Scattering by a Multi-Electron Atom, Atomic Scattering Factors; Wave Propagation and Refractive Index

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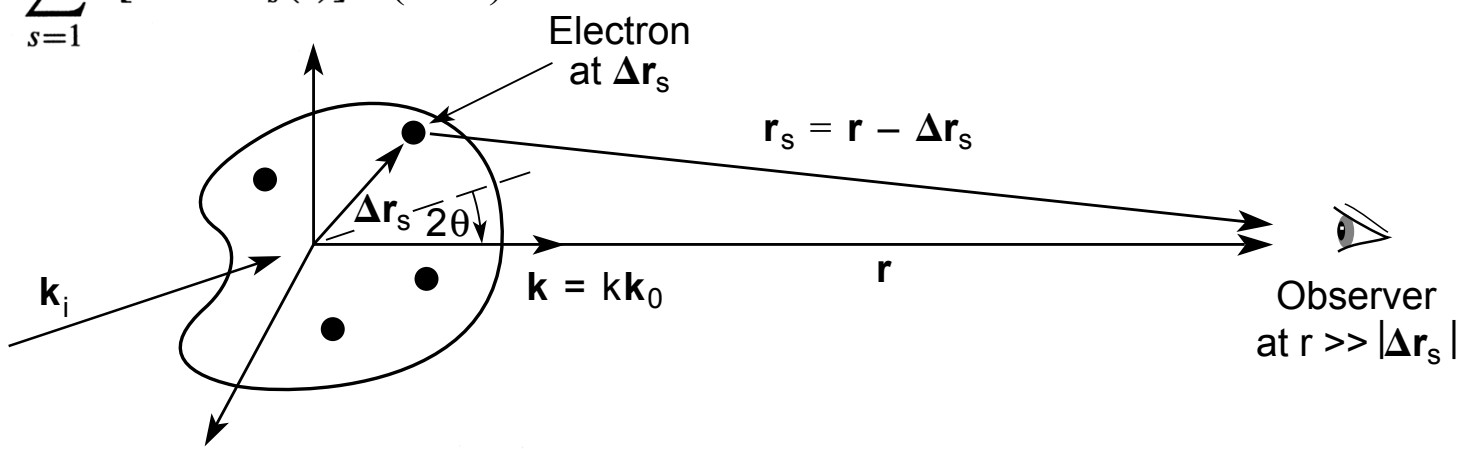
Scattering by a Multi-Electron Atom

Semi-classical model of an atom with Z electrons and nucleus of charge +Ze at $\mathbf{r} = 0$.

$$n(\mathbf{r}, t) = \sum_{s=1}^{Z} \delta[\mathbf{r} - \Delta\mathbf{r}_s(t)] \quad (2.53)$$



For each electron
$$m\frac{d^2\mathbf{x}_s}{dt^2} + m\gamma\frac{d\mathbf{x}_s}{dt} + m\omega_s^2\mathbf{x}_s = -e(\mathbf{E}_i + \underbrace{\mathbf{v}_s \times \mathbf{B}}_{\simeq 0}) \quad (2.58)$$

The acceleration has an additional phase term due to the position, $\Delta\mathbf{r}_s$, within the atom:

$$\mathbf{a}_s(t) = \frac{-\omega^2}{\omega^2 - \omega_s^2 + i\gamma\omega}\frac{e}{m}\mathbf{E}_i\, e^{-i(\omega t - \mathbf{k}_i\cdot\Delta\mathbf{r}_s)} \quad (2.61)$$

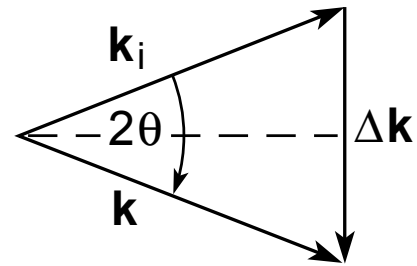
The scattered electric field at a distance $\mathbf{r}$ summed for all Z electrons, is

$$E(\mathbf{r}, t) = \frac{-e^2}{4\pi\epsilon_0 mc^2}\sum_{s=1}^{Z}\frac{\omega^2 E_i \sin\Theta}{\omega^2 - \omega_s^2 + i\gamma\omega}\frac{1}{r_s}e^{-i[\omega(t - r_s/c) - \mathbf{k}_i\cdot\Delta\mathbf{r}_s]}$$

where $\mathbf{r}_s \equiv \mathbf{r} - \Delta\mathbf{r}_s$ and $r_s = |\mathbf{r}_s|$. For $r >> \Delta\mathbf{r}_s$, $r_s \simeq r - \mathbf{k}_0 \cdot \Delta\mathbf{r}_s$ (2.62)

Scattering by a Multi-Electron Atom (continued)

$$E(\mathbf{r},t) = -\frac{r_e}{r}\underbrace{\left[\sum_{s=1}^{Z}\frac{\omega^2 e^{-i\Delta\mathbf{k}\cdot\Delta\mathbf{r}_s}}{\omega^2-\omega_s^2+i\gamma\omega}\right]}_{f(\Delta\mathbf{k},\,\omega)} E_i \sin\Theta\, e^{-i\omega(t-r/c)} \qquad (2.65)$$



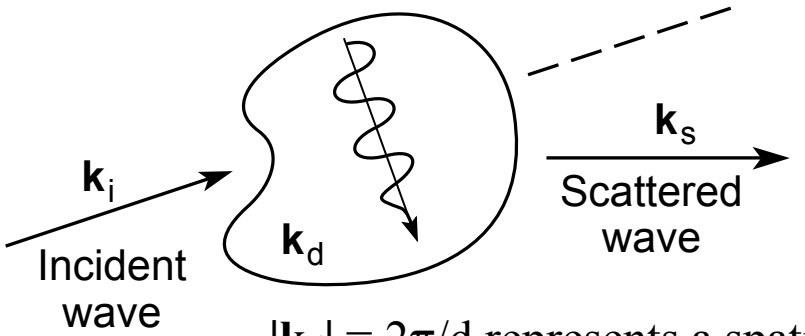
$\Delta\mathbf{k} = \mathbf{k} - \mathbf{k}_i$

$|\Delta\mathbf{k}| = 2k_i \sin\theta$

where the quantity f($\Delta\mathbf{k}$, ω) is the complex atomic scattering factor, which tells us the scattered electric field due to a multi-electron atom, relative to that of a single free electron (eq. 2.43). Note the dependence on frequency ω (photon energy $\hbar\omega$), the various resonant frequencies ω_s (resonant energies $\hbar\omega_s$), and the phase terms due to the various positions of electrons within the atom, $\Delta\mathbf{k} \cdot \Delta\mathbf{r}_s$.

A General Scattering Diagram

$$\mathbf{J}(r,t) = -e\, n(r,t)\mathbf{v}(r,t) \qquad (2.10)$$



$$\mathbf{J}_{\text{scatt}} e^{-i(\omega_s t - \mathbf{k}_s\cdot\mathbf{r})} = -e f^0(\omega_i) n_d e^{-i(\omega_d t - \mathbf{k}_d\cdot\mathbf{r})} \frac{-e\mathbf{E}_i}{-i\omega_i m} e^{-i(\omega_i t - \mathbf{k}_i\cdot\mathbf{r})}$$

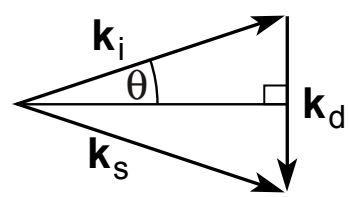
matching exponents

$$\omega_s = \omega_i + \omega_d$$
$$\mathbf{k}_s = \mathbf{k}_i + \mathbf{k}_d$$

$|\mathbf{k}_d| = 2\pi/d$ represents a spatial non-uniformity in the medium, such as atoms of periodicity d, a grating, or a density distribution due to a wave motion.

If the density distribution is stationary

$$\left.\begin{aligned} |\mathbf{k}_i| &= \frac{\omega}{c} = \frac{2\pi}{\lambda} \\ |\mathbf{k}_s| &= \frac{\omega}{c} = \frac{2\pi}{\lambda} \end{aligned}\right\} \therefore \text{ the scattering diagram is isosceles}$$



$$\mathbf{k}_i + \mathbf{k}_d = \mathbf{k}_s$$

$$\sin\theta = \frac{k_d/2}{k_i}$$

$$\sin\theta = \frac{\lambda}{2d}$$

$$\lambda = 2d\sin\theta \qquad (2.62)$$

(Bragg's Law, 1913)

(Reference: See chapter 4, eqs. 4.1 to 4.6)

The Atomic Scattering Factor

$$f(\Delta\mathbf{k},\omega) = \sum_{s=1}^{Z} \frac{\omega^2 e^{-i\Delta\mathbf{k}\cdot\Delta\mathbf{r}_s}}{\omega^2 - \omega_s^2 + i\gamma\omega} \qquad (2.66)$$

In general the $\Delta\mathbf{k} \cdot \Delta\mathbf{r}_s$ phase terms do not simplify, but in two cases they do. Noting that $|\Delta\mathbf{k}| = 2k_i \sin\theta = 4\pi/\lambda \sin\theta$, and that the radius of the atom is of order the Bohr radius, a_0, the phase factor is then bounded by

$$|\Delta\mathbf{k} \cdot \Delta\mathbf{r}_s| \leq \frac{4\pi a_0}{\lambda} \sin\theta \qquad (2.70)$$

The atomic scattering factor $f(\Delta\mathbf{k},\omega)$ simplifies significantly when

$$|\Delta\mathbf{k} \cdot \Delta\mathbf{r}_s| \to 0 \quad \text{for } a_0/\lambda \ll 1 \quad \text{(long wavelength limit)} \qquad (2.71a)$$

$$|\Delta\mathbf{k} \cdot \Delta\mathbf{r}_s| \to 0 \quad \text{for } \theta \ll 1 \quad \text{(forward scattering)} \qquad (2.71b)$$

In each of these two cases the atomic scattering factor $f(\Delta\mathbf{k},\omega)$ reduces to

$$f^{0}(\omega) = \sum_{s=1}^{Z} \frac{\omega^2}{\omega^2 - \omega_s^2 + i\gamma\omega} \qquad (2.72)$$

where we denote these special cases by the superscript zero.

Complex Atomic Scattering Factors

$$f^0(\omega) = \sum_{s=1}^{Z} \frac{\omega^2}{\omega^2 - \omega_s^2 + i\gamma\omega} = f_1^0(\omega) - i f_2^0(\omega) \qquad \begin{matrix}(2.72)\\(2.79)\end{matrix}$$

which some write as

$$f(\omega) = Z - f_1(\omega) - i f_2(\omega)$$

Atomic Scattering Cross-Sections

Comparing the scattered electric field for a multi-electron atom (2.65) with that for the free electron (2.43), the atomic scattering cross-sections are readily determined by the earlier proceedures to be

$$\frac{d\sigma(\omega)}{d\Omega} = r_e^2 |f^0(\omega)|^2 \sin^2 \Theta \qquad (2.75)$$

$$\sigma(\omega) = \frac{8\pi}{3} r_e^2 |f^0(\omega)|^2 \qquad (2.76)$$

where

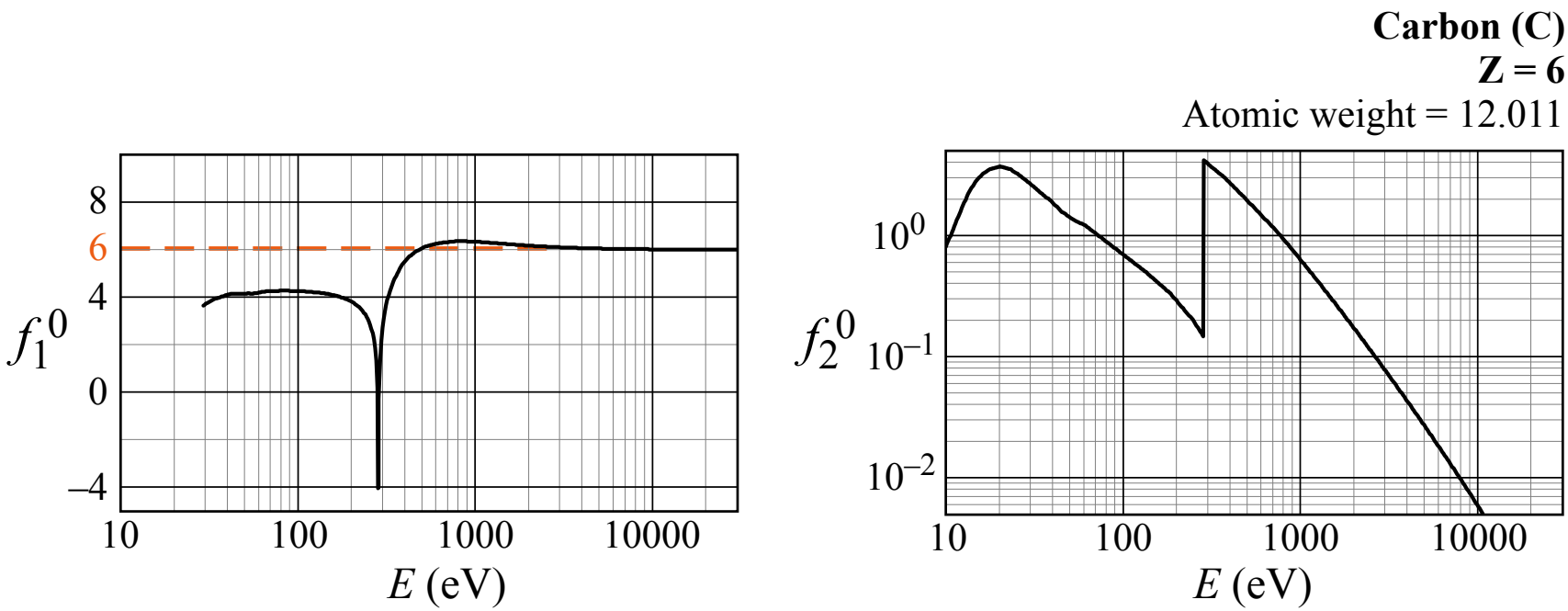
$$f^0(\omega) = \sum_s \frac{g_s \omega^2}{\omega^2 - \omega_s^2 + i\gamma\omega} \qquad (2.77)$$

and where the super-script zero refers to the special circumstances of long wavelength ($\lambda >> a_0$) or forward scattering ($\theta << 1$). With the Bohr radius $a_0 = 0.529$ Å, the long wavelength condition is easily satisfied for soft x-rays and EUV. Note too that we have introduced the concept of oscillator strengths, g_s, associated with each resonance, normalized by the condition

$$\sum_s g_s = Z \qquad (2.73)$$

Example:
Complex Atomic Scattering Factor for Carbon

$$f^0(\omega) = f_1^0(\omega) - i f_2^0(\omega) \qquad (2.79)$$



(Henke and Gullikson; www-cxro.LBL.gov)

Note that for $\hbar\omega >> \hbar\omega_s$, $f_1^0 \rightarrow Z$. This works here for carbon $f_1^0 \rightarrow 6$, but note that in general this conflicts with the condition $\lambda >> a_0$. For the case of carbon at 4 Å wavelength ($\lambda >> a_0$), and thus $\hbar\omega = 3$ keV ($>> \hbar\omega_s \sim 274$ eV), the atomic scattering cross-section (2.76) becomes

$$\sigma(\omega) \simeq \frac{8\pi}{3} r_e^2 Z^2 = Z^2 \sigma_e \qquad (2.78c)$$

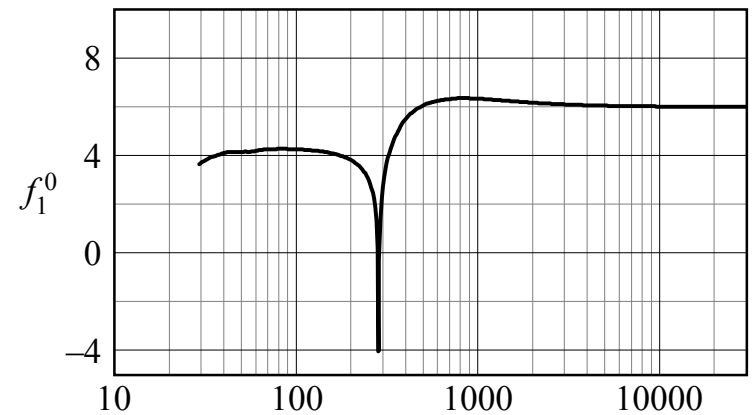
that is, all Z electrons are scattering cooperatively (in-phase) - the so-called N^2 effect.

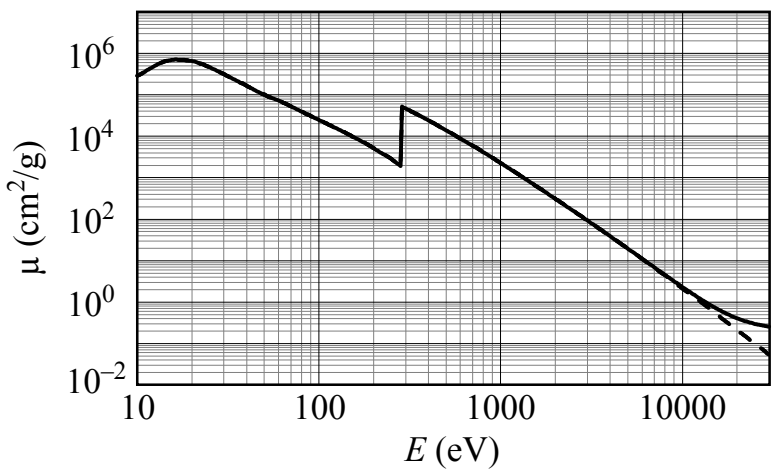
Atomic Scattering Factors for Carbon (Z = 6)

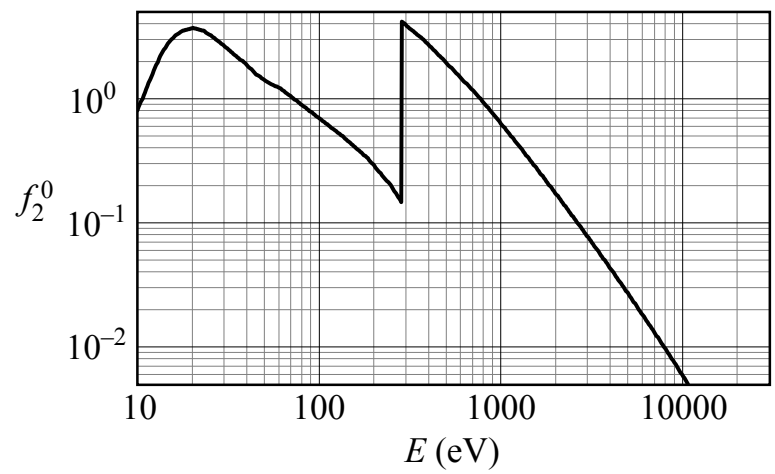
σ_a(barns/atom) = μ(cm^2/g) × 19.95
E(keV)μ(cm^2/g) = f_2^0 × 3503.31

Carbon (C)
Z = 6
Atomic weight = 12.011

Energy (eV)	f_1^0	f_2^0	μ (cm^2/g)
30	3.692	2.664E+00	3.111E+05
70	4.249	1.039E+00	5.201E+04
100	4.253	6.960E-01	2.438E+04
300	2.703	3.923E+00	4.581E+04
700	6.316	1.174E+00	5.878E+03
1000	6.332	6.328E–01	2.217E+03
3000	6.097	7.745E–02	9.044E+01
7000	6.025	1.306E–02	6.536E+00
10000	6.013	5.892E–03	2.064E+00
30000	6.000	4.425E–04	5.168E–02







Edge Energies: K 284.2 eV

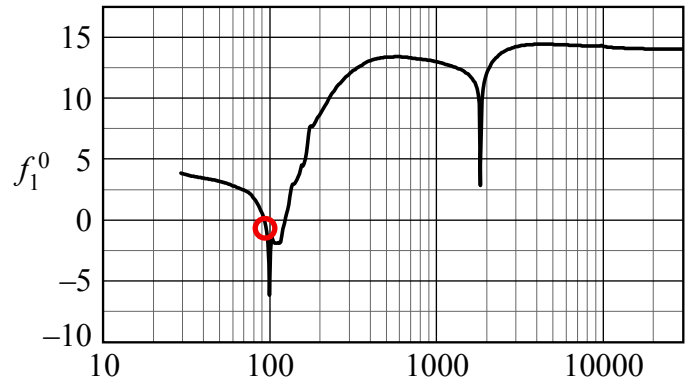
(Henke and Gullikson; www-cxro.LBL.gov)

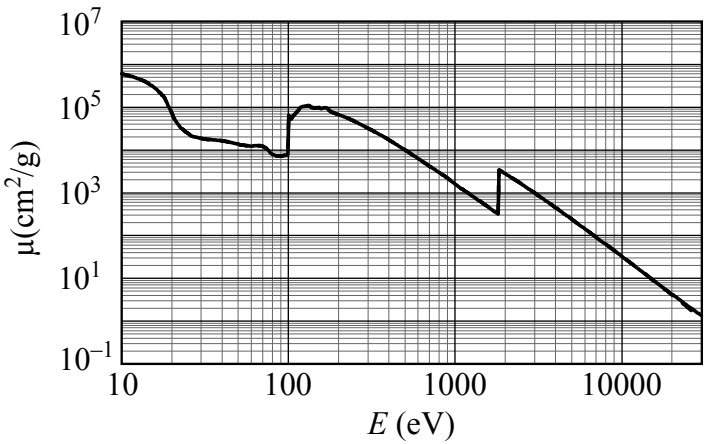
Atomic Scattering Factors for Silicon (Z = 14)

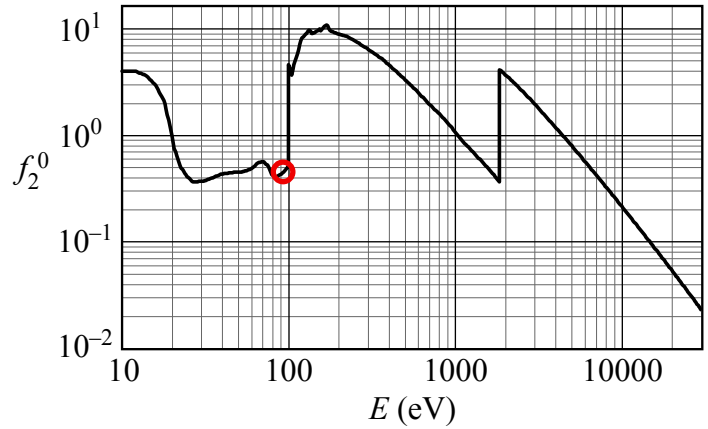
σ_a(barns/atom) = μ(cm^2/g) × 46.64
E(keV)μ(cm^2/g) = f_2^0 × 1498.22

Silicon (Si)
Z = 14
Atomic weight = 28.086

Energy (eV)	f_1^0	f_2^0	μ(cm^2/g)
30	3.799	3.734E–01	1.865E+04
70	2.448	5.701E–01	1.220E+04
100	–5.657	4.580E+00	6.862E+04
300	12.00	6.439E+00	3.216E+04
700	13.31	1.951E+00	4.175E+03
1000	13.00	1.070E+00	1.602E+03
3000	14.23	1.961E+00	9.792E+02
7000	14.33	4.240E–01	9.075E+01
10000	14.28	2.135E–01	3.199E+01
30000	14.02	2.285E–02	1.141E+00







Edge Energies: K 1838.9 eV L_1 149.7 eV
L_2 99.8 eV
L_3 99.2 eV

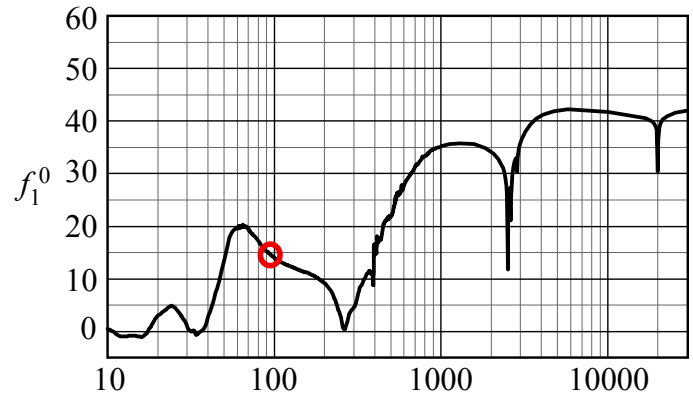
(Henke and Gullikson; www-cxro.LBL.gov)

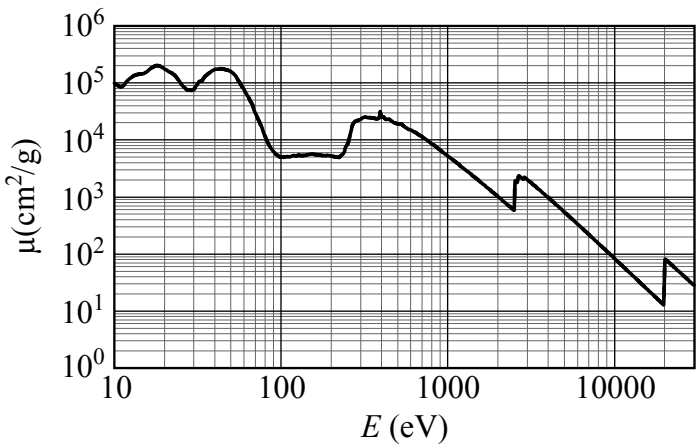
Atomic Scattering Factors for Molybdenum (Z = 42)

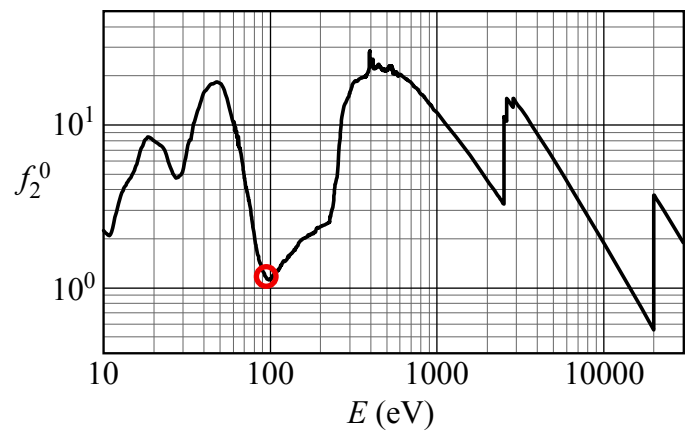
σ_a(barns/atom) = μ(cm^2/g) × 159.31
E(keV)μ(cm^2/g) = f_2^0 × 438.59

Molybdenum (Mo)
Z = 42
Atomic weight = 95.940

Energy (eV)	f_1^0	f_2^0	μ(cm^2/g)
30	1.071	5.292E+00	7.736E+04
70	19.38	4.732E+00	2.965E+04
100	14.02	1.124E+00	4.931E+03
300	4.609	1.568E+01	2.292E+04
700	31.41	1.819E+01	1.140E+04
1000	35.15	1.188E+01	5.210E+03
3000	35.88	1.366E+01	1.997E+03
7000	42.11	3.493E+00	2.189E+02
10000	41.67	1.881E+00	8.248E+01
30000	42.04	1.894E+00	2.769E+01



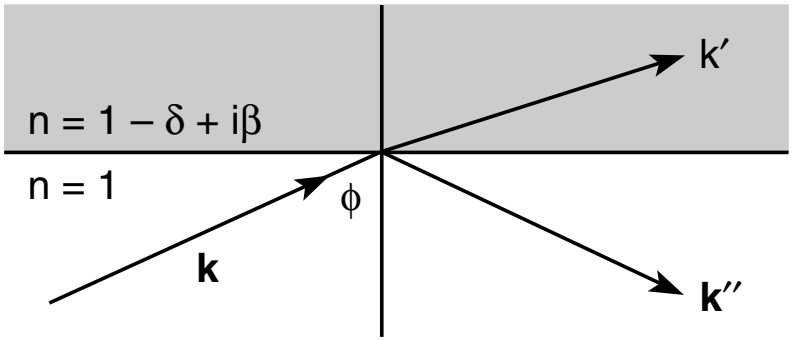




Edge Energies:	K	19999.5 eV	L_1	2865.5 eV	M_1	506.3 eV	N_1	63.2 eV
			L_2	2625.1 eV	M_2	411.6 eV	N_2	37.6 eV
			L_3	2520.2 eV	M_3	394.0 eV	N_3	35.5 eV
					M_4	231.1 eV		
					M_5	227.9 eV		

(Henke and Gullikson; www-cxro.LBL.gov)

Wave Propagation and Refractive Index at X-Ray Wavelengths



$$n(\omega) = 1 - \frac{n_a r_e \lambda^2}{2\pi}\left(f_1^0 - i f_2^0\right) \qquad (3.9)$$

$$n(\omega) = 1 - \delta + i\beta \qquad (3.12)$$

$$l_{\text{abs}} = \frac{\lambda}{4\pi\beta} \qquad (3.22)$$

$$\sigma_{\text{abs.}} = 2 r_e \lambda f_2^0(\omega) \qquad (3.28)$$

$$\Delta\phi = \left(\frac{2\pi\delta}{\lambda}\right)\Delta r \qquad (3.29)$$

$$\theta_c = \sqrt{2\delta} \qquad (3.41)$$

$$R_{s,\perp} \simeq \frac{\delta^2 + \beta^2}{4} \qquad (3.50)$$

$$\phi_B \simeq \frac{\pi}{4} - \frac{\delta}{2} \qquad (3.60)$$

The Wave Equation and Refractive Index

The transverse wave equation is

$$\left(\frac{\partial^2}{\partial t^2} - c^2\nabla^2\right)\mathbf{E}_{\mathrm{T}}(\mathbf{r}, t) = -\frac{1}{\epsilon_0}\frac{\partial \mathbf{J}_{\mathrm{T}}(\mathbf{r}, t)}{\partial t} \qquad (3.1)$$

For the special case of forward scattering the positions of the electrons within the atom ($\Delta\mathbf{k} \cdot \Delta\mathbf{r}_s$) are irrelevant, as are the positions of the atoms themselves, n($\mathbf{r}$, t). The contributing current density is then

$$\mathbf{J}_0(\mathbf{r}, t) = -en_a \sum_s g_s \mathbf{v}_s(\mathbf{r}, t) \qquad (3.2)$$

where n_a is the average density of atoms, and

$$\sum_s g_s = Z$$

The Wave Equation and Refractive Index (Continued)

The oscillating electron velocities are driven by the incident field **E**

$$\mathbf{v}(\mathbf{r}, t) = \frac{e}{m} \frac{1}{\left(\omega^2 - \omega_s^2\right) + i\gamma\omega} \frac{\partial \mathbf{E}(\mathbf{r}, t)}{\partial t} \tag{3.2}$$

such that the contributing current density is

$$\mathbf{J}_0(\mathbf{r}, t) = -\frac{e^2 n_a}{m} \sum_s \frac{g_s}{\left(\omega^2 - \omega_s^2\right) + i\gamma\omega} \frac{\partial \mathbf{E}(\mathbf{r}, t)}{\partial t} \tag{3.4}$$

Substituting this into the transverse wave equation (3.1), one has

$$\left(\frac{\partial^2}{\partial t^2} - c^2 \nabla^2\right) \mathbf{E}_{\mathrm{T}}(\mathbf{r}, t) = \frac{e^2 n_a}{\epsilon_0 m} \sum_s \frac{g_s}{\left(\omega^2 - \omega_s^2\right) + i\gamma\omega} \frac{\partial^2 \mathbf{E}_{\mathrm{T}}(\mathbf{r}, t)}{\partial t^2}$$

Combining terms with similar operators

$$\left[\left(1 - \frac{e^2 n_a}{\epsilon_0 m} \sum_s \frac{g_s}{\left(\omega^2 - \omega_s^2\right) + i\gamma\omega}\right) \frac{\partial^2}{\partial t^2} - c^2 \nabla^2\right] \mathbf{E}_{\mathrm{T}}(\mathbf{r}, t) = 0 \tag{3.5}$$

Refractive Index in the Soft X-Ray and EUV Spectral Region

Written in the standard form of the wave equation as

$$\left[\frac{\partial^2}{\partial t^2} - \frac{c^2}{n^2(\omega)}\nabla^2\right]\mathbf{E}_{\mathrm{T}}(\mathbf{r}, t) = 0 \qquad (3.6)$$

The frequency dependent refractive index n(ω) is identified as

$$n(\omega) \equiv \left[1 - \frac{e^2 n_a}{\epsilon_0 m}\sum_s \frac{g_s}{\left(\omega^2 - \omega_s^2\right) + i\gamma\omega}\right]^{1/2} \qquad (3.7)$$

For EUV/SXR radiation ω^2 is very large compared to the quantity $e^2 n_a/\epsilon_0 m$, so that to a high degree of accuracy the index of refraction can be written as

$$n(\omega) = 1 - \frac{1}{2}\frac{e^2 n_a}{\epsilon_0 m}\sum_s \frac{g_s}{\left(\omega^2 - \omega_s^2\right) + i\gamma\omega} \qquad (3.8)$$

which displays both positive and negative dispersion, depending on whether ω is less or greater than ω_s. Note that this will allow the refractive index to be more or less than unity, and thus the phase velocity to be less or greater than c.

Refractive Index in the Soft X-Ray and EUV Spectral Region (continued)

$$n(\omega) = 1 - \frac{1}{2}\frac{e^2 n_a}{\epsilon_0 m}\sum_s \frac{g_s}{\left(\omega^2 - \omega_s^2\right) + i\gamma\omega} \qquad (3.8)$$

Noting that

$$r_e = \frac{e^2}{4\pi\epsilon_0 m c^2}$$

and that for forward scattering

$$f^0(\omega) = \sum_s \frac{g_s \omega^2}{\omega^2 - \omega_s^2 + i\gamma\omega}$$

where this has complex components

$$f^0(\omega) = f_1^0(\omega) - i f_2^0(\omega)$$

The refractive index can then be written as

$$\boxed{n(\omega) = 1 - \frac{n_a r_e \lambda^2}{2\pi}\left[f_1^0(\omega) - i f_2^0(\omega)\right]} \qquad (3.9)$$

which we write in the simplified form

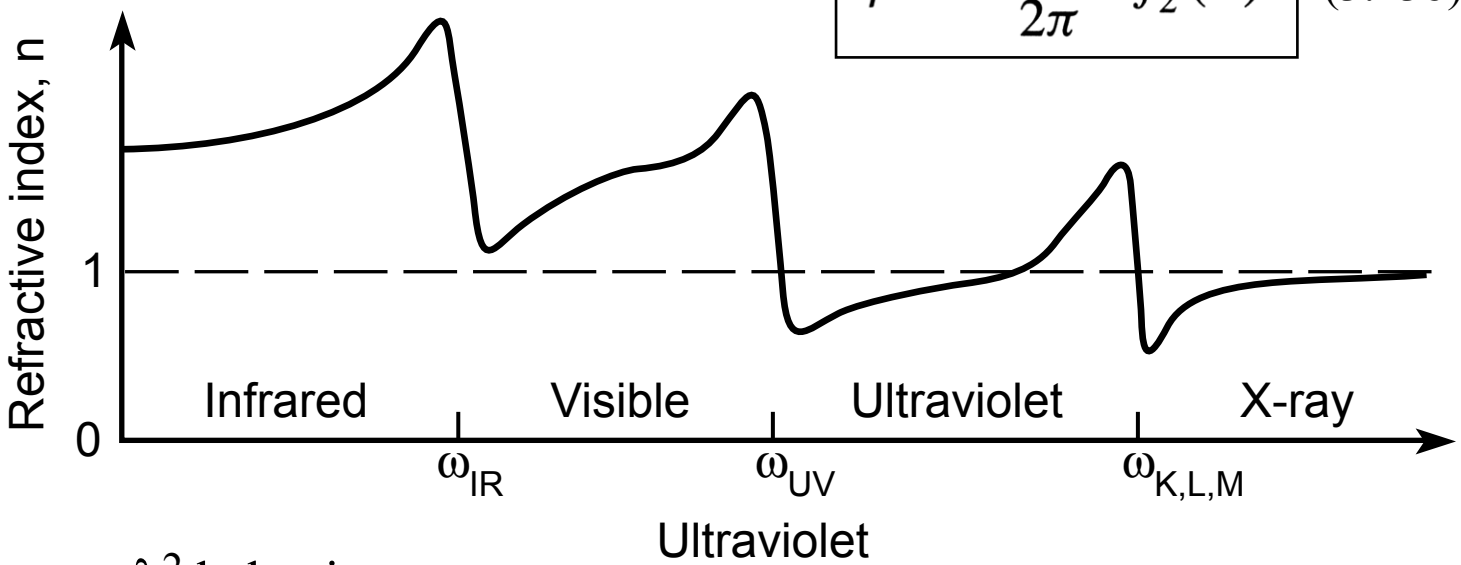
$$\boxed{n(\omega) = 1 - \delta + i\beta} \qquad (3.12)$$

Refractive Index from the IR to X-Ray Spectral Region

$$n(\omega) = 1 - \delta + i\beta \quad (3.12)$$

$$\delta = \frac{n_a r_e \lambda^2}{2\pi} f_1^0(\omega) \quad (3.13a)$$

$$\beta = \frac{n_a r_e \lambda^2}{2\pi} f_2^0(\omega) \quad (3.13b)$$



- λ^2 behavior
- δ & $\beta << 1$
- δ-crossover

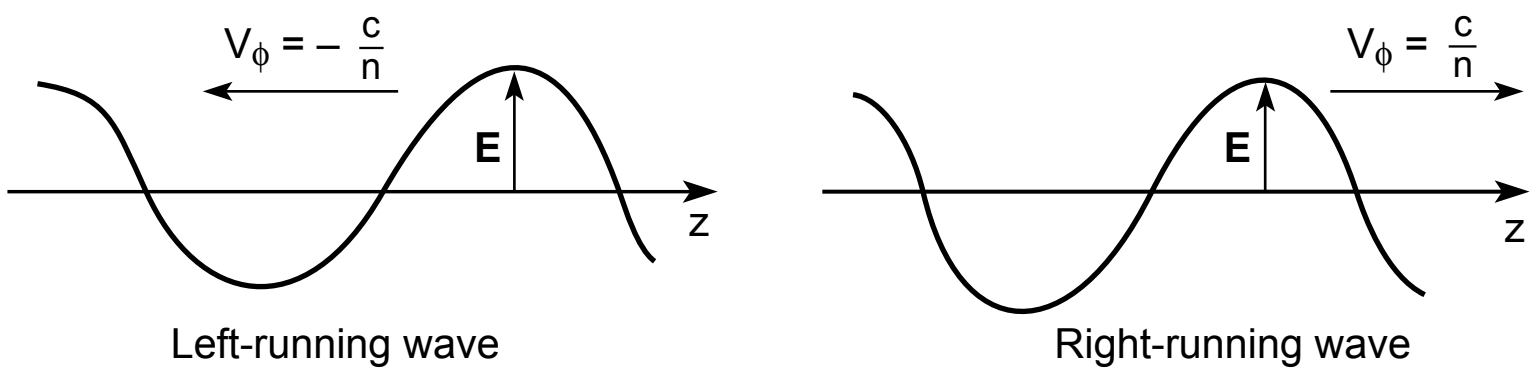
Phase Velocity and Refractive Index

The wave equation can be written as

$$\left(\frac{\partial}{\partial t} - \frac{c}{n(\omega)}\nabla\right)\left(\frac{\partial}{\partial t} + \frac{c}{n(\omega)}\nabla\right)\mathbf{E}_{\mathrm{T}}(\mathbf{r}, t) = 0 \qquad (3.10)$$

The two bracketed operators represent left and right-running waves

$$\left(\frac{\partial}{\partial t} - \frac{c}{n}\frac{\partial}{\partial z}\right)E_x = 0 \qquad \left(\frac{\partial}{\partial t} + \frac{c}{n}\frac{\partial}{\partial z}\right)E_x = 0$$



where the phase velocity, the speed with which crests of fixed phase move, is not equal to c as in vacuum, but rather is

$$v_\phi = \frac{c}{n(\omega)} \qquad (3.11)$$

Phase Velocity and Refractive Index (continued)

Recall the wave equation

$$\left(\frac{\partial}{\partial t} - \frac{c}{n(\omega)}\nabla\right)\left(\frac{\partial}{\partial t} + \frac{c}{n(\omega)}\nabla\right)\mathbf{E}_{\mathrm{T}}(\mathbf{r}, t) = 0 \qquad (3.10)$$

Examining one of these factors, for a space-time dependence

$$\mathbf{E}_T = \mathbf{E}_0 \exp[-i(\omega t - kz)]$$

$$-i\left(\omega - \frac{ck}{n}\right) = 0$$

Solving for ω/k we have the phase velocity

$$V_\phi = \frac{\omega}{k} = \frac{c}{n}$$

Phase Variation and Absorption of Propagating Waves

For a plane wave $$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0 e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})} \tag{3.14}$$

in a material of refractive index n, the complex dispersion relation is

$$\frac{\omega}{k} = \frac{c}{n} = \frac{c}{1 - \delta + i\beta} \tag{3.15}$$

Solving for k

$$k = \frac{\omega}{c}(1 - \delta + i\beta) \tag{3.16}$$

Substituting this into (3.14), in the propagation direction defined by $\mathbf{k} \cdot \mathbf{r} = kr$

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0 e^{-i[\omega t - (\omega/c)(1-\delta+i\beta)r]}$$

or

$$\mathbf{E}(\mathbf{r}, t) = \underbrace{\mathbf{E}_0 e^{-i\omega(t-r/c)}}_{\text{vacuum propagation}} \underbrace{e^{-i(2\pi\delta/\lambda)r}}_{\phi\text{-shift}} \underbrace{e^{-(2\pi\beta/\lambda)r}}_{\text{decay}} \tag{3.17}$$

where the first exponential factor represents the phase advance had the wave been propagating in vacuum, the second factor (containing $2\pi\delta r/\lambda$) represents the modified phase shift due to the medium, and the factor containing $2\pi\beta r/\lambda$ represents decay of the wave amplitude.

Intensity and Absorption in a Material of Complex Refractive Index

For complex refractive index n

$$\mathbf{H}(\mathbf{r}, t) = n\sqrt{\frac{\epsilon_0}{\mu_0}}\mathbf{k}_0 \times \mathbf{E}(\mathbf{r}, t) \tag{3.18}$$

The average intensity, in units of power per unit area, is

$$\bar{I} = |\bar{\mathbf{S}}| = \tfrac{1}{2}|\mathrm{Re}(\mathbf{E} \times \mathbf{H}^*)| \tag{3.19}$$

or

$$\bar{I} = \frac{1}{2}\mathrm{Re}(n)\sqrt{\frac{\epsilon_0}{\mu_0}}|\mathbf{E}|^2 \tag{3.20}$$

Recalling that

$$\mathbf{E}(\mathbf{r}, t) = \underbrace{\mathbf{E}_0 e^{-i\omega(t-r/c)}}_{\text{vacuum propagation}} \underbrace{e^{-i(2\pi\delta/\lambda)r}}_{\phi\text{-shift}} \underbrace{e^{-(2\pi\beta/\lambda)r}}_{\text{decay}} \tag{3.17}$$

$$\bar{I} = \frac{1}{2}\mathrm{Re}(n)\sqrt{\frac{\epsilon_0}{\mu_0}}|\mathbf{E}_0|^2 e^{-2(2\pi\beta/\lambda)r}$$

or

$$\bar{I} = \bar{I}_0 e^{-(4\pi\beta/\lambda)r} \tag{3.21}$$

the wave decays with an exponential decay length

$$\boxed{l_{\mathrm{abs}} = \frac{\lambda}{4\pi\beta}} \tag{3.22}$$

Absorption Lengths

$$\boxed{l_{\text{abs}} = \frac{\lambda}{4\pi\beta}} \qquad (3.22)$$

Recalling that $\beta = n_a r_e \lambda^2 f_2^{\circ}(\omega)/2\pi$

$$l_{\text{abs}} = \frac{1}{2 n_a r_e \lambda f_2^0(\omega)} \qquad (3.23)$$

In Chapter 1 we considered experimentally observed absorption in thin foils, writing

$$\frac{\bar{I}}{\bar{I}_0} = e^{-\rho\mu r} \qquad (3.24)$$

where ρ is the mass density, μ is the absorption coefficient, r is the foil thickness, and thus $l_{\text{abs}} = 1/\rho\mu$. Comparing absorption lengths, the macroscopic and atomic descriptions are related by

$$\mu = \frac{2 r_e \lambda}{A m_u} f_2^0(\omega) \qquad (3.26)$$

where $\rho = m_a n_a = A m_u n_a$, m_u is the atomic mass unit, and A is the number of atomic mass units

Phase Shift Relative to Vacuum Propagation

For a wave propagating in a medium of refractive index $n = 1 - \delta + i\beta$

$$\mathbf{E}(\mathbf{r}, t) = \underbrace{\mathbf{E}_0 e^{-i\omega(t-r/c)}}_{\text{vacuum propagation}} \underbrace{e^{-i(2\pi\delta/\lambda)r}}_{\phi\text{-shift}} \underbrace{e^{-(2\pi\beta/\lambda)r}}_{\text{decay}} \qquad (3.23)$$

the phase shift $\Delta\phi$ relative to vacuum, due to propagation through a thickness Δr is

$$\boxed{\Delta\phi = \left(\frac{2\pi\delta}{\lambda}\right)\Delta r} \qquad (3.29)$$

- Flat mirrors at short wavelengths
- Transmissive, flat beamsplitters
- Bonse and Hart interferometer
- Diffractive optics for SXR/EUV