.THE ROUTE TO DIRECT MULTIPHOTON MULTIPLE IONIZATION

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A basic question in multiple ionization

- If you submit any system with bound electrons to an EM field of any photon energy, given sufficient intensity and appropriate pulse duration, it will eject several electrons; a fact known for 40 years.
- A question that arose, explored and debated at least since the mid-80s, is whether the electrons are ejected sequentially, or more than one at the "same time", i.e. without a succession of ionic stages.
- This is what I will be discussing in this talk.

VOLUME 54, NUMBER 14 PHYSICAL REVIEW LETTERS 8 APRIL 1985

Atomic Inner-Shell Excitation Induced by Coherent Motion of Outer-Shell Electrons

K. Boyer and C. K. Rhodes
Department of Physics, University of Illinois at Chicago, Chicago, Illinois 60680
(Received 28 January 1985)

Outer-shell electrons coherently driven by intense radiation can transfer energy in a direct intra-atomic process to inner-shell excitations. Provided that the effective momentum transfer Δq is sufficiently low ($\Delta q \lesssim \hbar / a_0$), the amplitudes governing the coupling of the outer electrons to the atomic core constructively sum. The effective cross section, which can be related to fast atom-atom collisions ($\gtrsim 10$ MeV/u), is evaluated in a limiting form closely resembling the Bethe result for inelastic electron scattering from atoms.

PRL **102**, 163002 (2009) PHYSICAL REVIEW LETTERS week ending 24 APRIL 2009

Extreme Ultraviolet Laser Excites Atomic Giant Resonance

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(Received 15 January 2009; published 24 April 2009)

Exceptional behavior of light-matter interaction in the extreme ultraviolet is demonstrated. The photoionization of different rare gases was compared at the free-electron laser in Hamburg, FLASH, by applying ion spectroscopy at the wavelength of 13.7 nm and irradiance levels of thousands of terawatts per square centimeter. In the case of xenon, the degree of nonlinear photoionization was found to be significantly higher than for neon, argon, and krypton. This target specific behavior cannot be explained by the standard theories developed for optical strong-field phenomena. We suspect that the collective giant $4d$ resonance of xenon is the driving force behind the effect that arises in this spectral range.

Multiple Ionization of Xenon by UV Radiation of ps duration and intensity up to 10^16 W/cm^2, around 1983-84

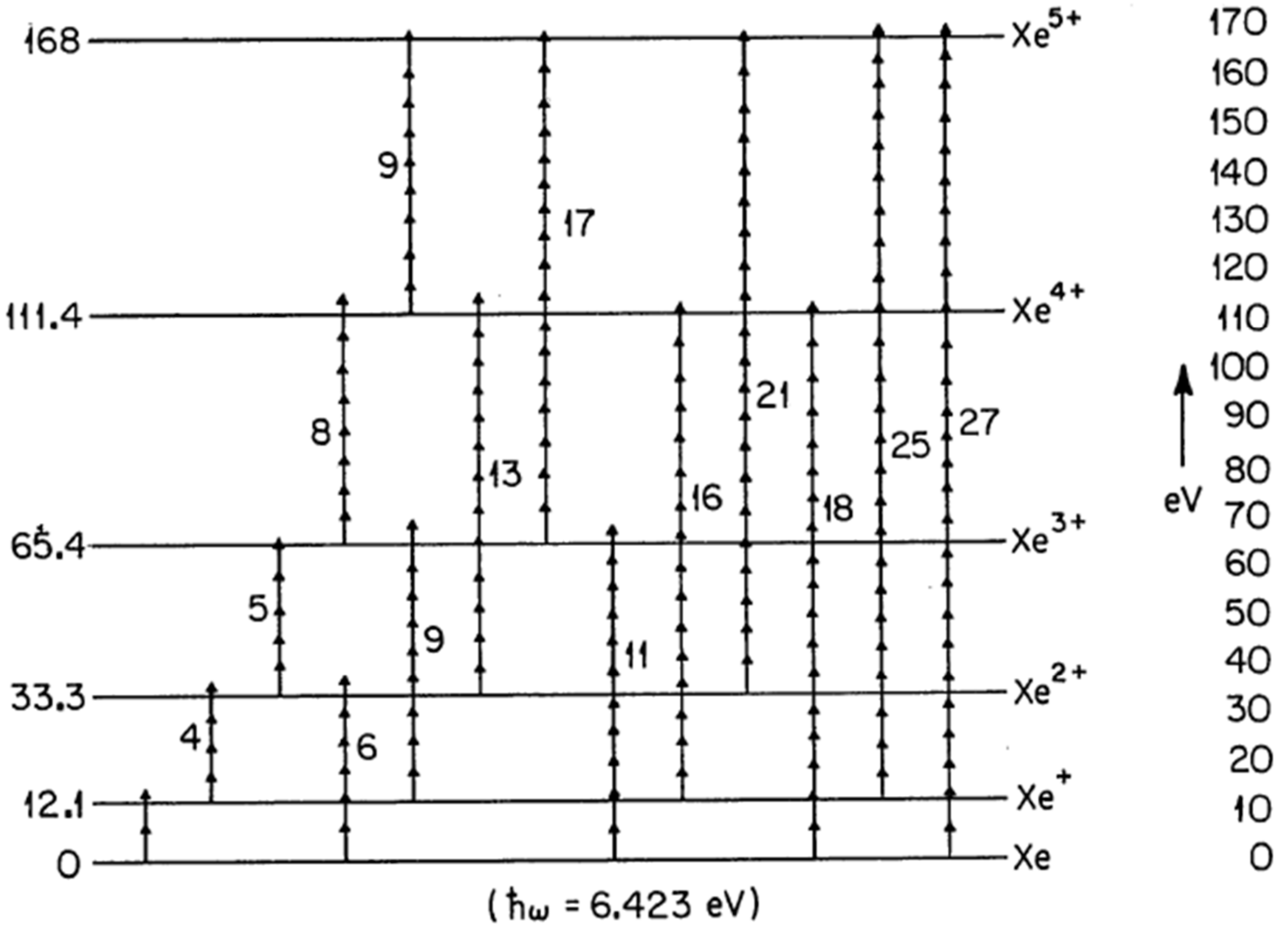
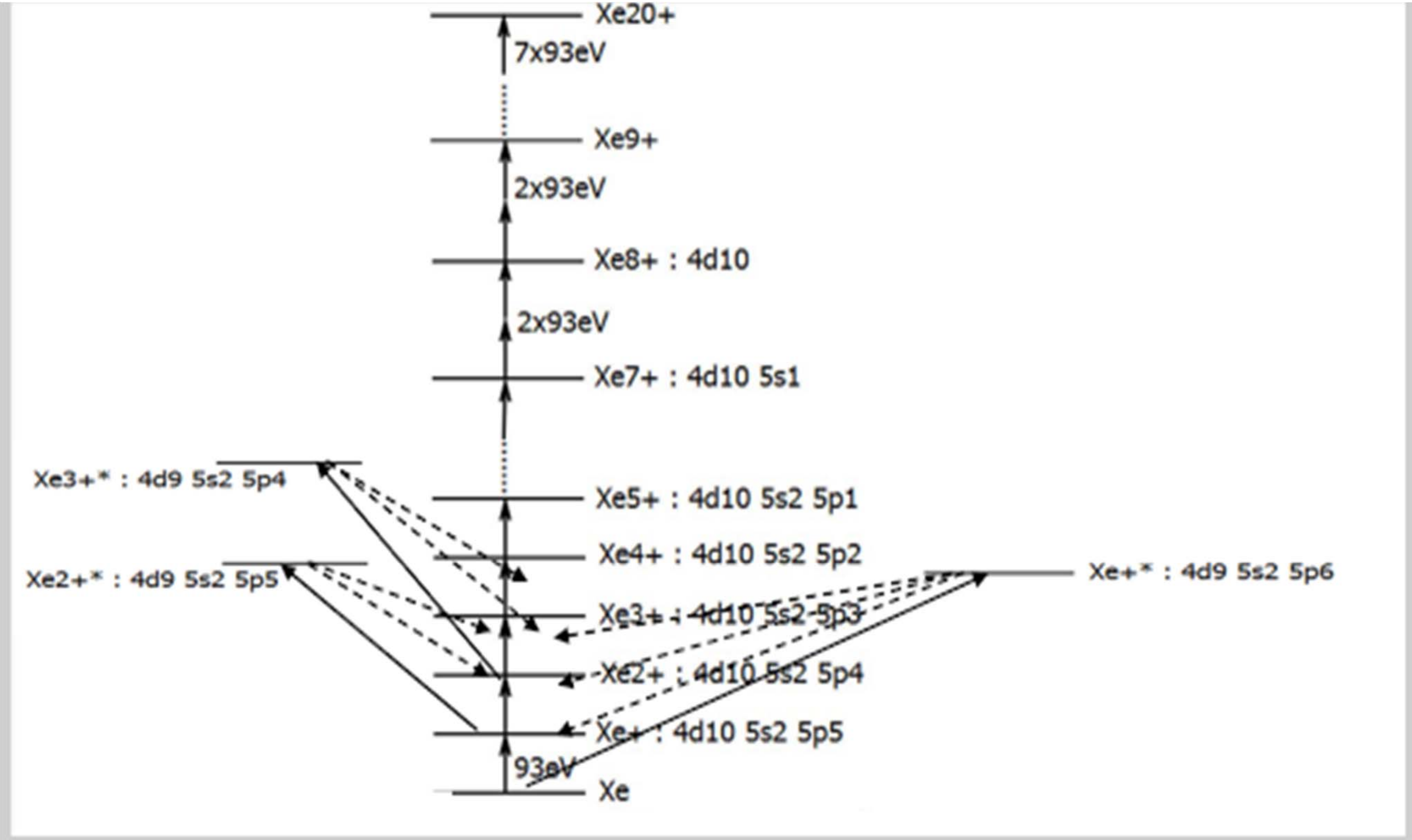


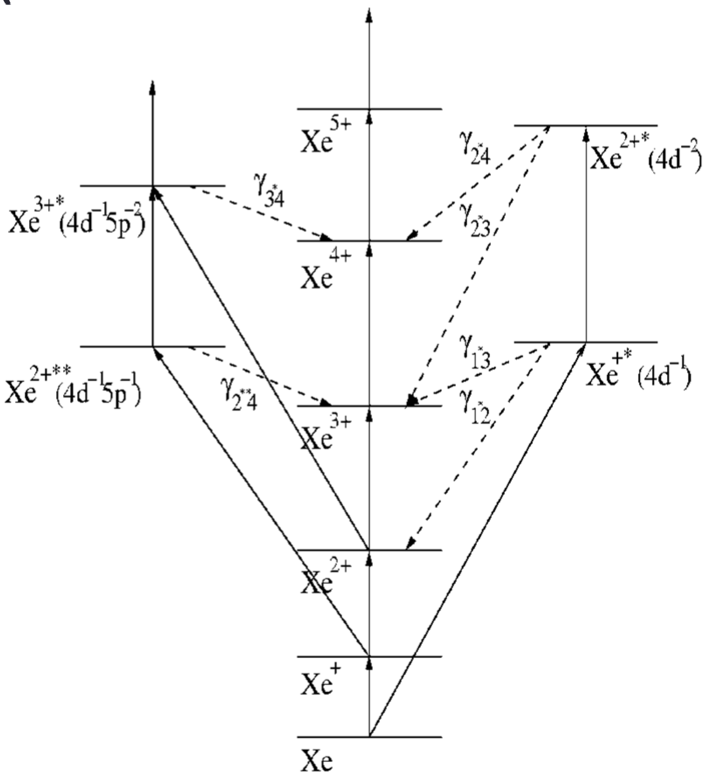
Fig. 3. Ionization potential of Xe and its first four ions. Also shown are the orders of ionization corresponding to sequential and direct pathways for photons of 198 nm ($\hbar\omega$ = 6.43 eV).

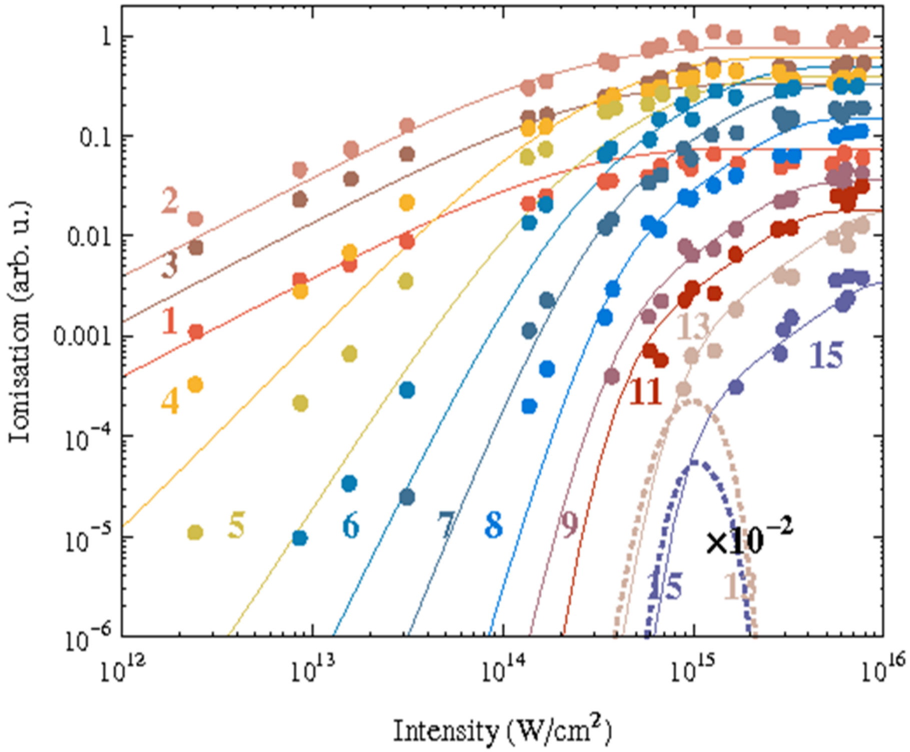
Xenon under 93 eV Radiation (FLASH)



Xenon under 93 eV radiation

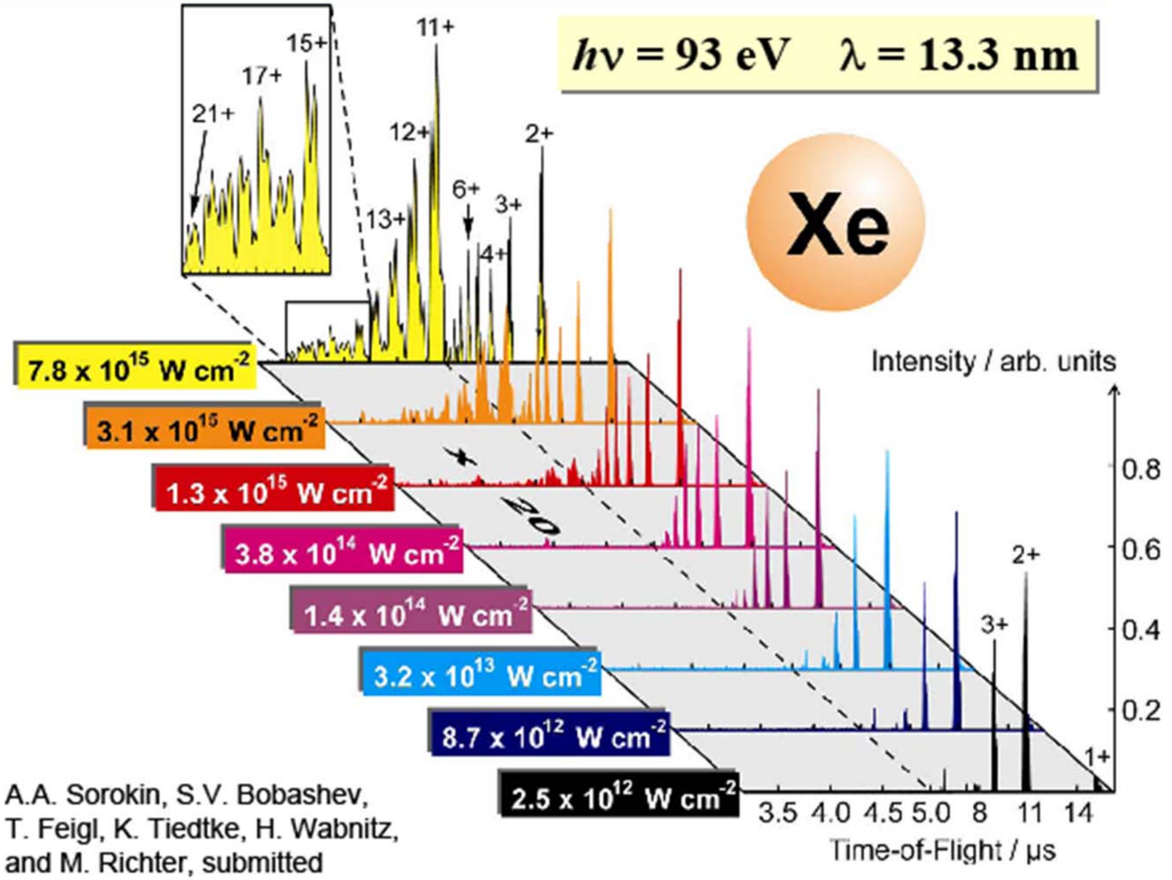
- Data: Sorokin et al. PRL 99, 213002 (2007) – (Photoelectric Effect at Ultrahigh Intensities)





Theory: Makris, Mihelic and PL , PRL 102, 033002 (2009)

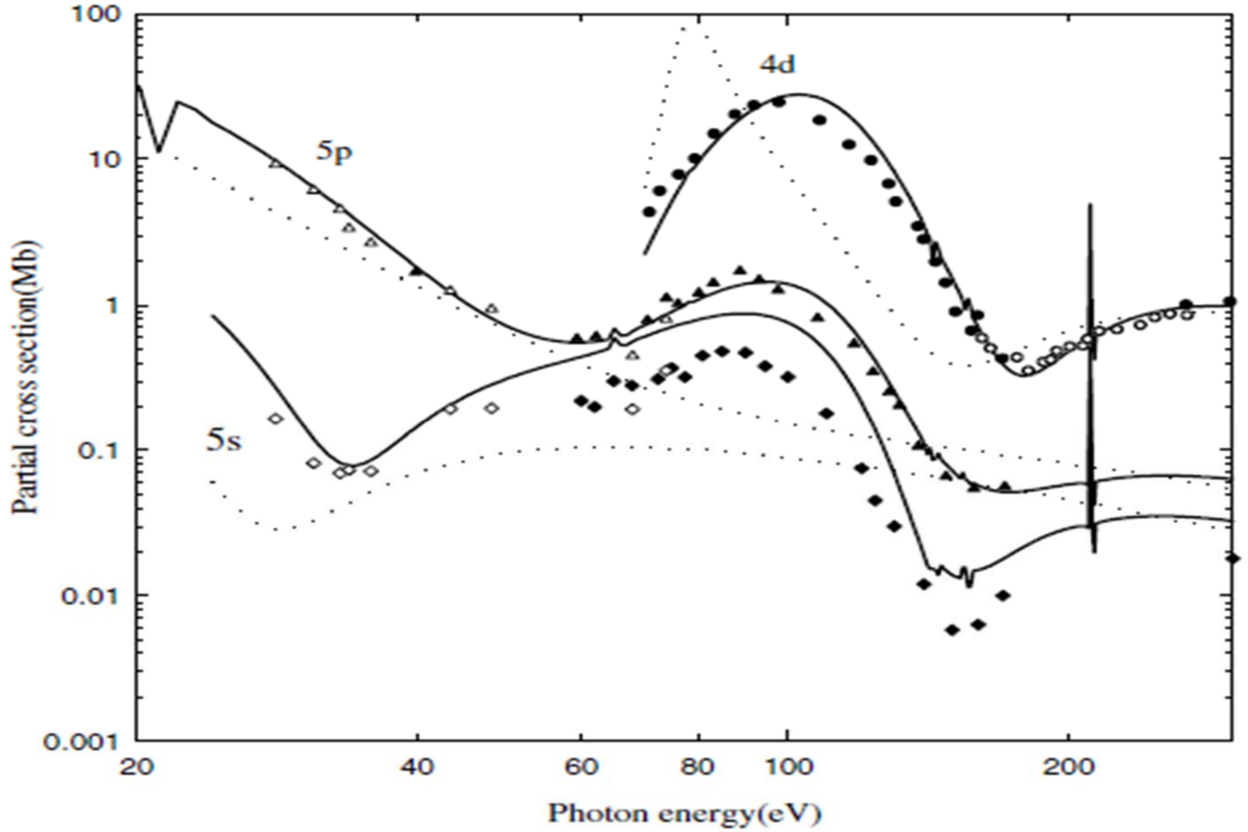
Photoionization of xenon atoms in the EUV at ultra-high intensities: ion time-of-flight spectra



A.A. Sorokin, S.V. Bobashev, T. Feigl, K. Tiedtke, H. Wabnitz, and M. Richter, submitted

The Giant Resonance in Xenon (Toffoli et al JPB 35, 1275 (2002))

- It has nothing to do with Xe(21+) under 93 eV. (see PL et al. JPB 44, 175402 (2011))

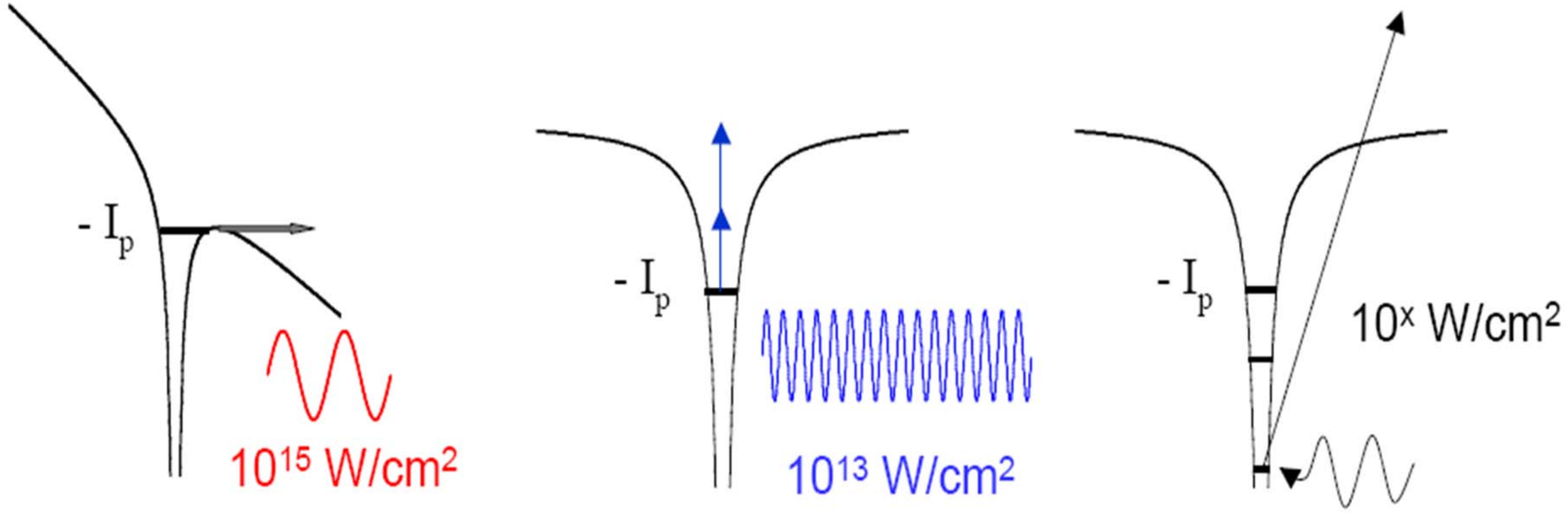


Bound electrons in EM fields

IR: quasistatic description

VUV FEL: intense photon source

XFEL FEL: highly ionizing



IR:

- Keldysh parameter Γ<<1
- Tunnel / over the barrier ionisation
- Ponderomotive energy 10 – 100 eV

VUV FEL:

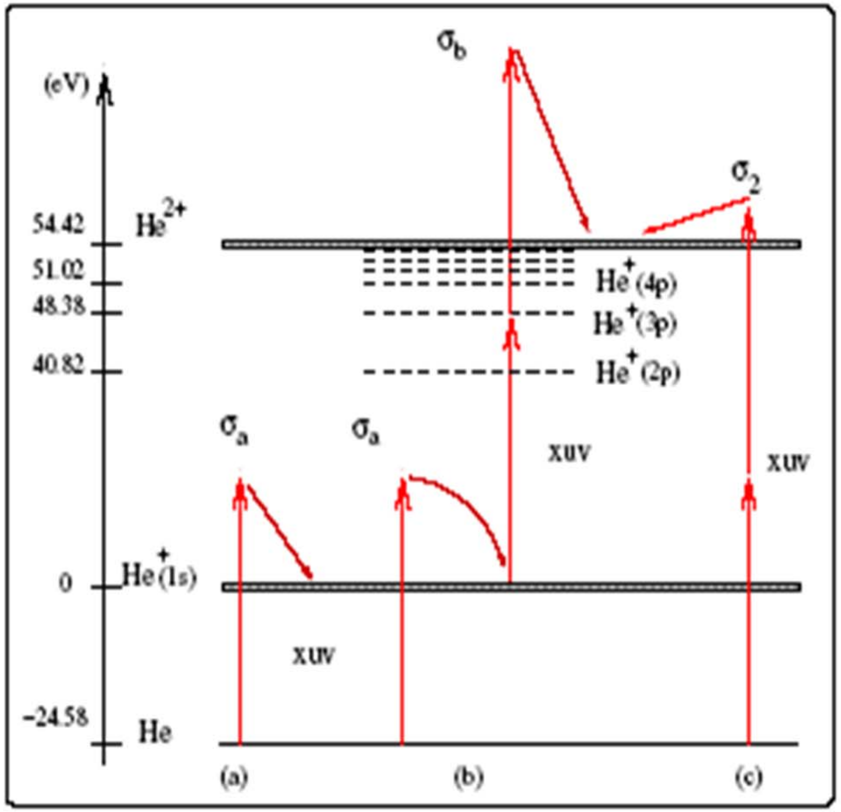
- Keldysh parameter Γ>>1
- Multi-photon ionisation
- Ponderomotive energy 10 meV

XFEL FEL:

- Angstrom wavelength
- Direct ionisation
- Secondary processes

2-photon double ionization of He

Channels of single and double ionization



RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 44, NUMBER 1 1 JULY 1991

Theory of the photoelectron spectrum in double ionization through two-photon absorption from He($2s^2$)

H. Bachau* and P. Lambropoulos

Department of Physics, University of Southern California, Los Angeles, California 90089-0484

(Received 15 February 1991)

We analyze in this paper the role of the correlations during double-electron ejection through two-photon absorption from the lowest autoionizing state $^1S^e$ of helium. Emphasis will be put on the pertinence of the notion of simultaneity for double-electron ejection when this process does not require electron interaction.

J. Phys. B: At. Mol. Opt. Phys. **32** (1999) L603–L613. Printed in the UK PII: S0953-4075(99)07080-7

LETTER TO THE EDITOR

Photoelectron energy spectrum in 'direct' two-photon double ionization of helium

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and

Department of Physics, University of Crete, Greece

T. NAKAJIMA AND L. A. A. NIKOLOPOULOS PHYSICAL REVIEW A **66**, 041402(R) (2002)

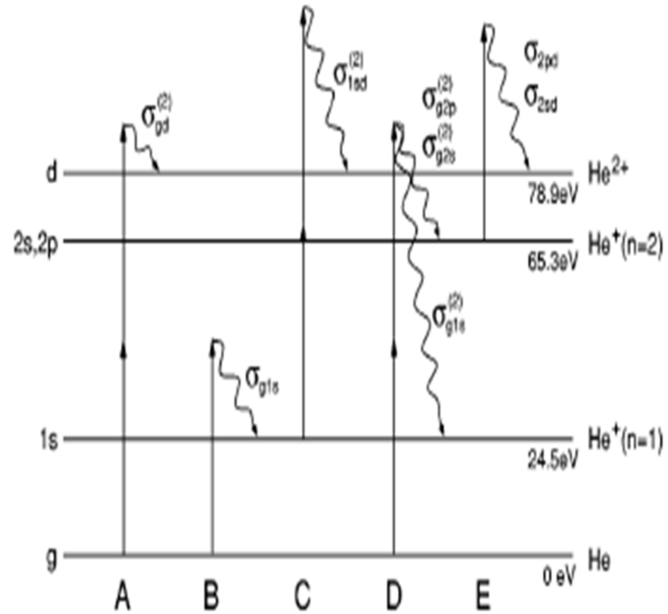


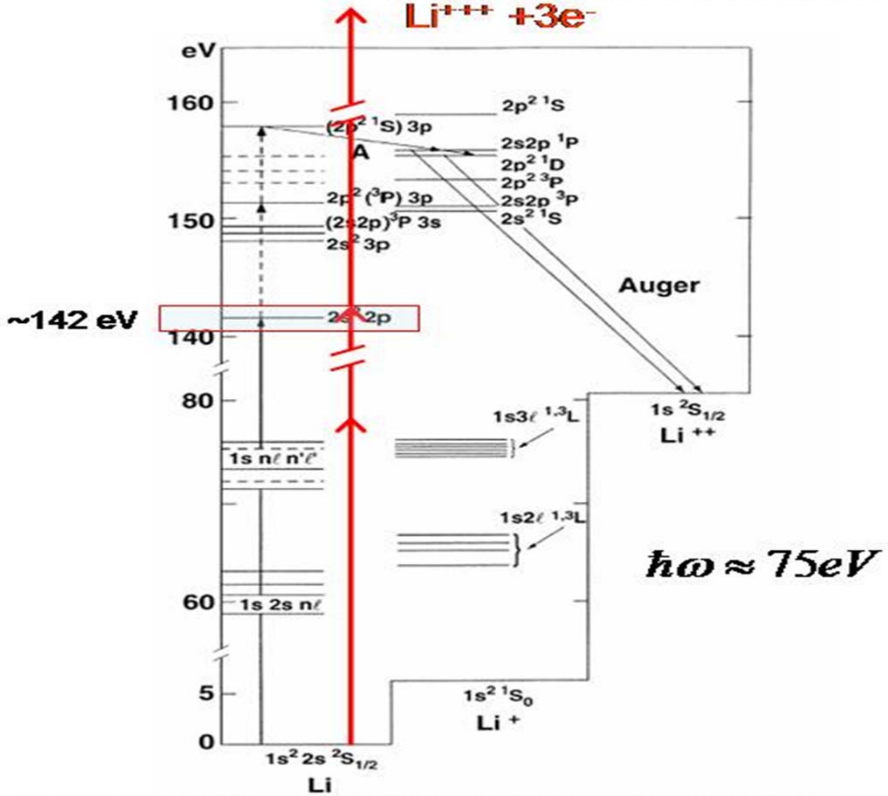
FIG. 1. Level scheme considered in this paper.

For the present study, we specifically assume the photon energy of 45 eV. Using a multichannel B-spline code developed in recent years [19,20], we obtain the cross sections as $\sigma_{gd}^{(2)}=8.1\times10^{-52}$ cm^4 s, $\sigma_{g1s}^{(2)}=1.0\times10^{-52}$ cm^4 s, $\sigma_{g2s}^{(2)}=2.3\times10^{-51}$ cm^4 s, $\sigma_{g2p}^{(2)}=3.8\times10^{-51}$ cm^4 s, $\sigma_{g1s}=2.4\times10^{-18}$ cm^2, $\sigma_{1sd}^{(2)}=1.0\times10^{-53}$ cm^4 s, $\sigma_{2sd}=2.4\times10^{-19}$ cm^2, and $\sigma_{2pd}=9.0\times10^{-20}$ cm^2. In our scheme the

Also Tang & PL, PRL 58, 108 (1987). 4-photon, 2e ionization in Carbon.

A simple generalization: Direct, 3-photon, Triple Ionization of Li

Diehl et al. J. Phys. B 30, L 595 (1997)



Schematic energy level diagram for neutral lithium, Li^{+} and Li^{2+} in the energy region corresponding to the production of hollow Li and hollow Li^{+} states. A selection of the hollow atom and hollow ion levels are shown to illustrate the excitation (indicated by the upward pointing arrow) and decay paths (indicated by the arrows A and Auger)

That, namely Li at 75-90 3V, should be possible with current parameters at FLASH.

But why not something more ambitious…like

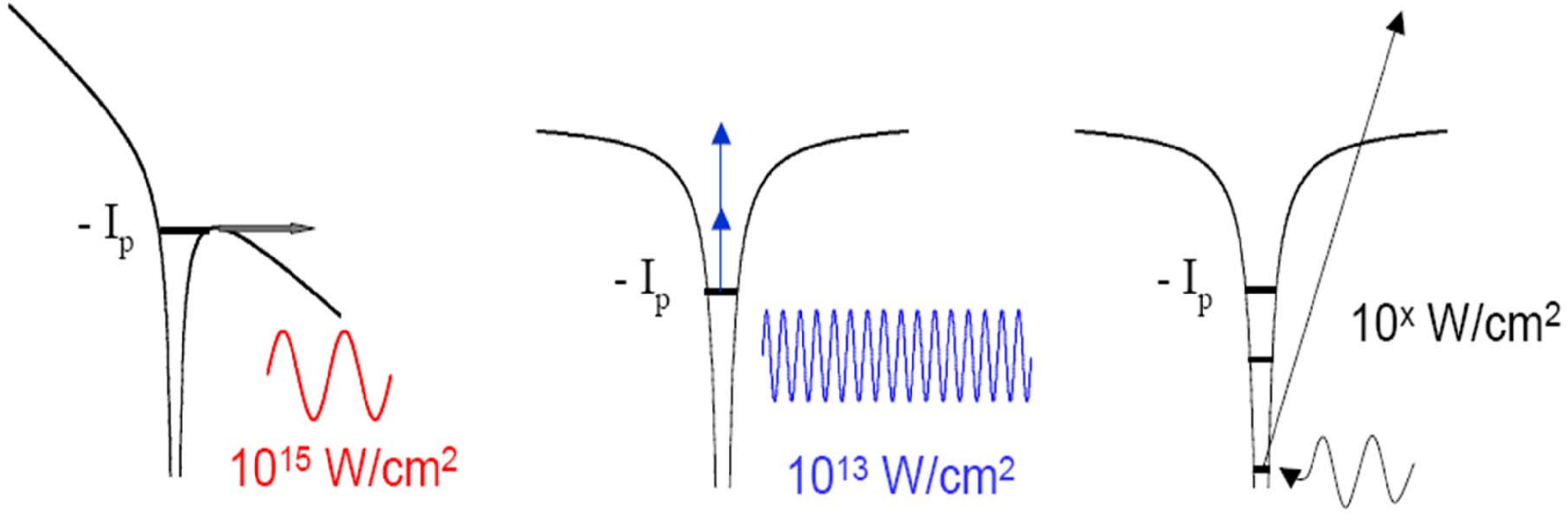
Direct 6-photon 6-electron ejection in …
let us say Neon; or,

why not, N-photon
18-electron (4d,5s,5p shells) ejection in Xe.

Let us take a look;after a short qualitative discussion.

Bound electrons in EM fields

IR: quasistatic description | **VUV FEL:** intense photon source | **XFEL FEL:** highly ionizing



IR:
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VUV FEL:
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- Multi-photon ionisation
- Ponderomotive energy 10 meV

XFEL FEL:
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- Direct ionisation
- Secondary processes

Measures of Intensity

Scaling of Ponderomotive Energy with Photon Energy

$\hbar\omega(eV)$	λ (nm)	$U_p(I=10^{13}W/cm^2)$	$I(U_p=\hbar\omega)(W/cm^2)$
1	1242.00	1.27 eV	$7.8x10^{12}$
10	124.00	$1.27x10^{-2}$	$7.8x10^{14}$
30	41.33	$1.44x10^{-3}$	$1.6x10^{17}$
50	24.80	$5x10^{-4}$	$1.1x10^{18}$
75	16.53	$2x10^{-4}$	$3.75x10^{18}$
100	12.40	$1.27x10^{-4}$	$7.87x10^{18}$
150	8.26	$5.65x10^{-5}$	$2.65x10^{19}$

Ionization Rates under LOPT

- PonderomotiveEnergy: $U_p \approx I/\omega^2 \ll \omega$
- Single-photon Ionization Rate: $W \approx \sigma I$
- N-photon Ionization Rate: $W_N \approx \sigma_N I^N$?
- Under these conditions a log-log plot of ion signal versus intensity should be a straight line of slope N. If not, something is wrong ?
- Keldysh parameter $\gamma > 1$ under these conditions

N-photon Ionization Cross Section

- General formal expression. (Generalized Cross Section)

$$\sigma^{(N)} = \frac{(2\pi\alpha)^N}{4\pi^2}\frac{mk}{\hbar}\omega^N \left| \sum\!\!\!\!\!\!\int_{a_{N-1}} \cdots \sum\!\!\!\!\!\!\int_{a_1} \frac{\langle f_{\mathbf{k}}|\mathbf{r}\cdot\hat{\epsilon}|a_{N-1}\rangle \ldots \langle a_1|\mathbf{r}\cdot\hat{\epsilon}|g\rangle}{[E_{a_{N-1}} - E_g - (N-1)\hbar\omega]\ldots(E_{a_1} - E_g - \hbar\omega)} \right|^2$$

The above equation involves summation over N-1 complete sets of atomic/molecular states, with final state in the continuum.

- Can something like this be calculated for, say, N=16 ?

Well….actually, yyyyes. Scaling helps.

Calculated 6-photon ionization (generalized) cross section of hydrogen

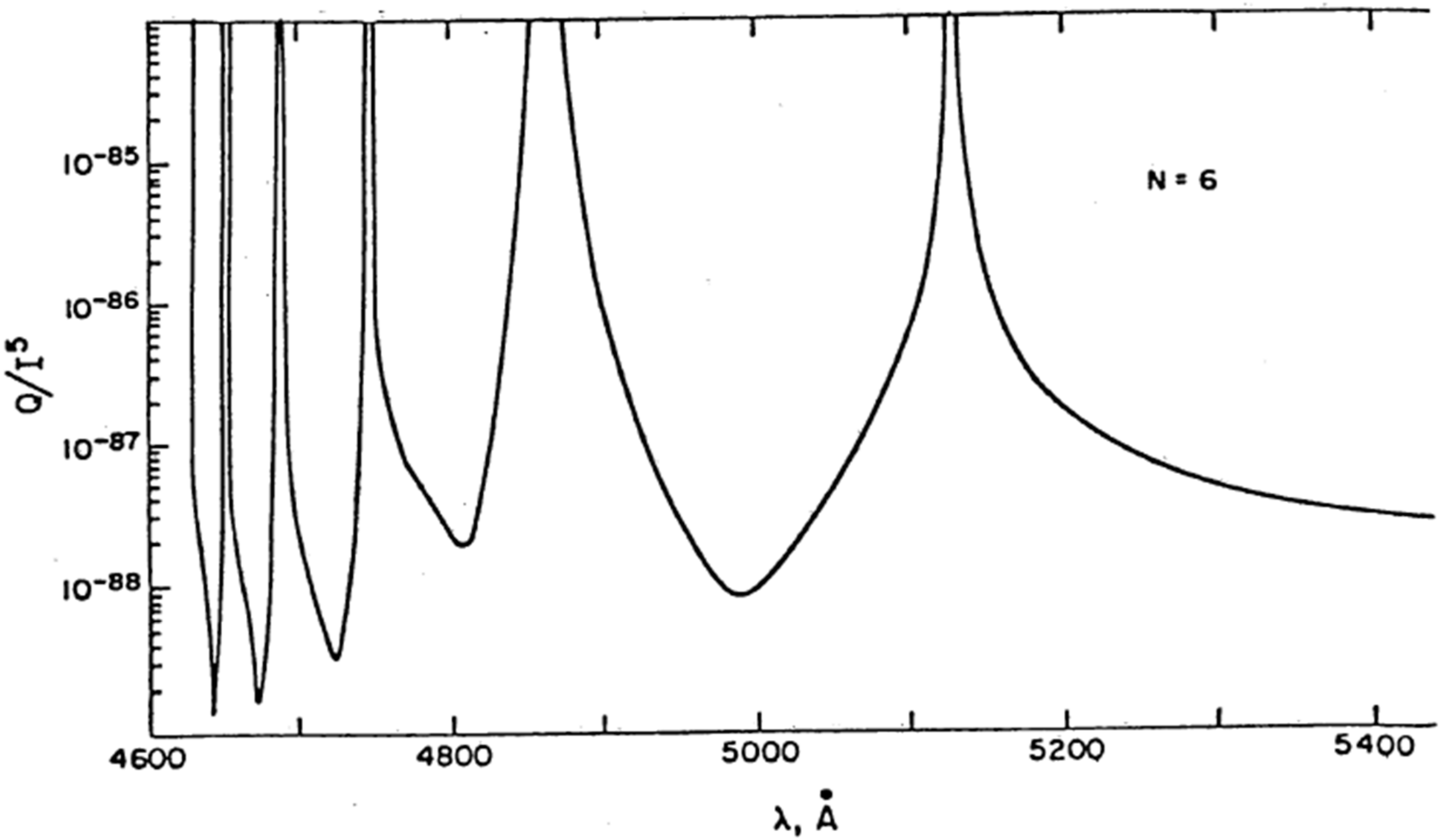

Fig. 1. Six-photon ionization generalized cross section of 1*s* H as calculated by Karule.[18]

Multiple excitation and ionization of atoms by strong lasers

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Department of Physics, University of Southern California, Los Angeles, California 90089-0484

Received December 29, 1986; accepted February 7, 1987

Experimental information on laser-induced multiple ionization is analyzed in terms of a model that includes the scaling of the generalized cross sections and the time dependence during the pulse. A comparison of direct with sequential processes reveals the dominance of the latter. The role of multiply excited states in strong-laser studies is also discussed.

PHYSICAL REVIEW A VOLUME 59, NUMBER 6 JUNE 1999

Scaling of hydrogenic atoms and ions interacting with laser fields: Positronium in a laser field

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[2]*Foundation for Research and Technology Hellas, Institute of Electronic Structure and Laser, P.O. Box 1527, 71110 Heraklion, Crete, Greece*

(Received 9 December 1998)

Scaling laws are derived for hydrogenlike atoms and ions interacting with laser fields. In particular, the scaling of (appearance) intensities is derived. This scaling is independent of the physical mechanism responsible for the ionization process, be it tunneling or multiphoton ionization. It provides a firm basis and explanation for the validity of earlier models for the estimation of appearance intensities and of the extrapolation into the tunneling regime of scaling laws obtained through lowest-order perturbation theory. As an example of the applicability of the scaling laws, we calculate two-, three-, and six-photon generalized cross sections and ionization rates for positronium at laser frequencies of current experimental interest. [S1050-2947(99)04806-4]

PACS number(s): 32.80.Rm, 36.10.Dr, 32.80.−t

N-photon Ionization Cross Sections σ_N

Gas	N	λ(nm)	σ_N	Pulse Duration	References
Cs	2	528	$(6.7 \pm 1.9) \times 10^{-50}$	19ns	D. Normand and J. Morellec, J. Phys. B **13**, 1551 (1980)
$He^*\ {}^2S^1$	2	347	$(2.7 \pm 2.0) \times 10^{-49}$	12ns	Lompré *et al.*, J. Phys. B **13**, 1799 (1980)
$He^*\ {}^2S^3$	2	347	$(1.5 \pm 1.4) \times 10^{-49}$	12ns	Lompré *et al.*, J. Phys. B **13**, 1799 (1980)
$He^*\ {}^2S^1$	3	694	$(3.3 \pm 1.9) \times 10^{-80}$	7ns	Lompré *et al.*, J. Phys. B **13**, 1799 (1980)
$He^*\ {}^2S^3$	3	694	$(3.0 \pm 2.2) \times 10^{-81}$	7ns	Lompré *et al.*, J. Phys. B **13**, 1799 (1980)
Cs	4	1056	$(7.5 \pm 2.8) \times 10^{-109}$	25ns	D. Normand and J. Morellec, J. Phys. B **13**, 1551 (1980)
Xe	6	532	6.0×10^{-172}	50ps	L'Huillier *et al*, Phys. Rev. A **27**, 2503 (1983)
Ne	10	532	1.0×10^{-303}	50ps	L'Huillier *et al*, Phys. Rev. A **27**, 2503 (1983)
He	11	532	4.0×10^{-338}	50ps	Lompré *et al.*, Phys. Letters **112**, 219 (1985)
O_2	6	532	1.0×10^{-176}	40ps	L'Huillier *et al*, Chemical Phys. Lett. **103**, 447 (1984)
N_2	7	532	1.0×10^{-207}	40ps	L'Huillier *et al*, Chemical Phys. Lett. **103**, 447 (1984)
Xe	11	1064	1.0×10^{-336}	50ps	L'Huillier *et al*, J. Phys. B **16**, 1363 (1983)
Kr	13	1064	1.0×10^{-407}	50ps	L'Huillier *et al*, J. Phys. B **16**, 1363 (1983)
Ar	14	1064	1.0×10^{-440}	50ps	L'Huillier *et al*, J. Phys. B **16**, 1363 (1983)
Ne	19	1064	1.0×10^{-622}	50ps	L'Huillier *et al*, J. Phys. B **16**, 1363 (1983)
He	22	1064	1.0×10^{-726}	50ps	L'Huillier *et al*, J. Phys. B **16**, 1363 (1983)
Xe^+	10	532	1.0×10^{-297}	50ps	L'Huillier *et al*, Phys. Rev. A **27**, 2503 (1983)
Ne^+	18	532	1.0×10^{-565}	50ps	L'Huillier *et al*, Phys. Rev. A **27**, 2503 (1983)
He^+	24	532	1.0×10^{-789}	50ps	Lompré *et al.*, Phys. Letters **112**, 219 (1985)

Scaling of Multiphoton Cross Sections

Cross section scaling

Given the calculated hydrogenic values $\Lambda_N^{(H)}$, for an atom A:

$$\Lambda_N^{(A)} = \Lambda_N^{(H)} \frac{R_A^2}{R_H^2} \frac{E_\infty^{(H)}}{E_\infty^{(A)}}$$

$$\sigma^{(1)} \sim 10^{-18}\, cm^2$$

$$\sigma^{(2)} \sim 10^{-49}\, cm^4 \cdot \sec$$

$$\sigma^{(3)} \sim 10^{-79}\, cm^6 \cdot \sec^2$$

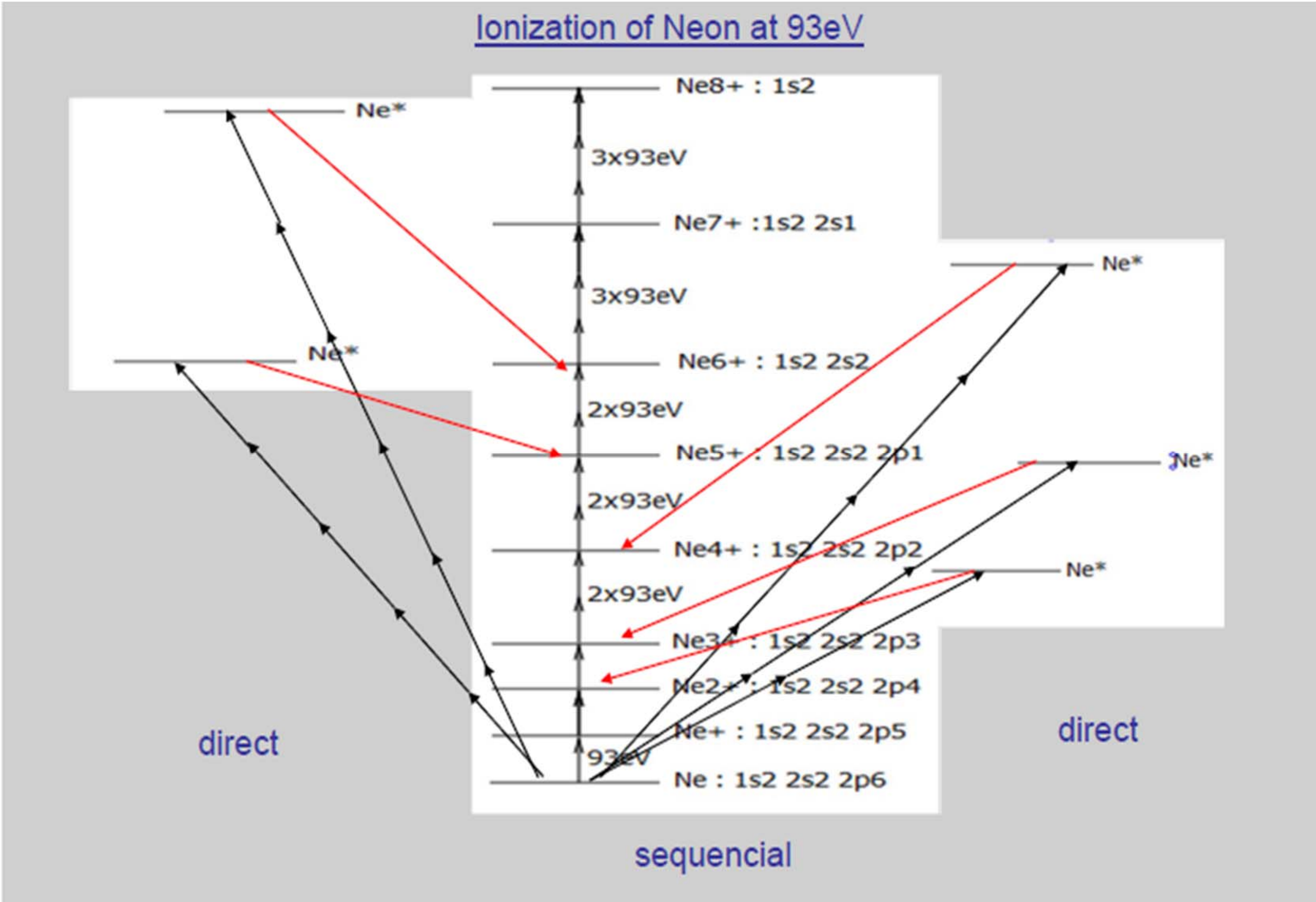
$$\sigma^{(4)} \sim 10^{-114} - 10^{-112}\, cm^8 \cdot \sec^3$$

$$\sigma^{(5)} \sim 10^{-143}\, cm^{10} \cdot \sec^4$$

$$\sigma^{(6)} \sim 10^{-172.8}\, cm^{12} \cdot \sec^5$$

$$\sigma^{(7)} \sim 10^{-203}\, cm^{14} \cdot \sec^6$$

Direct Multiple Ionization?
Neon at 93 eV (6-photon → 6e)



Mulitple Ionization of Neon at 93 eV

Sequential + Direct Channels

$$\dot{N}_0 = -(\sigma_{p01} + \sigma_{s01})FN_0 - \sigma_{2e}^{(2)}F^2N_0 - \sigma_{3e}^{(3)}F^3N_0$$
$$-\sigma_{4e}^{(4)}F^4N_0 - \sigma_{5e}^{(5)}F^5N_0 - \sigma_{6e}^{(6)}F^6N_0$$
$$\dot{N}_1 = (\sigma_{p01} + \sigma_{s01})FN_0 - (\sigma_{p12} + \sigma_{s12})FN_1$$
$$\dot{N}_2 = \sigma_{2e}^{(2)}F^2N_0 + (\sigma_{p12} + \sigma_{s12})FN_1 - (\sigma_{p23} + \sigma_{s23})FN_2$$
$$\dot{N}_3 = \sigma_{3e}^{(3)}F^3N_0 + (\sigma_{s23} + \sigma_{p23})FN_2 - (\sigma_{p34}^{(2)} + \sigma_{s34}^{(2)})F^2N_3$$
$$\dot{N}_4 = \sigma_{4e}^{(4)}F^4N_0 + (\sigma_{p34}^{(2)} + \sigma_{s34}^{(2)})F^2N_3 - (\sigma_{p45}^{(2)} + \sigma_{s45}^{(2)})F^2N_4$$
$$\dot{N}_5 = \sigma_{5e}^{(5)}F^5N_0 + (\sigma_{s45}^{(2)} + \sigma_{p45}^{(2)})F^2N_4 - (\sigma_{s56}^{(2)} + \sigma_{p56}^{(2)})F^2N_5$$
$$\dot{N}_6 = \sigma_{6e}^{(6)}F^6N_0 + (\sigma_{s56}^{(2)} + \sigma_{p56}^{(2)})F^2N_5 - \sigma_{s67}^{(3)}F^3N_6$$
$$\dot{N}_7 = \sigma_{s67}^{(3)}F^3N_6 - \sigma_{s78}^{(3)}F^3N_7$$
$$\dot{N}_8 = \sigma_{s78}^{(3)}F^3N_7$$

Direct Multiphoton Multiple Ionization Cross Sections

A direct N-photon, n-electron ejection cross Section is of the same order as an N-photon one-electron cross section as long as $N \geq n$.
(See, for example PRA 83, 021407 (2011))

Why can we accept that conjecture ?
Known cases and structure of the equation

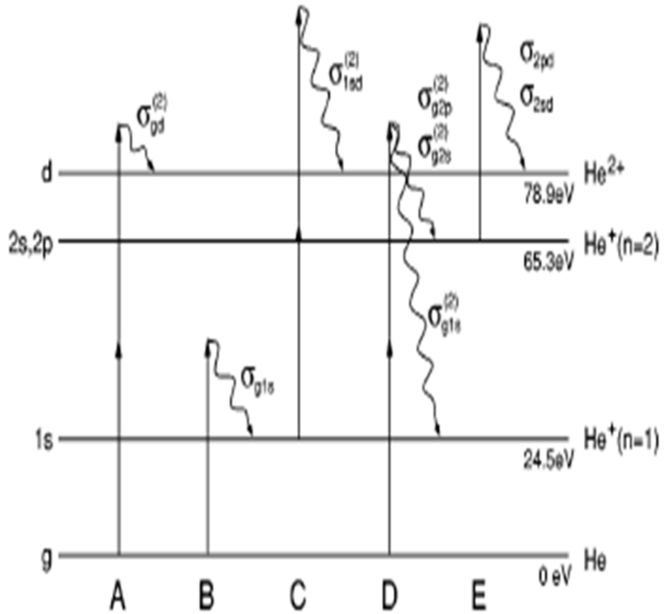


FIG. 1. Level scheme considered in this paper.

For the present study, we specifically assume the photon energy of 45 eV. Using a multichannel *B*-spline code developed in recent years [19,20], we obtain the cross sections as $\sigma_{gd}^{(2)}=8.1\times10^{-52}$ cm^4 s, $\sigma_{g1s}^{(2)}=1.0\times10^{-52}$ cm^4 s, $\sigma_{g2s}^{(2)}=2.3\times10^{-51}$ cm^4 s, $\sigma_{g2p}^{(2)}=3.8\times10^{-51}$ cm^4 s, $\sigma_{g1s}=2.4\times10^{-18}$ cm^2, $\sigma_{1sd}^{(2)}=1.0\times10^{-53}$ cm^4 s, $\sigma_{2sd}=2.4\times10^{-19}$ cm^2, and $\sigma_{2pd}=9.0\times10^{-20}$ cm^2. In our scheme the

Also Tang & PL, PRL 58, 108 (1987). 4-photon, 2e ionization in Carbon.

III. JUSTIFICATION OF THE CONJECTURE ON THE MAGNITUDE OF CROSS SECTIONS FOR MULTIPHOTON MULTIPLE IONIZATION.

The general expression for an N-photon transition amplitude from an initial state $|g\rangle$ to a final state $|f\rangle$, within LOPT, is proportional to:

$$\sum_{a_{N-1}} \cdots \sum_{a_1} \frac{\langle f|\hat{\mathrm{D}}|a_{N-1}\rangle \ldots \langle a_1|\hat{\mathrm{D}}|g\rangle}{[E_{a_{N-1}} - E_g - (N-1)\hbar\omega] \ldots (E_{a_1} - E_g - \hbar\omega)} \tag{2}$$

where $\hat{\mathrm{D}}$ is the electric dipole operator. For an M-electron atom, in principle, all states entering this expression are M-electron states. In reality, certain approximations are involved in calculating a multiphoton transition. Thus for N-photon single electron ejection, which may include above threshold ionization (ATI), as it certainly does for Neon under 93 eV photons, the main contribution comes through the single active electron (SAE) approximation. The terms that would contribute in the matrix elements are schematically

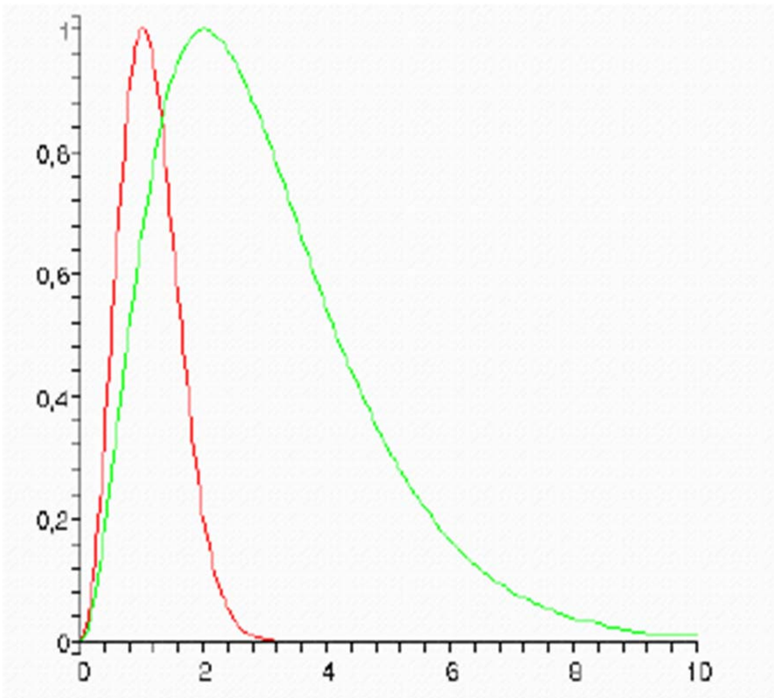
$$2p \to \{ns, nd\} \to \{np, nf\} \to \{ns, nd, ng\} \to \{np, nf, nh\} \to \ldots, \tag{3}$$

where n here denotes the principal quantum number, with the maximum angular momentum for, say, a 6-photon process being 7. In this case, $|f\rangle$ involves one electron in the continuum. Consider now, as an example, the case of a direct 6-photon, 6-electron transition from an initial state $2s^2 2p^6$, the corresponding transition amplitude would have the form

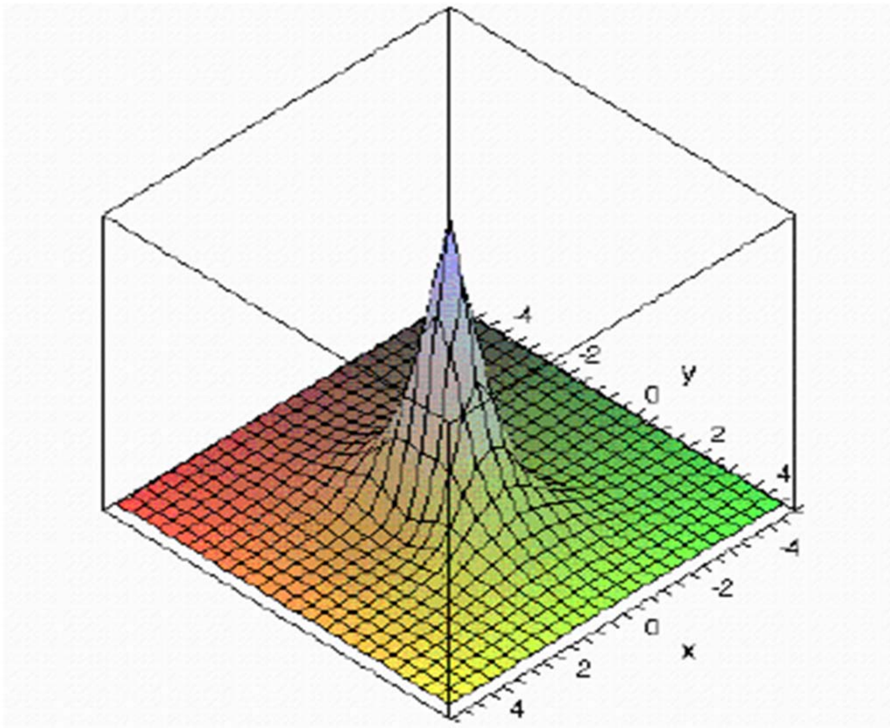
$$\sum_{\mathbf{k}_1} \cdots \sum_{\mathbf{k}_5} \frac{\langle \mathbf{k}_6 \ldots \mathbf{k}_1; 2s^2|\hat{\mathrm{D}}|\mathbf{k}_5 \ldots \mathbf{k}_1; 2p2s^2\rangle \langle \mathbf{k}_5, \ldots \mathbf{k}_1; 2p2s^2|\hat{\mathrm{D}}|\mathbf{k}_4 \ldots \mathbf{k}_1; 2p^2 2s^2\rangle \ldots \langle \mathbf{k}_1; 2p^5 2s^2|\hat{\mathrm{D}}|2p^6 2s^2\rangle}{\Delta_5 \Delta_4 \ldots \Delta_1} \tag{4}$$

Temporal and spatial distribution of intensity in focused laser pulses, the details of which depend on the particular experimental arrangement.

Temporal



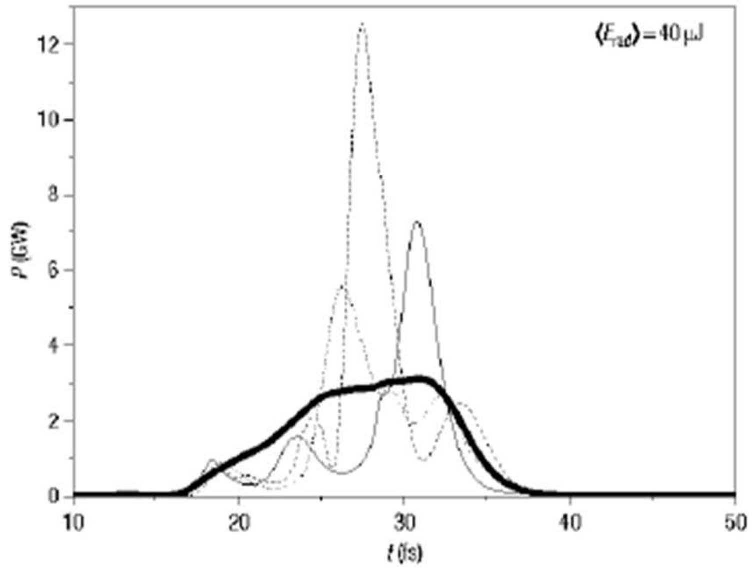
Spatial

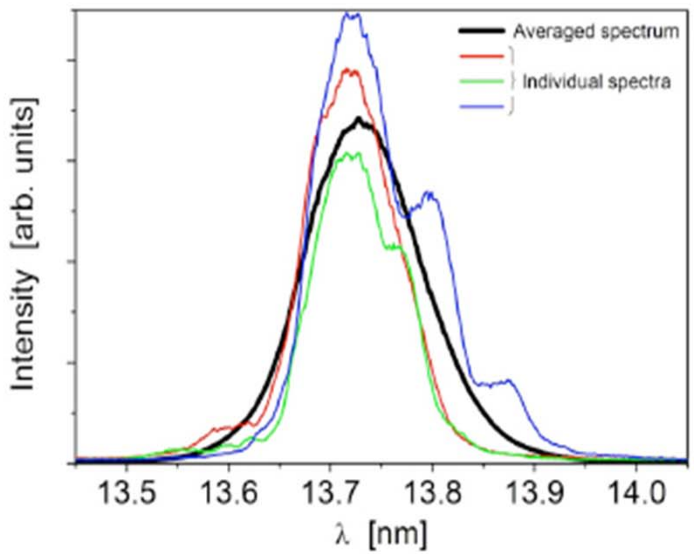


Features of FEL Pulses I

FEL pulses in the time and frequency domain

Spiky behavior, with the width of the main peaks determined by the coherence time T_c.





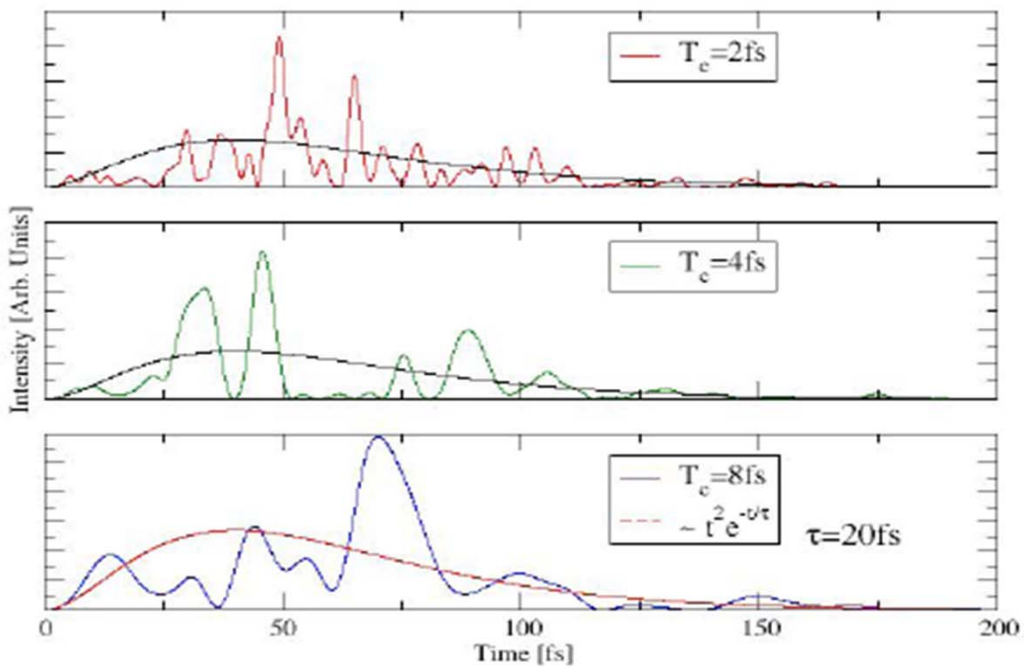
Ackermann *et al.*, Nature **1**, 336 (2007).

Photon statistics and non-linear processes.

- N-photon ionization in LOPT is proportional to the Nth order intensity correlation function.
- For a chaotic field the Nth order intensity correlation function is N! times the Nth power of the average intensity (Flux).
- Equivalent to increasing the N-photon cross section by a factor of N!

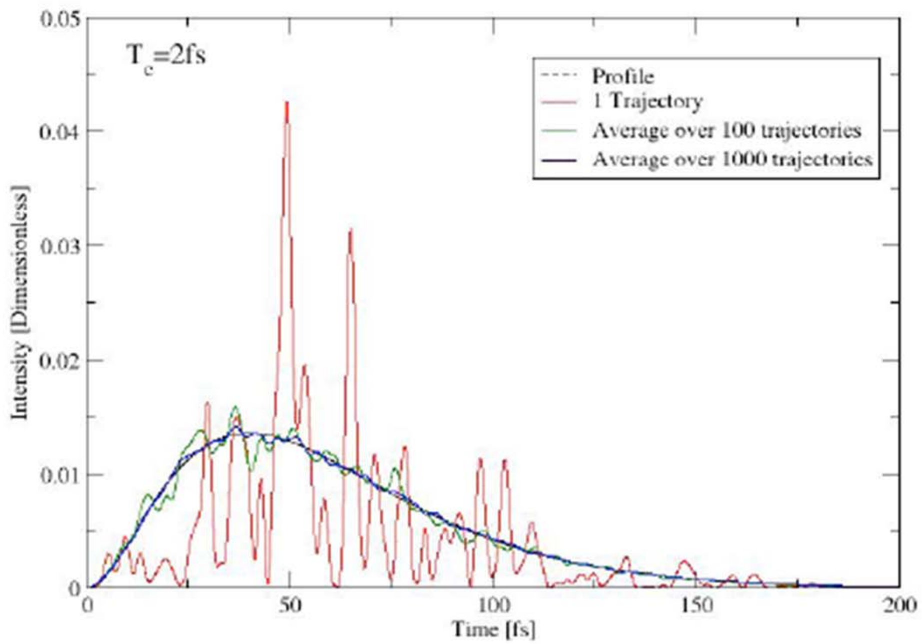
Features of FEL Pulses II

Gaussian spectral linewidth: Typical random pulses

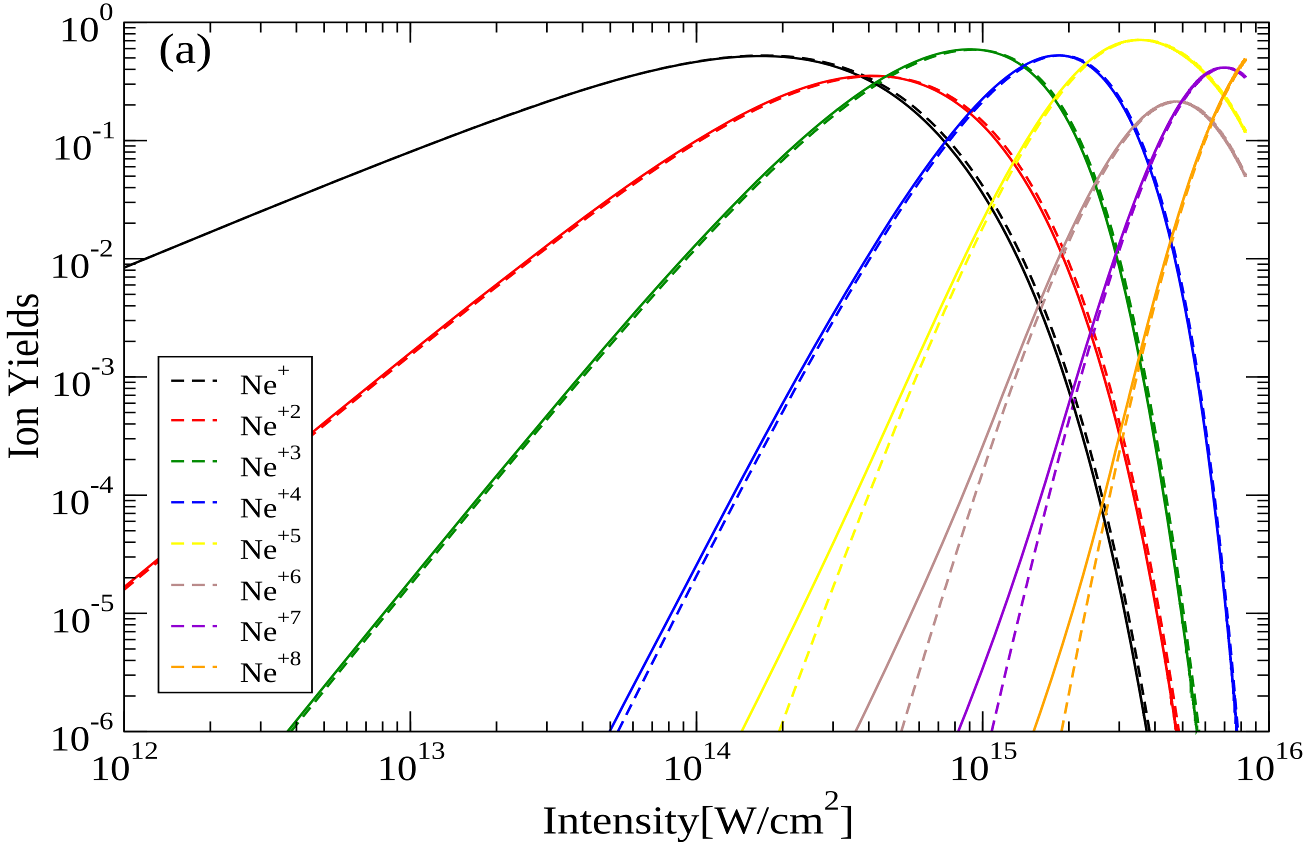


Averaging over random pulses

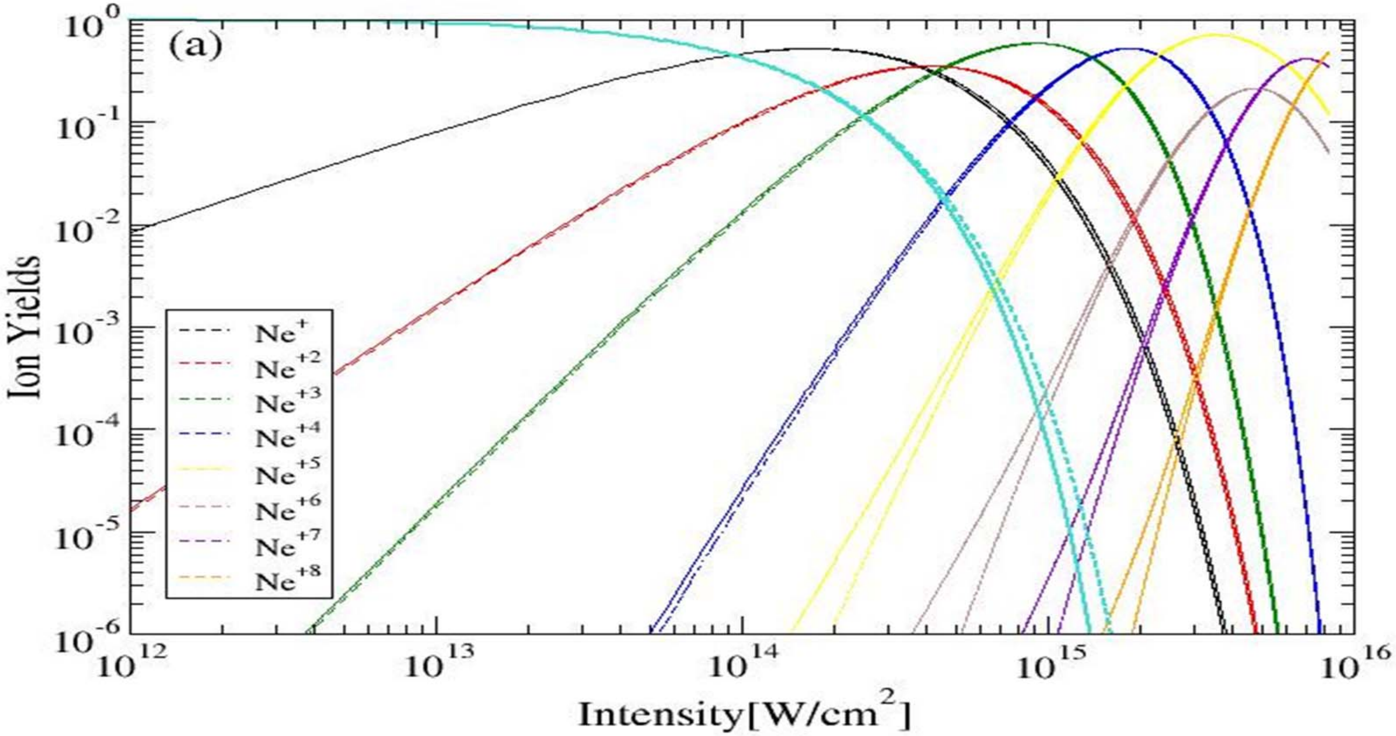
Gaussian spectral linewidth: Averaging over random pulses



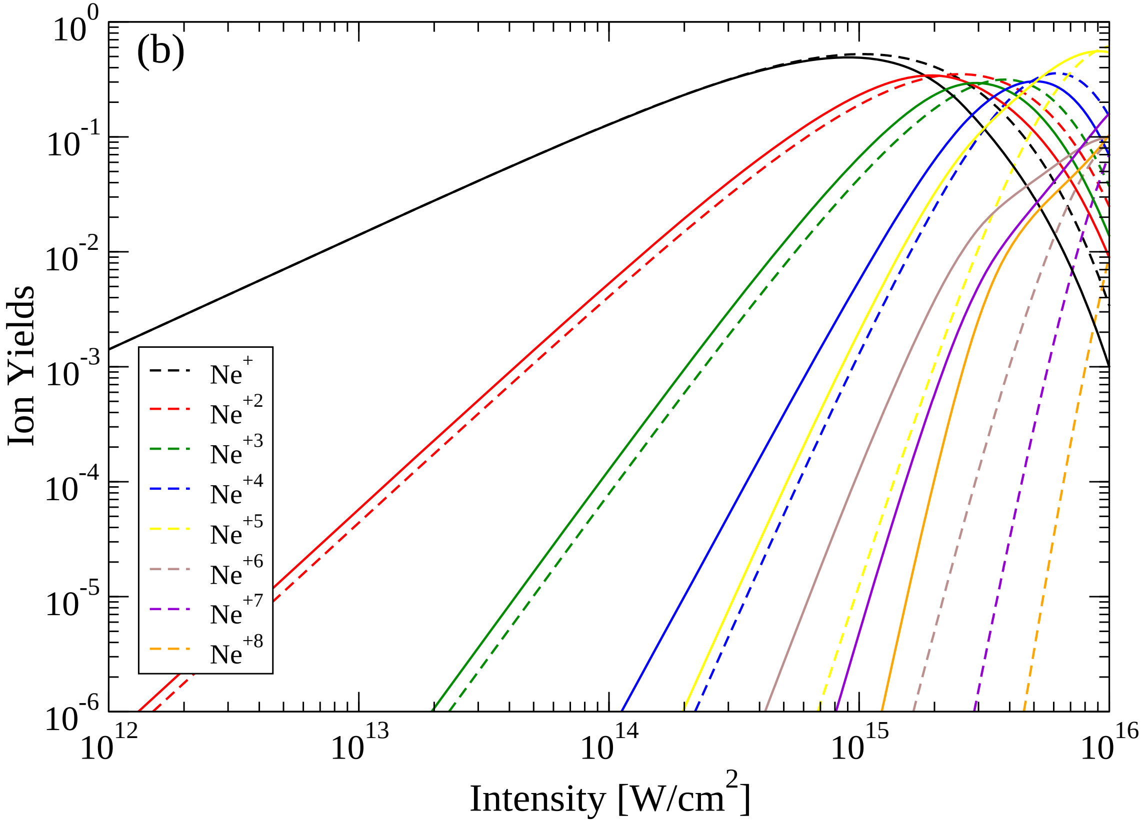
Deterministic pulse 30 fs (solid lines with and dashed lines without the direct channels, PRA 83, 021407 (2011))



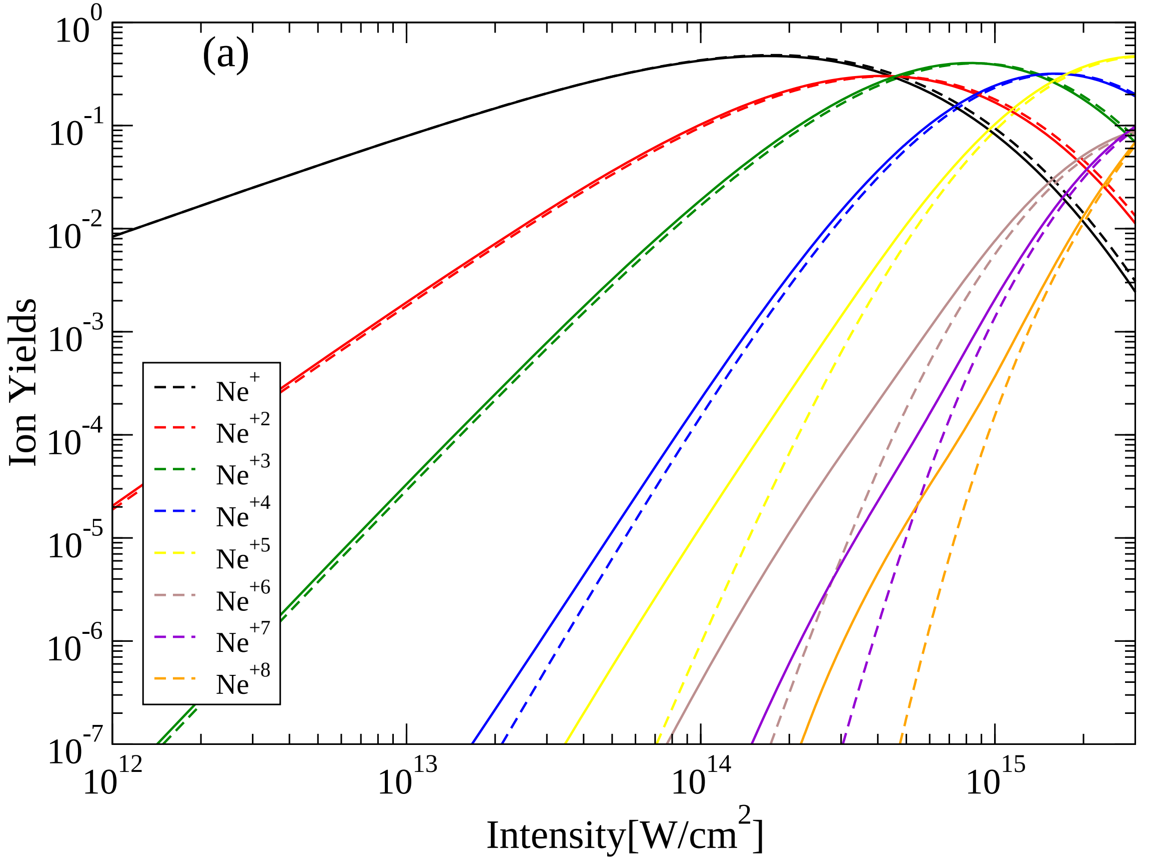
Evolution of the neutral under a 30 fs pulse.



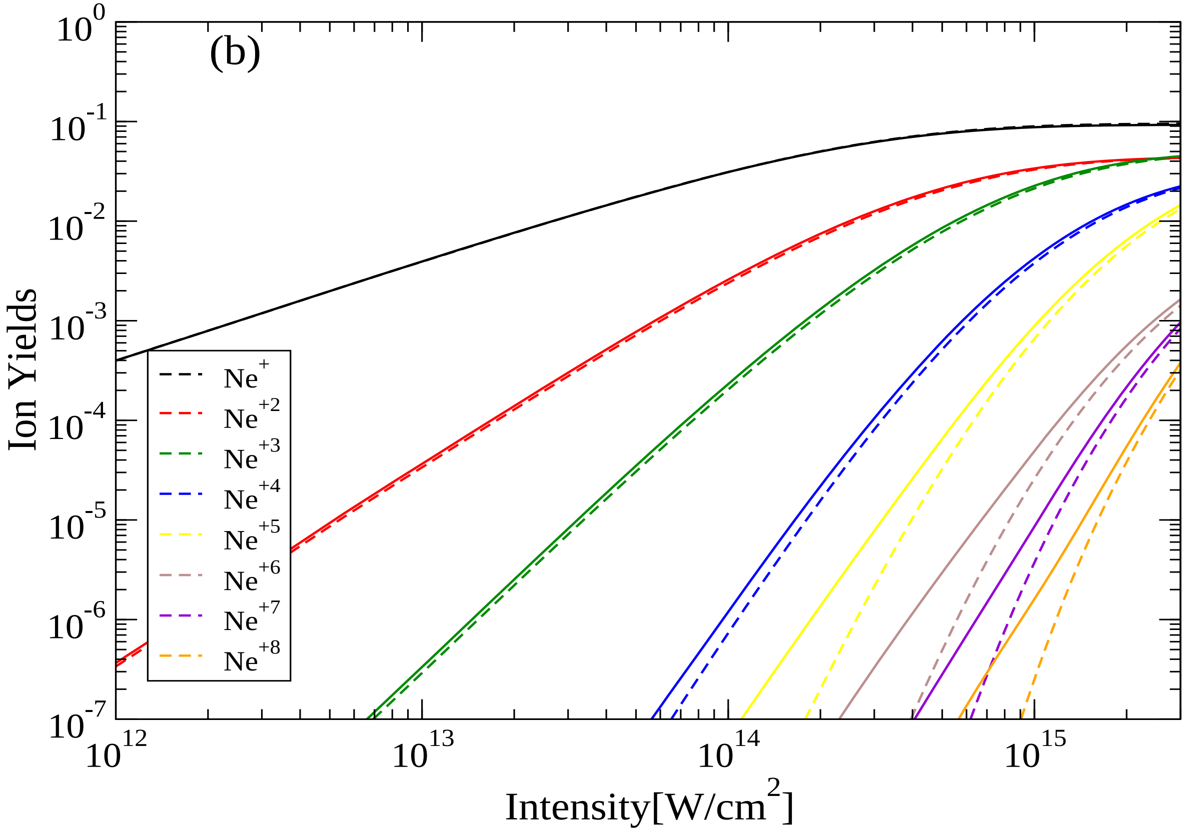
Deterministic pulse 5fs



Chaotic pulse 30fs.



Space integrated (30 fs, chaotic)



Exp. Data, Richter et al. PRL 102 163002 (09)

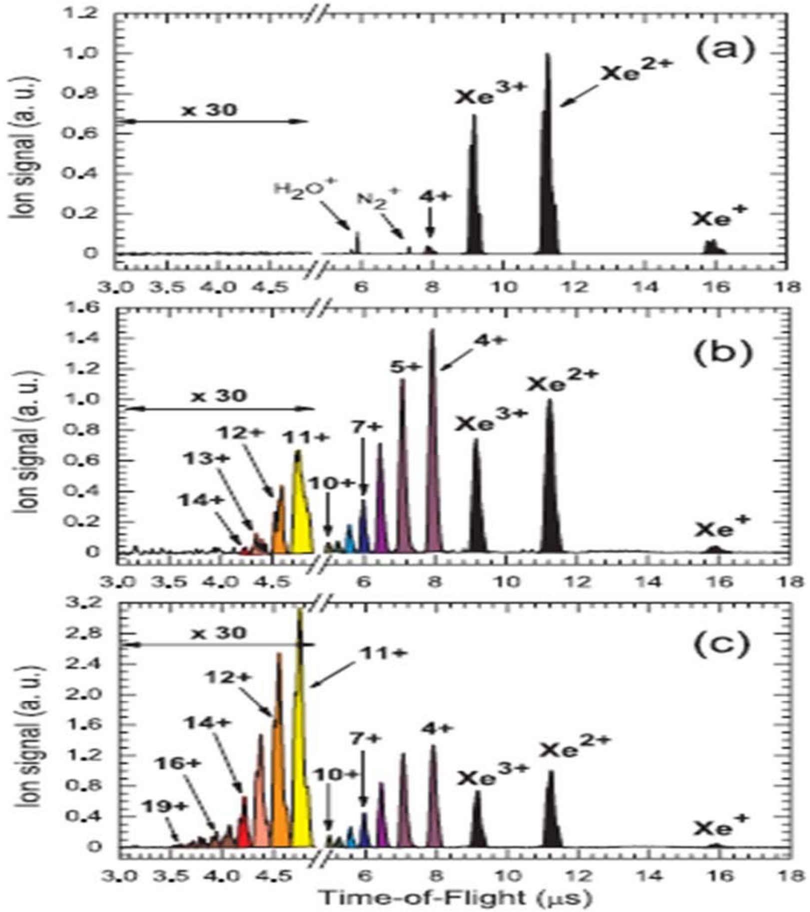


FIG. 1 (color online). Ion time-of-flight (TOF) mass-to-charge spectra of xenon (Xe) taken at 90.5 eV photon energy and irradiance levels of (a) 2.5×10^{12} W cm^{-2}, (b) 1.7×10^{15} W cm^{-2}, and (c) 2.0×10^{15} W cm^{-2}. Signals from residual gas are also indicated.

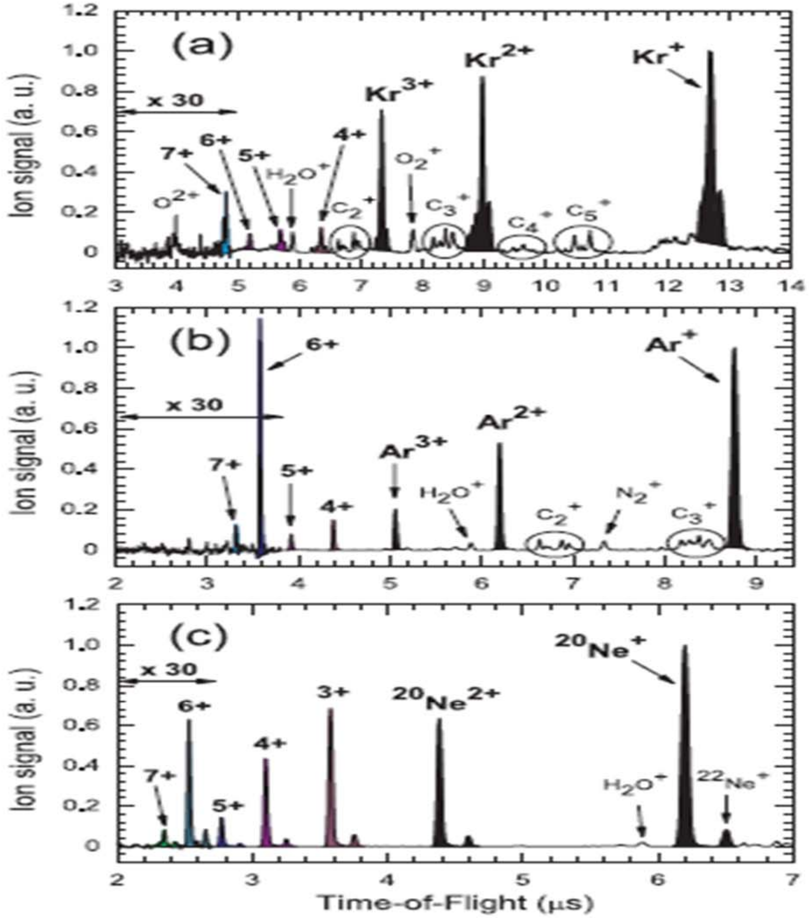
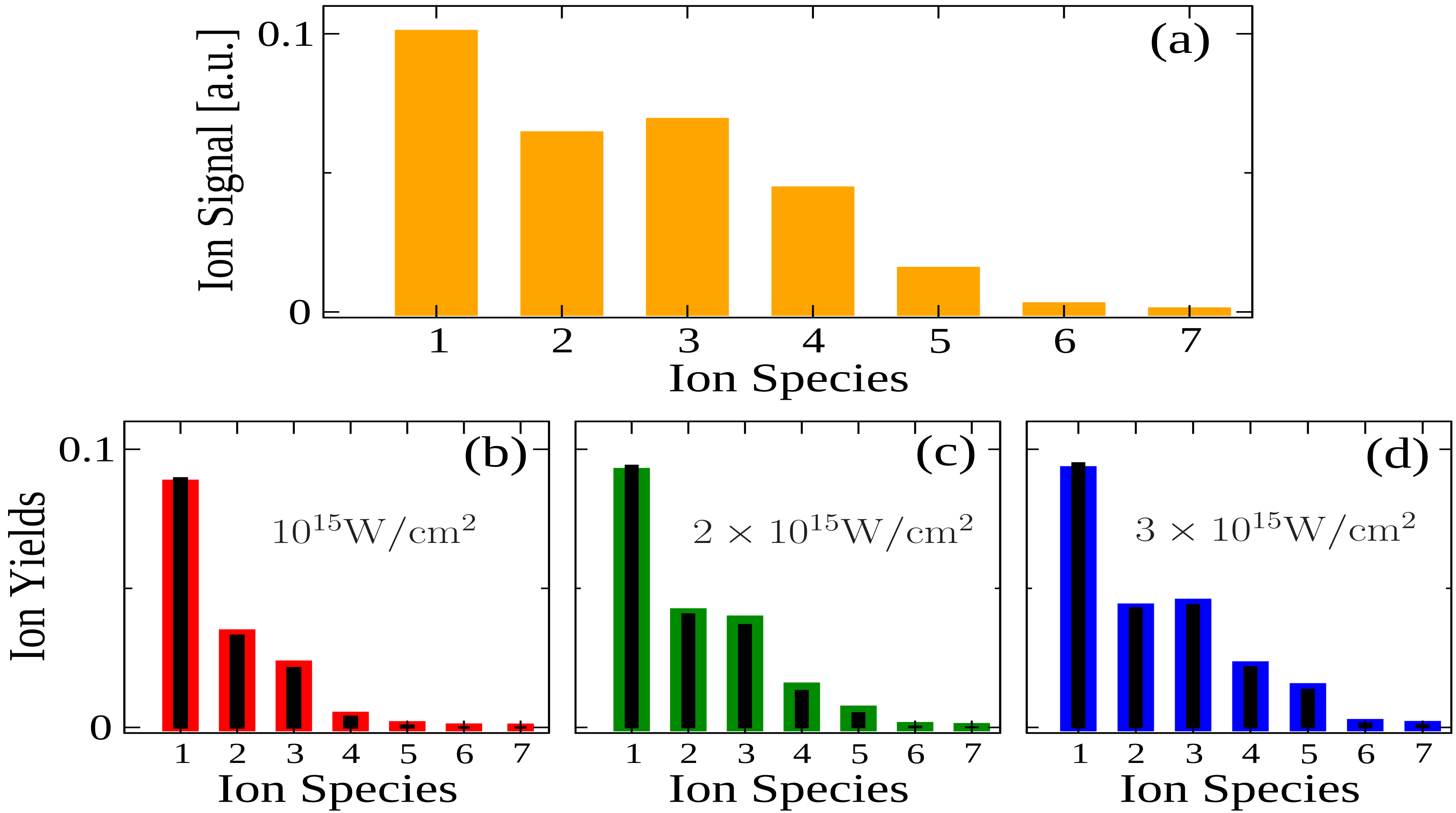


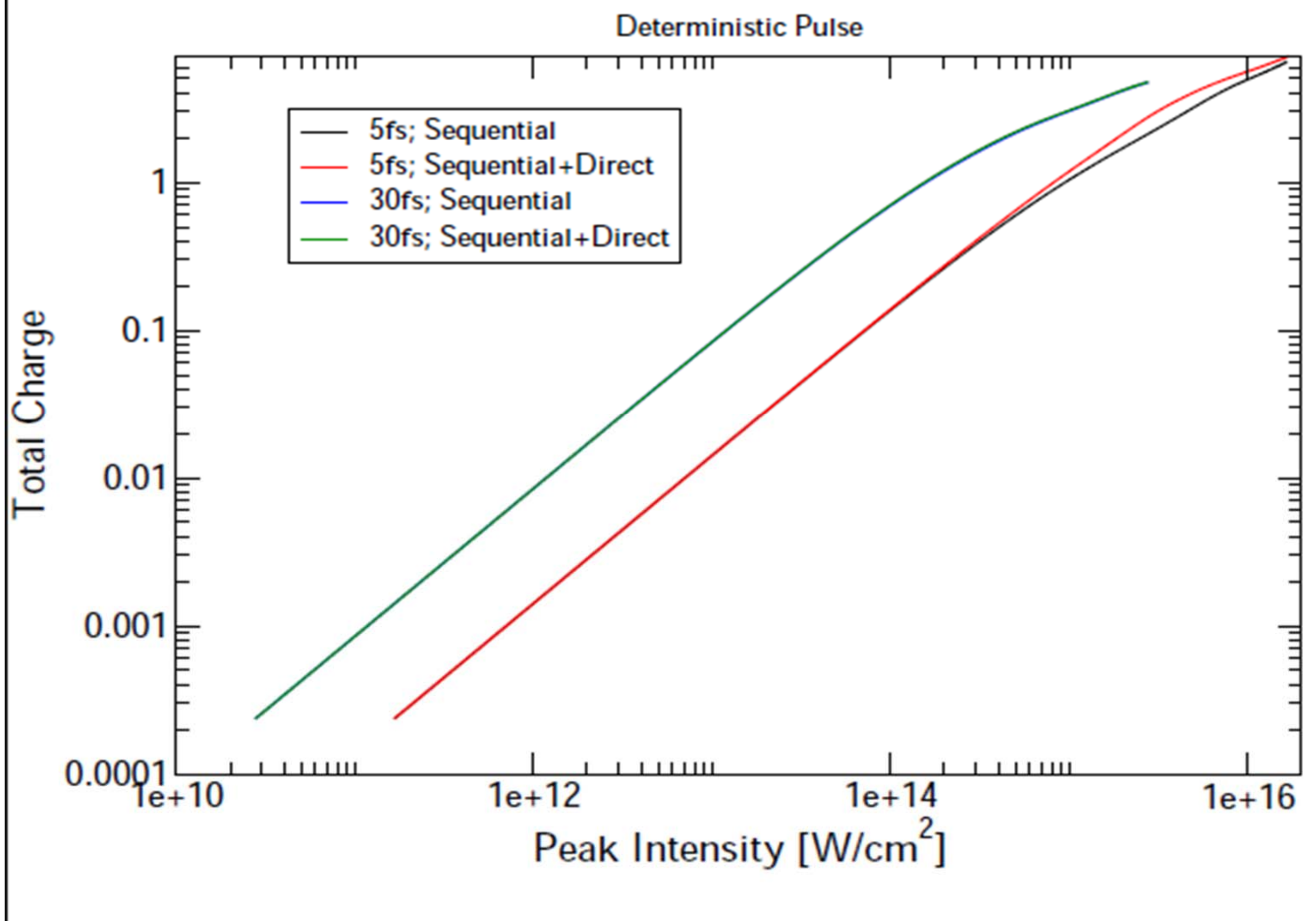
FIG. 2 (color online). Ion time-of-flight (TOF) mass-to-charge spectra of (a) krypton (Kr), (b) argon (Ar), and (c) neon (Ne), taken at 90.5 eV photon energy and irradiance levels between 1.5 and 1.8×10^{15} W cm^{-2}. The $C_n{}^+$ signals ($n = 2$ to 5) possibly arise from carbon clusters desorbed from the carbon coated BL2 focusing mirror.

Comparison to experiment

Richter PRL 102, 163002 (09); theory PRA 83, 021407 (2011).



Ne at 93 eV: Total charge versus laser intensity



Xe at 93 eV: Evolution of ionic populations as a function of peak laser intensity (10 fs pulse)

J. Phys. B: At. Mol. Opt. Phys. **44** (2011) 175402 P Lambropoulos *et al*

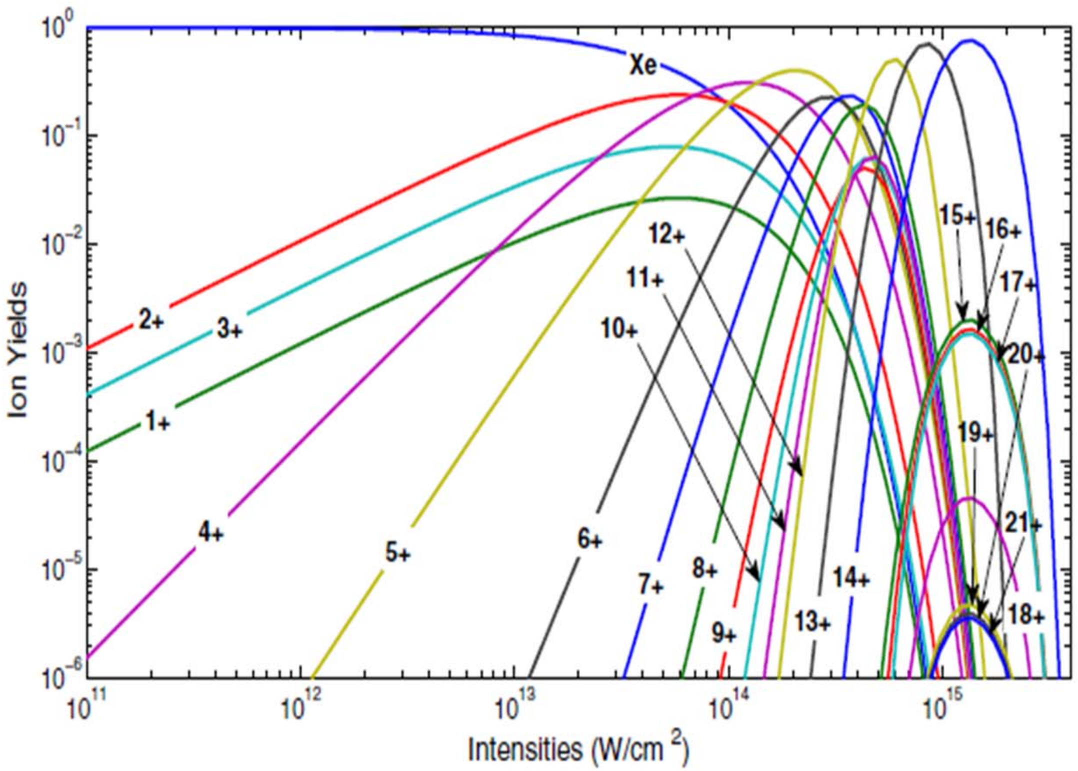


Figure 2. Calculated population of the neutral and generated ionic yields of Xe, as a function of FEL peak intensity, under 93 eV radiation of 10 fs pulse duration.

Xe at 93 eV: Population of the neutral as a function of laser peak intensity (10 fs pulse)

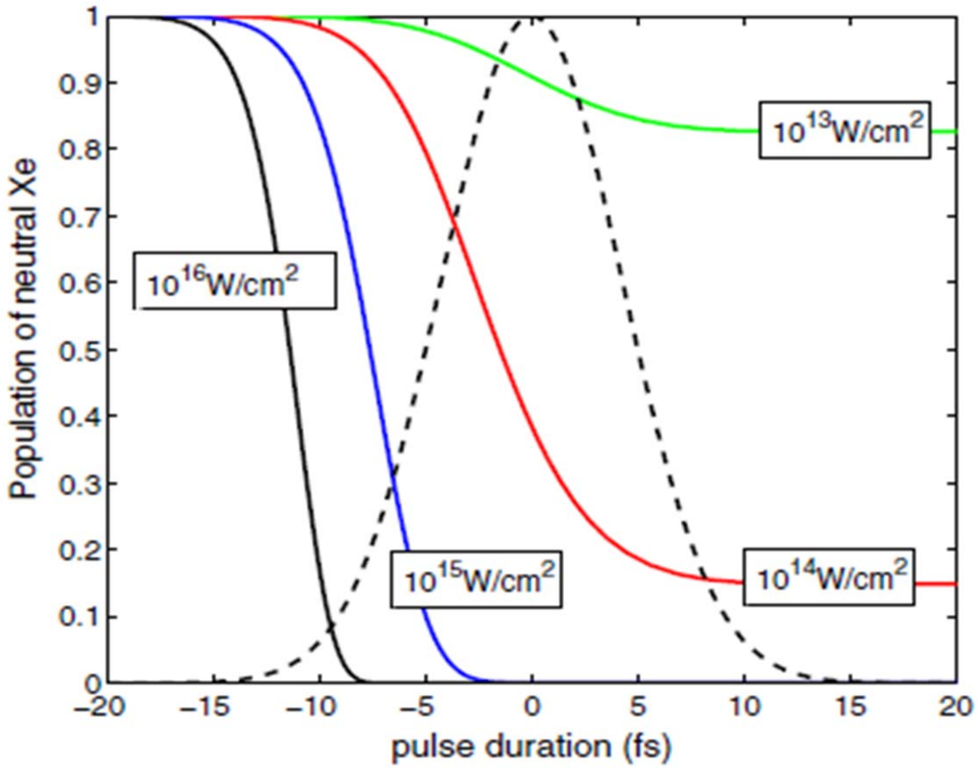


Figure 3. Evolution of the population of the neutral during the pulse for various peak intensities. The dashed line represents a pulse of 10 fs duration (FWHM).

$$\sigma_7 \cong 10^{-205}\ \mathrm{cm}^{14}\ \mathrm{sec}^{6}$$

Photon flux F (photons/cm^2sec)

7-photon rate → $\sigma_7 F^7$

An uncertainty of a factor of 3 in F implies an uncertainty of a factor of

2.18×10^3 in the rate.

So, an uncertainty of a factor of 100 in σ_7 is within experimental uncertainties.

Summary and Outlook

These multielectron direct processes should be present in essentially any atom and certainly in all rare gases.

END- Thank you