X-Ray Photoionization of Free and Confined Atoms

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AT HIGH PHOTON ENERGY PHOTOIONIZATION IS NOT GENERALLY A ONE-ELECTRON PROCESS

This is due to correlation—the fact that initial and final states in the photoionization process are often not well-represented by single-particle wave functions

This correlation takes many different forms resulting in various consequences

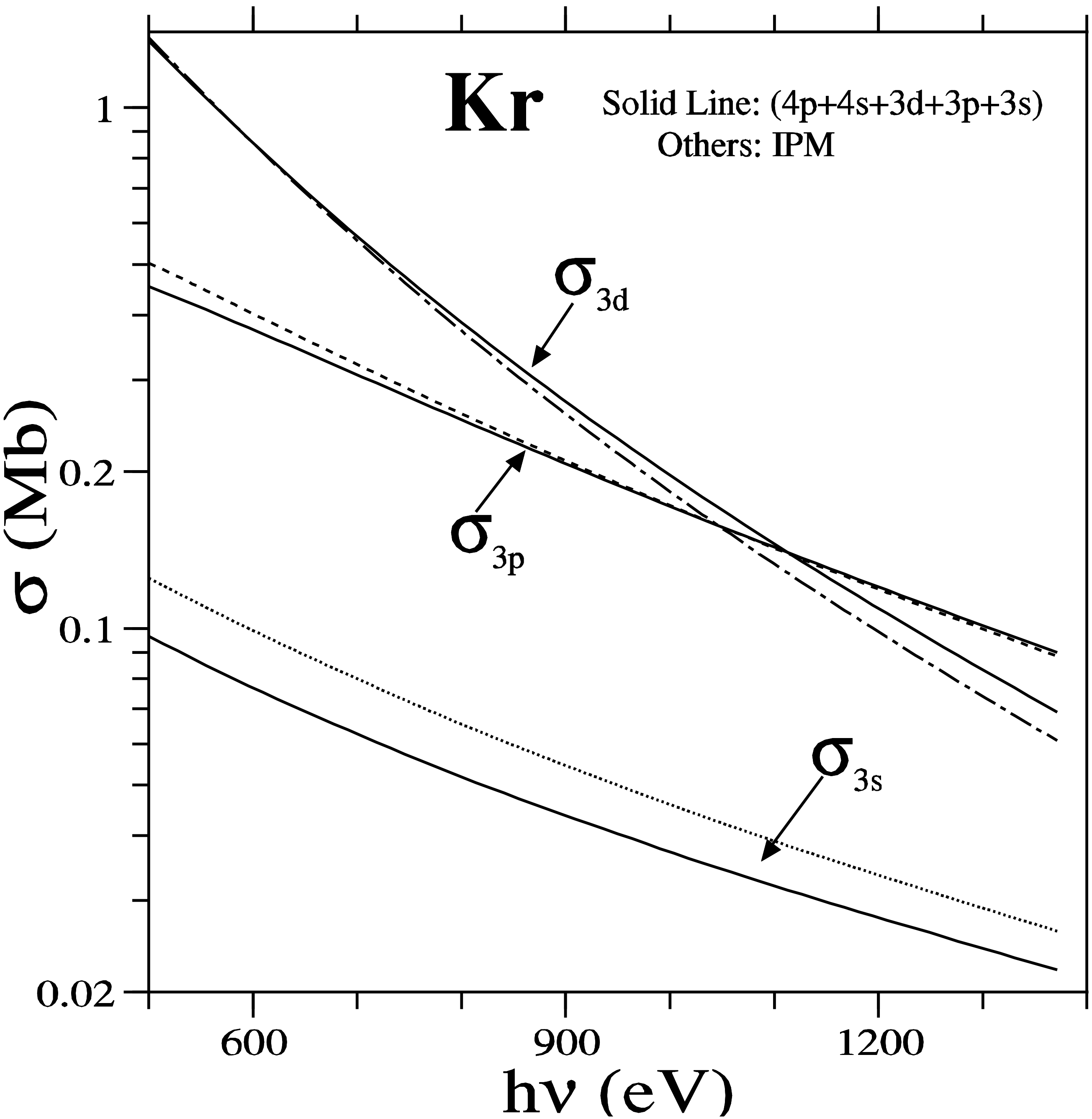
Correlation in the final (continuum) state wave function is generally known as interchannel coupling—it is really just configuration interaction in the continuum.

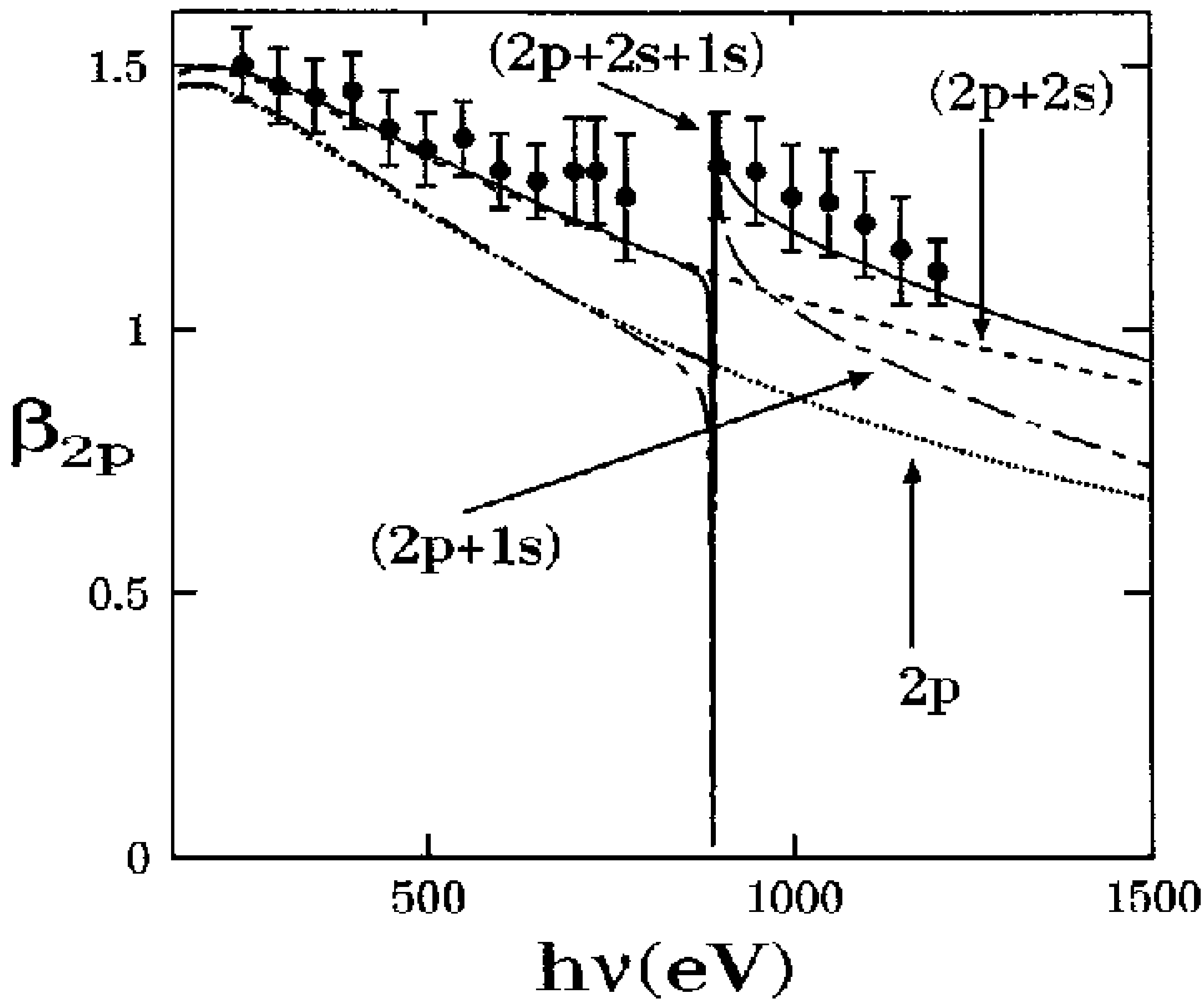
As a consequence, when there are degenerate photoionization processes, i.e., from different atomic subshells, weak cross sections can be altered significantly by strong ones.

HIGH-ENERGY PHOTOIONIZATION CROSS SECTIONS

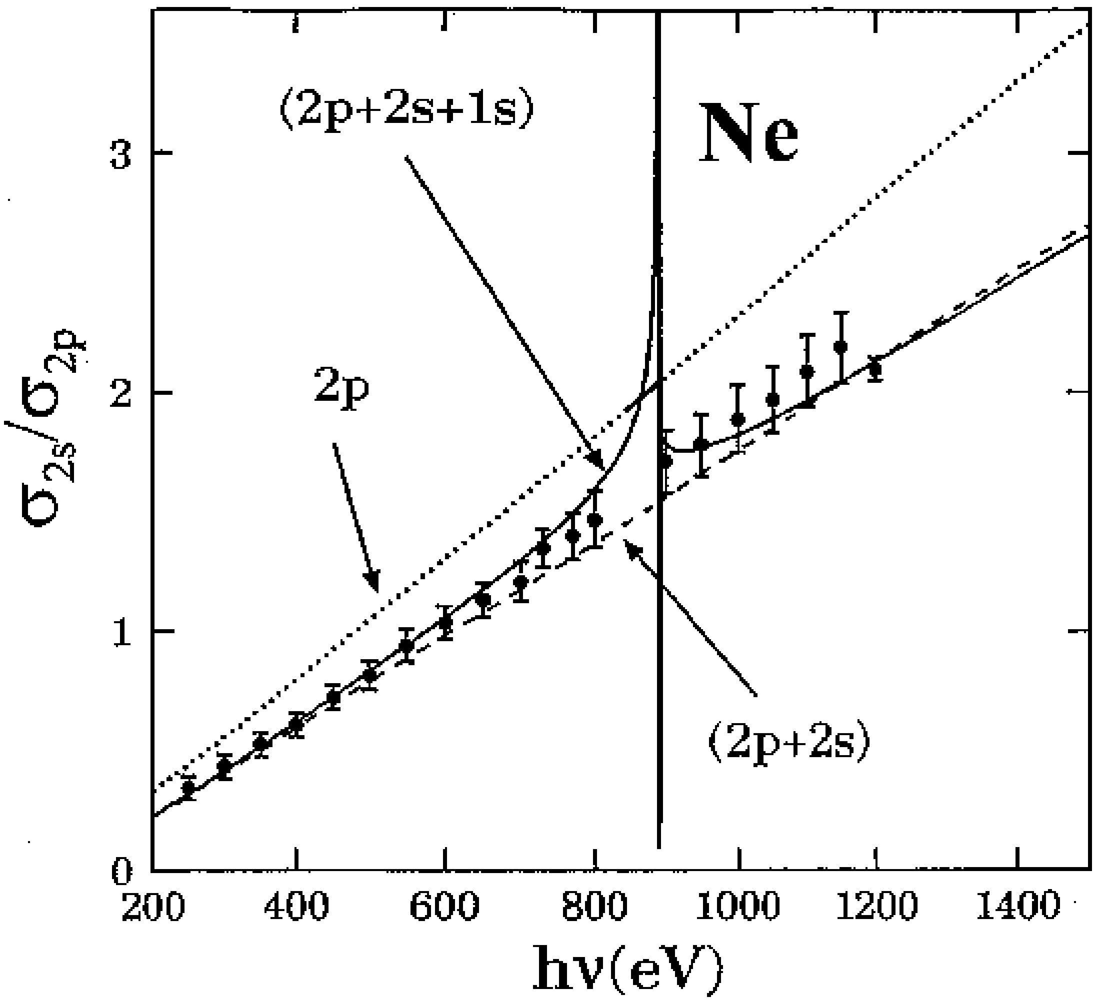
In the single-particle approximation, the asymptotic cross section for photo-ionization of an *nl* state goes as $E^{-(l+7/2)}$.

Owing to interchannel coupling, it is found that this dependence remains correct for ns states, but the dependence becomes $E^{-(9/2)}$ for all other states.


Kr
Solid Line: (4p+4s+3d+3p+3s)
Others: IPM
σ3d
σ3p
σ3s
σ (Mb)
1
0.2
0.1
0.02
600
900
1200
hν (eV)



1.5
1
0.5
0
500
1000
1500
β2p
hν(eV)
(2p+2s+1s)
(2p+2s)
(2p+1s)
2p



Ne
(2p+2s+1s)
2p
(2p+2s)
σ_{2s}/σ_{2p}
0
1
2
3
200
400
600
800
1000
1200
1400
hν(eV)

Initial state correlation can also have important consequences.

Example: Photoionization of an atom or ion with outer-shell structure $(4p)^2\ {}^1S$

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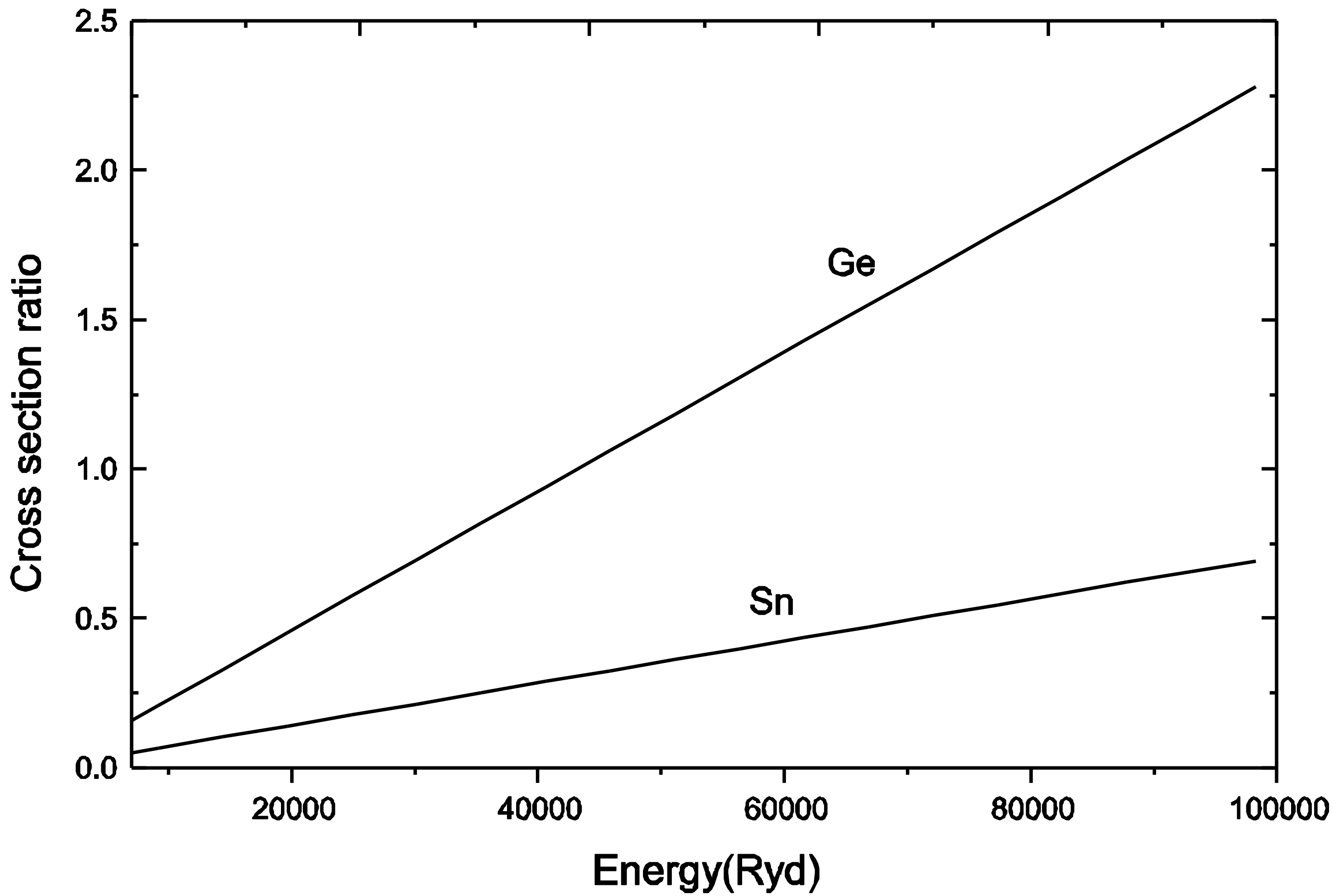
Initial state wave function: $\alpha\ (4p)^2 + \beta\ (5s)^2$, where β is quite small.

Final state wave functions: $4p\varepsilon s$, $4p\varepsilon d$, $5s\varepsilon p$

Cross section for leaving target in the 4p state, $4p \rightarrow \varepsilon s$, $4p \rightarrow \varepsilon d$, behaves at high energy as $E^{-(9/2)}$

Cross section for leaving target in the 4s state, $4s \rightarrow \varepsilon p$, behaves at high energy as $E^{-(7/2)}$

Thus, at high enough energy, ionization plus excitation will dominate!

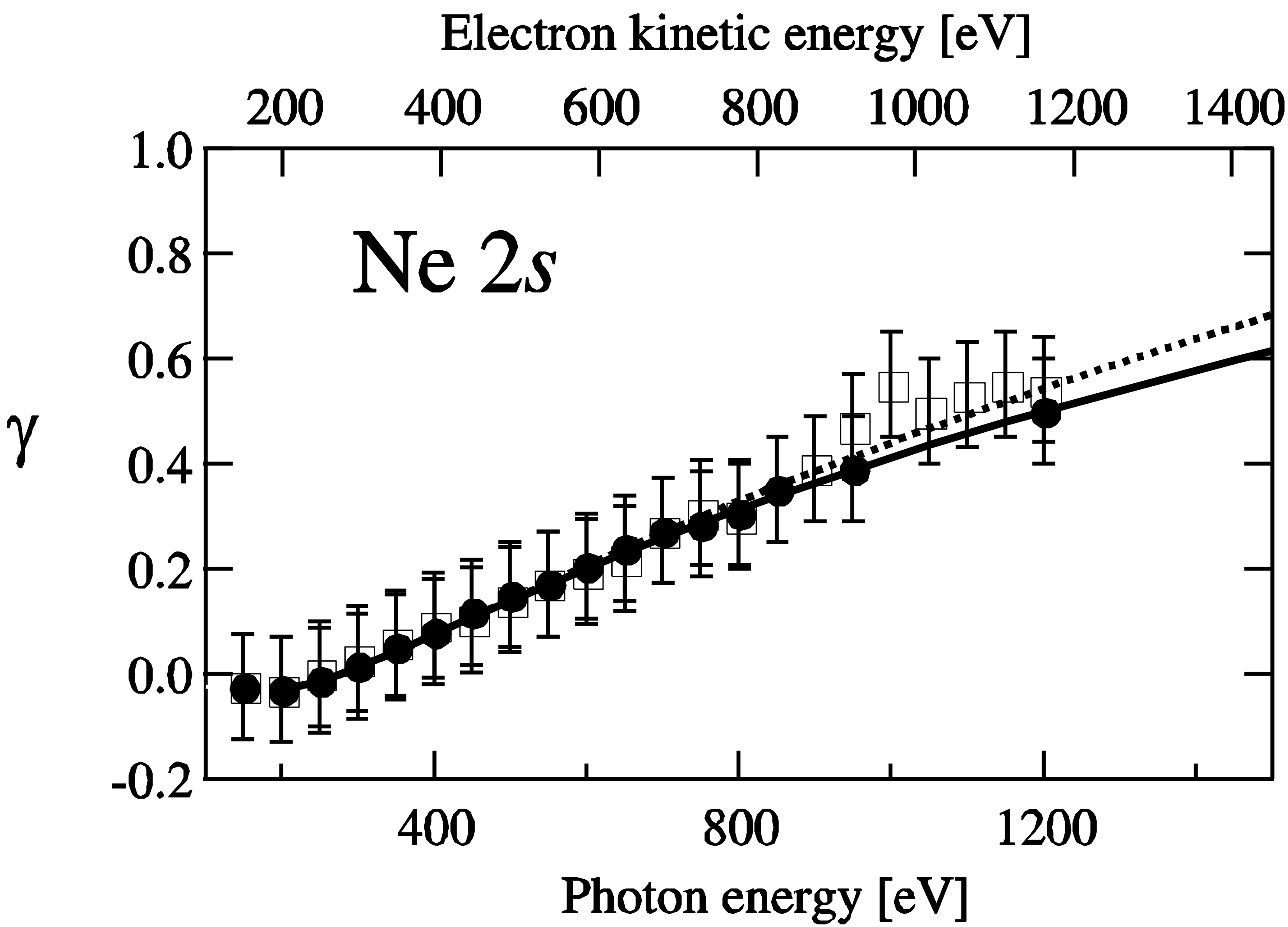

Cross section ratio
Energy(Ryd)
Ge
Sn
0.0
0.5
1.0
1.5
2.0
2.5
20000
40000
60000
80000
100000

Photoelectron Angular Distribution

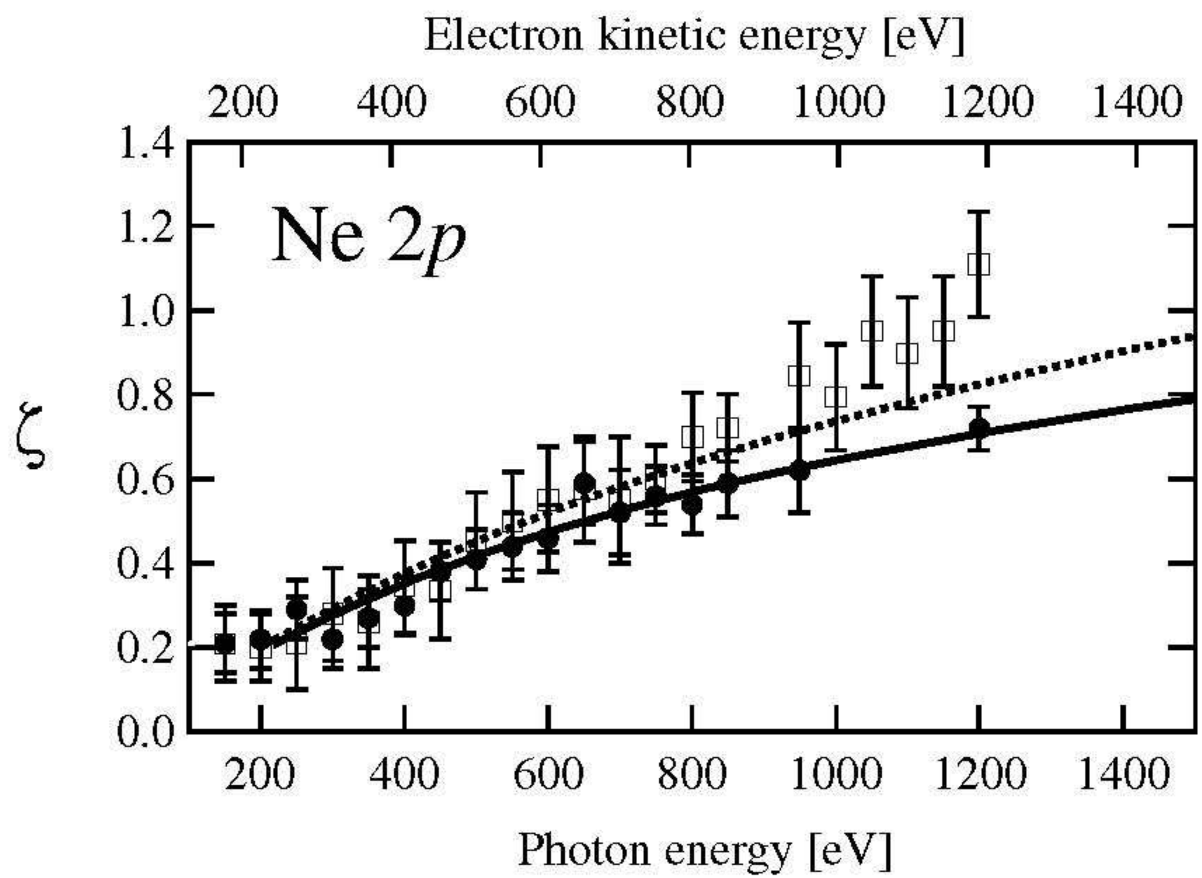
$$d\sigma/d\Omega = (\sigma/4\pi)[1 + (\beta+\Delta\beta)P_2(\cos\theta) + (\delta+\gamma\cos^2\theta)\sin\theta\cos\varphi + \lambda P_2(\cos\theta)\cos 2\varphi + \mu\cos 2\varphi + \nu(1+\cos 2\varphi)P_4(\cos\theta)$$

Red—First order in photon momentum; dipole-quadrupole interference

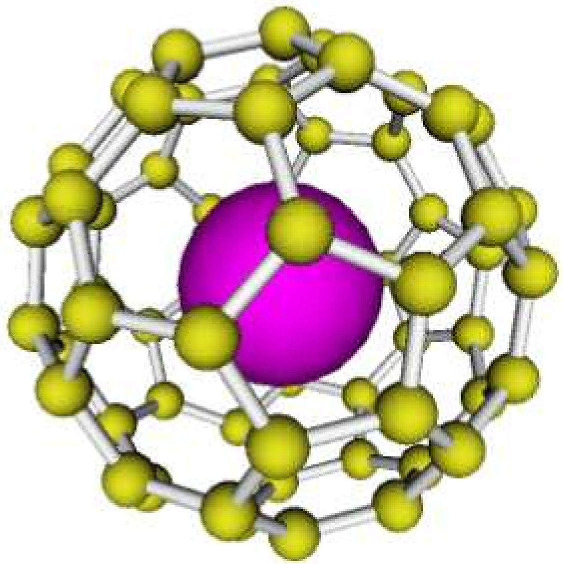
Violet—Second order in photon momentum-quadrupole quadrupole and dipole-octupole interference



Electron kinetic energy [eV]
200
400
600
800
1000
1200
1400
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
Ne 2s
γ
400
800
1200
Photon energy [eV]


Electron kinetic energy [eV]
200
400
600
800
1000
1200
1400
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Ne 2*p*
ζ
200
400
600
800
1000
1200
1400
Photon energy [eV]

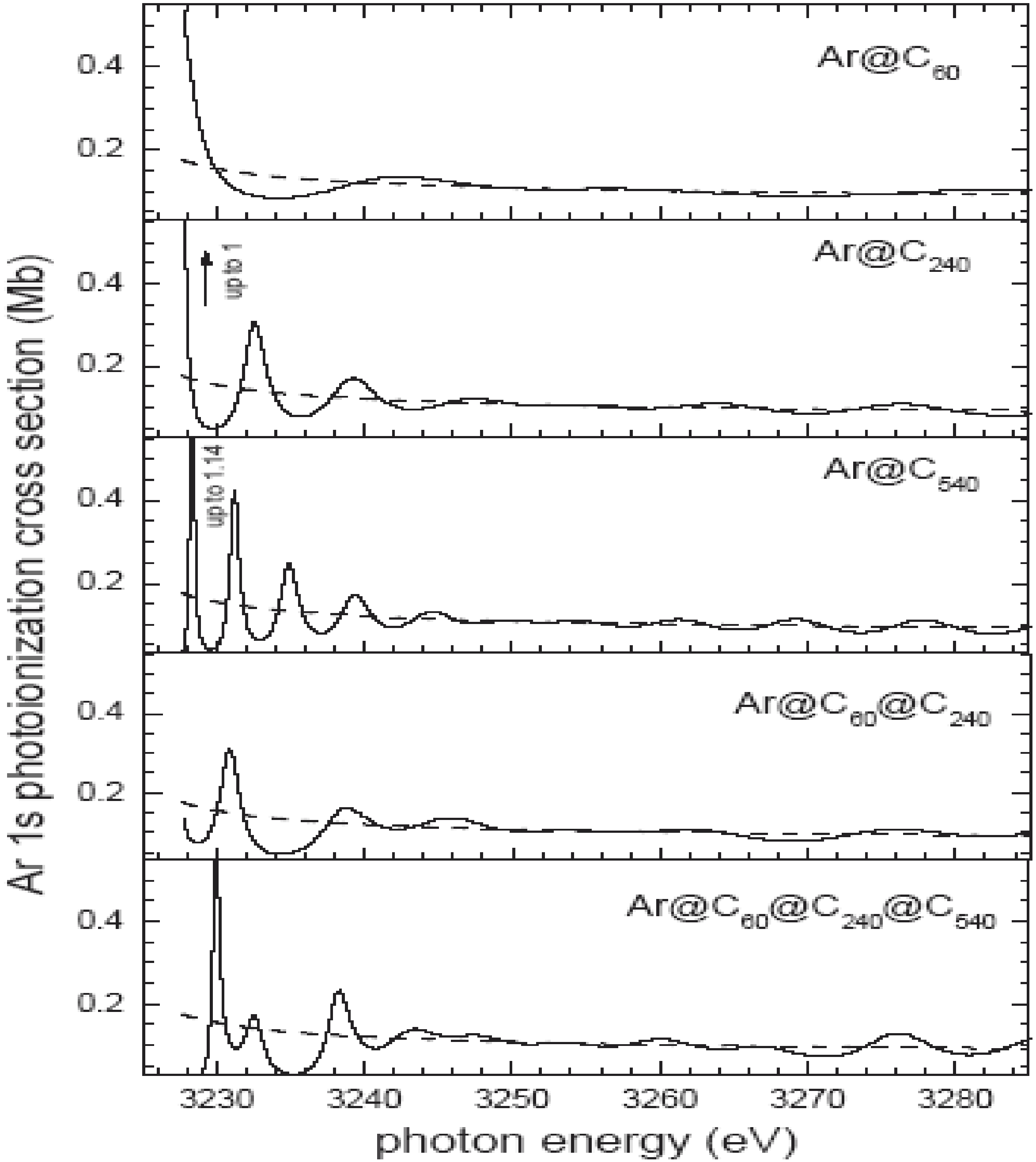
Ionization of Endohedral Atoms



Calculations based on a variety of approximations to describe the fullerene shell and its influence upon the encaged atom

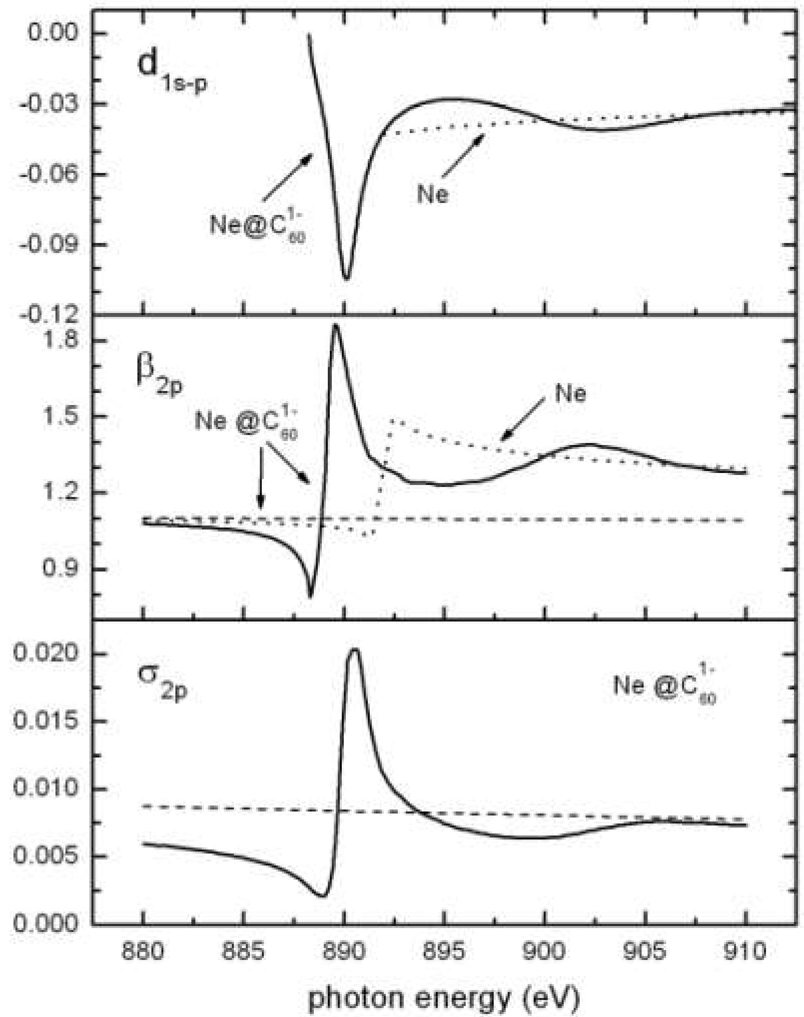
Ionization of Endohedral Atoms: Confinement Resonances

- Confinement resonances, oscillations in the photoionization cross section, were first predicted in the early 1990’s
- Their physical origin was explained about a decade ago as the interference of the photoelectron wave emitted directly with the waves that scatter from the confining potential.



Ar@C60
Ar@C240
up to 1
Ar@C540
up to 1.14
Ar@C60@C240
Ar@C60@C240@C540
0.4
0.2
Ar 1s photoionization cross section (Mb)
3230
3240
3250
3260
3270
3280
photon energy (eV)

Photoionization of $Ne@C_{60}$ in the vicinity of the 1s threshold



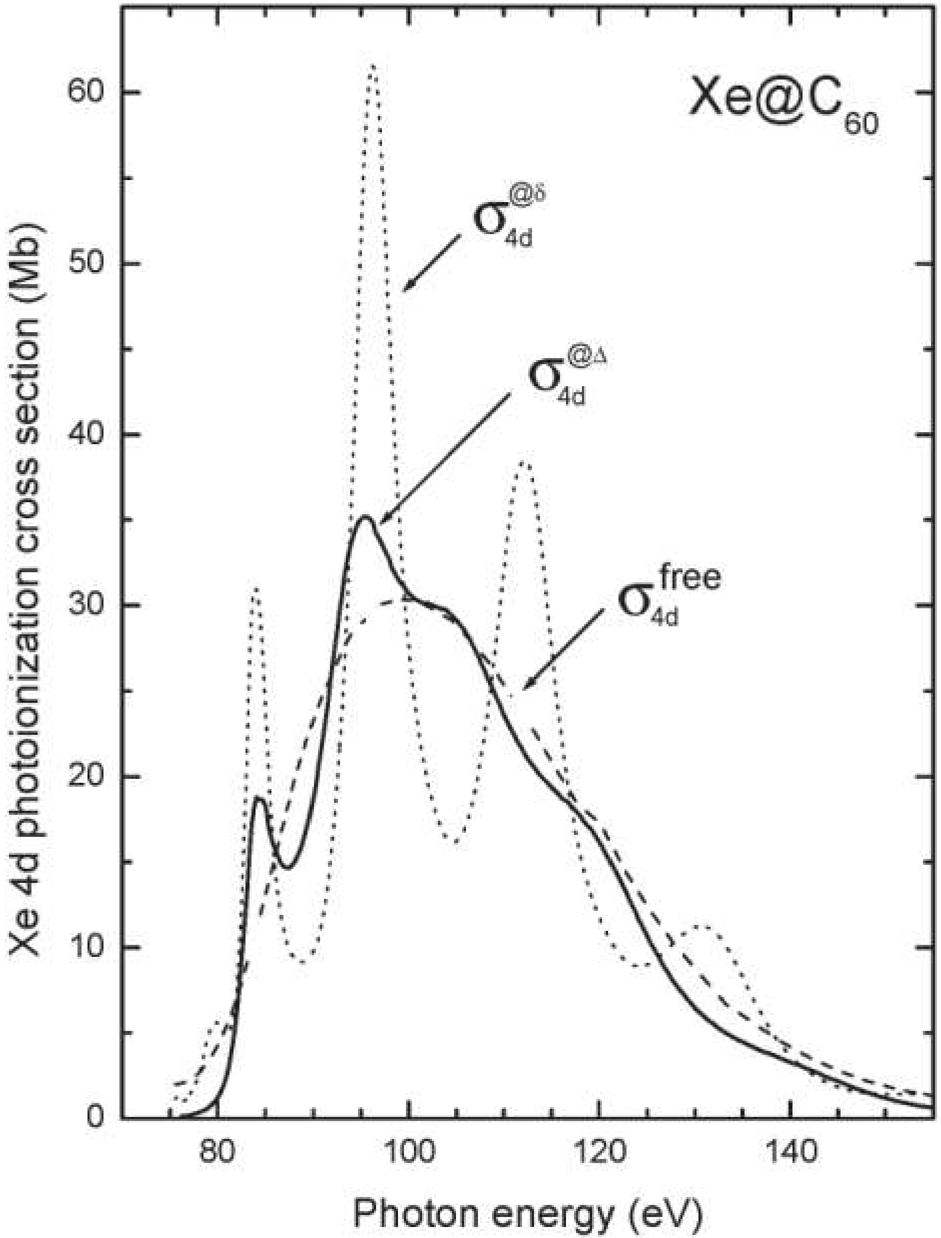
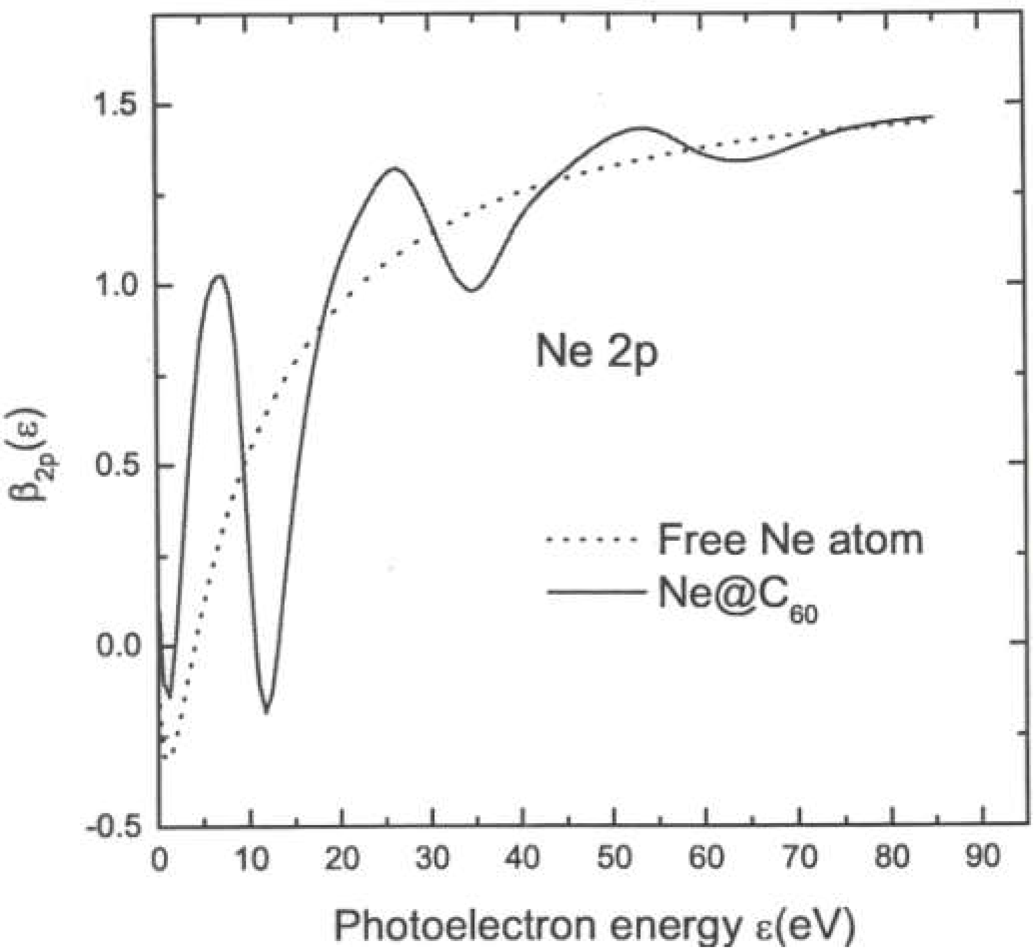
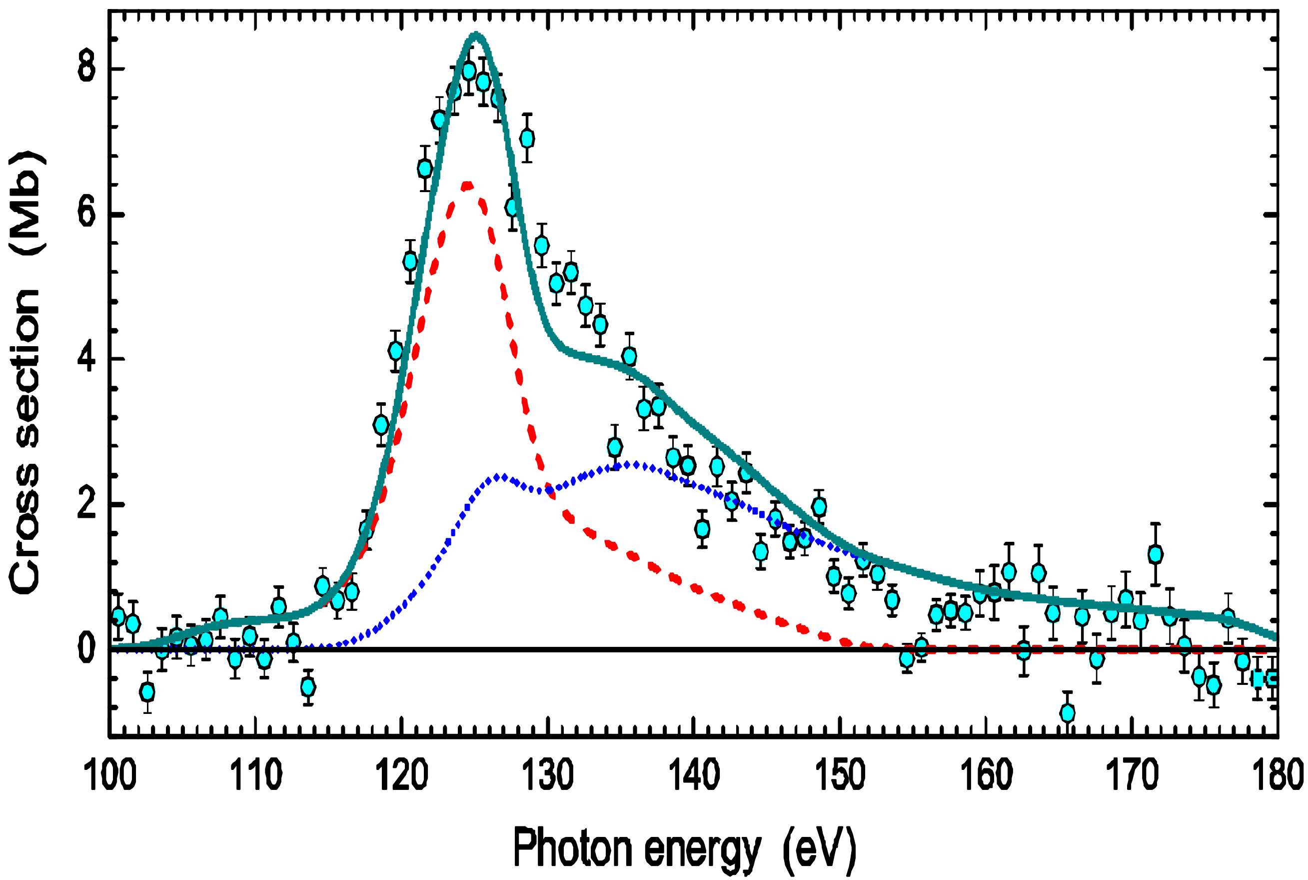


Figure 1. Calculated RPAE results for the 4d photoionization cross section of free Xe, σ_{4d}^{free}, (- - - -), as well as of endohedral Xe@C$_{60}$ calculated within the framework of both the zero-thickness, δ-potential model, $\sigma_{4d}^{@\delta}$ [4] $(\cdots\cdots)$ and a spherical, short-range, attractive potential of finite thickness Δ, $\sigma_{4d}^{@\Delta}$ (——, present work).

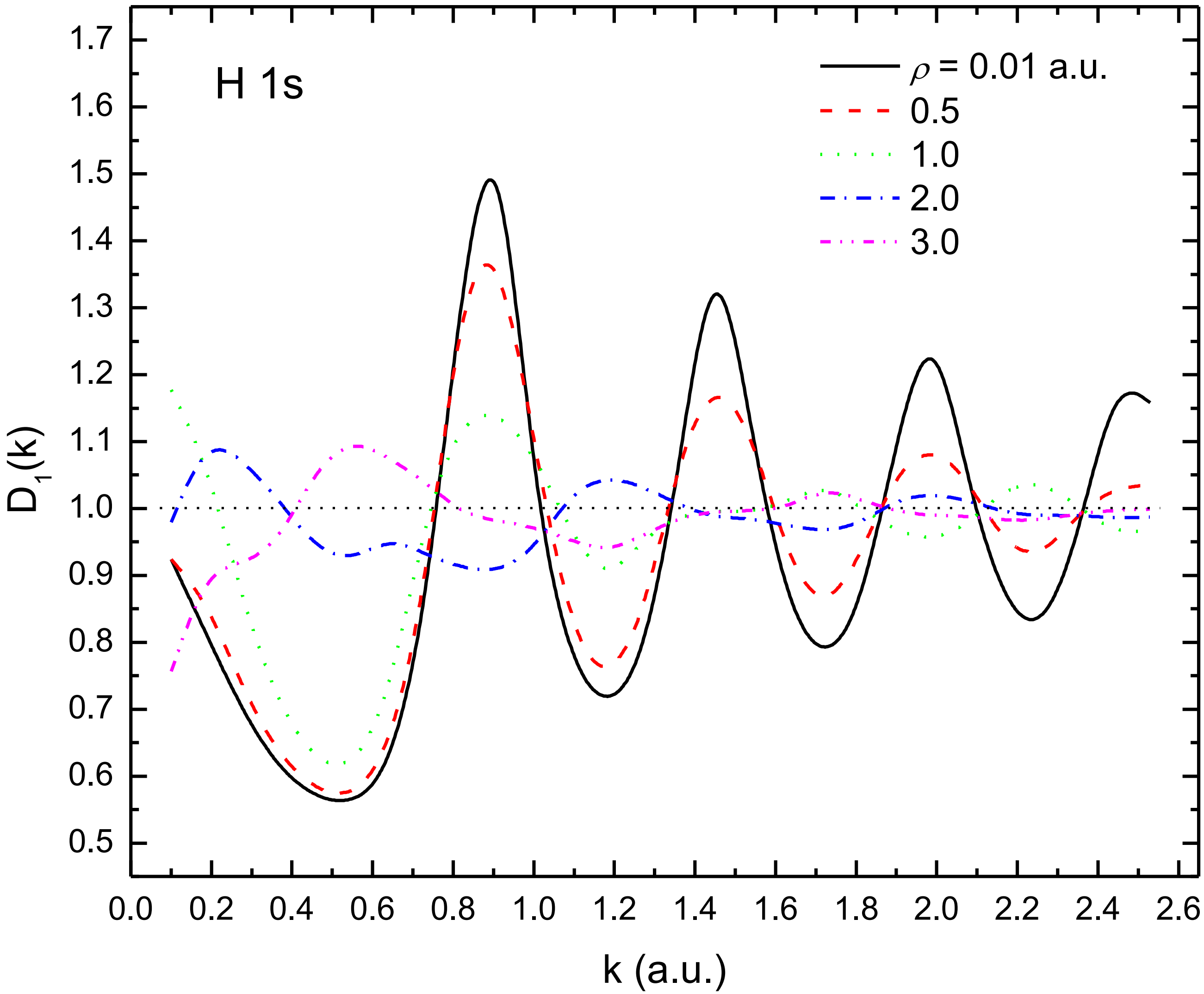


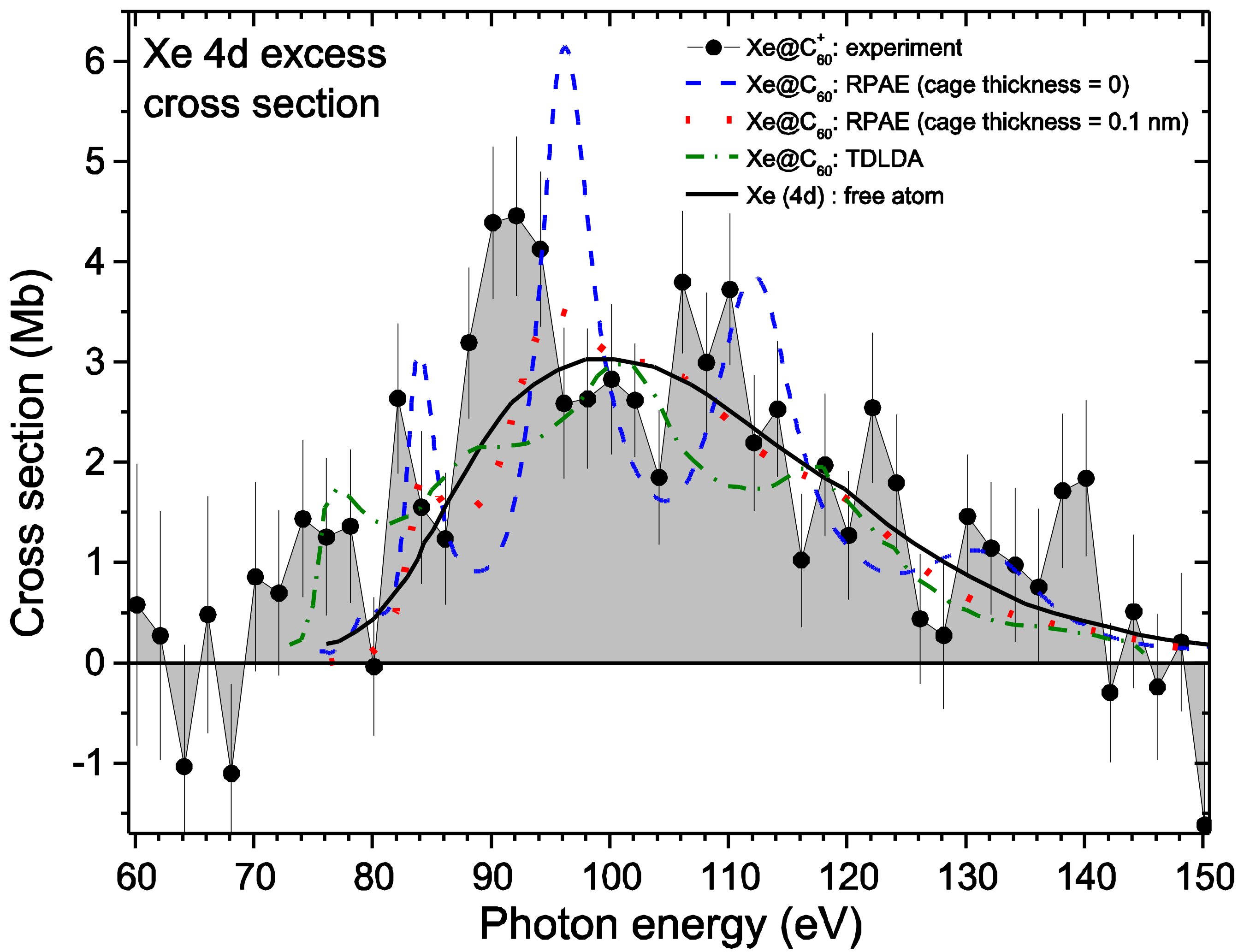
1.5
1.0
0.5
0.0
-0.5
0
10
20
30
40
50
60
70
80
90
Ne 2p
Free Ne atom
Ne@C60
β2p(ε)
Photoelectron energy ε(eV)



Cross section (Mb)
8
6
4
2
0
100
110
120
130
140
150
160
170
180
Photon energy (eV)

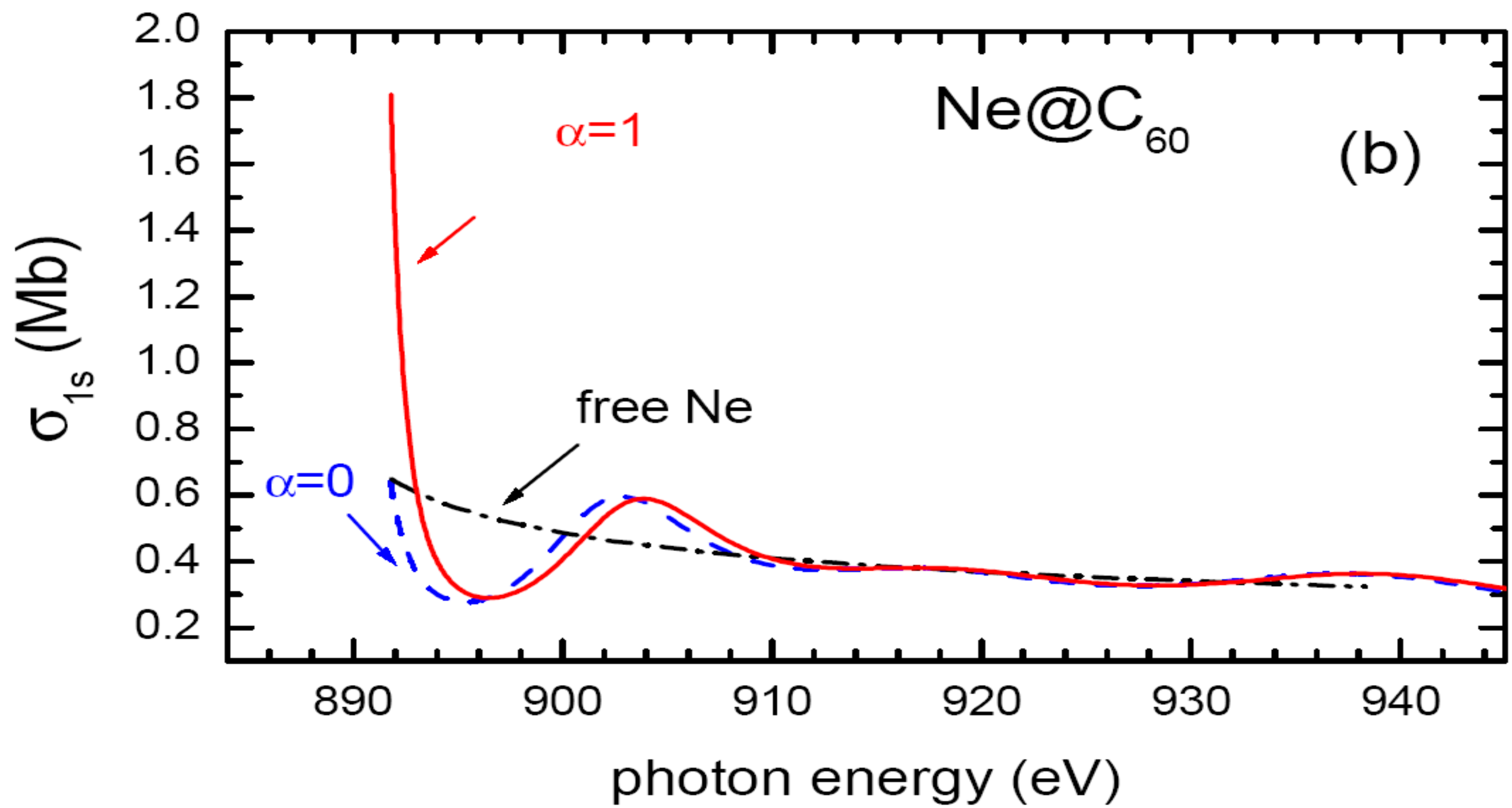
Photoionization of Off-Center H@C60





Xe 4d excess
cross section
Xe@C$_{60}^{+}$: experiment
Xe@C$_{60}$: RPAE (cage thickness = 0)
Xe@C$_{60}$: RPAE (cage thickness = 0.1 nm)
Xe@C$_{60}$: TDLDA
Xe (4d) : free atom
Cross section (Mb)
Photon energy (eV)
6
5
4
3
2
1
0
-1
60
70
80
90
100
110
120
130
140
150

Photoionization of Ne@C60 including polarization nof the cage



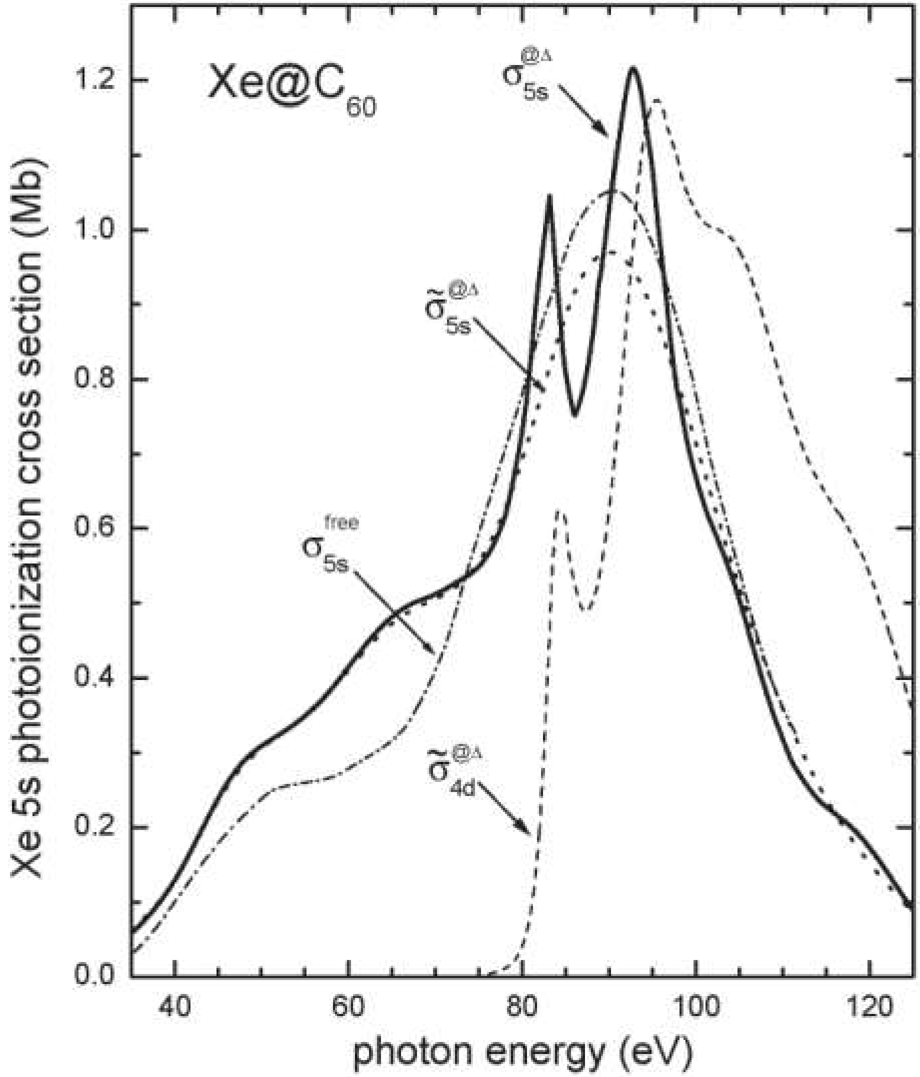


Figure 2. Calculated RPAE results (accounting for interchannel couplings among photo-transitions from $4d^{10}$, $5s^2$, and $5p^6$ subshells) for the 5s photoionization cross section of Xe: — · —, free Xe (σ_{5s}^{free}); ——, Xe@C_{60} in the framework of the present spherical, short-range, finite thickness potential model [$\sigma_{5s}^{@\Delta}(\omega)$]; $\cdots\cdots$, [$\tilde{\sigma}_{5s}^{@\Delta}(\omega)$] , the same as the previous but omitting ordinary confinement resonances in $4d \to f$, p transitions; - - - -, $(1/30)\sigma_{4d}^{@\Delta}(\omega) \equiv \tilde{\sigma}_{4d}^{@\Delta}(\omega)$, $\sigma_{4d}^{@\Delta}$ being the 4d photoionization cross section.