CHAPTER I

INTRODUCTION

AND

MOTIVATION FOR THE PRESENT WORK
Study of interaction of nuclear radiation with matter has been a subject of theoretical and experimental interest in view of its applications in fields like material science, radiation biology, forensic science, medical physics, X ray fluorescence, agriculture, industry and environmental science. The nuclear radiations are classified into charged and uncharged particles. The charged particles can lose their energy in matter by ionization and radiation processes. The loss of energy due to ionization processes is proportional to mass of the incident particle and inversely proportional to the energy of the particle. Consequently the rate of energy-loss due to ionization process is large for heavy charged particle and small for a high energy particle. Charged particles can also interact with the Coulomb field of a nucleus and lose their energy in the form of bremsstrahlung. The rate of energy loss due to bremsstrahlung process is inversely proportional to mass of the interacting particle and proportional to square of the atomic number of the target. Consequently radiation loss is significant for a low mass particle such as electron and for a target element of high atomic number. Therefore, significant bremsstrahlung production occurs for interaction of high-energy electrons with high atomic number target elements. Such a continuous spectrum of bremsstrahlung radiation is being used in nuclear physics experiments to understand nuclear properties.

Study of interaction of gamma and X ray photons with matter is aimed at the development of useful theoretical and experimental data bank for a variety of applications in several fields ranging from material to biological science. The study basically involves understanding the cause and effect of absorption and scattering of radiation by matter. With ever-increasing use of the radioactive isotopes for various applications it has became a concerning problem to safeguard the operating personnel from health hazards of exposure to harmful radiations. A solution to the problem is possible only through increased knowledge of interaction of radiation with organic and inorganic matter. The gamma and X ray photons can lose their energy in matter by several processes such as Rayleigh scattering, photoelectric effect, Compton scattering, pair production, nuclear Thomson scattering, Delbruck scattering, nuclear resonance
scattering, photodisintegration of the nuclei and meson production. However, only photoelectric effect, Compton scattering, Rayleigh scattering and pair production are significant processes. Among these, the photoelectric effect is a predominant mode of interaction of photons in all medium and high Z targets for photon energies less than 100 keV. In this process the incident gamma photon removes the most tightly bound (K shell) electron and subsequently the electron is emitted with an energy equal to the energy of the absorbed photon minus binding energy of electron. The vacancy so created in the shell will be filled by an electron from higher shells (L, M etc.) by emitting either the characteristic X ray photons or electrons. The emission of characteristic X ray photons is known as X ray fluorescence (XRF) and emission of electrons is known as the Auger process. The XRF technique is being used very recently for producing a true three-dimensional picture of the object using confocal arrangement.

The K shell electrons are the most tightly bound and the most important contributors to the photoelectric cross section in most of the cases. When energy of photon is greater than the K shell binding energy of the target atom, the total photoelectric cross section is the sum of the cross section due to K, L, M, N, ... shells. If energy of the incident photon is less than the K shell binding energy, the K shell photoelectric cross section is zero and the higher shells such as L, M, N, ... cross sections are significant. For medium and high Z elements, a plot of photoelectric cross section versus photon energy gives saw tooth structures, indicating absorption edges at the binding energy of the electrons. Just above the K edge the dramatic resonance structures of the order of 10 to 20% in cross sections have been observed using high-resolution spectrometers. Such structures are known as EXAFS (Extended X ray Absorption Fine Structure) and they are used as a tool to understand the structure of condensed matter.

Rayleigh scattering is the coherent or the elastic scattering process in which the photons are scattered by the bound electrons, and the atom is neither ionized nor excited. The photon loses only a negligible fraction of its
energy because the recoil is experienced by the entire atom unlike in the case of Compton effect where an individual atomic electron experiences the recoil. Most of these gamma photons are emitted in the forward direction, as the recoil experienced by the atom during the collision is very small. The probability of Rayleigh scattering is found to increase with atomic number Z of the target atom and decrease with the energy of the incident photon.

In Compton effect, the incident photon is scattered by electrons of target atom. In this process the incident gamma photon gives a part of its energy to the struck electron and gets scattered at different angle with different energy. The recoil electron receives the energy equal to the energy of incident photon minus the energy of scattered photon. Compton scattering from bound electrons is an inelastic scattering of photons by atoms in which an electron is ejected from the atom. In the case of scattering from free electrons, the outgoing photon energy is determined uniquely by its scattering angle, while scattering from bound electrons is characterized by outgoing photons having a spectral distribution at given scattering angle.

The process of pair production is another predominant mode of photon interaction with matter for incident energies above 1.02 MeV. In this process the incident photon is completely absorbed in the Coulomb field of a nucleus giving rise to production of electron – positron pair whose total energy is just equal to the energy of the photon. The cross section for pair production is proportional to square of atomic number of the target atom.

Therefore, for a given element, the photoelectric effect is predominant at low energy region, the Compton scattering at intermediate energy region and the pair production process at higher energy region.

A beam of gamma or X ray photons passing through matter is progressively attenuated due to above four significant processes namely Rayleigh scattering, photoelectric effect, Compton effect, and pair production.
Each interaction effectively removes a photon from the beam, which leads to an exponential decrease in the intensity of primary photons traversing the matter. For an intensity of photons $I_o$ incident on a thin slab of material of thickness $t$, the transmitted photon intensity $I_t$ is given by,

$$I_t = I_o e^{-\mu t}$$

Here the constant $\mu$ is characteristic of the target material atom and known as linear attenuation coefficient and expressed in cm$^{-1}$. It represents the probability of removal of a photon from the beam per cm path in the absorber-material by all possible modes of interaction. Hence the total linear attenuation coefficient is the sum of the linear attenuation coefficients for the individual interactions such as photoelectric absorption ($\mu_{pe}$), Compton scattering ($\mu_{coh}$), Rayleigh scattering ($\mu_{coh}$), and pair production ($\mu_{pp}$) i.e.

$$\mu = \mu_{pe} + \mu_{coh} + \mu_{coh} + \mu_{pp}$$

Such a linear attenuation coefficient is found to be dependent on the physical density of the absorber. Hence a constant normalized by density, called mass attenuation coefficient ($\mu / \rho$) is defined for materials by equation,

$$I_t = I_o e^{\frac{\mu}{\rho} t}$$

where, $\rho t$ [gm/cm$^2$] is called mass-thickness or areal density and $(\mu / \rho)$ mass-attenuation coefficient. The mass-attenuation coefficient is always to be distinguished from mass energy absorption coefficient, which measures the energy absorbed by the medium. Mass attenuation coefficient $(\mu / \rho)$ [cm$^2$/gm] is related to the total attenuation cross section $\sigma$ [barn/atom] according to the relation,

$$\sigma \left[ \frac{\text{barn}}{\text{atom}} \right] = \frac{\mu}{\rho} \left[ \frac{\text{cm}^2}{\text{g}} \right] \times \mathbb{U}[g] \times \mathbb{A}$$

It is a constant factor given as $\frac{\mu}{\rho}$.
In this equation, $u = 1.6605402 \times 10^{-24}$ g is the atomic mass unit [1 amu = $1/12$ of an atom of the nuclide $^{12}\text{C}$], $A$ is the atomic weight [g/mol] of the target element and 1 barn = $10^{-24}$ cm$^2$ is the unit of area on atomic scale. Obviously the total cross section for photon interaction $\sigma_{\text{tot}}$ is the sum of the cross sections due to individual processes such as photoelectric, Compton, Rayleigh and pair production as given by equation,

$$\sigma_{\text{tot}} = \sigma_{\text{pe}} + \sigma_{\text{coh}} + \sigma_{\text{incoh}} + \sigma_{\text{pp}}$$

Accurate values of total cross sections for various organic and inorganic materials, elements, compounds and mixtures are required in variety of applications. The study of gamma attenuation in biological system provides information about functioning of organs and tissues of human body. The method of radiography is the imaging gamma or X ray absorption-power of the specimen under investigation for diagnostic purposes. The ideal image of photon absorption represents a three dimensional function ($\mu / \rho$)[$x$, $y$, $z$] which describes the distribution of the linear attenuation coefficient in the object. The computer assisted X ray-tomography that produces images of thin slices of human tissues is proved to be ideal for the display of three-dimensional information of X ray absorption through normal and cancerous tissues in the form of films. Similarly the world of molecular imaging (MI) and genomics are the most current topics of research interest where complex structures of DNA, RNA and genes are investigated using irradiation and attenuation methods. In pharmaceutical industry the packing of the drug and uniformity of the powder in compressed tablets & filled capsules is monitored by gamma ray attenuation technique.

Apart from the medical and pharmaceutical fields there are many other areas where the study of attenuation of gamma and X ray photons find applications. A few such interesting areas of applications are as follows. Railway sleepers of wood are inspected by gamma attenuation methods for termite damages. In housing industry the valuation of an already constructed building is
based on strength of concrete and quantity of steel used which is assessed by using gamma attenuation study. In agriculture-fields gamma ray attenuation method is used for studying soil-moisture. In forestry the wood density and moisture content in the forest are measured by gamma ray transmission measurements. Thus the study of attenuation of gamma and X rays in matter is helping the humankind to harness the radiation for prosperity.

The mass attenuation coefficients are measured by using gamma and X-ray photons from radioisotopes. These radioisotopes are prepared by bombarding the elements by high-energy neutrons and charged particles. Majority of these radio-nuclei undergo electron capture or beta decay and subsequently the daughter nuclei are found in the excited state. The excitation energy is emitted in the form of gamma radiation. Electron capture process can also create vacancies in the atomic shells and filling of these vacancies leads to emission characteristic X-ray photons of the daughter atom.

Apart from these, X-ray photons from X-ray tubes and synchrotron radiation from synchrotron sources are used for measuring attenuation coefficients. Synchrotron sources are one of the most successful types of modern high-energy RF accelerators. Synchrotron radiation occurs over a wide range of energy confined to narrow cone of emission. It is interesting to note that the intensity of synchrotron radiation available now decreases rapidly above 50 keV and therefore most of the researchers have concentrated their studies in low and medium atomic number targets. At present more than 50 synchrotron sources are in operation in the world serving many areas of science ranging from physics, chemistry, biology, material science, medicine and industrial applications. But very recently the third generation synchrotrons have come up and these synchrotrons are capable of producing large fluxes of photons even above 100 keV. Using these synchrotrons, it is undoubtedly possible to study attenuation coefficient in high Z elements.

The External bremsstrahlung (EB) can also be used for measuring the attenuation coefficients. The EB photons are produced by passage of electrons
through moderately thick elemental targets. When foils of metals like Ni, Nb, Pd, Sn, Ta and Pb are exposed to $\beta$ particles from a radioactive source, such as $^{32}$P or $^{90}$Sr-$^{90}$Y, a continuous bremsstrahlung spectrum is produced. It is observed that the peak intensity of the EB spectrum increases with increasing beta energy and atomic number $Z$ of the radiator foil. In order to produce significant EB intensity from medium or high $Z$ elements, high-energy $\beta$ sources of long half-life are preferred.

There are two types of geometrical configurations generally employed in attenuation measurements: narrow beam geometry and broad beam geometry. An ideal geometry for a transmission experiment is the one consisting of point source, point absorber and point detector, such that photons that are scattered in the absorber incoherently even at very small angle do not reach the detector. This imposes many restrictions such as arrangement of one or more collimators, large separation between source and the detector, use of strong radio-active sources, huge radiation shielding for source and for the detector, and biological shielding. If lead is used for shielding, the X ray photons emanating from the shielding material may also contribute to the measured intensities; to absorb these lead X ray photons the secondary shielding material is necessary.

When attenuation experiments are performed with narrow beam geometry, the error in the measured values is very small. Such a narrow beam geometrical arrangement is referred to as "good-geometry" set up. Attenuation parameters measured in good geometry arrangement are proved to be useful in nuclear and radiation physics. Even though the narrow beam geometry is usually preferred for accurate determination of attenuation cross section, it does not have many practical applications because in this geometry only a small portion of the target is exposed to incident beam. However, in broad beam geometry, the incident photons that are scattered from the target may also reach the detector. Since a large portion of the target is exposed to the incident beam in broad beam geometry, this geometry has several practical
applications in medicine and industry. In broad beam geometry, inelastic scattering from the target can be minimized by choosing a thin target. Heavy shielding for the radioactive source and detector can be avoided by using a weak radioactive source of strength of the order of a few $\mu$Ci. Hence broad beam geometrical arrangement with a weak radioactive source and a thin target can be an approximate good geometry arrangement and is very useful from the practical point of view.

As we mentioned earlier, the photoelectric absorption process is the important process by which the X-ray and gamma photons interact with matter because in this process energy of photon is completely converted into photoelectron. Since K shell electrons are tightly bound, the photoelectric contribution from K shell electrons is significant provided the energy of the photon is greater than K shell binding energy of the target atom. If energy of photon is less than K shell binding energy, the photoelectric contribution comes from L and higher shell electrons. For a given target, the photoelectric effect is significant in the low energy region (< 100 keV). Several researchers have measured K shell photoelectric cross sections at different energies and for various targets by adopting good geometry and broad beam geometry arrangements. Normally two methods are adopted for measuring the K shell photoelectric cross sections; namely the direct method and indirect method. In the direct method the photoelectric cross sections are measured directly either by detecting the photoelectrons or by measuring the intensity of X-ray photons that follow the photoelectric process. In indirect method the photoelectric cross section is obtained by subtracting theoretical incoherent cross section from the experimental total attenuation cross section. However, our method that is adopted for measuring K shell photoelectric cross section is the direct one in view of the fact that the contribution of the incoherent cross section to the total cross section is negligibly small.

Theoretical values of the atomic photoelectric cross sections for all the elements at various energies have been calculated by several researchers. In
early theoretical developments various simplifying assumptions were made to reduce the complexity of calculations and to obtain simple analytical expressions for photoelectric cross sections. But after 1960, with the advent of fast computers it was possible to avoid almost all simplifying assumptions. Schmickley and Pratt (1967) have done computer based analytical work for relativistic photoelectric cross sections. The sophisticated calculations of Scofield (1973) provide the shell and sub shell photoelectric cross sections in the energy region 1 to 1500 keV for all elements from Z=1 to 101. However there are hardly few experiments to determine the K shell photoelectric parameters for heavy elements close to K edge.

**Motivation for the present work:**

Several Investigators have adopted different experimental techniques for measuring the K shell photoelectric parameters such as,

a) K shell photoelectric cross section at K edge ($\sigma_K$)
b) K shell jump ratio ($r_K$)
c) K shell jump factor ($J_K$)
d) Ratio of total to photoelectric cross section at K edge (Davisson-Kirchner ratio) ($\sigma_\mu/\sigma_K$)
e) K edge energy ($E_K$)
f) K shell oscillator strength ($g_K$)
g) Imaginary form factor ($f^*$)
h) Chemical shift in K edge ($\Delta E_K$)

Most of the researchers have adopted a good geometry arrangement and have measured gamma ray attenuation around K edge energy of the target atom. In this method monoenergetic gamma photons from strong radioactive source are allowed to pass through a target and the transmitted gamma photons are measured with either a scintillation detector or with a high resolution HPGe detector. This method requires many thin foils and many
gamma and X ray sources whose energies should lie around the K edge of the target. But in this method the K edge value is selected theoretically. Some researchers have adopted the reflection geometry; in this geometry, monoenergetic gamma radiation from a strong radioactive source (about 100 mCi) is used to produce the K X ray photons in the target whose K shell photoelectric parameters are to be measured. By knowing the K X ray intensity, the K shell fluorescence yield and fraction of X ray emission, the K shell photoelectric parameters have been determined. Recently synchrotron radiation is being used to study above parameters. In this method the energy of the synchrotron radiation is varied around the K edge of the target and the transmitted X ray photon intensity is measured with a high-resolution detector. The use of synchrotron radiation to study the exact variation of cross-section very close to K edge has provided enough data in the literature for low and medium Z elements. Since synchrotron radiation intensity decreases above 50 keV, very few researchers have carried out experiments in high Z targets close to K edge.

A detailed survey of literature clearly reveals that study of photoelectric cross sections at and around the K edge for high Z target elements is very interesting. The study of K shell photoelectric parameters around K edge of elements would help to explore valuable information of the atomic environment responsible for the processes such as EXAFS and RRS. The huge discrepancies of the order of 10-20% in the measured photoelectric cross sections just above the K edge for medium Z targets indicate one may expect similar trend in high Z target elements. Most of the experiments are carried out using either transmission geometry or the reflection geometry. The explorations of new experimental methods are essential which can yield accurate values of the photoelectric parameters at and around the K edge energy. Hence it is considered worthwhile to undertake a systematic study of K shell photoelectric parameters including the chemical shift by adopting altogether a new method.

In the present thesis we have proposed a novel method for determining K shell photoelectric parameters such as, a) K shell photoelectric cross section at
K edge, b) K shell jump ratio, c) K shell jump factor, d) Ratio of total to photoelectric cross section at K edge, e) K edge energy, f) K shell oscillator strength, g) Imaginary form factor and h) Chemical shift in K edge of heavy elements.

In this method, we have produced external bremsstrahlung (EB) photons of continuous energy in a nickel converter using beta particles from a weak $^{90}$Sr - $^{90}$Y beta source. Since the end point energies of $^{90}$Sr source are 546 keV and $^{90}$Y source is 2281 keV, the EB spectrum is continuous in energy from 0 to 2281 keV. The thickness of EB converter is selected in such way that the intensity of EB spectrum is maximum around 30 keV. The intensity of EB photon at 30 keV is 11,000 counts per 8 hours and that at 160 keV is 2000 counts per 8 hours. These EB photons are allowed to pass through target elements of Gd, Hf, Ta, Au and Pb whose K shell photoelectric parameters are to be determined. The spectrum of the transmitted photons is then recorded with an ORTEC type GMX 10P HPGe detector coupled to an ORTEC type 8K multichannel analyzer, which has MAESTRO software for acquiring the data. In order to stop the unwanted beta particles that are transmitted through the Ni converter, a thick perspex absorber is placed between the nickel converter and the target. The measured transmitted spectrum exhibits a sharp fall in intensity at the K shell binding energy of the target, which we have identified as K absorption edge of the target element.

We have used the incident EB spectrum and the transmitted EB spectrum at and around the K edge and estimated the total cross section at various photon energies. The plot of total cross section as a photon energy shows the cross section is almost constant just above the K edge indicating that the contribution of the inelastic scattering cross section from high energy EB photons is negligibly small and the cross section is also constant just below the K edge indicating that contribution of inelastic scattering cross section from high energy EB photons is negligible. Therefore, the total cross sections above the K edge is essentially due to photoelectric cross sections from K and higher shells and the
total cross sections below the K edge is attributed to the photoelectric cross sections from L and higher shells but not due to K shell. In other words, the contribution from inelastic scattering (Compton scattering) of high energy EB photons from the target in the narrow energy range of our interest in the present experimental arrangement is negligibly small. We have least square fitted a sigmoidal function to the data at the region of sharp fall of the spectrum for determining the above-mentioned K shell photoelectric parameters accurately. We have determined the K shell photoelectric parameters such as a) K shell photoelectric cross section at K edge, b) K shell jump ratio, c) K shell jump factor, d) Ratio of total to photoelectric cross section at K edge, e) K edge energy, f) K shell oscillator strength, g) Imaginary form factor for Gd, Hf, Ta, Au and Pb elemental foils. We have fitted a least square fit sigmoidal function to the sharp fall region of the transmitted spectrum of the elements and compounds. The chemical shift is obtained by taking the derivative of the sigmoidal fitting for Gd, Dy and Pb compounds.

The present thesis contains five chapters. The first chapter deals with introduction of the interaction of gamma and X ray photons with matter and motivation for the present investigations. In Chapter II, we give a brief survey of literature on theoretical and experimental aspect of interaction of gamma and X ray photons with matter with special reference to photoelectric absorption process. Chapter III deals with experimental arrangement adopted for measuring the K shell photoelectric parameters. In the same chapter we describe the various components such as radioactive sources, EB converter, beta stopper, HPGe detector, electronic modules and multichannel analyzer. We also explain in the same chapter standardization of gamma ray spectrometer using monoenergetic gamma sources. In Chapter IV, we present the procedure adopted for measuring the K shell photoelectric parameters. We have also presented how the following K shell photoelectric parameters are determined.

a) K shell photoelectric cross section at K edge,
b) K shell jump ratio,
c) K shell jump factor,
d) Ratio of total to photoelectric cross section at K edge,
e) K edge energy,
f) K shell oscillator strength,
g) Imaginary form factor,
of Gd, Hf, Ta, Au and Pb and also
(h) Chemical shift in K edge of Gd, Dy and Pb.

In the same chapter we present our measured values of K shell photoelectric parameters along with the theoretical values and others experimental values. In Chapter V we have presented consolidated values of K photoelectric parameters for Gd, Hf, Ta, Au and Pb targets and also presented results and discussion on our experimental K shell photoelectric parameters. In the same chapter we have presented our conclusions on the experimental investigations.
References: