bonded clusters $(H_20)_n$, $(HF)_n$*
  Van der Waals clusters (Ne)n; Ne-Ar; He-He

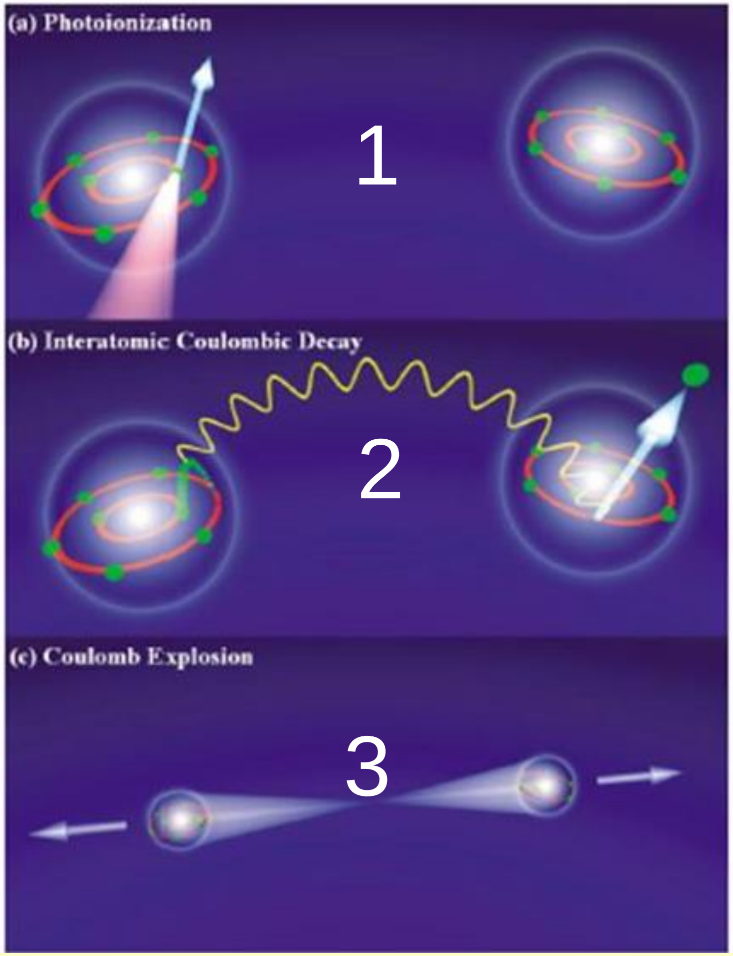
- *Source of low energy electrons*

1) Ionization by XUV radiation
   of a 2s electron.

2) a 2p electron fills the 2s hole;
   emission of a 2p electron from
   the neighboring atom

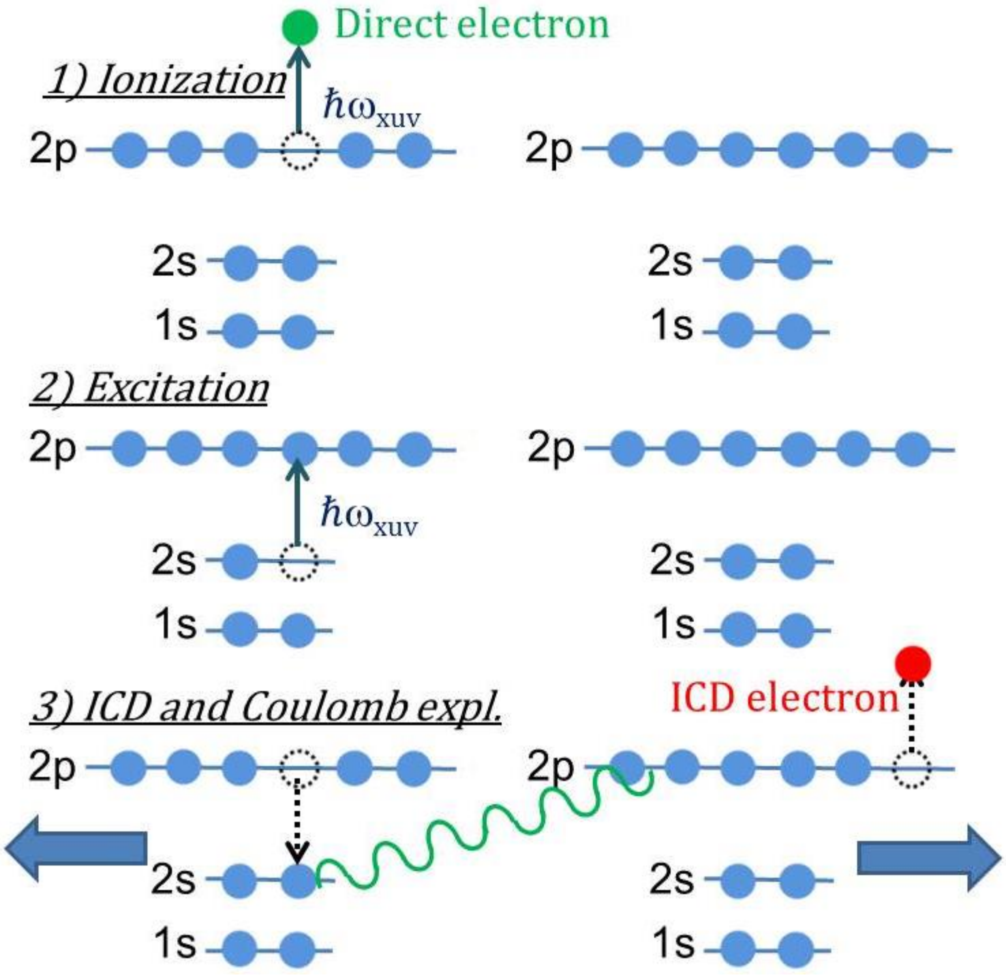
3) Coulomb explosion of the molecules.
   timescale ~80 fs

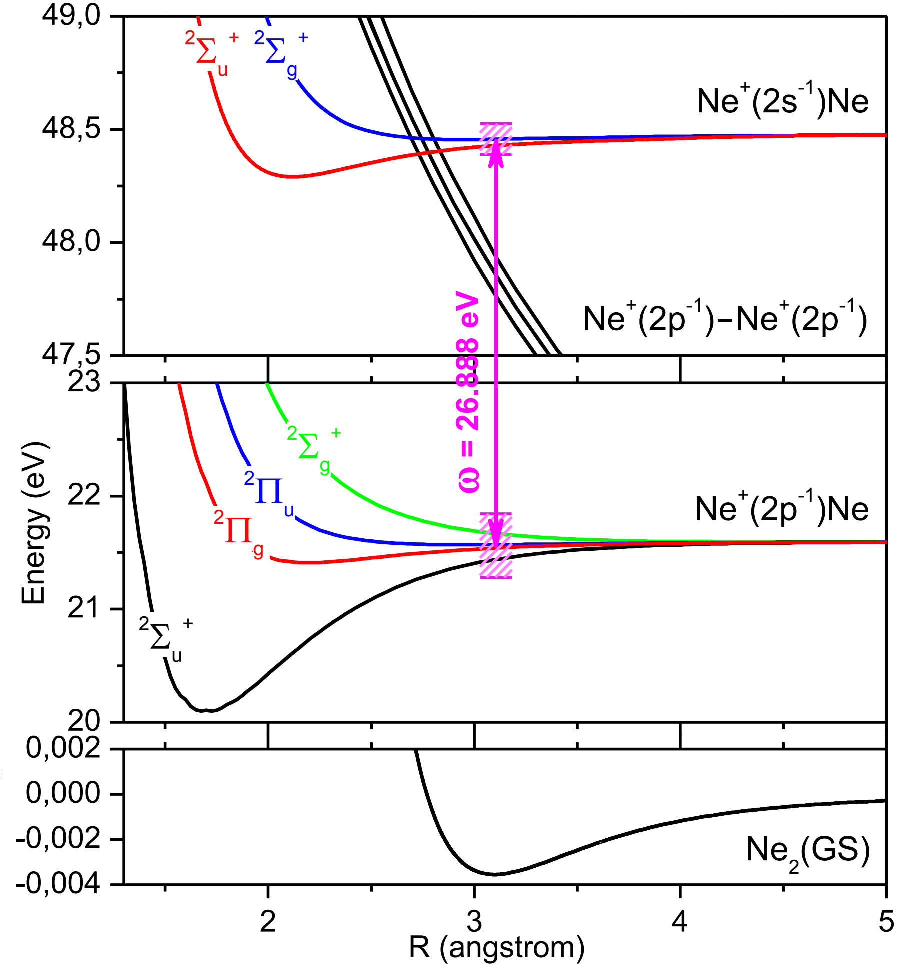
Neon Dimer



L.S. Cederbaum et al. Phys. Rev. Lett. **79**, 4778 (1997)
T. Jahnke et al. Phys. Rev. Lett. **93**, 163401 (2004)

Nonlinear excitation of ICD





E_{2s}-E_{2p}=$\hbar\omega = 26.9\ eV$

P. V. Demekhin *et al.*, Phys Rev Lett **107** (2011).

FERMI@ELETTRA: *seeded* FEL

First seeded FEL user facility

Energy 1-100 μJ

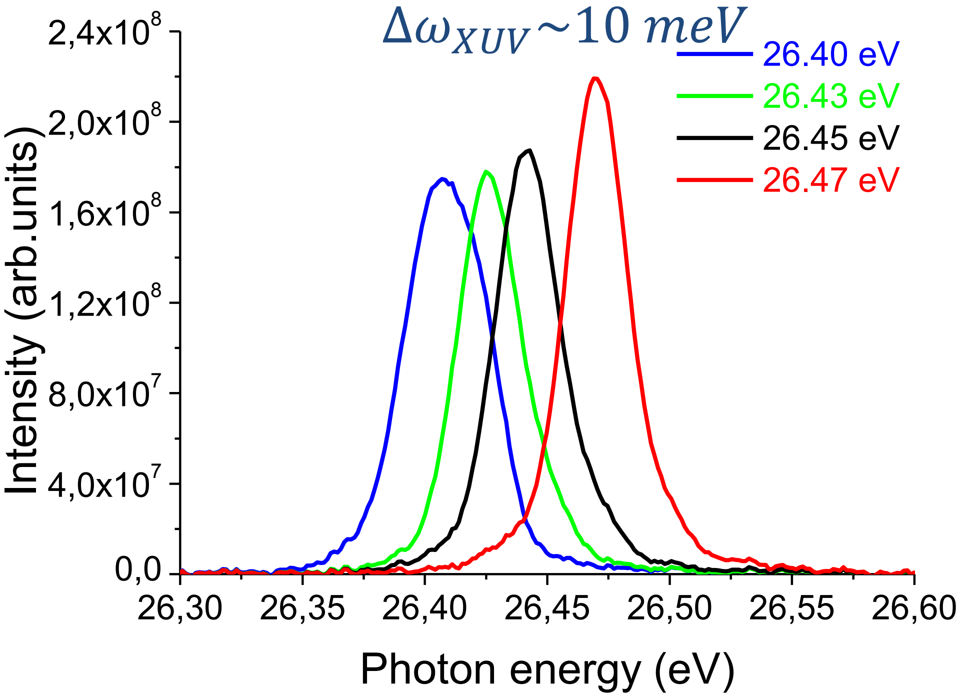
Photon energy tunability
19-62 eV

Pulse duration: 100 fs

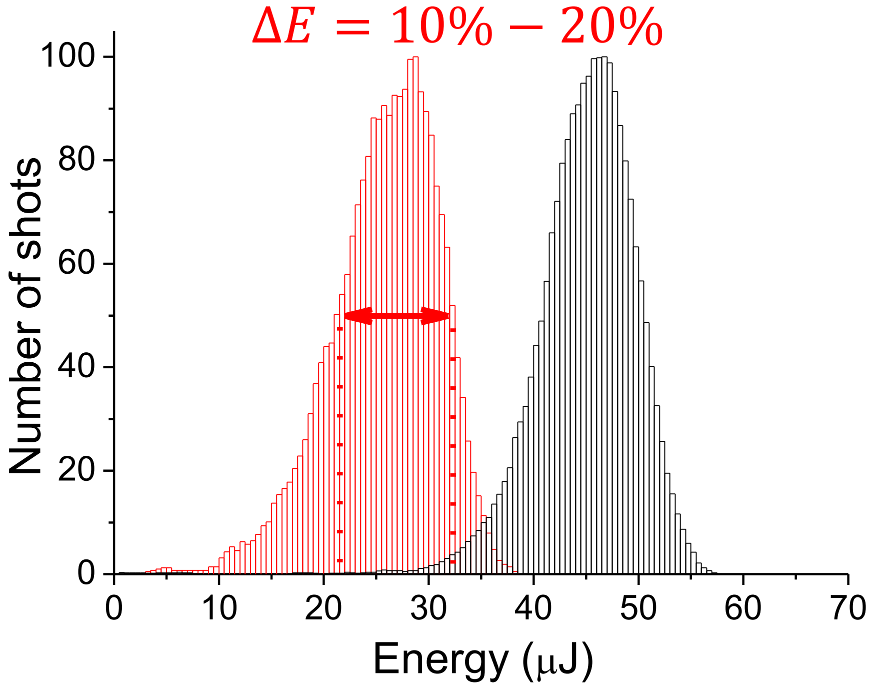
https://www.elettra.trieste.it/FERMI/index.php?n=Main.Parameter

FERMI@ELETTRA: characteristics

Photon energy tunability



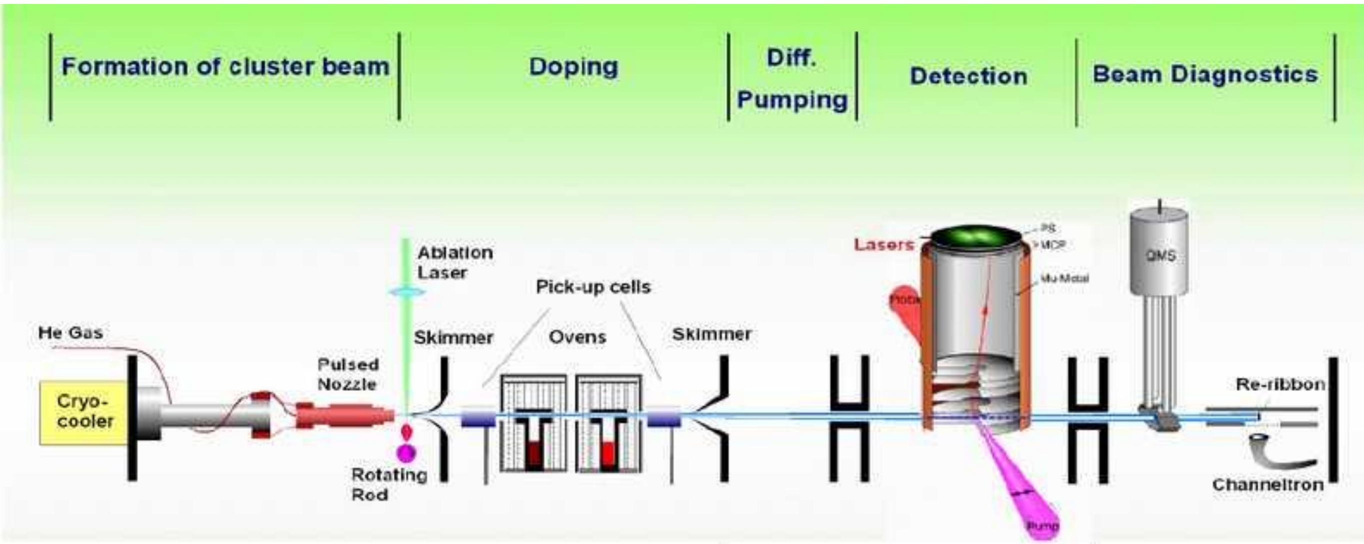
Pulse energy stability

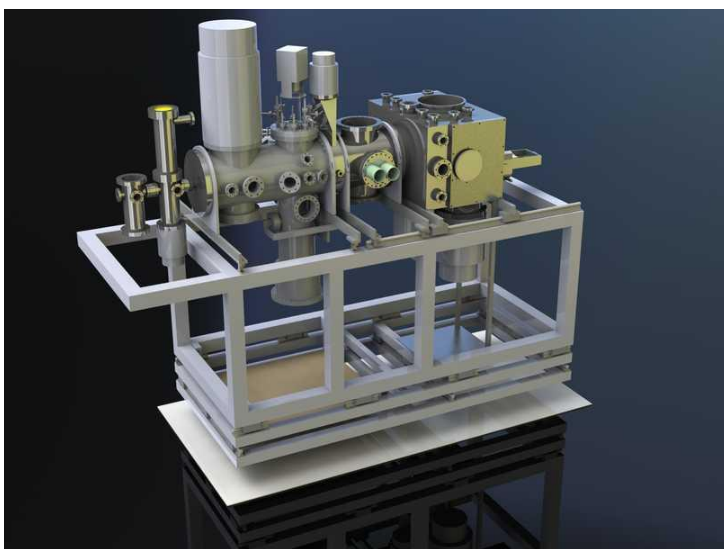


Single shot information (energy-spectrum)

Ideal source for investigating nonlinear resonant effects in the XUV region !

Low Density Matter (LDM) end station





Two supersonic Even-Lavie valves
(monomers, cluster, doped clusters)

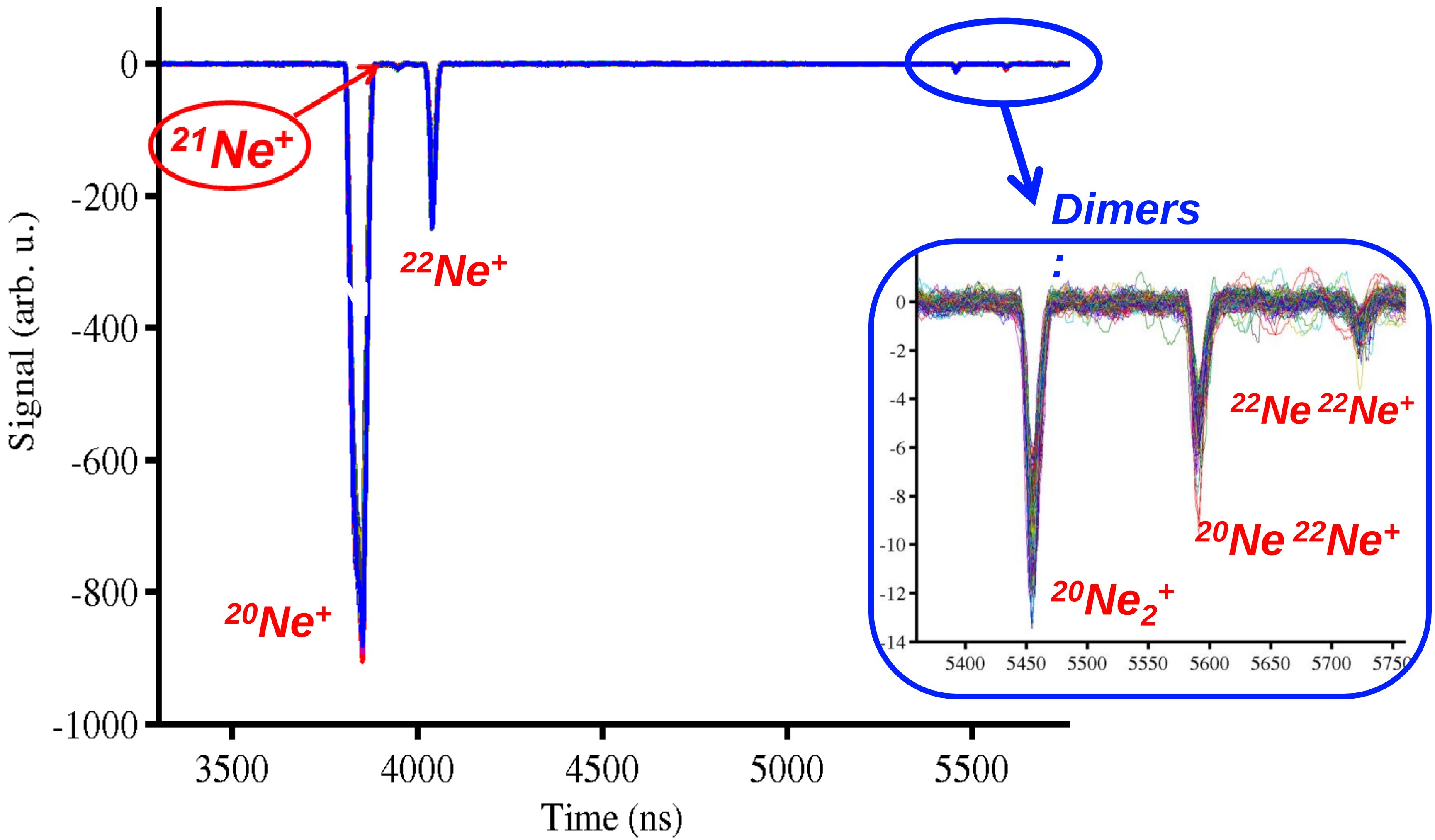
Experimental chamber:

1) Velocity map imaging spectrometer

2) Time-of-flight ion spectrometer

www.elettra.eu/lightsources/fermi/fermi.html

Signature of neon dimers: ions



1) Clear signature of singly ionized neon dimers in the TOF mass spectrum
2) 1% concentration of neon dimers (quadrupole mass spectometer)

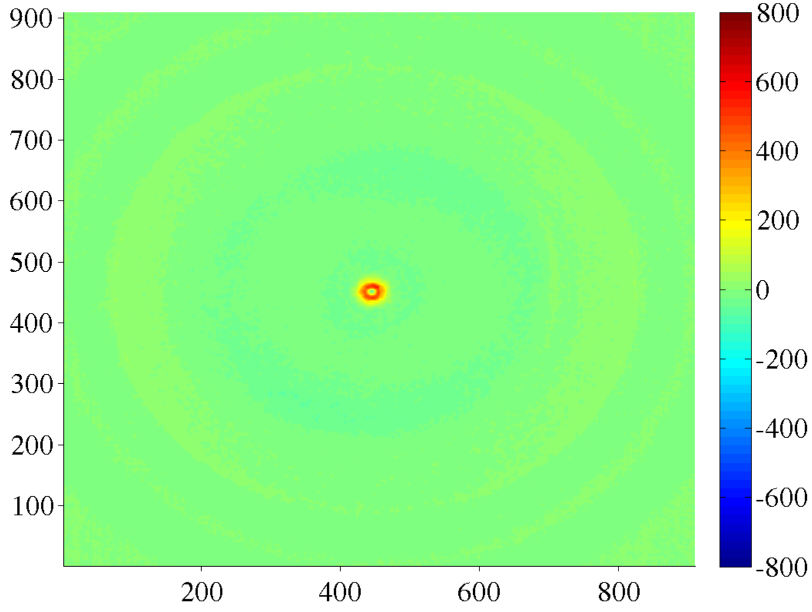
Signature of neon dimers : electrons (below ionization threshold of neon)

Differences of VMI images (for the same XUV pulse energy); no artefact introduced by the inversion procedure

E_0=27 μJ

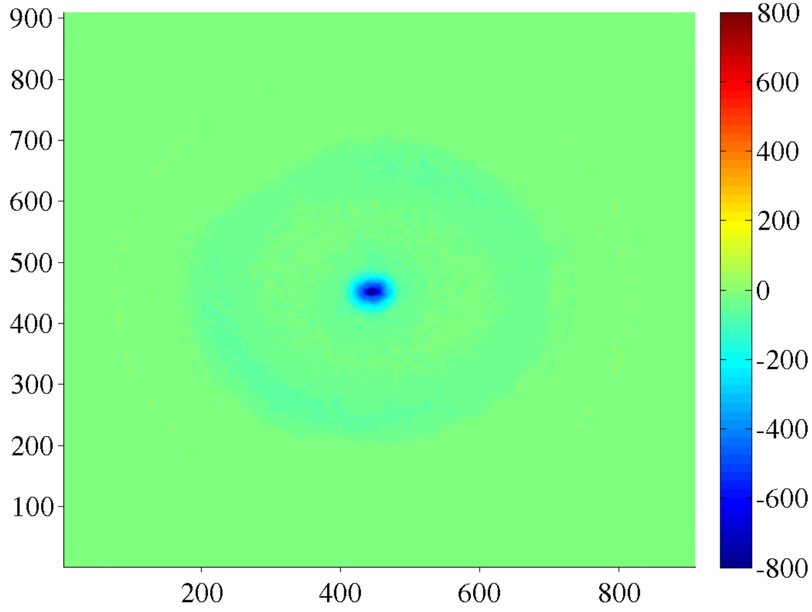
$S_{ref}=S(\lambda_1)$ λ_1=60.13 nm → 20.62 eV

λ_2=60.19 nm → 20.60 eV



ΔS= **$S(\lambda_2)$- $S(\lambda_1)$ >0**

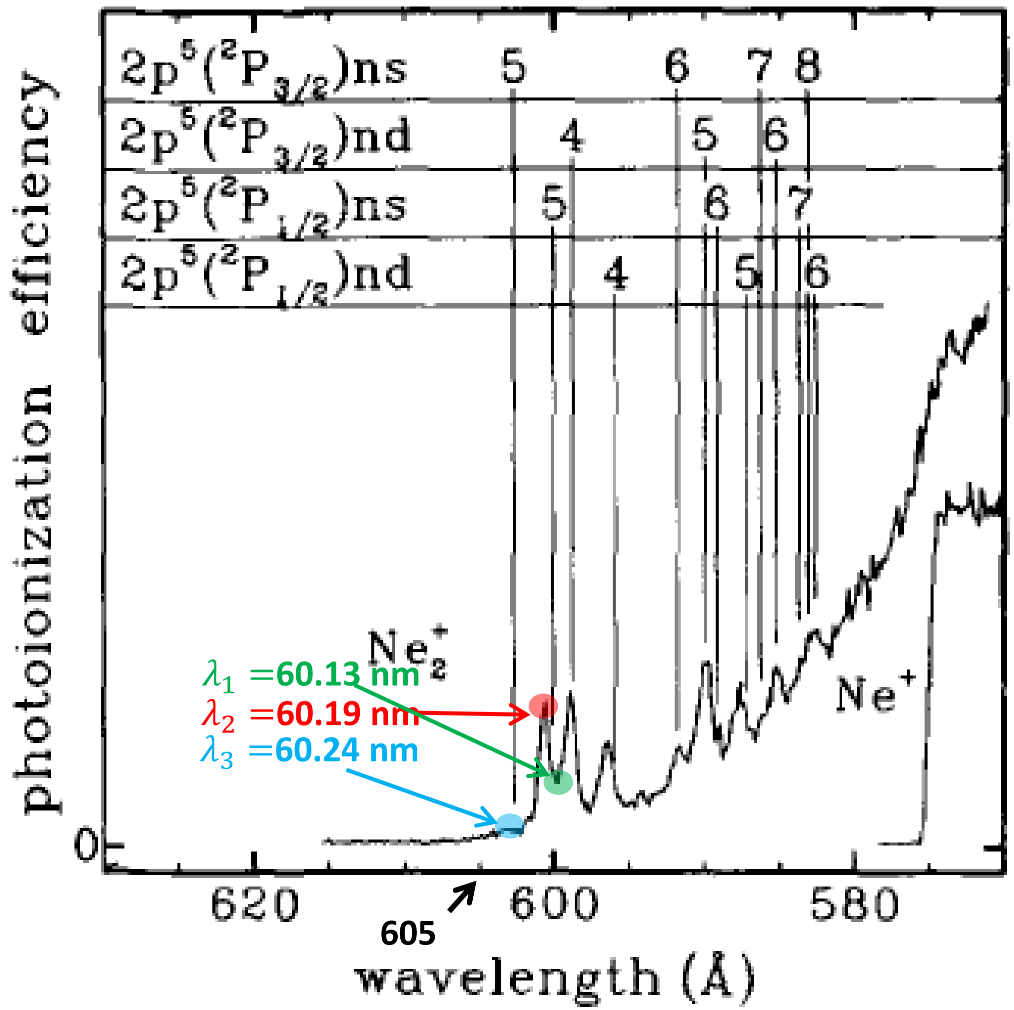
λ_3=60.24 nm→20.58 eV



ΔS= **$S(\lambda_3)$- $S(\lambda_1)$ <0**

Differences in the low energy electron region

Single photon ionization of neon dimers



Autoionization features in Neon dimers

Trevor et al. J. Chem. Phys. 80 6083 (1984)

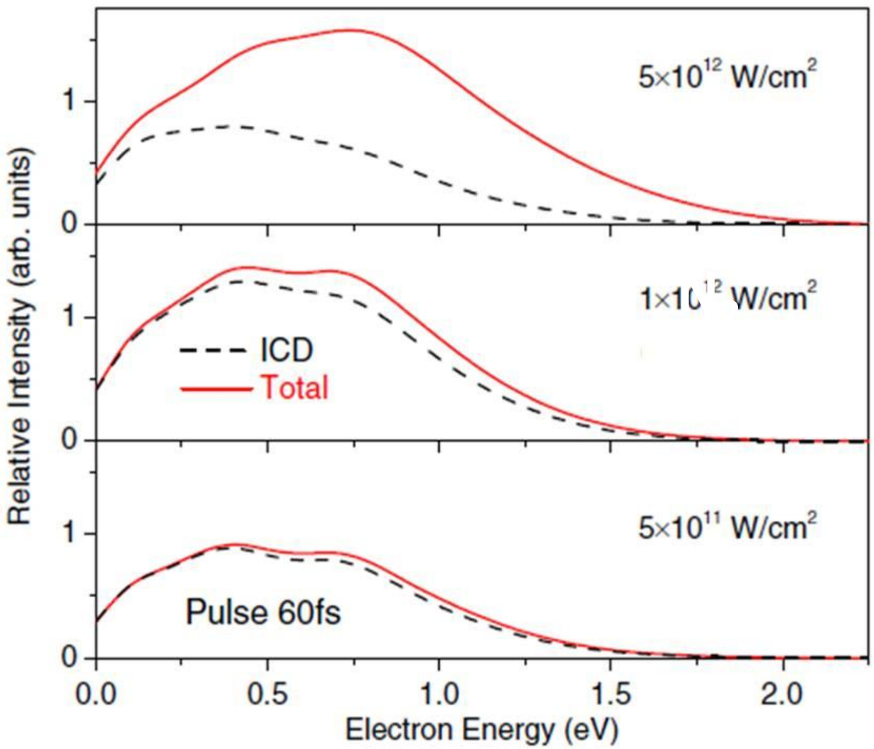
$$\lambda_1(60.13\ nm) < \lambda_2(60.19\ nm) < \lambda_3(60.24\ nm)$$

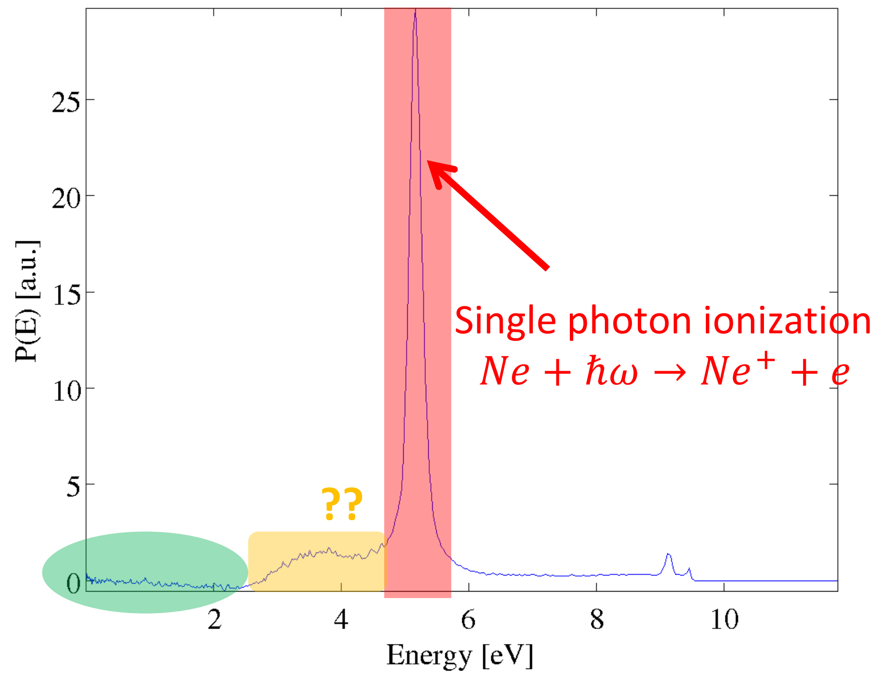
$$S(\lambda_3) < S(\lambda_1) < S(\lambda_2)$$

Nonlinear excitation of ICD: electrons

Photoelectron spectra dominated by ICD for the dimers

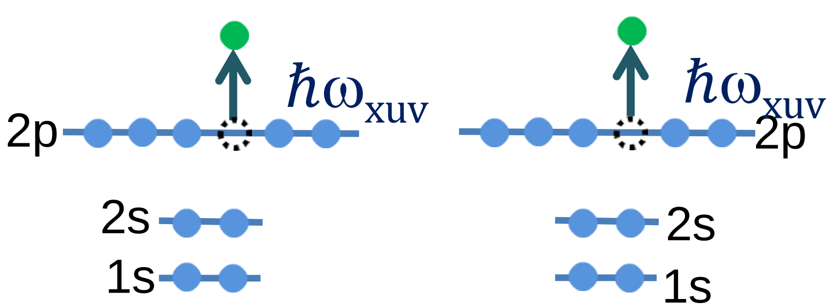
$\hbar\omega_{xuv} = 26.9$ eV





Competing channel: two-site single photon ionization of neon monomers

No clear indication of ICD electrons

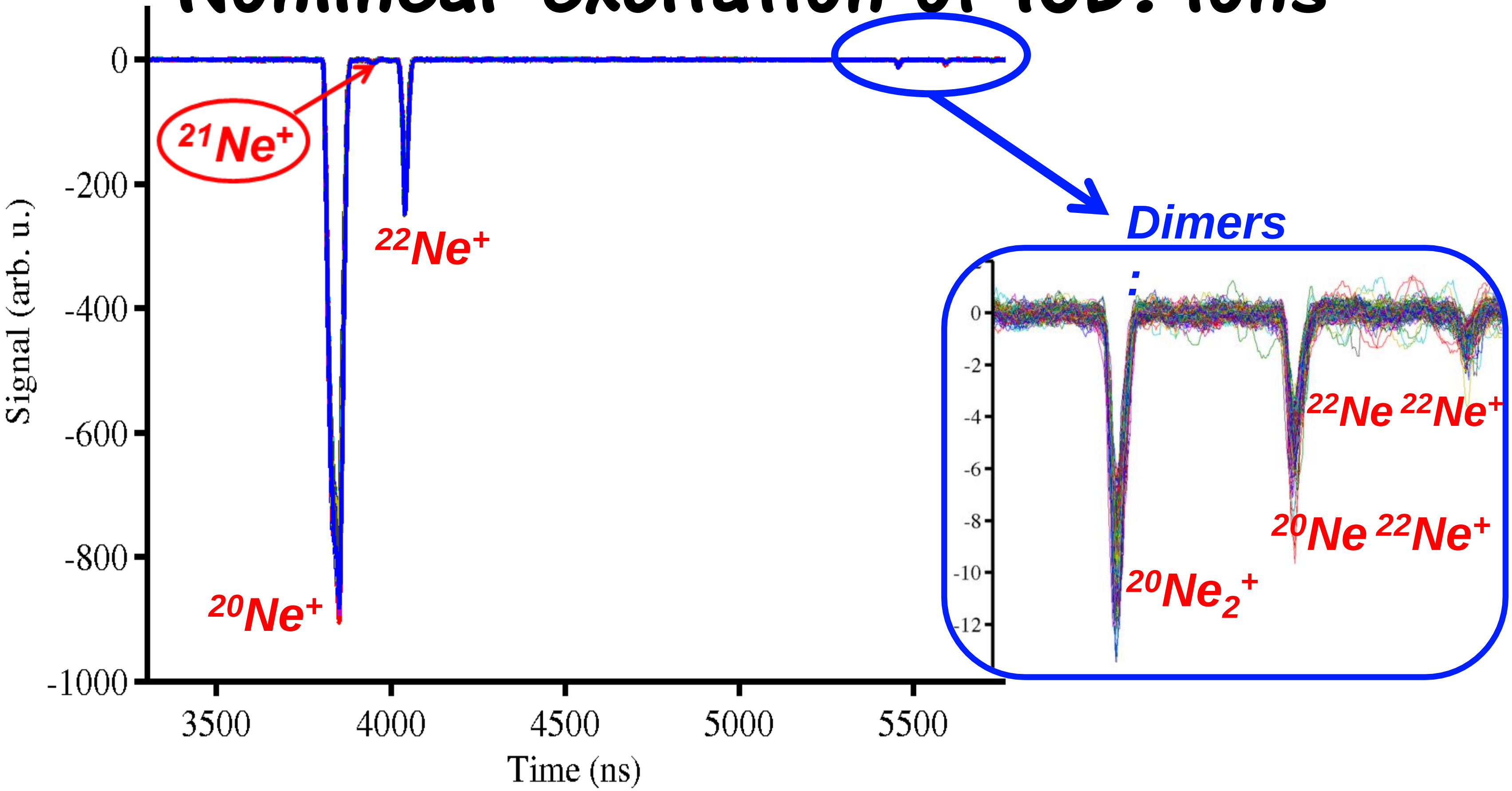


In the gas jet 1% of neon dimer

The photoelectron signal is dominated by the single photon ionization of the monomer

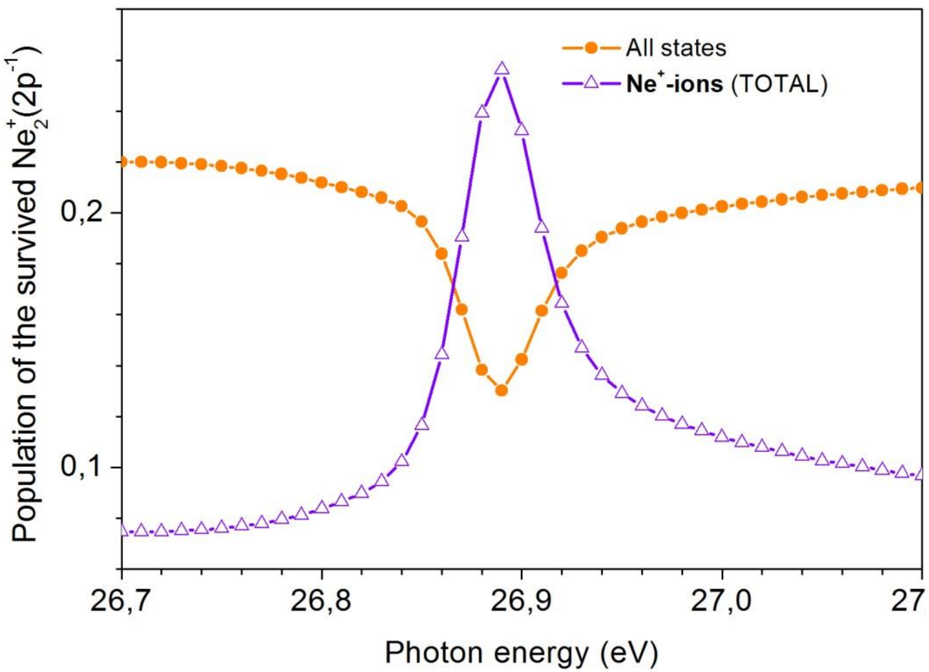
P. V. Demekhin *et al.*, Phys Rev Lett **107** (2011).

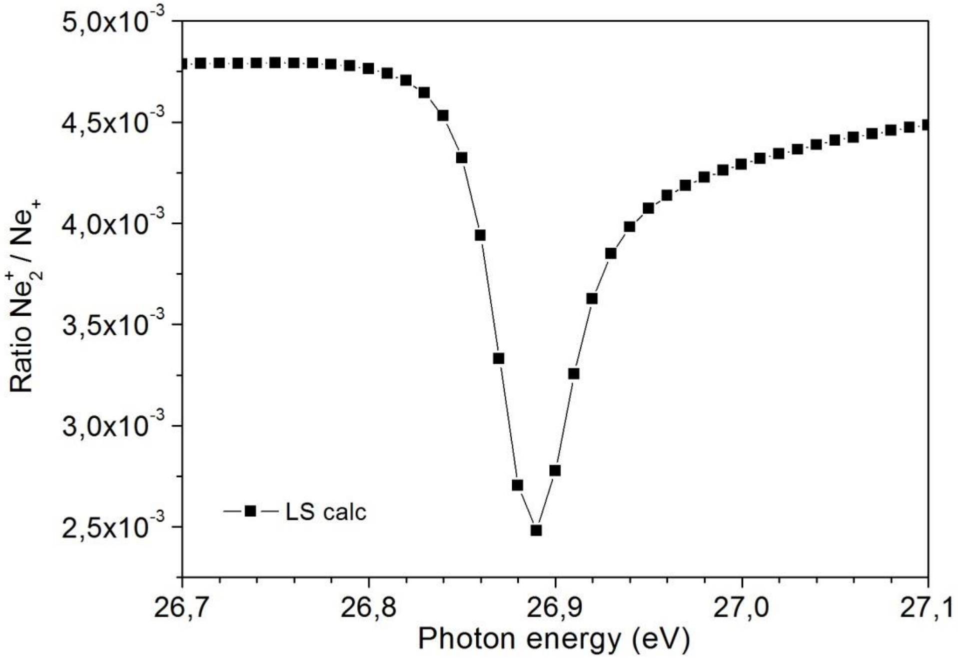
Nonlinear excitation of ICD: ions



	Neon monomer Ne^+	Neon dimers Ne^+_2
Off resonance	Weakly dependent on the photon energy	Weakly dependent on photon energy
On resonance (around 26.9 eV)	Weakly dependent on thephoton energy	**Reduction of neon dimers because ICD and Coulomb explosion**

Nonlinear excitation of ICD: dimers vs monomers

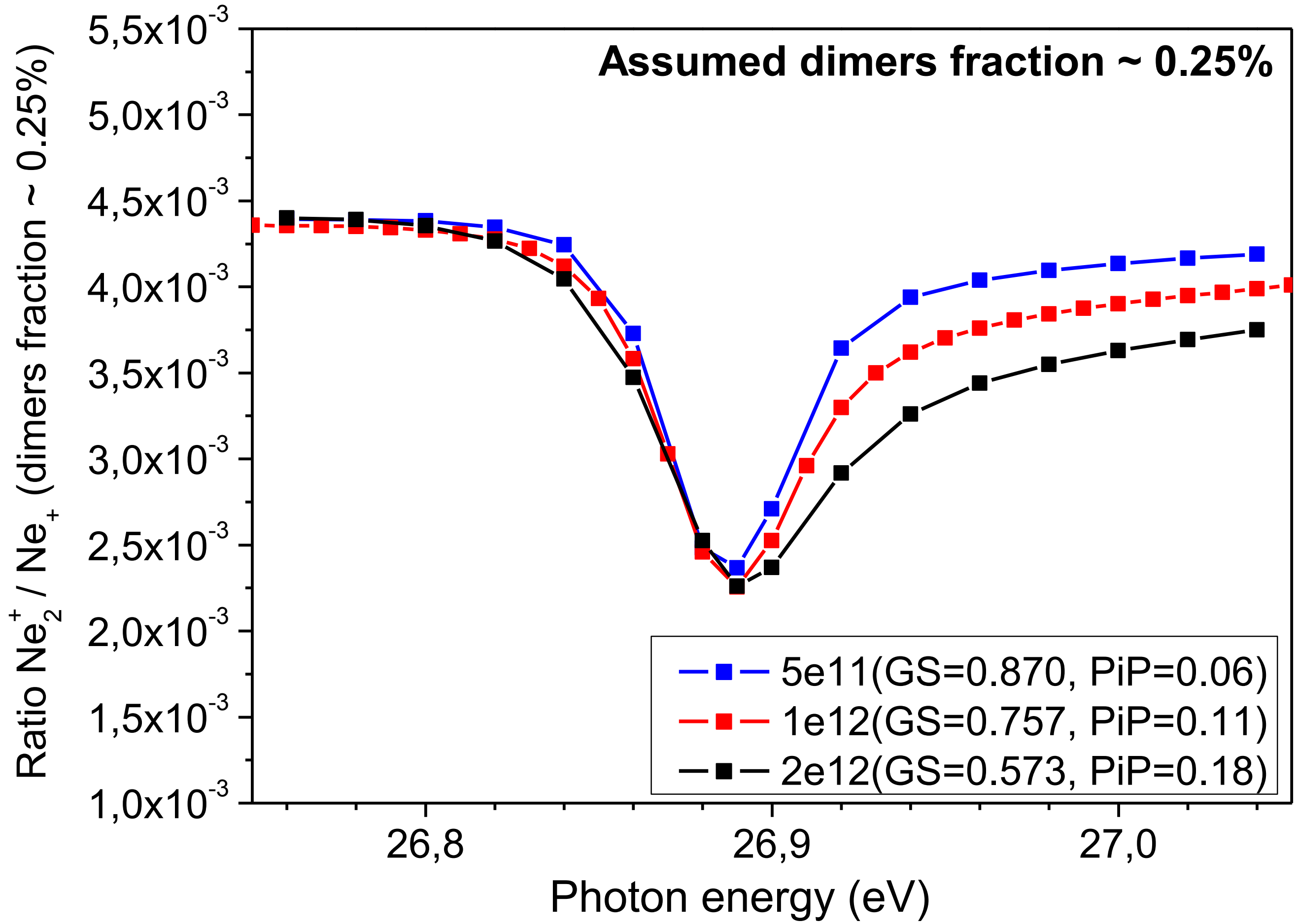




$$\frac{Ne^{+}{}_{2}}{Ne^{+}} = \frac{Ne^{+}{}_{2}}{Ne^{+}(monomers) + Ne^{+}(dimers)} \backsimeq \frac{Ne^{+}{}_{2}}{Ne^{+}(monomers)}$$

Ratio dimers/monomers (theory)

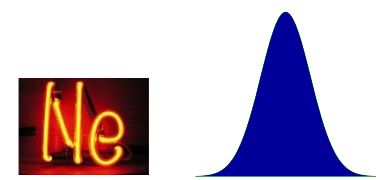
$$\textbf{Ratio} = \frac{Ne^{+}{}_{2}}{Ne^{+}} = \frac{Ne^{+}{}_{2}}{Ne^{+}(monomers) + Ne^{+}(dimers)} \backsimeq \frac{Ne^{+}{}_{2}}{Ne^{+}(monomers)}$$



Experiments

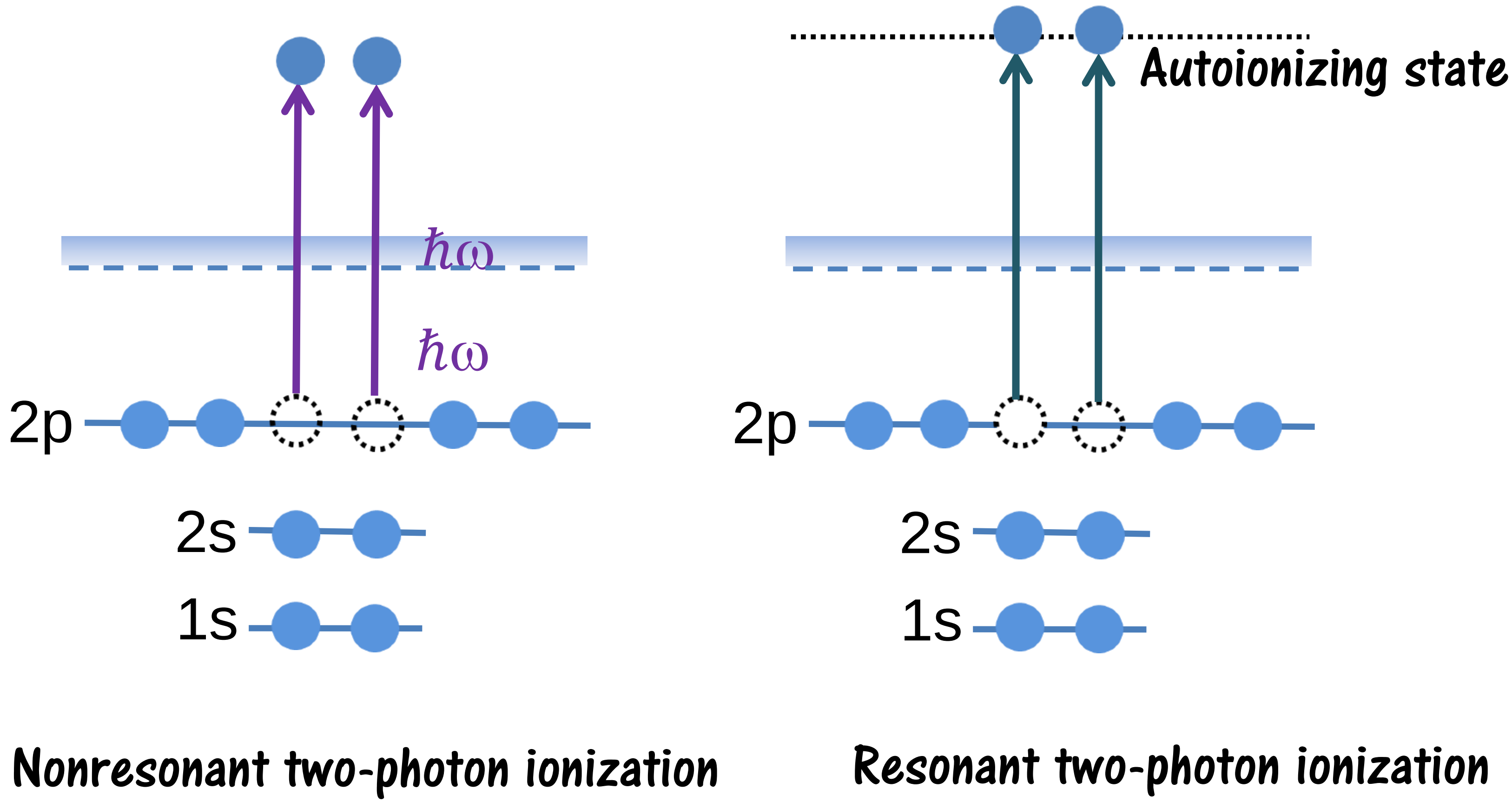
1) XUV nonlinear excitation of neon dimers (FEL)

2) Two-photon double ionization of neon



Motivation

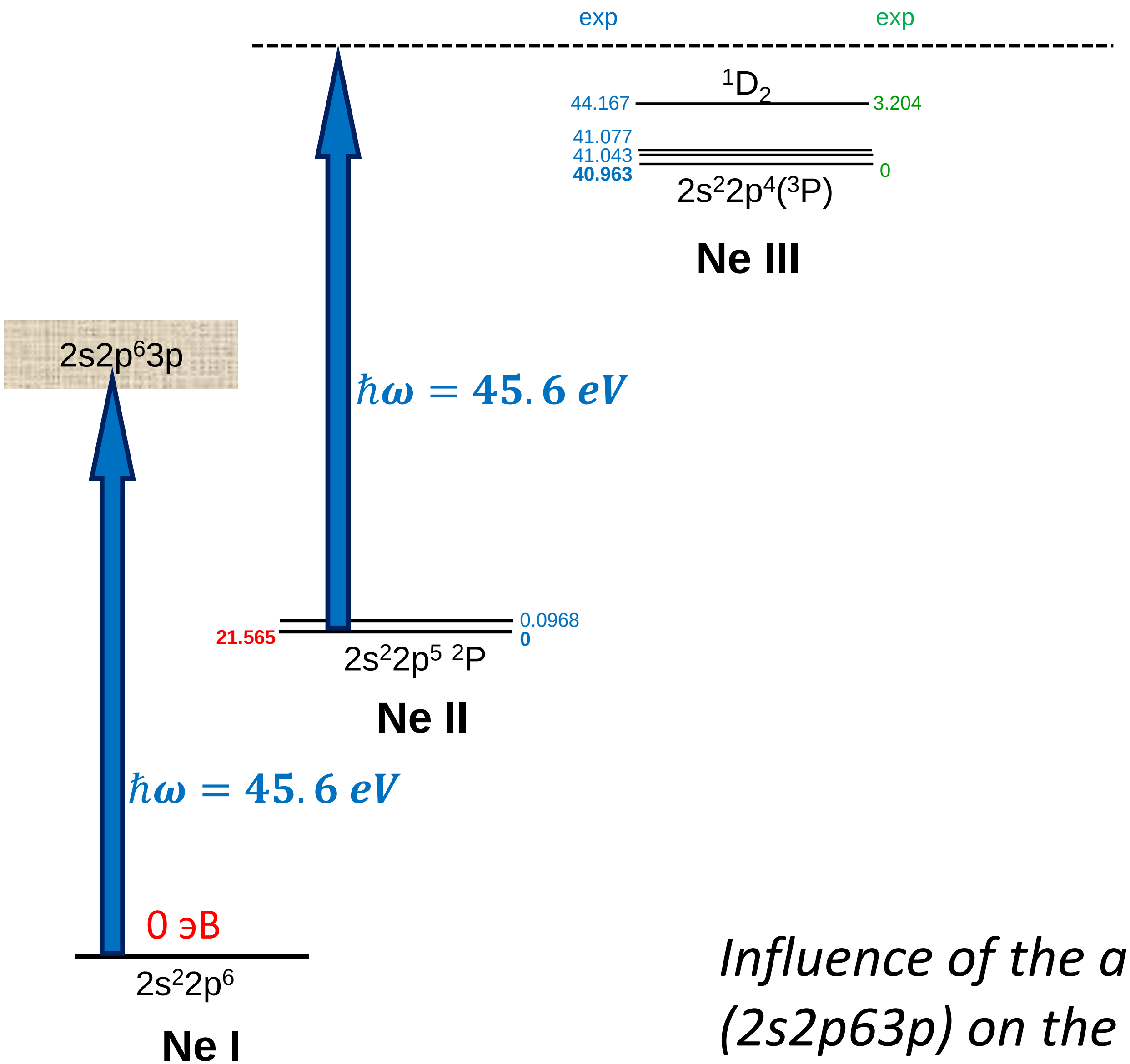
Resonant (vs nonresonant) two-photon double ionization in neon atoms

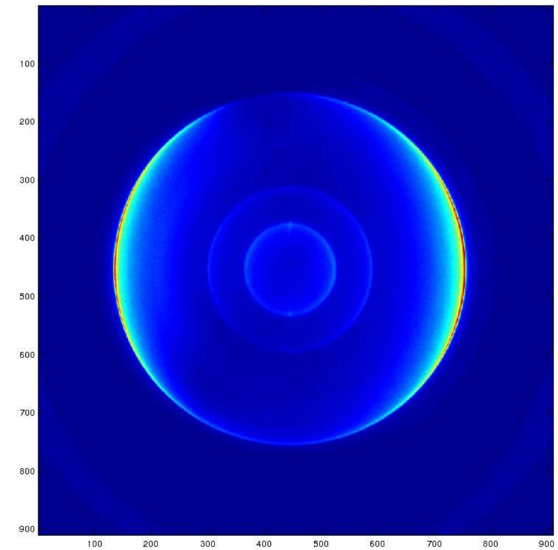


- *Electron yield*
- *Angular distribution*

Towards time-resolved dynamics (pump-probe experiment)

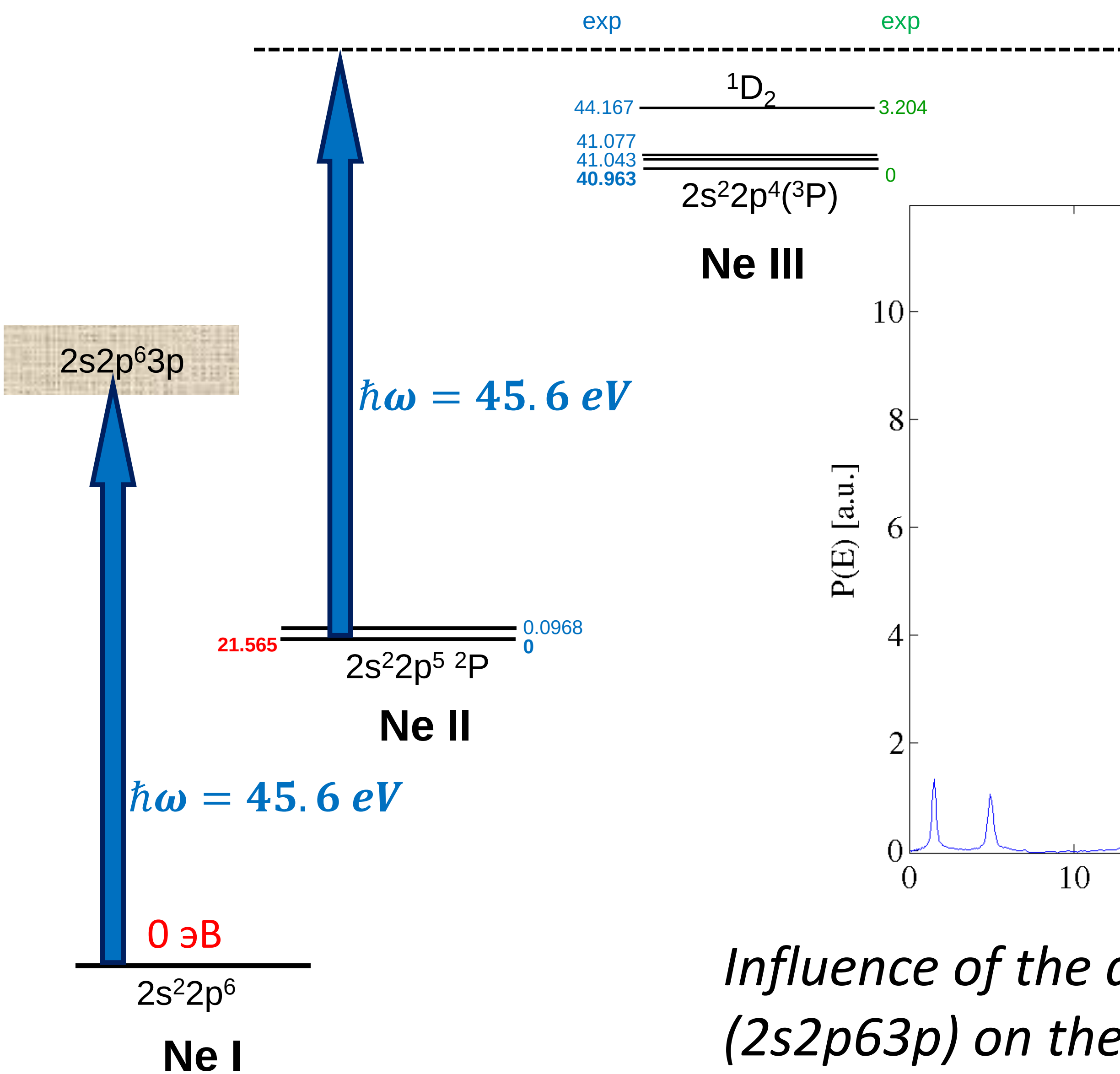
Two-photon double ionization of Neon

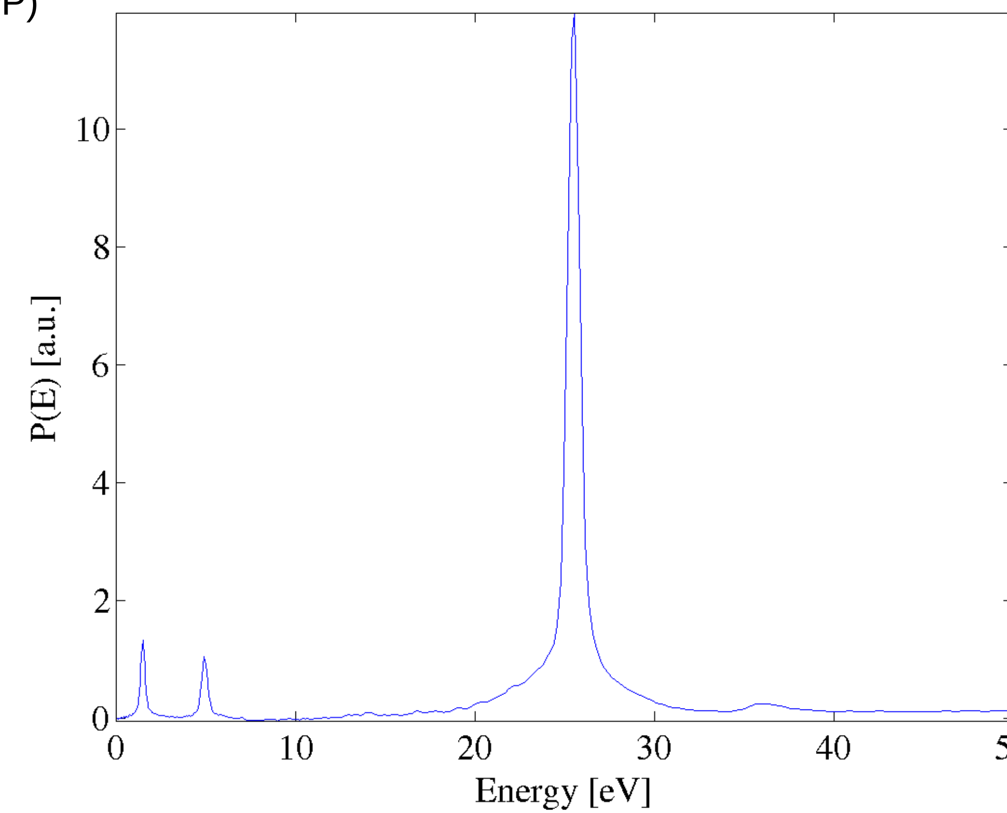




Influence of the autoionization resonance (2s2p63p) on the second ionization step

Two-photon double ionization of Neon





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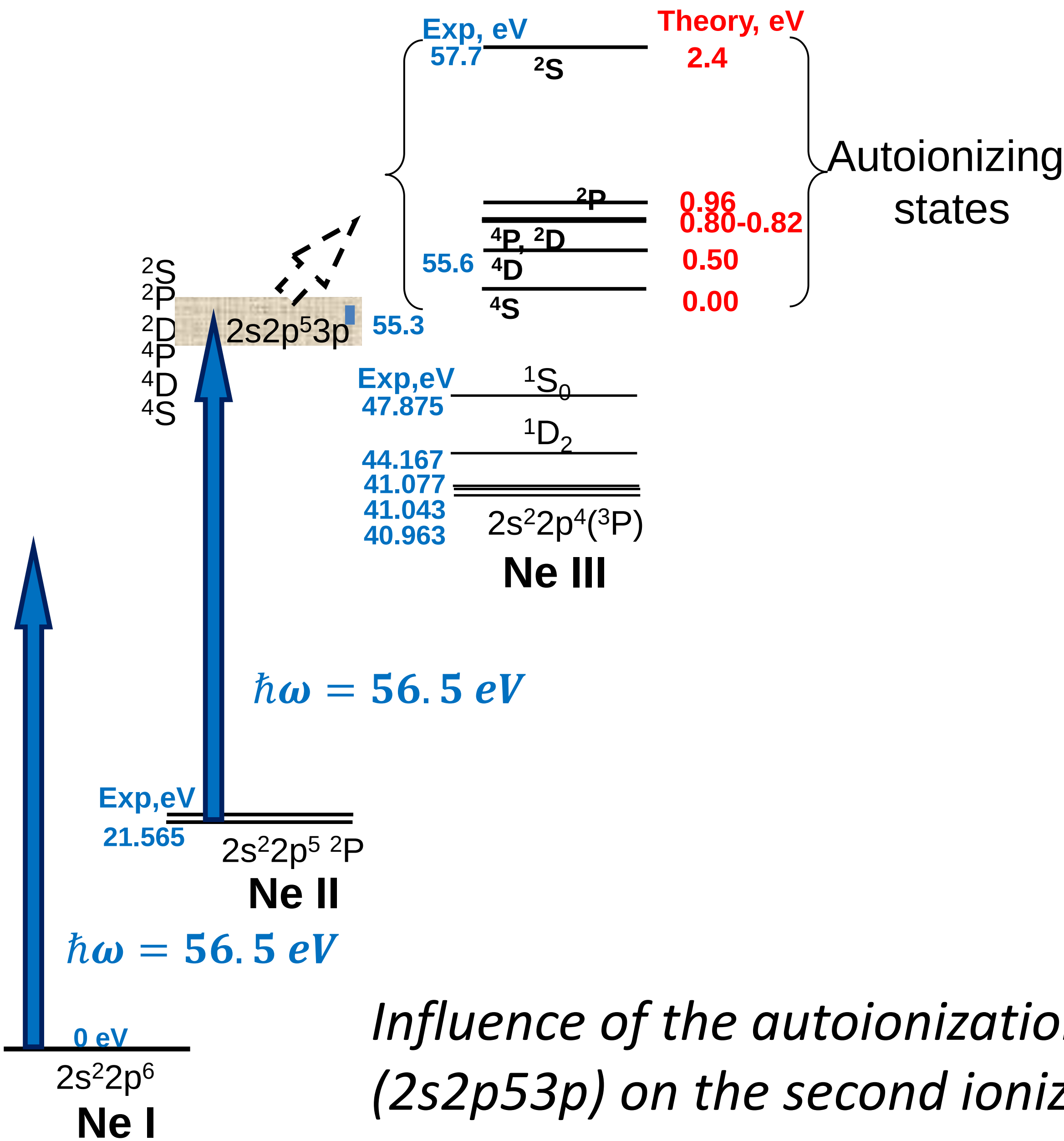
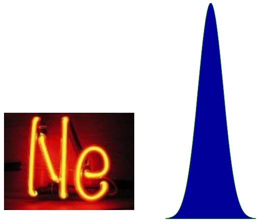


Influence of the autoionization resonance (2s2p53p) on the second ionization step

Conclusions and outloook

1) Nonlinear excitation of neon dimers

2) Two-photon double ionization of neon



- I) FERMI: pump-probe experiments for time-resolved ICD dynamics

- II) FERMI: theoretical simulations and data interpretation for two-photon double ionization