

CHAPTER - 3

RAYLEIGH SCATTERING NEAR EDGES - ANOMALOUS SCATTERING FACTOR PREDICTIONS

The fundamental problem in the theory of atomic Rayleigh scattering of photons arises from the binding of electrons in the atom. For scattering of low energy photons, effects of electron binding are crucial. When the incident photon energy happens to be close to the excitation energy of the target atom, electrons couple very strongly with the photons, and large variations in the elastic scattering cross section may occur. Such rapid variation of the elastic scattering cross section at energies near regions of atomic excitation or ionization thresholds is known as the anomalous scattering. This affects the scattering cross section through its modification of the forward scattering amplitude called the anomalous scattering factor or the anomalous dispersion correction.

3.1 Dispersion correction to form factor:

The simple form factor approach to the atomic elastic scattering of photons from bound electrons makes the simplifying assumption that each electron can be regarded as a Thomson free electron. The effects of electron binding are neglected in the calculation of scattering cross sections by

the form factor corrected Thomson formula. The free point charge of Thomson is simply replaced by a charge distribution of density equal to $|\psi|^2$ and scattering from this distribution is then determined. It is natural to expect that the value of the form factor for photon energy near the atomic absorption edge will differ appreciably from the value calculated using the formula

$$f(q) = \int |\psi|^2 e^{i\vec{q}\cdot\vec{r}} d^3r$$

This is expected even on purely classical grounds for when the incident frequency approaches that of one of the free vibrations of the system, resonance effects occur. The effect is closely related to anomalous dispersion.

The resonant phenomena in elastic photon-atom scattering has to be accounted for by modifying the atomic form factor as follows:

$$f_{\text{true}} = f_0(q) + f'(\omega) + i f''(\omega) \quad \text{--- (3.1)}$$

Here, $f_0(q)$ is the normal or non-anomalous form factor. The quantities $f'(\omega)$ and $f''(\omega)$ are called the real and imaginary parts of the correction to $f_0(q)$ arising from the anomalous dispersion effect.

3.2 Classical theory of anomalous scattering :

In the classical theory an electron is supposed to be bound to the atom with such a force that it oscillates with a natural frequency ω_0 . The damped harmonic oscillator model is then applied to an assembly of N atoms per unit volume, each containing an electron of mass m and charge e whose vibrations have definite angular frequency ω_0 and are characterized by a damping parameter γ . When subjected to the oscillating electric field of the incident electromagnetic radiation, they give rise to electric dipole oscillators. The oscillating dipole is the source of the scattered radiation at the same frequency.

The one dimensional motion of an electron under the influence of an oscillating field $E = E_0 e^{-i\omega t}$ can be described by the equation

$$\frac{d^2x}{dt^2} + \gamma \frac{dx}{dt} + \omega_0^2 x = \left(\frac{e}{m}\right) E_0 e^{-i\omega t}$$

After any transients have disappeared, the time dependence of the forced oscillation of the electron is $x = x_0 e^{-i\omega t}$.

This gives an expression for the dipole moment induced by the radiation:

$$p = \frac{(e^2/m) E_0 e^{-i\omega t}}{\omega_0^2 - \omega^2 + i\gamma\omega}$$

The amplitude A of the scattered wave at unit distance in the equatorial plane is then shown to be given by

$$A = \left(\frac{e^2}{mc^2} \right) \frac{\omega^2 E_0}{\omega_0^2 - \omega^2 + i\gamma\omega}$$

If we put $\omega_0 = 0$ and $\gamma = 0$, we obtain the amplitude of scattering by a free electron.

$$A' = - \left(\frac{e^2}{mc^2} \right) E_0$$

We can now define the scattering factor f for the dipole as the ratio A/A' and obtain

$$\begin{aligned} f &= - \frac{\omega^2}{\omega_0^2 - \omega^2 + i\gamma\omega} \\ &= \frac{\omega^2 (\omega^2 - \omega_0^2)}{(\omega^2 - \omega_0^2)^2 + \gamma^2 \omega^2} + i \frac{\gamma \omega^3}{(\omega^2 - \omega_0^2)^2 + \gamma^2 \omega^2} \\ &= \eta + i\xi \end{aligned}$$

unless ω is very nearly equal to ω_0 and so far as the damping is small,

$$\eta = \frac{\omega^2}{\omega^2 - \omega_0^2}$$

This is, however, for a single dipolar radiation field. If the electrons are thought of as occupying energy levels within the atom characterized by frequency ω_n and damping factor γ_n , then the real part of the scattering factor is written as

$$\eta = \sum \frac{g_n \omega^2}{\omega^2 - \omega_n^2}$$

We call g_n the oscillator strength corresponding to the natural frequency ω_n . Now

$$\sum \frac{g_n \omega^2}{\omega^2 - \omega_n^2} = \sum g_n - \sum \frac{g_n \omega_n^2}{\omega_n^2 - \omega^2}$$

and hence $\eta = f_0 + f'$ where we have put

$$\begin{aligned} f_0 &= \sum g_n \\ f' &= \sum \frac{g_n \omega_n^2}{\omega^2 - \omega_n^2} \end{aligned} \quad \text{--- (3.2)}$$

Also, for X-rays, the refractive index n of a medium is given by

$$n = 1 - \frac{2\pi N e^2}{m \omega^2} f$$

When ω is comparable with ω_n , f will be complex. The medium will have a complex refractive index. The imaginary part of the complex refractive index corresponds to an absorption of radiation, that is, to a decrease in intensity of the wave with increasing penetration. $\text{Im } n$ is thus related to the linear absorption coefficient.

$$\mu_{\ell} = \left(\frac{4\pi}{\lambda} \right) \text{Im } n \quad \text{--- (3.3) a}$$

Taking

$$n = 1 - \frac{2\pi N e^2}{m \omega^2} (f_0 + f' + i f'')$$

We obtain

$$\text{Im } n = - \frac{2\pi N e^2}{m \omega^2} f''$$

and hence

$$\begin{aligned} f'' &= - \frac{m \omega^2}{2\pi N e^2} \text{Im } n = - \frac{m \omega^2}{2\pi N e^2} \cdot \frac{\lambda}{4\pi} \mu_{\ell} \\ &= - \frac{\omega}{4\pi \nu_0 c} \sigma_{\text{abs}} \quad \text{--- (3.3) b} \end{aligned}$$

3.3 Dispersion relation between f' and f'' :

The results for anomalous dispersion correction terms f' and f'' which were derived in the preceding section using purely classical arguments have been found to be identical with the results gained from quantum mechanics. The result in eqn. (3.3) b is the same as the optical theorem in the quantum theory of scattering connecting the total cross section and the imaginary part of the elastic scattering amplitude in the forward direction. It is a general practice in the description of anomalous scattering of photons from atoms to regard f' a real number representing the dispersion effect in scattering arising from the proximity of the incident photon energy to the binding energy of the scattering electron and f'' an imaginary number representing absorption due to damping of the radiation in the vicinity of resonance level.

With the corrections for anomalous dispersion effect included in the atomic scattering factor, the expressions for Rayleigh amplitudes become

$$A_{\perp}^R = -r_0 \left[f_0(q) + f'(\omega) + i f''(\omega) \right]$$

$$A_{\parallel}^R = -r_0 \left[f_0(q) + f'(\omega) + i f''(\omega) \right] \cos \Theta$$

If we consider only the forward amplitudes for an isolated atom we may write

$$\begin{aligned} \operatorname{Re} A^R &= -r_0 [f_0 + f'(\omega)] \\ \operatorname{Im} A^R &= -r_0 f''(\omega) \end{aligned}$$

The forward imaginary amplitude is given by $\operatorname{Im} A^R = \frac{\omega}{4\pi c} \sigma_{abs}$ and it follows that

$$f''(\omega) = -\frac{\omega}{4\pi r_0 c} \sigma_{abs}$$

σ_{abs} is the total photoelectric cross section from a photon of the given energy off the atom in question.

To obtain real forward anomalous amplitude f' , a dispersion relation connecting f' and f'' is usually employed. It was observed by Kramers and Kronig³³ that in the theory of dispersion of light by atom there exists a relation between the real and imaginary parts of the complex refractive index. Such relations were later found to be of very general kind, and the integral relation between the real and imaginary parts of any frequency dependent quantity came to be known as the dispersion relation. For scattering, the dispersion relation is between the real and imaginary parts of the complex amplitude. Consequently

$$\begin{aligned} f'(\omega) &= \left(\frac{2}{\pi}\right) P \int_0^{\infty} d\omega' \frac{\omega' f''(\omega')}{\omega'^2 - \omega^2} \\ &= \left(\frac{1}{2\pi r_0^2 c}\right) P \int_0^{\infty} d\omega' \frac{\omega'^2}{\omega^2 - \omega'^2} \sigma_{abs}(\omega') \end{aligned}$$

The symbol P refers to the integration about the point where the integrand becomes infinite. This implies that a small interval centred about $w = w'$ is excluded from the range of integration. The procedure is known as taking the 'principal value' of the integral.

3.4 Angular dependence of f' and f'' :

The dispersion relation linking f' and f'' is valid only for forward angle scattering ($\theta = 0$) and provides no information concerning the angular dependence of the anomalous dispersion corrections. It had often been thought that the dispersion corrections should exhibit some functional dependence on scattering angle θ . The work of Wagenfeld³⁴ based on non-relativistic quantum mechanics predicted that the values of f' and f'' were functions of momentum transfer q . This prediction is not supported by modern approaches which use relativistic quantum mechanics. Some authors ascribe to these corrections the same functional dependence on the angle of scattering as the form factor. The difficulties with such assumption have been pointed out by Pratt and co-workers³⁵⁻³⁶. It has been found more useful to treat f' and f'' as the angle independent correction terms to be added to form factor in obtaining the Rayleigh amplitudes.

3.5 Tables of f' and f'' :

table

The first acceptable of the X-ray dispersion terms was made by H. Honl³⁷ in 1933. The work was extended by Wagenfeld³⁴. The method used by them is based on a completely fundamental calculation of oscillator density $\frac{dg}{d\omega}$ and an evaluation for each electron sub shell such as

$$f'_k(\omega) = \int_0^{\infty} \frac{\omega^2 \left(\frac{dg}{d\omega} \right)_k}{\omega^2 - \omega_k^2} d\omega$$

$$f''_k(\omega) = \left(\frac{\pi}{2} \right) \omega \left(\frac{dg}{d\omega} \right)_k$$

Another method makes use of the relationship between the theoretical photoelectric absorption cross section $\sigma_{abs}^{PE}(\omega)$, hence f'' and the real part f' of the anomalous scattering factor. In 1970, Cromer and Liberman³⁸ made an extensive calculation of the anomalous X-ray atomic scattering factors using the optical theorem and dispersion relation together with theoretical photoeffect cross sections. To develop their tables, Cromer and Liberman used the Brysk and Zerby³⁹ computer code for the calculation of photoelectric cross sections based on Dirac-Slater relativistic wave functions. More recently Henke et al⁴⁰ have made an extensive compilation of experimental and theoretical photoelectric cross sections for the elements $Z = 1$ to $Z = 94$. These data were then used to compute f' between 100 eV and 2 keV from the dispersion integral.

Experimental measurements of the dispersion corrections involve techniques which measure refractive indices or attenuation coefficients. The oldest technique for measuring refractive index is based on the measurement of deviation produced when a prism of the material under investigation is placed in the photon beam. X-ray interferometer techniques are now used extensively for the measurement of refractive index of materials and hence of f' . Linear attenuation coefficient measurements over an extended energy range can also be used as a basis for the determination of f'' by virtue of the relation in equation (3.3 a). Creagh and co-workers⁴¹⁻⁴⁵ have made use of this method to determine the dispersion corrections for a number of elements.

In the relativistic treatment presented by Cromer and Liberman³⁸, an energy-independent term was given as a correction to the usual dispersion formula for f' . The non-relativistic value for f' was reduced by an amount $\frac{5 E_{tot}}{3 mc^2}$ (where E_{tot} is the total ground state energy of the atom) for all photon energies. The use of dipole approximation by Cromer and Liberman to compute the high energy limit results in an error that affects their f' values at all energies. Kissel has found that correcting the high energy limit of the anomalous scattering factor predictions of Cromer and Liberman brings those predictions into much better agreement with S-matrix calculations near absorption edges.

Table 8. Anomalous scattering factor predictions of elastic scattering of 59.54 keV photons based on relativistic form factors plus dispersion corrections of Creagh and McAuley

$${}^{42}\text{Mo} (E_k = 20.002 \text{ keV})$$

Scattering angle (deg) Θ	Momentum transfer x (\AA^{-1})	$d\sigma/d\Omega$ (b/atom/Sr)	$(R_e A_{11}, I_m A_{11})$		$(R_f A_{11}, I_m A_{11})$	
0	0	142.00	-42.10	0.548	-42.10	0.548
5	0.209	85.80	-32.70	0.546	-32.80	0.548
10	0.418	45.70	-23.70	0.539	-24.10	0.548
30	1.24	7.73	- 9.09	0.474	-10.50	0.548
60	2.40	1.33	- 2.57	0.274	- 5.14	0.548
90	3.39	0.502	0	0	- 3.50	0.548
120	4.16	0.344	1.28	-0.274	- 2.57	0.548
150	4.63	0.347	1.87	-0.474	- 2.16	0.548

Table 9.

 $^{48}\text{Cd} (E_k = 26.712 \text{ keV})$

Θ (deg)	α ($\text{A}^{\circ-1}$)	$d\sigma/d\Omega$ (b/atom/Sr)	$(R_{eA_{11}}$	$I_{mA_{11}})$	$(R_{eA_1}$	$I_{mA_1})$
0	0	185.00	-48.10	0.913	-48.10	0.913
5	0.209	117.00	-38.20	0.909	-38.40	0.913
10	0.418	60.30	-27.20	0.899	-27.70	0.913
30	1.24	12.40	-11.50	0.790	-13.30	0.913
60	2.40	1.71	-2.89	0.456	-5.78	0.913
90	3.39	0.745	0	0	-4.22	0.913
120	4.16	0.553	1.60	-0.456	-3.20	0.913
150	4.63	0.593	2.39	-0.790	-2.77	0.913

Table 10.

 $^{50}\text{Sn} (E_k = 29.190 \text{ keV})$

Θ (deg)	α ($\text{A}^{\circ-1}$)	$d\sigma/d\Omega$ (b/atom/Sr)	$(R_{eA_{11}}$	$I_{mA_{11}})$	$(R_{eA_1}$	$I_{mA_1})$
0	0	200.00	-50.00	1.06	-50.00	1.06
5	0.209	130.00	-40.30	1.05	-40.50	1.06
10	0.418	67.10	-28.70	1.04	-29.20	1.06
30	1.24	13.90	-12.20	0.917	-14.10	1.06
60	2.40	1.86	-3.01	0.530	-6.02	1.06
90	3.39	0.819	0	0	-4.40	1.06
120	4.16	0.634	1.70	-0.530	-3.40	1.06
150	4.63	0.691	2.56	-0.917	-2.96	1.06

Table 11.

 ^{54}Xe ($E_k = 34.56$ keV)

Θ (deg)	α (\AA^{-1})	$d\sigma/d\Omega$ (b/atom/Sr)	(R_{e11}, I_{m11})		(R_{e1}, I_{m1})	
0	0	232.00	-53.90	1.40	-53.90	1.40
5	0.209	143.00	-42.20	1.39	-42.40	1.40
10	0.418	79.70	-31.30	1.37	-31.80	1.40
30	1.24	16.40	-13.20	1.21	-15.30	1.40
60	2.90	2.27	-3.30	0.70	-6.60	1.40
90	3.39	0.943	0	0	-4.65	1.40
120	4.16	0.792	1.86	-0.70	-3.73	1.40
150	4.63	0.892	2.84	-1.21	-3.29	1.40

Table 12.

 ^{64}Gd ($E_k = 50.24$ keV)

Θ (deg)	α (\AA^{-1})	$d\sigma/d\Omega$ (b/atom/Sr)	(R_{e11}, I_{m11})		(R_{e1}, I_{m1})	
0	0	317.00	-62.90	2.57	-62.90	2.57
5	0.209	206.00	-50.70	2.55	-50.90	2.57
10	0.418	114.00	-37.50	2.53	-38.10	2.57
30	1.24	20.50	-14.70	2.22	-17.00	2.57
60	2.40	3.62	-4.06	1.28	-8.12	2.57
90	3.39	1.13	0	0	-4.66	2.57
120	4.16	0.980	1.80	-1.28	-3.61	2.57
150	4.63	1.19	2.81	-2.22	-3.24	2.57

Table 13. ^{66}Dy ($E_k = 53.79$ keV)

θ (deg)	α ($\text{A}^{\circ-1}$)	$d\sigma/d\Omega$ (b/atom/Sr)	$(R_{eA_{11}}, I_{mA_{11}})$	(R_{eA_1}, I_{mA_1})
0	0	332.00	-64.40 2.86	-64.40 2.86
5	0.209	222.00	-52.50 2.84	-52.80 2.86
10	0.418	123.00	-38.90 2.81	-39.50 2.86
30	1.24	21.00	-14.80 2.47	-17.10 2.86
60	2.40	3.70	- 4.06 1.43	- 8.12 2.86
90	3.39	1.10	0 0	- 4.40 2.86
120	4.16	0.936	1.62 -1.43	- 3.25 2.86
150	4.63	1.14	2.48 -2.47	- 2.87 2.86

Table 14. ^{68}Er ($E_k = 57.49$ keV)

θ (deg)	α ($\text{A}^{\circ-1}$)	$d\sigma/d\Omega$ (b/atom/Sr)	$(R_{eA_{11}}, I_{mA_{11}})$	(R_{eA_1}, I_{mA_1})
0	0	344.00	-65.53 3.366	-65.53 3.366
5	0.209	233.00	-53.83 3.352	-54.05 3.366
10	0.418	129.00	-39.86 3.314	-40.48 3.366
30	1.24	20.40	-14.50 2.914	-16.75 3.366
60	2.40	3.53	- 3.86 1.683	- 7.71 3.366
90	3.39	1.01	0 0	- 3.76 3.366
120	4.16	0.874	1.24 -1.683	- 2.48 3.366
150	4.63	1.098	1.81 -2.914	- 2.09 3.366

Table 15.

 ^{70}Yb ($E_k = 61.33$ keV)

θ (deg)	α ($\text{A}^{\circ-1}$)	$d\sigma/d\Omega$ (b/atom/Sr)	$(R_{eA_{11}}, I_{mA_{11}})$		(R_{eA_1}, I_{mA_1})	
0	0	359.10	-67.00	.663	-67.00	.663
5	0.209	246.90	-55.40	.660	-55.70	.663
10	0.418	144.80	-42.20	.652	-42.90	.663
30	1.24	19.98	-14.60	.574	-16.90	.663
60	2.40	3.00	-3.86	.331	-7.72	.663
90	3.39	0.429	0	0	-3.21	.663
120	4.16	0.255	1.08	-.331	-2.16	.663
150	4.63	0.242	1.50	-.574	-1.75	.663

Table 16.

 ^{73}Ta ($E_k = 67.42$ keV)

θ (deg)	α ($\text{A}^{\circ-1}$)	$d\sigma/d\Omega$ (b/atom/Sr)	$(R_{eA_{11}}, I_{mA_{11}})$		(R_{eA_1}, I_{mA_1})	
0	0	402.00	-70.90	.787	-70.90	.787
5	0.209	276.00	-58.70	.784	-58.90	.787
10	0.418	161.00	-44.60	.775	-45.30	.787
30	1.24	25.00	-16.40	.682	-18.90	.787
60	2.40	4.41	-4.68	.394	-9.36	.787
90	3.39	1.03	0	0	-5.03	.787
120	4.16	0.542	1.60	.394	-3.20	.787
150	4.63	0.641	2.53	.682	-2.92	.787

Table 17. Comparison of differential cross sections for Rayleigh scattering of 59.54 keV photons calculate in the RFF model, dispersion corrected RFF + CM model with the KPR cross sections in the S-matrix formalism.

Z	θ (deg)	$\frac{d\sigma}{d\Omega}$ (barns/atom/Sr)		
		RFF	RFF + CM	KPR
68	0	367.15	344.00	342.40
	5	258.92	233.00	231.10
	10	151.92	129.00	129.20
	30	25.67	20.40	19.20
	60	5.14	3.53	3.49
	90	1.66	1.01	0.988
	120	1.21	0.874	0.835
	150	1.44	1.09	0.983
70	0	389.06	359.00	355.30
	5	278.82	246.00	242.70
	10	165.69	144.00	136.40
	30	27.40	19.98	18.30
	60	5.65	3.00	2.88
	90	1.66	0.429	0.516
	120	1.30	0.255	0.266
	150	1.54	0.243	0.237

Z	Θ (deg)	$d\sigma/d\Omega$ (barns/atom/Sr)		
		RFF	RFF + CM	KPR
73	0	423.12	402.00	400.00
	5	300.88	276.00	275.00
	10	183.79	161.00	161.00
	30	30.63	25.00	23.50
	60	6.43	4.41	4.41
	90	2.15	1.03	1.07
	120	1.45	0.542	0.668
	150	1.70	0.641	0.673

59.54 keV are of particular interest. In table 17 we compare the forward angle dispersion corrected RFF cross section results with corresponding uncorrected RFF predictions and the S-matrix predictions of KPR for the elements ^{68}Er , ^{70}Yb , and ^{73}Ta . In the case of other elements included in our calculations, K-shell threshold energies are not very close to the chosen photon energy of 59.54 keV. In Fig 11 we have compared graphically the cross sections calculated in the three models on the element ^{54}Xe at the photon energy of 21.2 keV. An inspection of the comparison table and graph leads us to observe how well the dispersion model can predict Rayleigh scattering in close agreement with

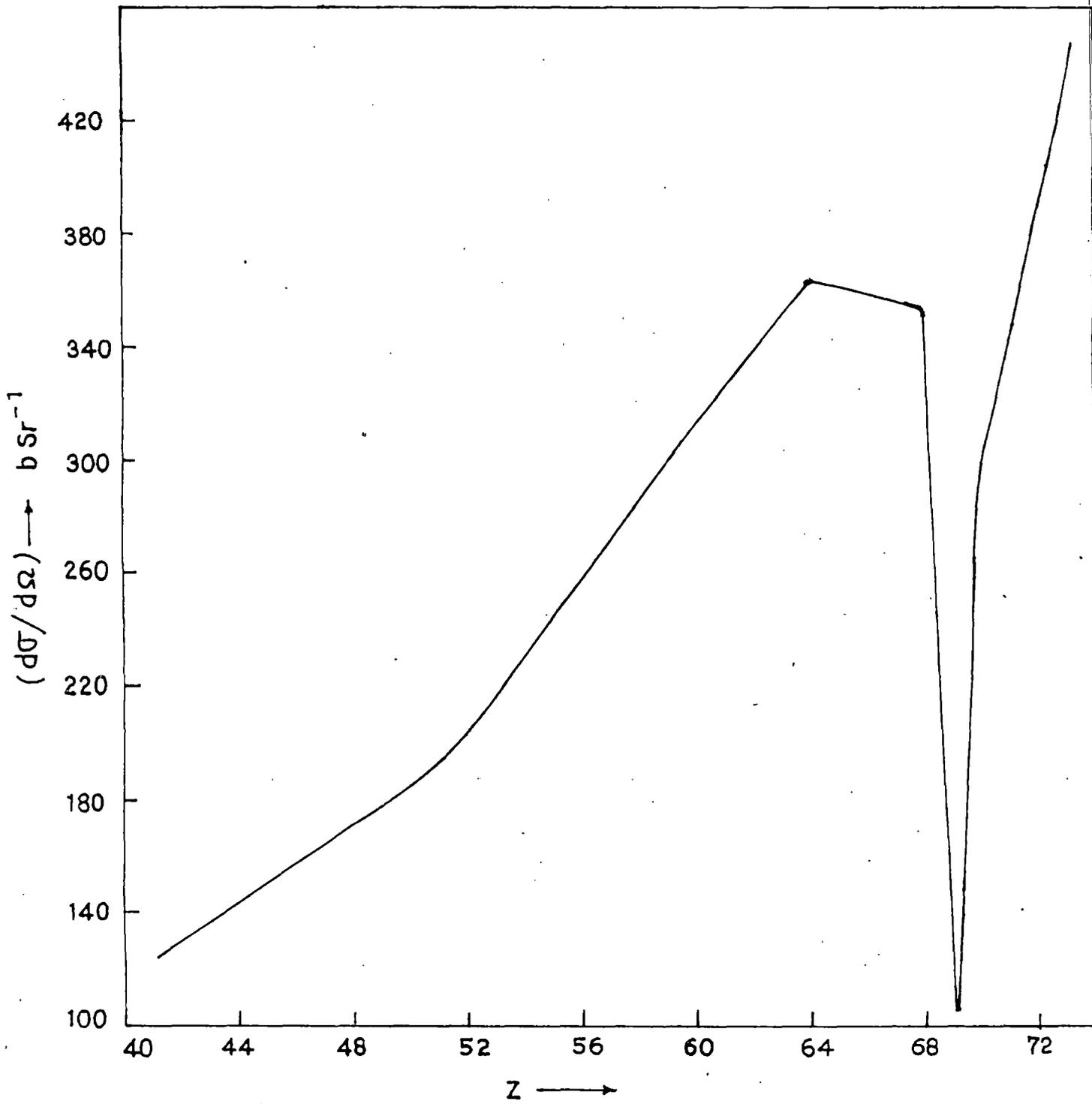


Fig 8.

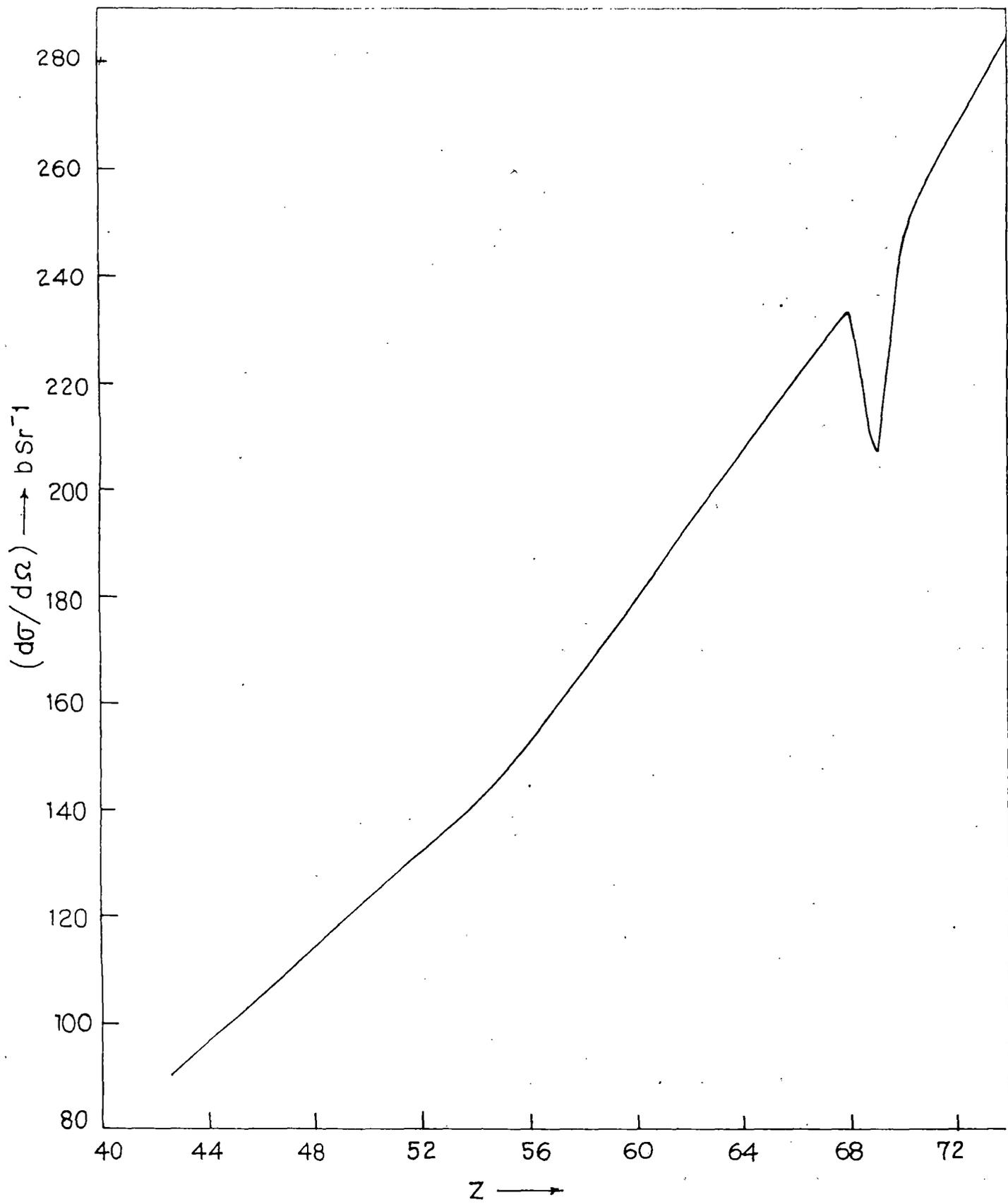


Fig 9.

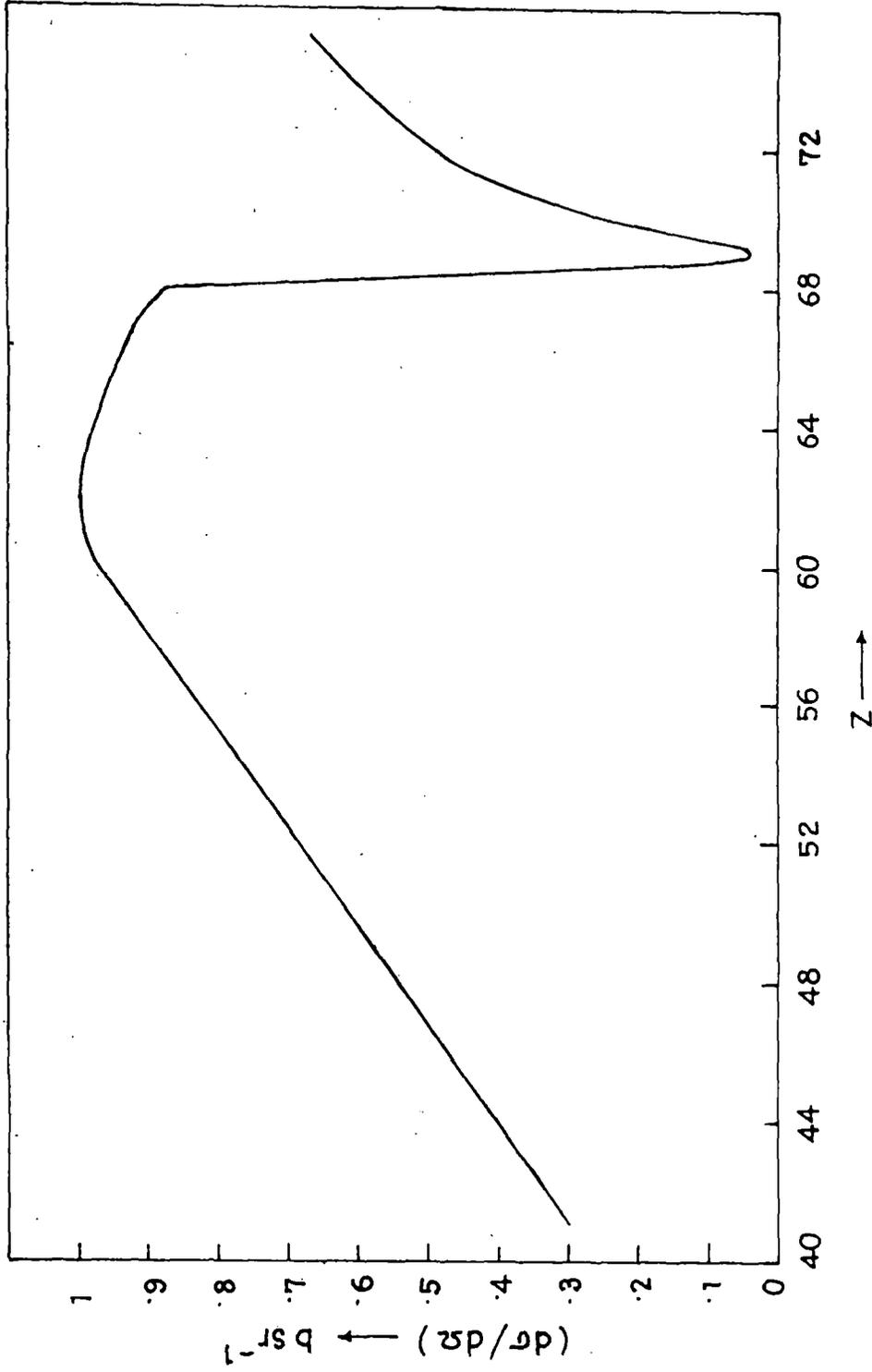


Fig 10.

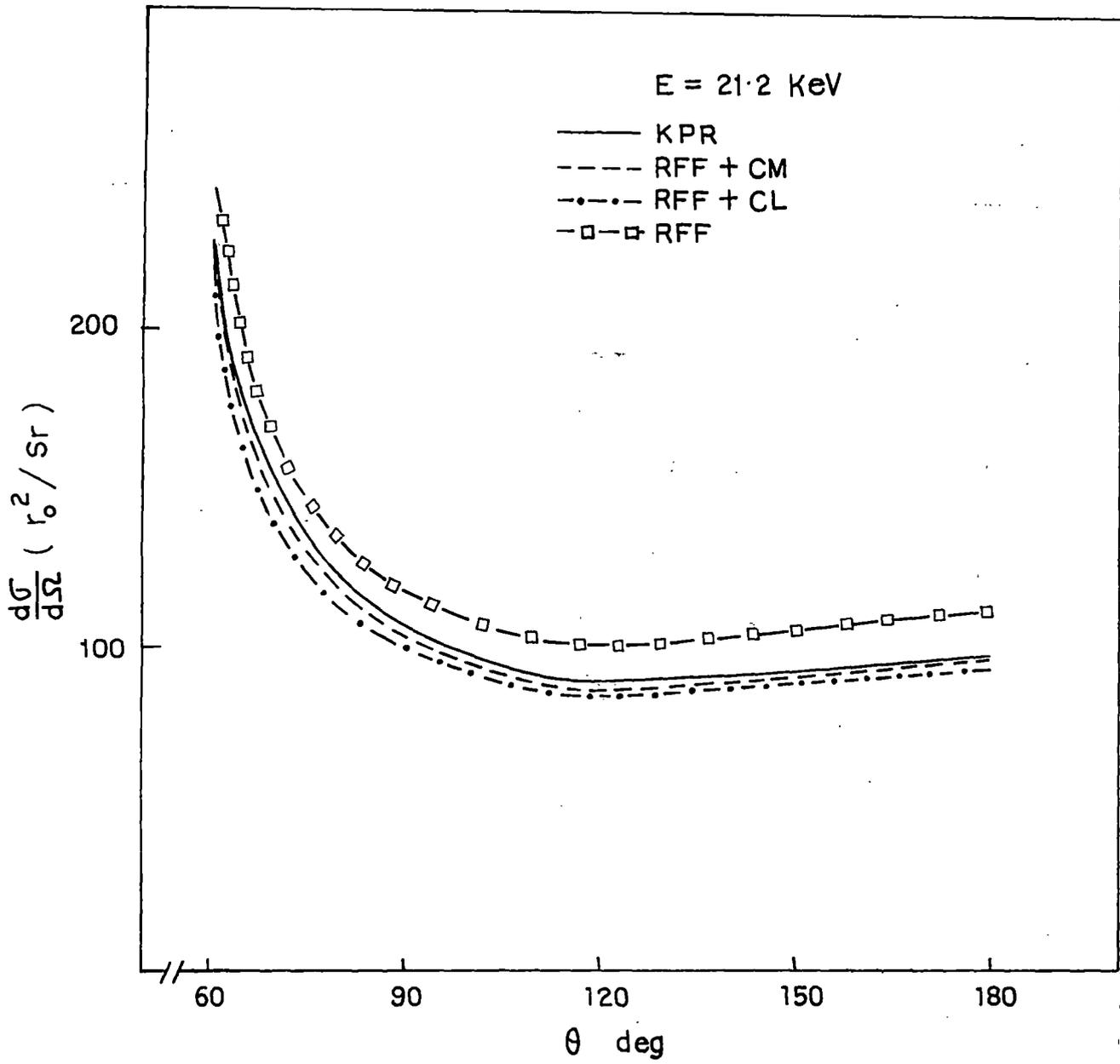


Fig 11.

the numerical partial wave calculations under the condition that the K-shell threshold of the target atom is near the primary photon energy.

The cross section results of our calculations for the elements in the range $Z = 42-73$ at 59.54 keV energy are plotted against Z in Figs. 8-10 corresponding to scattering angles 5° , 60° and 120° . As can be seen from the graphs, the cross section values follow the same general trend at small, intermediate, and large angle of scattering. For elements with K-shell binding energies far from the incident photon energy, the cross section increases more or less linearly with increasing Z . The cross section is observed to fall off rapidly as the elements with K-edge energy near 59.54 keV are approached. Beyond the region of anomalous scattering the cross section increases again with increasing Z . This is in accord with the expectation that pronounced dispersion effect will occur with K-edge close to the incident photon energy.