Two-electron photo-excited atomic processes near inner-shell threshold studied by RIXS spectroscopy

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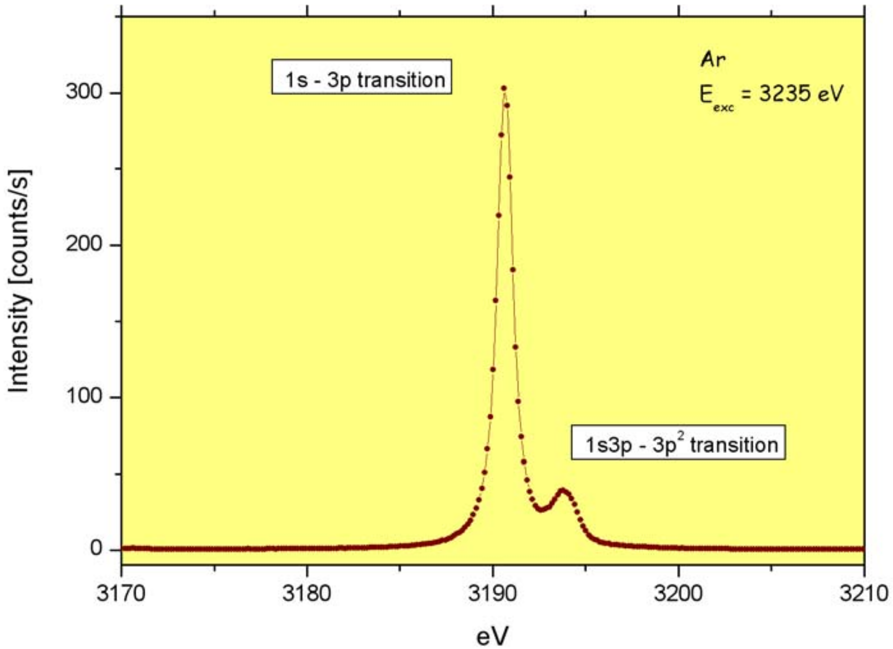
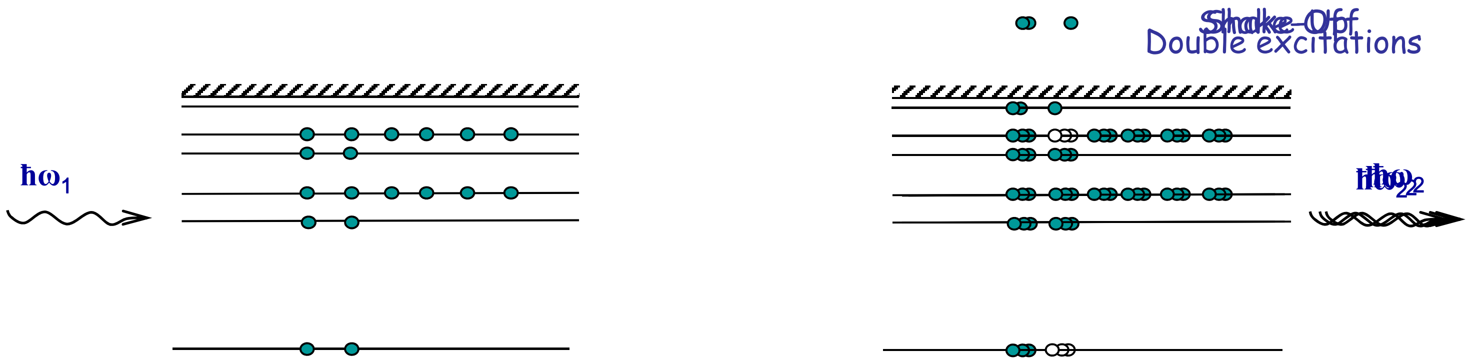
Single photon multi-electron excitations

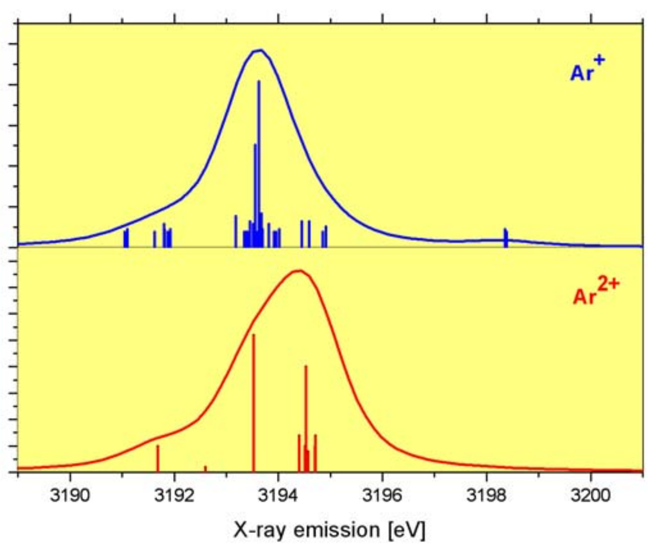
straightforward manifestations of the breakdown of the independent electron picture as they appear due to the electron-electron interactions.



Multi-electron excitations can be studied by high-resolution X-ray measurements.



The main experimental problem hindering the study of near-threshold multi-electron excitations is the overlap of the shake-up, shake-off and double excitation spectral contributions.



MCDF calculated 1s3p – $3p^2$ emission spectrum of Ar

An experimental method commonly used to study multielectronic excitations is near-edge photoabsorption on monoatomic gaseous targets

K edge of Ar:

M. Deutsch, N. Maskil, W. Drube, Phys. Rev. A 46, 3963 (1992).

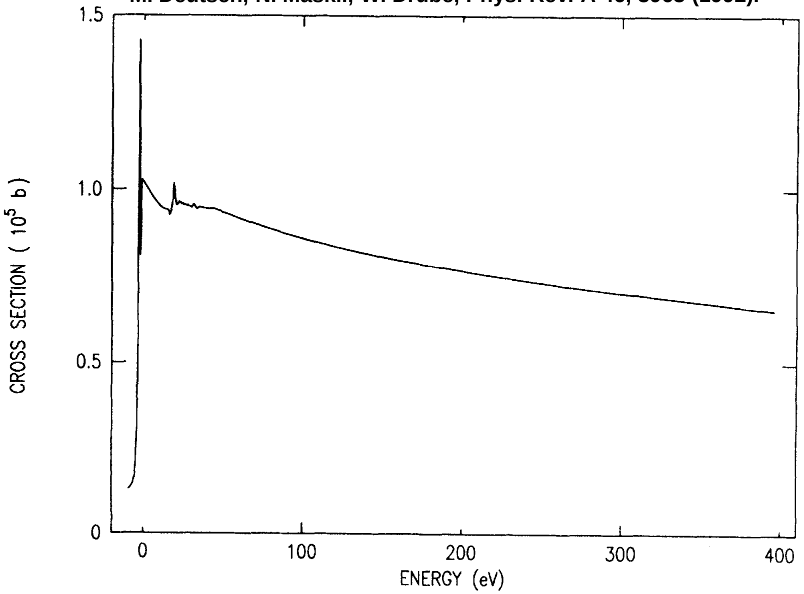
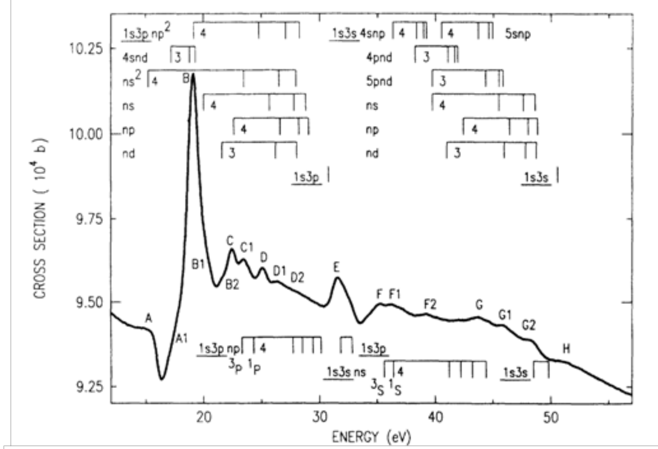
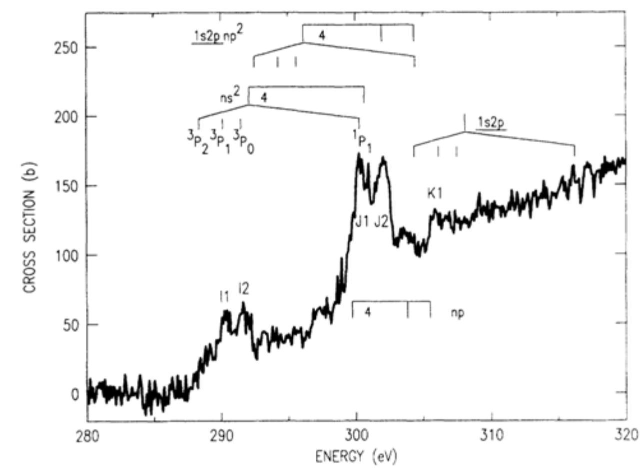


FIG. 1. Near-K-edge photoabsorption spectrum of Ar. Zero energy is at the edge, E_K=3206.26 eV.

1s3p excitations





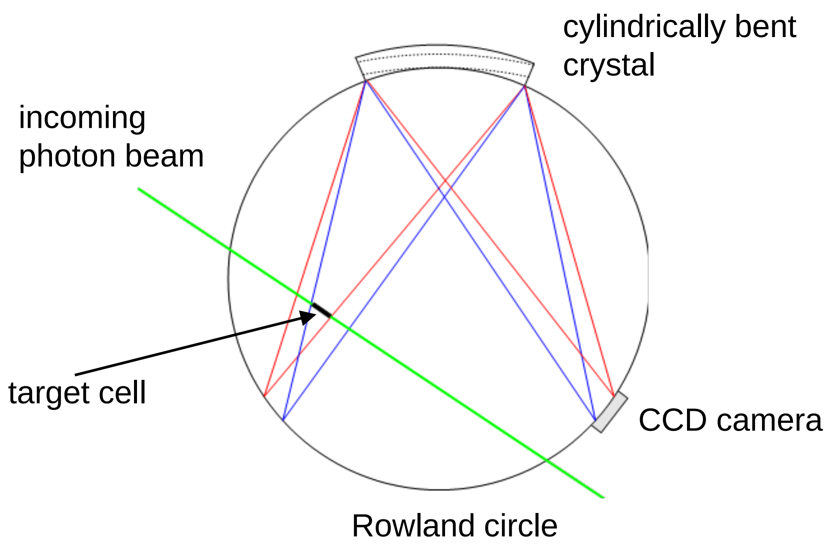
1s2p excitations

RIXS spectroscopy on doubly excited states:

⇓

RIXS spectroscopy on
atomic (gaseous) target

"Off-Rowland circle" geometry combined with position sensitive x-ray detector

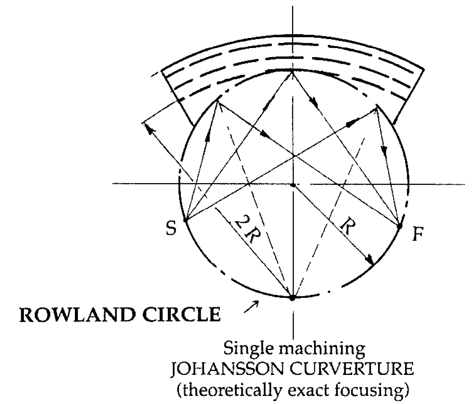


High resolution x-ray spectrometer (HRXRS) at J. Stefan Institute

→ cylindricaly bent crystal in Johansson geometry ($R_{Rowland}$ = 50 cm)

Angular range: **30^0 – 65^0**

crystal	refl. plane	2d[Å]	energy range
Quartz	**(110)**	**8.5096**	**1.6 – 2.9 keV (3.2 - 5.8 keV)**
Si	**(111)**	**6.271**	**2.2 – 4.0 keV**

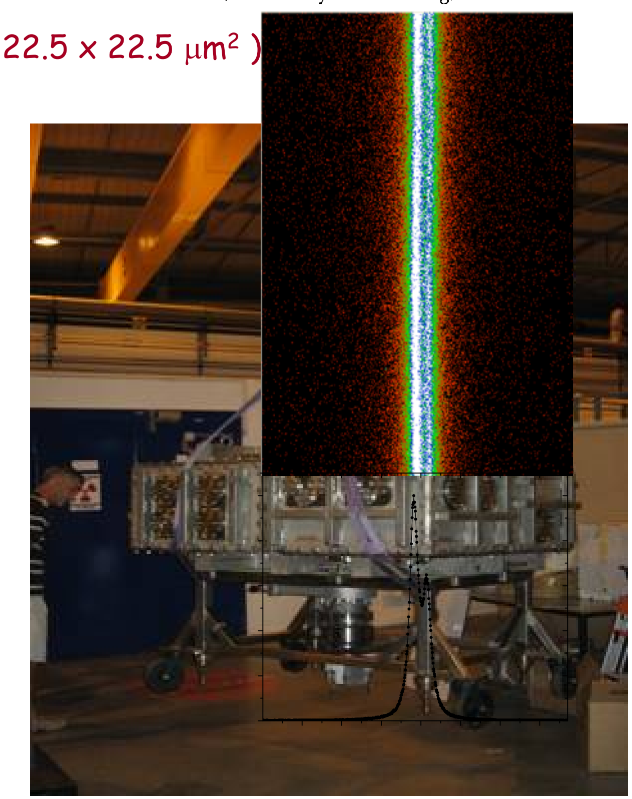


→ diffracted photons are detected by the CCD camera (pixel size 22.5 x 22.5 μm²)

Thermoelectricaly cooled BI CCD camera (ANDOR DX-438-BV)

chip Marconi 555-20, 770x1152 pixels, pixel size 22,5 x 22,5 μm²

CCI-010 controler, readout frequency 1 MHz, 16bit AD conversion,



→ spectrometer is enclosed in the 1,6 x 1,3 x 0,3 m³ stainless steel vacuum chamber

working pressure: 10^{-6} mbar

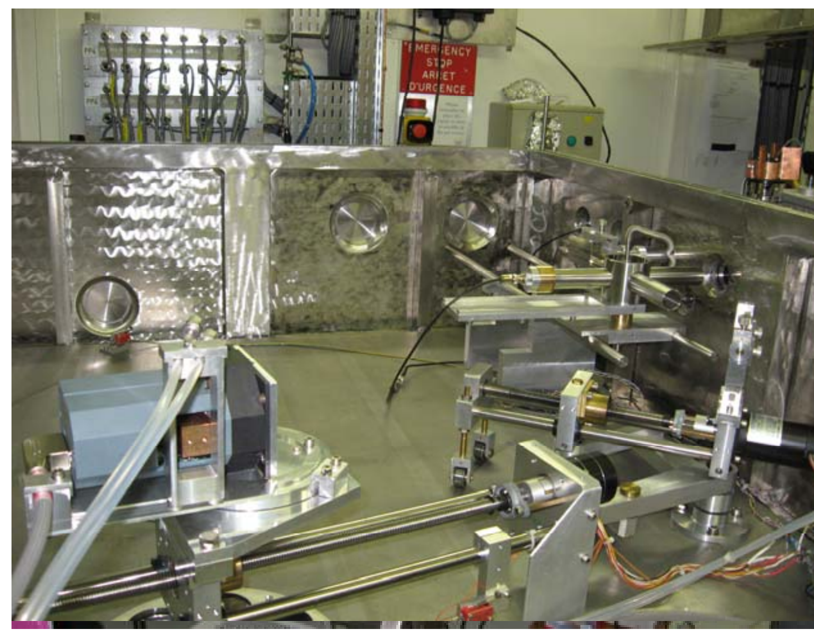
1s3p and 1s2p double excitations in Ar:

ID26 beamline

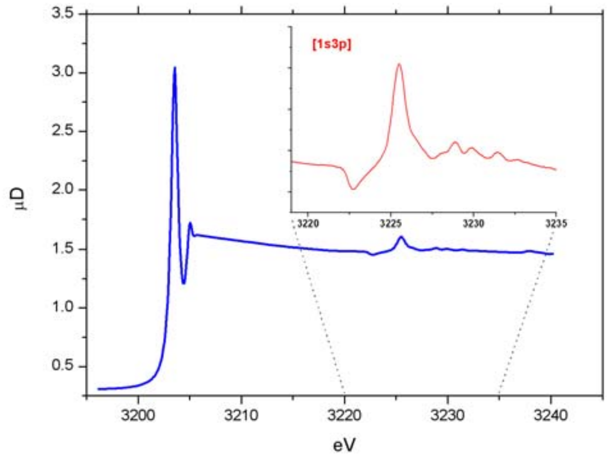
- cryogenically cooled double Si(111) crystal monochromator
- energy resolution ~ 0.5 eV at the Ar K edge
- photon flux ~ 5×10^{12} /s
- suppresion of higher harmonics by two Si mirrors
- beamsize 50(height) x 250(width) μm^2

JSI HRXRS

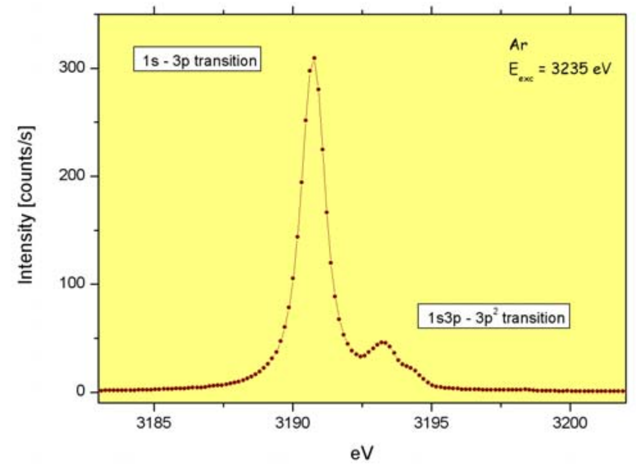
- Target cell (12.5 mm Kapton windows) filled with 500 mbar of Ar
- Si(111) diffraction crystal
- energy resolution ~ 0.6 eV at the Ar KM line



K absorption edge of Ar:

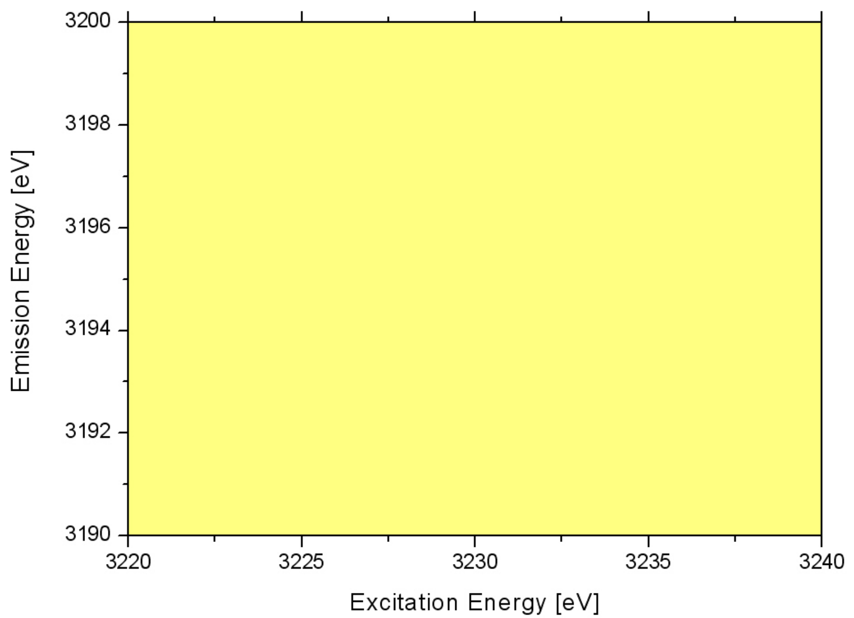


HR KM X-ray line of Ar:



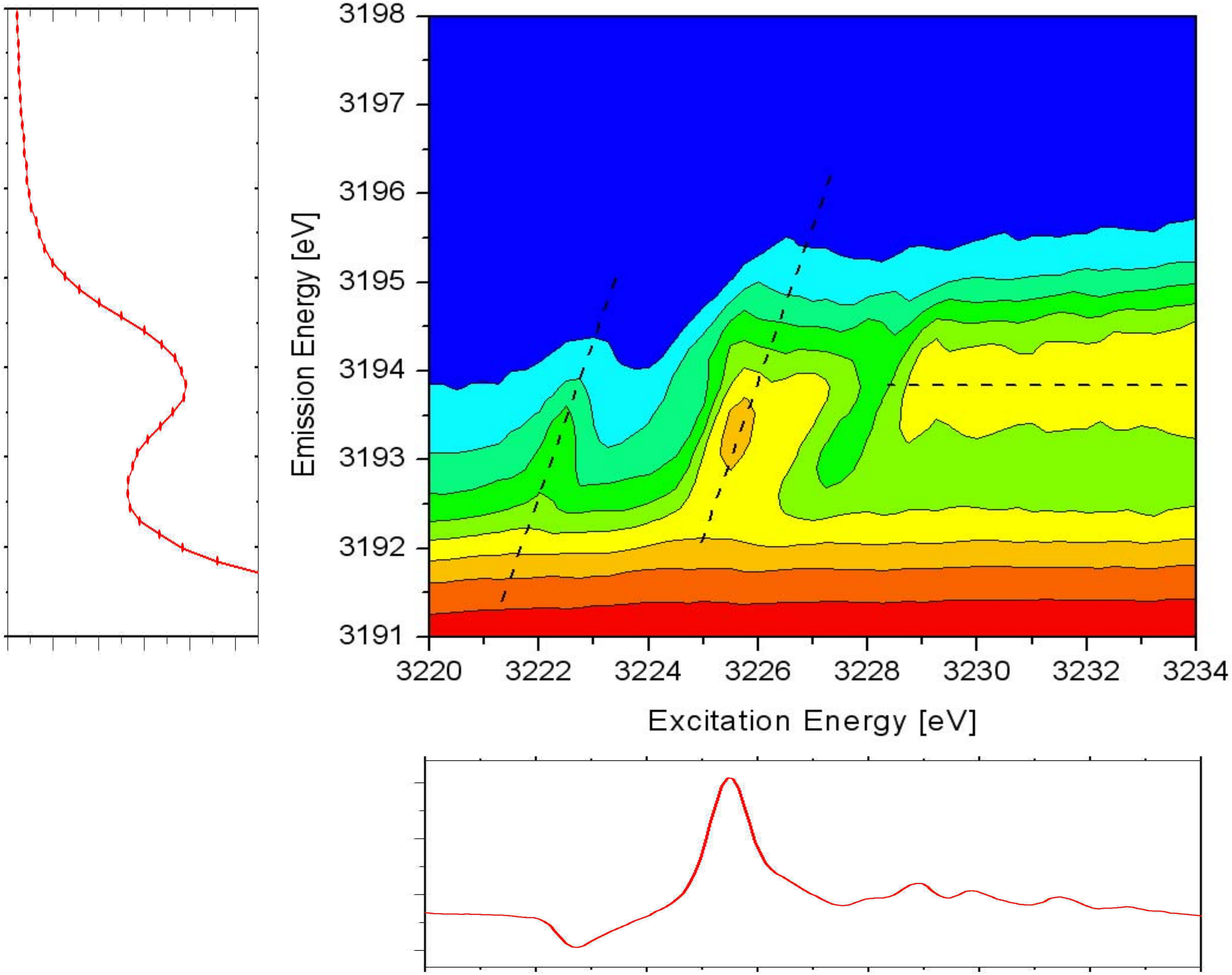
Build a full 2D RIXS map of [1s3p] doubly excited states:

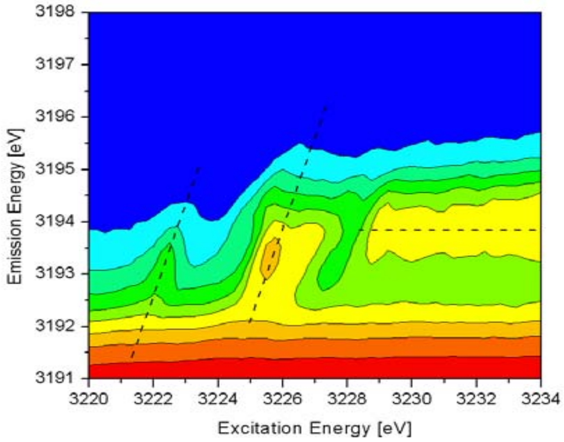
Energy resolution ~ 0.6 eV
Energy step ~ 0.15 eV

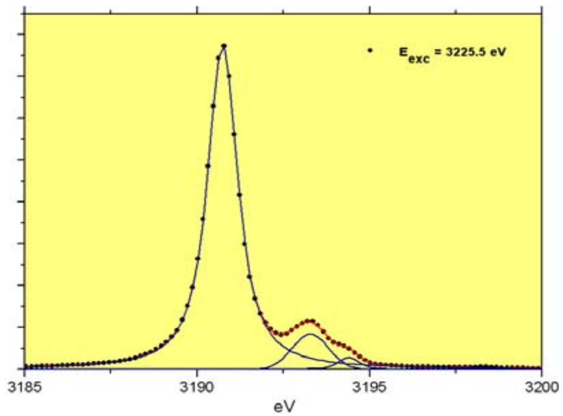


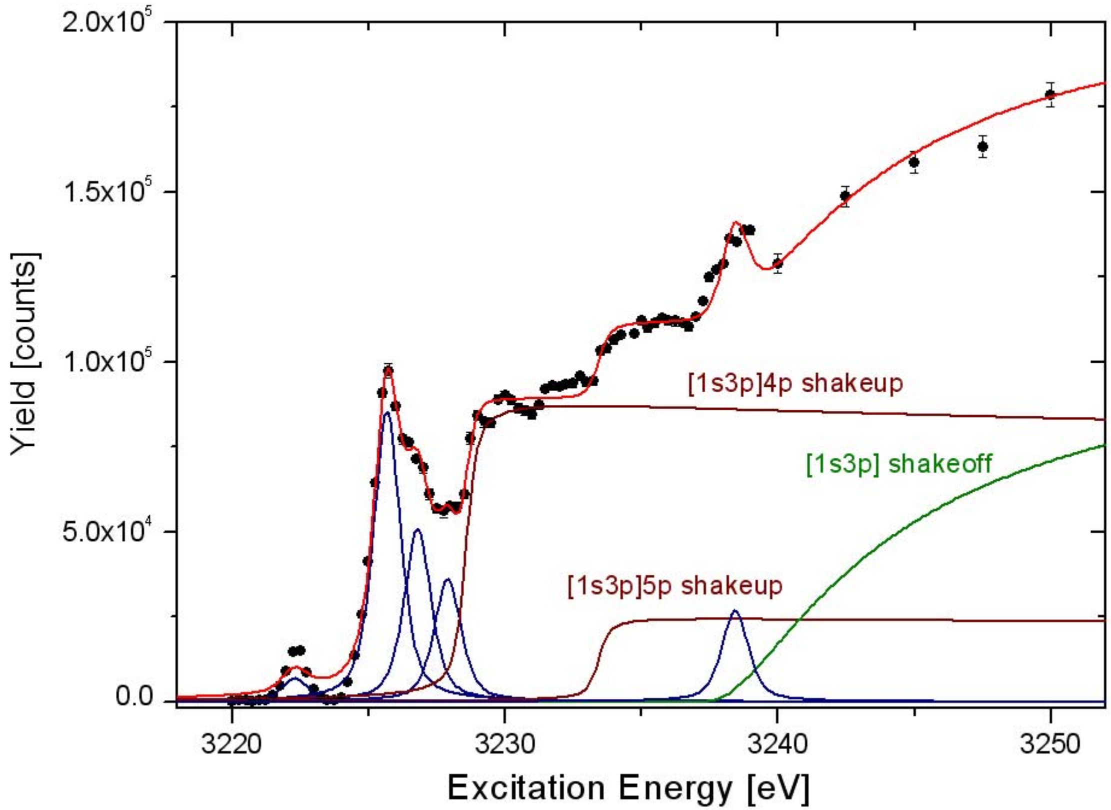
Energy resolution ~ 0.5 eV
Energy step ~ 0.25 eV

In this full RIXS plane a different character of separate multielectron contributions is clearly manifested!







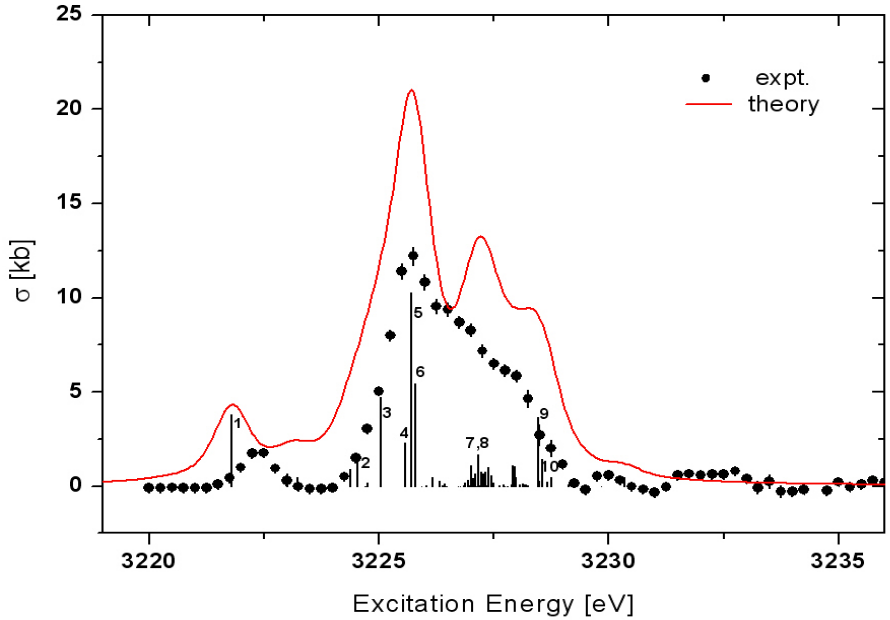


[1s3p] = 3236.3 ± 0.7 eV
[1s3p]4p = 3228.56 ± 0.04 eV
[1s3p]5p = 3233.43 ± 0.08 eV

Theoretical single configuration HF values

[1s3p] = 3237.1 eV
[1s3p]4p = 3228.7 eV
[1s3p]5p = 3232.9 eV

The experimental [1s3p]nln'l' excitation spectrum



Calculations of single configuration energy levels provide only a tentative assignment of the main spectral features.

A precise theoretical description requires elaborate multiconfiguration energy and cross-section calculation, which takes into account correlations as well as the relaxation effects.

Results of our MCDF calculated dipole oscillator strengths into [1s3p]nln'l' excited atomic states.

To represent excited states, the singlet P terms of eight configurations were mixed in the average level (AL) mode:

$4p^2$, $3d^2$, $4s^2$, 3d4s, 4p5p, 4s4d, 3d4d and $4d^2$.

N	E [eV]	$f \times 10^4$	
1	3221.80	0.473	0.89 $4s^2$ / 0.09 $4p^2$
2	3224.53	0.208	0.68 $4p^2$ / 0.19 $3d^2$
3	3225.04	0.586	0.39 4s4d / 0.24 $4p^2$
4	3225.59	0.288	0.73 4s4d / 0.13 $4p^2$
5	3225.71	1.274	0.58 4s4d / 0.25 $4p^2$
6	3225.80	0.676	0.48 4s4d / 0.22 $4p^2$
7	3227.16	0.209	0.54 4p5p / 0.37 3d4d
8	3227.17	0.178	0.47 3d4d / 0.42 4p5p
9	3228.46	0.452	0.52 3d4d / 0.37 4p5p
10	3228.55	0.181	0.82 3d4d / 0.11 4p5p

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Separation of Two-Electron Photoexcited Atomic Processes near the Inner-Shell Threshold

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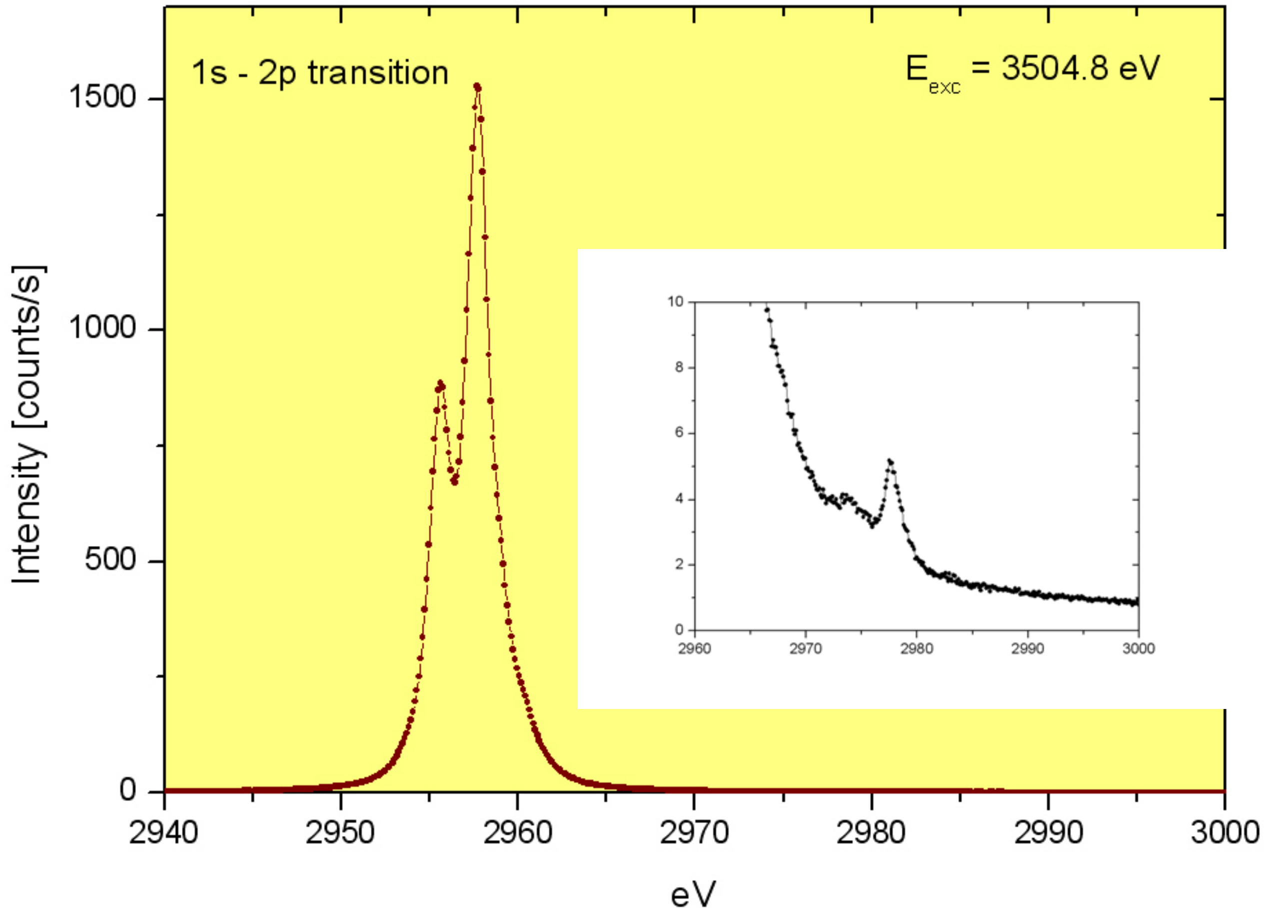
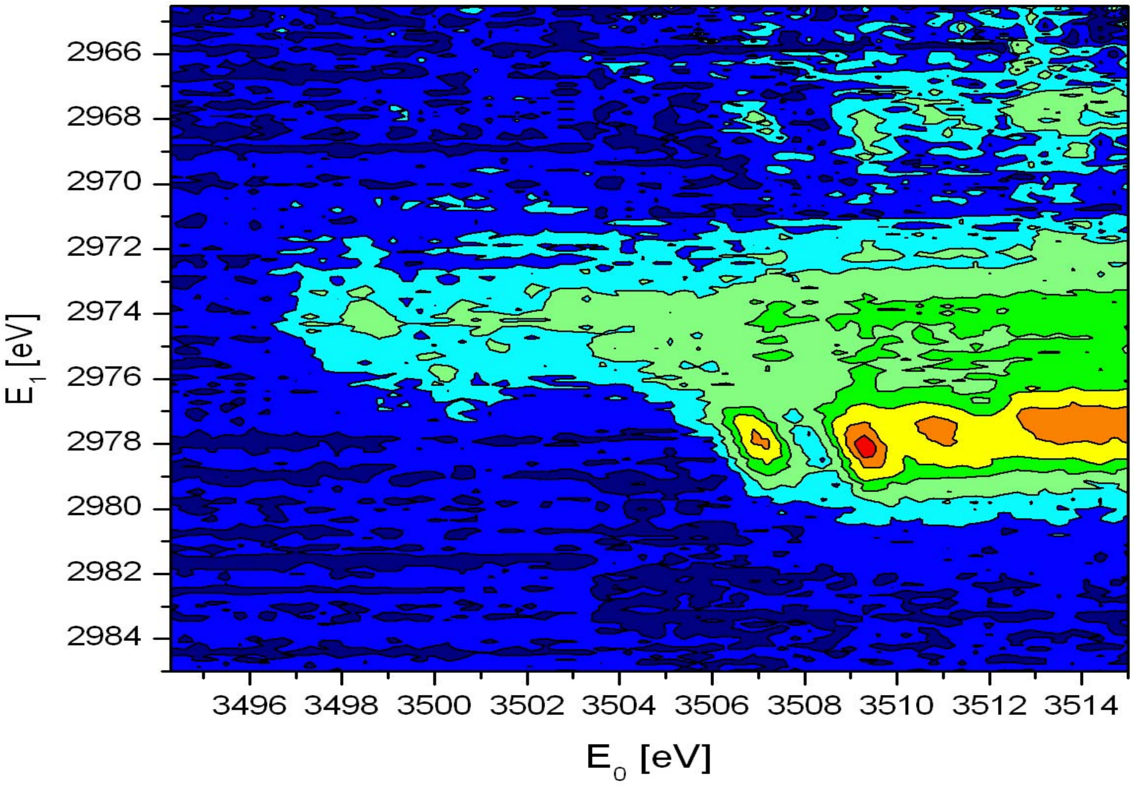
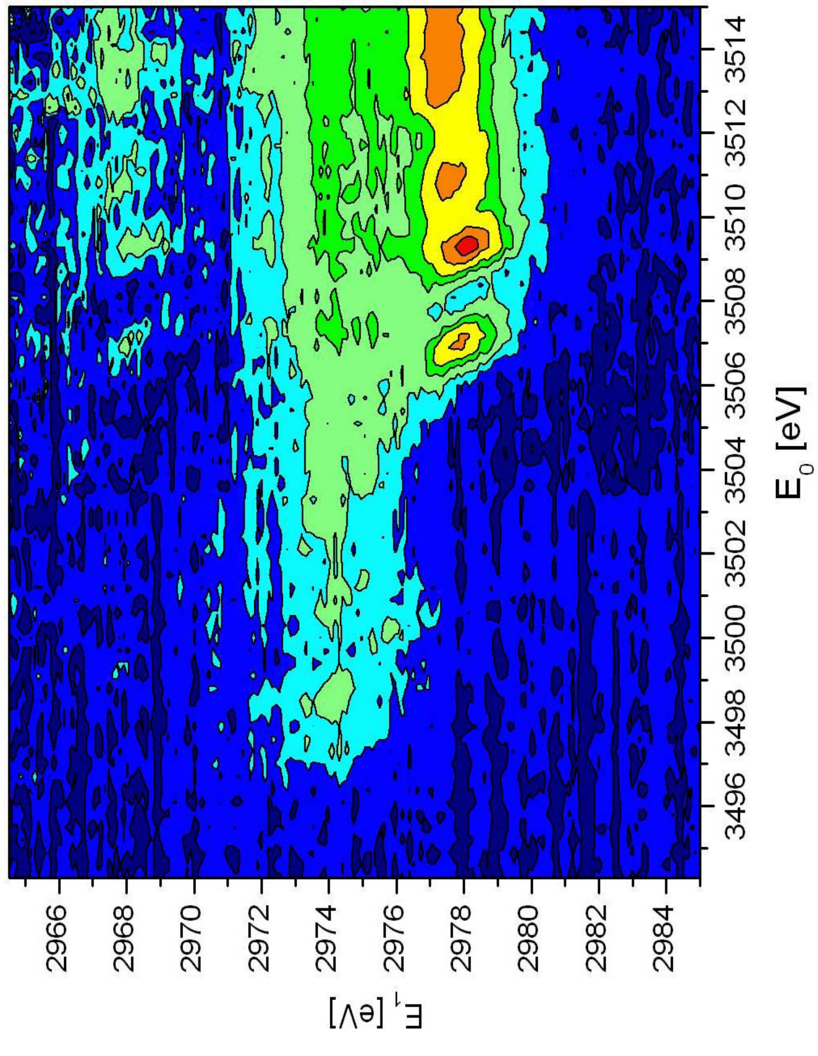
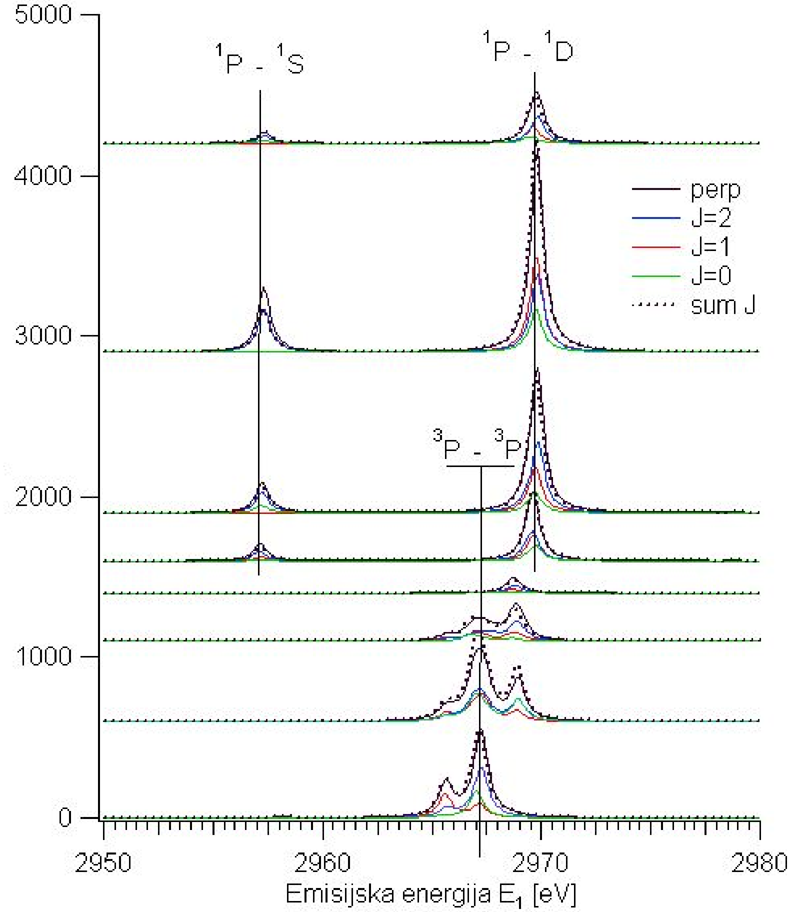
[1s2p] double excitations:

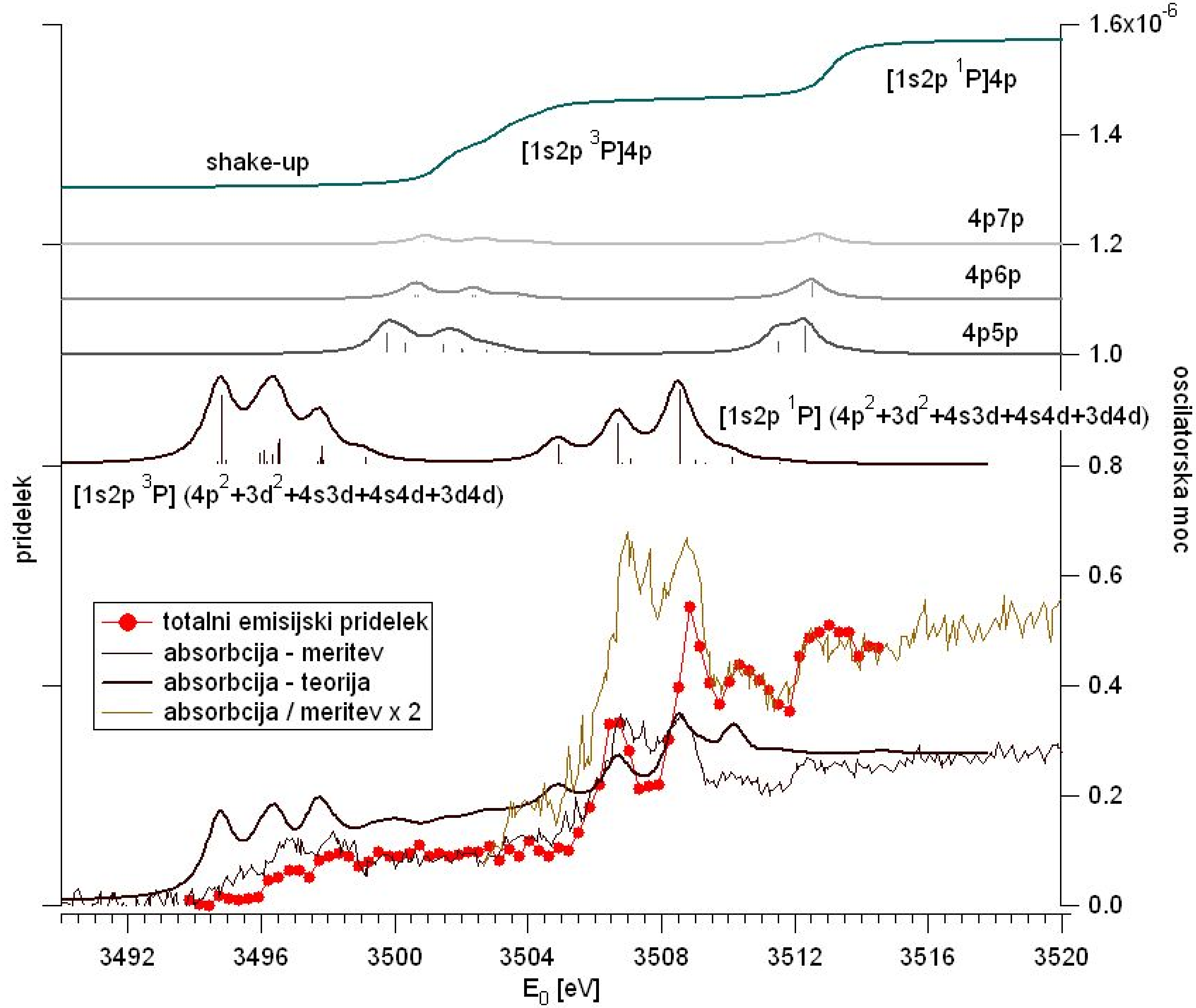


A 2D RIXS map of [1s2p] doubly excited states:




E1 [eV]
E0 [eV]


1P - 1S
1P - 1D
3P - 3P
perp
J=2
J=1
J=0
sum J
Emisijska energija E1 [eV]


1.6x10^-6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
shake-up
[1s2p 3P]4p
[1s2p 1P]4p
4p7p
4p6p
4p5p
[1s2p 1P] (4p2+3d2+4s3d+4s4d+3d4d)
[1s2p 3P] (4p2+3d2+4s3d+4s4d+3d4d)
pridelek
oscilatorska moc
totalni emisijski pridelek
absorbcija - meritev
absorbcija - teorija
absorbcija / meritev x 2
3492
3496
3500
3504
3508
3512
3516
3520
E0 [eV]

Conclusion:

- This work signs up RIXS as an important tool to study multielectron excitations in gaseous targets, with the potential that reaches beyond that of the absorption technique at least in two points:
  effective experimental separation of multielectron excitations from the dominant single inner hole decay,
  the energy resolution is not limited by energy width of the inner hole and the energy

- We are capable to perform high resolution RIXS studies of dilluted targets in a due time. Presented approach could be applied to other low density targets such as metallic vapors (involving open shell in the initial state), molecular targets (involving competitive decay channels, elaborated basis sets and structural effects) or gaseous targets in the external fields.