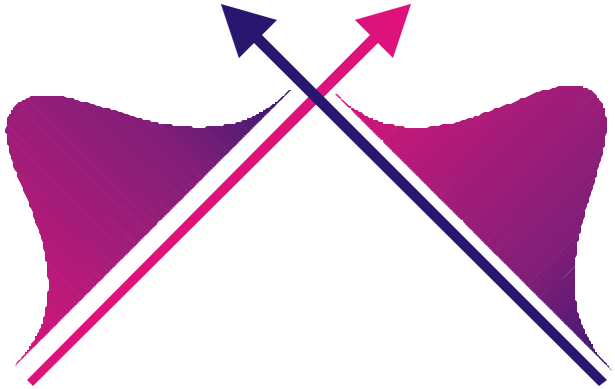
Time resolved photoelectron spectroscopy with ultrafast soft x-ray light



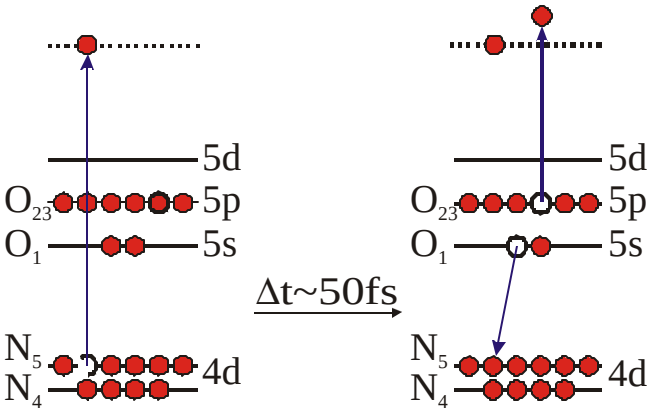
Lora Nugent-Glandorf

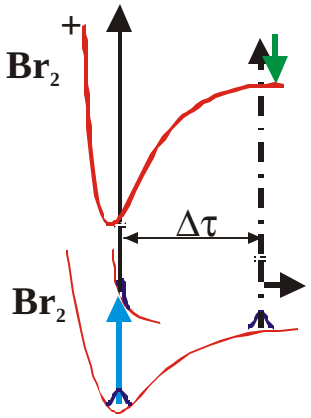
Michael Scheer, David Samuels, Anneliese Mulhisen
Edward Grant, Xeuming Yang
M. Krishnamurthy, Jennifer Odom, Ronnie Bierbaum

Stephen R. Leone

Femtosecond spectroscopy with an XUV light source

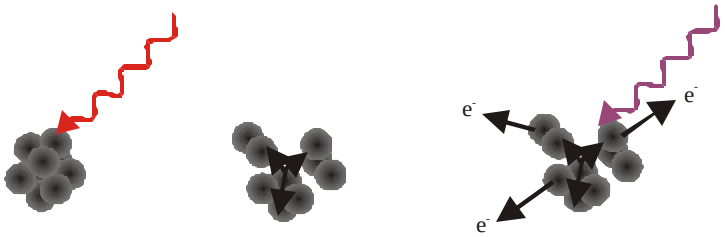
Atoms:
Core excited states
Rydberg Wavepackets





Molecules:
Bound wavepackets
Dissociation Dynamics
Valence and Core level

Clusters:
Fragmentation dynamics
Time-dependent energy
re-distrubution



Coherent UV, XUV ultrafast light source

800 nm

70 fs, 2.5 mJ/pulse, 1 kHz repetition rate

Non-linear crystal doubling

400 nm

100 μJ/pulse

~1×10^{14} photons/pulse

Wave-mixing in NLC

266 nm

60 μJ/pulse

~5×10^{13} photons/pulse

Rare gas high-order harmonic generation

ARGON

13th-29th harmonics

(62 nm -28 nm)

10^{6}-10^{7} photons/pulse

NEON

13th-65th harmonics

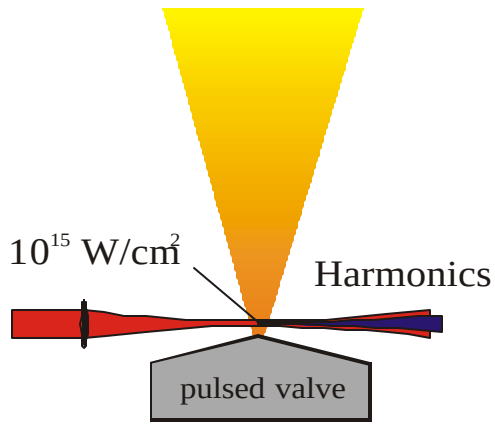
(62 nm-12 nm)

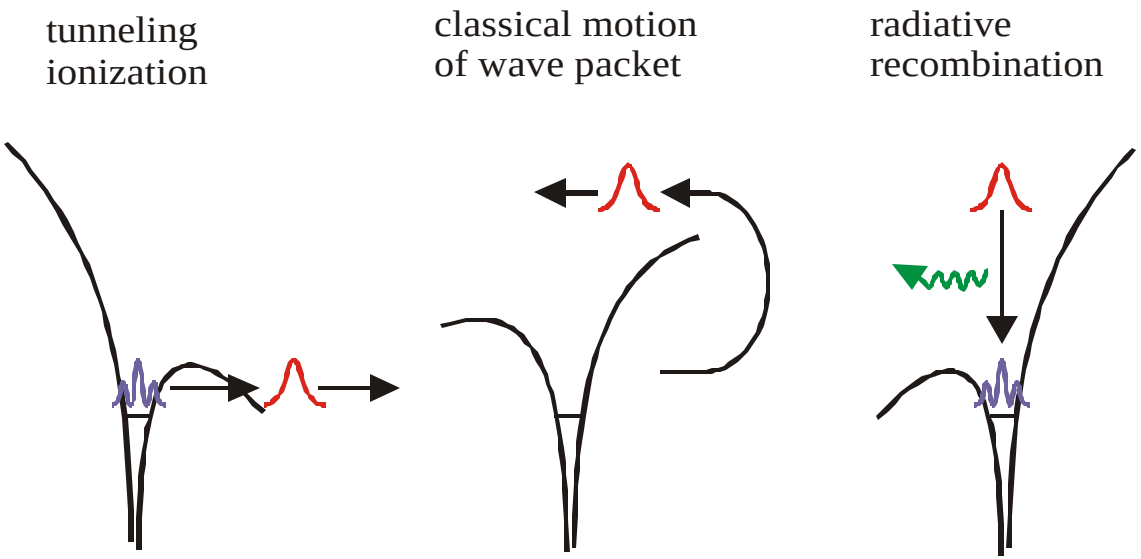
10^{4}-10^{5} photons/pulse

High-Order Harmonic Generation

Relevant parameters:

Pulse duration	70 fs
Peak power	10^{11} W
Spot size	50 mm
Focal position	-1 to 1 mm
Gas medium	Ne, Ar
Gas density	10^{18} cm^{-3}



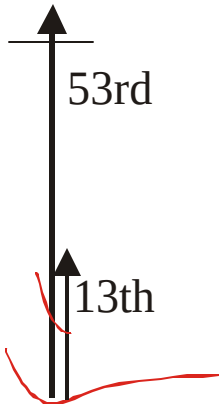


Harmonics:

Valence and Core level photoelectron spectra of atoms and molecules

Spectral Bandwidth of Harmonics

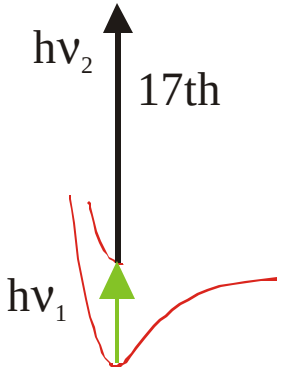
Tuning to resonant core-excitations



Excited State photoelectron spectroscopy:

Spectrum of the excited bound B state of Br_2

Initial and final electronic and vibrational states

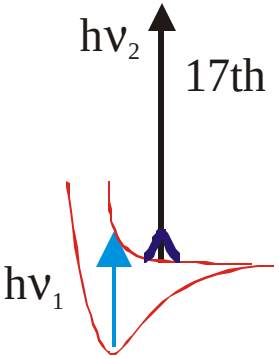


400 nm + harmonics time-resolved:

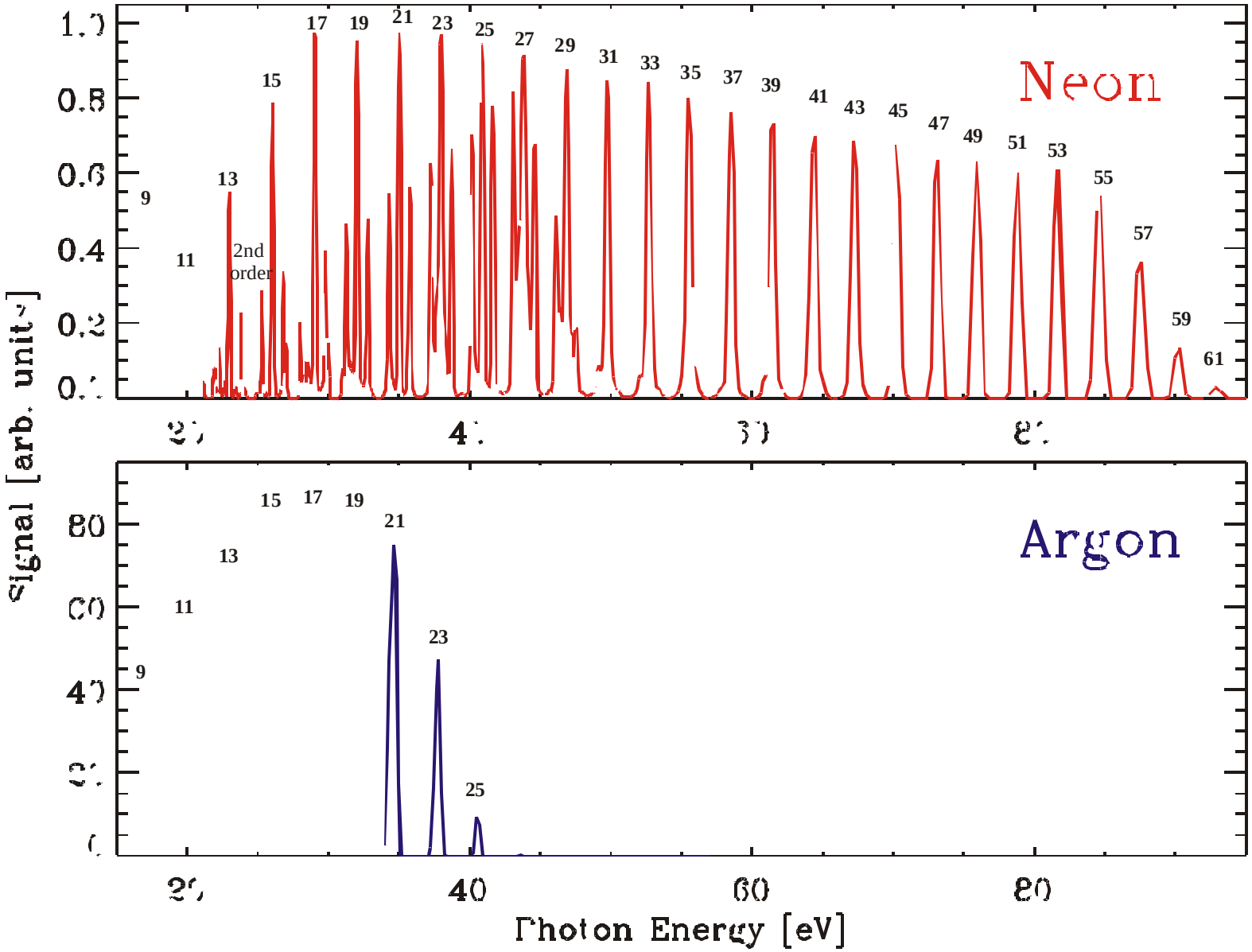
Cross correlation measurements

Compression of harmonic soft x-ray pulses

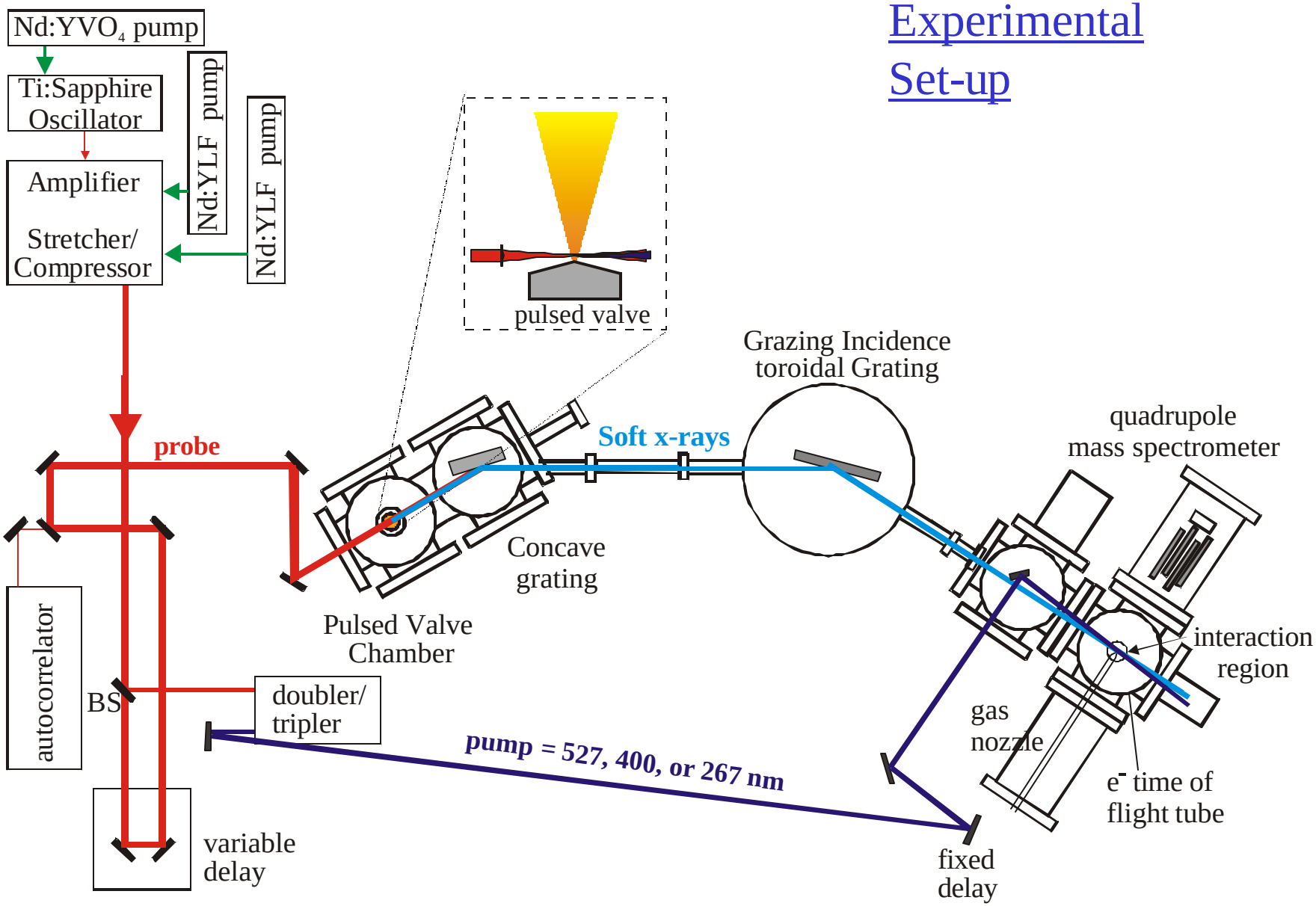
Photodissociation of Br_2

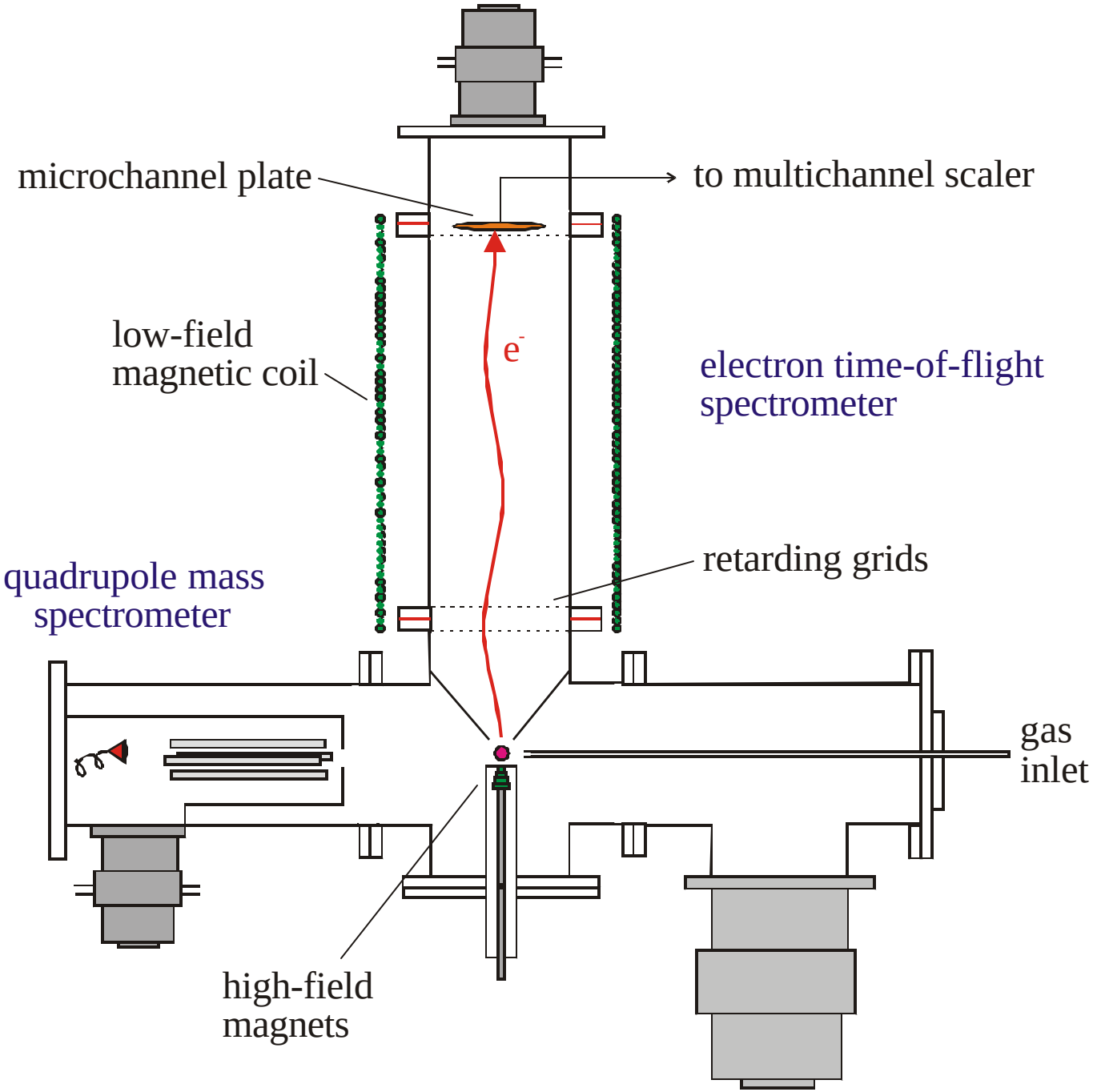


Harmonics of an 800 nm Ti:Sapphire laser

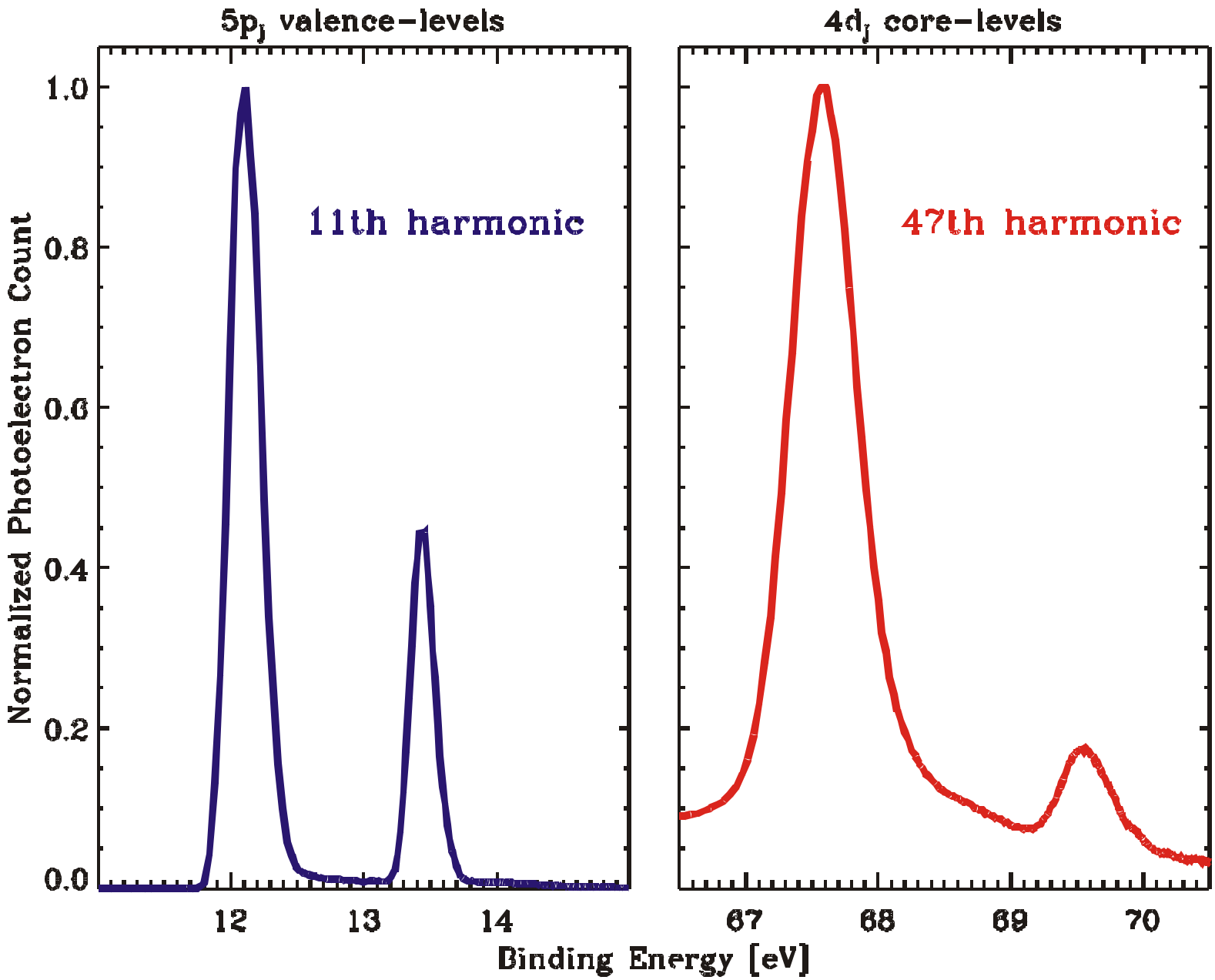


Experimental Set-up

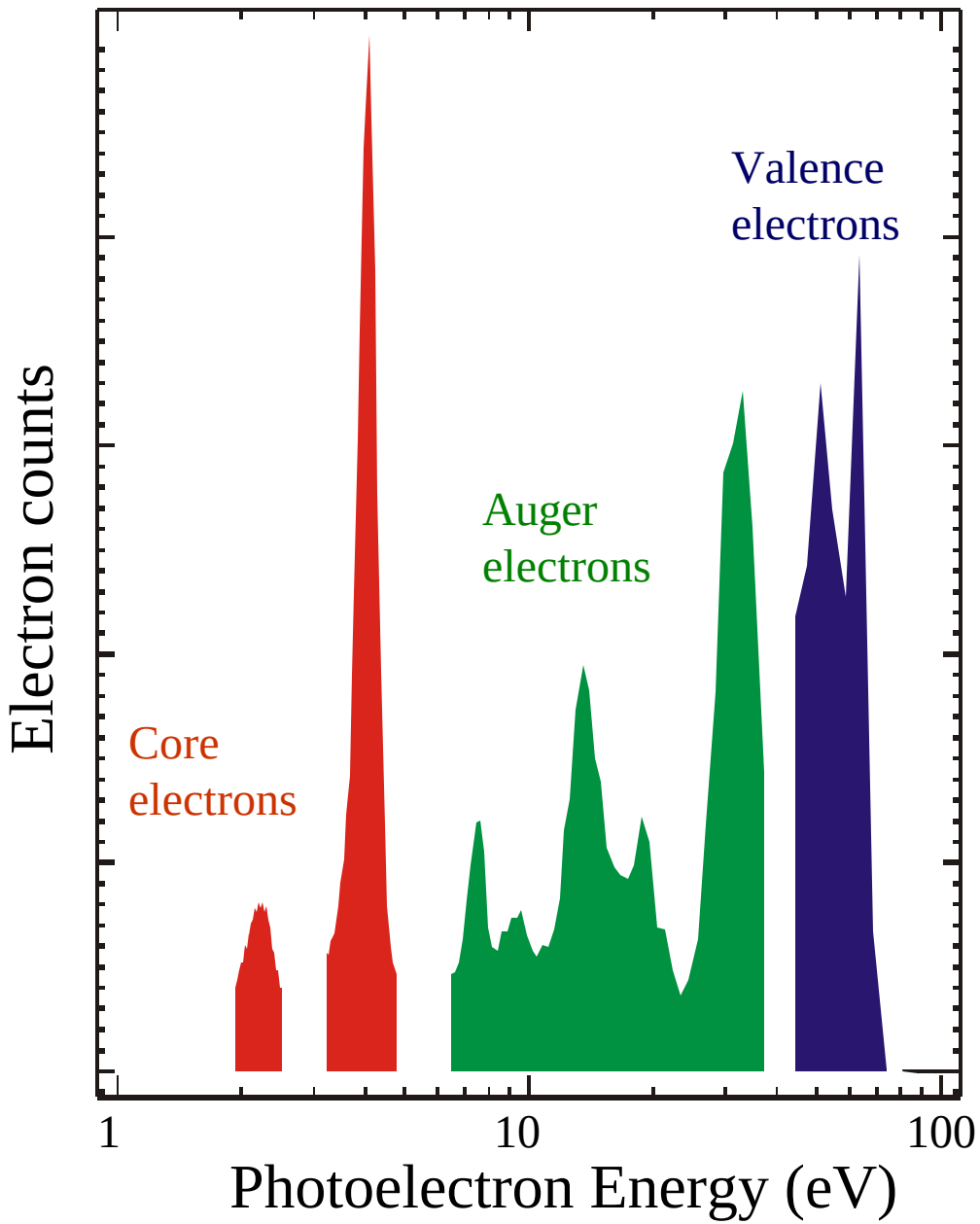


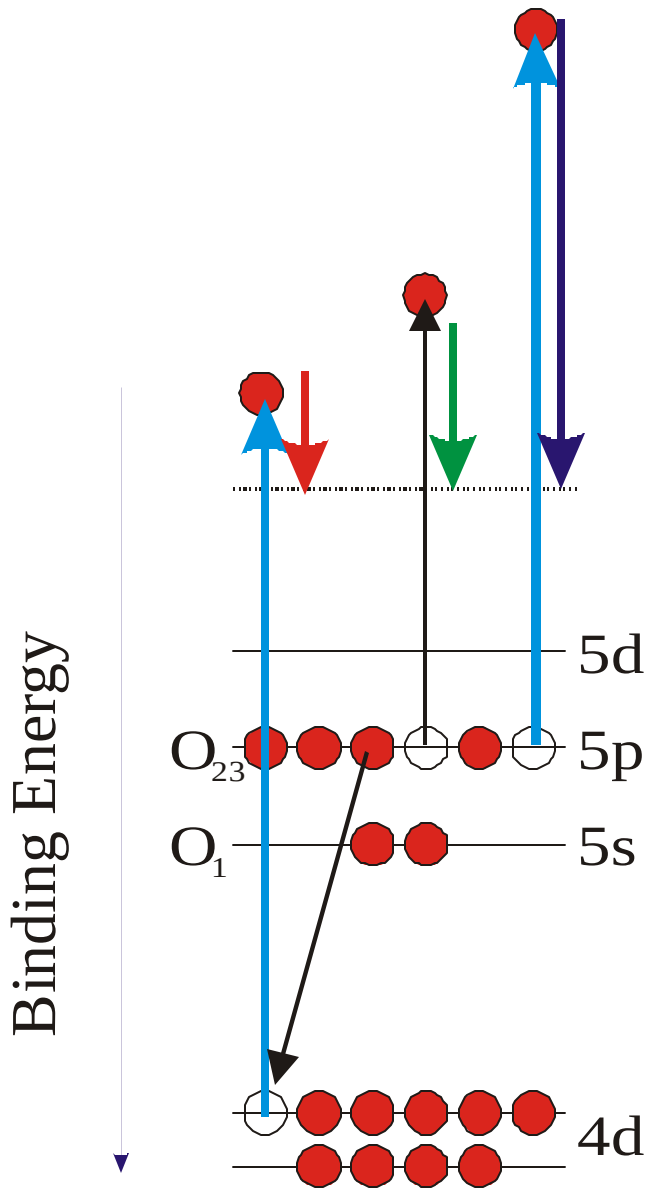

microchannel plate
to multichannel scaler
low-field
magnetic coil
e-
electron time-of-flight
spectrometer
retarding grids
quadrupole mass
spectrometer
gas
inlet
high-field
magnets

Photoelectron Spectra of Xenon

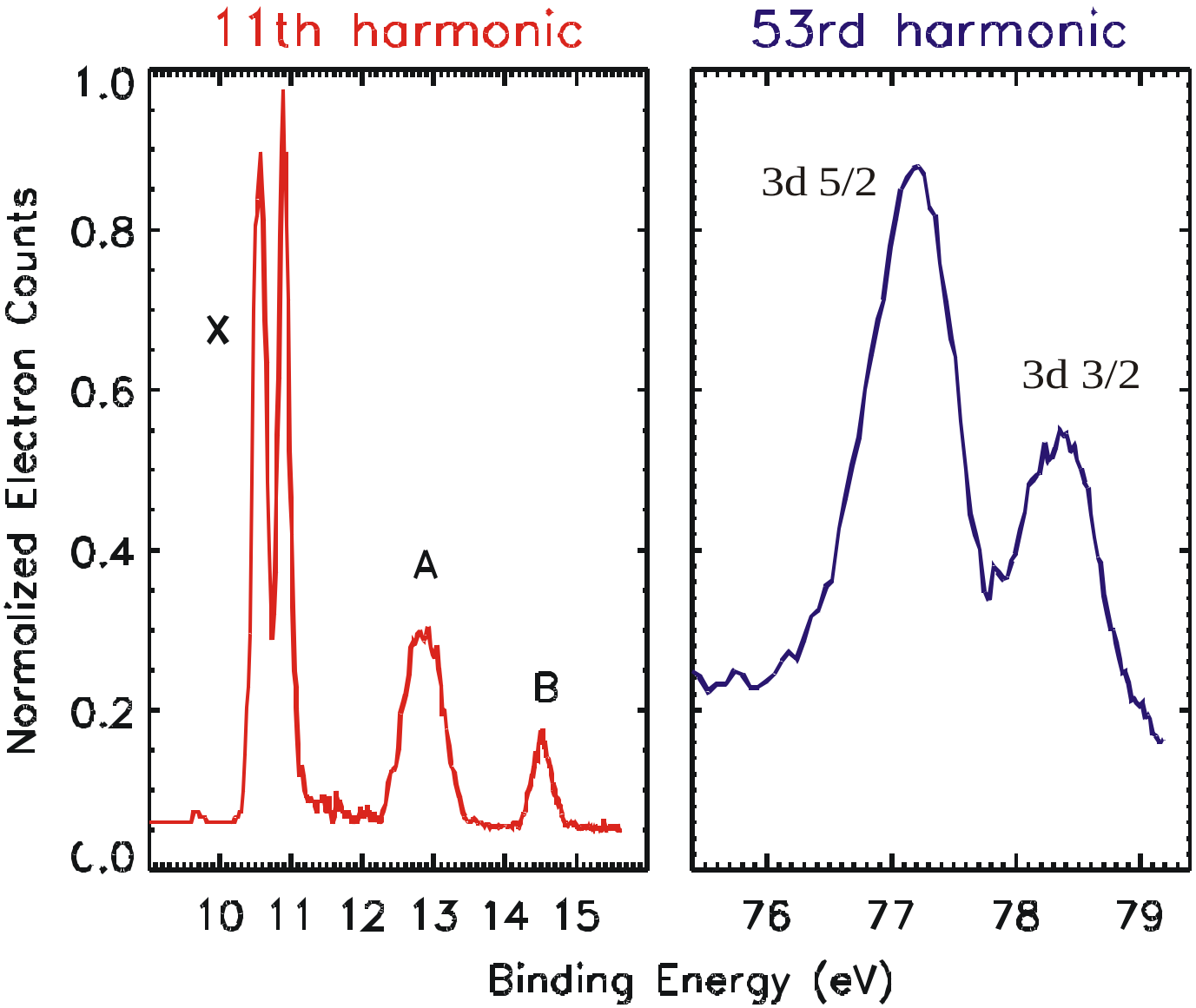


Photoelectron Spectrum of Xenon (47th harmonic)

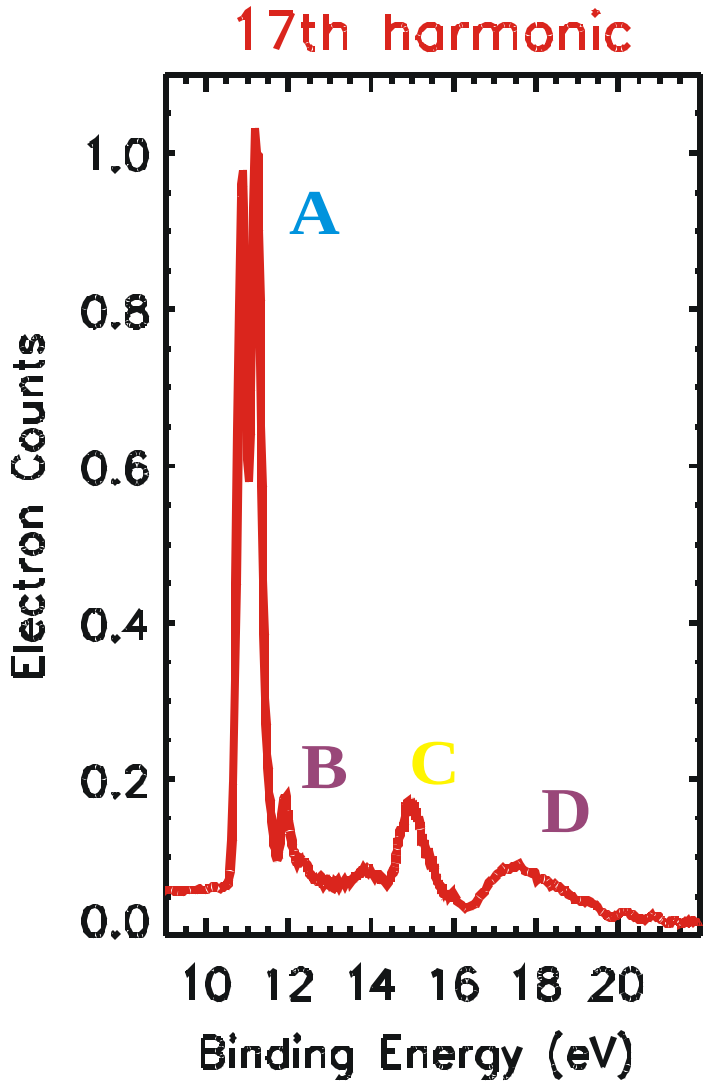


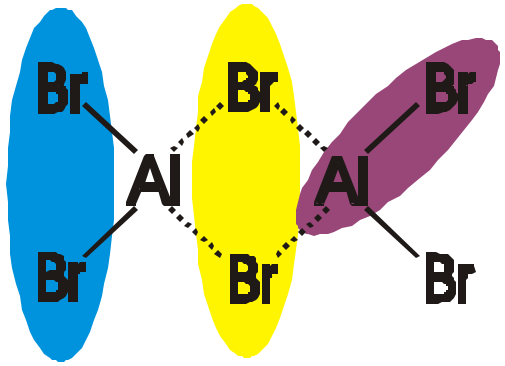


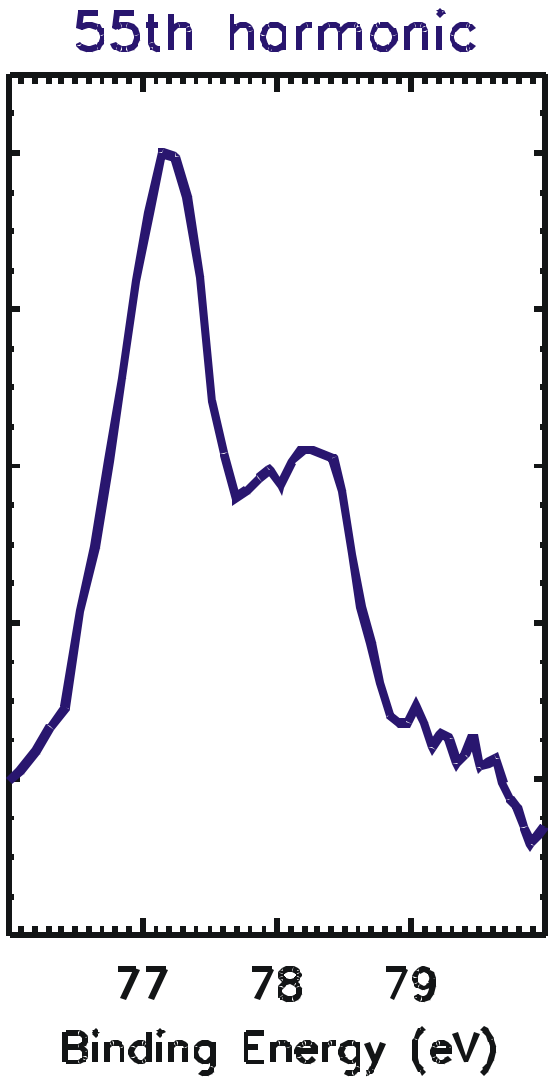
Photoelectron spectra of Br_2



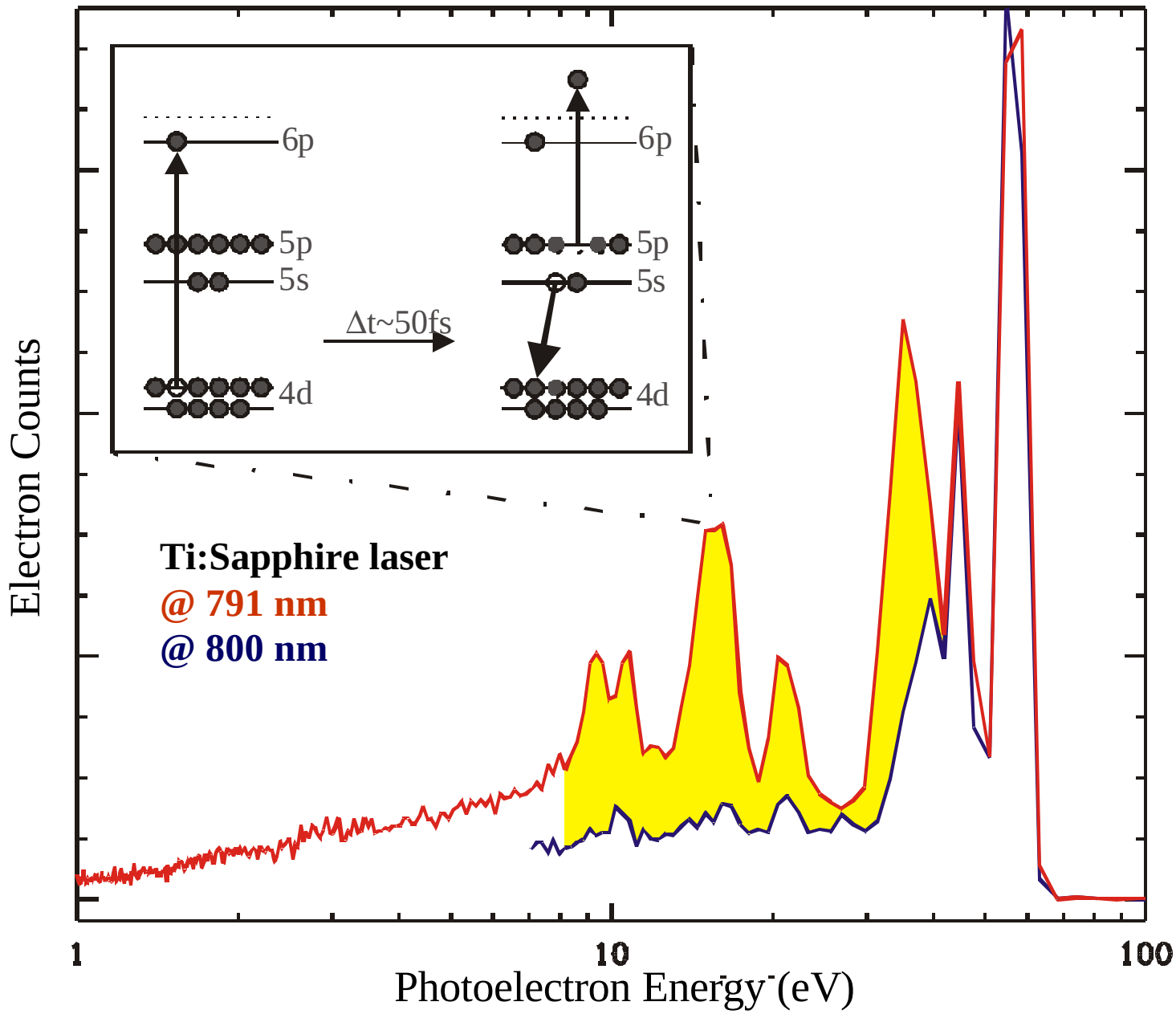
Photoelectron spectra of Al_2Br_6



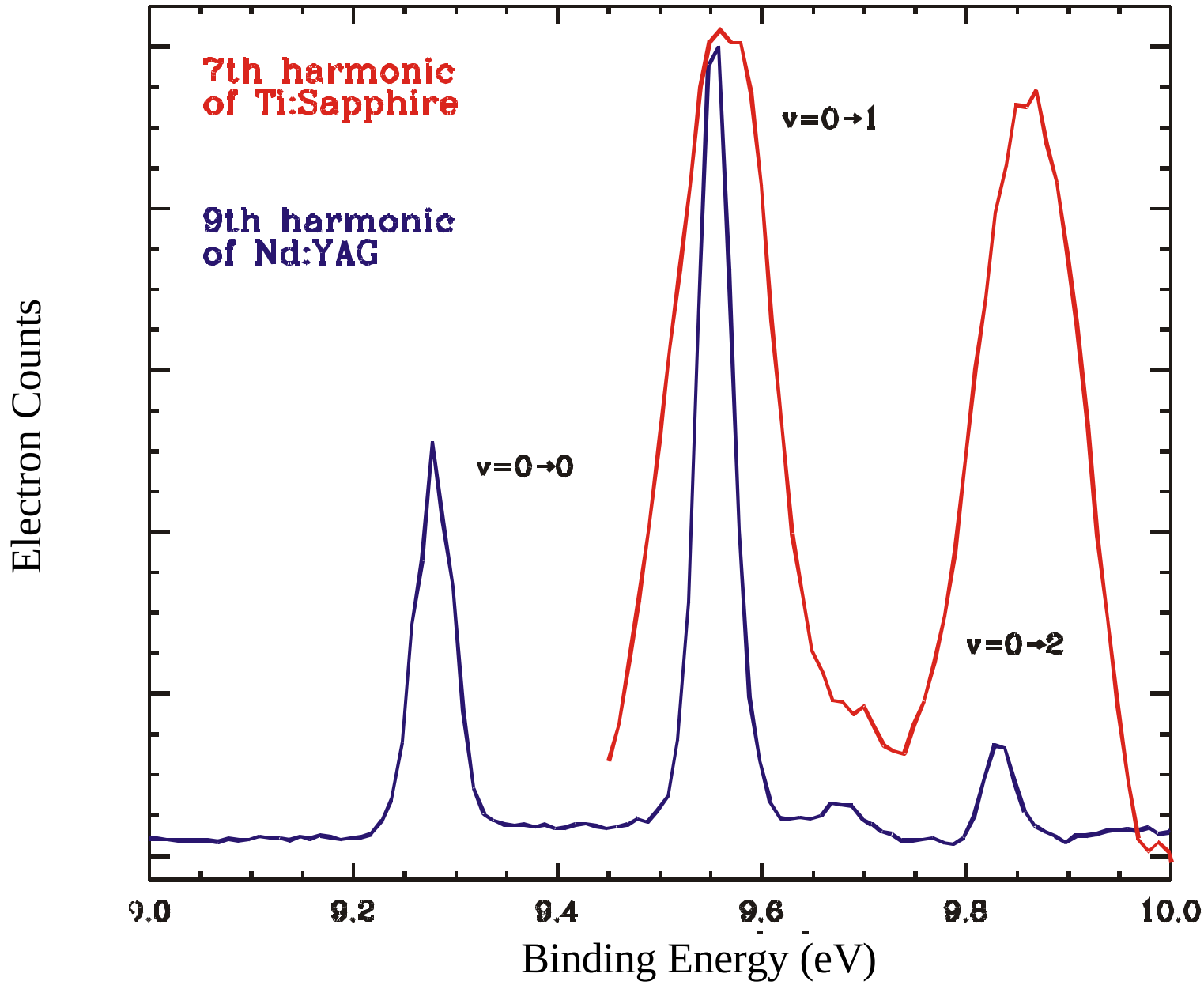




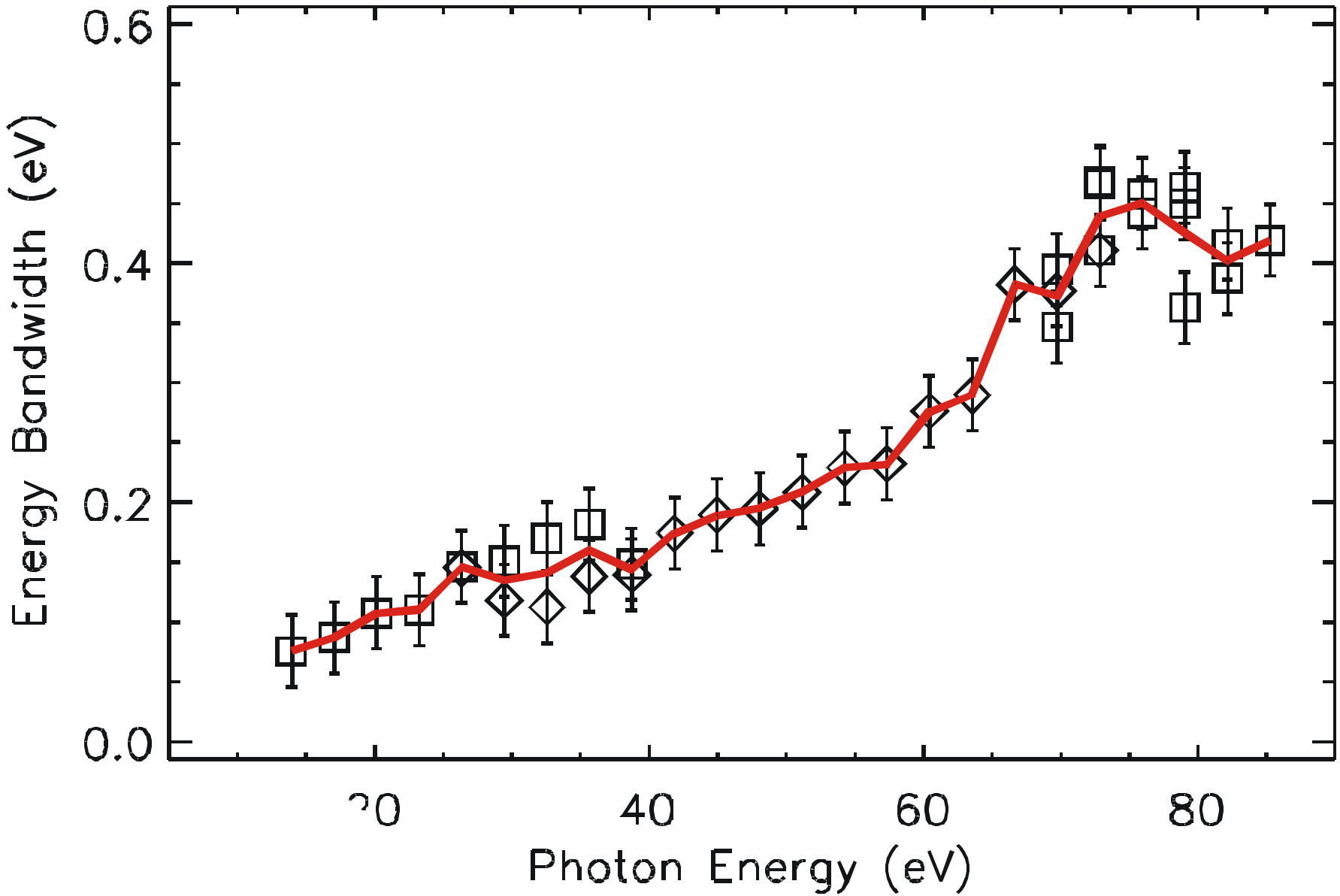
Resonant Core Excitation in Xenon

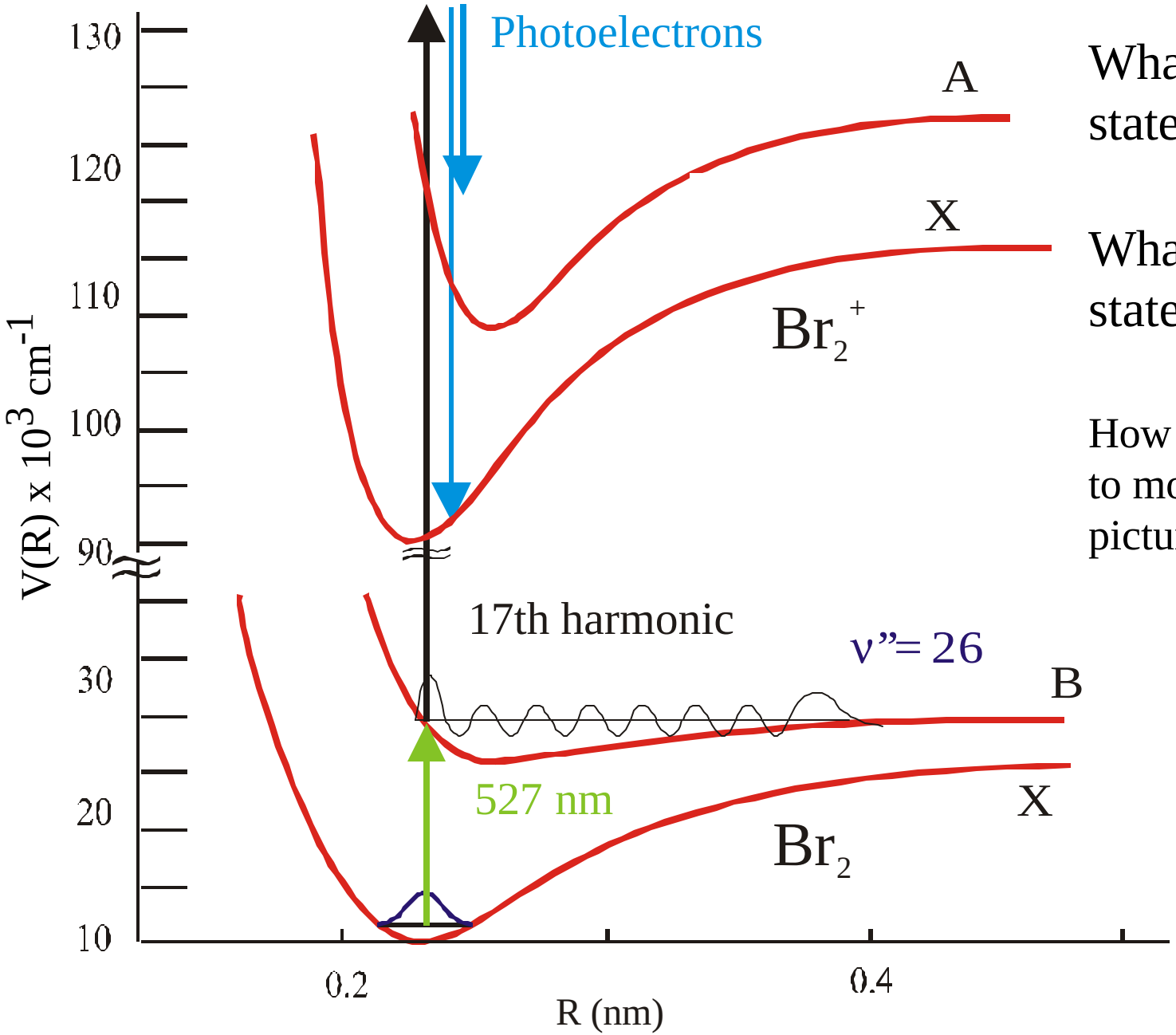


Photoelectron Spectra of NO



Energy Bandwidths of harmonics 7-55



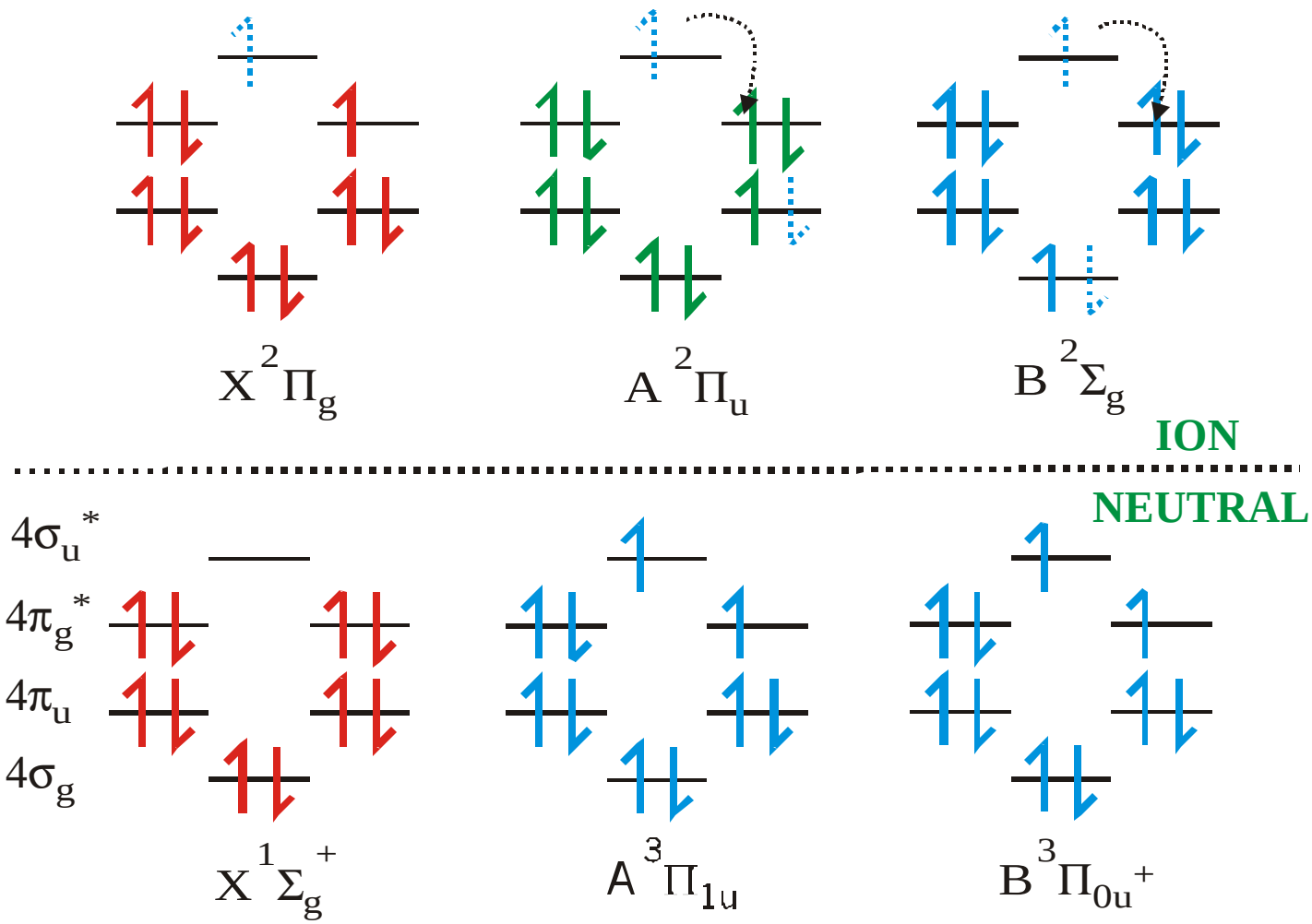


What electronic states?

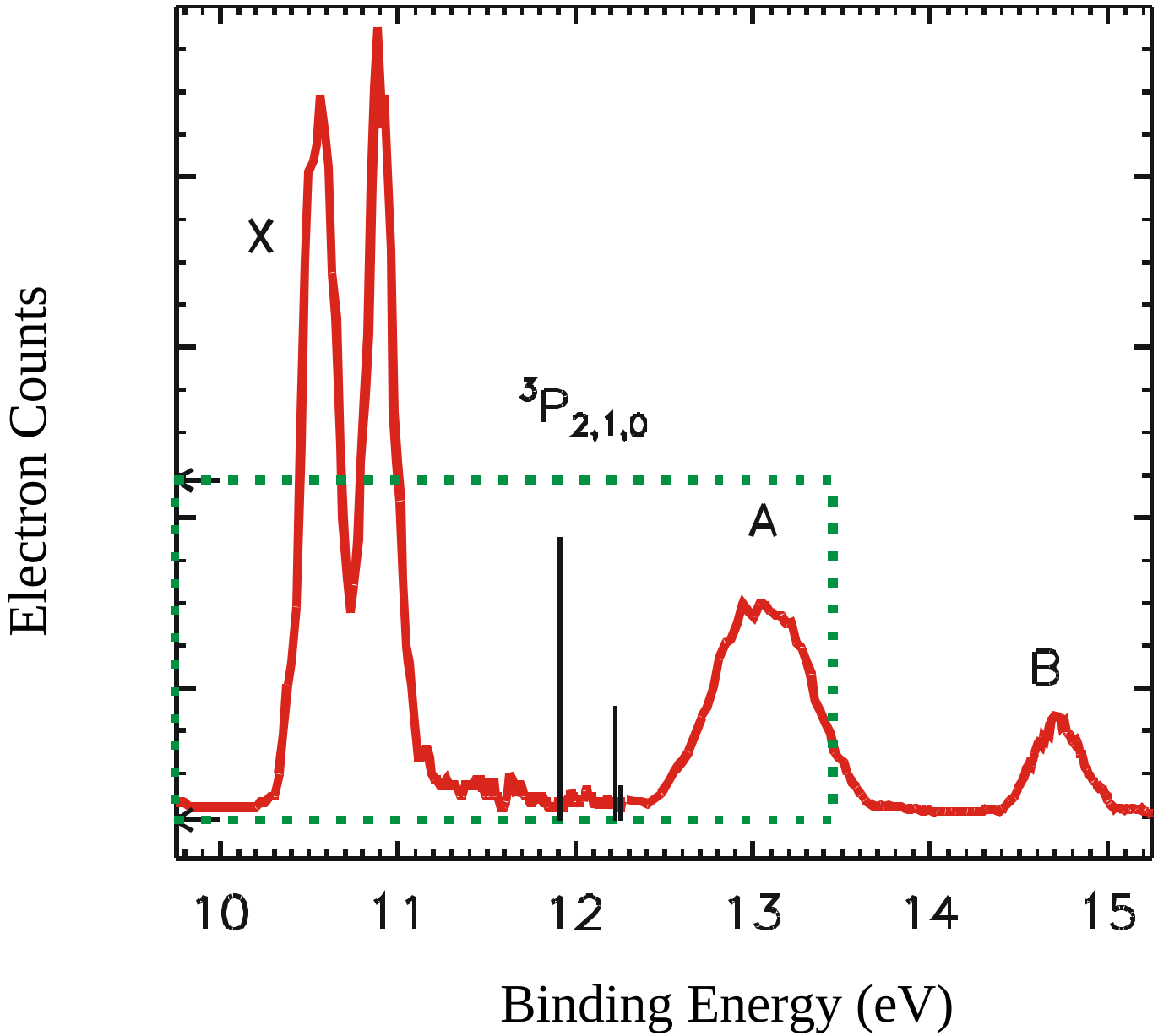
What vibrational states? (overlap)

How does it compare to molecular orbital picture?

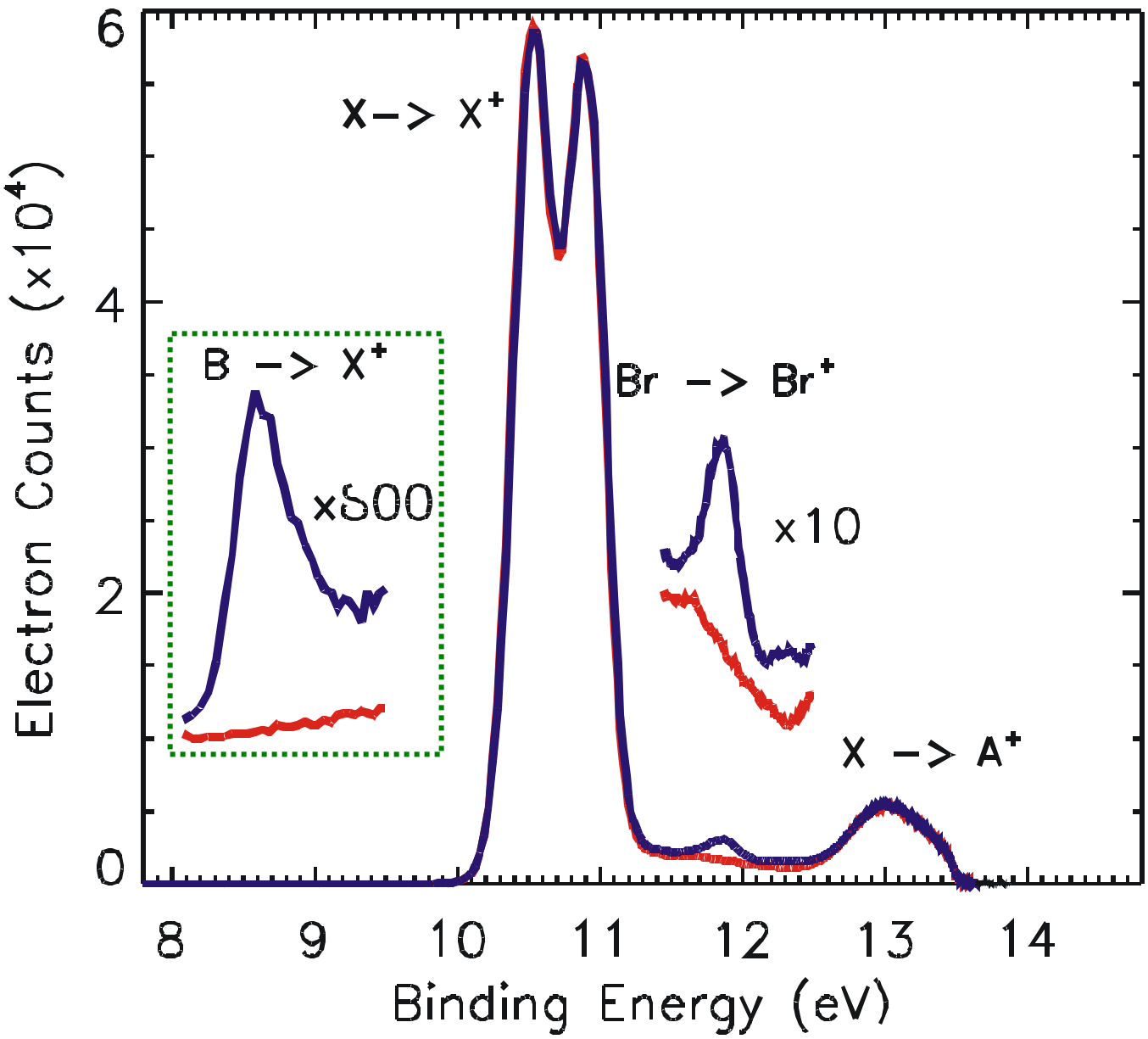
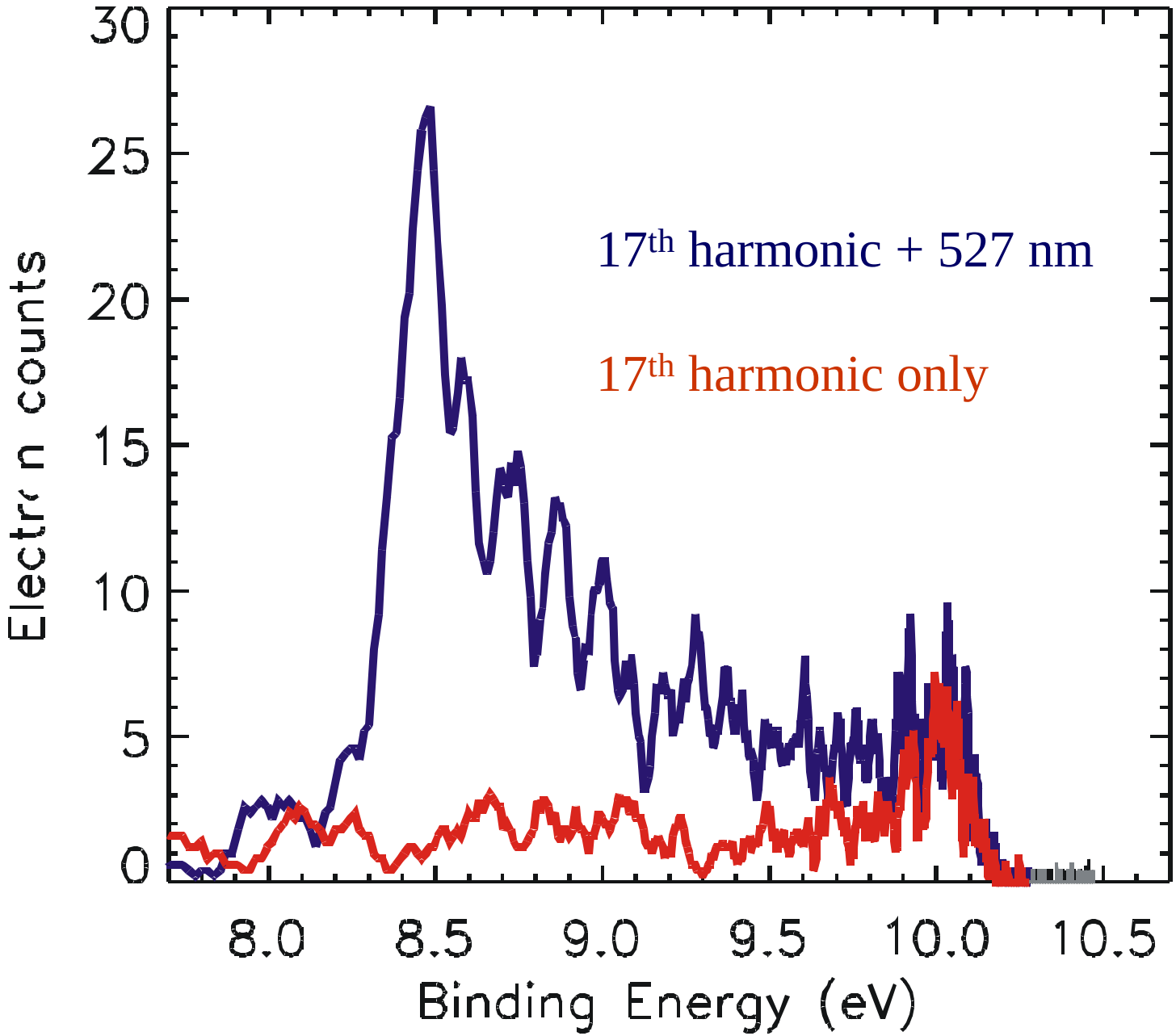
MOLECULAR ORBITALS OF Br_2/Br_2^+



Photoelectron spectrum of Br_2 (17th harmonic)



Photoelectron Spectrum of Br_2 (17th + 527 nm)



Photoelectron spectrum of the Br_2 bound B state