



XIII School on Synchrotron Radiation:
Fundamentals, Methods and Applications
Grado, Italy / 14-25 September 2015



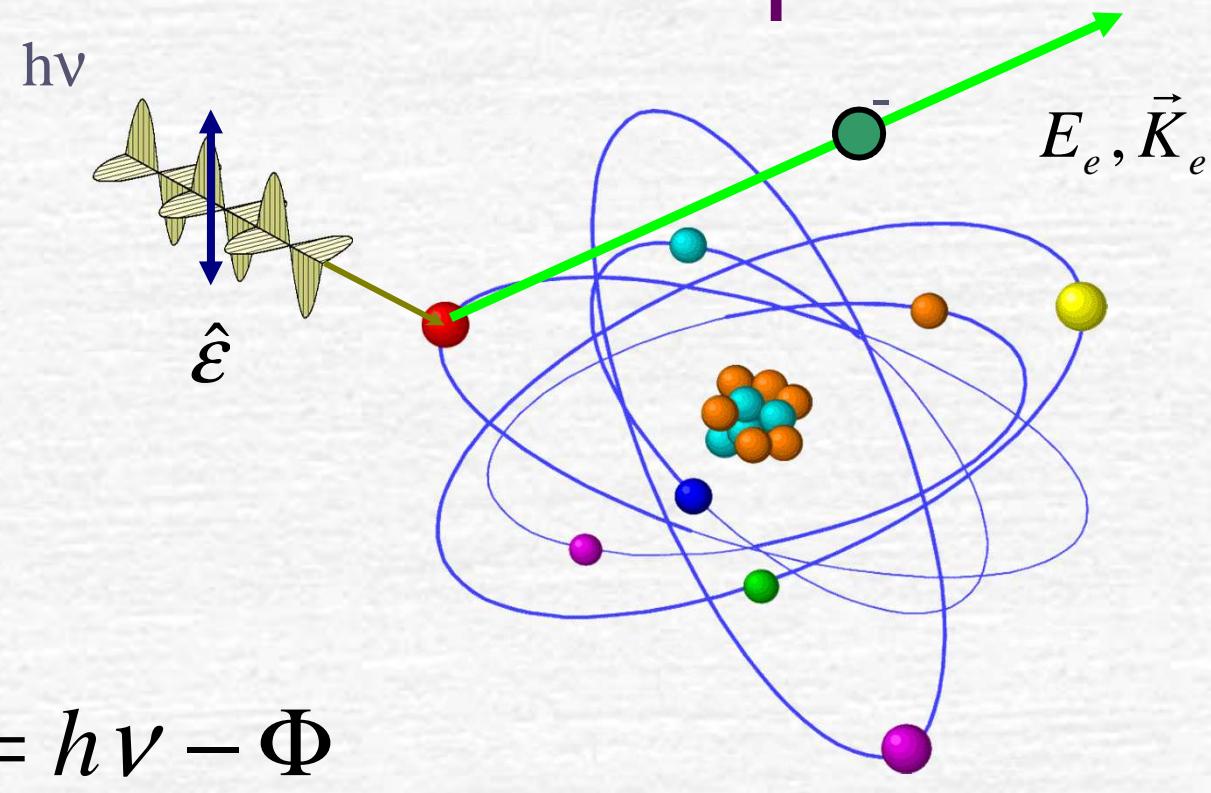
Photoemission Spectroscopy: Fundamental Aspects

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CNISM Unita' di Ricerca di Roma 3



Basic Concepts



$$E_e^{MAX} = h\nu - \Phi$$



outline

- 1. Introduction**
 - 2. Energy conservation, binding energy and photoelectron energy**
 - 3. Satellite structures and multiplet splitting**
 - 4. Chemical shift**
 - 5. Molecular photoelectron spectra**
 - 6. Photoelectron angular distributions**
 - 7. Hole state relaxation**
 - 8. Resonant photoemission**
 - 9. Photoemission in solids**
 - 10. EDC and core ionization**
 - 11. Angular resolved PES**
- 

Photoemission Schematics:

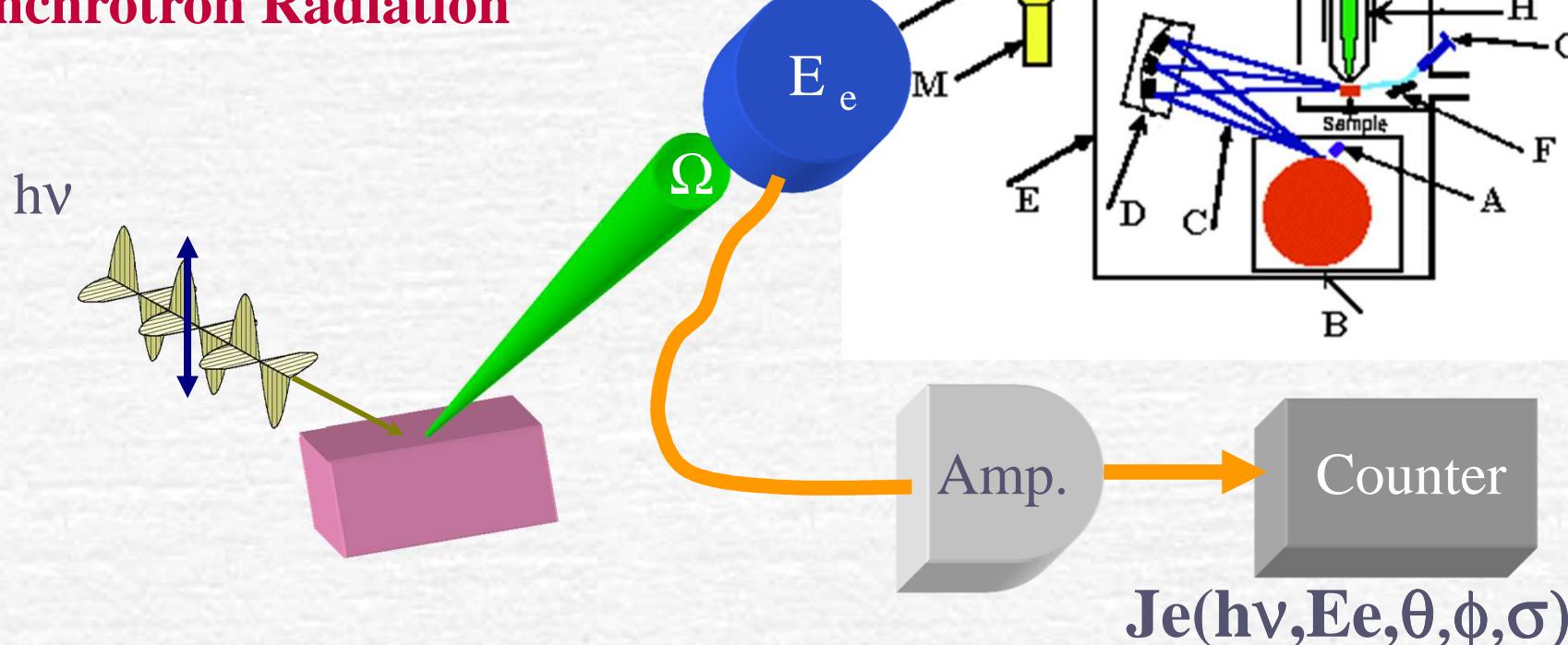
$\text{He I}\alpha=21.23\text{eV}$

$\text{He II}\alpha=40.82\text{eV}$

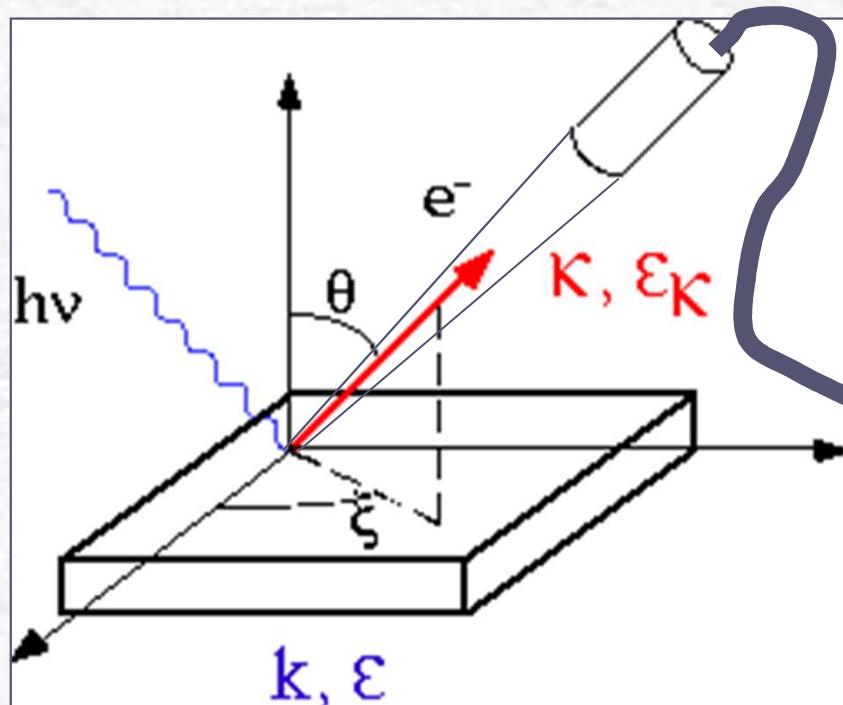
$\text{Mg K}\alpha_{1,2} = 1253,6 \text{ eV}$

$\text{Al K}\alpha_{1,2}=1486,6\text{eV}$

Synchrotron Radiation



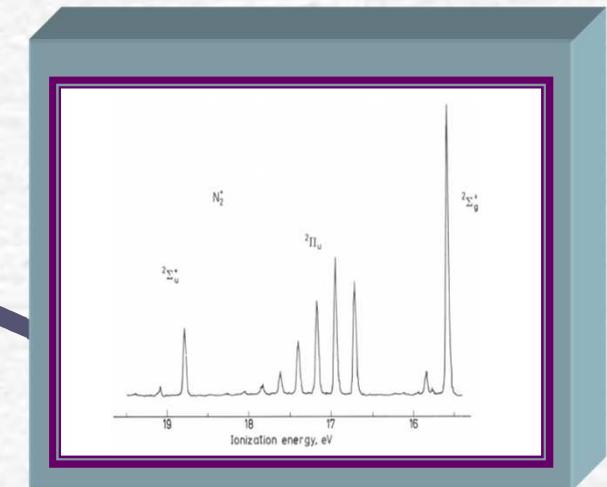
ENERGY CONSERVATION, BINDING ENERGY AND PHOTOELECTRON ENERGY



$$E_{\text{kin}} = \hbar\omega - \phi - |E_b|$$

$$E_{\text{kin}} = \frac{p^2}{2m}$$

$$p = \sqrt{2mE_{\text{kin}}}$$



X-section vs. Photoemission current

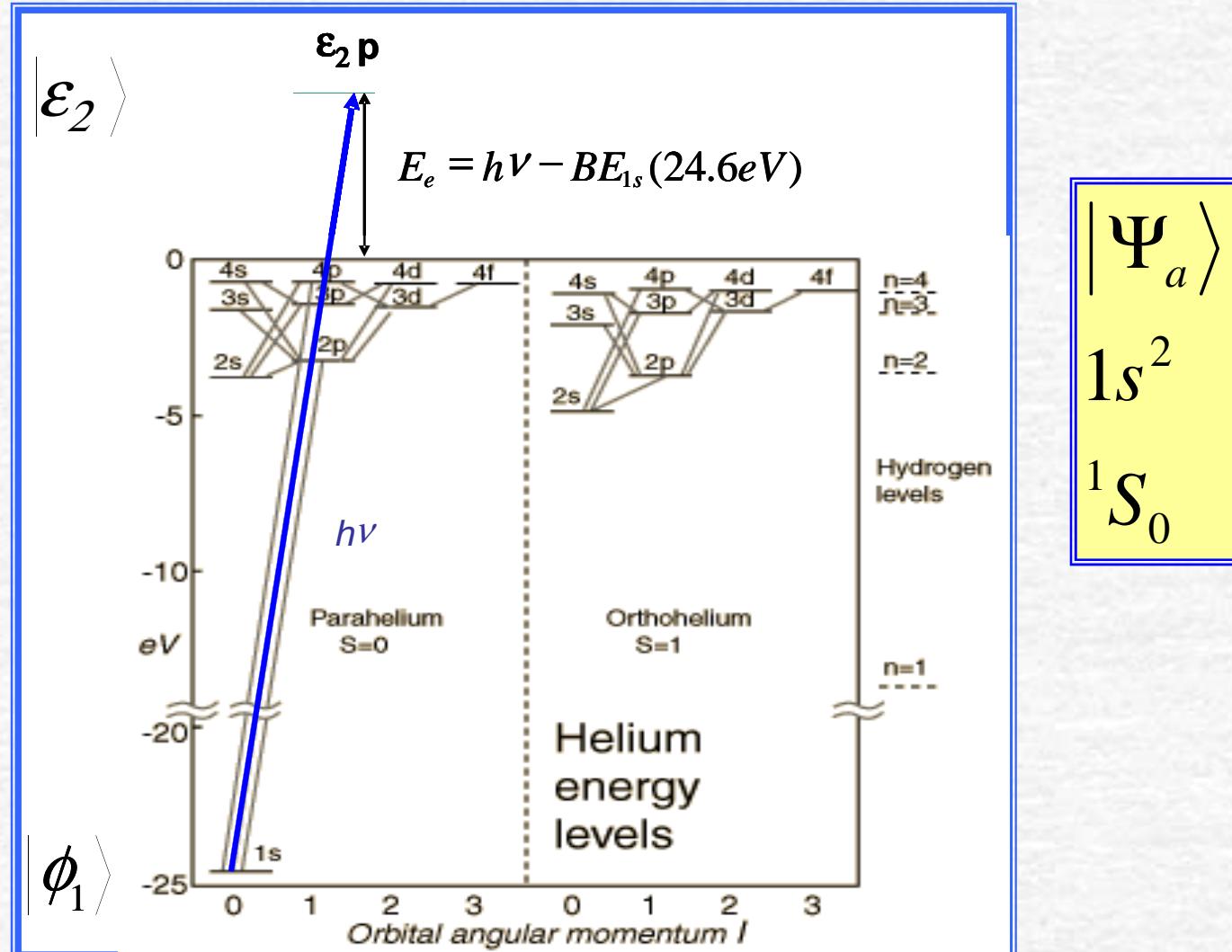
$$J_e(h\nu, \vartheta, \phi) = J_{h\nu}(\rho l) \int \int \frac{d\sigma}{d\Omega dE} F_{an}(E, \Omega) \eta_{\text{det}}(E) d\Omega dE$$

Photoemission peak lineshape

- | | |
|-------------------------------------------------|-----------|
| 1. Photon monochromaticity | Gaussian |
| 2. Electron analyzer resolution | Gaussian |
| 3. Final state lifetime (uncertainty principle) | Lorentian |

Lineshape = Convolution (1,2,3)

The photoemission process



Interaction radiation matter

$$\frac{d\sigma}{dh\nu} = 4\pi^2 \alpha h\nu \sum_B \left| \hat{\epsilon} \bullet \left\langle \Psi_B \left| \sum_i \vec{r}_i \right| \Psi_A \right\rangle \right|^2 \delta(E_B - E_A - h\nu)$$

Bertoni's lecture this school

Initial state A = Neutral ground(excited) state
Final state B = Residual ion + free electron(s)

Energy balance for 2e atom

$$E_B = E_A + h\nu$$

$$\Psi_A = \hat{A}\phi_1\phi_2$$

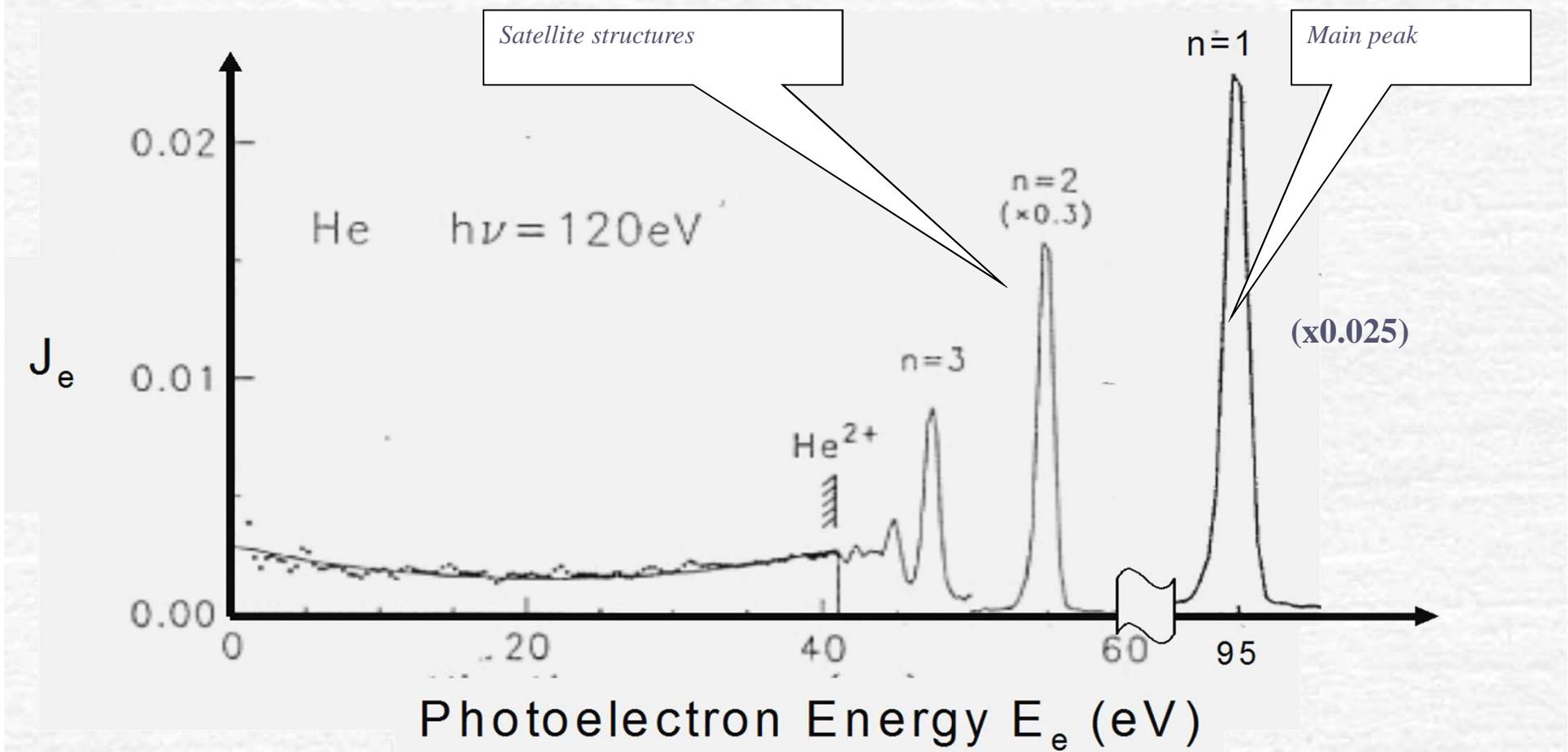
$$\Psi_B = \hat{A}\phi_1\epsilon_2$$

$$\cancel{E_{1s}} + E_e = \cancel{E_{1s}} + E_{1s} + h\nu$$

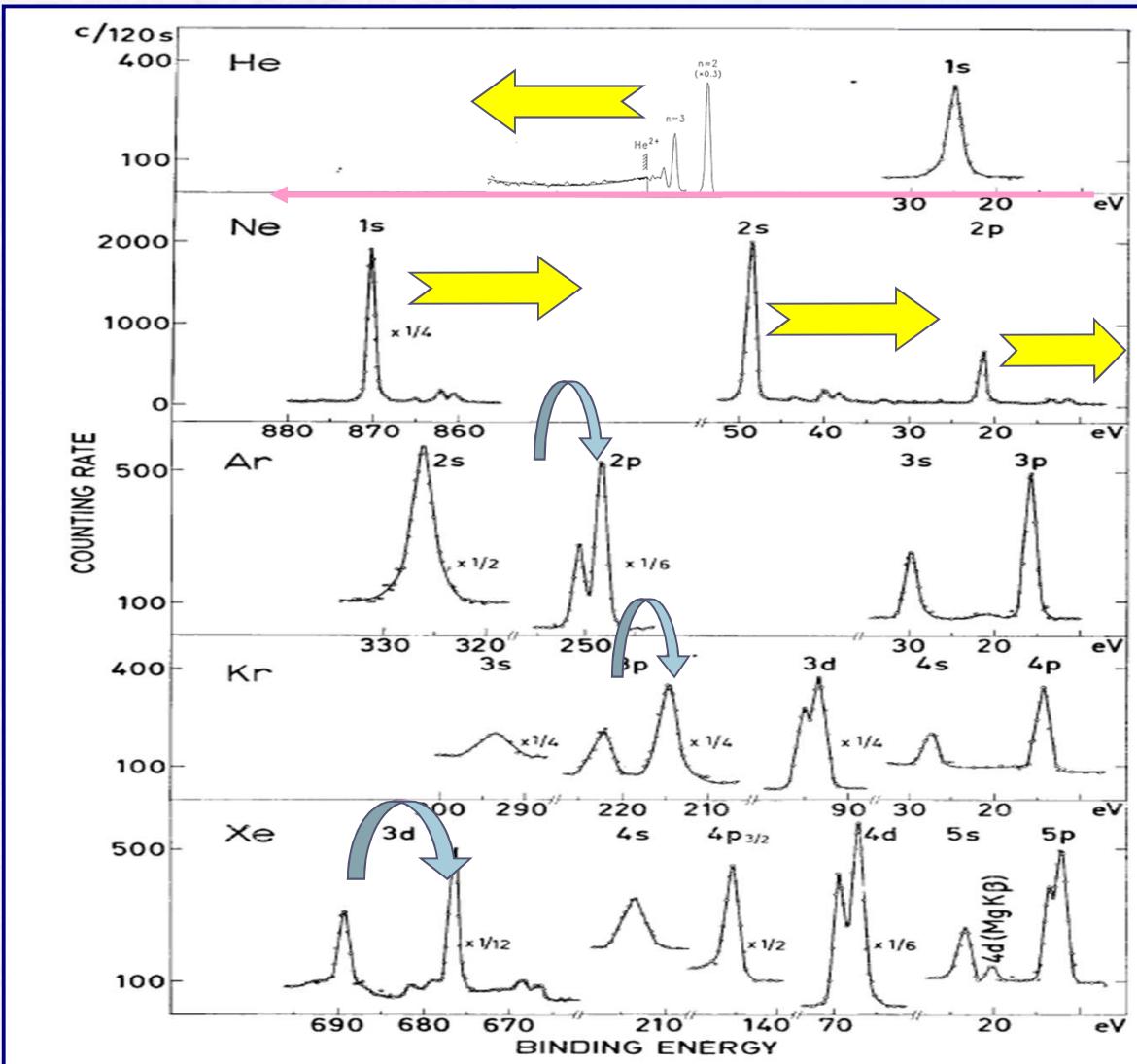
$$E_e = h\nu - BE_{1s} (24.6eV)$$

One single photoemission peak is expected
Energy and momentum are conserved

Complexity of the photoemission spectrum



The noble gas panorama



He $1s^2$

Ne $1s^2 2s^2 2p^6$

Ar $1s^2 2s^2 2p^6 3s^2 3p^6$

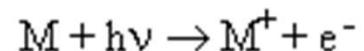
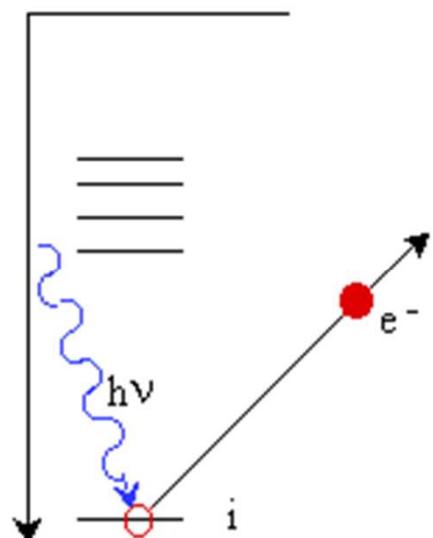
Kr $1s^2 \dots 3s^2 3p^6 3d^{10} 4s^2 4p^6$

Xe $\dots 3d^{10} 4s^2 4p^6 4d^{10} 5s^2 5p^6$

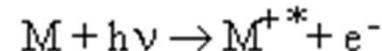
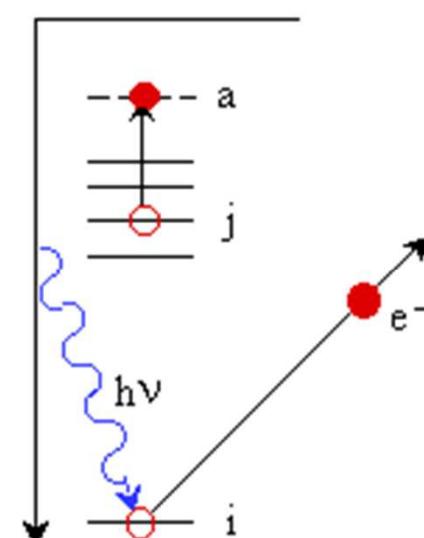
Primary photoionization process

PRIMARY PHOTOIONIZATION PROCESSES

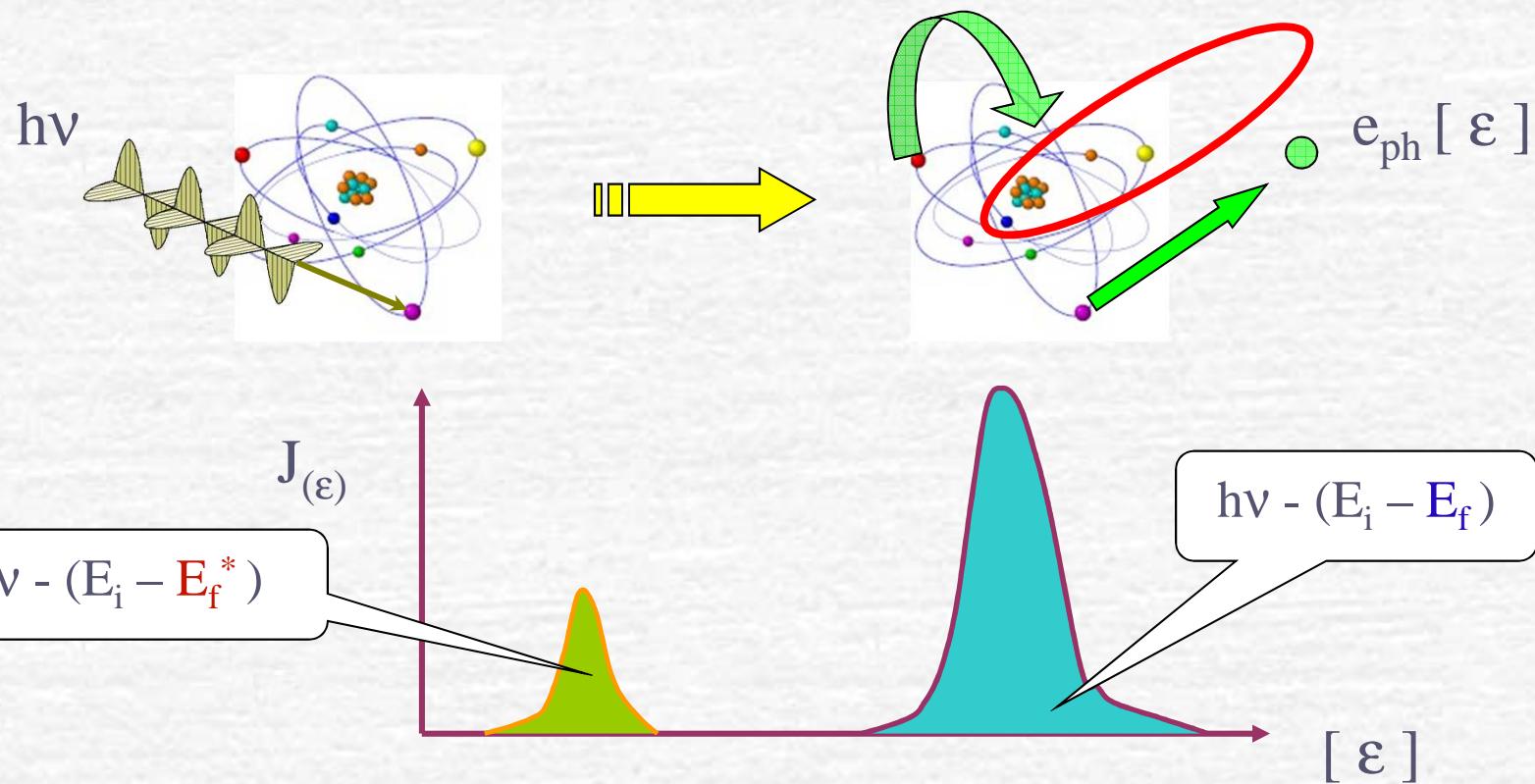
MAIN PROCESS



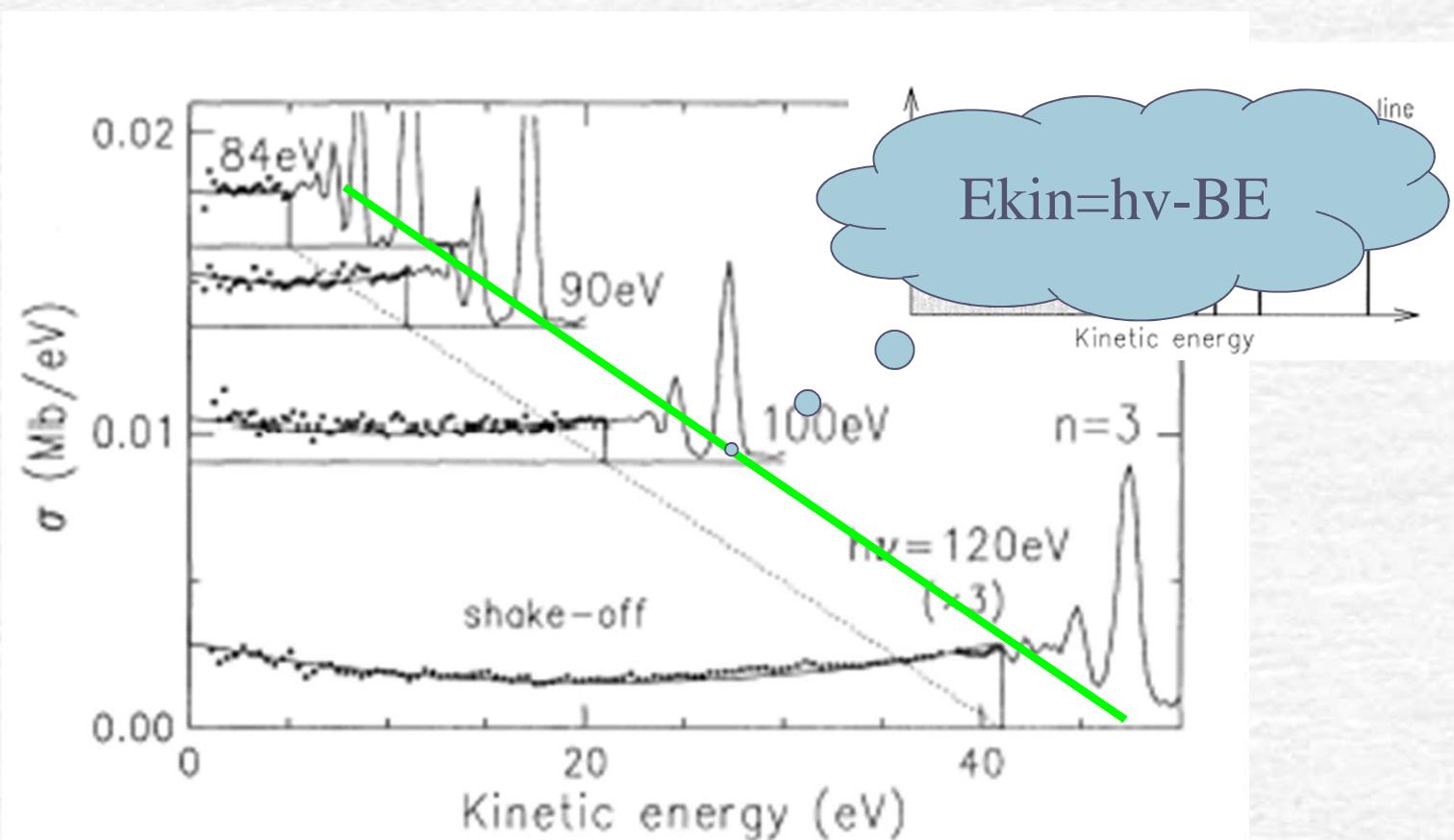
SATELLITE PROCESS



- Photon = single particle operator
- 2 or more particles involved in final state = e-e correlation
- Relaxation & e-e correlation in photoemission = satellite



The He satellite structure



A many electron atom

$$H_0 \left| \Psi_A^{(N)} \right\rangle = E_A^{(N)} \left| \Psi_A^{(N)} \right\rangle$$

$$\begin{aligned} H_0 &= H_0(kin) + H_0(e-n) + H_0(e-e) + H_0(s-o) = \\ &= \sum_1^N \frac{p_i^2}{2m} + \sum_1^N -\frac{Ze^2}{r_i} + \sum_{i>j}^N \frac{e^2}{r_{ij}} + \sum_1^N \zeta(r_j) \vec{l}_i \bullet \vec{s}_i \end{aligned}$$

$$\left| \Psi_A^{(N)} \right\rangle = \hat{A}(\phi_j(\vec{r}_i, \sigma_i); \Psi_R^{(N-1)})$$

Single
particle
orbital

$$H'_0 \left| \Psi_B^{(N)} \right\rangle = E_B^{(N)} \left| \Psi_B^{(N)} \right\rangle$$

sudden approximation

$$\left| \Psi_B^{(N)} \right\rangle = \hat{A}(\mathcal{E}_l; \left| \Psi_B^{(N-1)} \right\rangle)$$

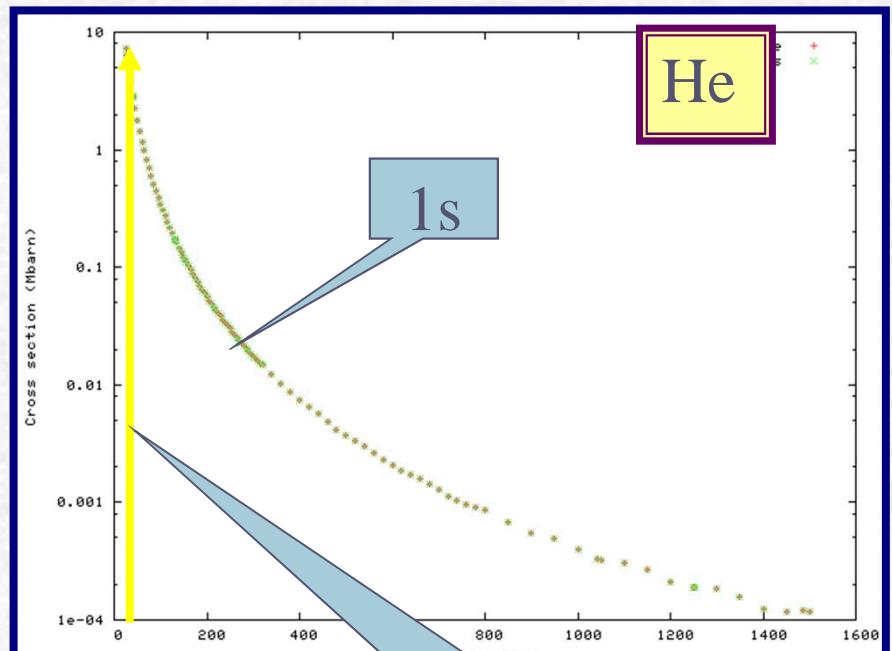
$$\frac{d\sigma}{d\Omega dE_e} \propto \frac{1}{h\nu} \sum_{A,B} \left| \hat{\epsilon} \bullet \langle \mathcal{E}_l | \vec{r}_j | \phi_j(\vec{r}_j, \sigma_j) \rangle \langle \Psi_B^{(N-1)} | \Psi_R^{(N-1)} \rangle \right|^2 \delta(E_e + E_B^{(N-1)} - E_A - h\nu)$$

frozen core approximation

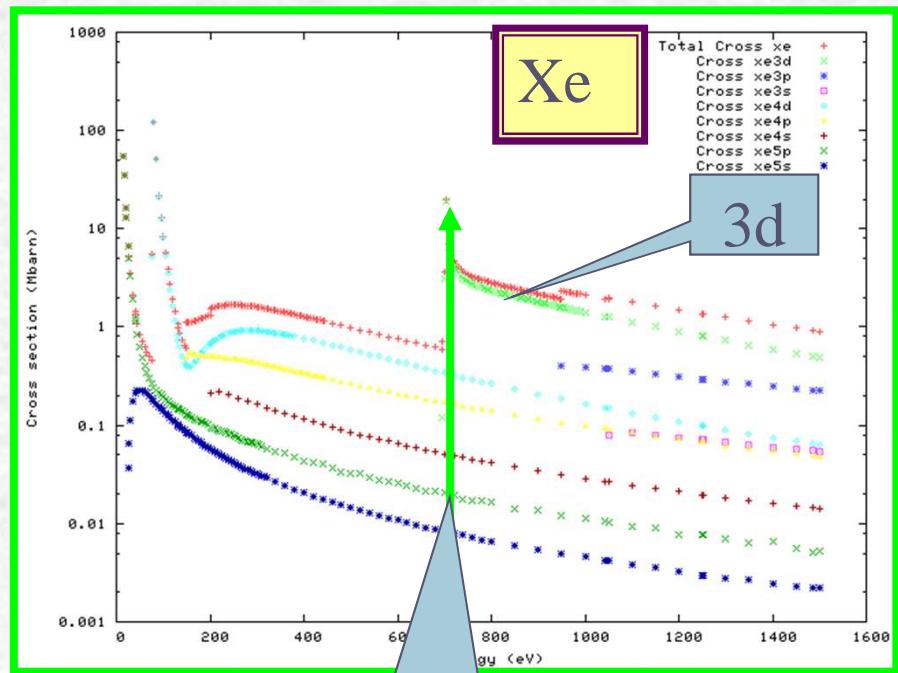
$$H_0' = H_0$$

$$\frac{d\sigma}{d\Omega dE_e} \propto \frac{1}{h\nu} \sum_{A,B} \left| \hat{\epsilon} \bullet \langle \mathcal{E}_l | \vec{r}_j | \phi_j(\vec{r}_j, \sigma_j) \rangle \right|^2 \delta(E_e + \varepsilon_j - h\nu)$$

Total photoemission cross section

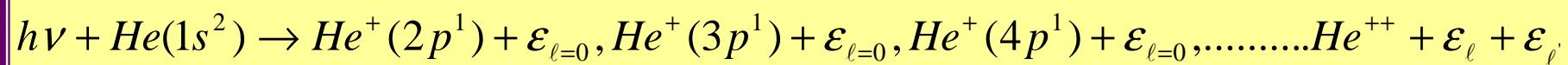
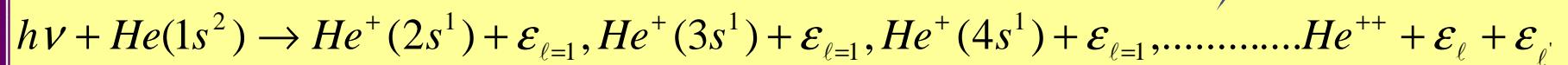
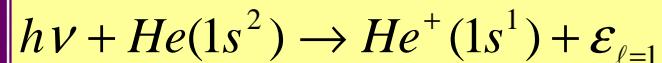
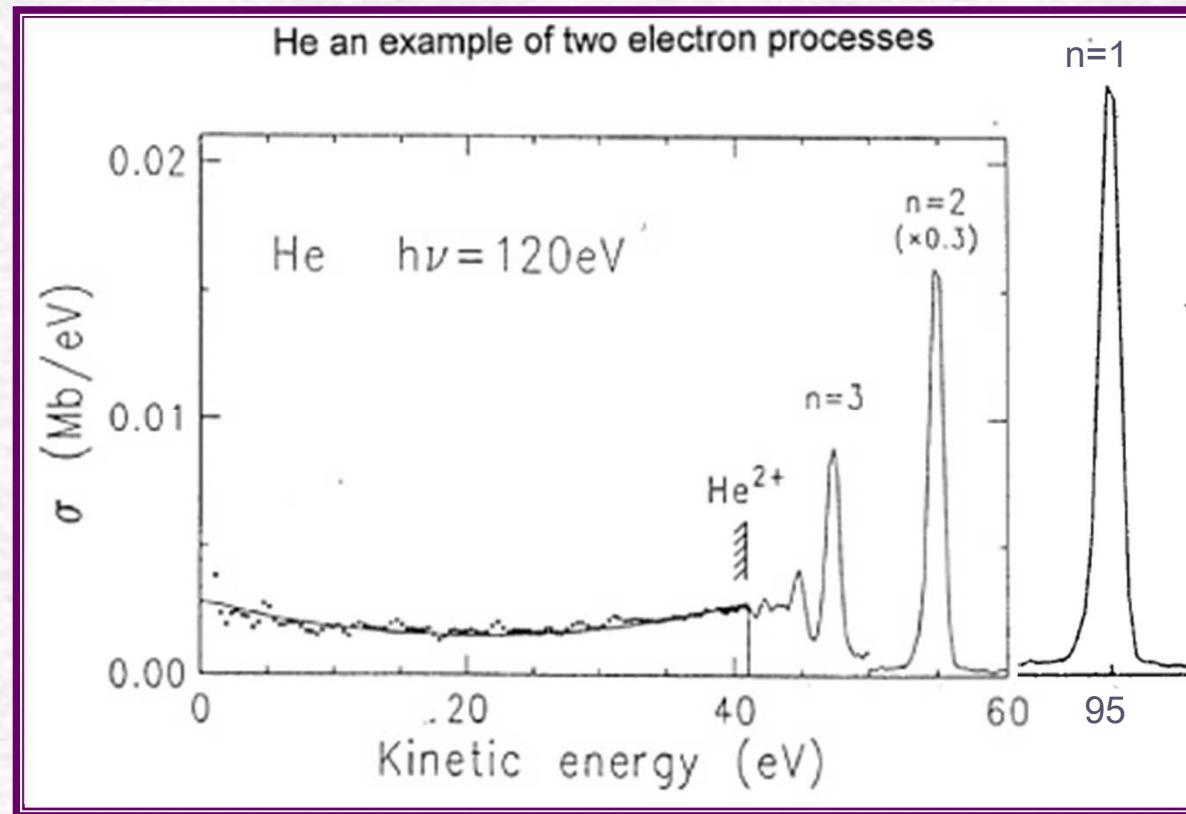


1s Threshold



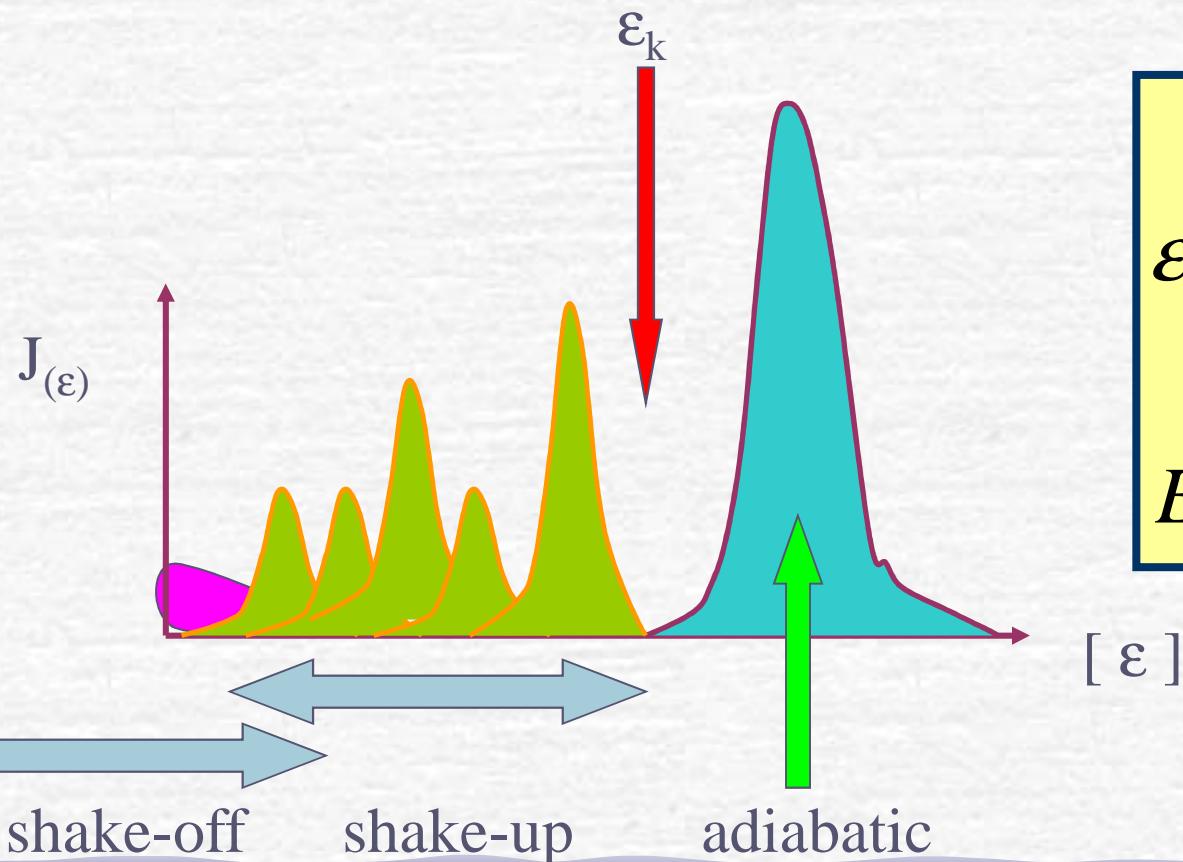
3d Threshold

Photoelectron current vs. photoelectron energy



Koopmans energy vs. photoemission peaks

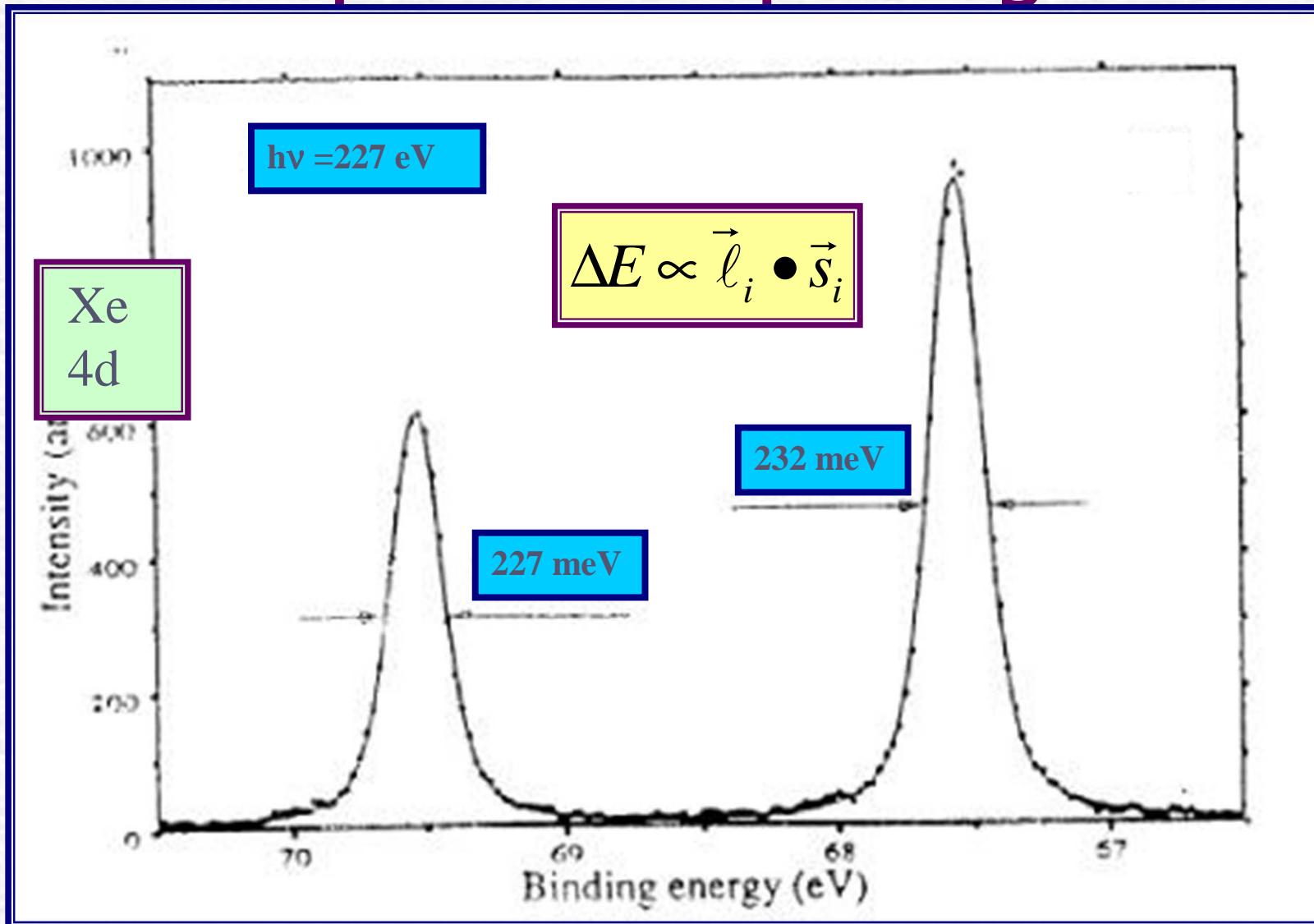
$$\frac{d\sigma}{d\Omega dE_e} \propto \frac{1}{h\nu} \sum_{A,B} \left| \hat{\varepsilon} \bullet \left\langle \varepsilon_l \left| \vec{r}_j \right| \phi_j(\vec{r}_j, \sigma_j) \right\rangle \left\langle \Psi_B^{(N-1)} \left| \Psi_R^{(N-1)} \right. \right\rangle \right|^2 \delta(E_e + E_B^{(N-1)} - E_A - h\nu)$$



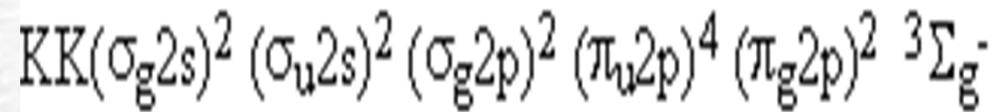
$$\varepsilon_k = \frac{\sum_i \varepsilon_i I_i}{\sum_i I_i}$$

$$E_{relax} = \varepsilon_{adiab} - \varepsilon_k$$

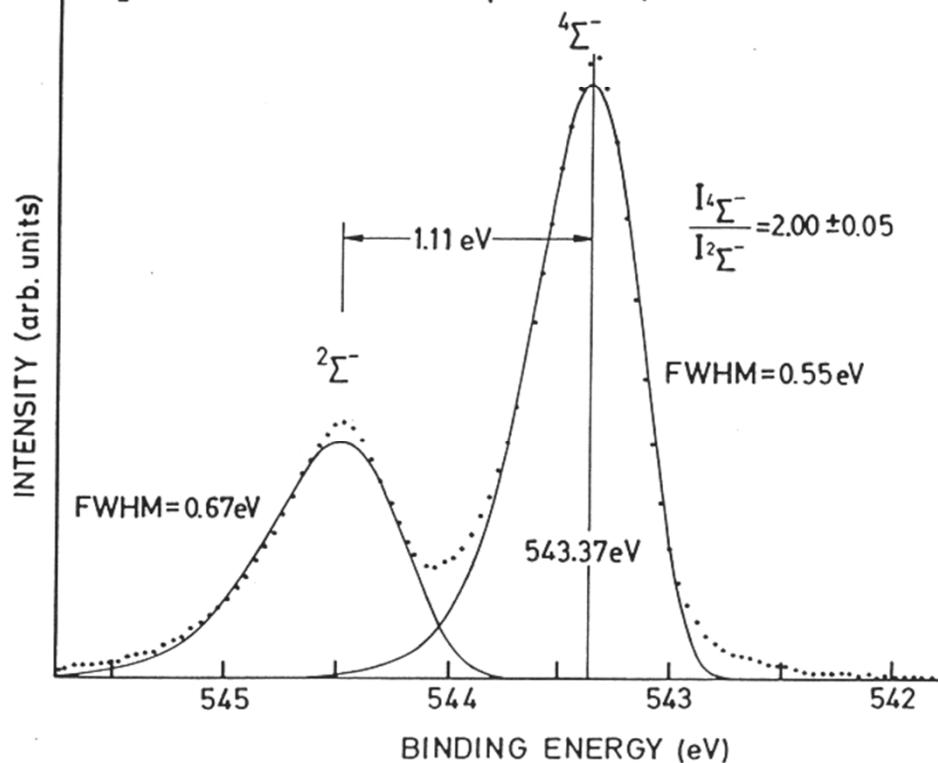
Spin orbit splitting



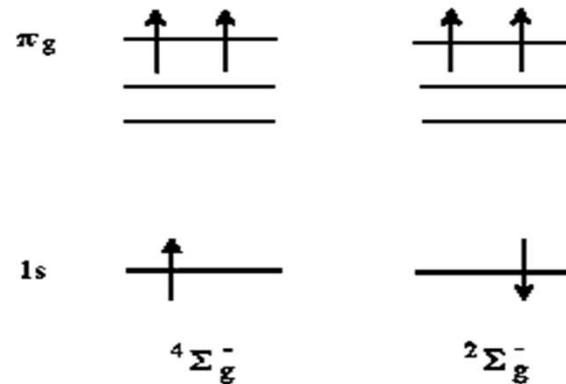
Molecular multiplet splitting O₂



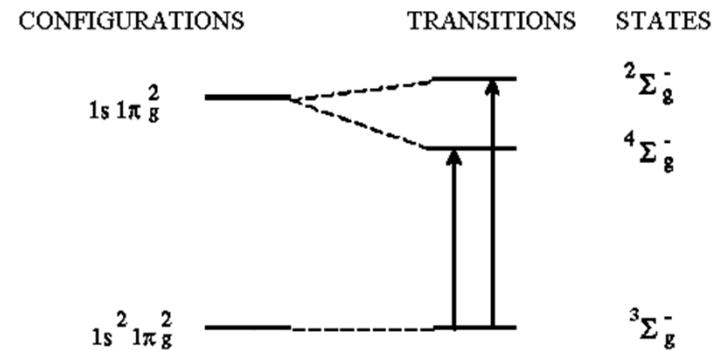
O₂ 01s Photoelectron spectrum ($h\nu = 1487 \text{ eV}$)



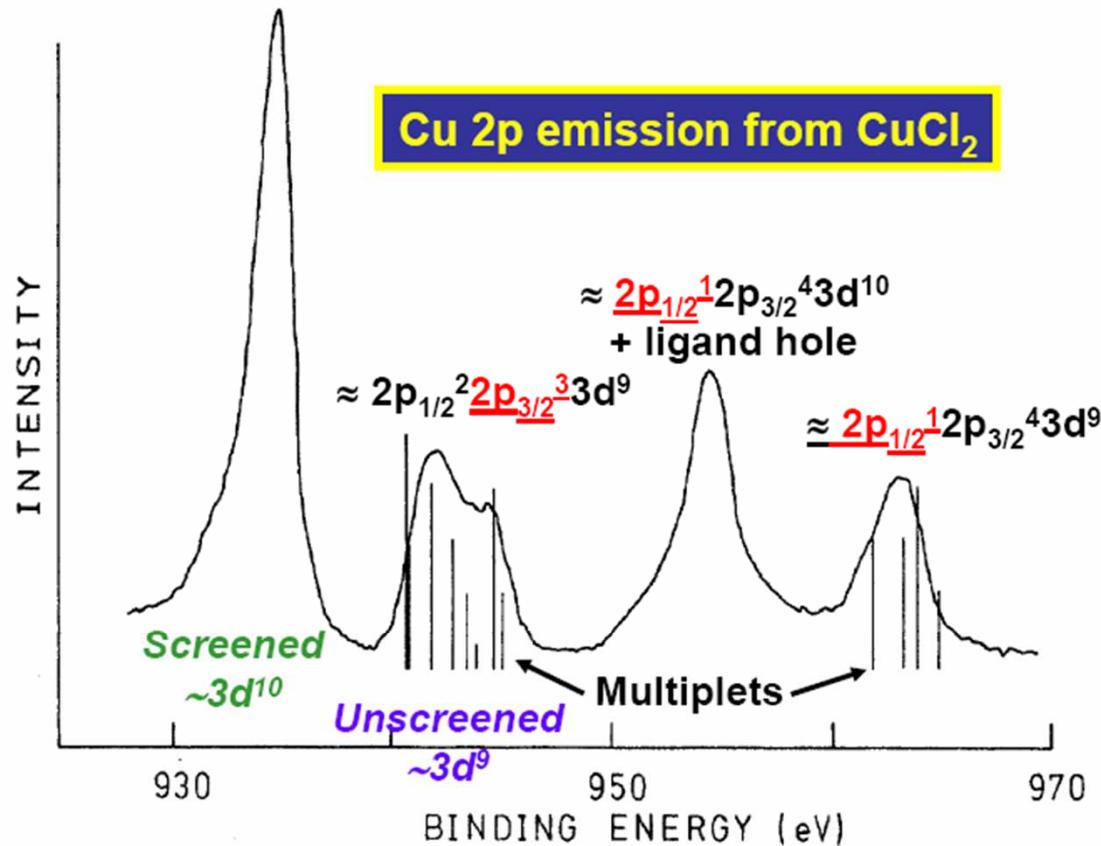
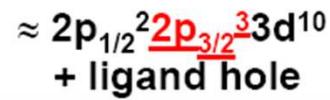
CORE HOLE MULTIPLET STATES OF O₂⁺



MULTIPLER TRANSITIONS IN O₂



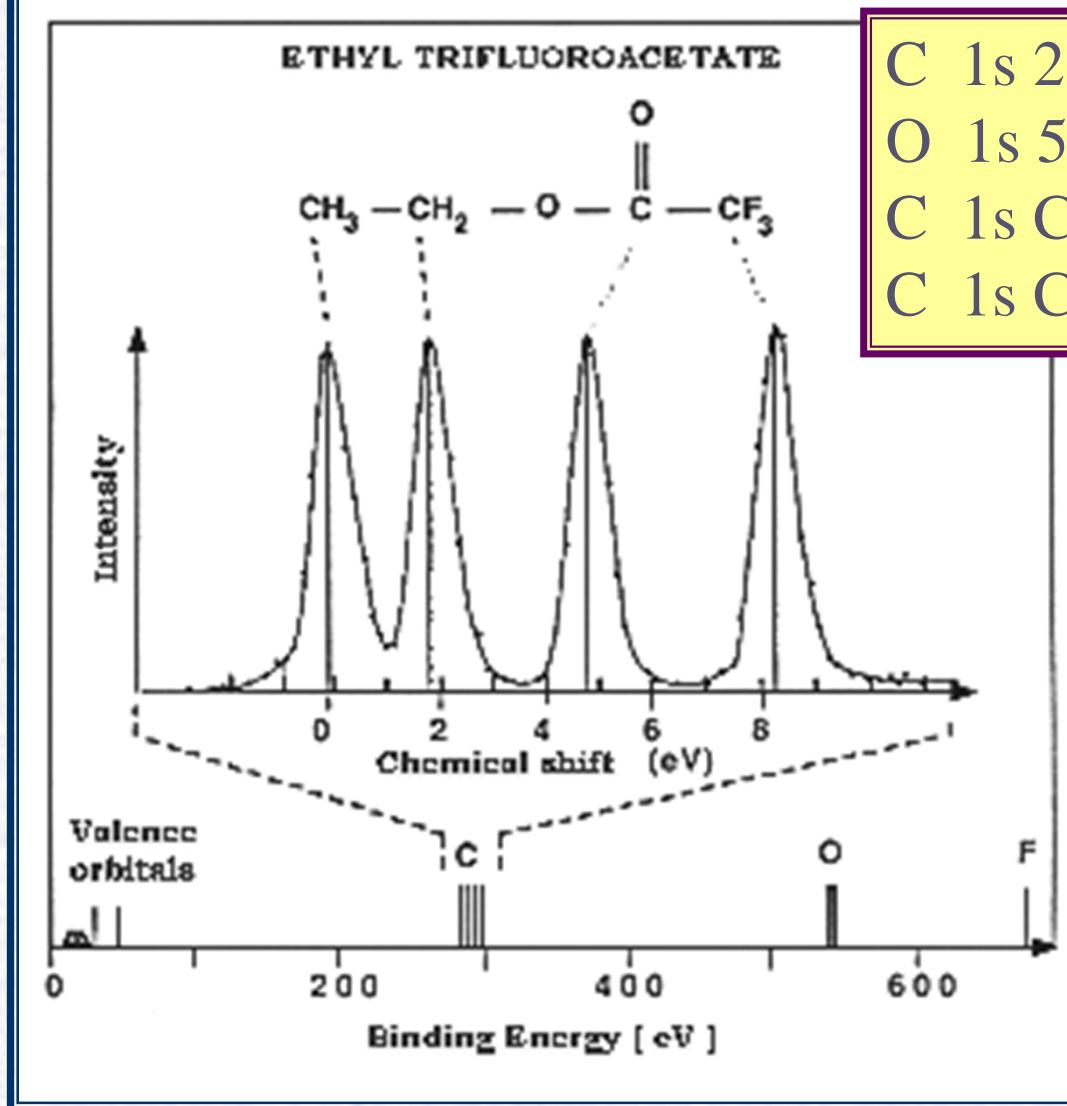
CuCl₂ multiplet & satellite



$$\Psi_{final,K}(N-1) = C_{1,K}(2p_{1/2}^2 2p_{3/2}^3 3d^{10} + \text{Cl hole}) + C_{2,K}(2p_{1/2}^2 2p_{3/2}^3 3d^9)$$

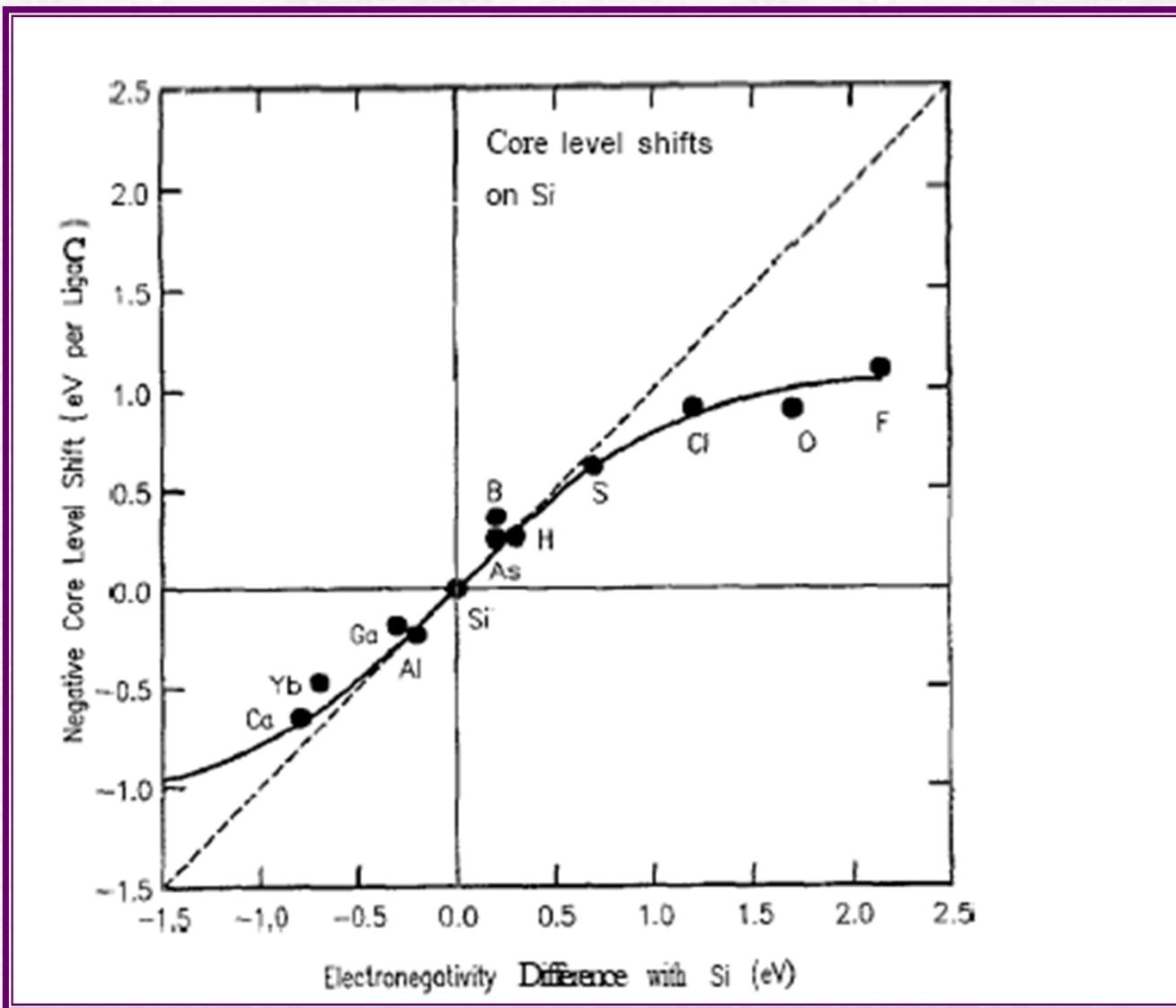
Van der Laan et al., Phys. Rev. B 23 (1981) 4369

Chemical shift

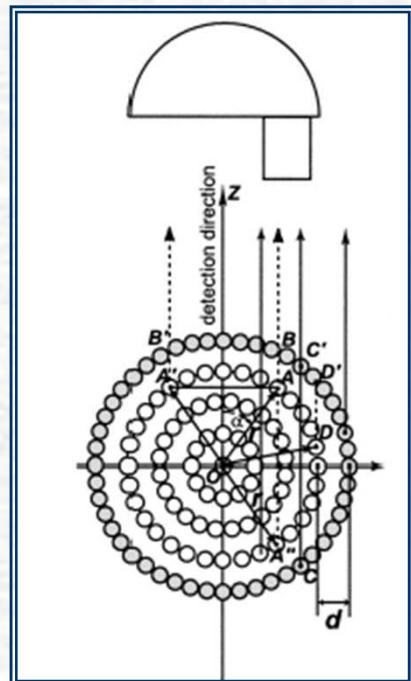


C 1s 285-300 eV
O 1s 530-540 eV
C 1s CO₂ 298 eV
C 1s CH₄ 291 eV

Chemical shift vs.electronegativity



Sensitivity to the local environment in free clusters



J. Chem. Phys.,
Vol. 120, No. 1, 1
January 2004

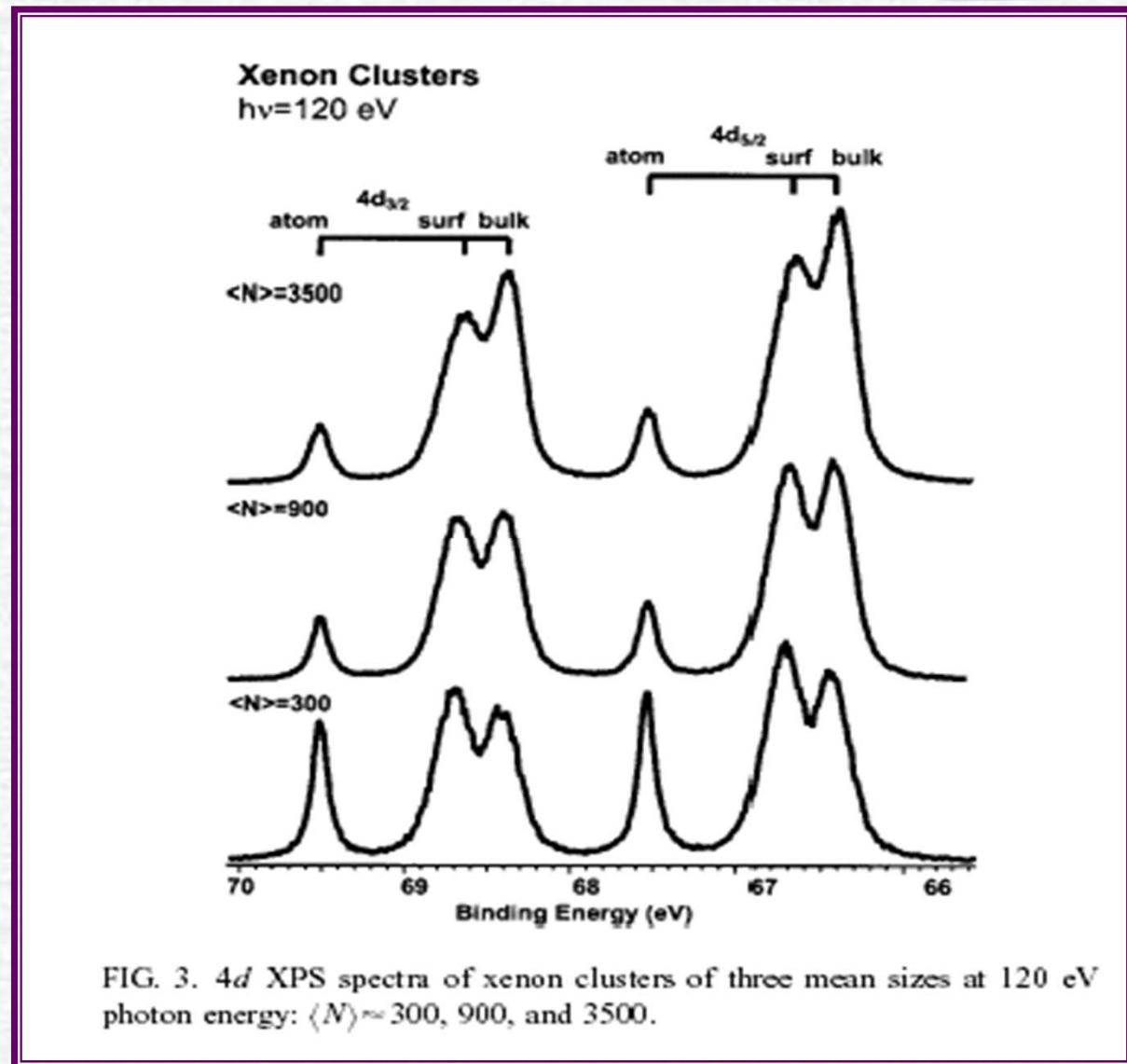
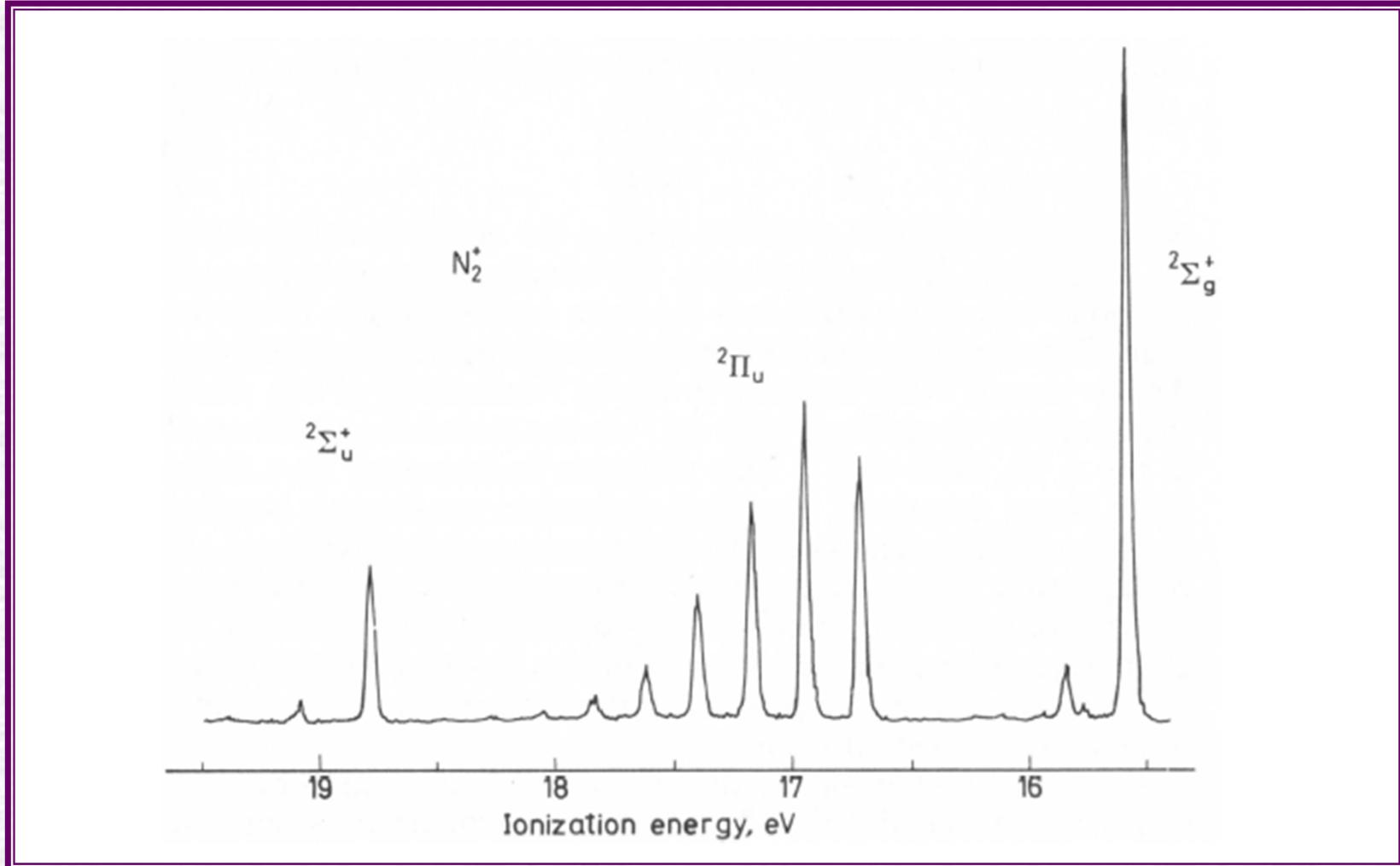
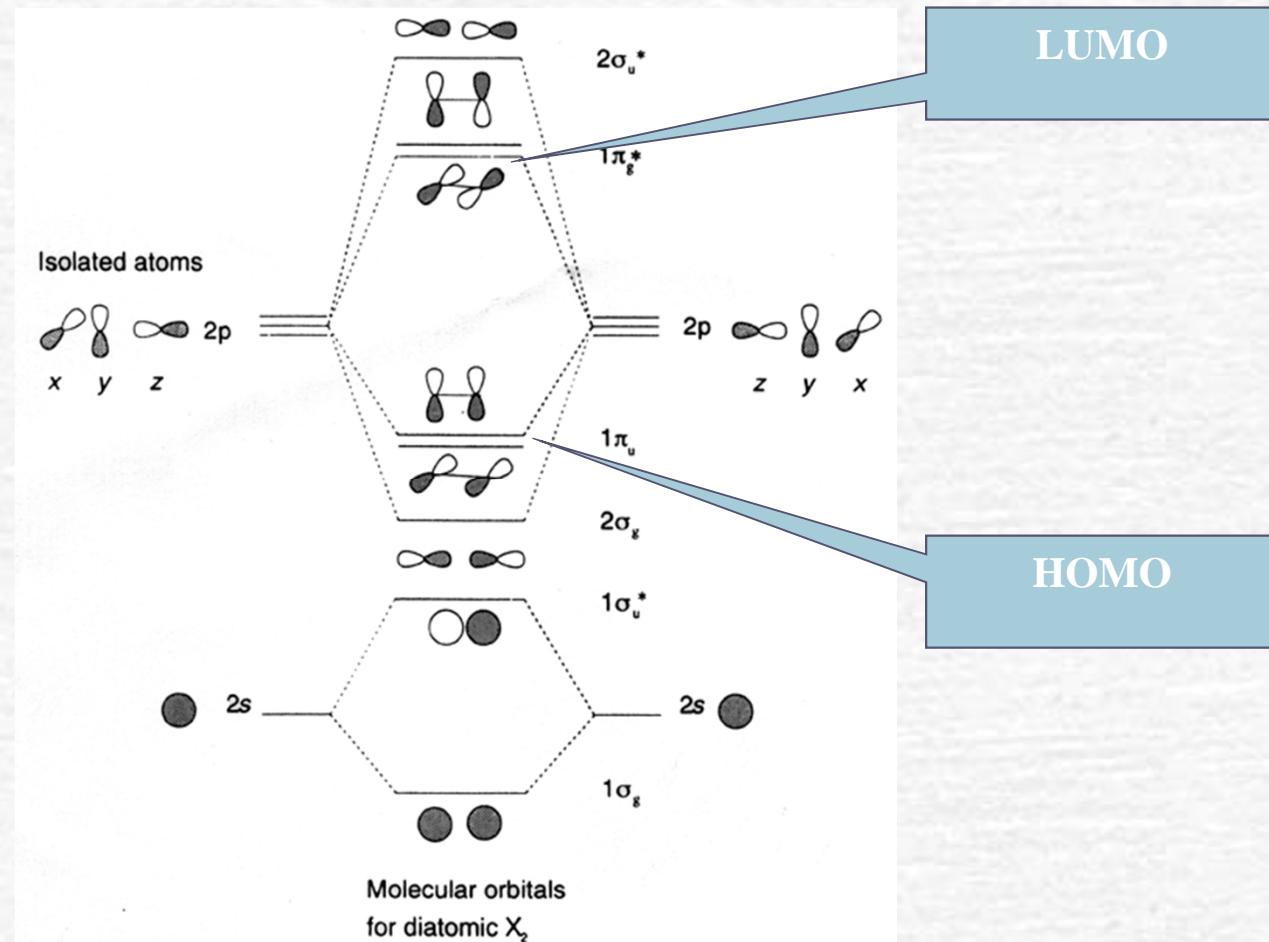


FIG. 3. $4d$ XPS spectra of xenon clusters of three mean sizes at 120 eV photon energy: $\langle N \rangle \approx 300, 900$, and 3500.

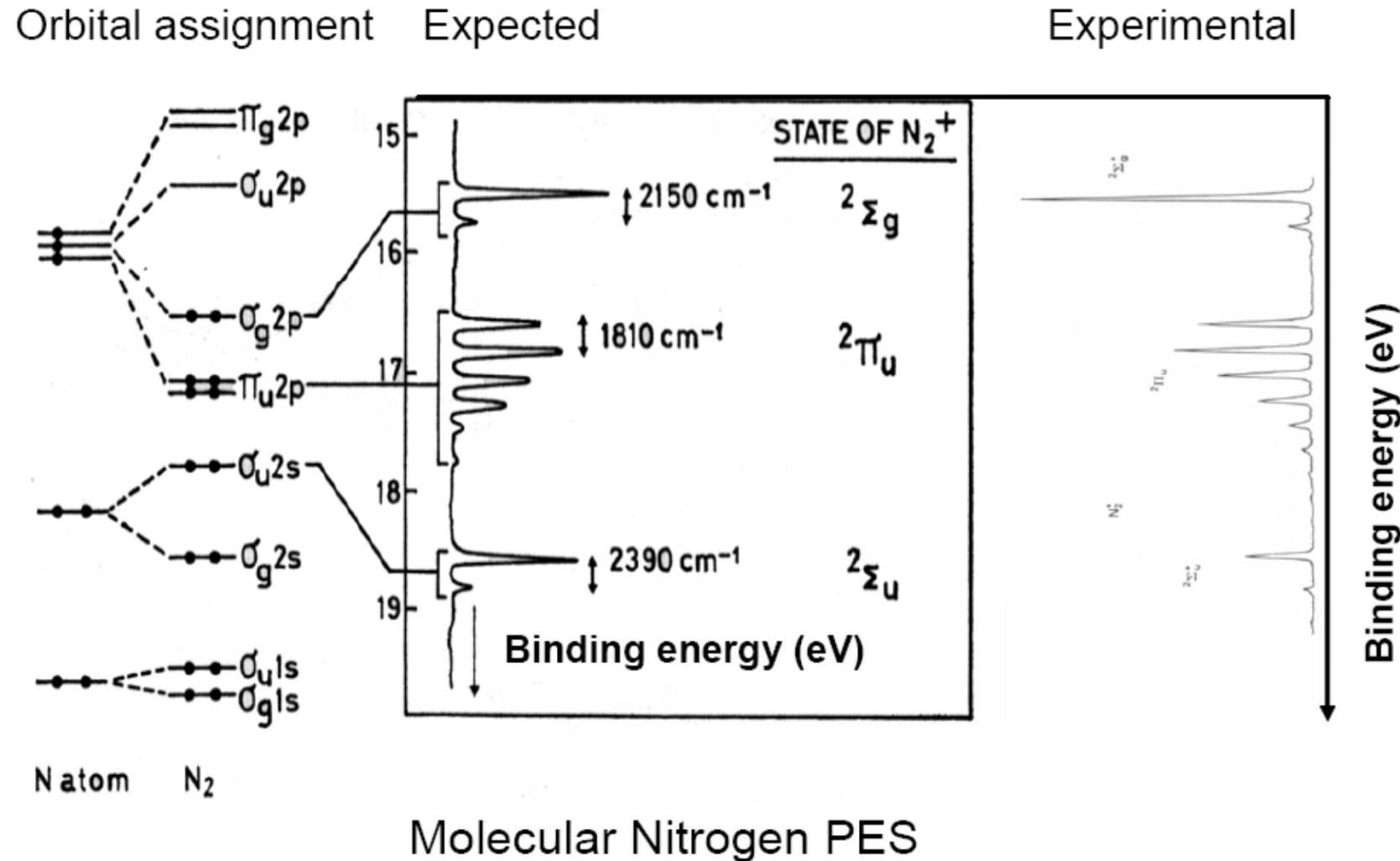
PES spectrum of N₂



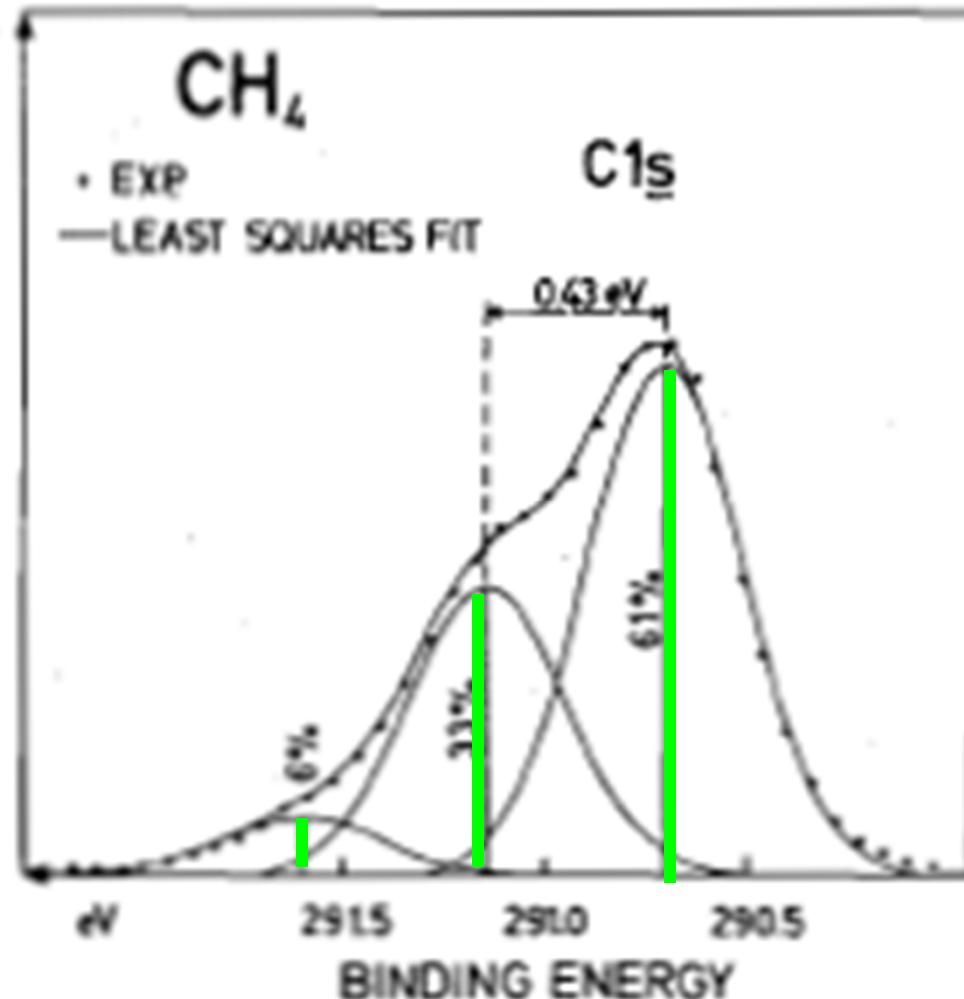
Diatomeric molecule e levels



Experimental PE spectrum of N₂, and MOs



Core PE vibrational spectrum



K. Siegbahn et al "ESCA Applied to free molecules"

Molecular PE x-section

$$\begin{aligned}
 H_0 &= H_0(kin) + H_0(e-n) + H_0(e-e) + H_0(s-o) + H_0(n-n) = \\
 &= \sum_1^N \frac{p_i^2}{2m} + \sum_1^N -\frac{Ze^2}{r_i} + \sum_{i>j}^N \frac{e^2}{r_{ij}} + \sum_1^N \zeta(r_j) \vec{l}_i \bullet \vec{s}_i + \sum_{i>j}^M \frac{e^2 Z_i Z_j}{r_{ij}}
 \end{aligned}$$

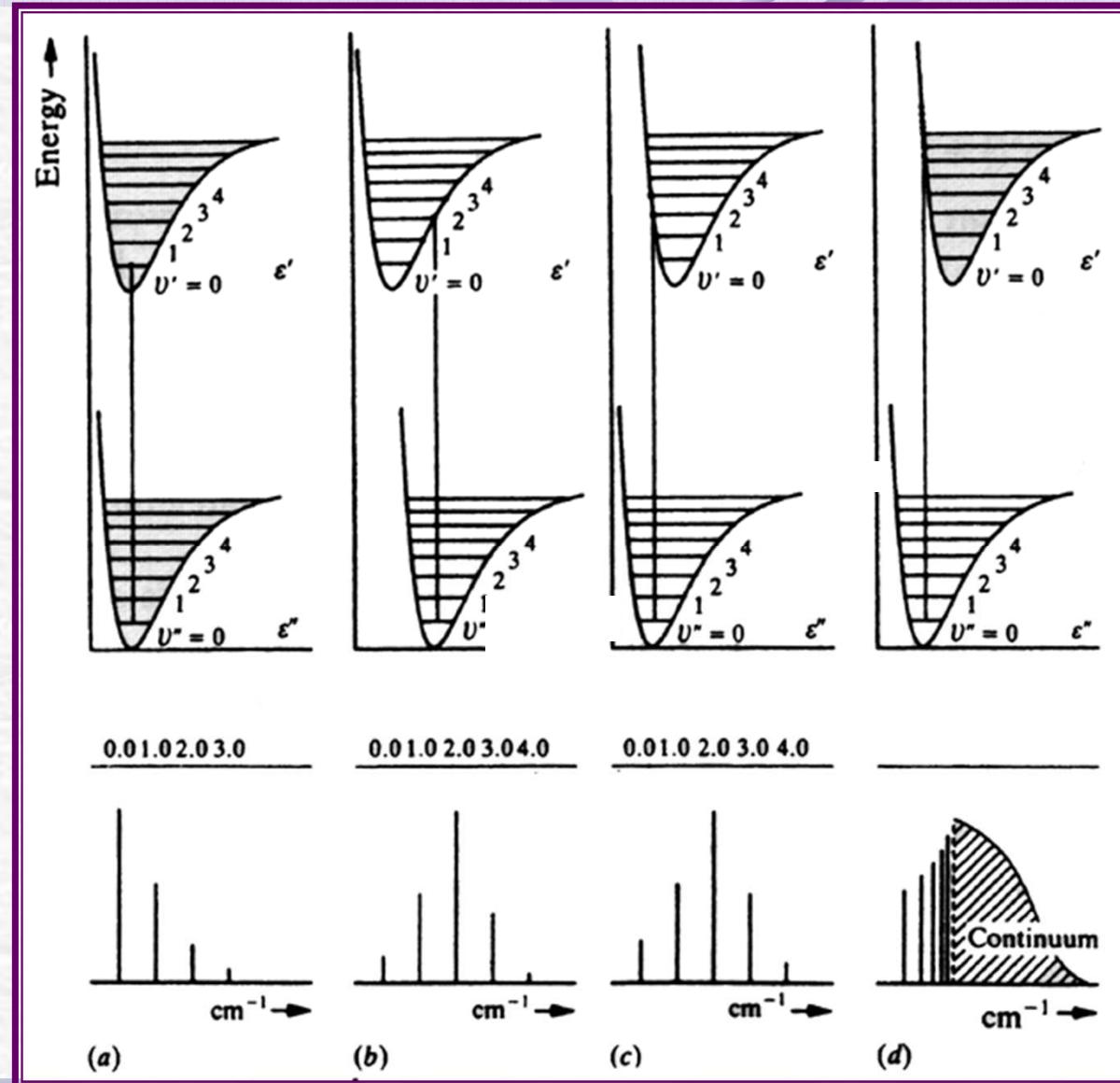
Born Oppenheimer

$$\left| \Psi_{A,B}^{(N)} \right\rangle = \left| \Psi_{A,B}^{(N)} \right\rangle \left| \Psi_{A,B}^{vib} \right\rangle$$

$$\frac{d\sigma}{d\Omega dE_e} \propto \frac{1}{h\nu} \sum_{A,B} \left| \hat{\epsilon} \bullet \left\langle \mathcal{E}_l \left| \vec{r}_j \right| \sum_{A\lambda} C_{A\lambda} \phi_{A\lambda} \right\rangle \left\langle \Psi_B^{(N-1)} \left| \Psi_R^{(N-1)} \right\rangle \right|^2 \right.$$

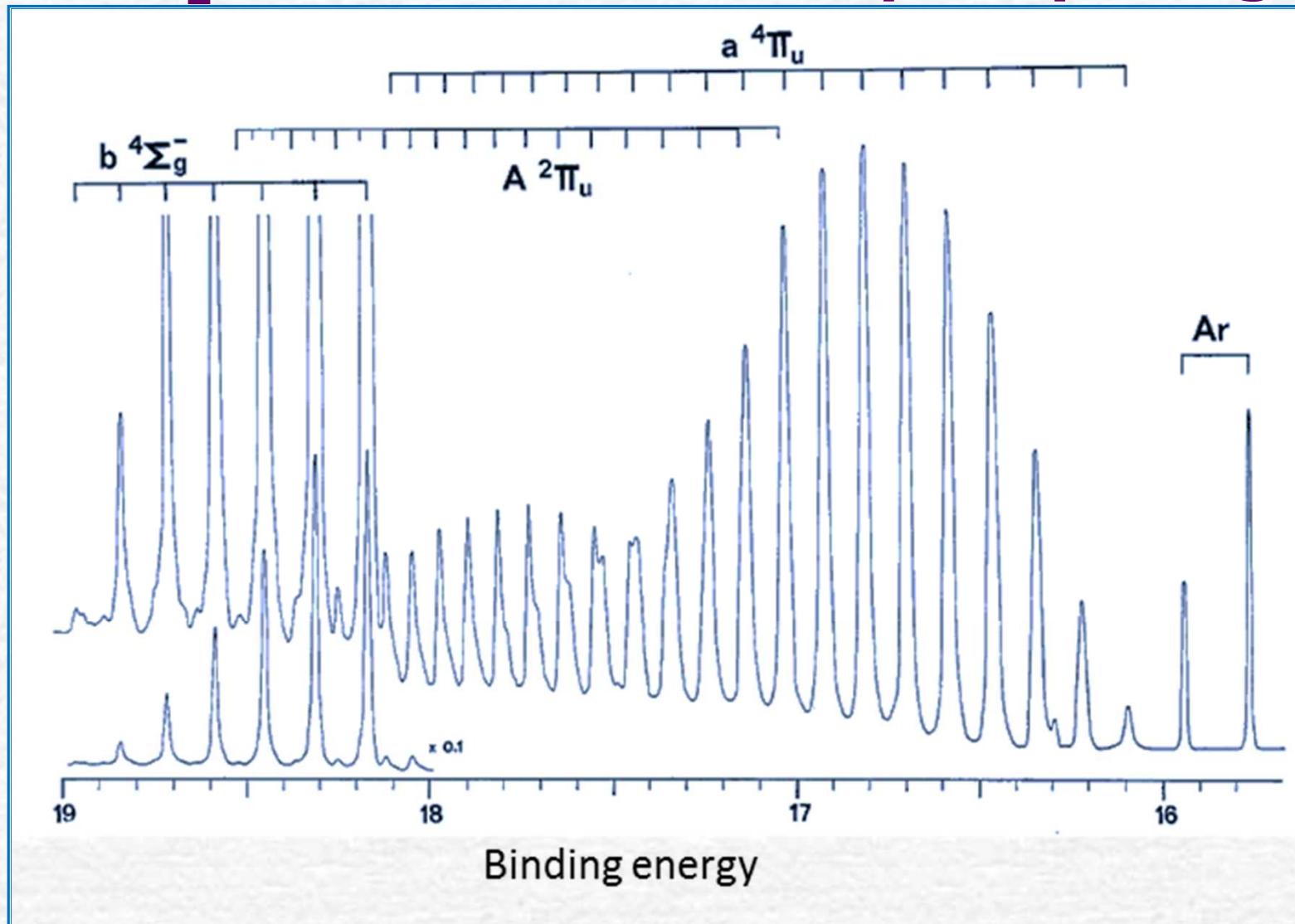
$$\left| \left\langle \Psi_B^{vib} \left| \Psi_A^{vib} \right\rangle \right|^2 \delta(E_e + E_B^{(N-1)} - E_A - h\nu) \right]$$

Frank Condon Factors



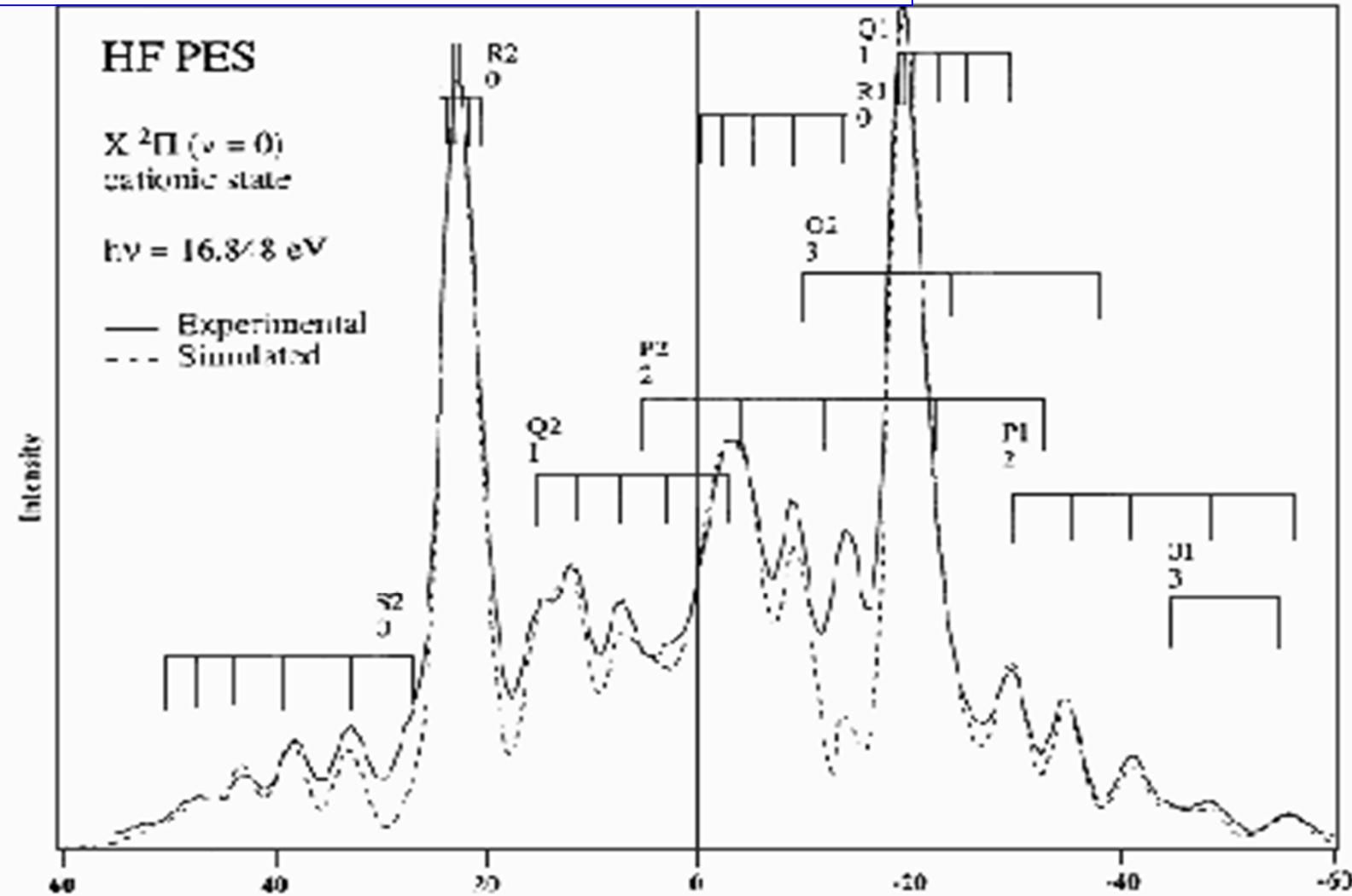
Photoelectron Spectroscopy
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PE O₂ vibrational and multiplet splitting

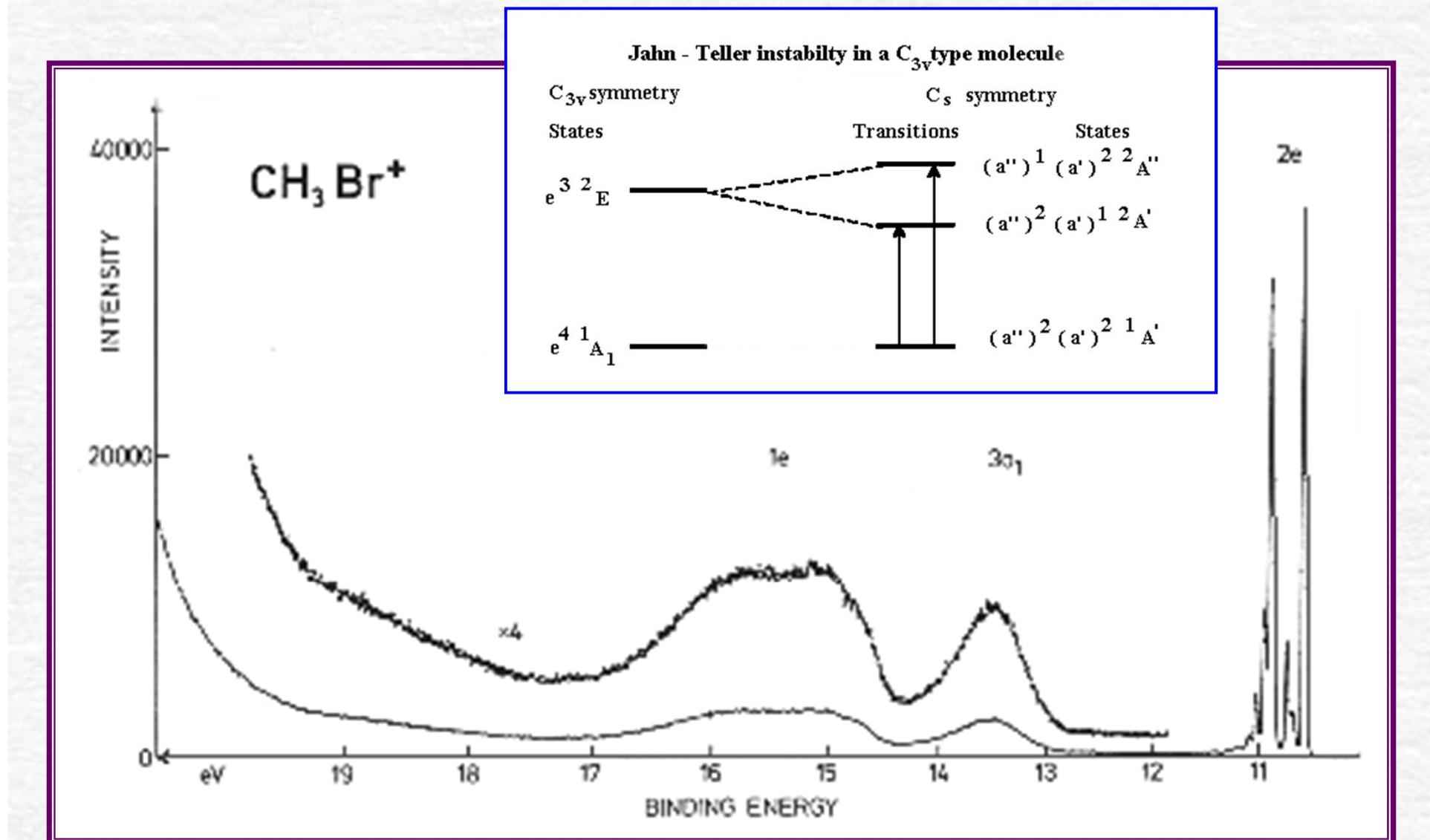


Rotational structure HF

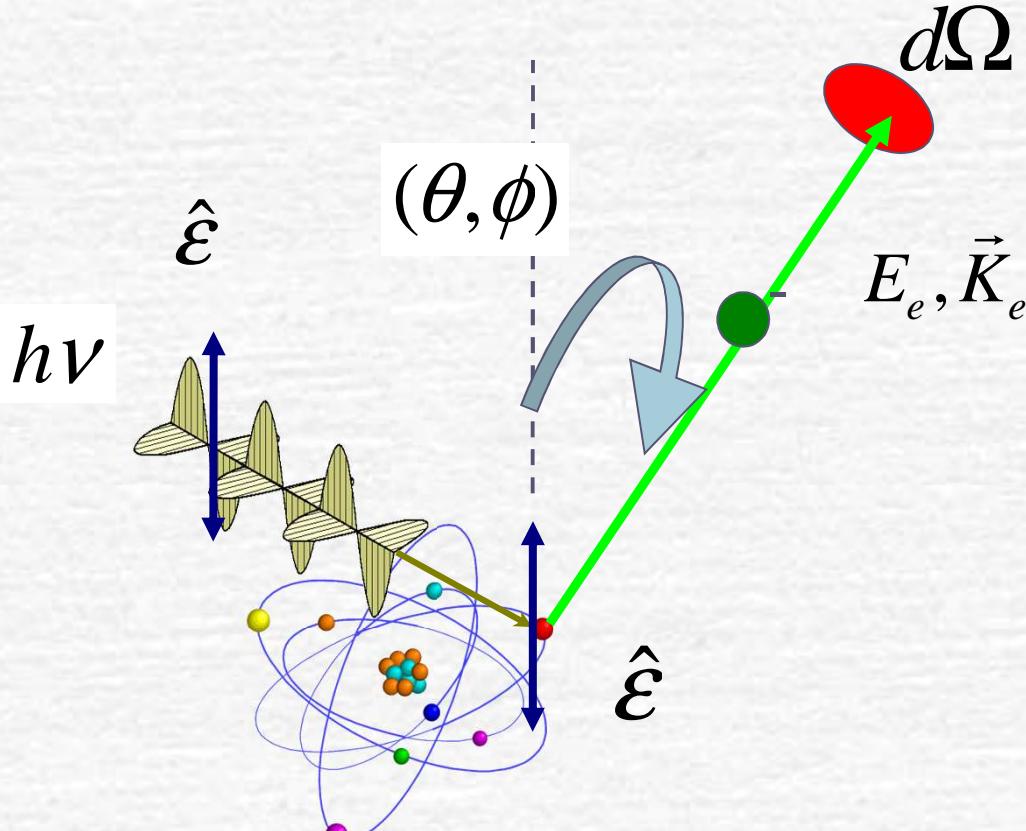
1 and 2 refer to the $^2\Pi_{3/2}$ and $^2\Pi_{1/2}$ spin-orbit split components



Jahn Teller splitting

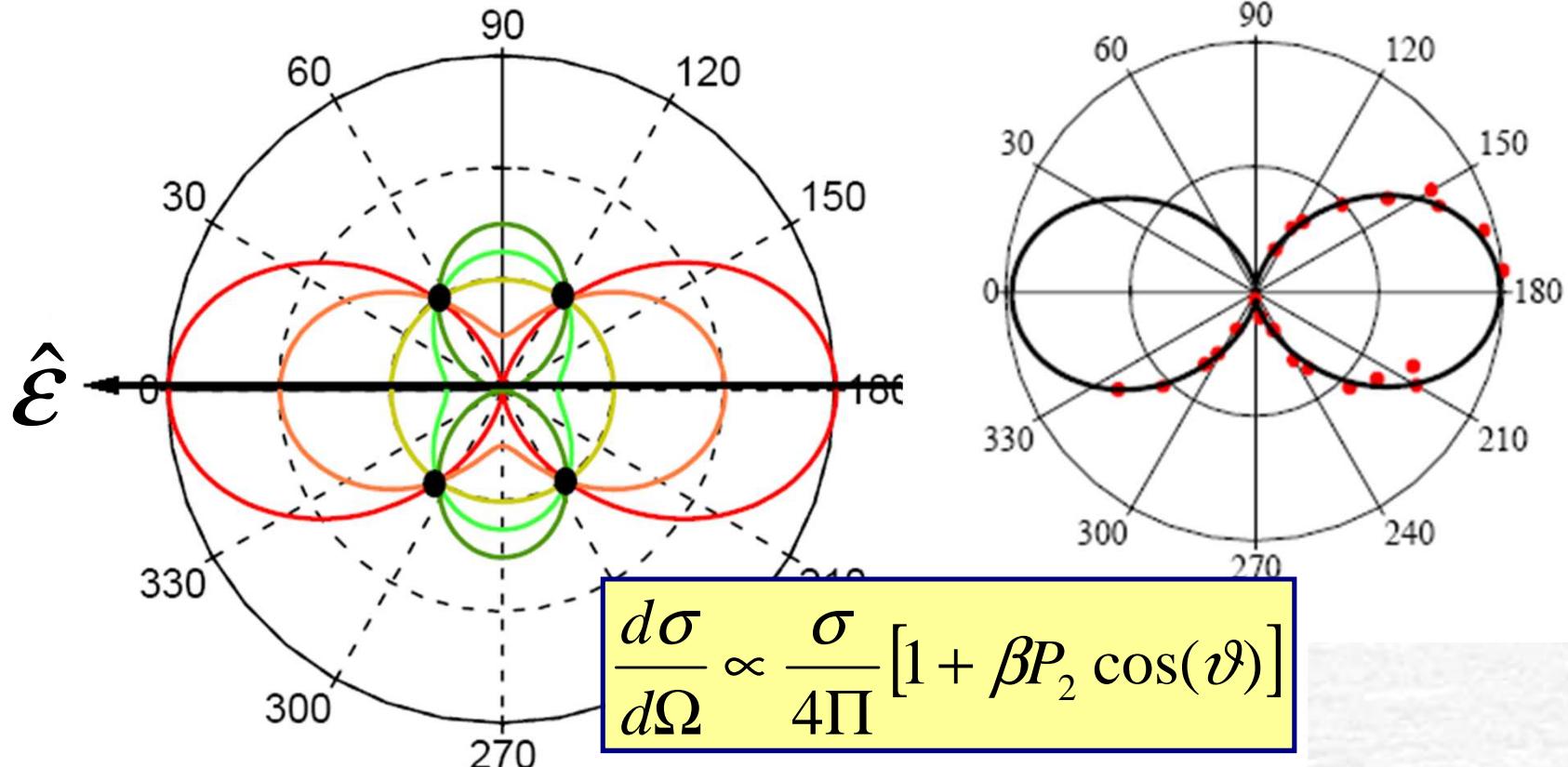


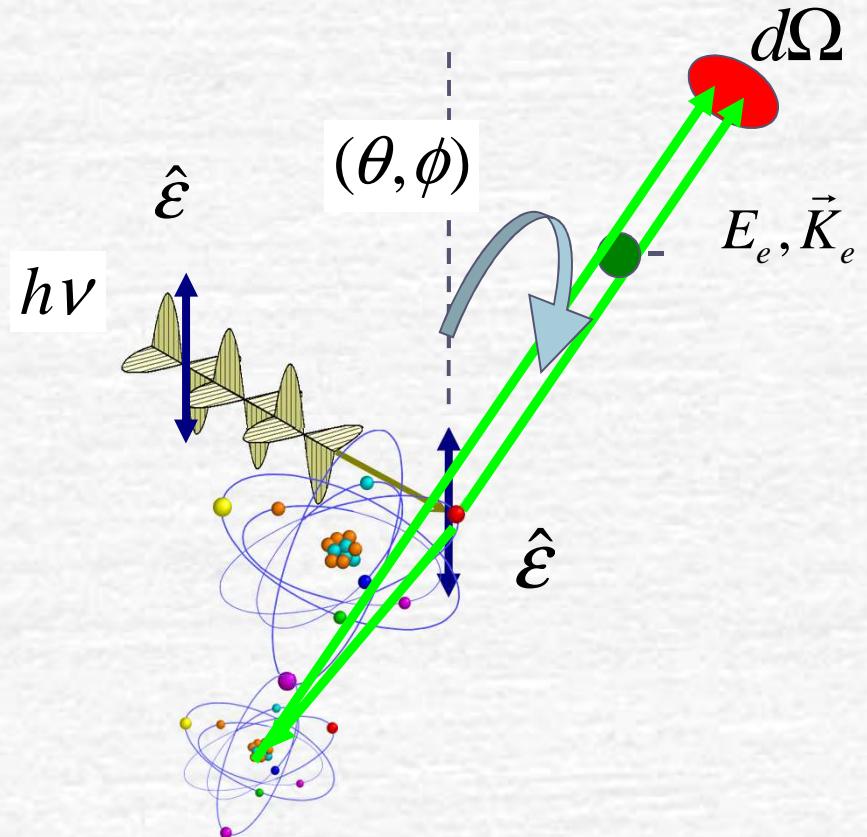
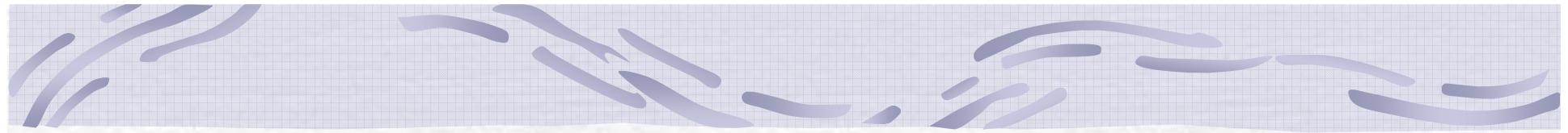
PE angular distribution



$$\frac{d\sigma}{d\Omega} \propto \frac{1}{h\nu} \sum_{A,B} \left| \hat{\epsilon} \bullet \left\langle \epsilon_l | \vec{r}_j | \phi_j(\vec{r}_j, \sigma_j) \right\rangle \left\langle \Psi_B^{(N-1)} | \Psi_R^{(N-1)} \right\rangle \right|^2$$

Angular distributions

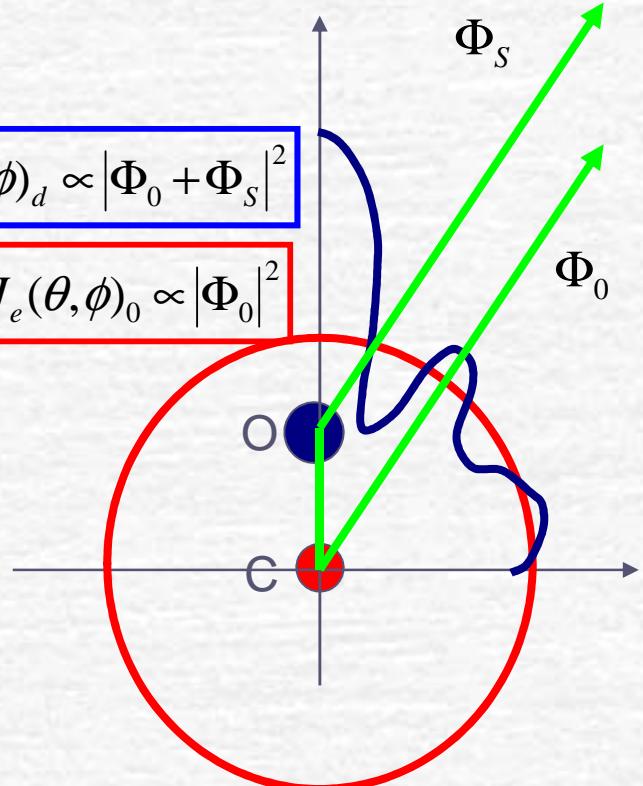




$$J_e \left| \Phi_{direct} + \sum_i \Phi_{scattered}^i \right|^2$$

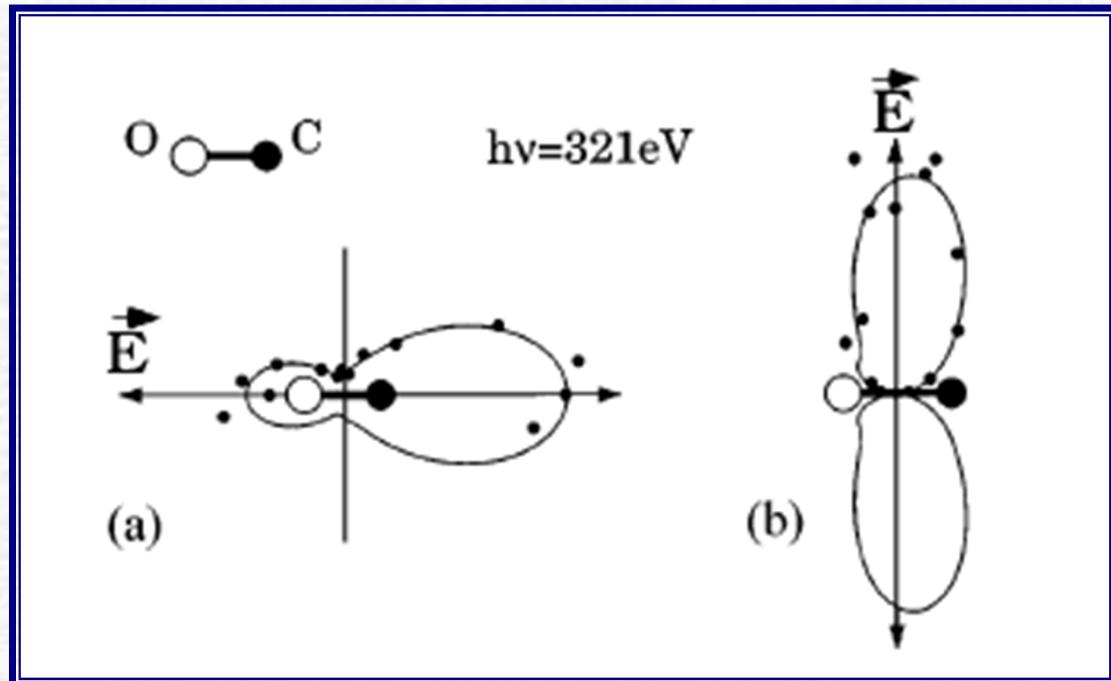
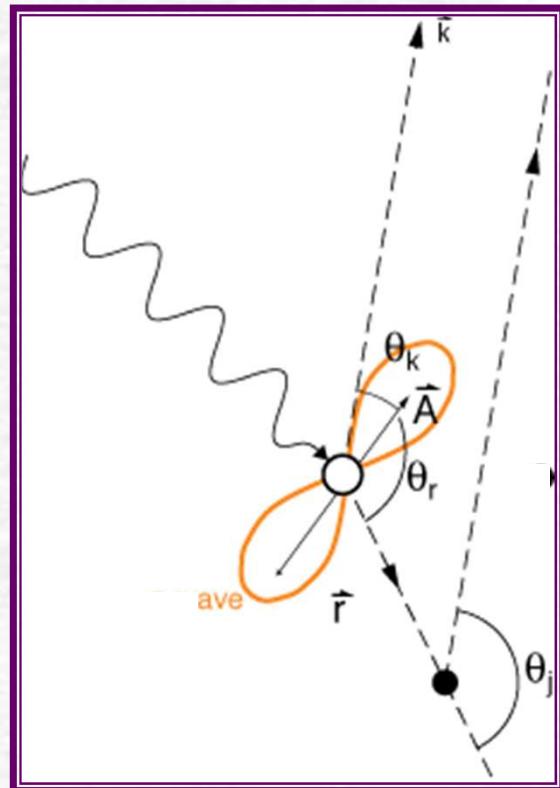
$$J_e(\theta, \phi)_d \propto |\Phi_0 + \Phi_S|^2$$

$$J_e(\theta, \phi)_0 \propto |\Phi_0|^2$$



Fixed in space molecules

CO C 1s



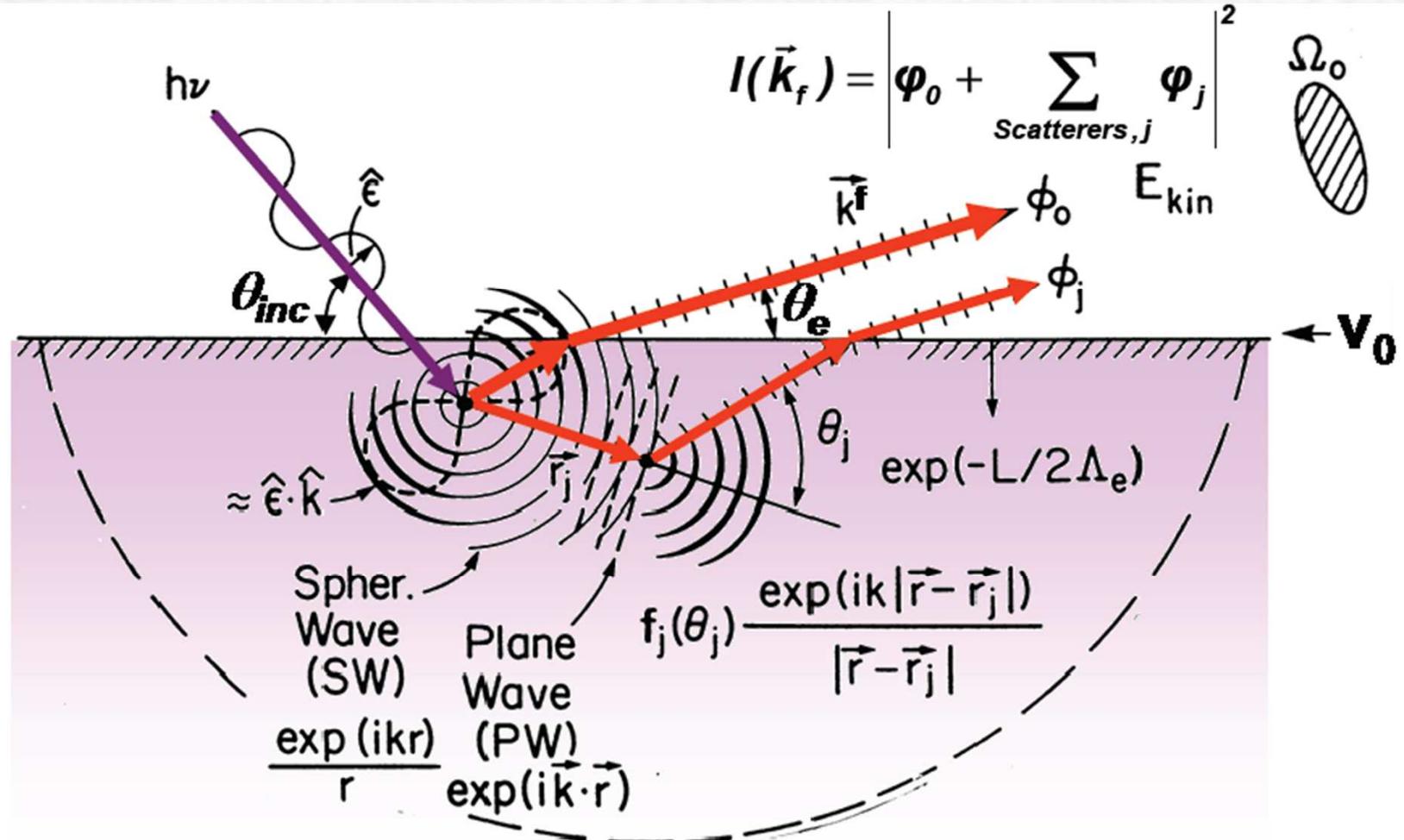
F. Heiser et al.

VOLUME 79, NUMBER 13

PHYSICAL REVIEW LETTERS

Photoelectron Spectroscopy
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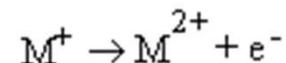
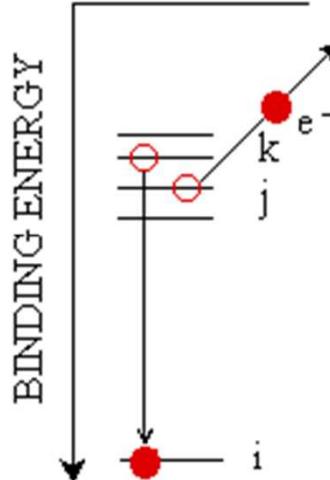
Application to surfaces



Core hole relaxation

SECONDARY PROCESSES

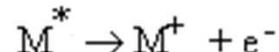
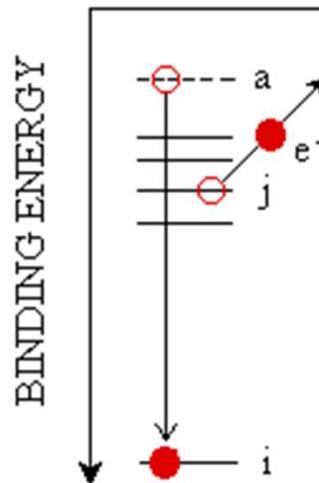
AUGER ELECTRON DECAY



Global energy, angular
Momentum and parity

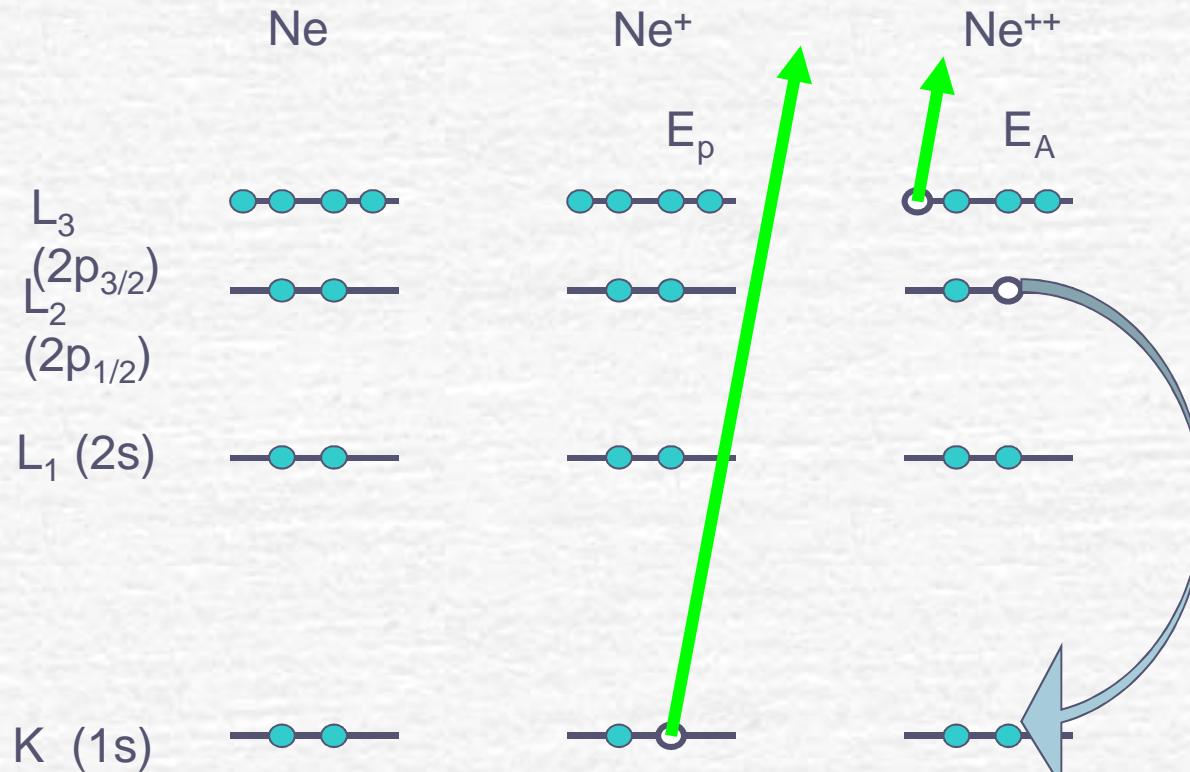
PROCESSES

AUTOIONIZATION



Energy, angular momentum,
Dipole selections at each step

Auger decay

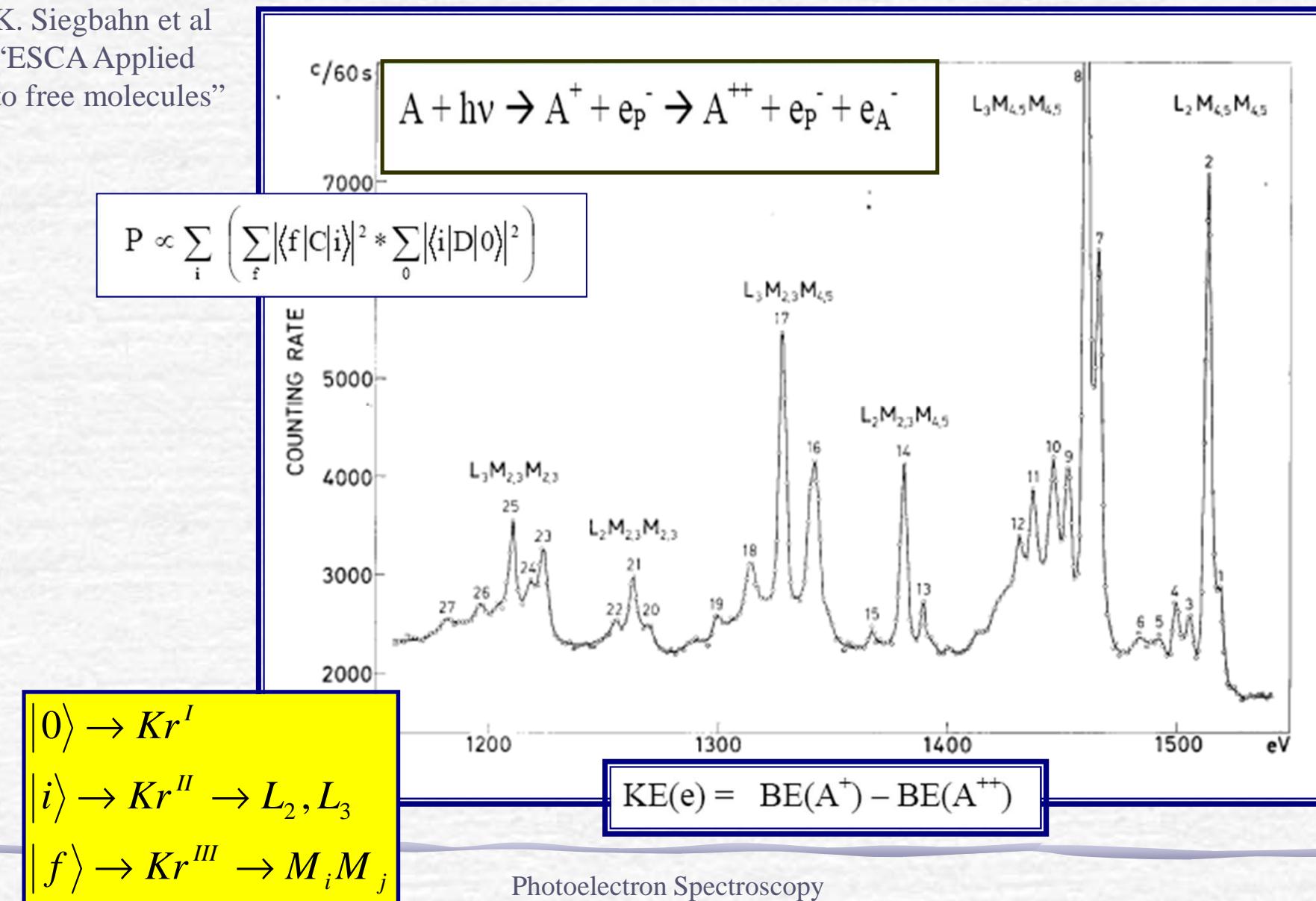


Auger Transition	Double ion valence configuration	Multiplet Terms
KL ₁ L ₁	2s ⁰ 2p ⁶	¹ S ₀
KL ₁ L _{2,3}	2s ¹ 2p ⁵	¹ P ₁ , ³ P ₀ , ³ P ₂ , ³ P ₁
KL _{2,3} L _{2,3}	2s ² 2p ⁴	¹ S ₀ , ³ P ₀ , ³ P ₂ , ¹ D ₂

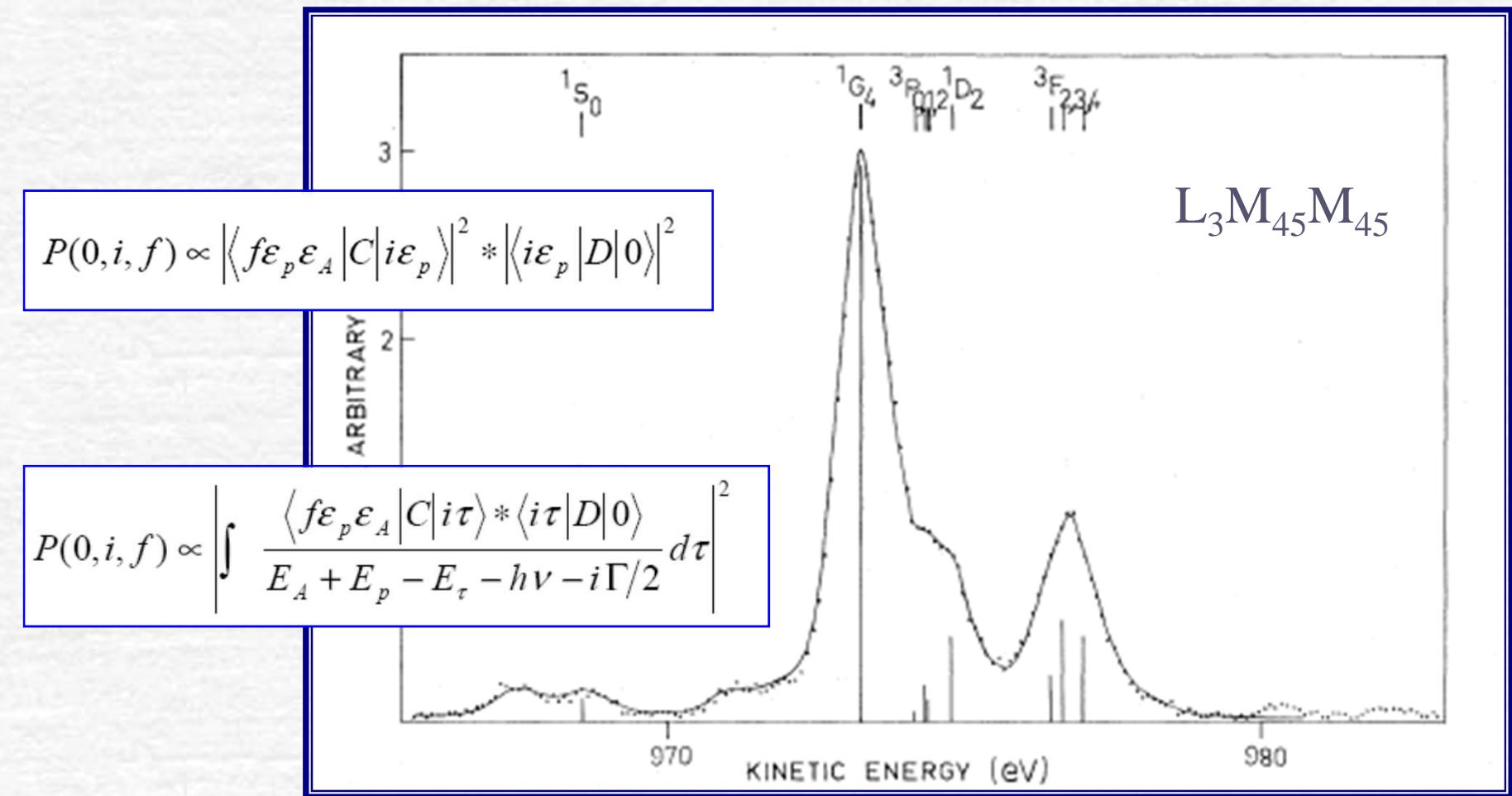
$$EA(KL_1L_2) = E(K) - E(L_1) - E(L_2, L_1)$$

Kr Auger spectrum

K. Siegbahn et al
“ESCA Applied
to free molecules”

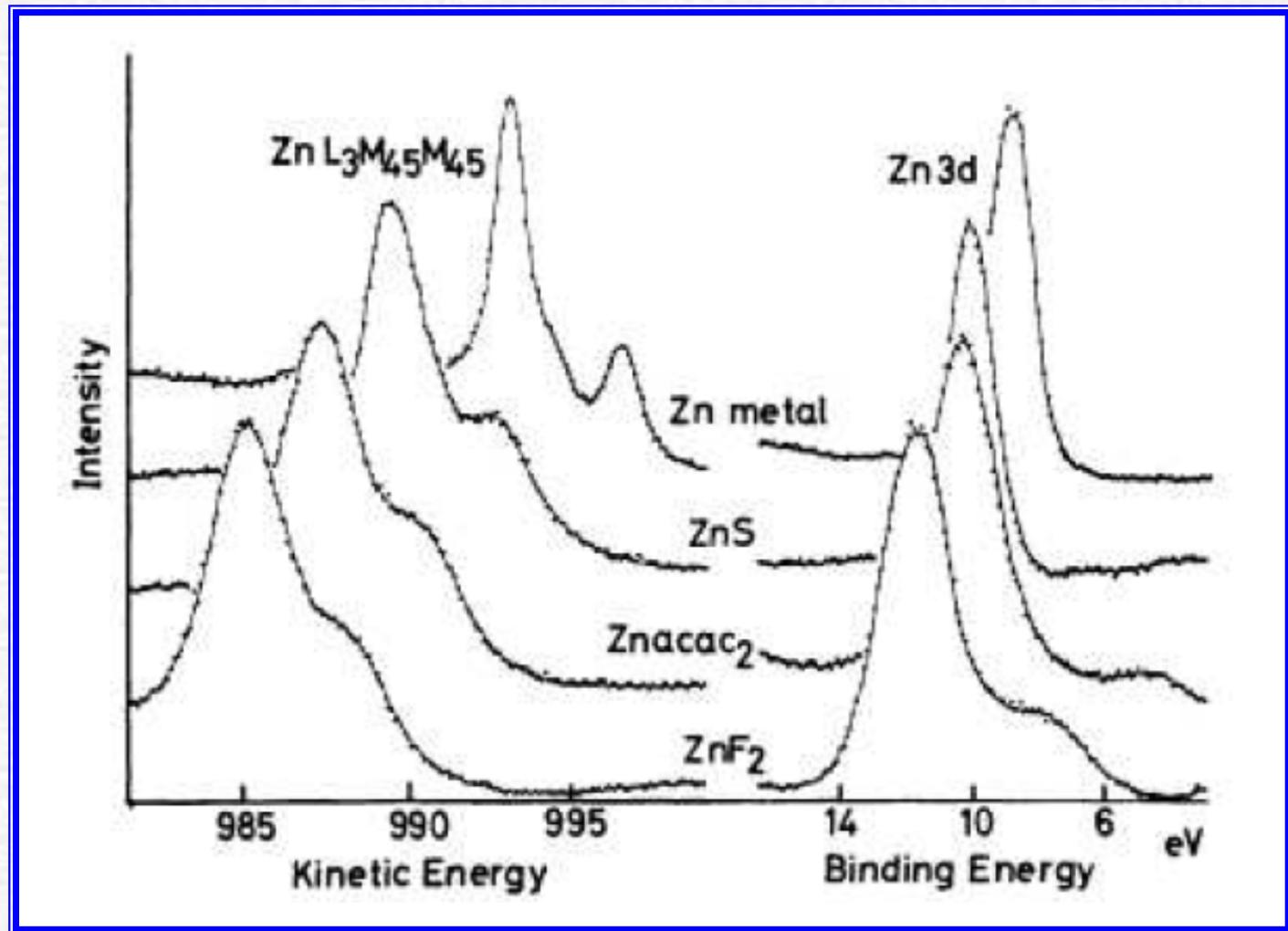


Zn Auger multiplet splitting

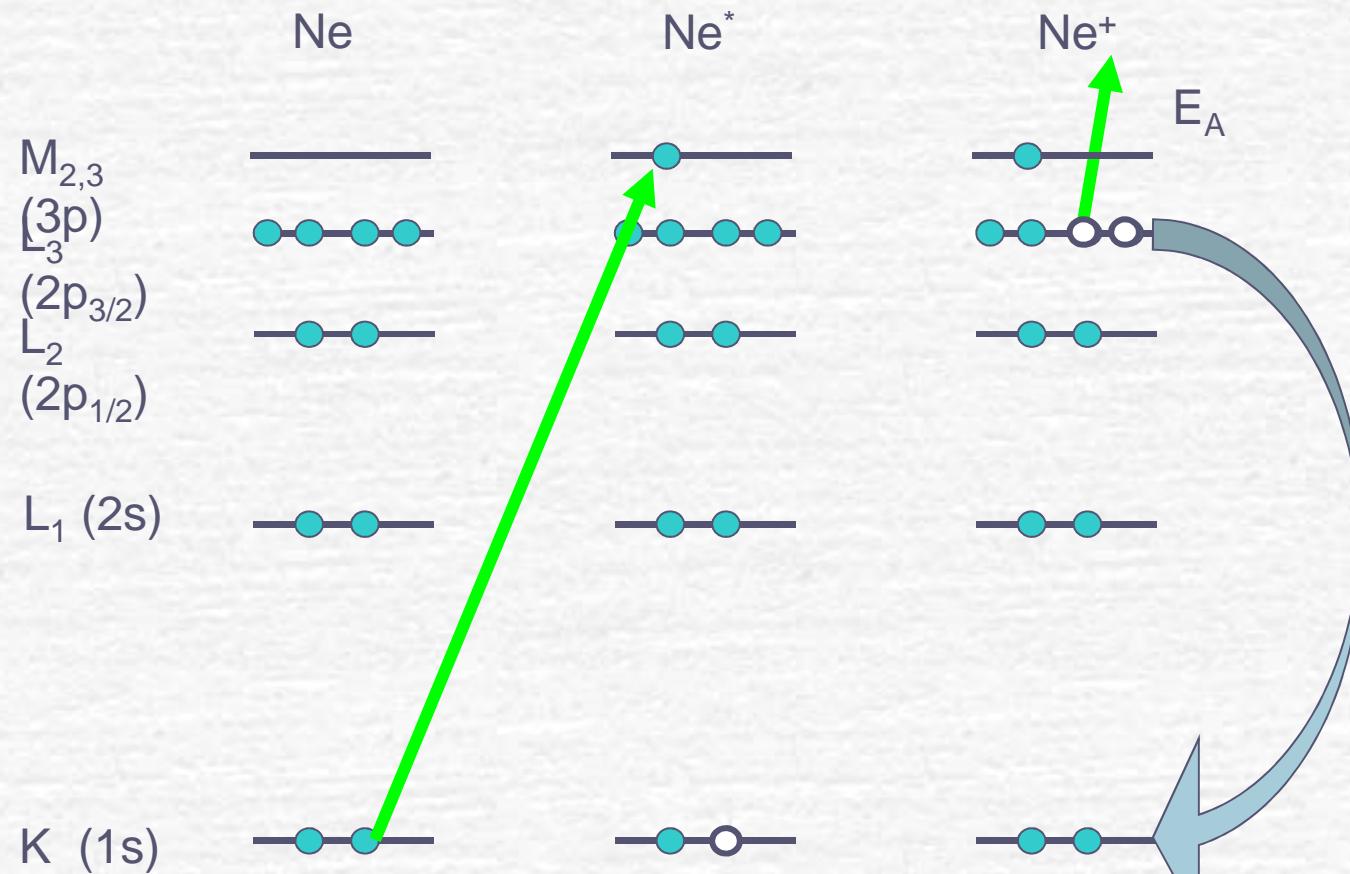


Aksela et al. PRL 33,999 (1974)

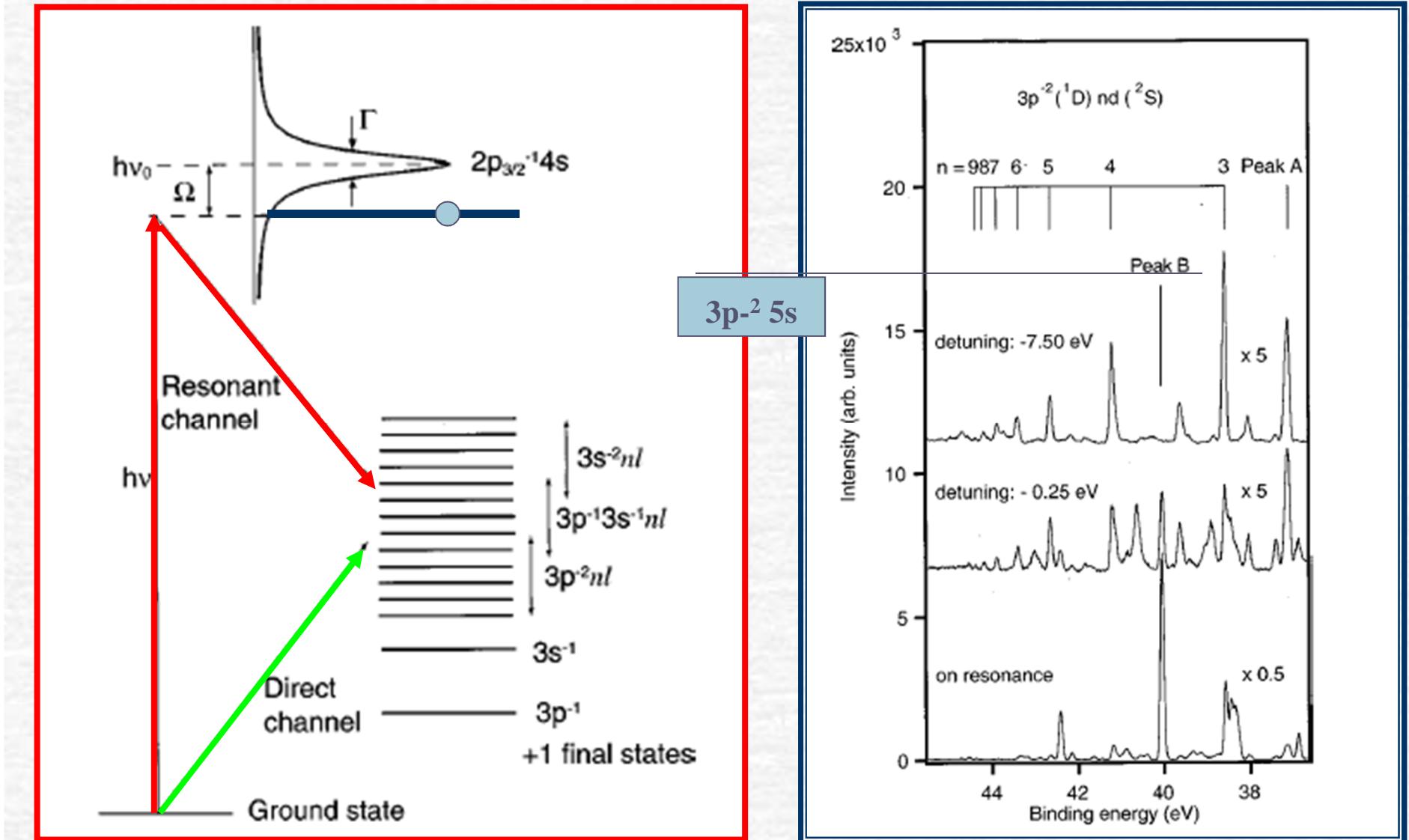
Auger chemical shift



Autoionizing decay



Ar Autoionization spectrum



Phys. Rev. A 63,032514

Photoelectron Spectroscopy
XIII SILS School G. Stefani

Quantum interference

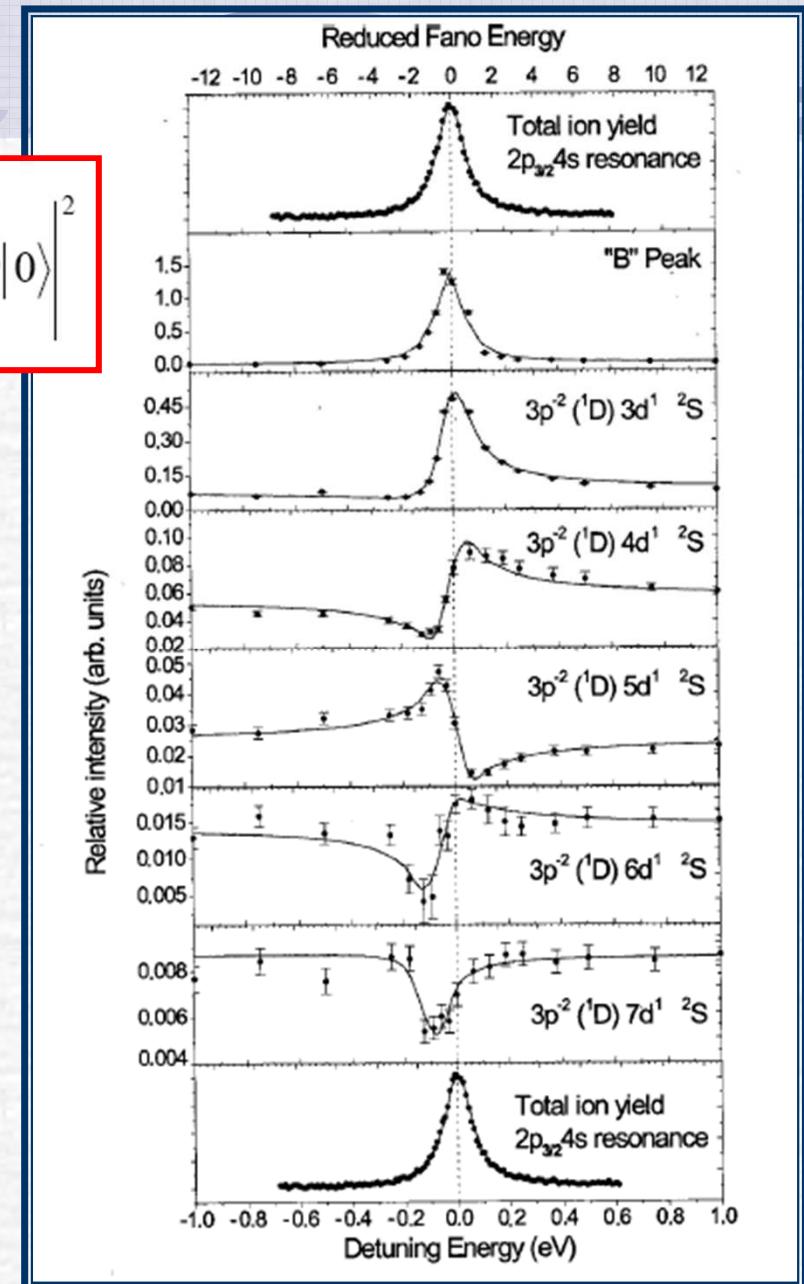
$$P(0, i, f) = \left| \int \frac{\langle f \varepsilon_A | C | i \tau \rangle^* \langle i \tau | D | 0 \rangle}{E_A + E_p - E_\tau - h\nu - i\Gamma/2} d\tau + \langle f \varepsilon_A | D | 0 \rangle \right|^2$$

$$\sigma(h\nu) = \sigma_T \left\{ \rho'^2 \left[\frac{(q+\varepsilon')^2}{\varepsilon'^2+1} + \frac{\Delta h\nu}{\Gamma} - 1 \right] + 1 \right\}.$$

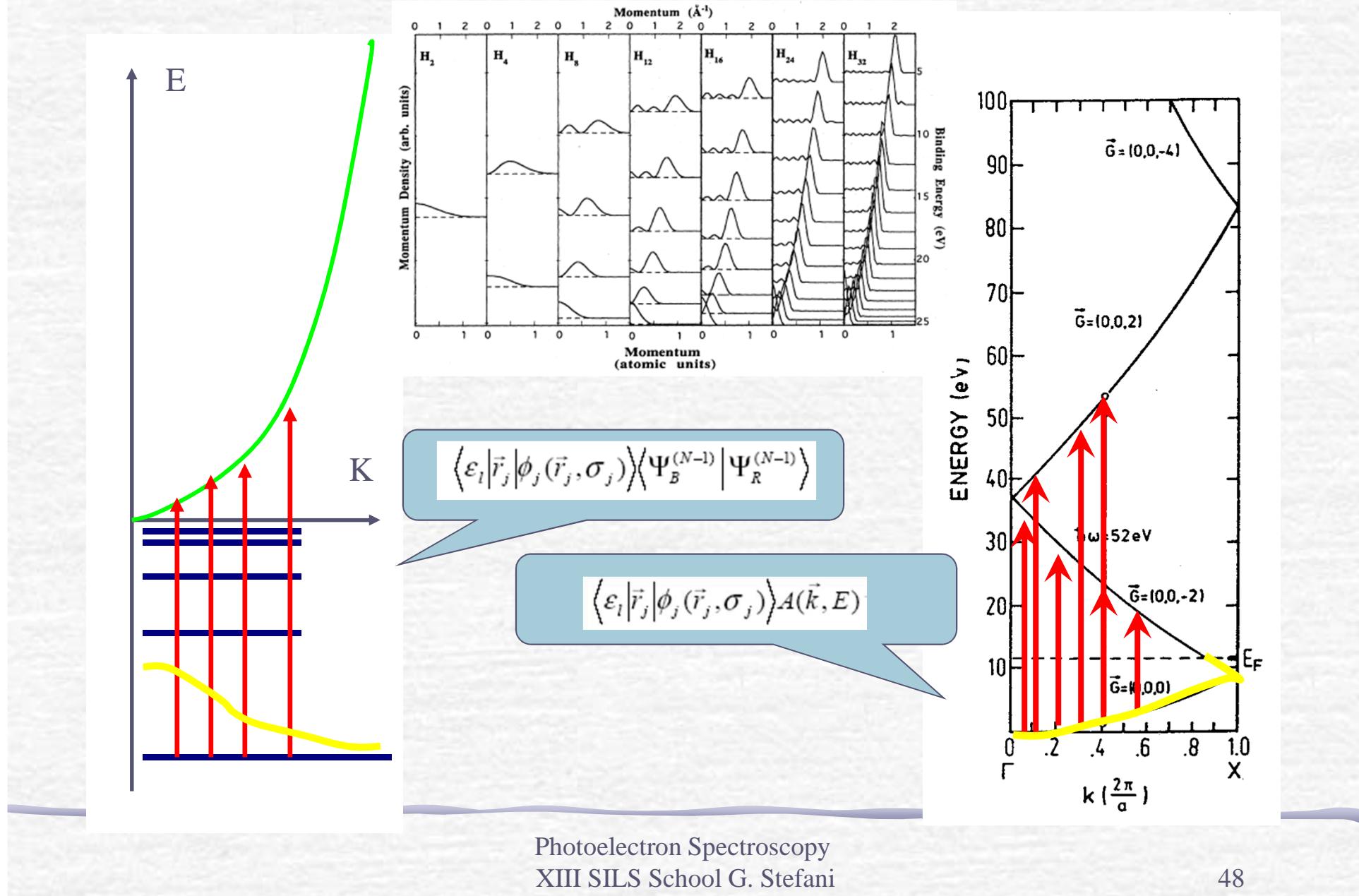
What does it happen when the photoemission line concides with the autoionization line?

Interference between direct and resonant channel

Phys. Rev. A 63,032514



From central to periodic potential



Spectral Function in Interacting Solids

$$\Psi_{Nf} = A \Psi^{N-1} f \Phi_{kf}$$

$$J_e \propto \sum_{if} |M_{if}^2| \sum_m |m_{im}|^2 \times \delta(E_i^N + h\nu - E_{m-kin}^{N-1})$$

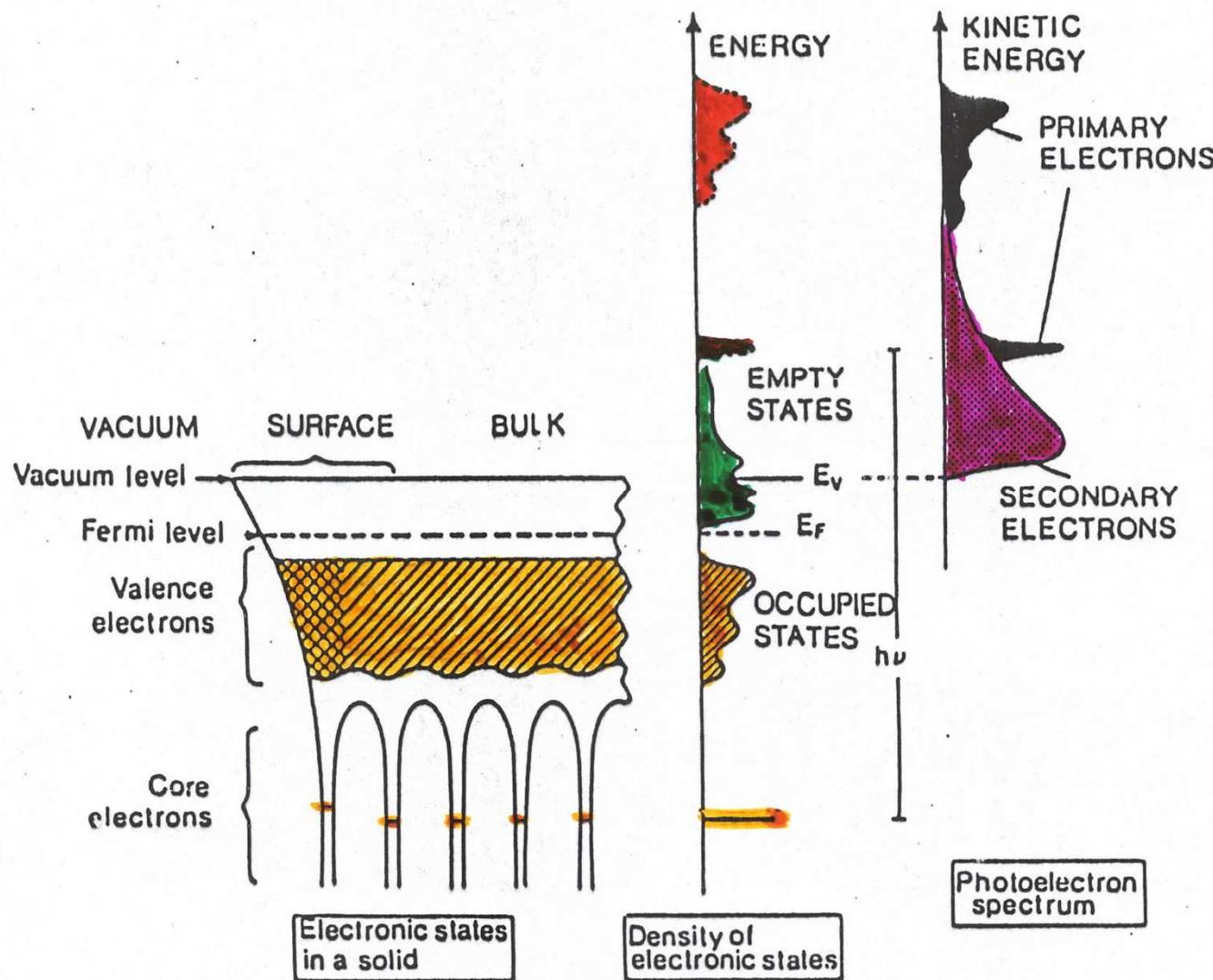
$$A(k, \varepsilon) = \sum_m (| \langle \Psi_{m-kin}^{N-1} | c_k | \Psi_i^N \rangle |^2 \times \delta(\varepsilon + E_{m-kin}^{N-1} - E_i^N))$$

$$J_e(k, \omega) \propto \sum_{if} |M_{if}|^2 A(k, E_{kin} - h\nu) f(E_{kin} - h\nu)$$

For non interacting particles $A(\varepsilon, k) = \delta(\varepsilon - E_k)$ where $E_k = E_i^{N-1} - E_i^N$

$$A(\varepsilon, k) = 1/\pi |\Sigma''(k, \varepsilon)| / [|\varepsilon - E_k - \Sigma'(k, \varepsilon)|^2 + |\Sigma''(k, \varepsilon)|^2]$$

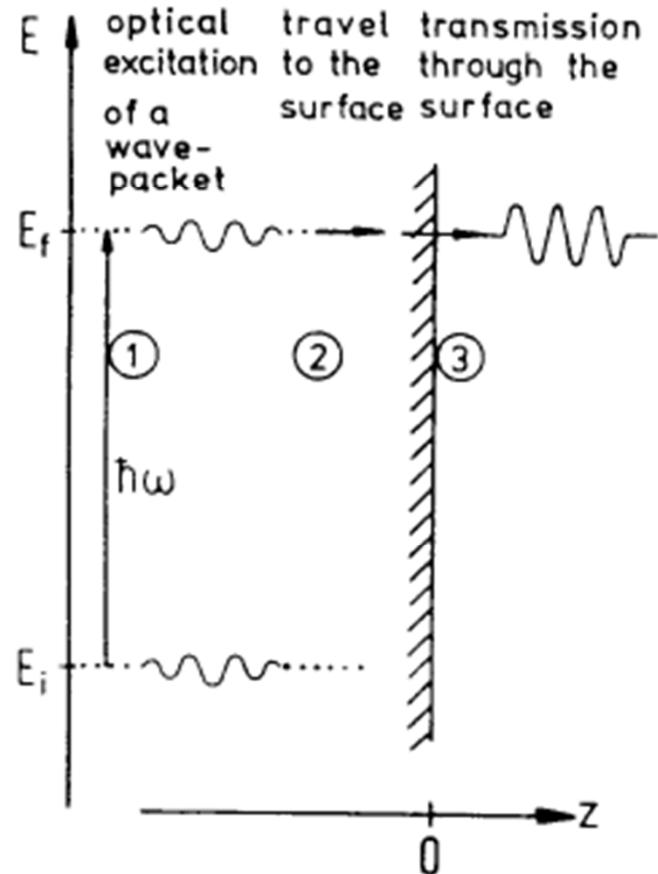
Photoemission Spectroscopy



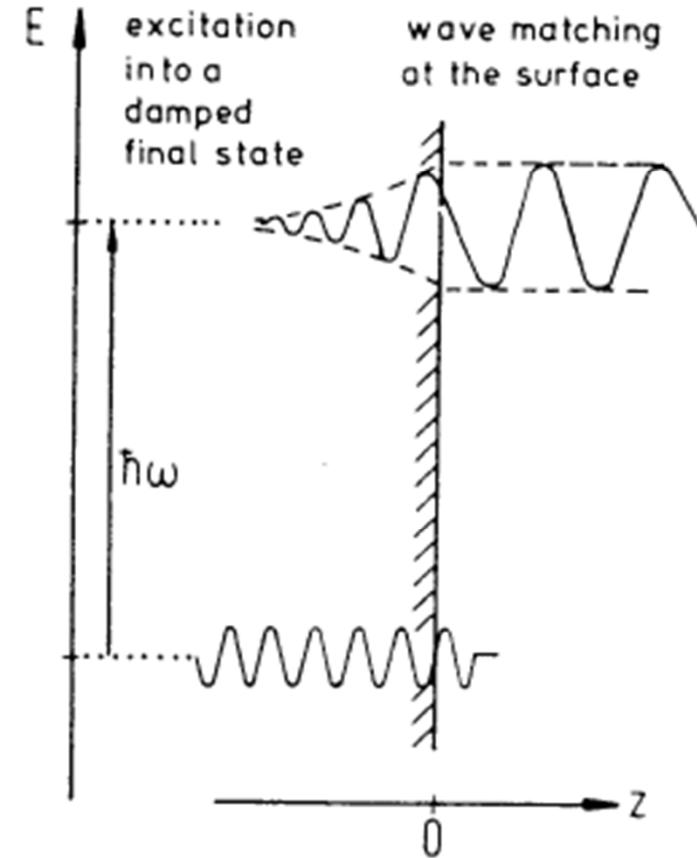
How real spectra look like:
Primary and Secondary Electrons

The Three-Step model 1

three-step model



one-step model



The three-step model 2

1. Dipole transition

$$J_e = \sum_{if} f(E_i) [1-f(E_f)] M_{if}^2 \times \delta[E_{kin} - (E_f - \Phi)] \delta(E_f - E_i - h\nu) \delta(\mathbf{k}_i + \mathbf{G} - \mathbf{k}_f)$$

2. Elastic transport

$$d(E_f, \mathbf{k}) = \alpha\lambda/(1+\alpha\lambda)$$

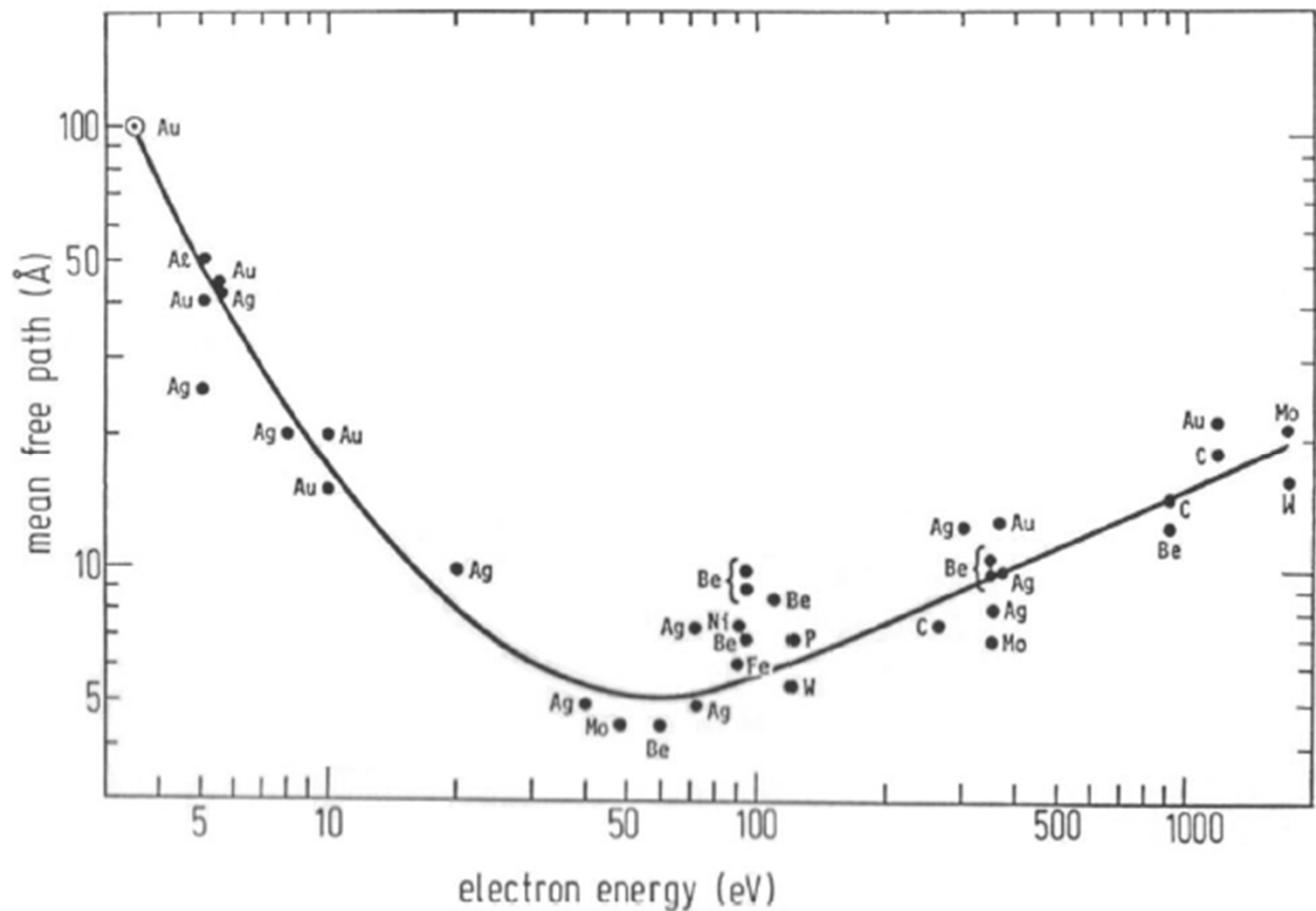
3. Exit to vacuum

$$T(E_f, \mathbf{k}_{ext}) = \begin{cases} 0 & \text{if } E_f < E_F + \Phi \\ 1/2 \sqrt{[1 - (E_F + \Phi)/E_f]} & \text{if } E_f > E_F + \Phi \end{cases}$$

Total current

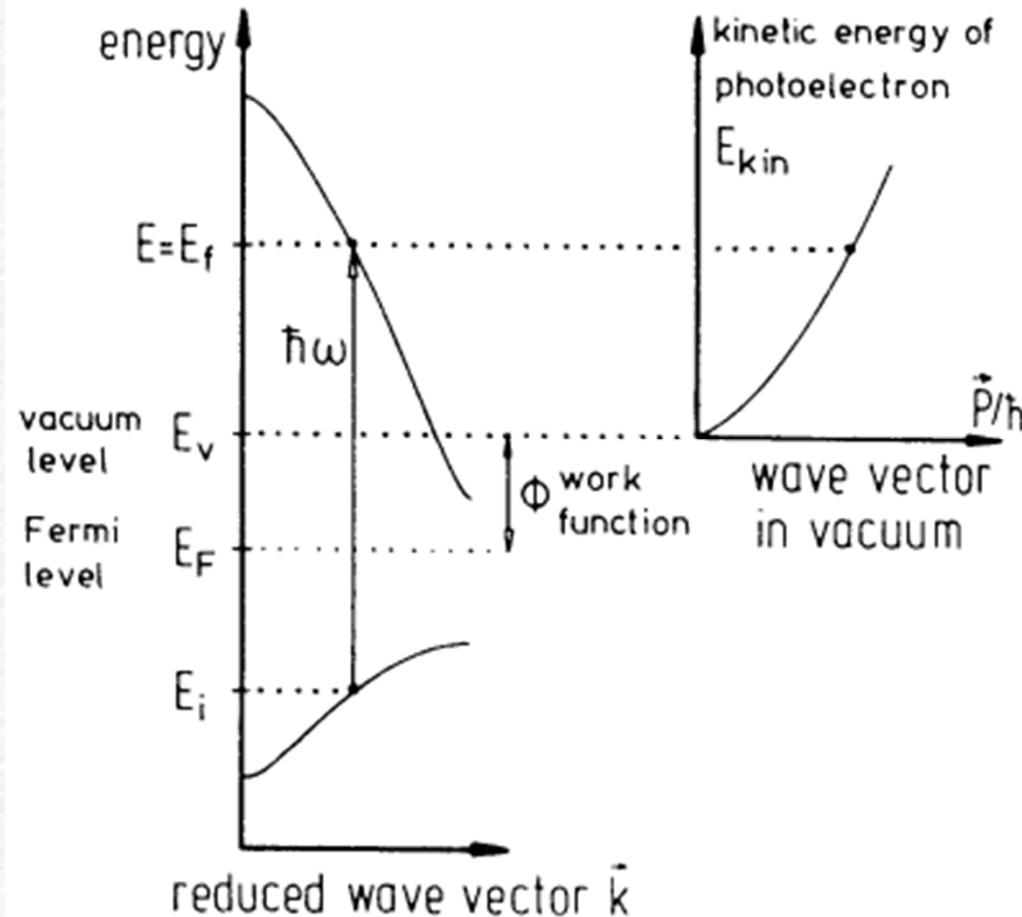
$$J_e \propto \sum_{if} f(E_i) [1-f(E_f)] M_{if}^2 \times T(E_f, \mathbf{k}_{ext}) \times d(E_f, \mathbf{k}) \times \delta[E_{kin} - (E_f - \Phi)] \delta(E_f - E_i - h\nu) \times \delta(\mathbf{k}_i + \mathbf{G} - \mathbf{k}_f) \times \delta(\mathbf{k}_i'' + \mathbf{G}'' - \mathbf{k}_f'')$$

Photoemission Spectroscopy

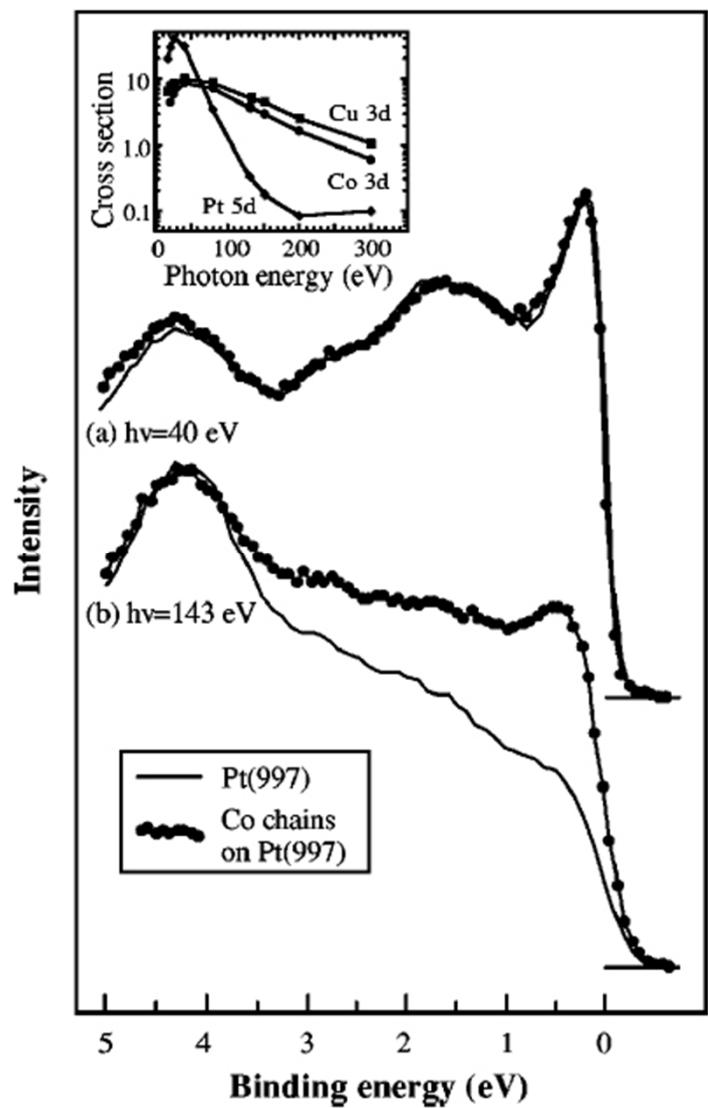


electron mean free path

How to reconstruct the initial state

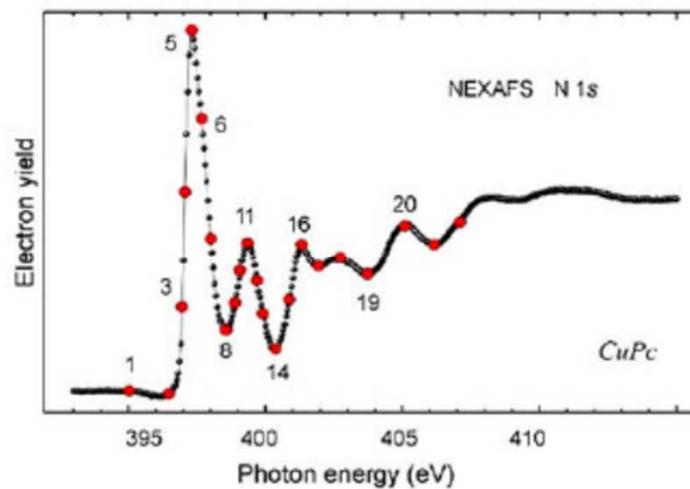
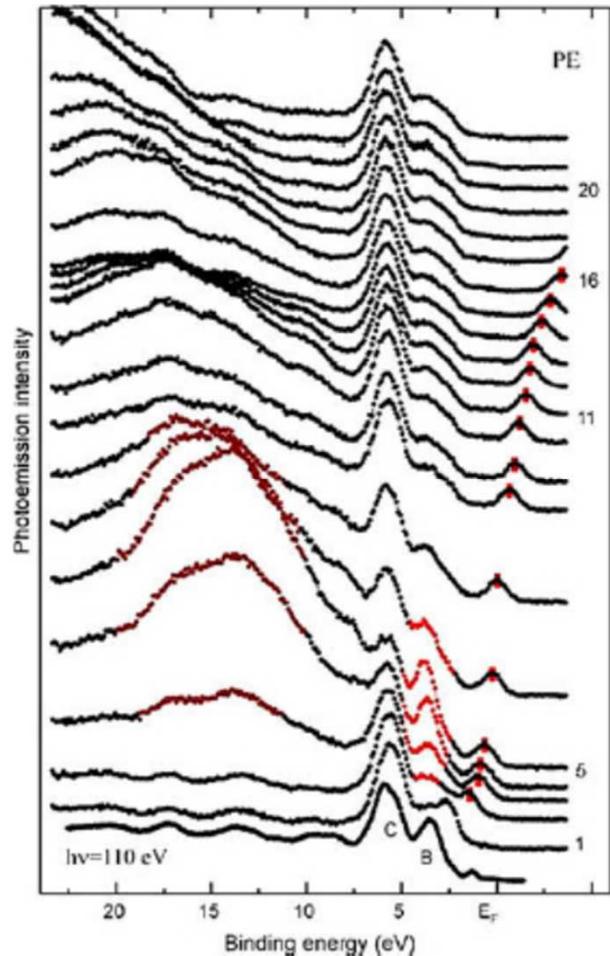


Valence band EDC and Cooper minimum



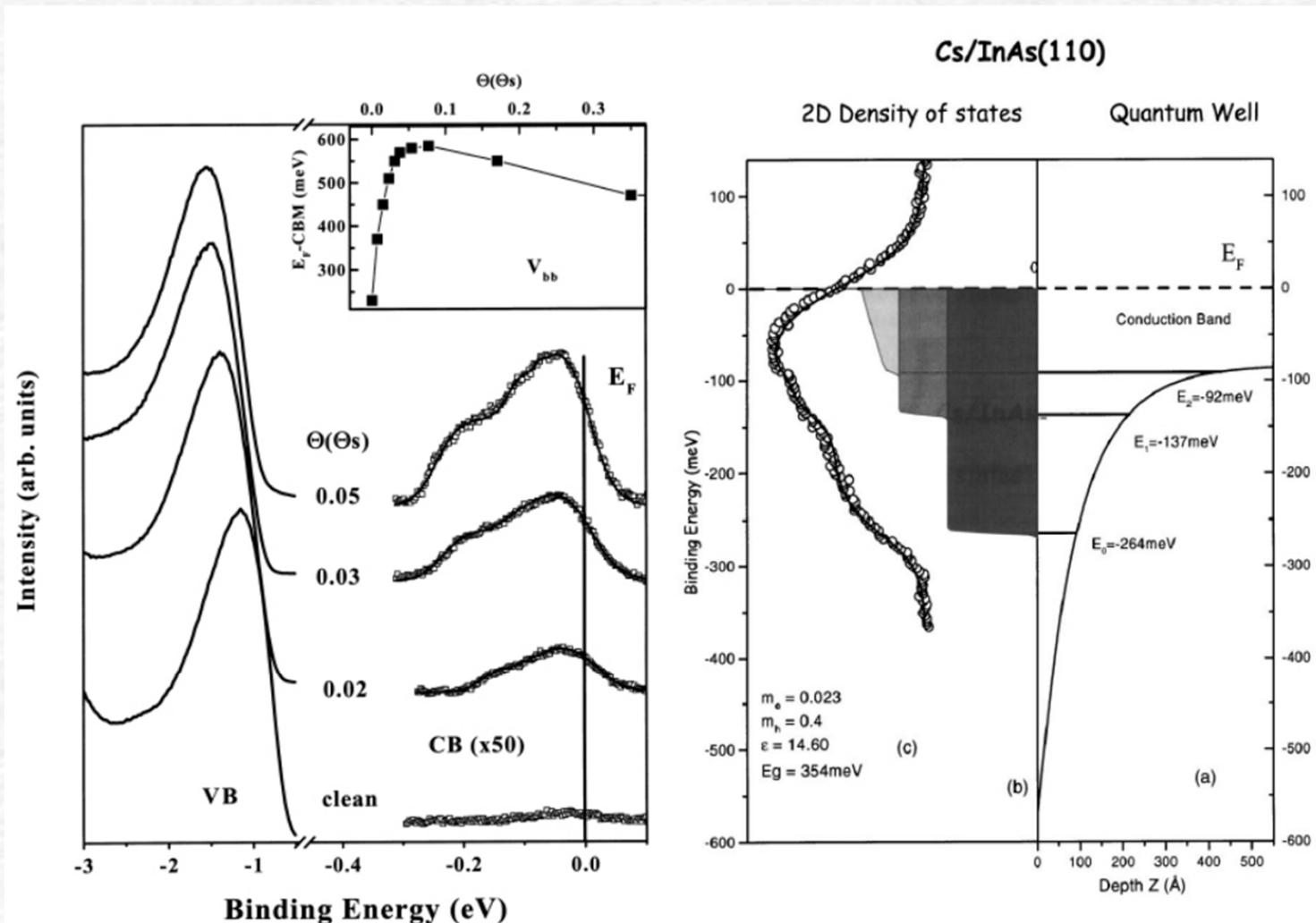
Valence Band EDCs of the clean Pt(997) surface (thin lines) and of Co-nanowires grown on Pt(997) (dots and thick lines), taken at different photon energies

Resonant photoemission



Valence Band EDCs of a CuPc thin-film taken at different photon energies (left panel) and X-ray Absorption Spectroscopy (XAS) from the same CuPc across the N K-edge (right panel).

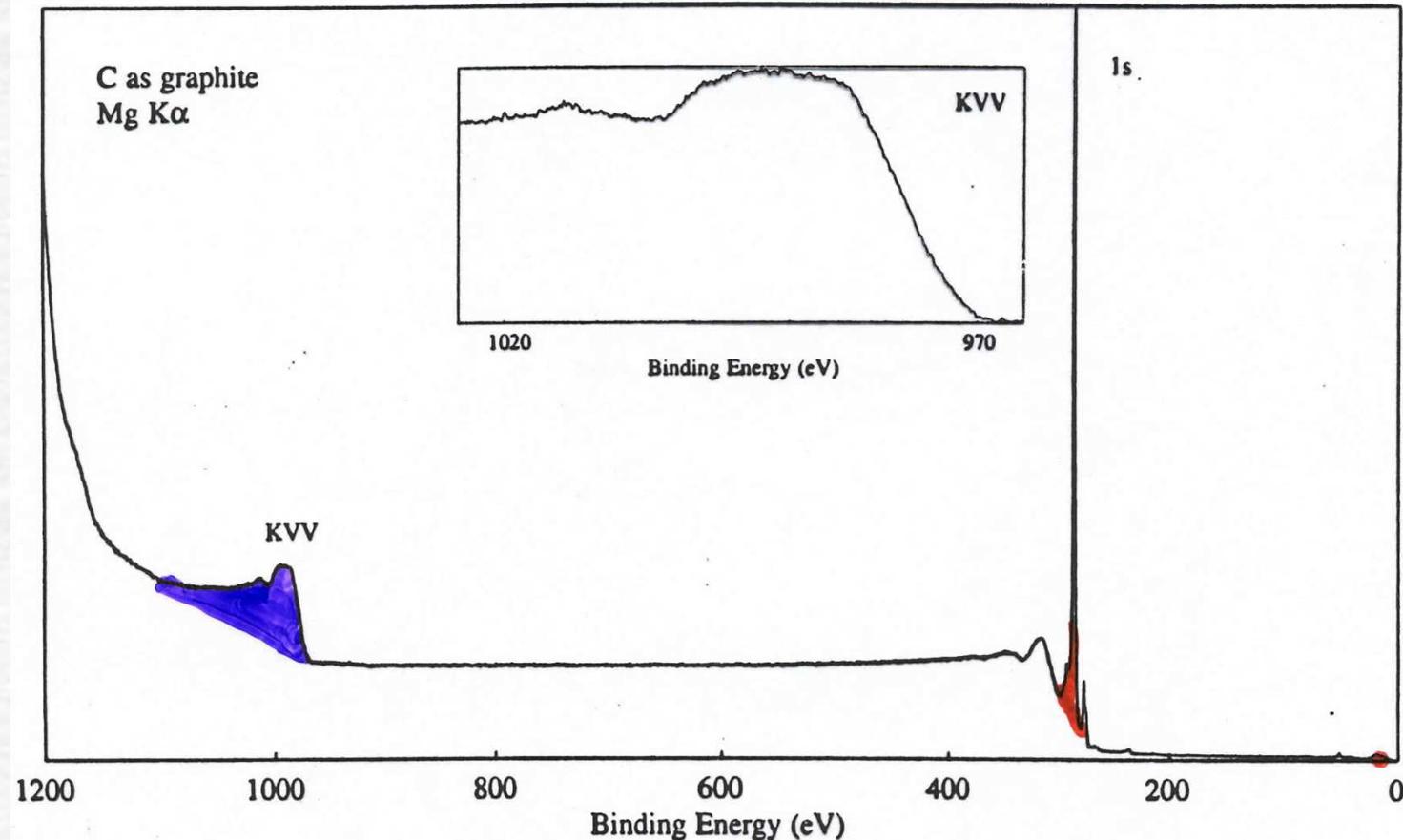
2D electron gas spatially confined Cs/InAs(110)



Photoemission - Core Level Spectroscopy

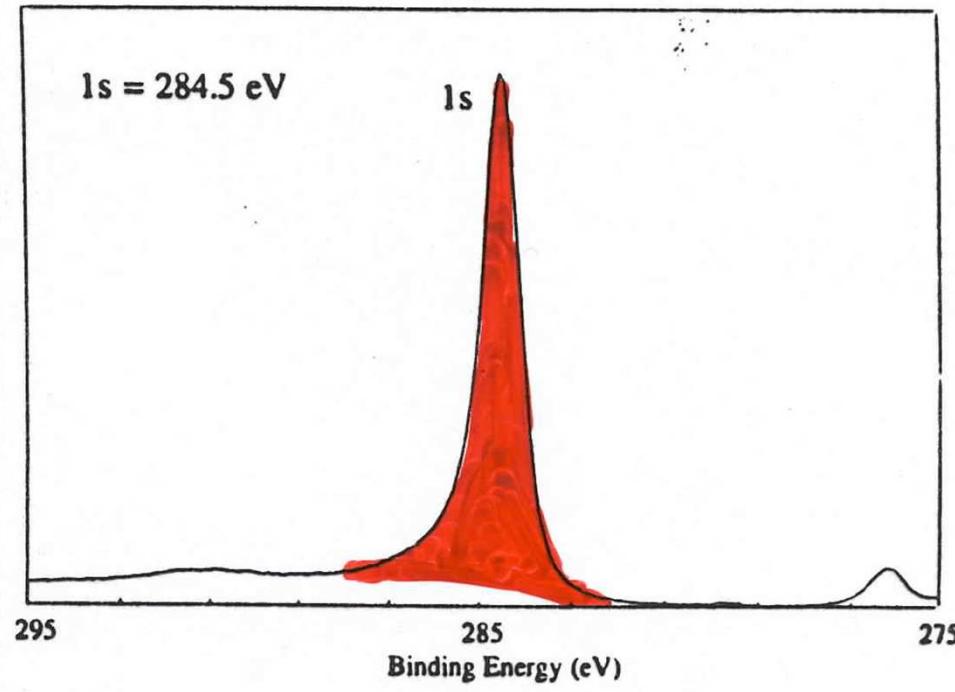


Wide XPS spectrum of graphite (C)



Photoemission Spectroscopy

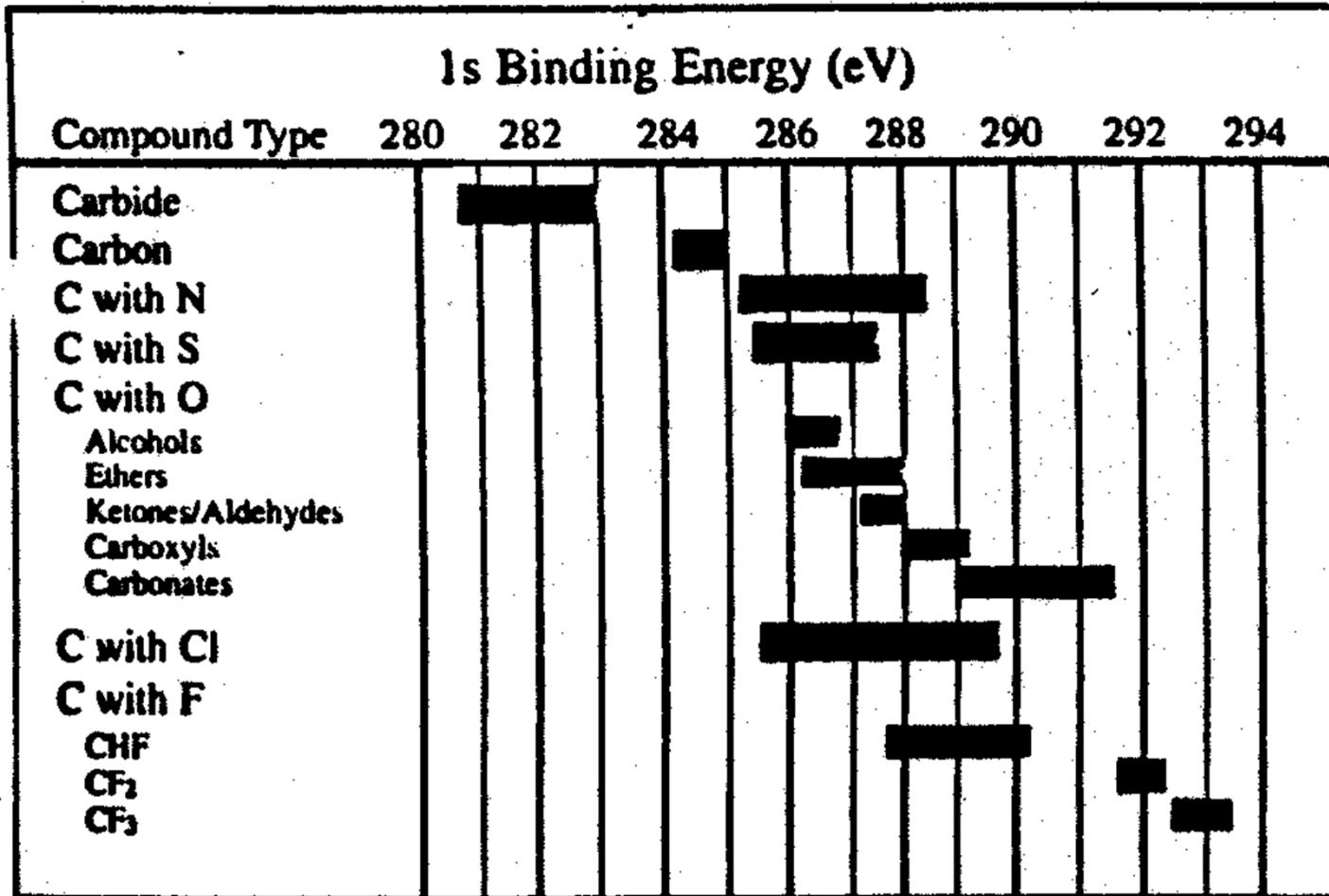
Core level XPS spectrum
of graphite (C)



The singlet **C 1s** line is characterized by:

- 1) A specific binding energy which reflects the specific atomic species (C) in a specific chemical environment
- 2) A finite width reflecting the instrumental resolution, lifetime broadening and other many-body effects

Photoemission Spectroscopy: Chemical Shift (ΔE_b)



How the C
1s binding
energy
reflects
differing
chemical
environme
nt local to
the
excited C
sites

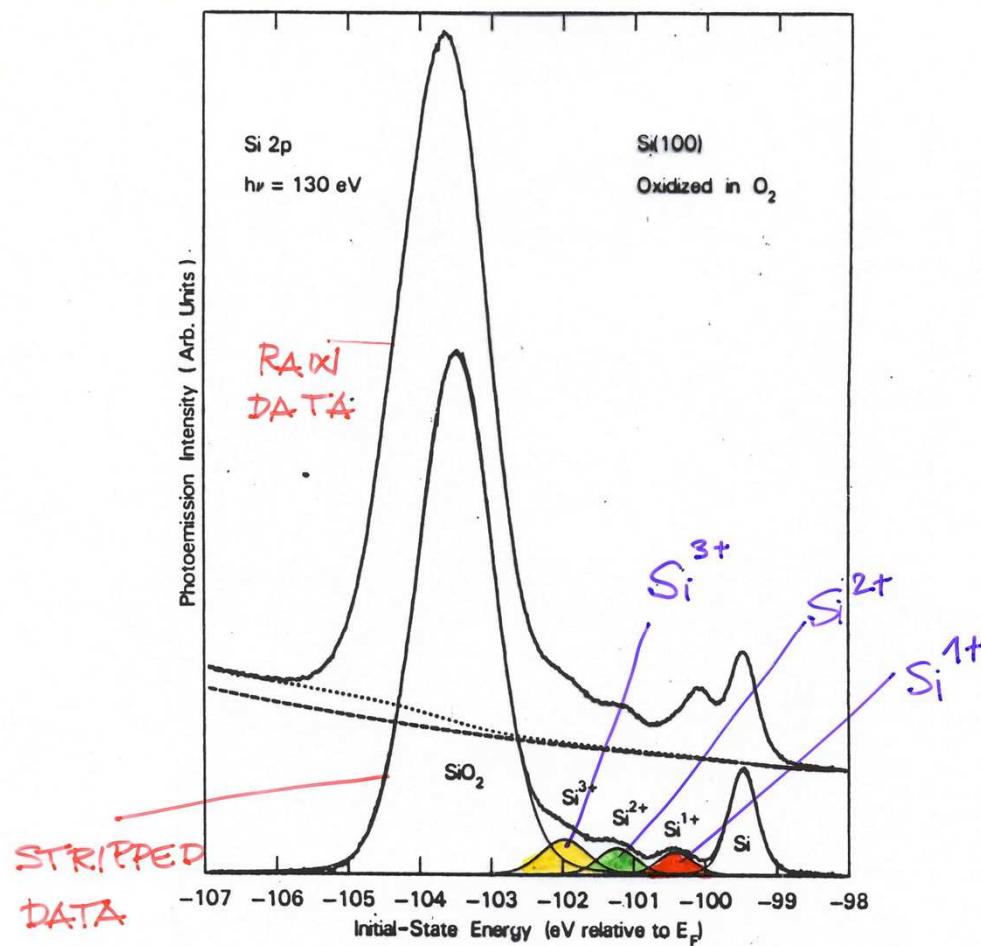


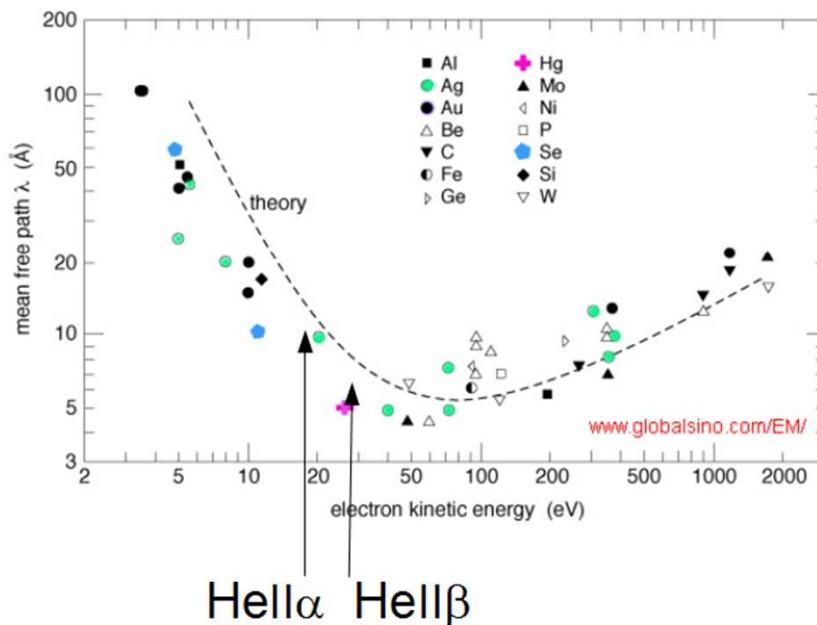
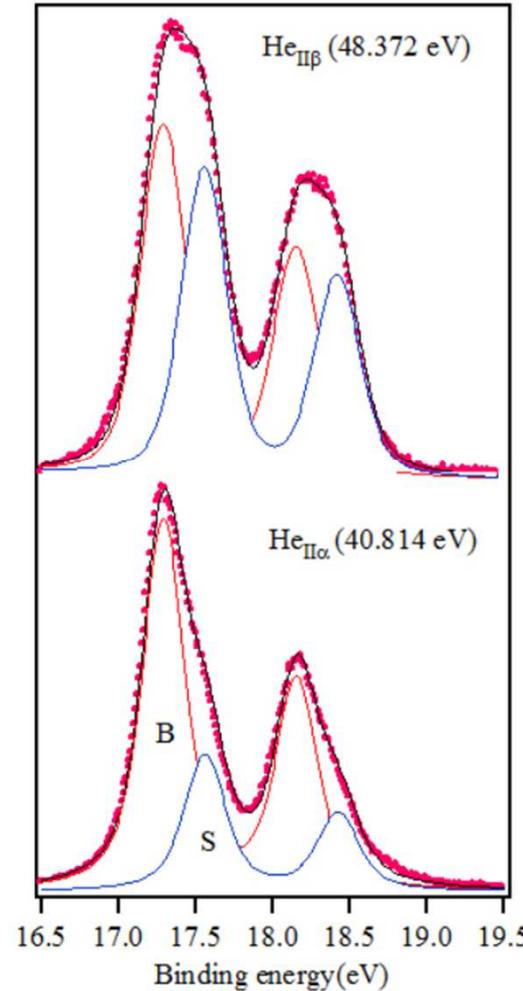
FIG. 1: Intermediate-oxidation states at the $\text{SiO}_2/\text{Si}(100)$ interface, identified by their $\text{Si} 2p$ core-level shifts. The top curve represents the raw photoemission data for the $\text{Si} 2p_{1/2,3/2}$ core levels. The bottom curve has the $\text{Si} 2p_{1/2}$ line and the secondary electron background subtracted. All three intermediate-oxidation states are seen. For a truncated bulk structure only Si^{2+} would be present since the $\text{Si}(100)$ surface has two broken bonds per atom.

Core Levels, chemical shift

The $\text{Si} 2p$ line is characterized by the occurrence of 5 chemically distinct components which reflect different chemical states of the Si atoms at the interface

Surface core level shift vs. mean free path In 4d

In 4d core-level



High-resolution In-4d core-levels at freshly cleaved InAs(110), taken with He $\text{II}\alpha$ and He $\text{II}\beta$ radiation; Voigt-profiled fit with surface (S, blue lines) and bulk (B, red lines) doublet components (left panel)

Photoemission Spectroscopy: Core Levels

Spin-Orbit Splitting

(iii) Degeneracies determine relative intensities of peaks comprising doublet

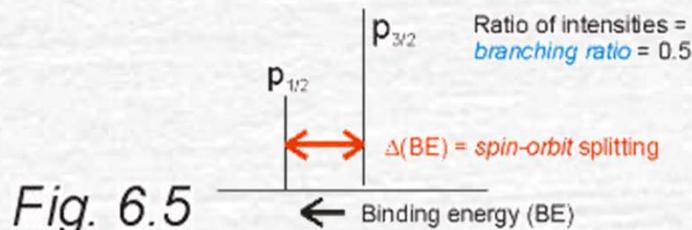


Fig. 6.5

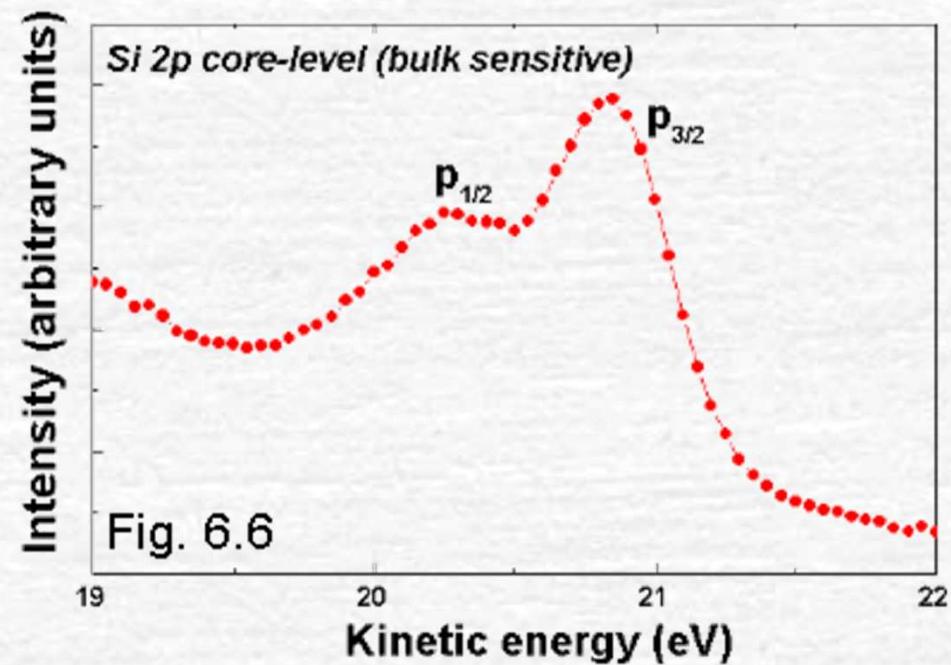
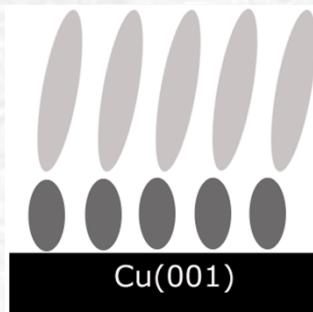


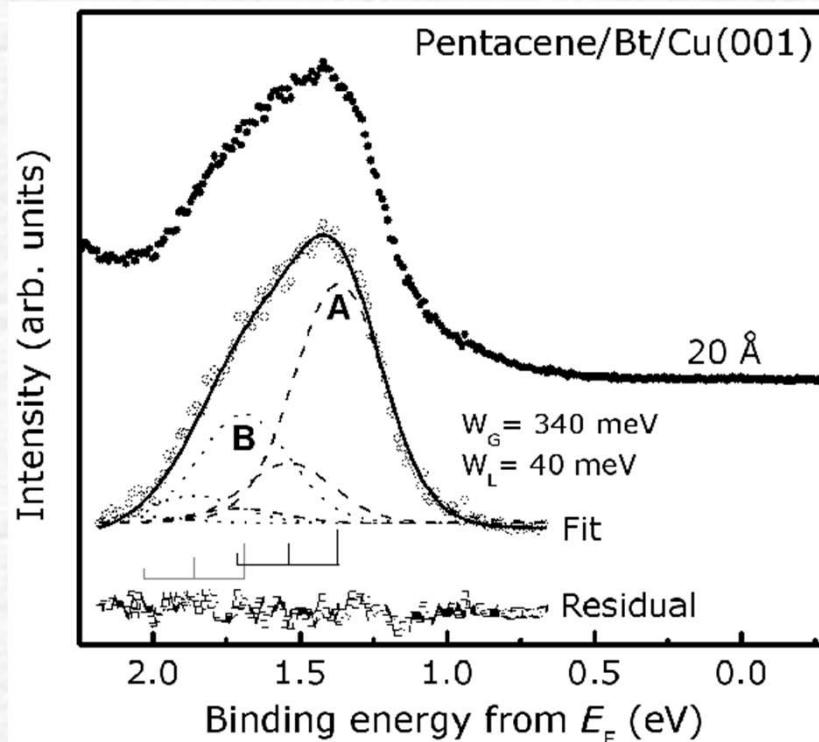
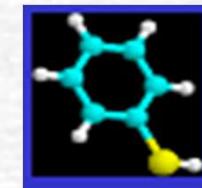
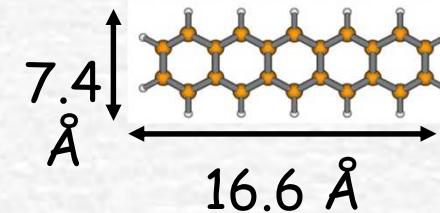
Fig. 6.6

valence PE vibrational spectrum



pentacene: $C_{22}H_{14}$

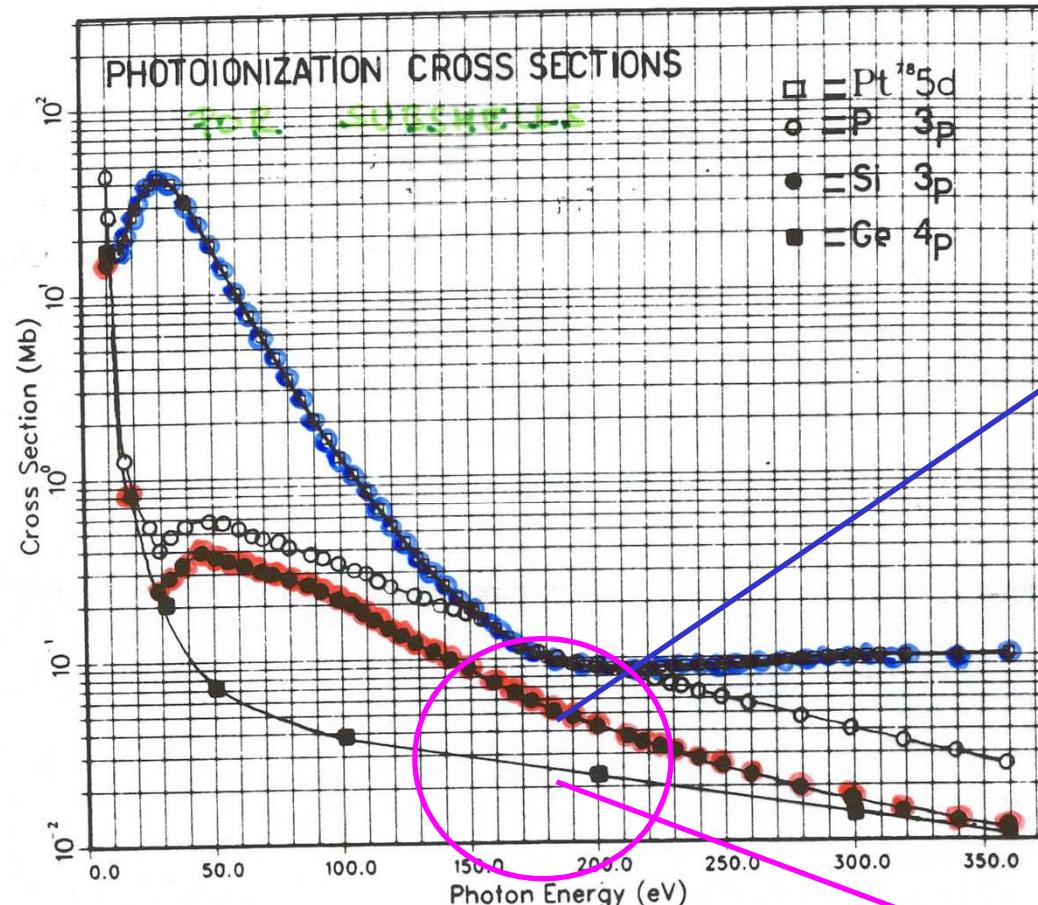
benzene-thiol: C_6H_5-SH



Betti, Kanjilal and Mariani, J. Phys. Chem. A **111**, 12454 (2007)

Cooper Minimum Photoemission

It is possible when one of the valence band orbital shows a Cooper minimum in the photoionization cross section



Photoionization cross section for 4d and 5d subshells in the energy range 0–200 eV compared to the cross sections for the 3ps and 4sp valence states of the semiconductors [21].

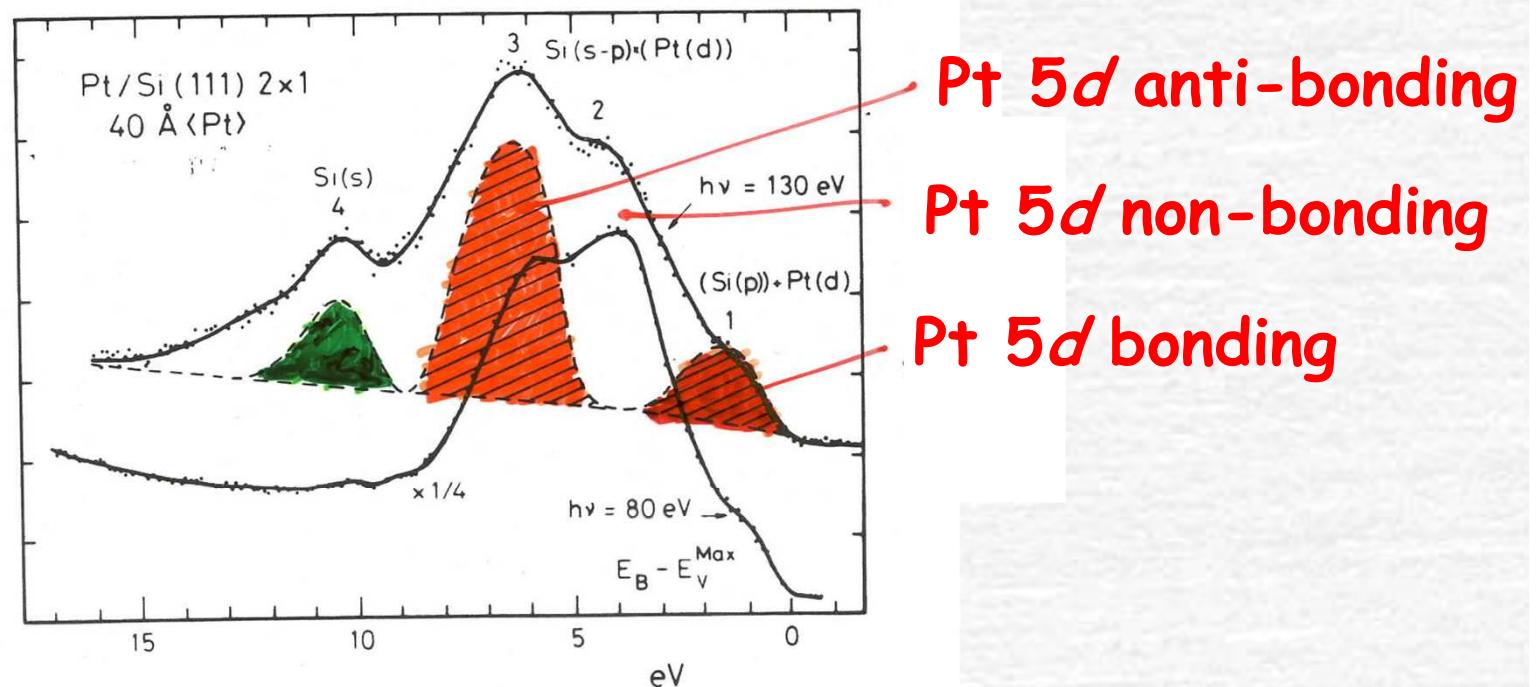
Cooper minimum
in the Pt 5d
cross section

A Cooper minimum exists when the radial part of the orbital wave function exhibits a node

The Pt 5d and Si 3p cross sections are comparable

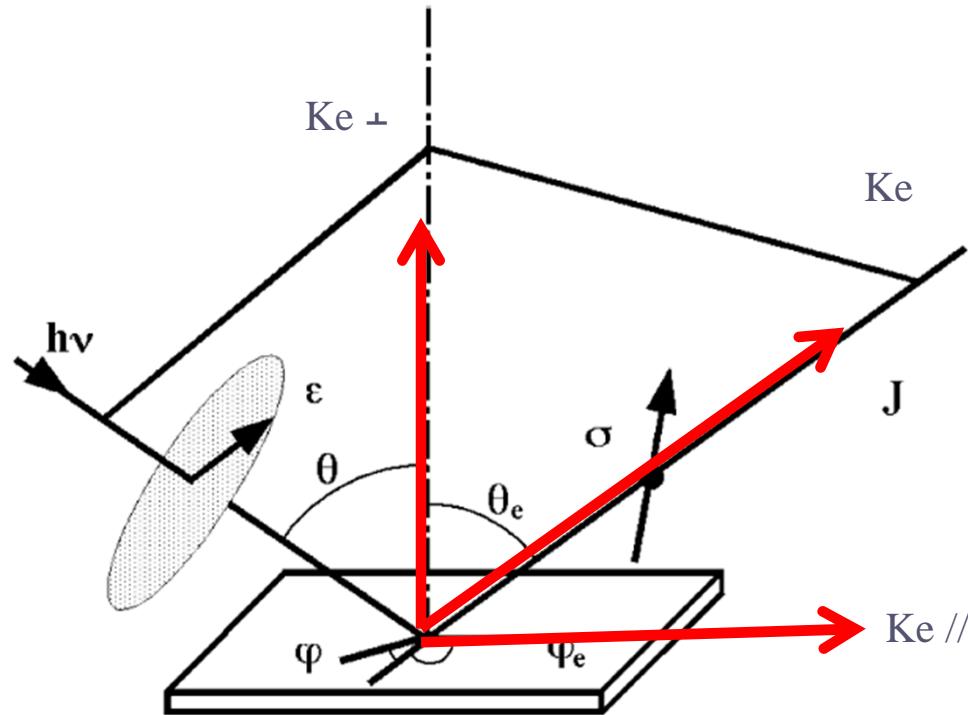
Cooper Minimum Photoemission

A joint analysis of VB photoemission spectra taken at and off the Cooper minimum enables one to disentangle the differing site- and orbital-specific contributions



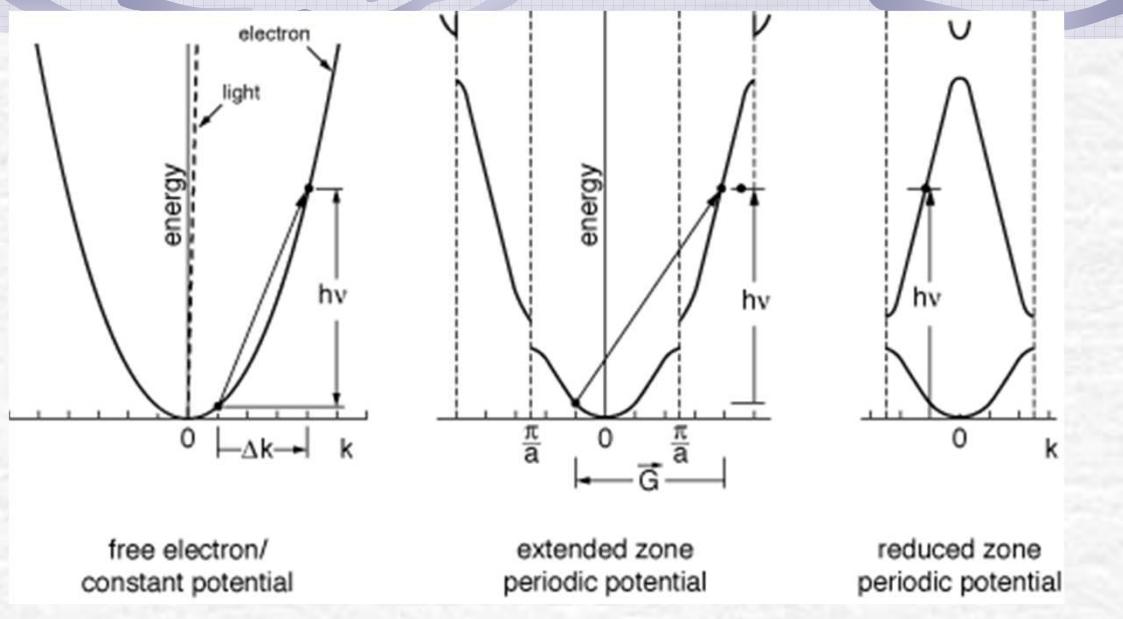
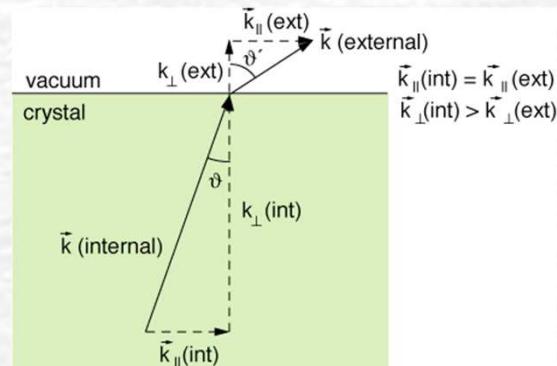
Analysis of the Si sp partial DOS at the Pt-Si(111) reacted interface (40 \AA Pt-Si(111) at room temperature). The top panel displays the CM and the $h\nu = 80 \text{ eV}$ photoemission data, and a three-peak partial DOS that accounts for the Si hybridized 3sp charge at the interface; a gap is present in correspondence to the localized Pt 5d states. The same three-peak partial DOS is then self-convoluted and compared to the integrated Si L_{2,3}VV lineshape. The correspondence of all peaks and relative intensities (a part of the known reduction of the Si 3s contribution) confirms the CM derivation of the Si sp partial DOS [159].

Angular resolved photoemission

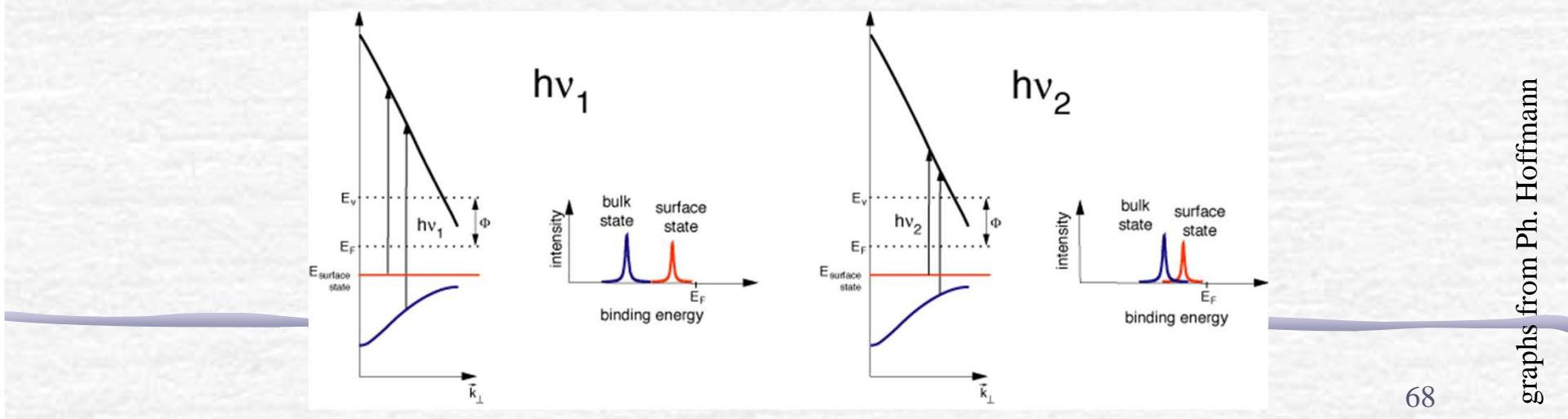


angular resolved photoemission

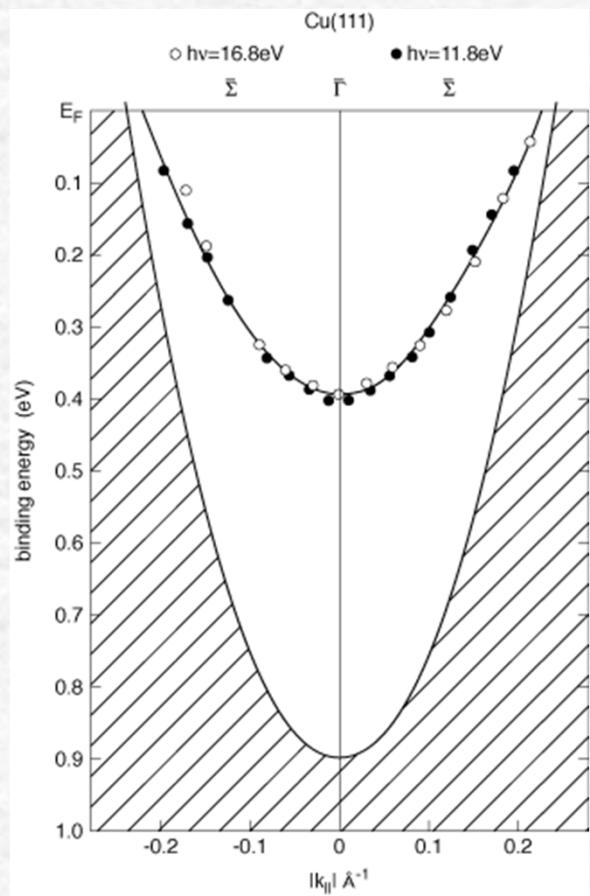
How to photo-excite electrons from a crystal in a periodic potential, by photoemission



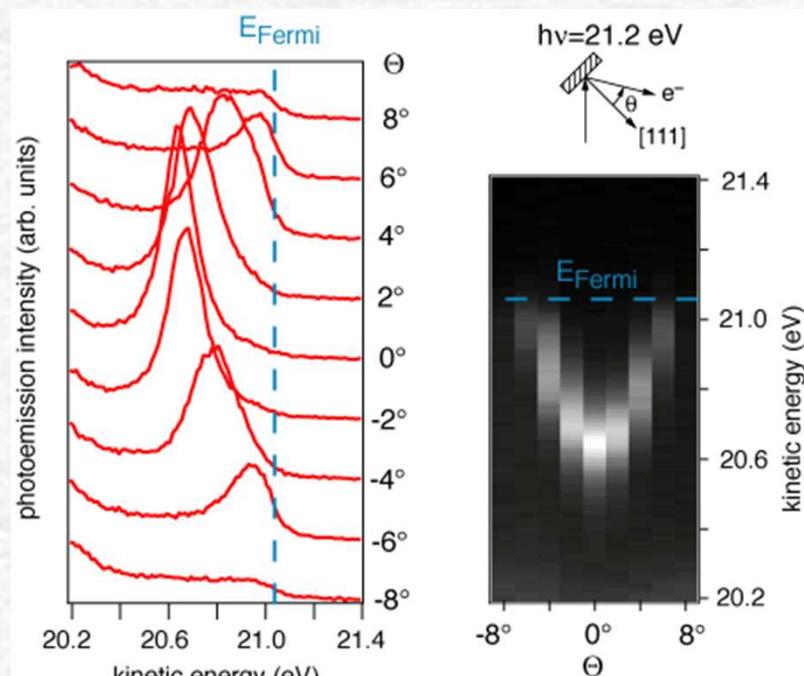
$$\vec{k}_{||i} = \vec{k}_{||f} = \sin(\Theta) \sqrt{\frac{2m}{\hbar^2}} \sqrt{E_{kin}} = \sin(\Theta) \sqrt{\frac{2m}{\hbar^2}} \sqrt{h\nu - E_{bin} - e\Phi}$$



electronic surface states at Cu(111)

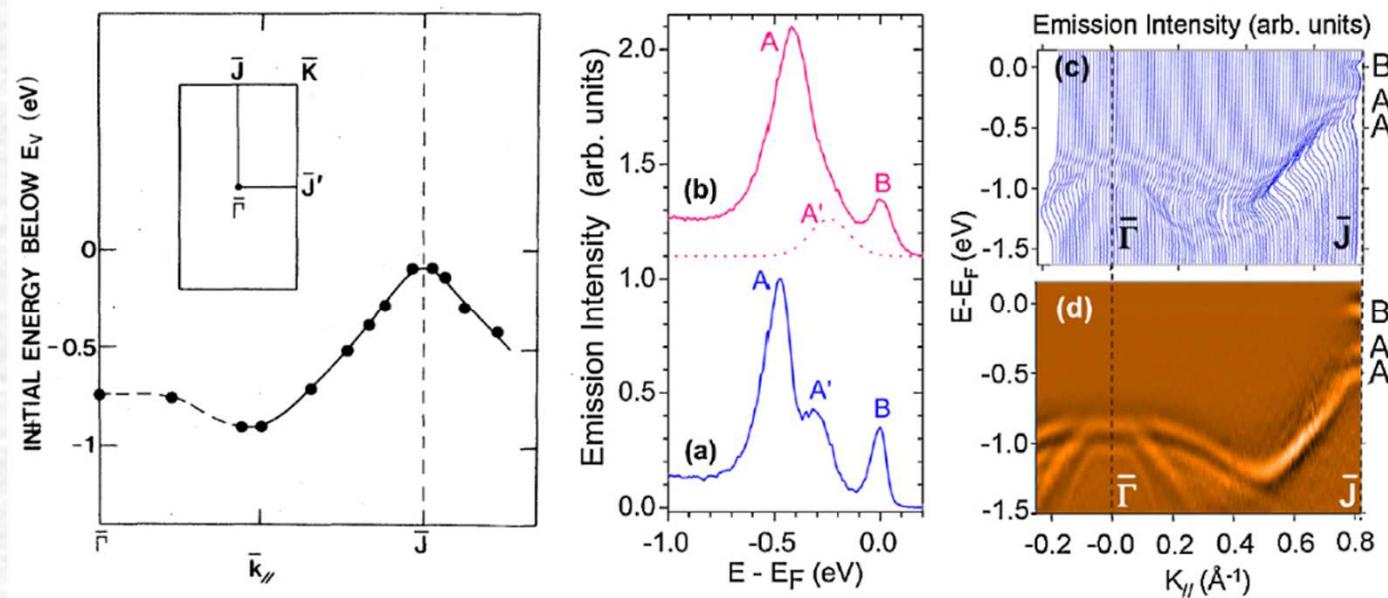


S.D. Kevan, Phys. Rev. Lett. 50, 526 (1983).



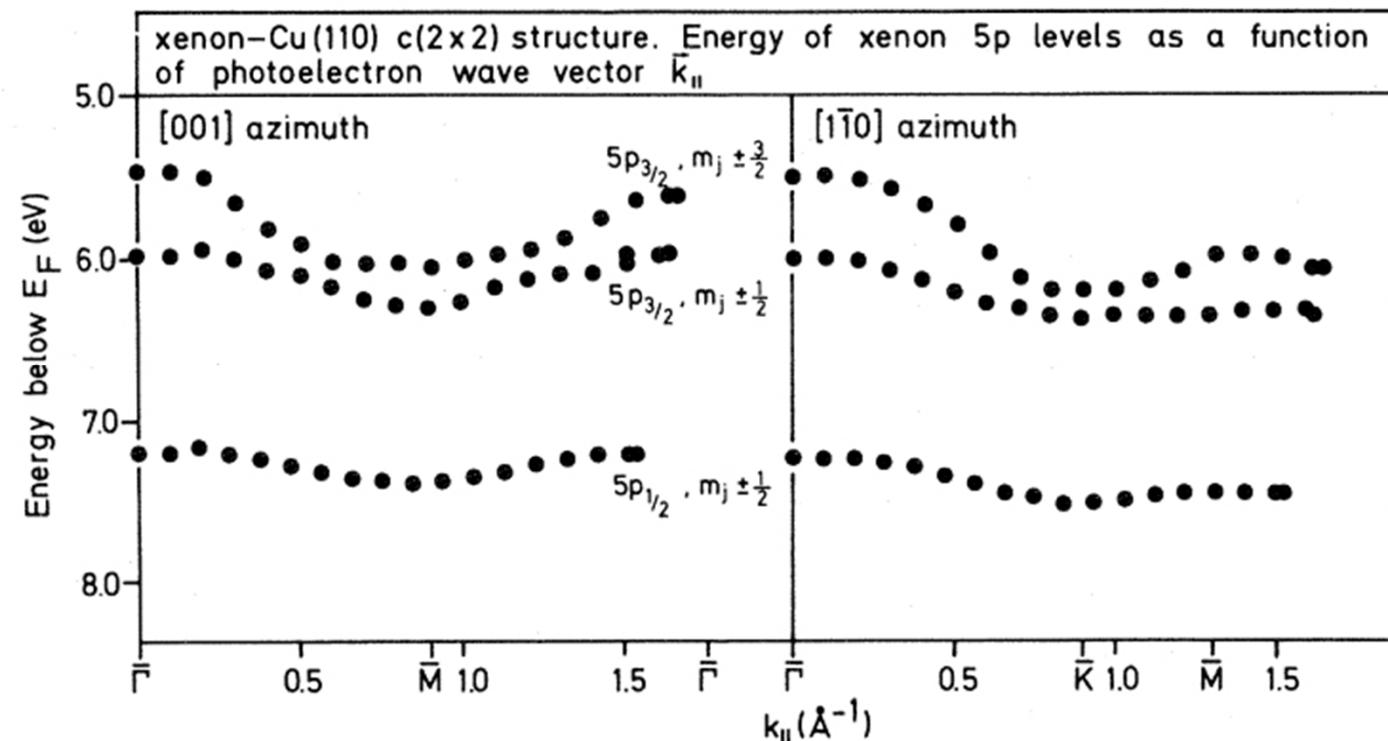
quasi-free electron surface state on Cu(111),
Schockley state, *s-like*

Dangling bonds Si(111)-(2x1)



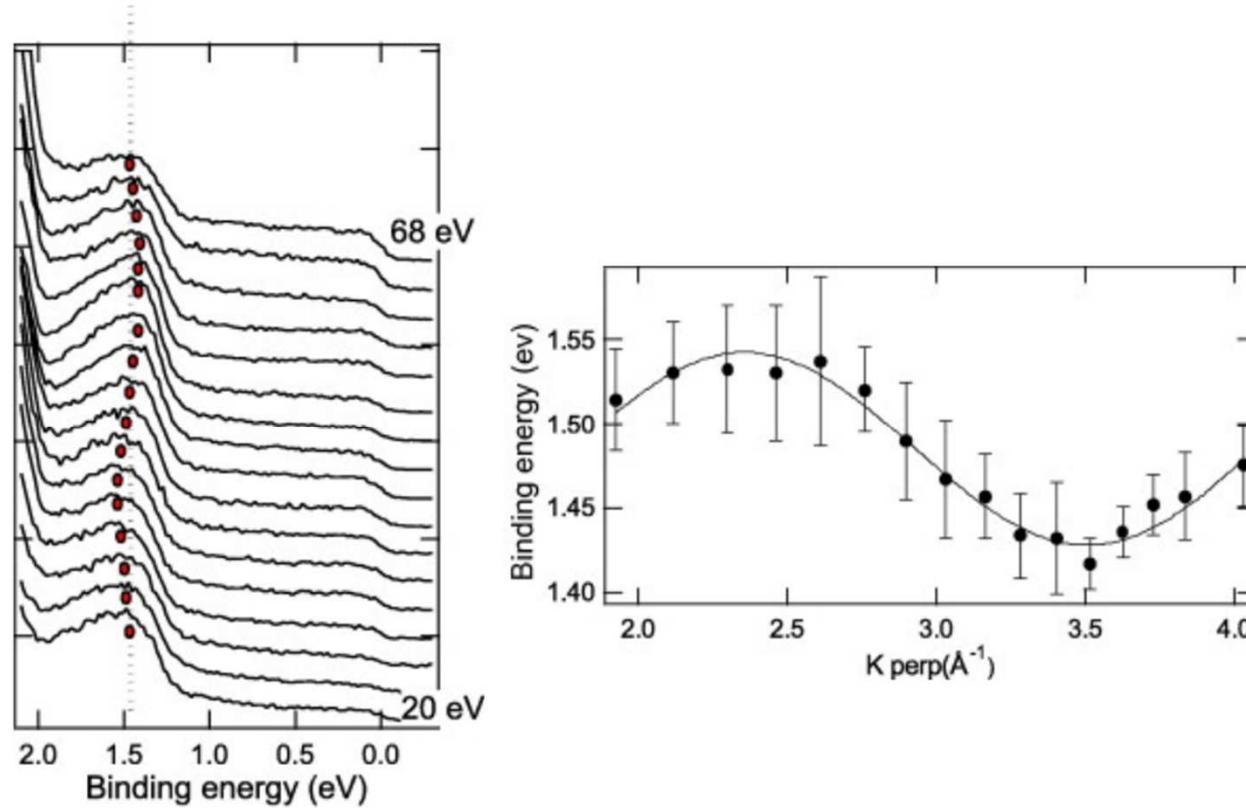
Dangling-bond surface state dispersion at the Si(111)-(2x1) reconstructed surface along the Γ -J direction of the Surface Brillouin Zone (SBZ). One of the first experimental ARPES dangling-bond dispersion (left panel); recent high-resolution ARPES dangling-bond dispersion.

Physisorbed Xe c(2x2)/Cu(110)



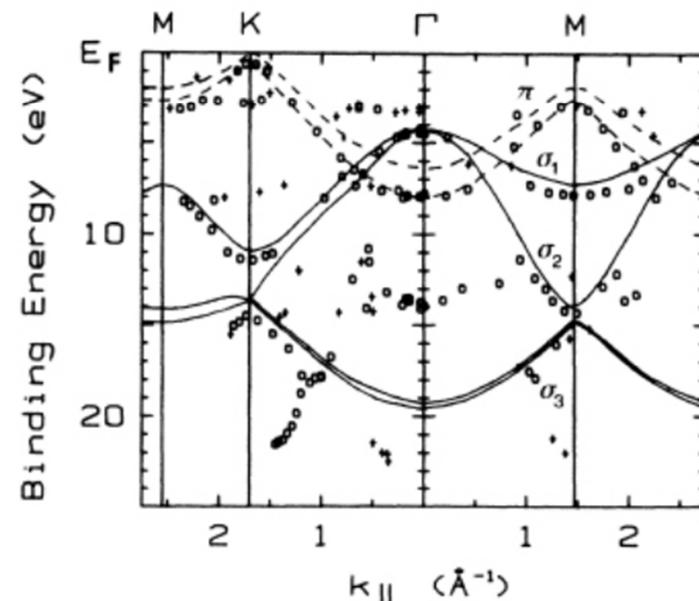
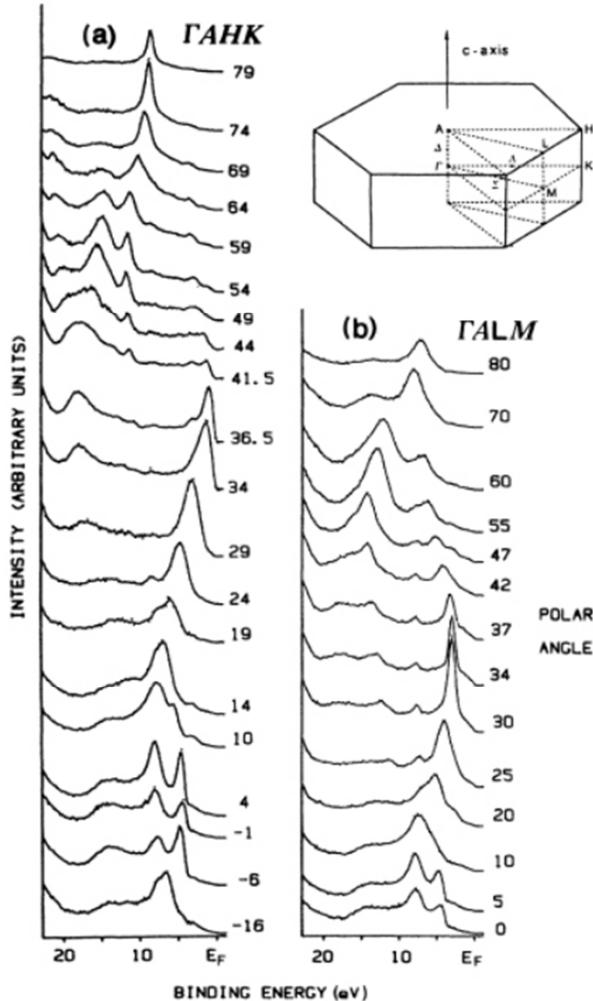
Experimental band structure of the 5p levels of Xe physisorbed in an ordered c(2×2) structure onto the Cu(110) surface. ARPES bands

Pentacene HOMO on Cu(119)



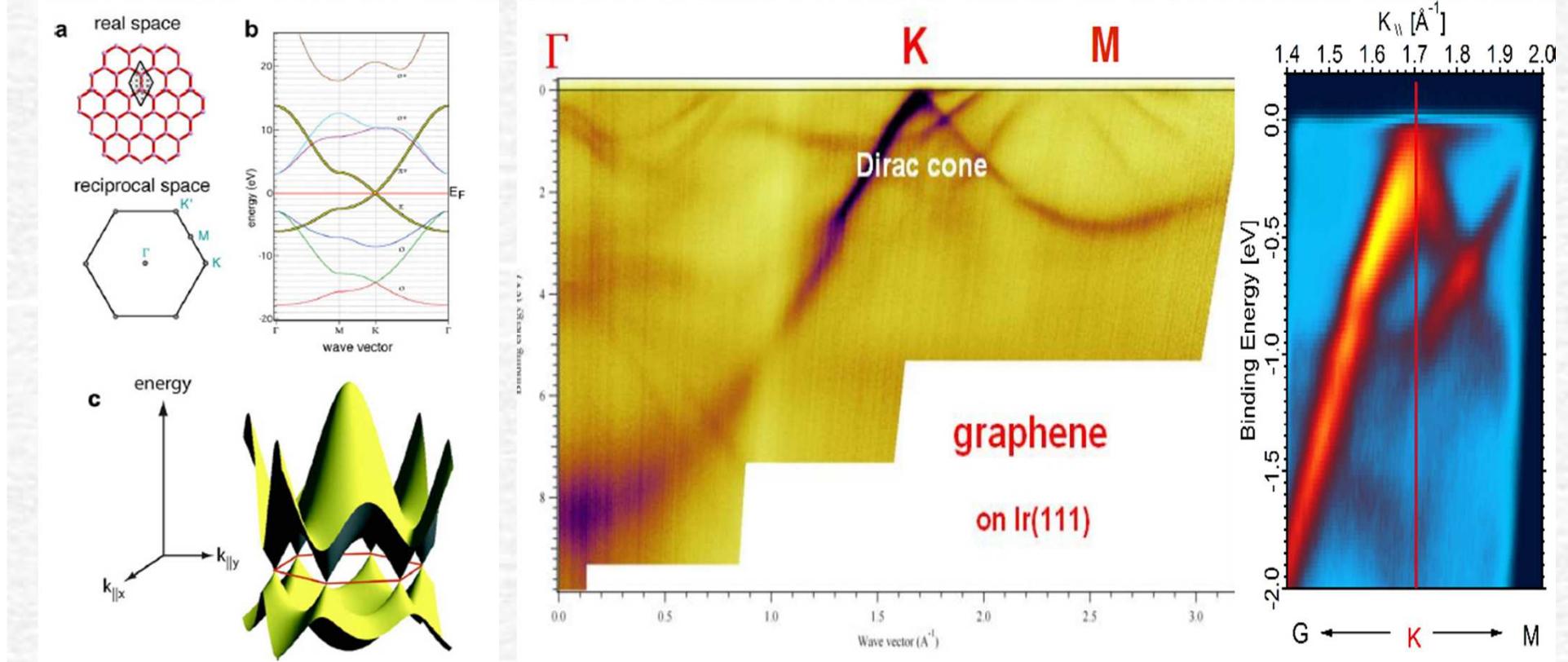
2-nm thick pentacene film grown on Cu(119). ARPES selection of spectra taken at normal emission and varying the photon energy (left); highest-occupied molecular-orbital (HOMO) band dispersion along k_{\perp} (right).

ARPES graphite (HOPG)



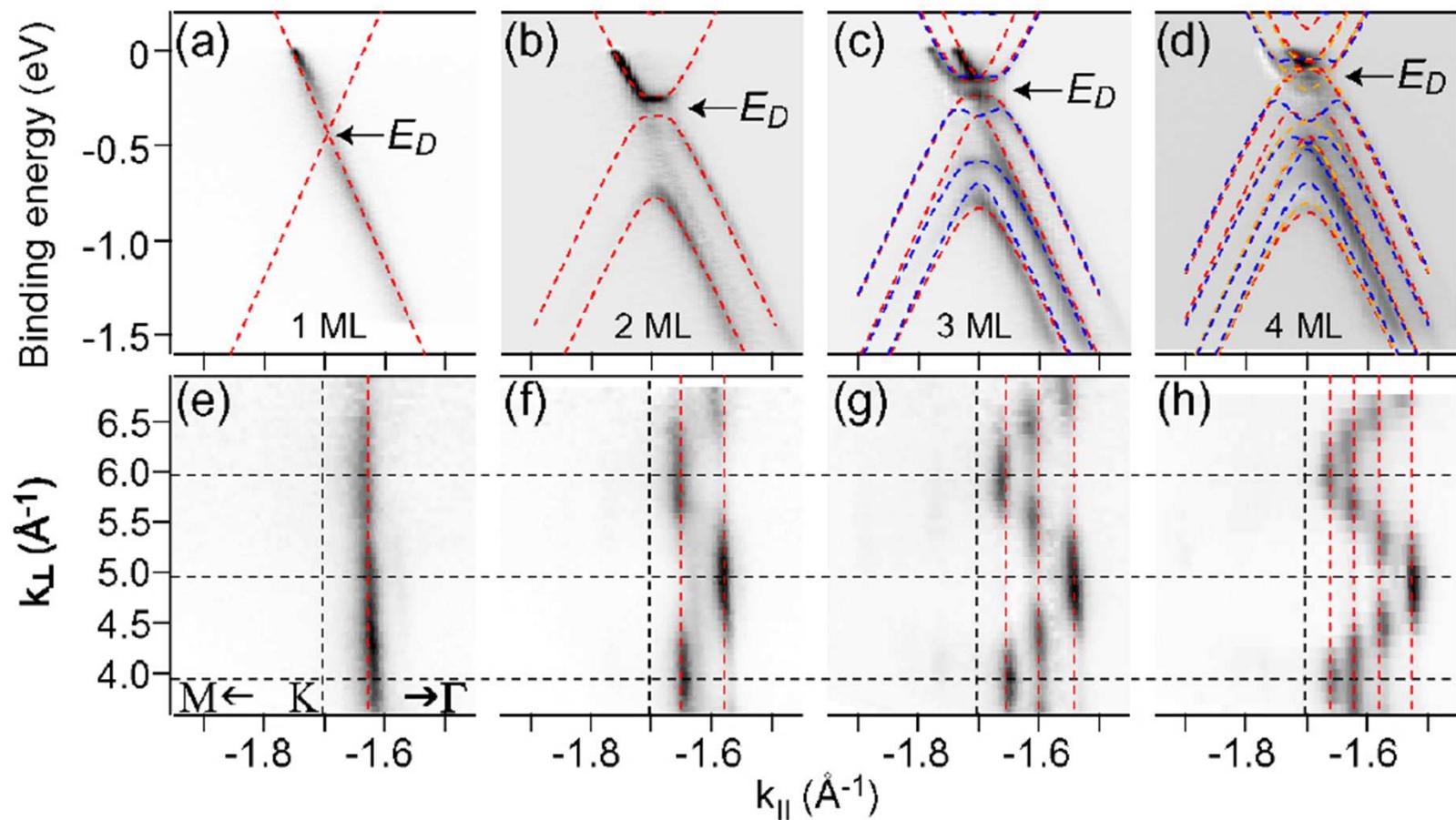
Valence band of graphite (HOPG), stacking of the ARPES spectra as a function of polar angle (left) and experimental band structure (right).

Graphene band structure



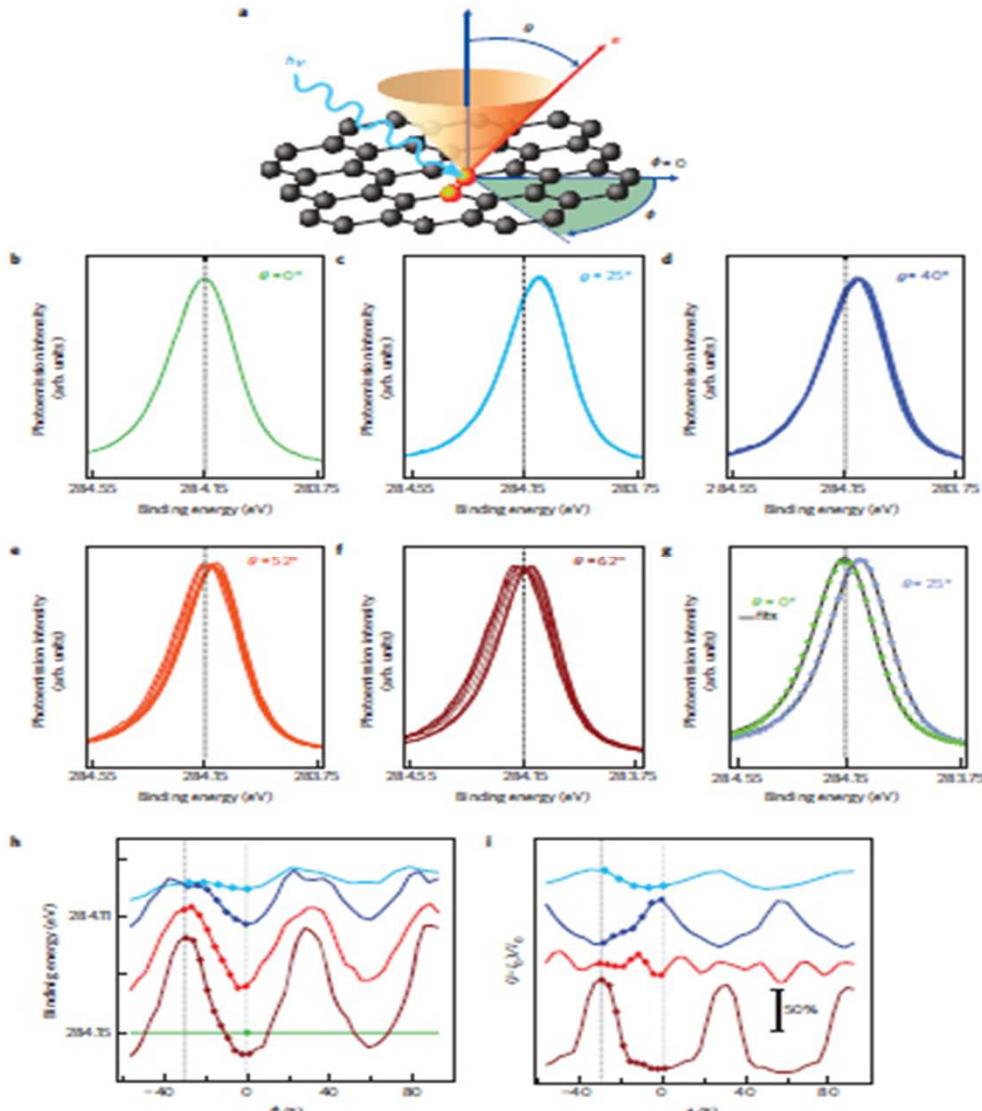
Graphene band structure along GKM and zoom of the Dirac cone around the K point of the SBZ. ARPES data taken with high-resolution ARPES and a He discharge source

Band formation in graphene multilayers



- . Formation of an electronic band, stepwise: from 1-layer (extreme left) to 4-layer (extreme right) graphene band structure along across the Dirac point.

K shell dispersion in graphene



Spectral function of the C 1s core-level in graphene as a function of the emission polar angle

Silvano Lizzit, et al.: Nature Physics **6**, 345-349 (2010)



The End

References

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- ❖ S. Hufner “Photoelectron Spectroscopy, principle and applications” (Berlin Springer 2003) 3rd Edition
- ❖ V. Schmidt “Photoionization of atoms using synchrotron radiation” Report on Progress in Physics 55(1992)1482
- ❖ C.M. Bertoni in “Synchrotron Radiation Basics, Methods and Applications (Springer Verlag Berlin Heidelberg 2015, pg. 145)
- ❖ C. Mariani and G. Stefani in “Synchrotron Radiation Basics, Methods and Applications (Springer Verlag Berlin Heidelberg 2015, pg. 275)