Measurement of Energy Loss Straggling of Relativistic Electrons in Thin Aluminum Foils

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(Received November 16, 2015; in final form March 6, 2016)

The mean energy loss straggling per unit path length of relativistic electrons in aluminum has been determined by recording electrons with a Si(Li) detector coupled to 8K multi channel analyzer. The energy loss straggling is determined by measuring the full width at half maximum of the spectrum of the incident and transmitted internal conversion electrons of energies 942 keV and 1016 keV from Bi^{207} source. Measured values have been compared with theoretical values indicating that the present method can be used for measuring mean energy loss straggling.

DOI: 10.12693/APhysPolA.129.1118
PACS/topics: 34.50.Bw

1. Introduction

Study of interaction of charged particle with matter has wide applications in various fields like astrophysics, space science, plasma physics, fusion research, material research, and biomedical research. All investigators have concentrated on stopping power and only few investigators [1–9] have studied the energy loss straggling (ELS) extensively.

ELS mainly deals with distribution of energy losses of incident particles. Accurate information on ELS is important in the application of the Rutherford backscattering spectrometry, nuclear reaction analysis, and scanning transmission ion microscopy. As ELS is one of the main factors limiting depth resolution, understanding the shape of energy loss distribution rather than energy distribution is important in the application of particle identification. ELS reveals the collisional and radiative interaction probabilities of a radiation in a medium and have technological implications such as the spatial dispersion of implantation profiles. The effective atomic number and the morphological changes of complex chemicals like polymers can be investigated from the knowledge of experimental and theoretical values of the ELS of electrons in them. In view of these applications, the study of ELS in matter has become an interesting topic in recent years.

In these directions, several theories and various computer programs [10–15] have been developed to compute ELS for different types of radiation at different energies. However, the validity of these theoretical predictions rests on the experimental verification. But survey of literature shows the ELS has been investigated in a lesser extent because of experimental difficulties. The determination of the ELS sets more severe requirements to the spectrometric methods and the target preparation. Even a small degree of inhomogeneity and roughness in the target introduces additional ELS. Due to these facts, only a few experimental data [16–21] are available for the light and heavy charged projectiles. Particularly, the experimental data available for electrons of intermediate energies are very few.

Keeping above points in view, we have measured the ELS of 942 and 1016 keV internal conversion electrons of Bi^{207} in the aluminum absorber and compared the results with the values predicted by different theoretical models.

2. Theory

When monoenergetic electrons pass through a foil, they emerge with a distribution of energies due to statistical nature of the collisional loss and this process is called energy loss straggling. It is measured by the standard deviation σ, the root-mean square deviation of the energy distribution about its mean value. Depending upon the absorber thickness, straggling theories are broadly divided into two categories as below.

2.1. Straggling theories for thick absorbers

For thick absorbers wherein the number of collisions is larger, central limit theorem predicts the energy loss distribution to be Gaussian. For high energies, where the projectile velocity is larger than the orbital velocities of the target atoms, Bohr [1] predicted that ELS is proportional to the square root of the number of target electrons per unit area traversed by the projectiles. Lindhard and Scharff [3] extended this theory to intermediate energies by correcting the Bohr formula with the dielectric function. Livingston and Bethe [22] improved the Bohr formula by incorporating the atomic number of the absorber, Z, and the distant collisions. They replaced Z by a reduced atomic number, Z', and incorporated a term which depends on the atomic structure of the absorber.

Bonderup and Hvelplund [4] refined the Lindhard and Scharff formalism by improving the target description by means of the Lenz–Jensen atomic model. With the Hartree–Fork–Slater model of electron density, Chu [5]

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calculated the straggling of helium ion in each element, which can be converted to that for proton energy straggling.

All these above analysis are based on the assumption that the energy loss is less than the change in velocity of the projectile in the absorber.

2.2. Straggling theories of thin absorbers

In case of thin absorbers as the number of collisions is too small, there are chances for large energy loss in a single collision and hence central limit theorem cannot be applied. For heavy particles, the large energy loss in single collision is kinematically limited, while for electrons, as much as half of the initial energy can be transferred apart from one shot energy loss by Bremstrahlung. All these rare events add a long tail to the energy loss probability distribution, making it skewed and asymmetric Gaussian called Landau curve, which can be fitted to an exponentially modified Gaussian (EMG). Here the mean energy loss no longer corresponds to the peak but it is displaced because of the high energy tail. The position of the peak defines the most probable energy loss.

The probability distribution $f(x, \Delta)$ that an electron penetrating the thickness, x of an absorber will have lost an energy Δ, which is small compared to its initial kinetic energy, is given by the Boltzmann transport equation as

$$\frac{\partial f(x,\Delta)}{\partial x} = \int_0^\infty w(E)\left[f(x,\Delta-E) - f(x,\Delta)\right] \mathrm{d}E, \quad (1)$$

where $w(E)$ is the differential collision cross-section (DCCS) for single collisions with an energy loss, E. This equation implicitly assumes that each collision suffered by the incident particle is a stochastic process. For example, channeling is not accounted for in this transport equation.

All the theoretical descriptions of the straggling process are based on this transport equation. The methods of obtaining $f(x, \Delta)$ can be divided into 3 groups as

1. Laplace transform method used in the original work of Landau [23] and Symon [24] and the modification by Blunck and Leisegang [25], Vavilov [26] and Shulek et al. [27].

2. Convolution method due to Williams [28] as applied by Tung [29] to calculate energy spectra of electrons transmitted through thin aluminum foils.

3. Method of moments employed by Tschalar [30].

The Laplace transform methods differ from each other chiefly in the form assumed for the DCCS and in the corrections employed if any, to account for the difference between the assumed DCCS and the realistic spectrum. Each of these methods have different region of applicability depending on the significance ratio, $k = (\xi/E_{\max})$ where $E_{\max}$ is the maximum transferable energy to atomic electrons and ξ is the mean energy loss. ξ can be obtained from the Bethe–Bloch formula by ignoring the logarithmic term as below

$$\xi = \frac{2\pi z^2 e^4}{mc^2\beta^2}\frac{N_A Z x}{A} = \frac{0.15354 Z z^2 x}{A\beta^2} \ \left[\mathrm{MeV\,cm^2}\right], \quad (2)$$

where z is the charge of the incident particle. x, Z, and A are the thickness, atomic number, and atomic weight of the target. Thin absorber region is generally taken to be $k < 10$, although the distribution begins to approach Gaussian for $k > 1$.

The Landau theory [23] is applicable for $k \leq 0.01$ with the following assumptions:

1. Large energy loss in a single collision, $E_{\max}$ is infinity, i.e., $k \to 0$.

2. Individual energy transfers are so large that the electrons are treated as free.

3. The decrease in velocity of the projectile is assumed to be negligible and ignored.

As per the above mentioned assumption (2), Landau used Rutherford cross-section $w(E) = \xi/(xE^2)$ for free electron as DCCS and solved the transport equation to get

$$f_L(x,\Delta) = \Phi(\lambda)/\xi, \quad (3)$$

where $\Phi(\lambda)$ is a universal function depending only on the variable λ as below

$$\Phi(\lambda) = \frac{1}{2\pi\,\mathrm{i}}\int_{C_k-\mathrm{i}\infty}^{C_k+\mathrm{i}\infty} \exp\left(u\ln u + \lambda u\right) \mathrm{d}u, \quad (4)$$

where $u = \xi p$ and $\lambda = \frac{\Delta-\bar{\Delta}}{\xi} - \beta^2 - \ln(k) - 1 + C_E$, C_k being an arbitrary real constant, C_E is the Euler constant $= 0.577215$, p is the momentum and $\bar{\Delta}$ — the mean energy loss of the incident particle. This universal function $\Phi(\lambda)$ must be evaluated numerically and a tabulation of $\Phi(\lambda)$ for various λ was done by Borsch-Supan [31], Seltzer and Berger [32]. The Landau distribution is asymmetric with a long tail extending to $E_{\max}$ with a maximum for $\lambda = 0.229$ and width of $W_L = 4.018\xi$ [33].

Rohrlich and Carlson [34] followed the Landau analysis but replaced the Rutherford cross-section by improved approximations to the Bhabha cross-sections [35] for positrons and the Moller cross-sections [36] for electrons to get the fractional energy transfers. The spin and exchange terms of the Bhabha and Moller cross-sections produce small corrections in the Landau distribution of energy losses by electrons and positrons in thin foils, the most probable energy loss being less affected than the shape of the distribution. The FWHM of the shape corrected Landau curve is

$$W_R = \xi(3.98 - 6.6\alpha), \quad (5)$$

where $\alpha = \frac{(2\gamma-1)}{\gamma^2}\frac{k}{2}$ is the curve shape correction factor and γ — the incident energy in the units of mc^2.

Sigmund and Winterbon [7] analyzed the transport equation theoretically using a steepest-descent evaluation of the Bethe–Landau integral to get a general expression for the energy loss spectrum, the most probable energy loss and half width, without reference to any particular DCCS.

The region between k covered by Landau and Gaussian limit is treated by Symon [24] and Vavilov [26]. Using the limiting distribution derived by Landau, Symon made a number of ingenious approximations to derive energy loss distribution in graphical form [24].

Vavilov [26] used the following form of DCCS to account for the spin of the incident particle and solved the transport equation to get a more generalized solution

$$w(E) = \frac{\xi}{x}\frac{1}{E^2}\left[1 - \frac{E\beta^2}{E_{\max}}\right]. \tag{6}$$

His result reduces to the Landau distribution in the limit $k \to 0$ and the Gaussian in the limit $k \to \infty$.

All the above theories are based on the assumption that scattering occurs on quasi-free electrons and follows the Rutherford collision probability. Thus they ignored the electron binding energies and hence the small energy transfers due to distant collisions. This resonant effect (resonance excitation of bound electrons of the target by the incident electron in distant collisions) is more pronounced especially when ξ is greater than the average ionization energy, I_{av} of the absorber as a thumb rule. Blunck and Leisegang [25], Shulek et al. [27] have modified the Landau–Vavilov energy straggling function to include the resonance correction. In these approaches, the Rutherford DCCS is replaced by a realistic DCCS so that, for close collisions, it tends to the Rutherford DCCS but for distant collisions, it takes into account the atomic shell structure.

Shulek et al. [27] convoluted the Landau distribution with a normal distribution of standard deviation, $\sigma_I = \sqrt{\text{variance}} = \sqrt{x\delta_2}$, which can be computed with relative ease. These results in a broader distribution with the peak value increased by a small amount compared to the Landau theory. They proposed the following form of δ_2 to estimate the resonant effect:

$$\delta_2 = \frac{8}{3}\frac{\xi}{x}\sum_i I f_i \left(\ln\frac{mc^2\beta^2}{I_i}\right), \tag{7}$$

where I_i — the effective ionization potential of the i-th shell and f_i — the fraction of electrons in that shell. The FWHM of the resulting improved energy-loss distribution, FWHM_I is related to the FWHM of the energy straggling distribution, FWHM_L and the FWHM of the Gaussian convolving distribution, FWHM_G as

$$\text{FWHM}_I^2 = \sqrt{\text{FWHM}_L^2 + \text{FWHM}_G^2}, \tag{8}$$

where

$$\text{FWHM}_L = 4.02\xi, \tag{9}$$

$$\text{FWHM}_G = 2.36\sigma_I = 2.36\sqrt{\delta_2}. \tag{10}$$

Blunck and Leisegang [25] have approximated the Landau universal function by a sum of Gaussians folded within a Gaussian function (a broadening Gaussian) in terms of a dimensionless quantity b, known as the broadening parameter. Findlay and Dusautoy [37] has improved it by folding the effect of resonant energy transfer into a sum of nine Gaussians as a good fit to the Landau distribution to get the FWHM as

$$\sigma = W\ [\text{MeV}] = 4.022 + 0.3600H + 0.4320H^2 - 0.04088H^3 + 0.01677H^4, \tag{11}$$

where

$$H = \ln\left(b^2 + 1\right) \tag{12}$$

and

$$b^2 = 2 \times 10^{-5} Z^{4/3}\frac{\bar{\Delta}}{\xi^2}, \tag{13}$$

where $\bar{\Delta}$ is the average energy loss in the absorber of atomic number Z.

3. Experimental details

The experimental arrangement to measure the ELS of relativistic electrons from Bi^{207} internal conversion (IC) source has been published in our earlier paper [38]. However for shake of continuity, we briefly describe the same here. The Bi^{207} radioactive source used in this work is electroplated on a platinum foil and encapsulated in stainless steel of 1.52 cm outer diameter with 18.8 mg/cm^2 thick beryllium window to protect the source from spilling and prevent the contamination. Bi^{207} IC source emits 481.69, 555.39, 975.69, and 1049.39 keV IC electrons. After correcting for the attenuation in beryllium window of the source and the air column between the source and detector, the effective energies of these emitted electrons become 443.98, 518.84, 941.74, and 1015.56 keV, respectively.

The energy of IC electrons are measured using selection-grade NE Si(Li) detector of 0.2 cm depletion area and 15 cm^2 active area. Output of the detector is connected to an ORTEC made charge sensitive preamplifier of charge sensitivity 15 mV/MeV (Si equivalent). This preamplifier output is fed to 8K Multi Channel Analyzer through an ORTEC made delay line amplifier. As the detector is sensitive for light, the whole assembly is placed in a light tight box.

The absorbers are pure aluminum foils obtained from Sigma-Aldrich Inc. Their thicknesses are determined using a travelling microscope and a sensitive balance. Uniformity of the foil thickness is confirmed by checking the FWHM of the peak transmitted through different regions of the foils.

4. Procedure

After confirming the long term stability of the instrument, the spectrum of incident IC electrons of Bi^{207} containing 4 major peaks is acquired as in Fig. 1. All these four peaks corresponding to 443.98, 518.84, 941.74 and 1015.56 keV are fitted to four EMG to get the channel numbers corresponding to their most probable energies and hence the calibration graph of the Si(Li) detector spectrometer as is given in the inset of Fig. 1.

From the spectrum transmitted by the aluminum foils of various thicknesses, the FWHM of the IC electrons and hence their ELS in the aluminum are determined. Figures 2 and 3 present the EMG fitted spectra of the

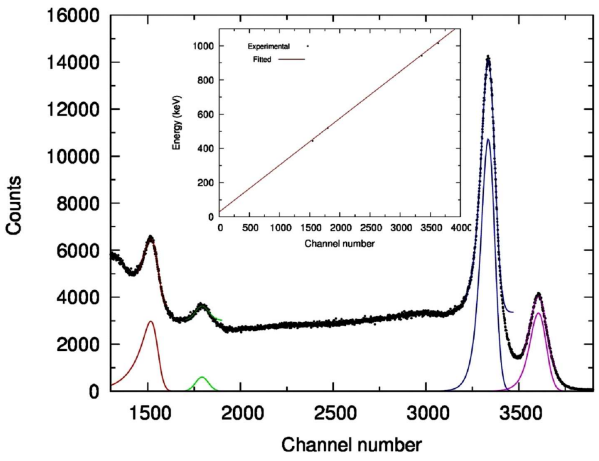

Fig. 1. EMG fitted incident Bi^{207} spectrum with calibration graph.

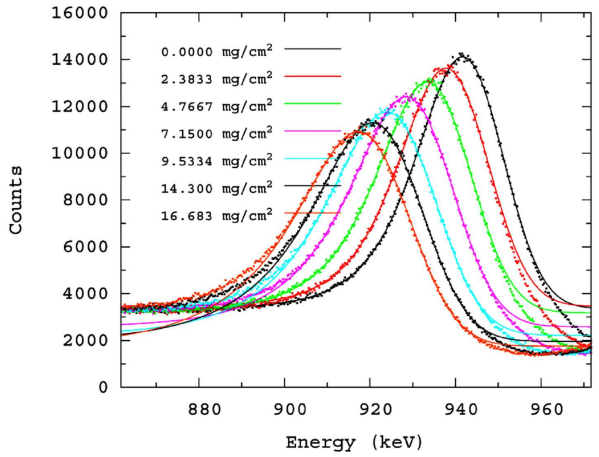

Fig. 2. Transmitted spectra of 942 keV IC electrons for Al foils of various thicknesses.

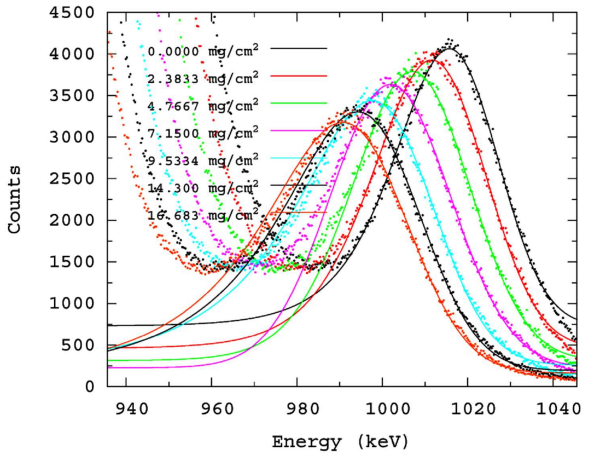

Fig. 3. Transmitted spectra of 1016 keV IC electrons for Al foils of various thicknesses.

IC electrons of energies 942 and 1016 keV transmitted through different thicknesses of the aluminum foil.

TABLE I

Experimental and theoretical values of ELS of 942 keV IC electrons in Al.

foil th.	measured	Landau	Rohrlich	Shulek	Findlay
[mg/cm^2]	[keV]	[keV]	[keV]	[keV]	[keV]
2.383	1.6786	0.3435	0.3401	1.1985	1.2070
4.767	2.1514	0.6869	0.6801	1.7632	1.8105
7.150	2.3410	1.0304	1.0200	2.2400	2.3228
9.533	2.8199	1.3738	1.3597	2.6761	2.7905
14.300	3.9877	2.0607	2.0387	3.4868	3.6532
16.683	4.3289	2.4042	2.3780	3.8742	4.0618
ELS/path length in [$\frac{\text{MeV cm}^2}{\text{g m}}$] with fitting error					
ELS/PL	0.1903	0.1441	0.1425	0.1847	0.1972
fit. error	0.0113	0.0000	0.0000	0.0054	0.0059

TABLE II

Experimental and theoretical values of ELS of 1016 keV IC electrons in Al.

foil th.	measured	Landau	Rohrlich	Shulek	Findlay
[mg/cm^2]	[keV]	[keV]	[keV]	[keV]	[keV]
2.383	1.6792	0.3389	0.3357	1.1900	1.2028
4.767	1.8562	0.6779	0.6712	1.7498	1.8028
7.150	2.0497	1.0168	1.0066	2.2220	2.3116
9.533	3.2568	1.3557	1.3419	2.6538	2.7758
14.300	3.8447	2.0336	2.0122	3.4558	3.6312
16.683	4.4871	2.3725	2.3471	3.8389	4.0361
ELS/path length in [$\frac{\text{MeV cm}^2}{\text{g m}}$] with fitting error					
ELS/PL	0.1953	0.1422	0.1407	0.1828	0.1957
fit. error	0.0229	0.0000	0.0000	0.0054	0.0059

Thus measured ELS values for the 942 keV IC electrons are as given in Table I and Fig. 4 as a function of absorber thickness. Since the variation is linear, the slopes of these lines are taken as the average ELS per unit path length in aluminum. For the sake of comparison we have also shown the energy loss values evaluated using theories of Landau [23], Rohrlich [34], Shulek et al. [27] and Findlay and Dusautoy [37]. Table II and Fig. 5 gives the measured and theoretical ELS for the IC electrons of energy 1016 keV in aluminum as a function of thickness.

5. Results and discussion

We opted the Bi^{207} IC source, as it emits wide ranges of electrons, all of which can be used simultaneously for the MCA calibration under the same environmental conditions. The spectral acquisition time is taken high enough to get counts of more than 10,000 under each peak and hence the counting error below 1%. The choice of aluminum as the target follows the fact that the dielectric theory used for its well established plasmon losses can be readily used for characteristic energy losses in organic

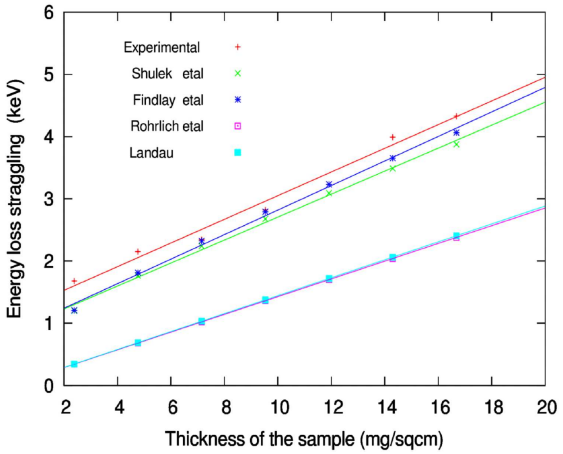

Fig. 4. ELS of 942 keV IC electrons as a function of Al foil thickness.

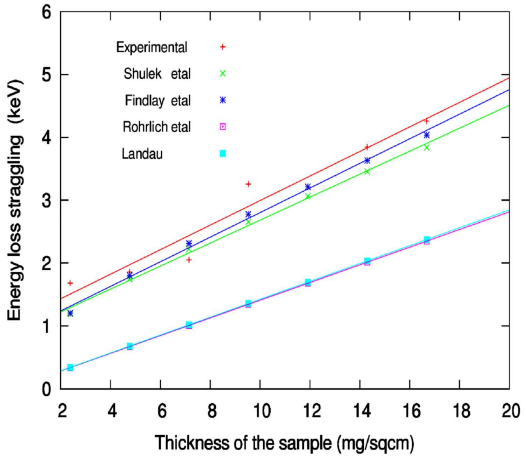

Fig. 5. ELS of 1016 keV IC electrons as a function of Al foil thickness.

and biological solids [39–42]. As the roughness of the foil increases the FWHM and the ELS, we used laboratory grade aluminum foils obtained from Sigma-Aldrich Inc. The foils are selected by checking the uniformity in their thickness over the beam area. Additional care is taken by fitting the spectrum to EMG using the Levenberg–Marquardt algorithm [43], a nonlinear least-squares fitting in GNUPLOT.

The measured ELS values as a function of thickness are compared with the straggling theories of Landau [23], Shulek et al. [27], Rohrlich [34] and Findlay and Dusautoy [37] as in Tables I and II. From these tables we notice that though all these theories predict the same ELS per path length, the straggling magnitudes predicted by them are all different. The Landau formula derived using the Rutherford cross-section and the Rohrlich formula derived using the Moller cross-section agrees well with each other but underestimates the magnitude of the straggling due to lack of resonant corrections. Though the resonant corrected formula by Shulek et al. [27] and Findlay and Dusautoy [37] leads to almost same result, the Findlay formula is closer to the measured values. Further the deviation between the Rohrlich [34] and Landau [23] predictions, as well as Shulek et al. [27] and Findlay and Dusautoy [37] predictions increases with the absorber thickness smoothly within their region of validity itself.

We have also plotted the energy loss straggling of 942 and 1016 keV IC electrons in aluminum foil as a function of the absorber thickness as in Figs. 4 and 5. As the variation is linear, the slopes of these plots are taken as the ELS per unit path length of the electrons in aluminum at that energy. The ELS evaluated in this manner are presented along with the least square fitting error in the last two rows of Tables I and II. This shows good agreement between our measurements and theoretical values predicted by Shulek et al. [27] and Findlay and Dusautoy [37]. The mean percentage deviation of measured values from that predicted by Findlay and Dusautoy [37] is +3% at 942 keV and +0.3% at 1016 keV. This may be due to the unavoidable roughness present in the foils.

6. Conclusion

We have measured the ELS of 942 and 1016 keV IC electrons in aluminum absorber of thickness varying from 2 mg/cm^2 to 17 mg/cm^2. Our results are compared with that predicted by theories of Landau [23], Shulek et al. [27], Rohrlich [34] and Findlay and Dusautoy [37]. The measured ELS values of the 942 and 1016 keV electrons agree closely with the results predicted by Findlay and Dusautoy [37].

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