PAUL SCHERRER INSTITUT

Rasmus Ischebeck

Interaction of X-Rays with Matter

Joint Universities Accelerator School

Quiz: Which Components Primarily Determine the Interaction of X-Rays with Matter?

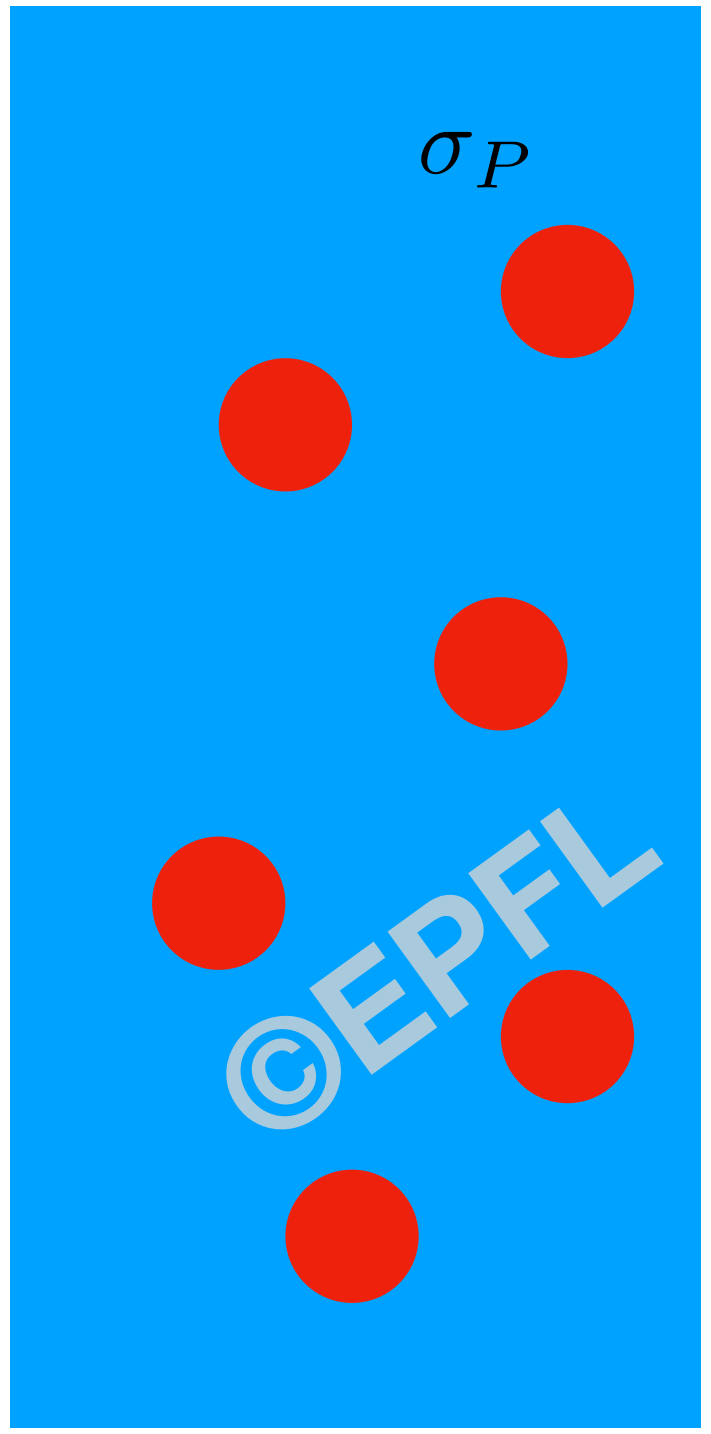
Protons

Electrons

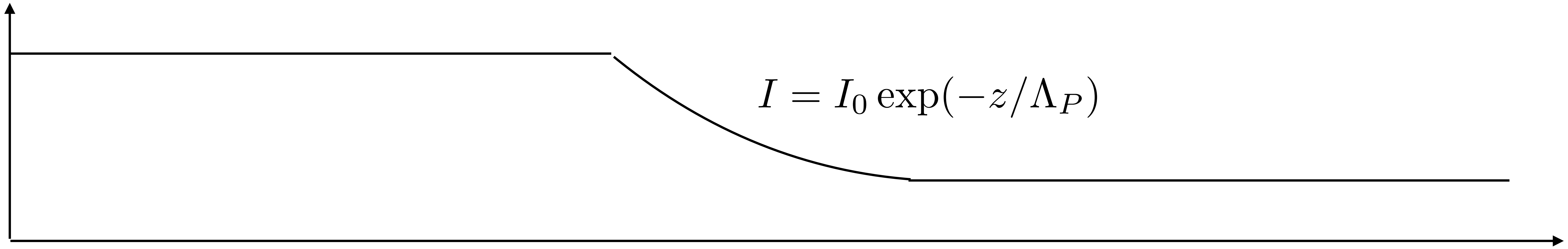
Neutrons

Klingons

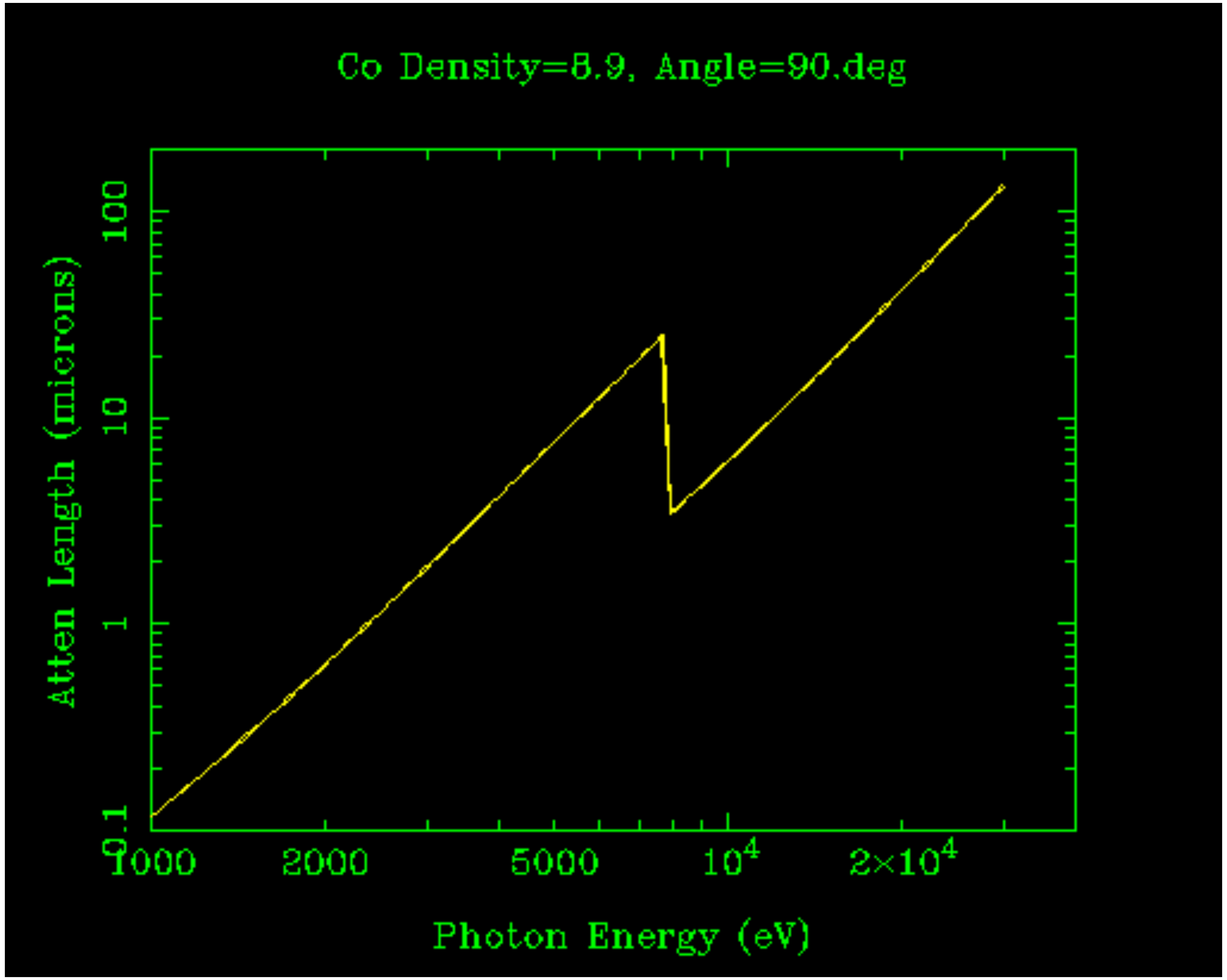
Cross Section



$$\Lambda_P = \frac{1}{\sigma_P \rho_a}$$



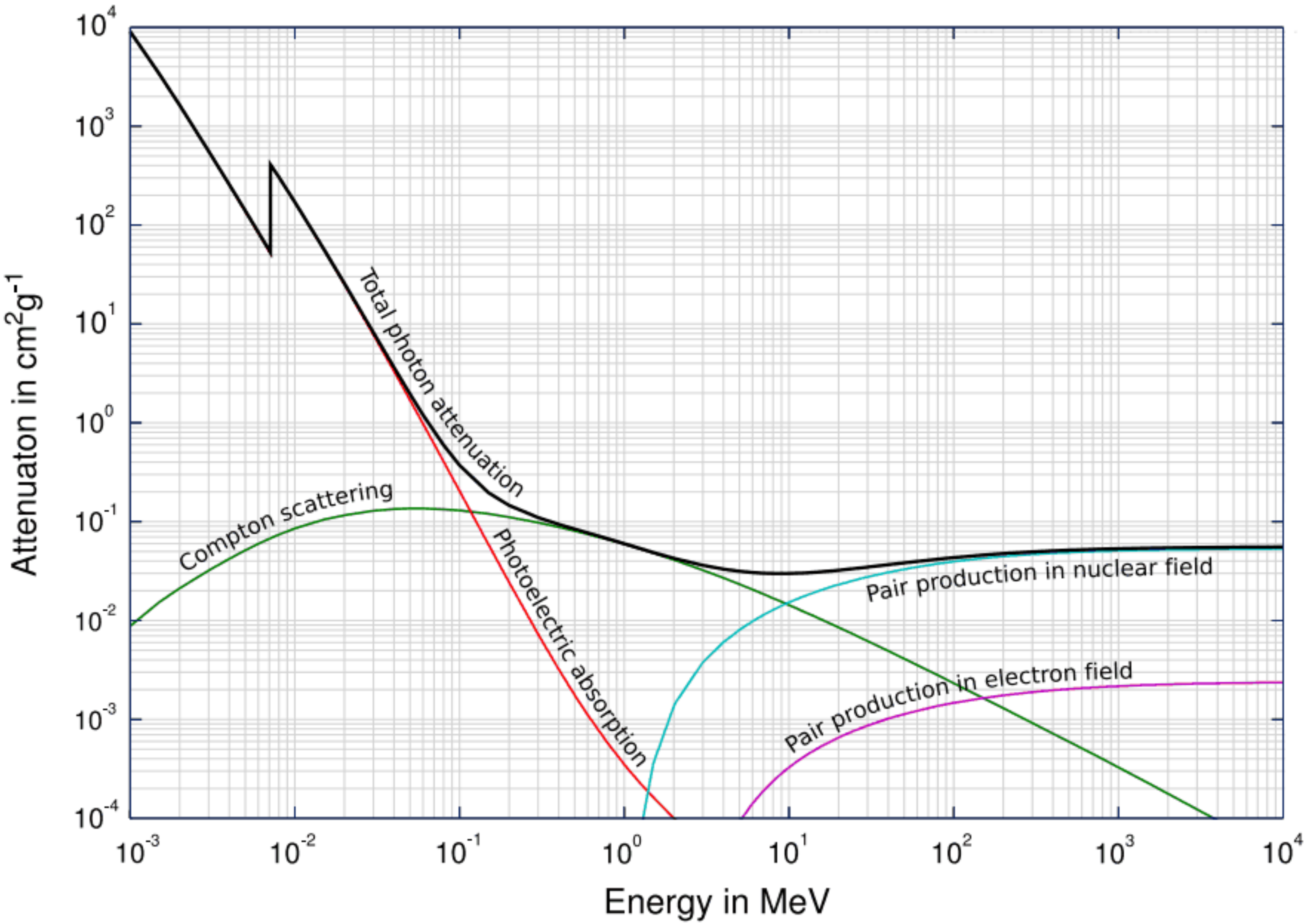
Cobalt Metal



$$\rho_a = 9.0943 \cdot 10^{10}/\mu\text{m}$$

http://henke.lbl.gov/cgi-bin/atten.pl

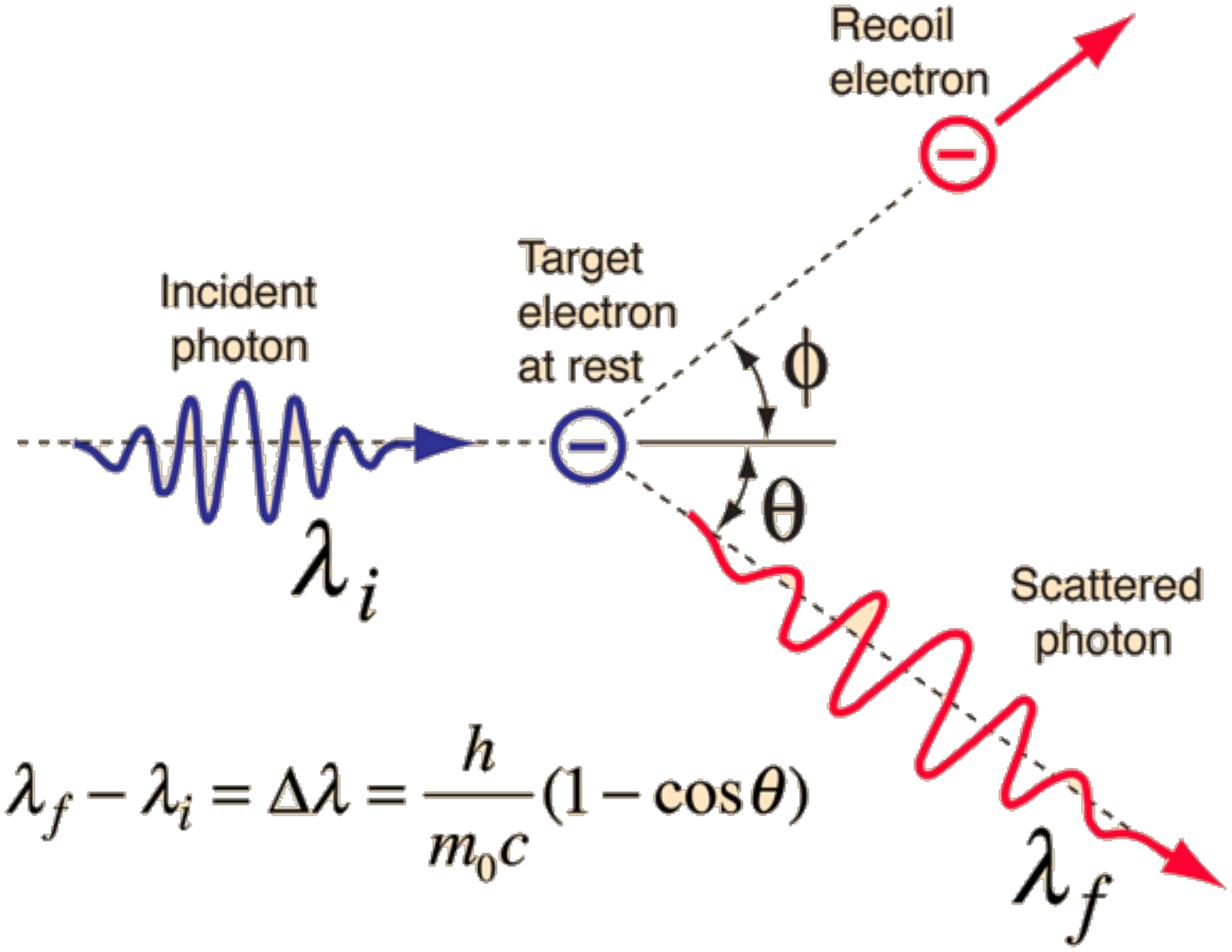
What Processes Contribute to X-Ray Cross Section?



Quiz: A photon undergoes an elastic collision. What does this mean? More than one answer possible.

- The photon has the same energy before and after collision
- The photon has the same momentum vector before and after collision
- The photon has the same wavelength before and after collision
- Complete transfer of the photon's energy to the collision partner

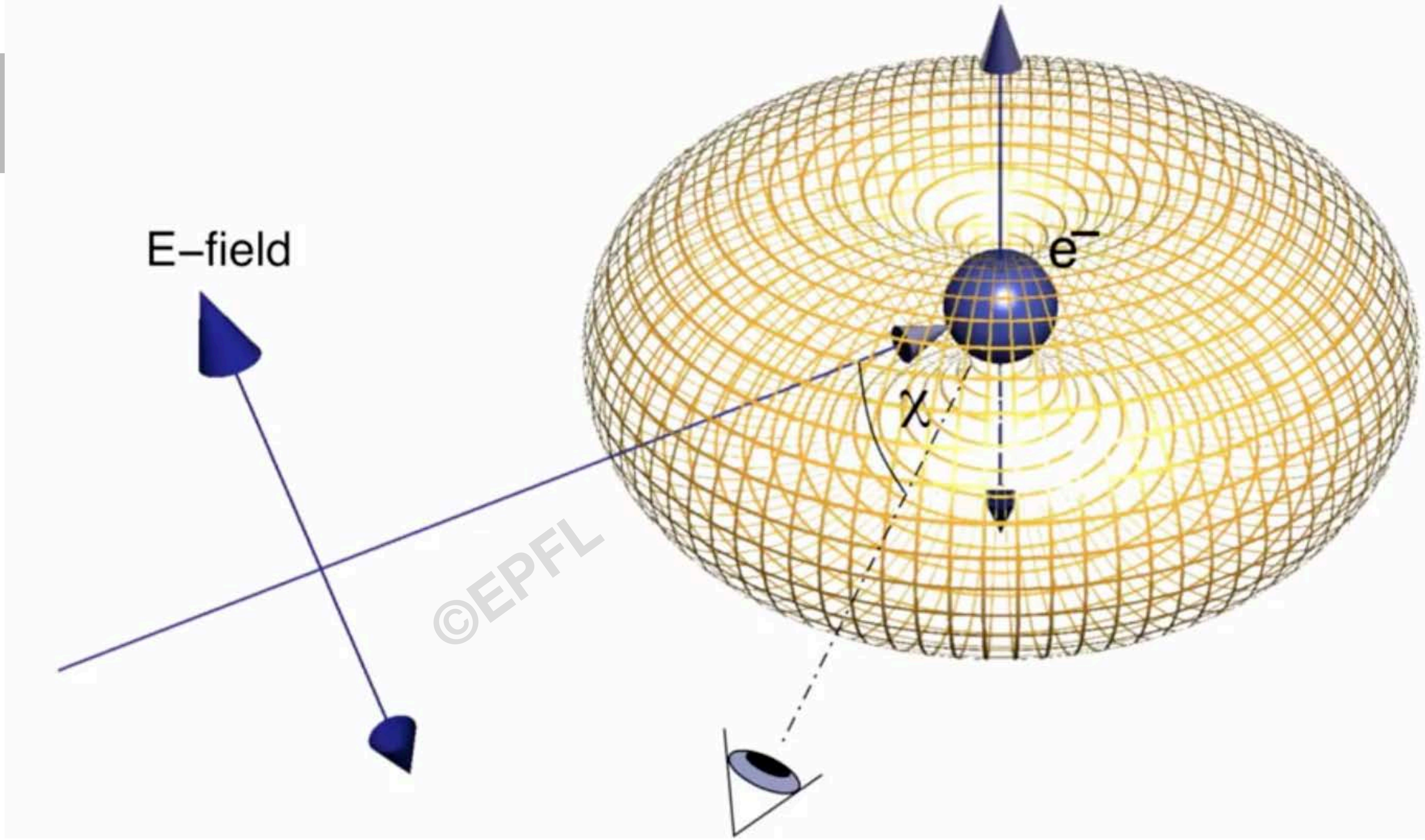
Compton Scattering



$$\lambda_f - \lambda_i = \Delta\lambda = \frac{h}{m_0 c}(1-\cos\theta)$$

http://hyperphysics.phy-astr.gsu.edu/hbase/quantum/compton.html

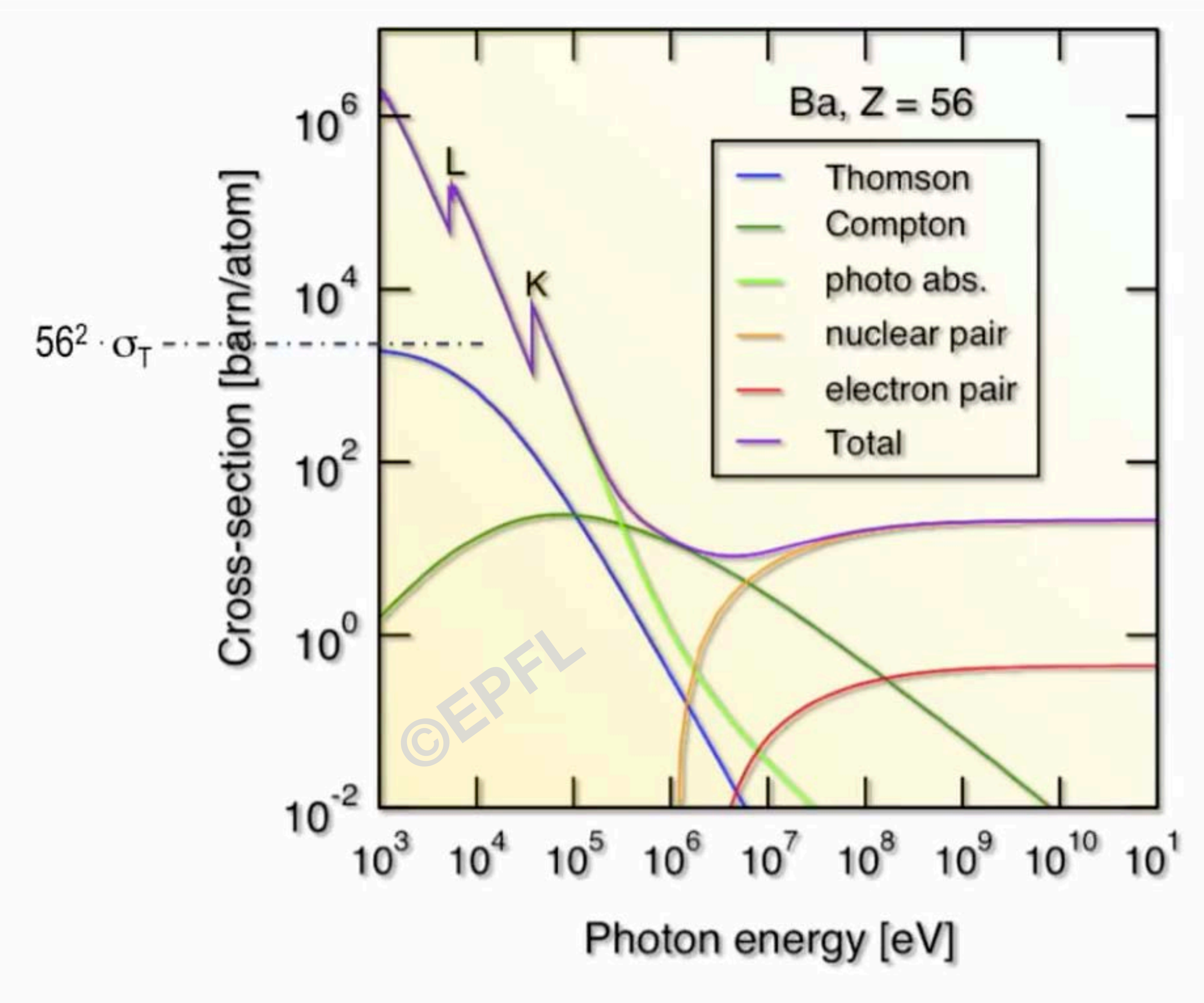
Thomson Scattering: Elastic Scattering on Free Electrons



$I = I_0 \cos^2\chi$

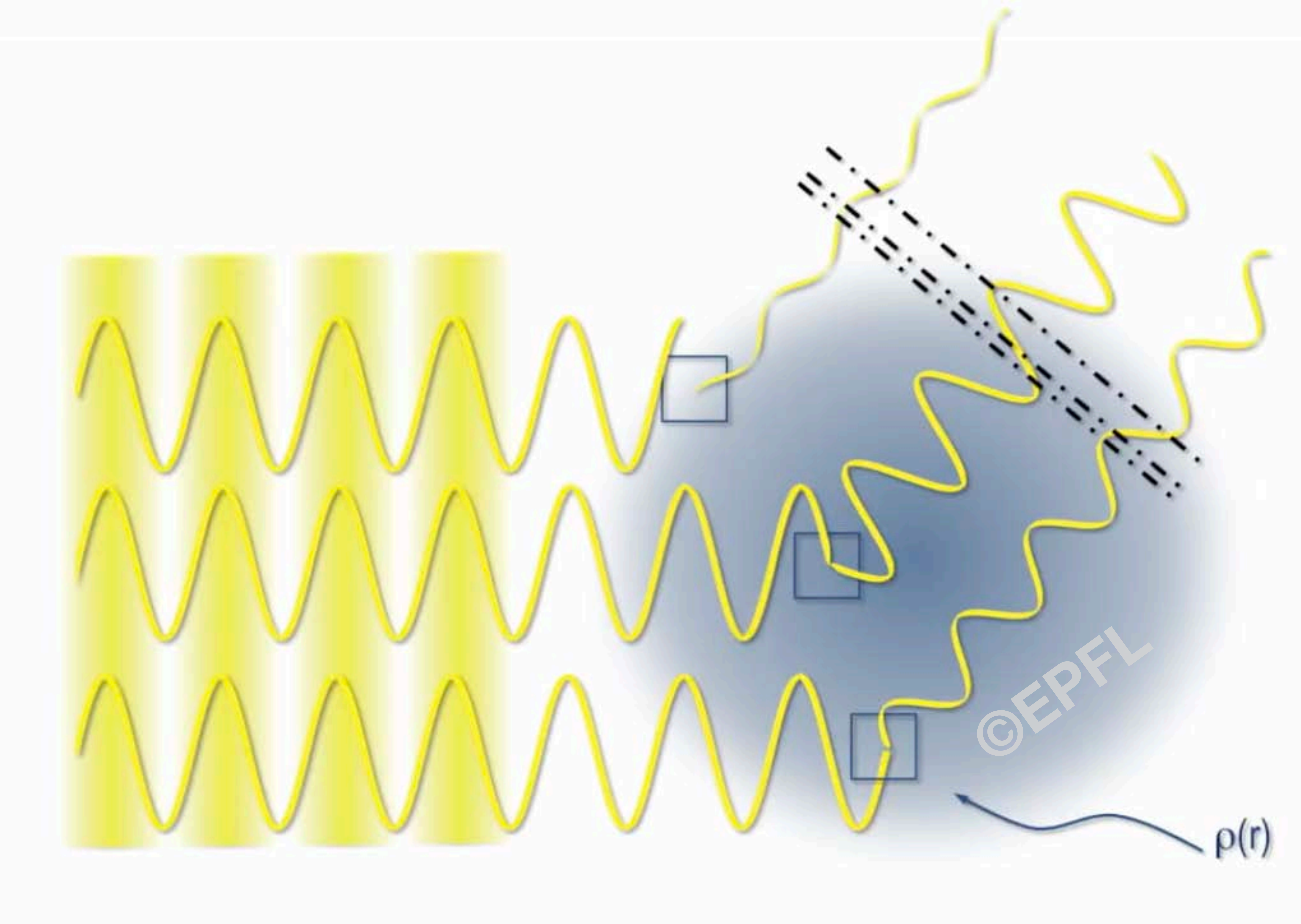
Absorption Cross Section

$$\sigma = Z^2 \sigma_T = Z^2 \frac{8\pi r_0^2}{3}$$



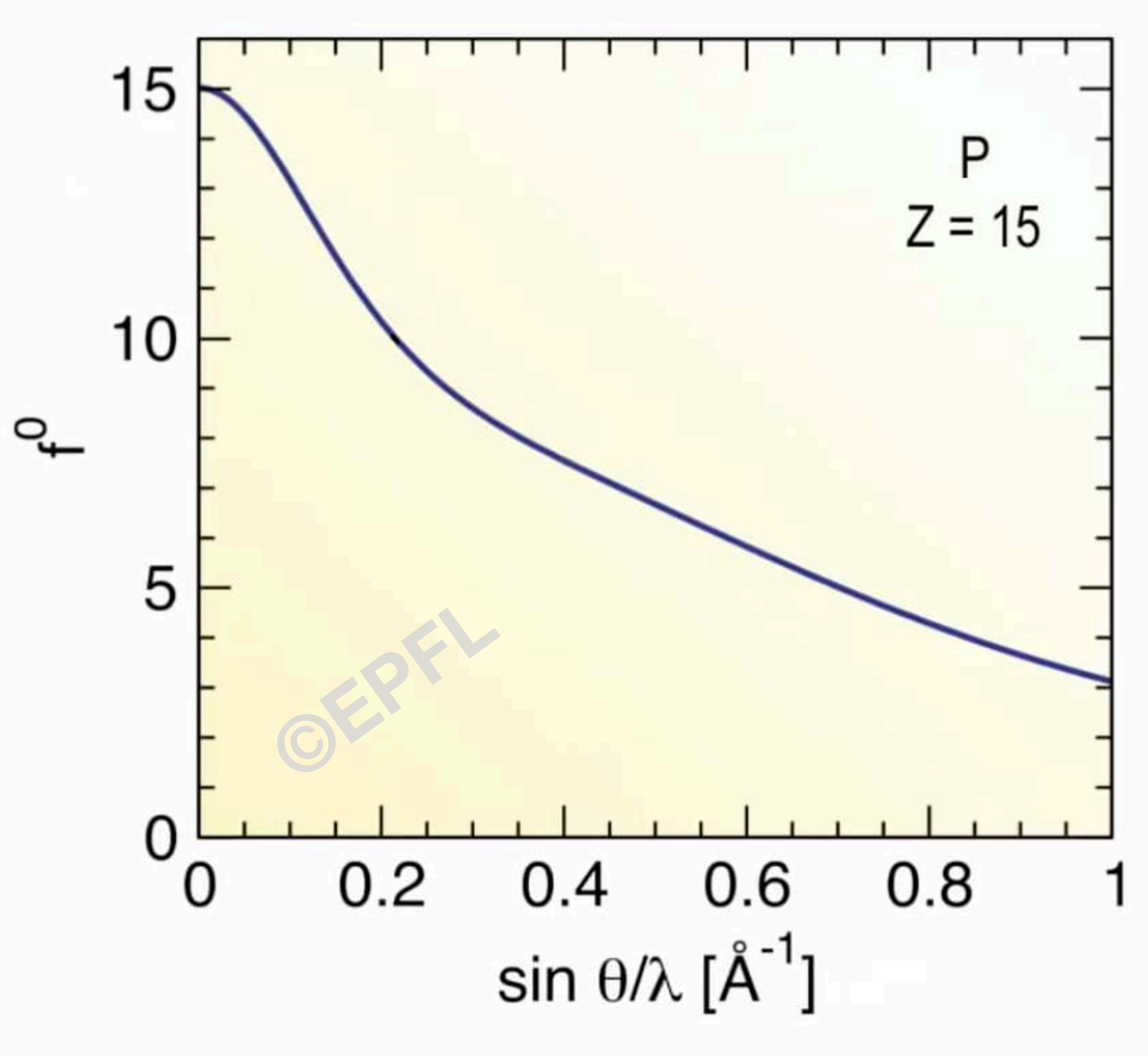
Phil Willmott

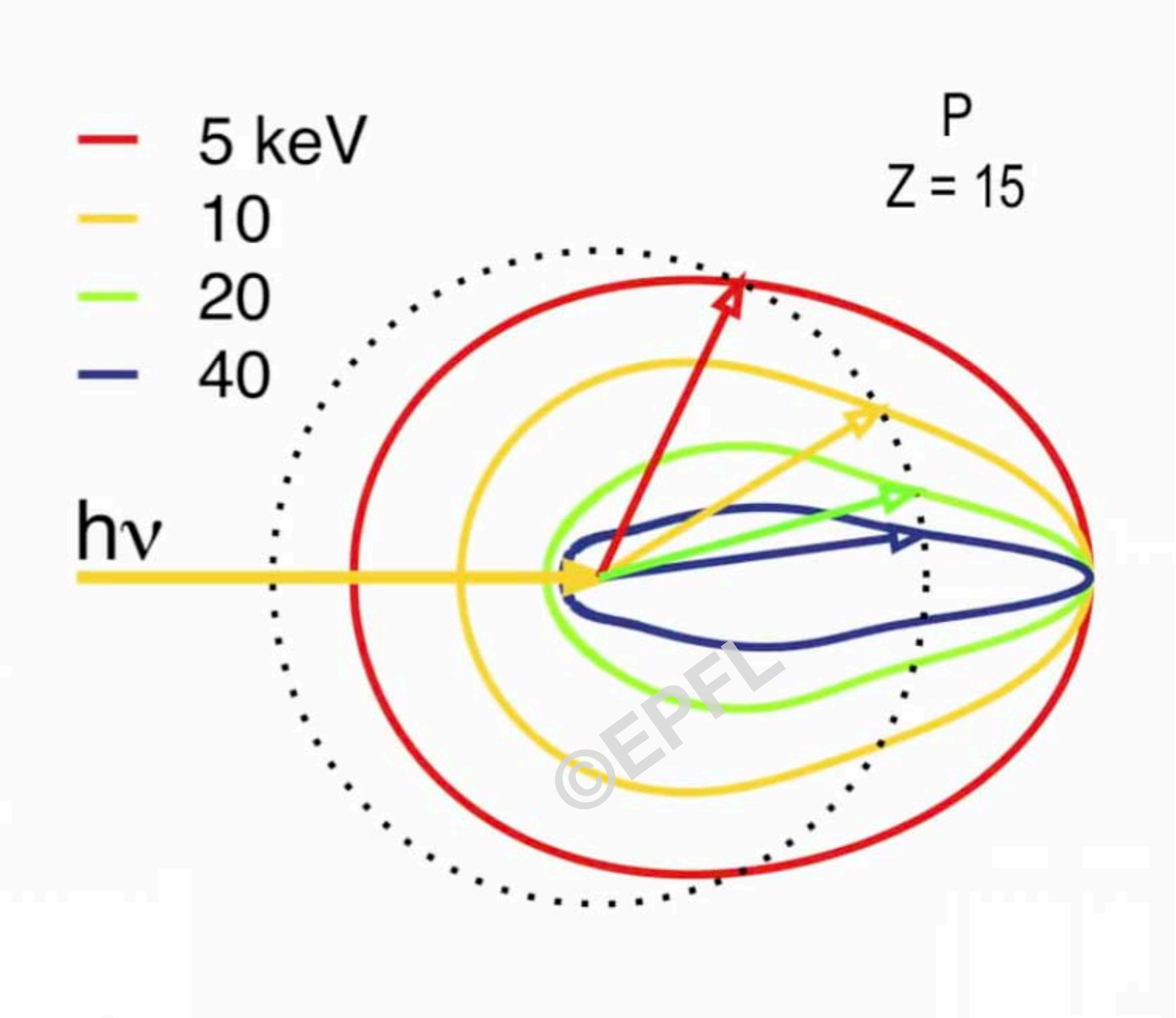
Absorption by Higher Energy Photons



Phil Willmott

Form Factor





Phil Willmott

PAUL SCHERRER INSTITUT

Parametrization of the Form Factor

$$f^0(\sin\,\theta/\lambda) = \sum_{i=1}^{4} a_i \exp(-b_i \sin^2\theta/\lambda^2) + c$$

International Tables for Crystallography

Summary: Elastic Scattering

- The electrons respond to the electric-field component of the incident x-rays and reradiate as a consequence at the same frequency - the process is elastic.
- At large wavelengths compared to the size of the atoms scattering is independent of the photon energy, as the cloud can be approximated as being a point source.
- However, as the wavelength becomes smaller (photon energy increases), scattering from the different volume elements within the electron cloud begin to be out of phase with each other, resulting in increasing destructive interference.
- As an approximation, the cross-section drops off as 1/E^2, and is described in detail by the atomic form factor, f, which is normally expressed as a function of Q, or sin theta/lambda.

Electrons Bound to an Atom

Wikimedia Commons

Quiz: In Which Element are the Core Electrons Most Strongly Bound to the Atom?

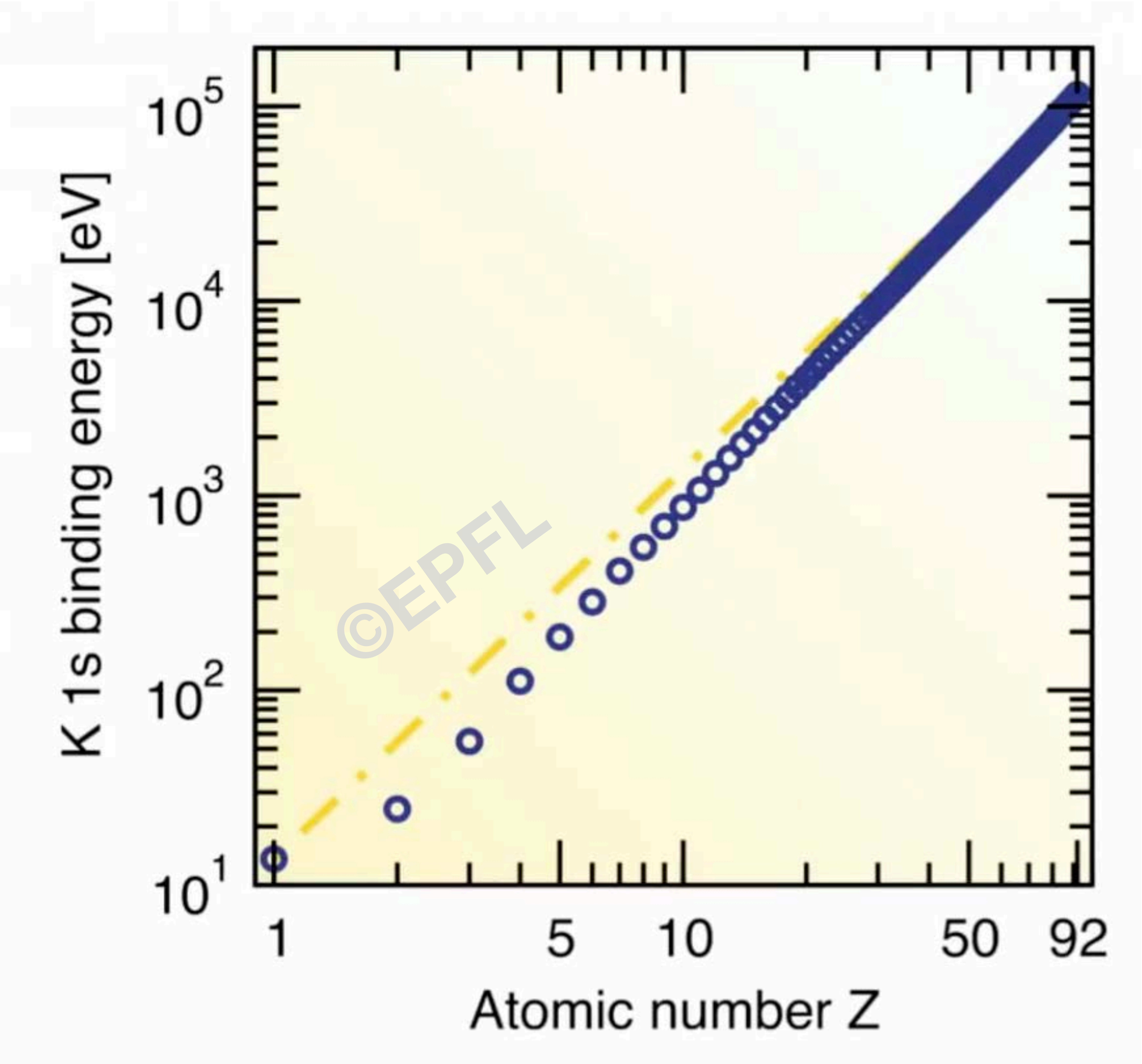
Hydrogen

Silicon

Iron

Gold

Binding Energies of the Atoms



Phil Willmott

Absorption and Refractive Index

The wave equation, suitably modified using the slowly varying envelope approximation:

$$2jk\frac{\partial E_0}{\partial z} = -\mu_0\omega^2 P_0(z)$$

combined with the classical forced oscillator model for the motion of a bound electron:

$$P_0(z) = \frac{Ne^2/m_e}{2\omega_0(\omega_0 - \omega - j\Gamma)} E_0(z)$$

tells us how an EM wave propagates in a medium:

$$E(z,t) = E_0(z=0)\cdot \mathrm{e}^{-\alpha z/2}\cdot \mathrm{e}^{jnkz - j\omega t}$$

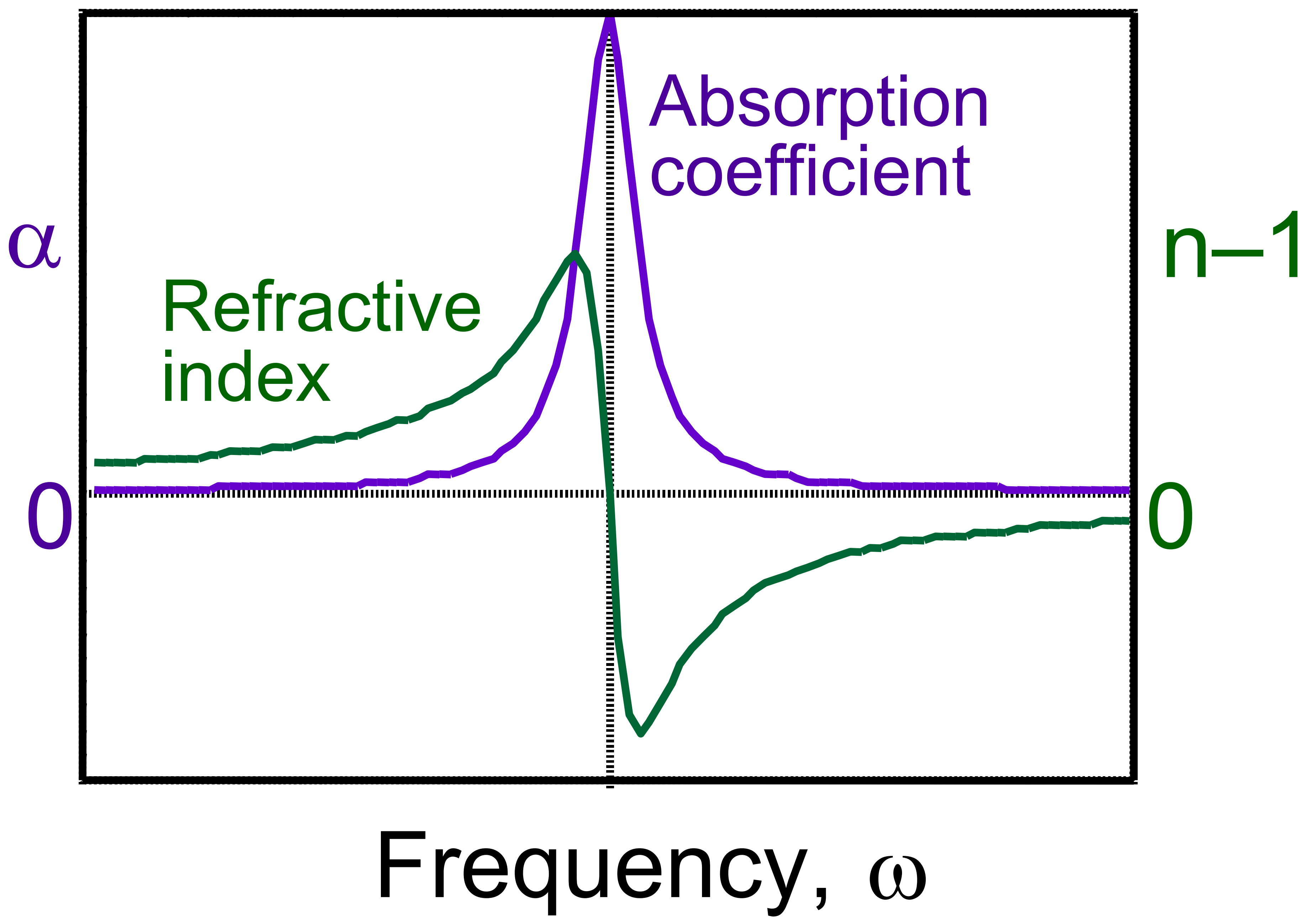
Absorption causes attenuation of the field with increasing z

Refractive index changes the k-vector

https://www.brown.edu/research/labs/mittleman/sites/brown.edu.research.labs.mittleman/files/uploads/lecture08_0.pdf

Absorption and Refractive Index

These functions are, together, a Complex Lorentzian (with some constants in front).



$$\alpha = \frac{Ne^2/m_e}{2\varepsilon_0 c_0} \cdot \frac{\omega}{\omega_0} \cdot \frac{\Gamma}{(\omega_0-\omega)^2+\Gamma^2} \qquad n-1 = \frac{Ne^2/m_e}{4\varepsilon_0\omega_0} \frac{(\omega_0-\omega)}{(\omega_0-\omega)^2+\Gamma^2}$$

https://www.brown.edu/research/labs/mittleman/sites/brown.edu.research.labs.mittleman/files/uploads/lecture08_0.pdf

Absorption and Refractive Index

When $\omega \ll \omega_0$, the electron vibrates 180° out of phase with the light wave.

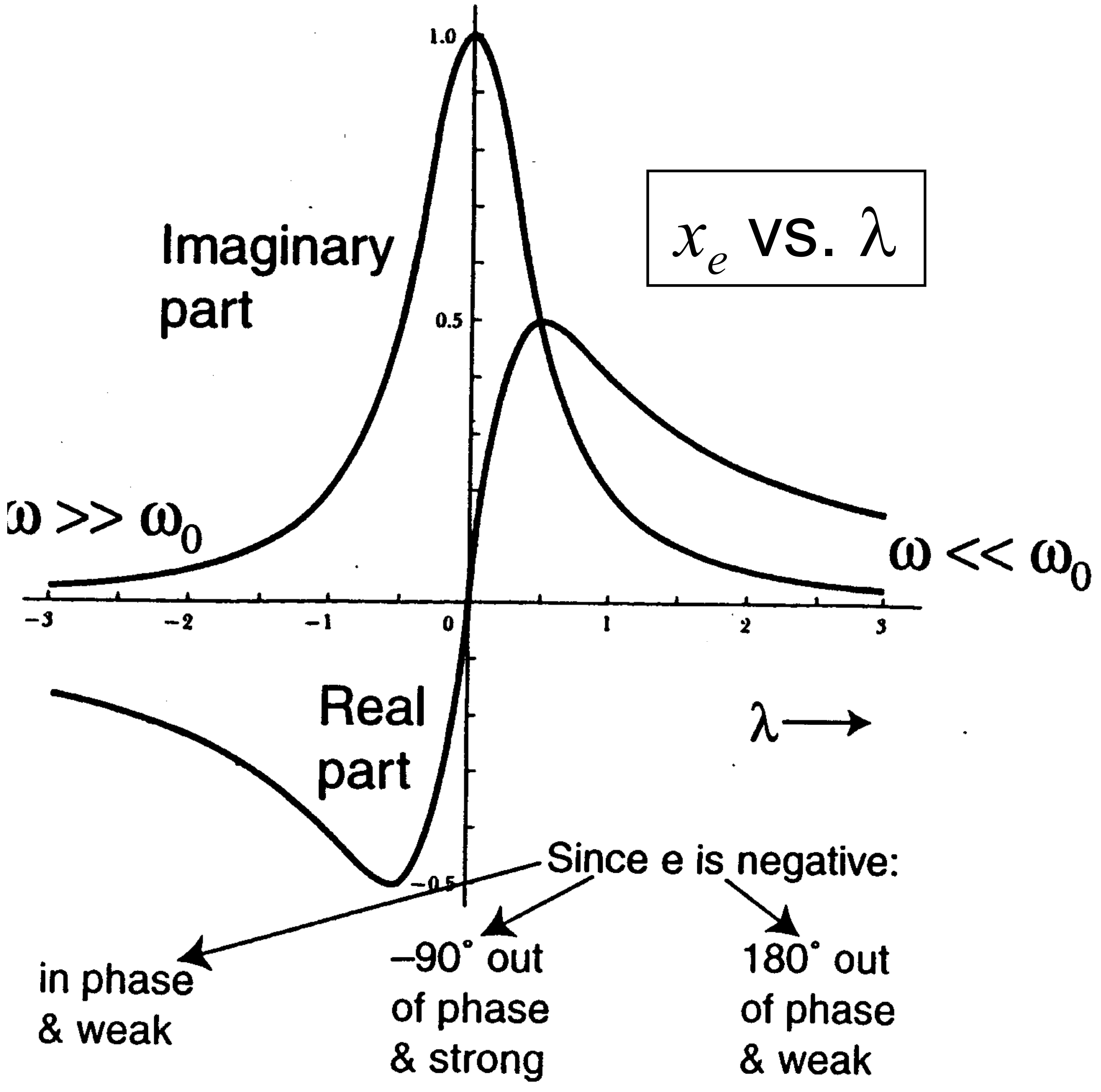
↳ absorption is low, but refractive index is still important.

When $\omega = \omega_0$, the electron vibrates 90° out of phase with the light wave.

↳ absorption is high and refractive index changes rapidly with frequency.

When $\omega \gg \omega_0$, the electron vibrates in phase with the light wave.

↳ absorption is low, but refractive index is still important.

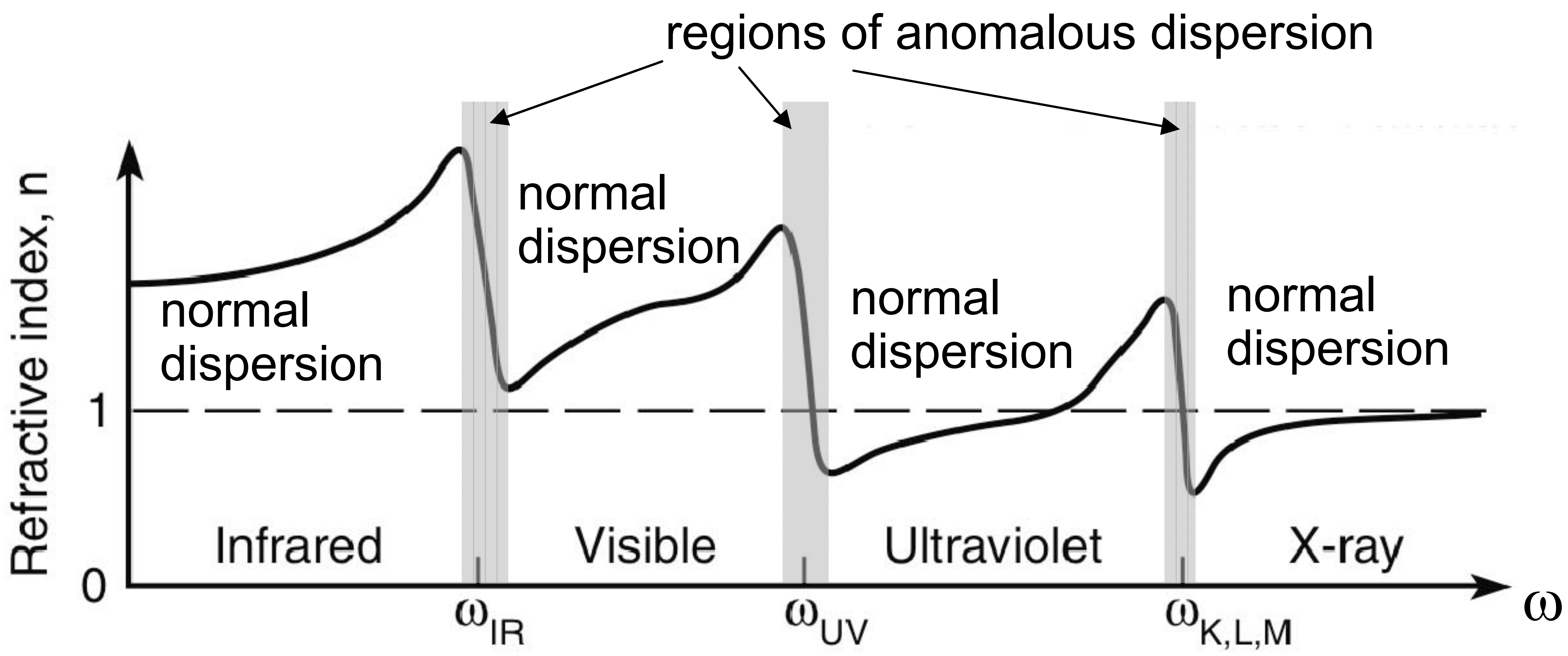


The atoms always oscillate at the frequency of the incident light. The light is not always absorbed by the atoms, but it is always changed by its interaction with the atoms.

https://www.brown.edu/research/labs/mittleman/sites/brown.edu.research.labs.mittleman/files/uploads/lecture08_0.pdf

Absorption and Refractive Index

Since resonance frequencies exist in many spectral ranges, the refractive index varies in a complicated manner.



This illustrates a typical distribution of resonances, with electronic resonances in the UV; vibrational and rotational resonances in the IR, and core electronic resonances occur in the x-ray region.

n increases with frequency, except in "anomalous dispersion" regions. But the overall trend is a decrease in n, as ω increases.

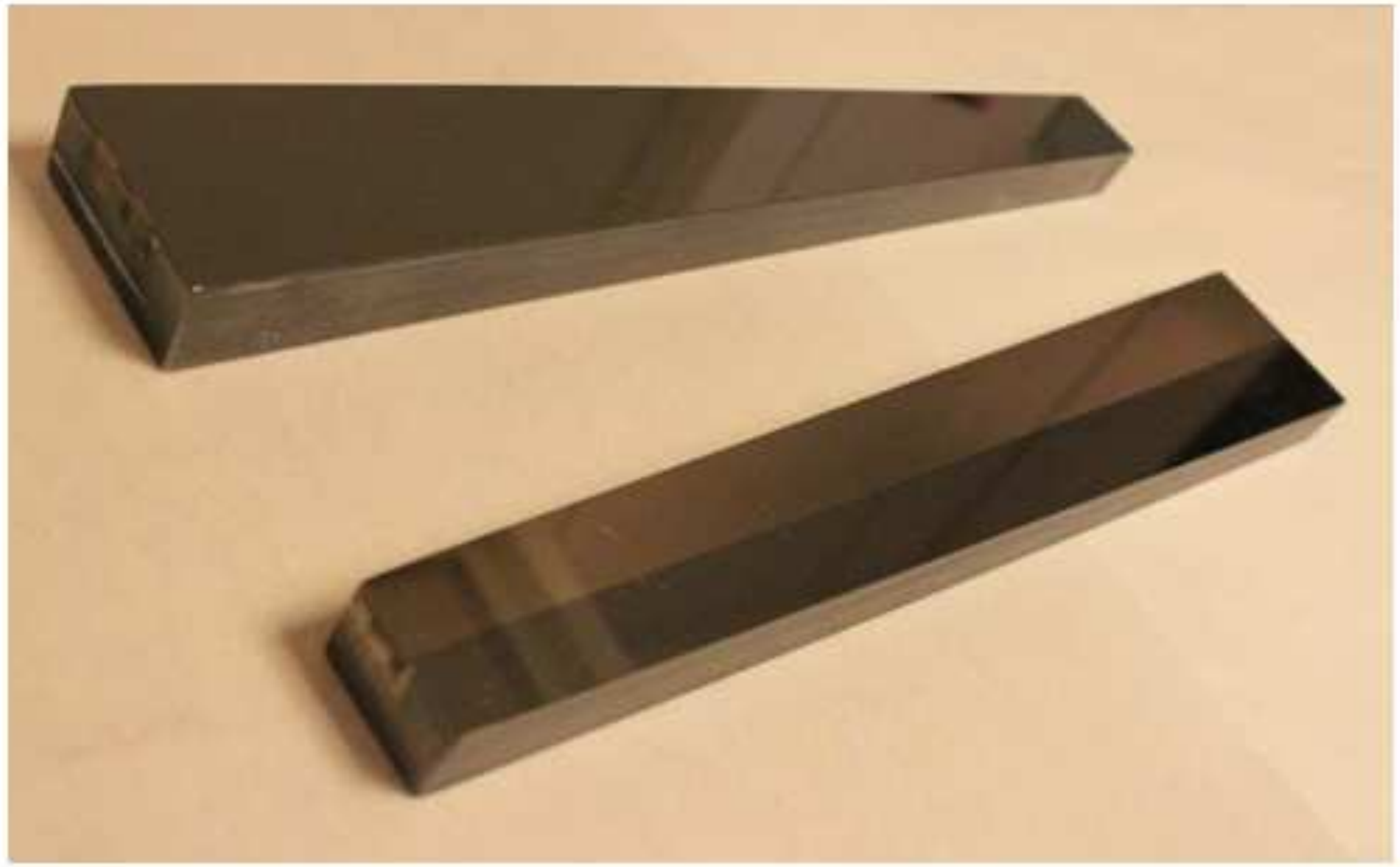
https://www.brown.edu/research/labs/mittleman/sites/brown.edu.research.labs.mittleman/files/uploads/lecture08_0.pdf

Does n<1 Imply that the Speed of the Wave is Larger than the Speed of Light in Vacuum?

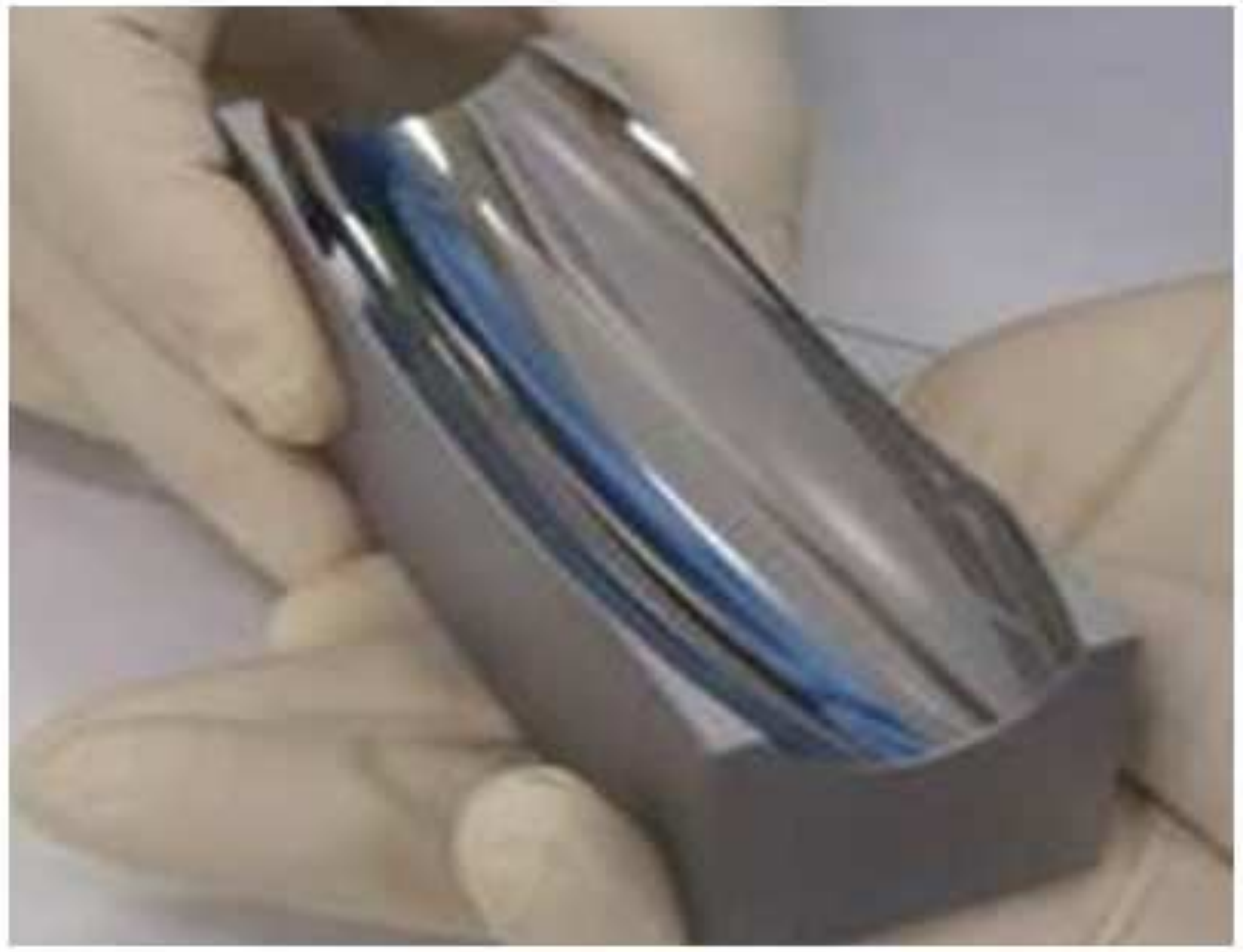
- Indeed, Einstein was wrong: the speed of X-ray photons in material is larger than c
- This applies to the phase velocity only; the group velocity of an X-Ray pulse is smaller than c
- Iron
- Gold

X-Ray Mirrors: Total External Reflection

Kirkpatrick-Baez Mirror



Ellipsoidal Mirror

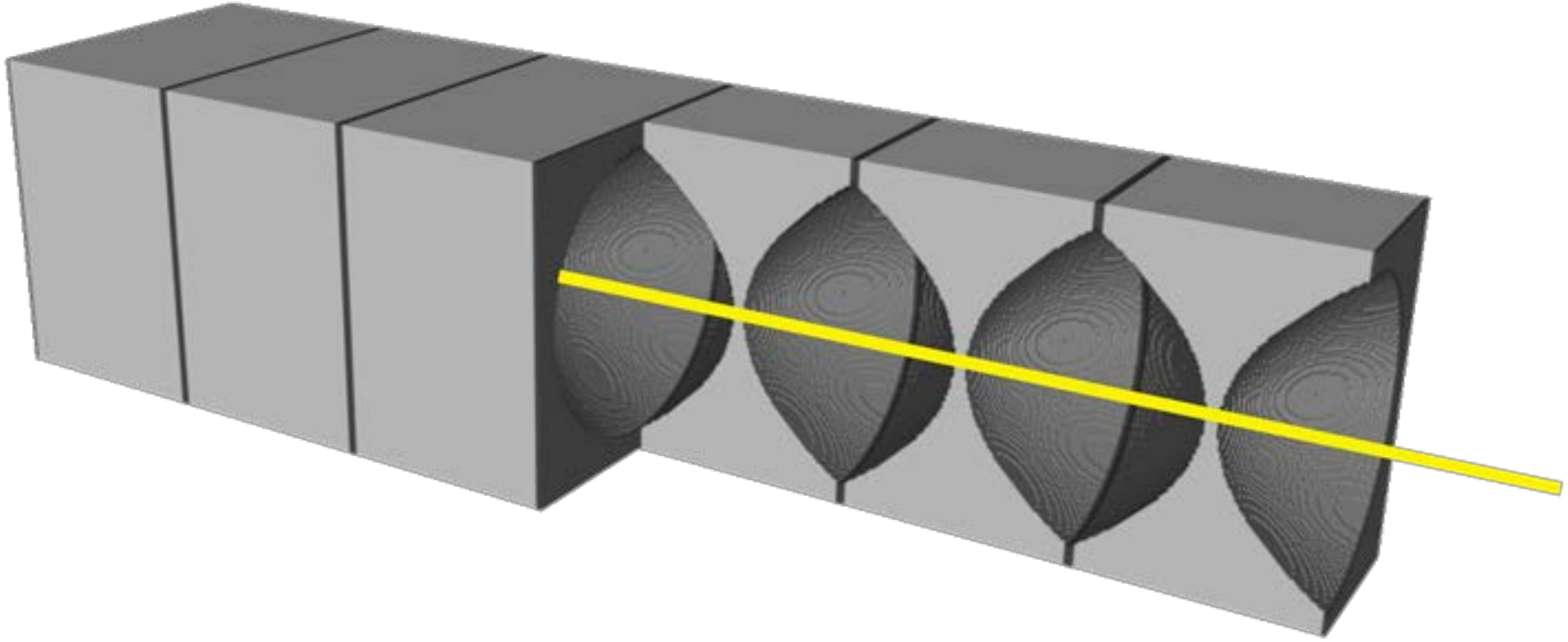


Toroidal Mirror

R. Radhakrishnan et al., DOI: 10.1149/07711.1255ecst

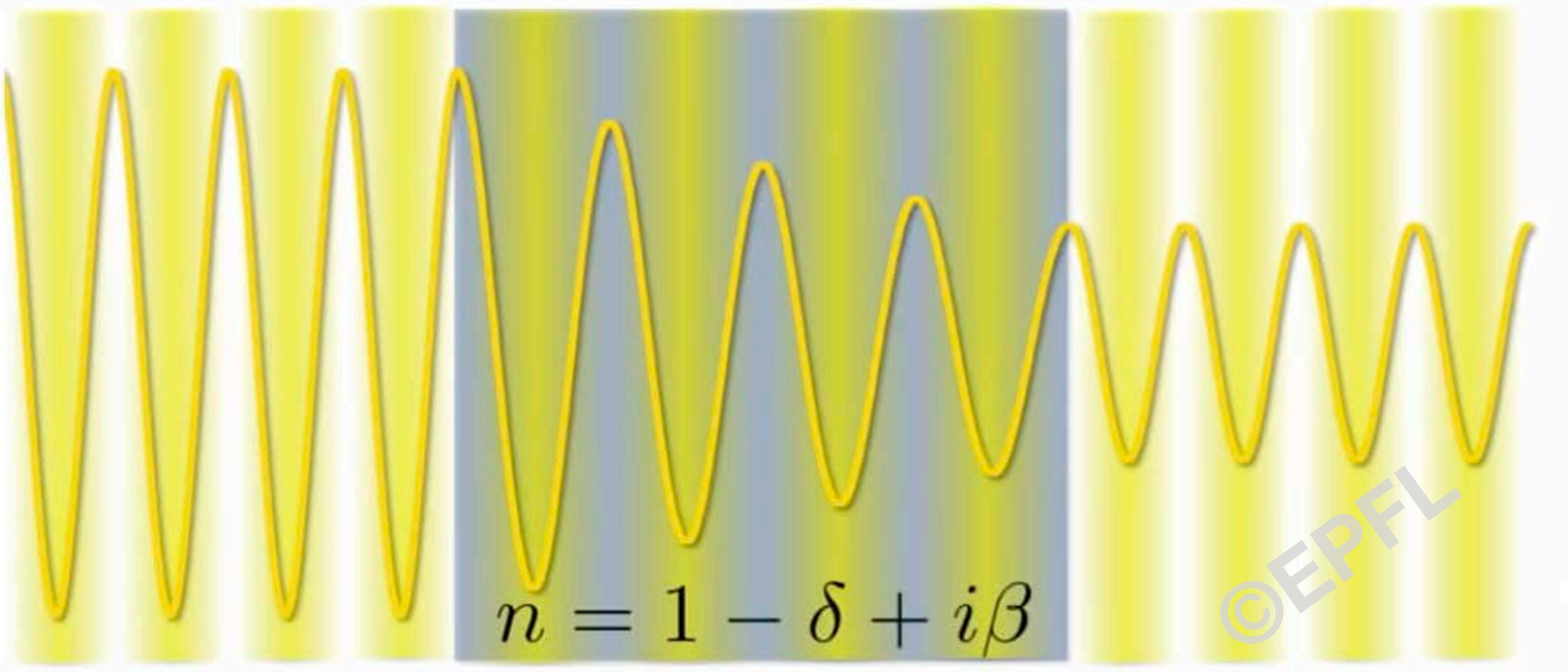
X-Ray Lenses

- Keep in mind that n<1



rxoptics.de

Absorption

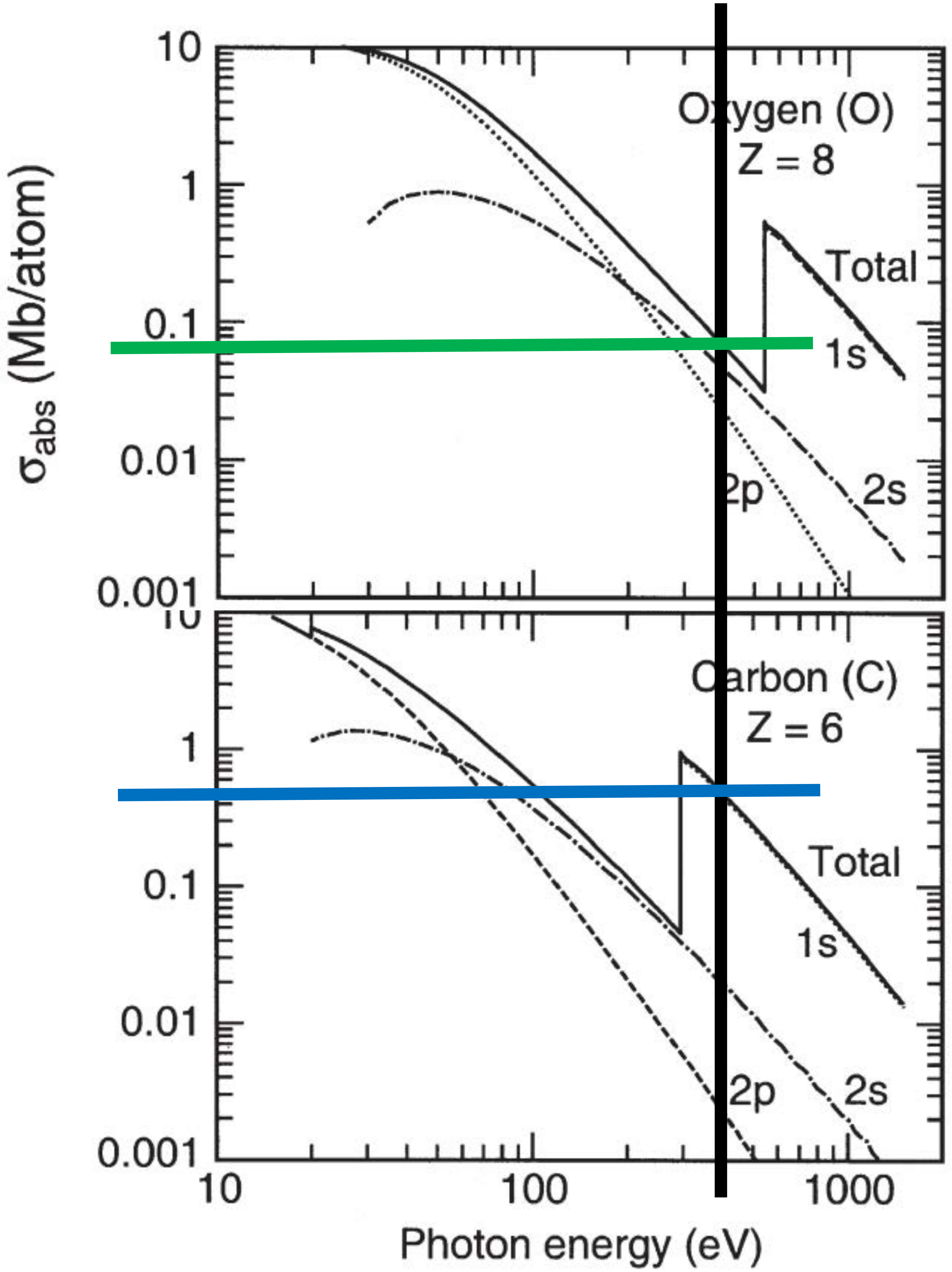


- Real part, $1 - \delta$: increases λ to $\lambda/(1-\delta)$
- Imaginary part, β: exponential decay of amplitude E_0

Which Atoms Absorb 0.4 keV X-Rays More Strongly?

Carbon (Z=6)

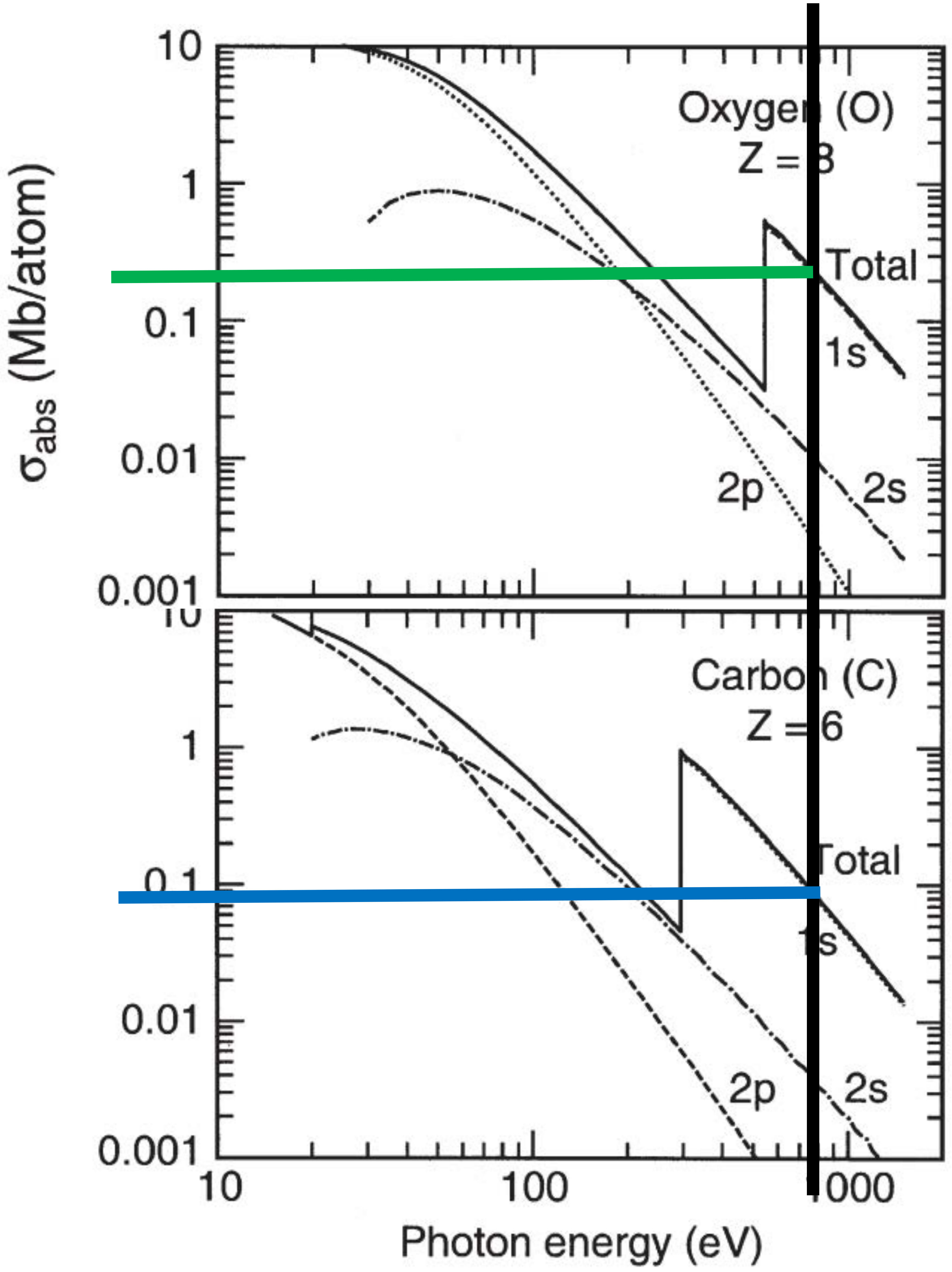
Oxygen (Z=8)



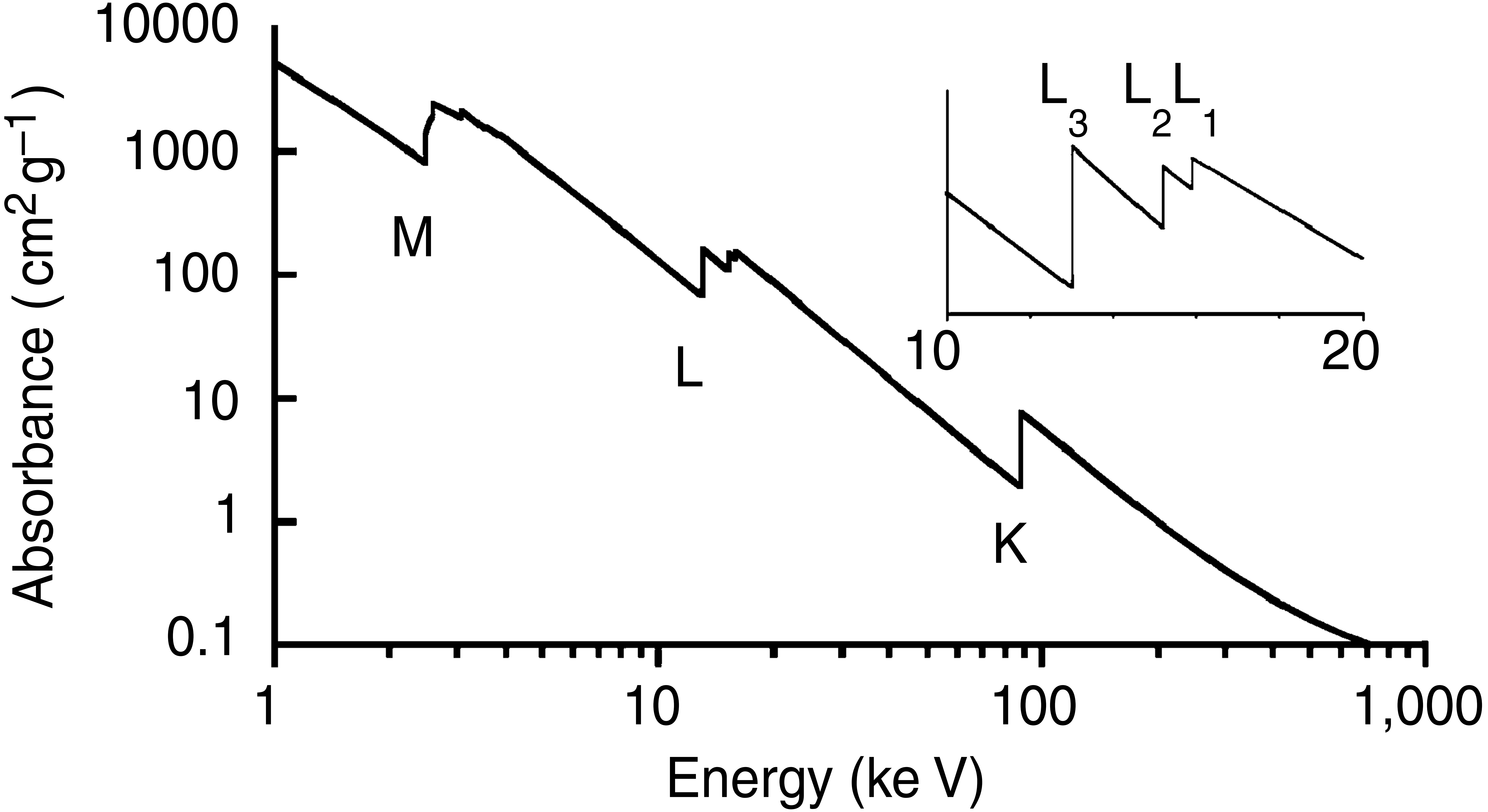
Which Atoms Absorb 0.8 keV X-Rays More Strongly?

Carbon (Z=6)

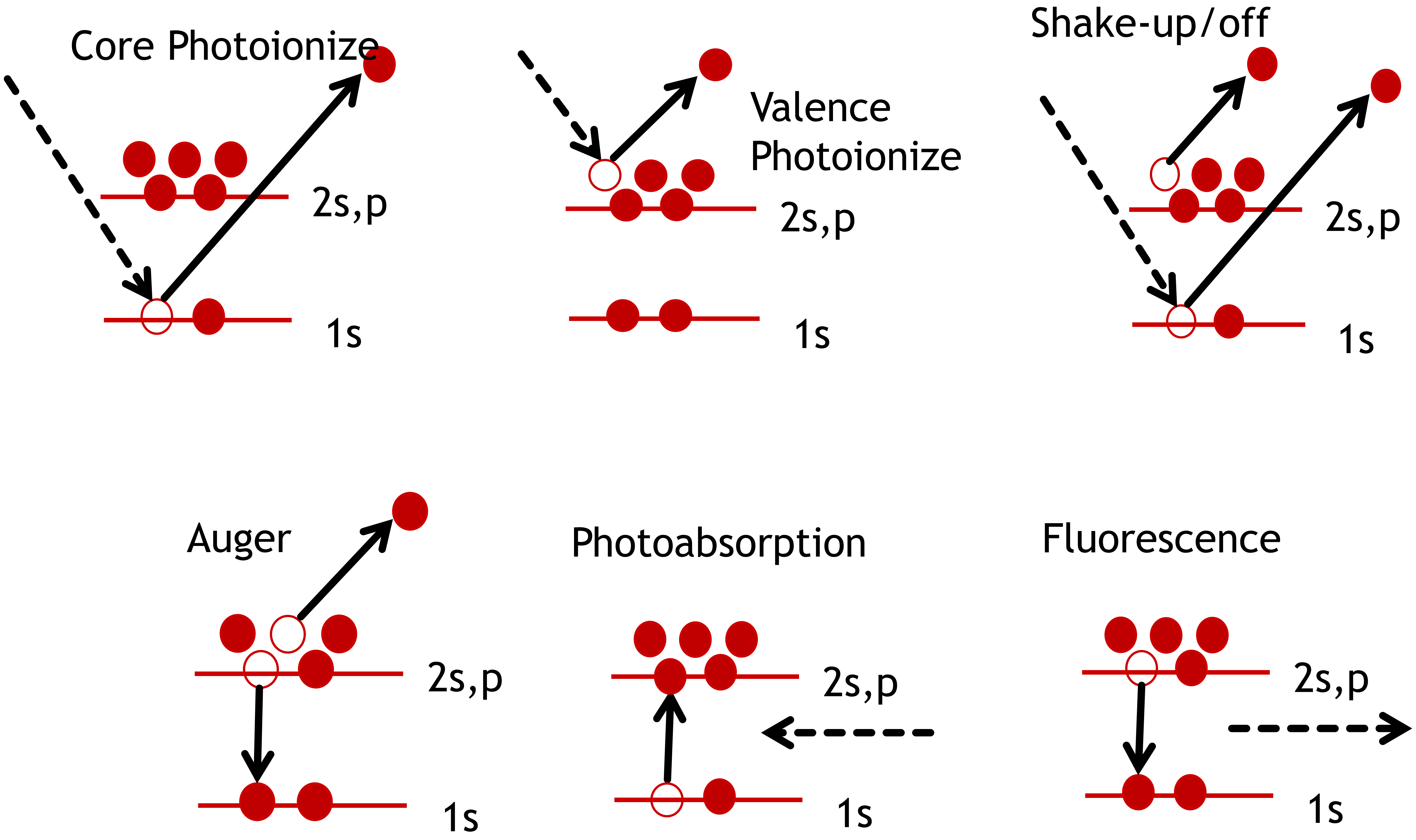
Oxygen (Z=8)



X-Ray Absorption Spectrum of Lead



Processes


Core Photoionize
2s,p
1s
Valence
Photoionize
2s,p
1s
Shake-up/off
2s,p
1s
Auger
2s,p
1s
Photoabsorption
2s,p
1s
Fluorescence
2s,p
1s

Calculation of Cross Section

$\psi(\boldsymbol{R}_{j=1,N\ nuclei}, \boldsymbol{r}_{i=1,M\ electrons}, t)$ is a complete dynamical description of the M electrons and N nuclei in a molecule interacting with x rays (YIKES!)

$\hat{H}\psi = i\dot{\psi}$ Looks simple enough, but wait:

$\hat{H} = \hat{H}_{\text{mol}} + \hat{H}_{\text{EM}} + \hat{H}_{\text{int}}$ The atoms, the x-rays, and their interaction

$$\hat{H}_{\text{mol}} = \hat{T}_{\text{N}} + \hat{V}_{\text{NN}} + \hat{H}_{\text{el}}$$

$$\hat{T}_{\text{N}} = -\frac{1}{2}\sum_{n}\frac{\nabla_n^2}{M_n} \qquad \hat{V}_{\text{NN}} = \sum_{n<n'}\frac{Z_n Z_{n'}}{|\boldsymbol{R}_n - \boldsymbol{R}_{n'}|}$$

$$\hat{H}_{el} = \sum_i\left[-\frac{1}{2}\nabla_i^2 - \sum_j\frac{Z_j}{|\boldsymbol{r}, -\boldsymbol{R}_j|} + \frac{1}{2}\sum_k\frac{1}{|\boldsymbol{r}_i - \boldsymbol{r}_k|}\right]$$

Simplifications

Born-Oppenheimer Approximation: Treat the nuclei as classical point particles at first, and just solve $\hat{H}_{el}\psi = i\dot{\psi}$ for the electrons.

$$\sum_i \left[-\frac{1}{2}\nabla_i^2 - \sum_j \frac{Z_j}{|\boldsymbol{r}, -\boldsymbol{R}_j|} + \frac{1}{2}\sum_k \frac{1}{|\boldsymbol{r}_i - \boldsymbol{r}_k|}\right]\psi(\boldsymbol{R_j}; \boldsymbol{r_{i=1,M}}) = i\dot{\psi}(\boldsymbol{R_j}; \boldsymbol{r_{i=1,M}})$$

Hartree-Fock Approximation: The wave function can be expressed as a product of single-electron orbitals (called spin orbitals if you include the electron spin)

$$\boldsymbol{\psi}(\boldsymbol{R_j}; \boldsymbol{r}_{i=1,M\ electrons}, t) = \prod_i \varphi(\boldsymbol{R_j}; \boldsymbol{r_i}) + antisymmetrized\ permutations$$

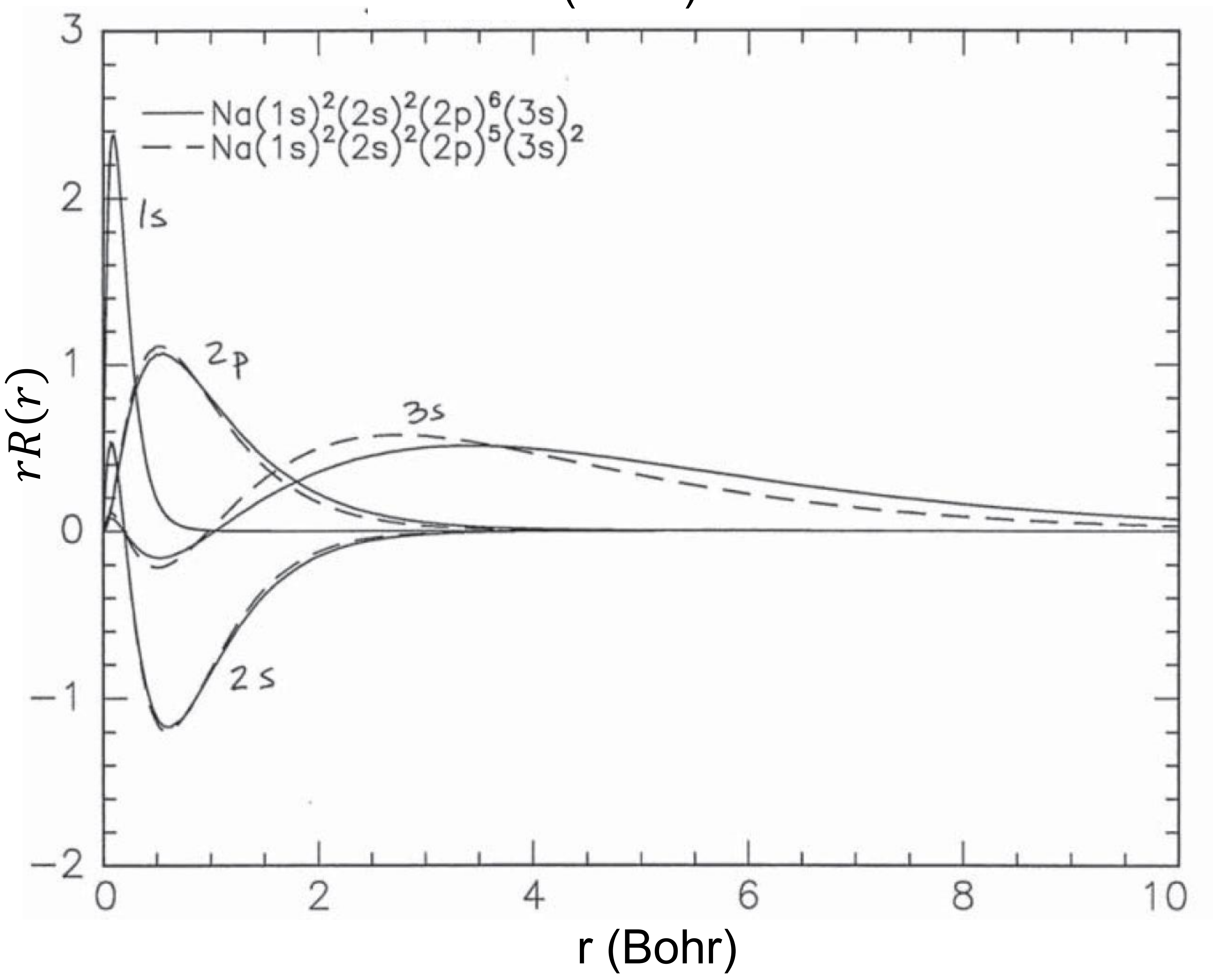
(This is called a "Slater determinant", because complete antisymmetrization of a list of symbols may be expressed as a determinant of the symbol matrix.)

Standard programs to calculate this using variational calculus

Hartree Fock Single Electron Orbitals

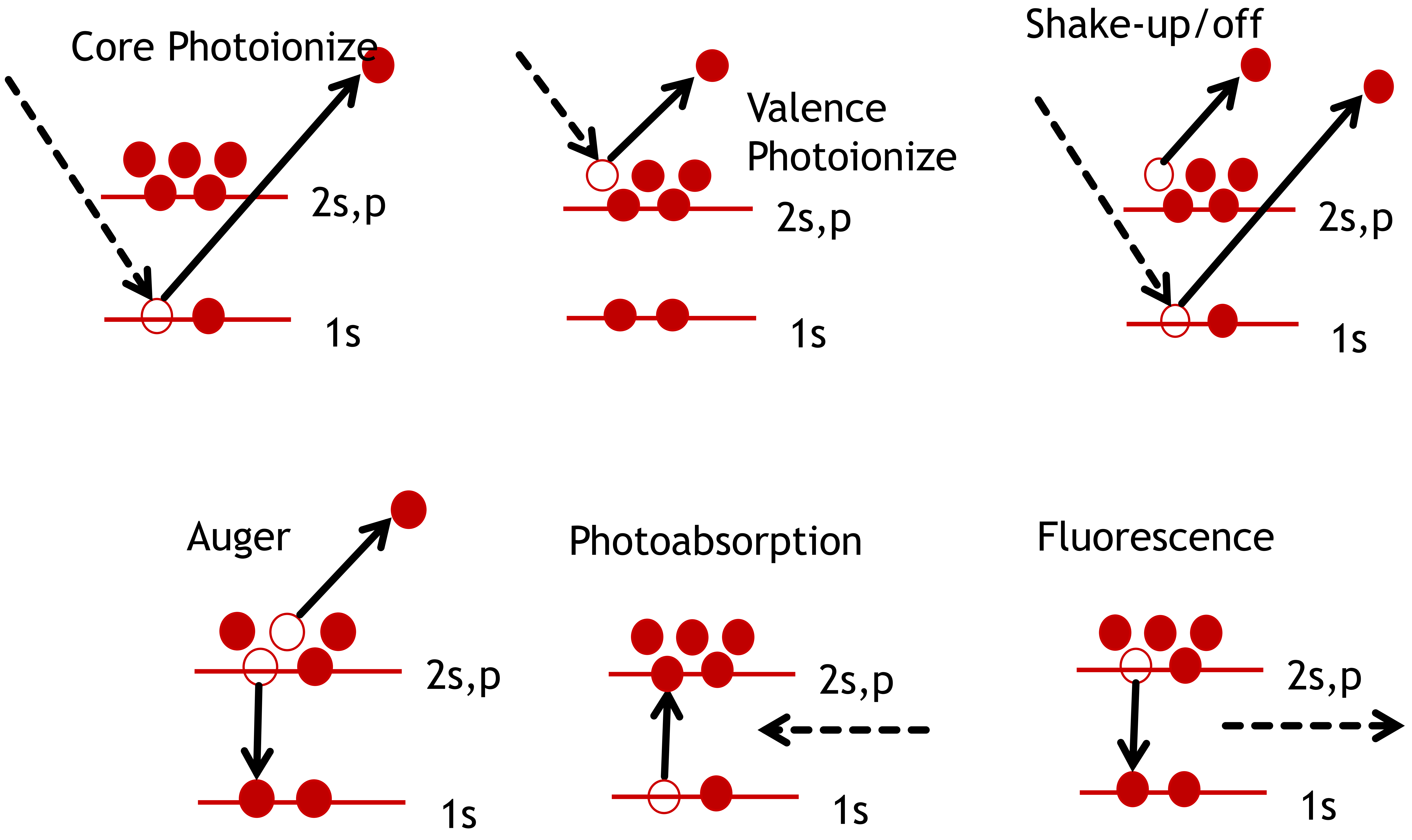
$$\varphi(\boldsymbol{r}) = R(r)Y_{lm}(\Omega)$$

Sodium (Z=11)



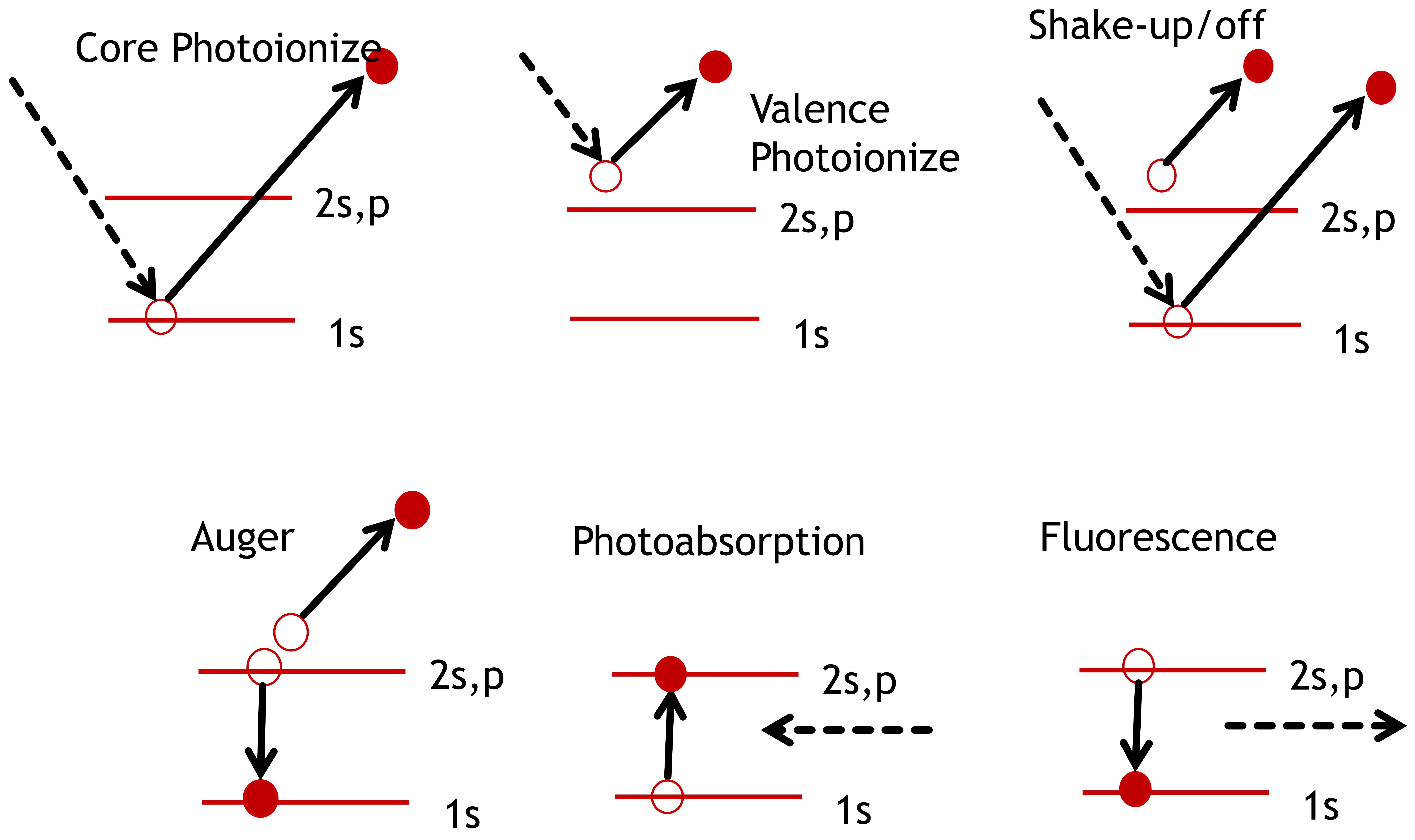
Orbital	Binding (eV)
1s	1070.8
2s	63.5
2p	30.6
3s	5.14

Processes



Phil Bucksbaum

Mean Field Approximation:
Only Keep Track of the Electron and the Hole



These electrons (and holes, or electron vacancies) are quantum excitations of the "mean field" of the original atom.

Second Quantization

Electron and hole excitations are created in the atom's mean field using "creation" and "annihilation" operators

The operator $\hat{\psi}_\sigma^\dagger(\boldsymbol{r})[\hat{\psi}_\sigma(\boldsymbol{r})]$ creates (annihilates) an electron at $\boldsymbol{r}$ with spin quantum number σ.

The operators obey anticommutation relations because of Fermi-Dirac statistics:

$$\{\hat{\psi}_\sigma(\boldsymbol{x}), \hat{\psi}_{\sigma'}(\boldsymbol{x}')\} = 0,$$

$$\{\hat{\psi}_\sigma(\boldsymbol{x}), \hat{\psi}^\dagger_{\sigma'}(\boldsymbol{x}')\} = \delta_{\sigma,\sigma'}\delta^{(3)}(\boldsymbol{x} - \boldsymbol{x}'),$$

$$\{\hat{\psi}^\dagger_\sigma(\boldsymbol{x}), \hat{\psi}^\dagger_{\sigma'}(\boldsymbol{x}')\} = 0.$$

- For more details, see: https://app.certain.com/accounts/register123/stanford/pulseinstitute/events/uxss2018/2018.UXSS.Bucksbaum.AMO.tutorial.pdf

Result: Calculation of Cross-Sections

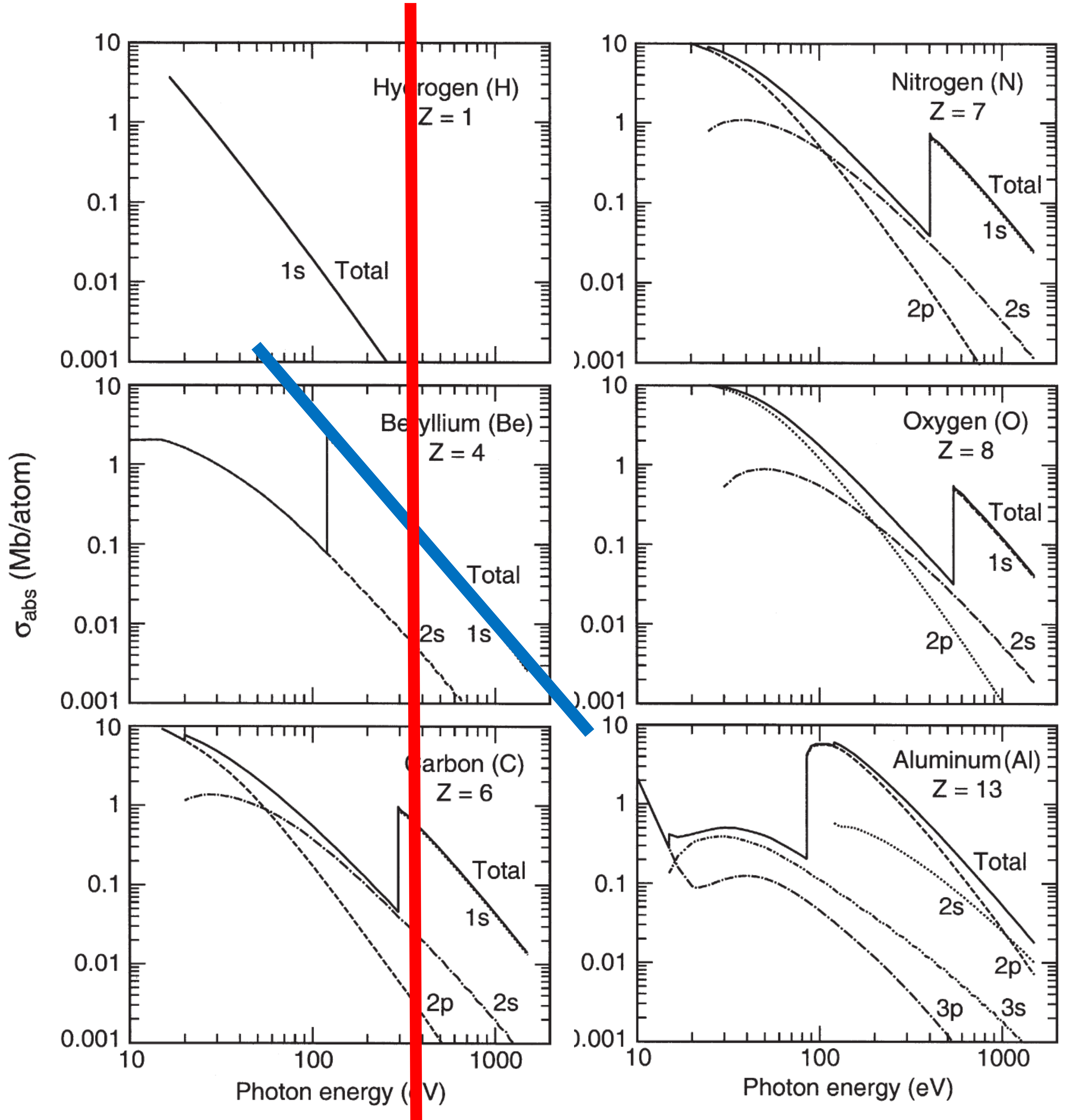


Fig. 1-4. *Plots of atomic subshell photoemission cross sections, calculated for isolated atoms.* *Subshell photoemission cross sections (continued)*

Cross section rises as Z^5 at fixed photon energy

And falls as $\omega^{-7/2}$ above threshold.

Crystals

Wikimedia Commons

Which of These are Crystals?

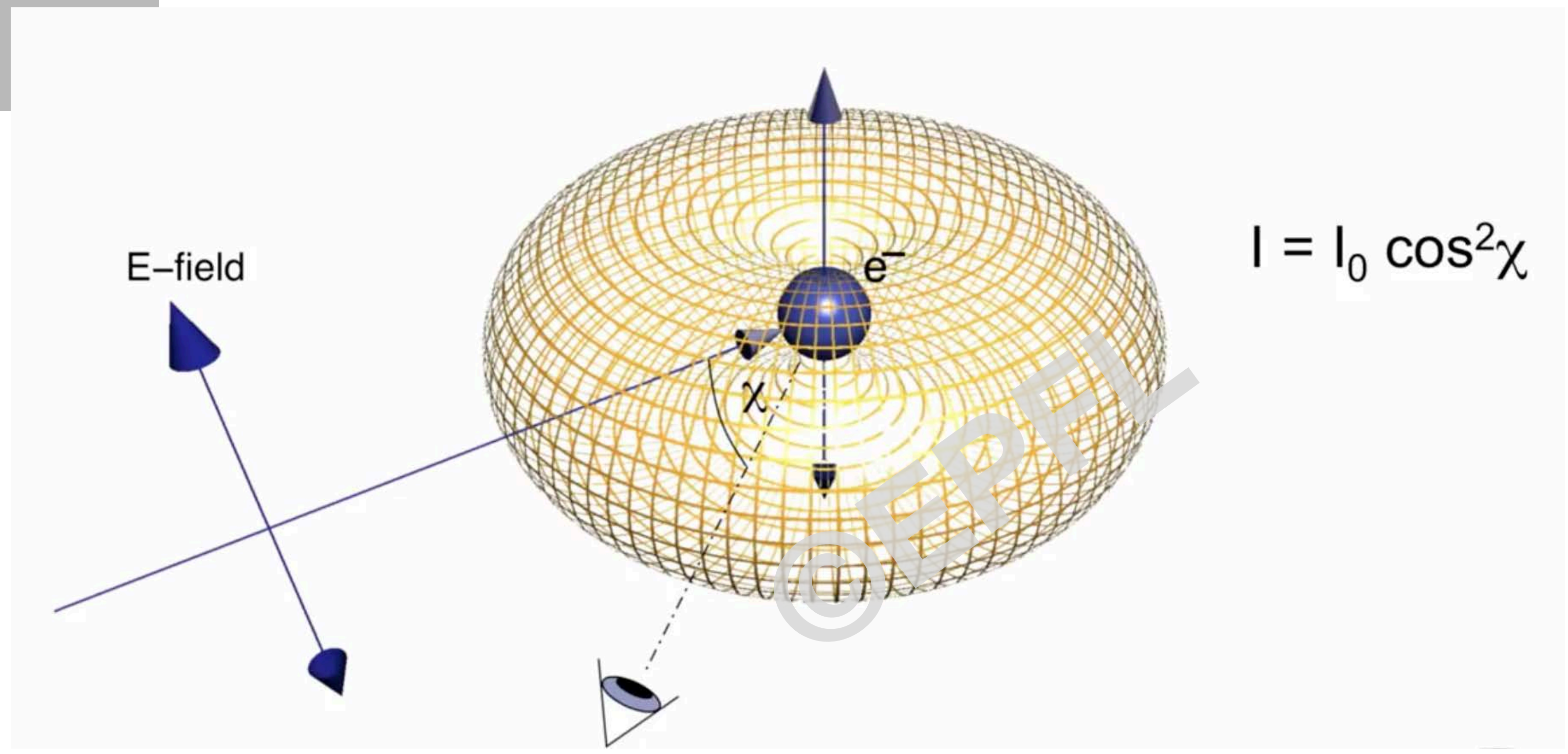
- Salt
- Silicon wafers for computer chips
- Diamonds
- All of the above

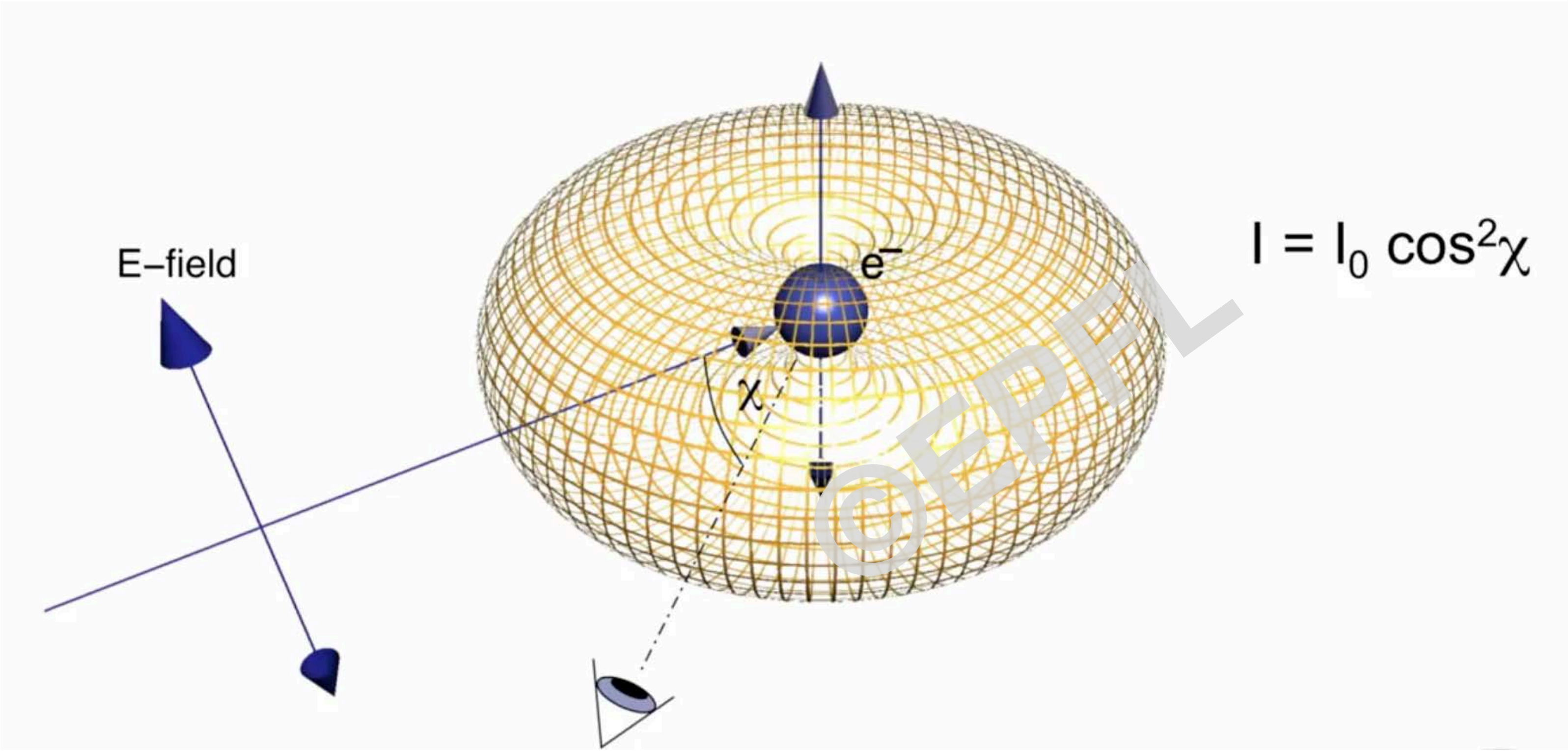
Interaction of X-Rays with Matter

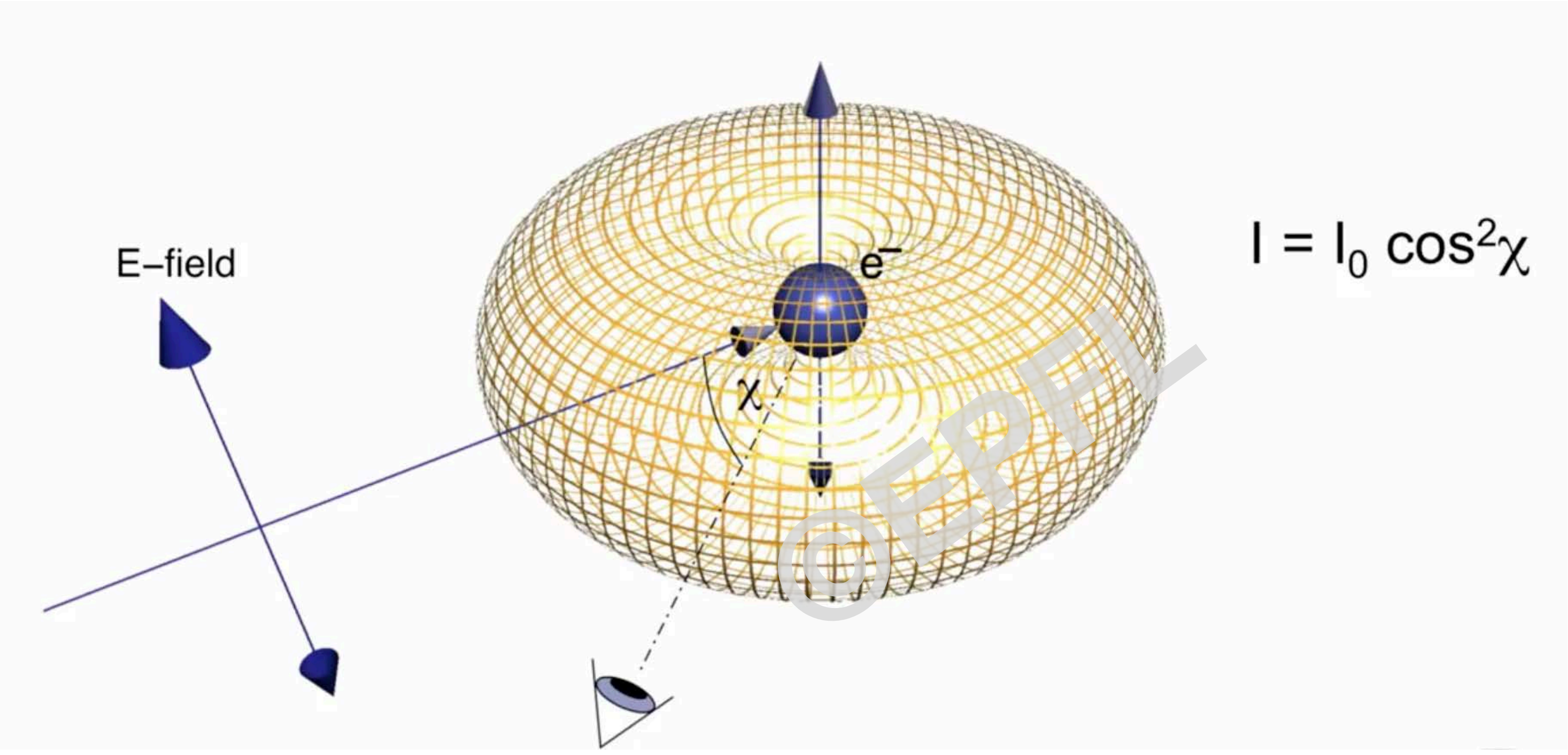
What are Not Crystals?

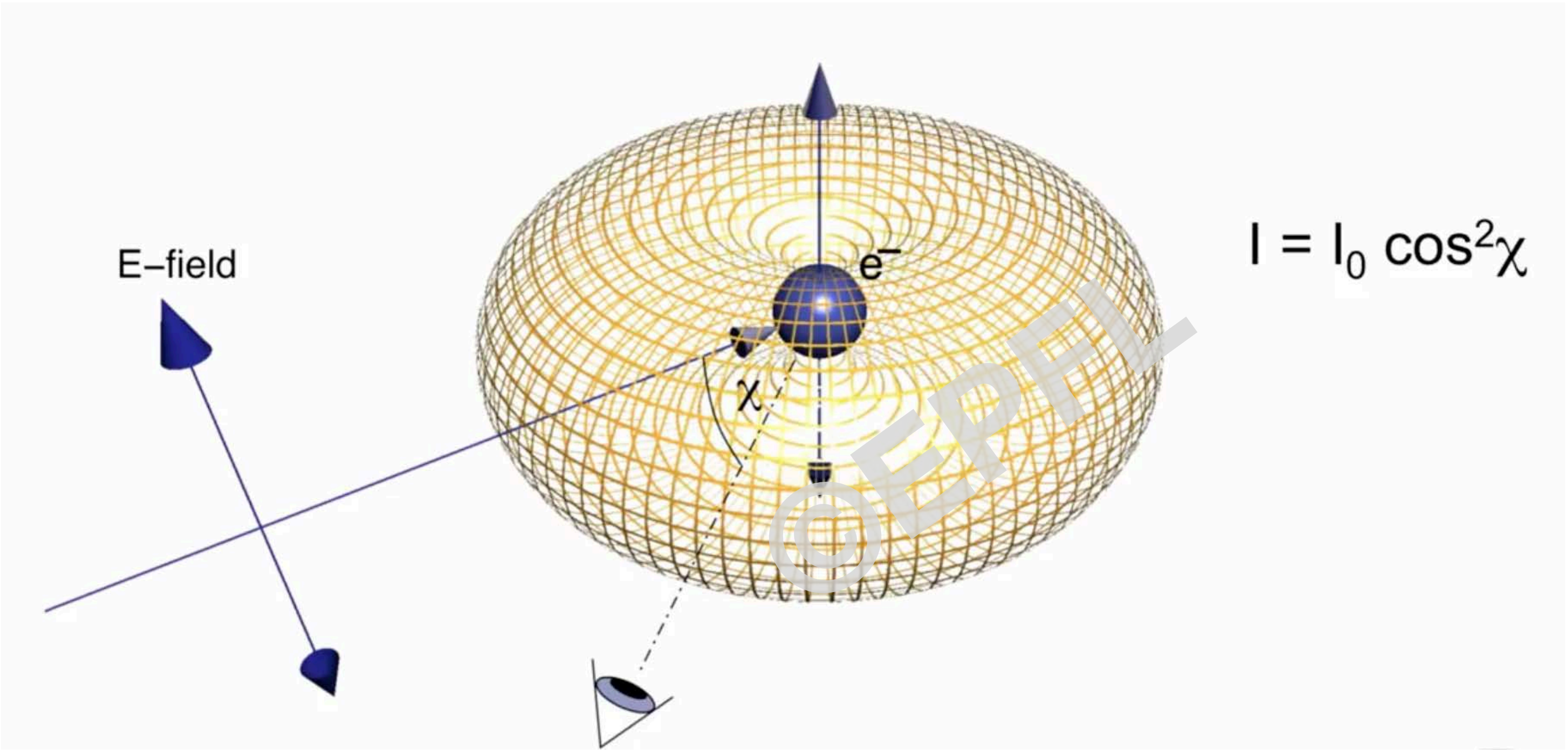
Nick Veasey

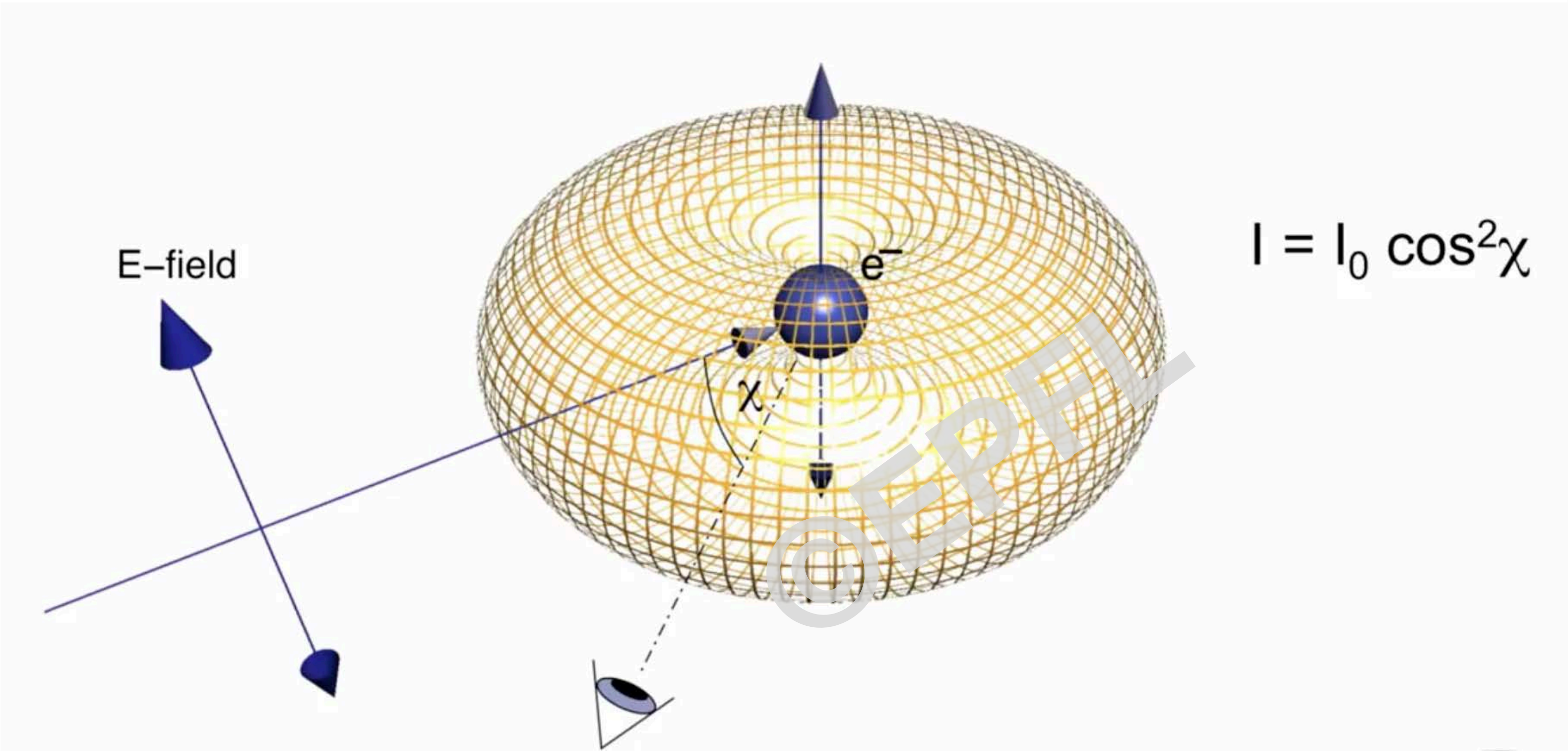
Diffraction

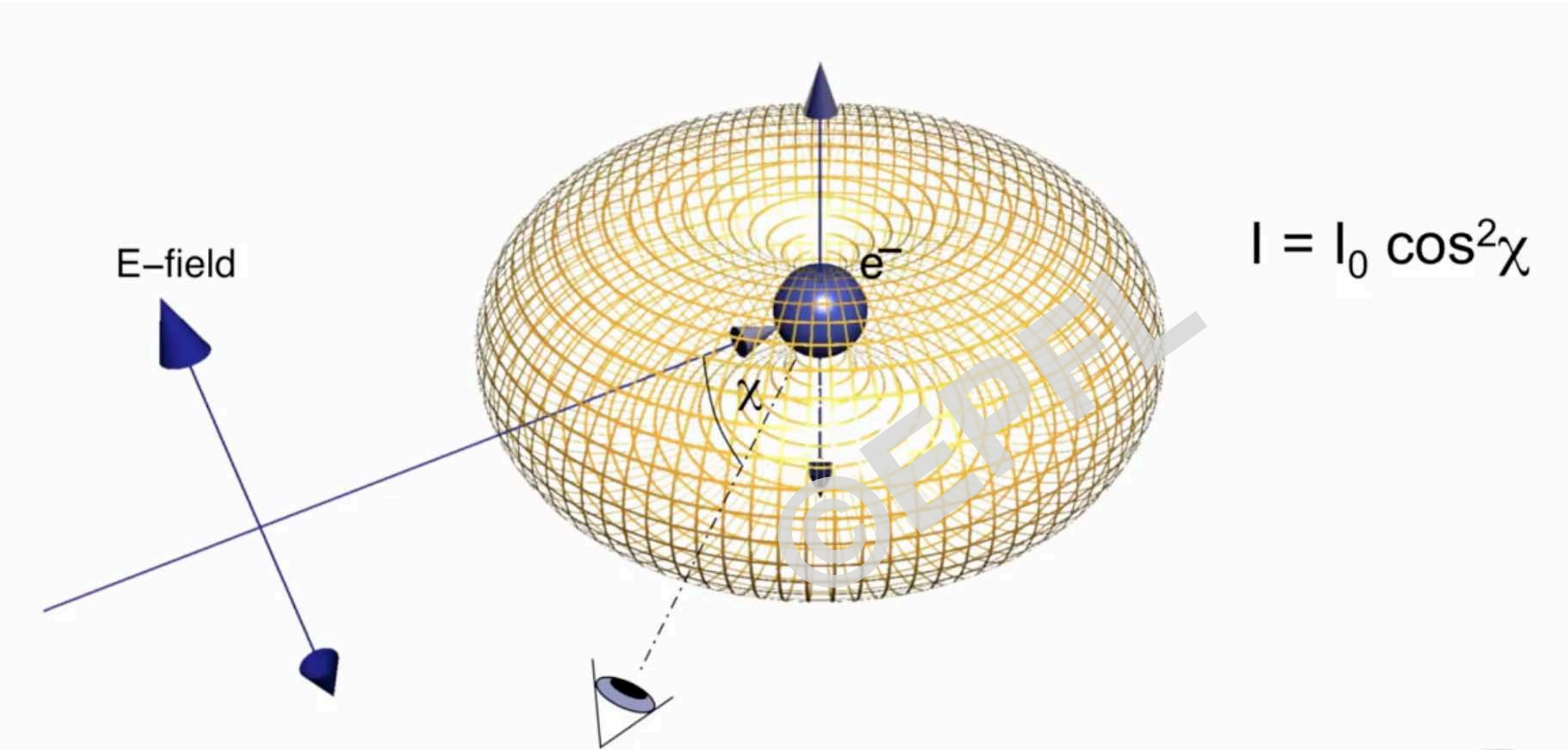




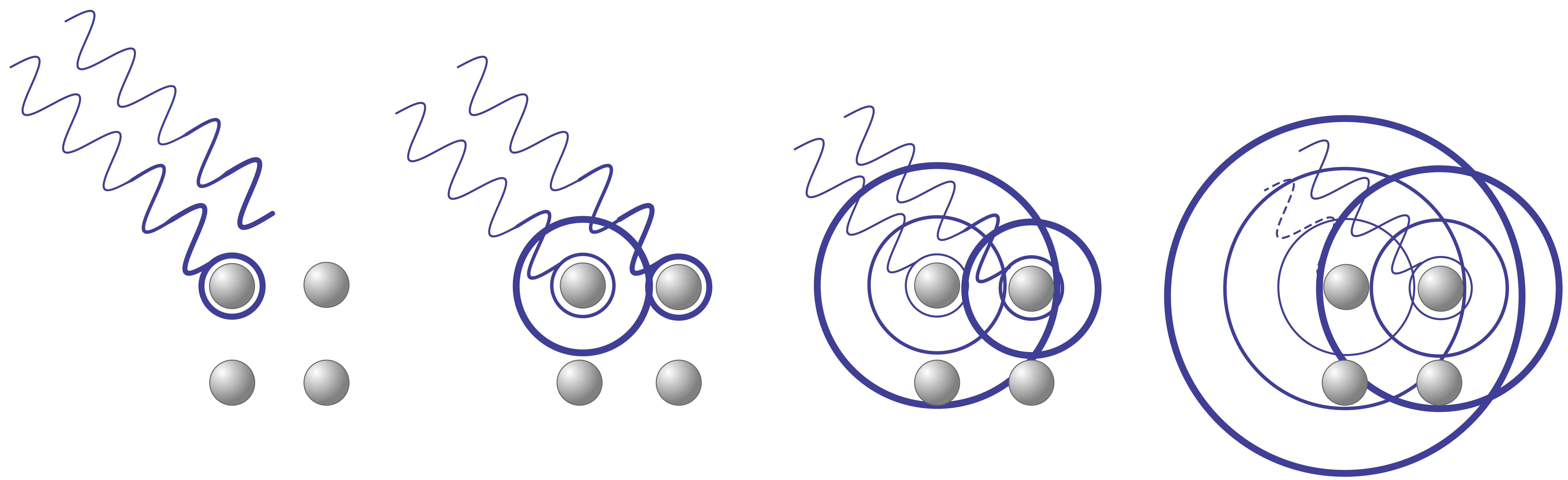


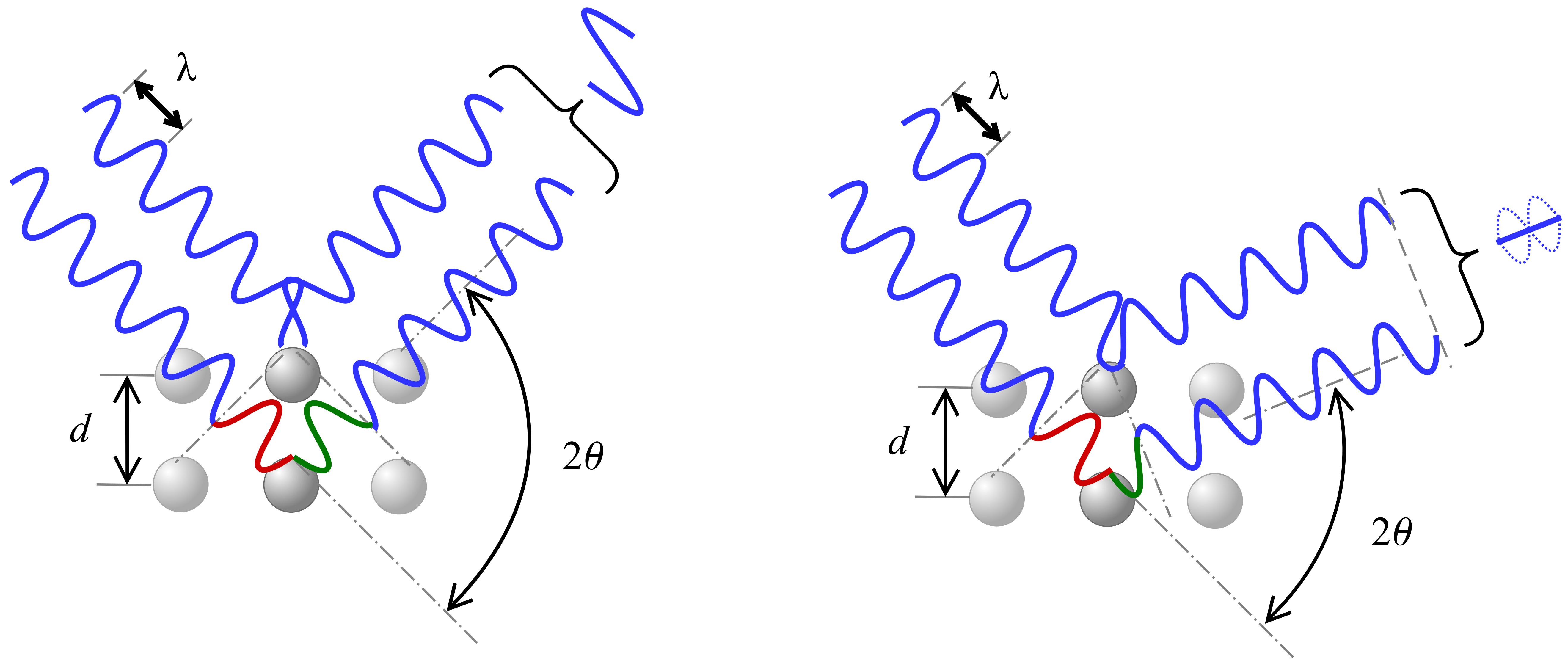






Diffraction





Wikimedia Commons

Bragg's Law

$$(AB + BC) - (AC') = n\lambda\,,$$

where the same definition of n and λ apply as above

Therefore,

$$AB = BC = \frac{d}{\sin\theta} \text{ and } AC = \frac{2d}{\tan\theta}\,,$$

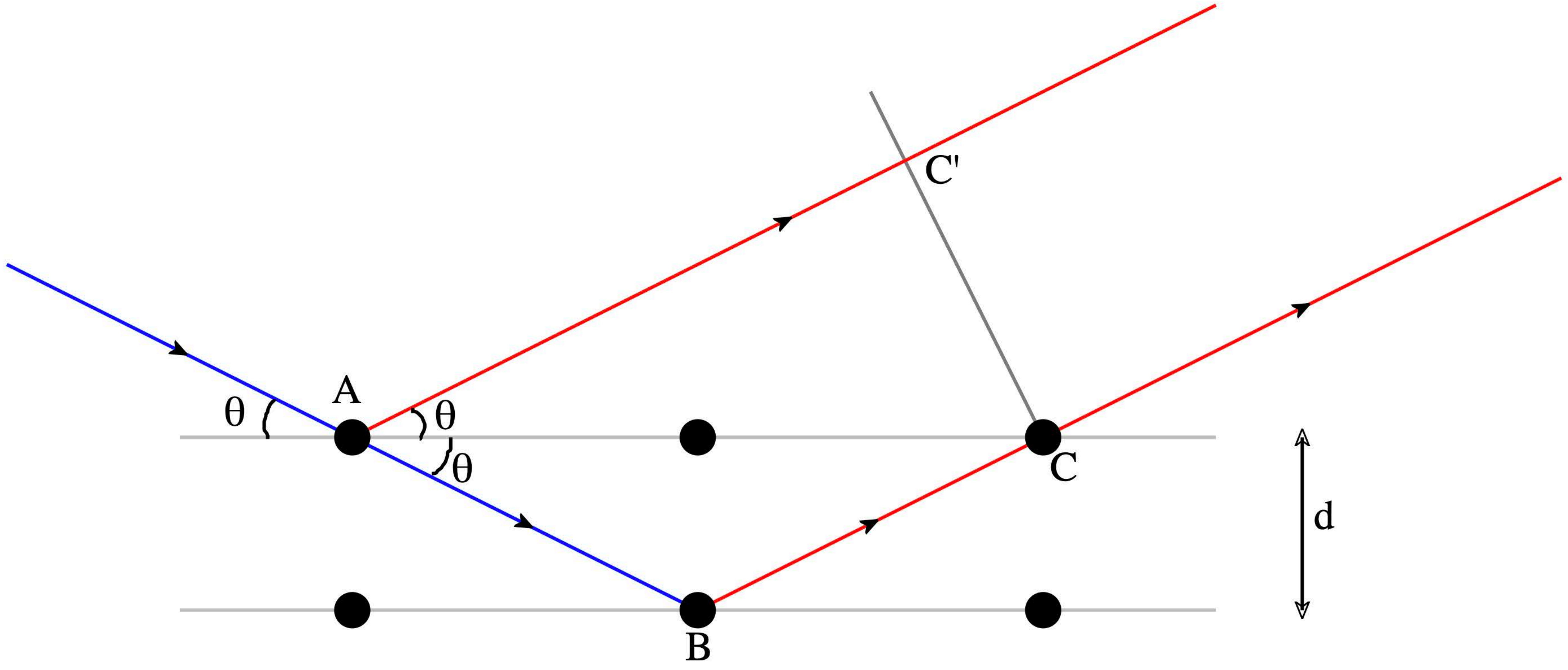
from which it follows that

$$AC' = AC \cdot \cos\theta = \frac{2d}{\tan\theta}\cos\theta = \left(\frac{2d}{\sin\theta}\cos\theta\right)\cos\theta = \frac{2d}{\sin\theta}\cos^2\theta$$

Putting everything together,

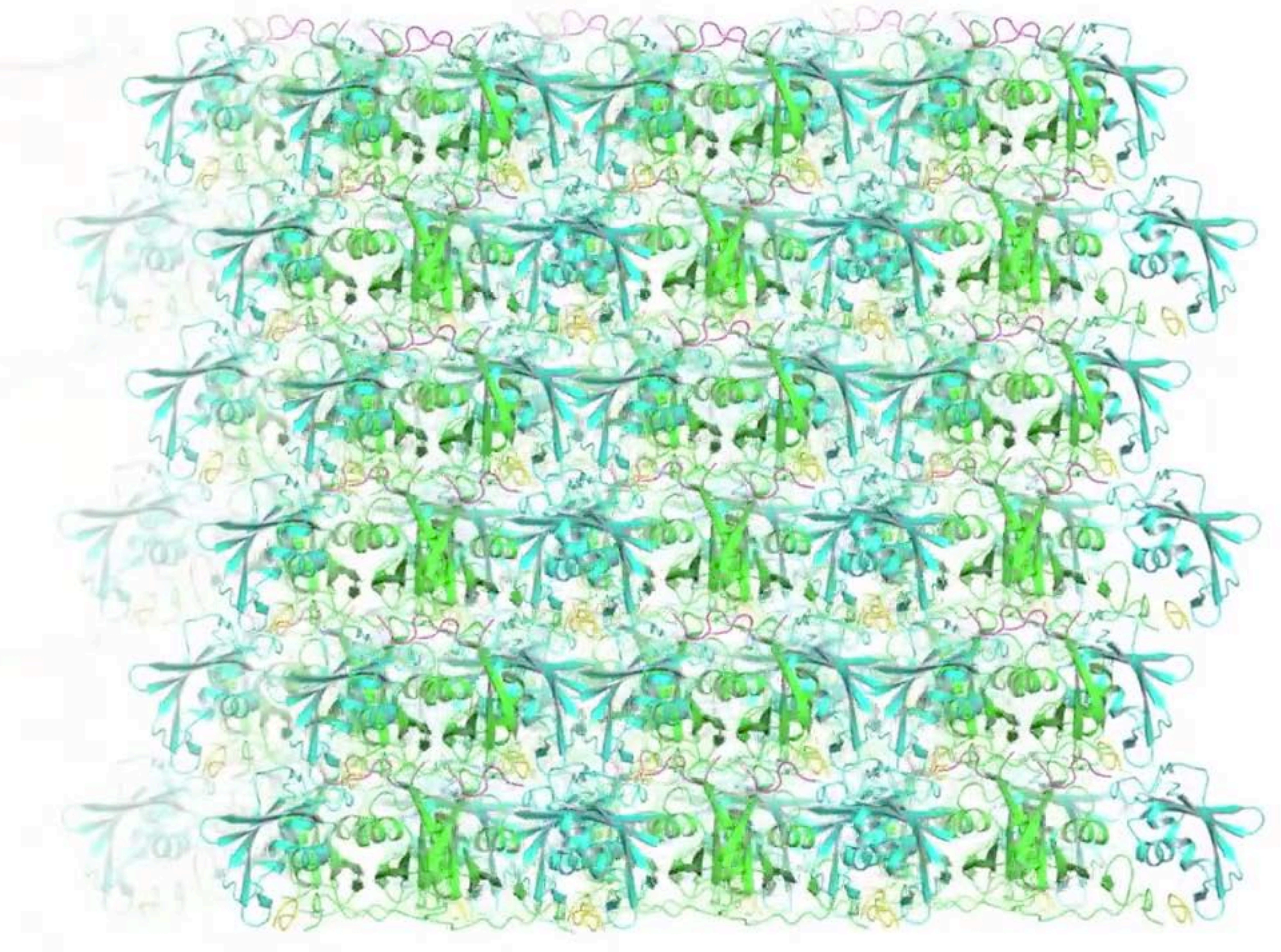
$$n\lambda = \frac{2d}{\sin\theta} - \frac{2d}{\tan\theta}\cos\theta = \frac{2d}{\sin\theta}\left(1 - \cos^2\theta\right) = \frac{2d}{\sin\theta}\sin^2\theta$$

which simplifies to $n\lambda = 2d\sin\theta$, which is Bragg's law.



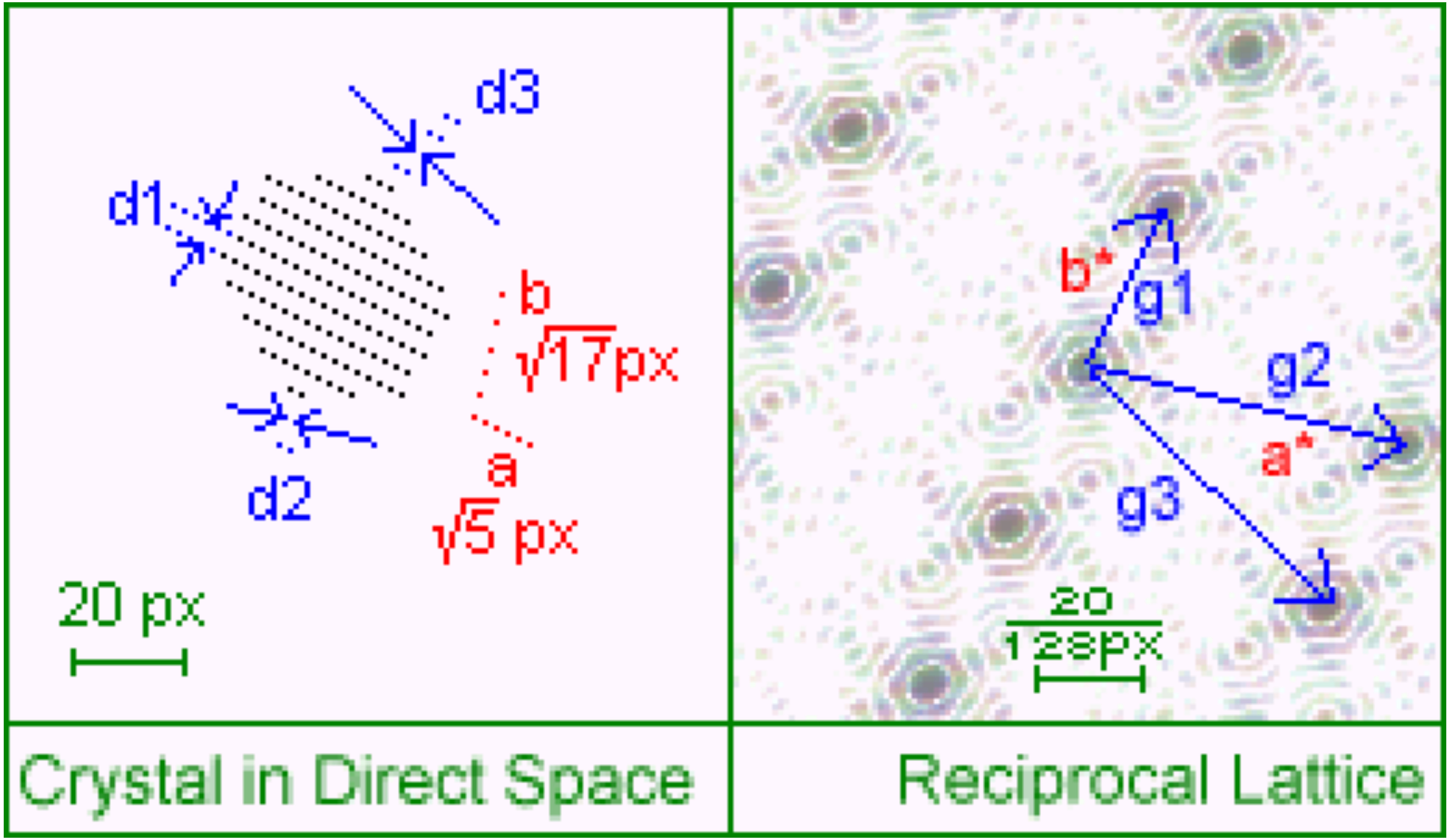
Wikimedia Commons

Diffraction on Molecular Crystals



The Royal Institution

Fourier Transform



Wikimedia Commons

Questions?

Nick Veasey