Chapter 2

ELECTRONIC STOPPING POWER (Random Case)

2.1 INTRODUCTION

Even after many decades of its experimental observation and theoretical interpretation, the study of energy loss of charged particle propagating through solids is still attracting attention of researchers and is used extensively for probing and analysing various phenomenon occuring in the field of atomic, nuclear physics and condensed matter physics [1,2]. Charged particle loses its energy through two processes. (i). electronic energy loss, where the particle loses energy through inelastic collision (excitation,ionization) with target electrons. (ii). nuclear energy loss, where the energy of the particle is transferred to the target nucleus resulting in atomic vibrations and possible displacement / dislodging of these atoms / ions. Nuclear stopping is predominant only at very low velocities (~ 10 keV/amu) and at higher velocities it can be neglected. Any correlation between nuclear and electronic stopping can be completely ignored except for single scattering studies [3] and for very thin targets. The total stopping cross sections of ions in the solids is the sum of contribution from the electronic and nuclear stopping cross sections. The velocity dependence of stopping power, as an ion passes through solid is elucidated below.

When an energetic ion moves in a solid (target) with a velocity higher than the velocity of innermost orbital of the solid, most of its electrons are stripped off and essentially a bare nucleus moves through the solid (Region I in Fig. 2.1.) [4]. As this energetic ion
loses energy to the target through various processes such as excitation and ionization of the target, its velocity decreases. When the velocity approaches a velocity equal to that of electrons in the innermost orbit, the probability of capturing an electron in that particular orbit increases, whereas the probability of losing an electron from that shell decreases, so that electrons are captured in that inner shell. When the ion velocity further decreases and becomes comparable to the velocities of successive orbital electrons, then the electrons are captured successively in those shells (Region II in Fig. 2.1.), and consequently at velocities of the order of critical velocity \( v_c = v_o Z''_1 \), a neutral atom moves in solid (where \( v_o \) is the Bohr velocity and \( Z'' \) is the atomic number of projectile ion). The region of velocities below these velocities is called low velocity region (Region III in Fig. 2.1.), where the rate of electronic energy loss decreases with decreasing velocity.

At low energies, nuclear stopping becomes significant. The main effect is dislodgement of atoms from their bonding states and creation of radiation damage in the target material [4]. When the energy of the primary displaced atom is large enough to dislodge more atoms, a damage cascade is induced giving rise to secondary and other higher order damage cascades. For still lower energies of incident ions the chemical binding among the atoms of the target material is more important and energy is given to the target crystal as a whole; in this case the ion is said to be stopped in the target.

Only a few energy loss data for heavy ions are available in the low velocity region [5]. A knowledge of accurate heavy ion stopping powers in various elements and compounds are essential from both fundamental and practical points of view. Heavy ion beams are routinely used in Ion Beam Analysis techniques for depth profiling and elemental analysis of materials [6]. In addition to their utility in Ion Beam Analysis techniques, energy loss values are essential input parameters for most nuclear physics experiments, such as life time measurements by the Doppler shift attenuation technique, analysis of data recorded by particle track detectors, etc. Therefore, applications of the basic energy...
loss or stopping power data are significant.

We have performed a series of energy loss measurements of various target-ion combinations in the energy range of 1.0 to 5.0 atomic units of velocity at Nuclear Science Centre (N.S.C.), New Delhi, India [7,8]. We have been systematically carrying out the stopping power experiments in the low velocity region using heavy ions \((Z_1 = 8 - 29)\) with a broader perspective to supplement the insufficient stopping power data available in this region. It was theoretically predicted that the \(Z_1\) oscillations in the channeling electronic stopping power [4] will damp out, when one increases the projectile velocity above \(2-3v_0\) [9]. So far this prediction eludes experimental verification due to inadequate stopping power values in this region as mentioned earlier. We hope we can verify this theory in near future specially when channeling experiments are possible at N.S.C. New Delhi. As of now, in the absence of Goniometer at this Palletron, we have been able to do only random stopping power measurements. To ensure optimum usage of the beam time, we have developed an ingenious method for doing energy loss measurement using Elastic Recoil Detection Analysis (ERDA) experiment. As a new approach, recoil ions produced in heavy ion scattering (ERDA) were utilised for energy loss measurements. Secondary recoils of the required \(Z_1\) value generated by the bombardment of high energy heavy ion on a particular target is utilized here. So the various secondary ion beam can be produced by merely changing the targets. The energy loss of the recoil ions was measured by keeping two surface barrier detectors, with and without target foil, at the same recoil angle, with their gain matched with pulsers before the experiment. Different energies of the secondary beam could be selected by changing the detection angle. A similar technique has also been used for stopping power measurements by another group [10-12]. The details of the experiment are outlined in the section 2.2, the theoretical models used to compare the experimental results are discussed in section 2.3 and results and future programme of ongoing experiment is discussed in the section 2.4
2.2 EXPERIMENTAL DETAILS

The electronic energy loss measurements were carried out at the 15 MV Palletron facility at the Nuclear Science Centre, New Delhi, India [7]. A Schematic diagram for experimental setup is shown in Fig. 2.2. A well collimated primary beam of $^{127}I$ and $^{192}Au$ ions (80 -130 MeV) were used to generate secondary recoil ions of various species of targets. Targets with different Z's were mounted on the target ladder of a scattering chamber facility of the Palletron. The recoiled ions were then detected by surface barrier detectors before and after their passage through the mylar (carbon) foils. Rectangular Ta slits of 4mm X 8mm area defined the solid angle for the detection geometry. As the incident energy is below the Coulomb barrier, only Rutherford scattering is possible.

The experiment was carried out in a scattering chamber of 1.5m diameter [13]. The rotatable detector arm mounted inside could be moved from outside the vacuum. The angle of detection could be changed with a precision of 0.05°. The vacuum in the scattering chamber was $1 \times 10^{-6}$ Torr. The target ladder could also be rotated and there was a provision for in vacuo transfer of the target. This facility therefore gave the advantage that the energy of the recoiled ion beams could be varied by changing the forward angle of detection. Since the elastic primary beam will not be scattered beyond $\theta_{max} = \sin^{-1}(M_t/M_p)$, the detection angles larger than $\theta_{max}$ could be chosen for different recoil ion beams, thereby avoiding large count rates from the elastic events. The targets were tilted with respect to the primary ion beam direction so that the recoils were obtained in the reflection geometry. The stopping cross sections were determined by measuring the energy of the recoil ions with and without the mylar (carbon) foils using surface barrier detectors with depletion depths of 60μm. The thickness of the mylar (carbon) film was measured by alpha - energy loss method. The surface barrier detectors were energy calibrated using an $^{241}$Am alpha source. Care was taken to monitor the energy calibrations
of the detectors at regular intervals by using the standard pulsers duly adjusted before the start of the experiments.

Two detectors, one with the mylar (Carbon) foil and another without the foil, were mounted in slots provided in the scattering chamber angular arms. These slots were 6° apart from each other. The detectors were swung into the path of the recoil beams one after the other and the energy spectrum of the secondary recoils were recorded. The difference in the measured energies of the particles in the two detectors gave us the energy loss in the foil.

The incident primary beam will produce secondary recoil beams of a range of energies, depending on the depth from which the recoils are emerging. For the recoils generated from the surface, the energy is given by

\[ E_{\text{rec}} = \frac{4M_p M_t}{(M_p + M_t)^2} E_0 \cos^2 \phi_R \]  

(2.1)

where \( M_p \) and \( M_t \) are the masses of the primary ion beam and target atom (in atomic-mass units) respectively, \( \phi_R \) is the recoil angle in degrees, which was varied to carry out the measurements at different energies.

The experimental values deduced have errors of about 3.5% which includes errors arising from the foil thickness measurements and errors in the calculations of recoil energies from spectra. To minimize the energy uncertainty resulting from kinematic broadening, the detector solid angles were taken to be about 0.2 msr. The surface barrier detectors generally had pulse height defects [14] for detection of heavy ions. This defect would lead to an incorrect energy determination of the particles. Since the pulse height defect increases with the mass of the ion and its energy, it was estimated [14] for Ti ions of energy 27 MeV and was found to be 0.05 MeV. Therefore, the error resulting from this effect is 0.05%.
2.3 THEORETICAL CALCULATIONS

An exhaustive collection of stopping power data and various theoretical models were reported elsewhere [15-18]. Most of our ion velocities belong to low velocity region and some are just above this region. LSS theory [19] which is based on the assumption that the electron density in the target varies slowly with position, can be used to get a theoretical estimation of electronic stopping power in this low velocity region. Varelas - Biersack approximation [20] holds good in intermediate energy region, and Bethe formula [18,21] can be used to calculate electronic stopping power in high velocity region. It is to be noted that, we have used Bethe formula only to apply Varelas - Biersack approximation to electronic stopping power in the intermediate velocity region.

At low velocities \((v < v_o Z_j = v_e)\), the stopping power can be written as \(N.S_e\), where \(N\) is the number of atoms per unit volume and \(S_e\) is the electronic stopping cross section per atom. According to LSS theory [19], which is based on Thomas Fermi model, the electronic stopping cross section can be written as

\[
S_{E,L} = \xi \frac{8 \pi e^2 a_o Z_1 Z_2 v}{Z \, v_n}
\]

(2.2)

Where \(e\) is the electronic charge and \(Z_1\) and \(Z_2\) are the atomic numbers of projectile ion and target atom respectively, \(a_o\) is the Bohr radius, the constant \(\xi\) is of the order of \(Z_1^{1/6}\) and \(Z_2^{2/3} = Z_1^{2/3} + Z_2^{2/3}\).

The electronic stopping power \((dE/dx)_e\) is given by [19]

\[
(dE/dx)_e = N.S_{E,L}
\]

(2.3)

where \(N\) is the atomic density. The electronic stopping power can be evaluated in \(MeV/mg/cm^2\) units and is given as
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\[
\left( \frac{dE}{dx} \right)_e = 11.532790 \cdot \xi \frac{Z_1 Z_2}{A_2 Z} \frac{v}{v_o} \tag{2.4}
\]

where \( A_2 \) is the mass number of target atom. Another related quantity of interest is Reduced stopping power \((dE/dx)_R\) defined as

\[
\left( \frac{dE}{dx} \right)_R = \left( \frac{dE}{dx} \right)_e \times \left( \frac{Z}{\xi Z_1 Z_2} \right) \tag{2.5}
\]

Both the energy loss and velocity of the ions are reduced in such a way that the LSS values fall on a straight line intersecting the origin [22] (shown in Fig.2.3). For Mylar, we have used Bragg’s Rule for calculating stopping power. According to Bragg’s Rule, energy loss of a compound target is the sum of the energy losses of the constituent elements weighted by the abundance of the elements [2]. We have also calculated the stopping power of Mylar by substituting the effective atomic number of Mylar \( (Z_{eff} = 6.46) \) in the equation 2.2. At high velocities \((v > v_c)\), Bethe formula can be used to calculate stopping power. Bethe formula is given by

\[
S_{E,H} = \frac{4\pi Z_1^2 Z_2 e^4}{mv^2} N \ln \frac{2mv^2}{I} \tag{2.6}
\]

Where \( I \) is the mean excitation energy of a target atom. The logarithmic term \( L(X) \) can be replaced by an universal function of a parameter \( X = v^2 / v_o^2 Z_2 \). For low velocities \([23]\) \( X < 10 \).

\[
L(X) = 1.36X^{1/2} - 0.016X^{3/2} \tag{2.7}
\]

Fano [24] found that above equation overestimate \( L(X) \) for stopping media of low atomic number, since the statistical treatment was not expected to be valid for atoms containing only a few electrons [25].
For intermediate energy region, there is no concrete theory that can satisfactorily explain the stopping power maximum. Varelas - Biersack [20] suggested an approximation for stopping power in the intermediate energy region. This is given in terms of values obtained using low velocity formula $S_{E,L}$ and high velocity formula $S_{E,H}$ as

$$S_{E,IM}^{-1} = S_{E,L}^{-1} + S_{E,H}^{-1}$$

we have used this approximation to estimate stopping powers in the intermediate region.

2.4 RESULTS AND DISCUSSION

2.4.1 MYLAR

To best of our knowledge very few energy loss measurements have been carried out for $^{48}$Ti and $^{16}$O ions in Mylar ($C_{10}H_8O_4$) foil [25-30]. The choice of the mylar was due to its wide application in ERDA experiments with thin windows for gaseous detectors. The energy loss of $^{16}$O and $^{48}$Ti ions in Mylar foil were theoretically calculated using LSS formula, Bethe formula and Varelas Biersacks approximation using these formulae with and without Bragg’s rule and results are tabulated in Table 2.1. Bragg’s rule calculations gave slightly lesser values of stopping power. Calculations based on LSS formulation (using effective $Z_2$ (Mylar)= 6.46) gave almost same values as experimental result except for 0.11 Mev/amu $^{48}$Ti ions. It is already a well known fact that the LSS Theory under estimates the stopping power values at low velocities. For 0.62 (MeV/amu) $^{16}$O ions, Varelas - Biersack approximation gave better value compared to the other methods. This is expected because the 0.62 (MeV/amu) $^{16}$O ions belong to the intermediate energy region. We also calculated the stopping power of Mylar for both ions using Ziegler scaling laws [31]. For calculating proton stopping power of mylar for equal velocities of $^{16}$O and
we used the scaling formula given in the appendix of Reference [32]. Both TRIM-92 and Ziegler scaling calculations gave slightly higher values than experimental values in all cases. Jin Chang wen et al [30] reported that at lower energies there is a remarkable disparity between TRIM-92 and experimental values. We didn’t observe such difference in the case of $^{48}$Ti ions.

2.4.2 CARBON

Electronic stopping powers of Carbon [thickness = $72\mu \text{gm/cm}^2$] were experimentally measured using ERDA technique for $^{16}$O, $^{27}$Al, $^{48}$Ti, $^{56}$Fe and $^{64}$Cu ions. Our experimental results were compared with experimental data obtained from M. Abdessclam et al. in the same energy range [10 12] and theoretical as well as semi empirical models. These results are shown in Table 2.2 - 2.4 respectively. Typical spectra obtained in these experiments and the method of analysis has already been indicated in our earlier study [7]. We notice from the table that LSS theory is valid up to the critical velocity limit $v_c$ and the corresponding critical energy $E_c$ is calculated for each projectile ion. For $^{16}$O, $^{27}$Al, $^{48}$Ti, $^{56}$Fe and $^{64}$Cu the $E_c$ values are 6.35, 20.47, 73.37, 106.96 and 141.40 MeV respectively. The LSS values agree better for the ion energies above 0.7 MeV/amu. For lower ion energies these values tend to lie increasingly lower than the experimental values as one goes towards the low ion energies. For lower energy values, (For $^{48}$Ti, $E/A < 0.945 \text{Mev/amu},$ $^{56}$Fe and $^{64}$Cu, all data in Table 2.4) LSS theory underestimates electronic stopping power and for higher energy values (For $^{27}$Al, $E/A > 0.45 \text{Mev/amu}$) it overestimates the stopping power. This behaviour is one of the shortcomings of LSS theory and is explicitly shown in Fig.2.3. For $^{16}$O case, Varelas Biersack approximation [20] gives a good estimation of electronic stopping power. LSS theory is not valid in this region ($v > v_c$) because all the energies of oxygen ions fall in the intermediate energy region.
(For this reason, we did not plot oxygen data in fig.2.3). There is a reasonable agreement between experimental data and values based on TRIM-92 code. However, there is a tendency of these values remaining somewhat higher than experimental values. The experimental data obtained from M. Abdesselam et. al. [10-12] in the same energy range are in good agreement with our results.

### 2.4.3 FUTURE PLANS

Our technique of performing the energy loss measurements is simple and convenient, specially when it is desired to cover a wide range of ion energies for various ion species. However, experimental data may require higher precision in order to discern oscillations in stopping power as a function of Z\textendash (for constant velocity values) in a given absorber. Also, it has been felt that the absorber thickness needs to be varied as the ion energies are changed over a wide range. It is planned to overcome these limitations by modifying the experimental set up to (i) permit a tilt in the absorber holder position, (ii) use of dE-E gas detector telescope or even a focal plane detector placed in a Recoil Mass Spectrometer (RMS). The dE (gas) - E (Semiconductor) detector can help in the present set up to provide Z\textendash discrimination, whenever required. The RMS helps in covering, forward recoil angles between 0° and 30°, not available presently. We have measured the electronic stopping power of mylar and carbon specifically in the projectile velocity region greater than 2 − 3 v_o (shown in Table 2.1 - 2.4). As mentioned earlier, it has been theoretically predicted [9] that above 2 − 3 v_o units of projectile velocities, Z_i oscillations in the electronic stopping power wash out. With the existing data, we can't deduce any such effect and we will be shortly measuring the stopping power of carbon and other target materials for various species of ions which were not covered so far in our experiments [7,8]. More importantly, the channeling stopping power measurements are planned for medium velocity region near maxima as soon as goniometer is acquired at
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N.S.C palletron facility. The channeling stopping power data is available either in high velocity region [33,34] or low velocity region where the oscillations exist and have been adequately explained.
References


References


Experimental and Theoretical Values of Stopping Power of Mylar for Ti and O ions.

<table>
<thead>
<tr>
<th>Ion</th>
<th>Energy (MeV/amu)</th>
<th>Expt</th>
<th>Trim-92</th>
<th>LSS</th>
<th>Bethe</th>
<th>Ziegler scaling</th>
<th>Varelas Biersack</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>0.11</td>
<td>0.27</td>
<td>0.27</td>
<td>0.19</td>
<td>0.19</td>
<td>0.25</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.29</td>
<td>0.32</td>
<td>0.36</td>
<td>0.31</td>
<td>0.31</td>
<td>(0.28)</td>
<td>0.40</td>
</tr>
<tr>
<td></td>
<td>0.42</td>
<td>0.37</td>
<td>0.40</td>
<td>0.37</td>
<td>0.37</td>
<td>(0.34)</td>
<td>0.43</td>
</tr>
<tr>
<td></td>
<td>0.55</td>
<td>0.41</td>
<td>0.42</td>
<td>0.42</td>
<td>0.42</td>
<td>(0.39)</td>
<td>0.44</td>
</tr>
<tr>
<td>O</td>
<td>0.62</td>
<td>0.13</td>
<td>0.14</td>
<td>0.27</td>
<td>0.27</td>
<td>(0.24)</td>
<td>0.29</td>
</tr>
</tbody>
</table>

Values shown in brackets are calculated using Bragg's rule.
Experimental and Theoretical Values of Stopping Power of Carbon for $O$ and $Al$ ions.

<table>
<thead>
<tr>
<th>Ion</th>
<th>$V \times 10^8$ (cm/sec)</th>
<th>$E$ (Mev/amu)</th>
<th>STOPPING POWER ($\text{MeV-cm}^2/\text{mg}$)</th>
</tr>
</thead>
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<tr>
<td></td>
<td></td>
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<td>EXPT</td>
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<tr>
<td>$O$</td>
<td>10.03</td>
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<td>9.3</td>
</tr>
<tr>
<td></td>
<td>11.55</td>
<td>0.69</td>
<td>8.3</td>
</tr>
<tr>
<td></td>
<td>13.05</td>
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<td>8.3</td>
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<td>14.39</td>
<td>1.07</td>
<td>8.3</td>
</tr>
<tr>
<td>$Al$</td>
<td>7.87</td>
<td>0.32</td>
<td>16.2</td>
</tr>
<tr>
<td></td>
<td>9.33</td>
<td>0.45</td>
<td>16.7</td>
</tr>
<tr>
<td></td>
<td>10.77</td>
<td>0.60</td>
<td>16.4</td>
</tr>
<tr>
<td></td>
<td>12.04</td>
<td>0.75</td>
<td>16.9</td>
</tr>
<tr>
<td></td>
<td>13.34</td>
<td>0.92</td>
<td>16.0</td>
</tr>
<tr>
<td></td>
<td>14.52</td>
<td>1.09</td>
<td>15.0</td>
</tr>
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</table>

The experimental stopping powers measured (for energies close to the energies of our experiment) for $O$ in carbon by M. Abdesselam et al. [11] are given below. (Incident energy value ($\text{MeV/amu}$) and corresponding stopping power value ($\text{MeV-cm}^2/\text{mg}$) is given in brackets).

$0.50\ (9.69),\ 0.73(9.32),\ 1.04(8.46)$. 


Experimental and Theoretical Values of Stopping Power of Carbon for Ti ion.

<table>
<thead>
<tr>
<th>Ion</th>
<th>V x 10^8 (cm/sec)</th>
<th>E (MeV/amu)</th>
<th>STOPPING POWER (MeV-cm²/mg)</th>
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</thead>
<tbody>
<tr>
<td></td>
<td></td>
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<td>EXPT</td>
</tr>
<tr>
<td>Ti</td>
<td>5.73</td>
<td>0.17</td>
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</tr>
<tr>
<td></td>
<td>6.67</td>
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<td>8.23</td>
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<td></td>
<td>8.90</td>
<td>0.41</td>
<td>28.9</td>
</tr>
<tr>
<td></td>
<td>9.74</td>
<td>0.49</td>
<td>31.2</td>
</tr>
<tr>
<td></td>
<td>10.41</td>
<td>0.56</td>
<td>31.7</td>
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<td></td>
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<td>0.79</td>
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<td></td>
<td>12.97</td>
<td>0.87</td>
<td>34.4</td>
</tr>
<tr>
<td></td>
<td>13.48</td>
<td>0.94</td>
<td>33.9</td>
</tr>
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</table>

The experimental stopping powers measured (for energies close to the energies of our experiments) for Ti in carbon by M. Abdesselam et. al. [11] are given below:

(Incident energy value (MeV/amu) and corresponding stopping power value (MeV - cm²/mg) given in brackets).

0.45 (31.09), 0.69 (31.71), 0.78 (32.06), 0.89 (32.36), 0.98 (32.37)
Experimental and Theoretical Values of Stopping Power of *Carbon* for *Fe* and *Cu* ions.

<table>
<thead>
<tr>
<th>Ion</th>
<th>V x 10^6 (cm/sec)</th>
<th>E (Mev/amu)</th>
<th>STOPPING POWER (Mev·cm²/mg)</th>
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<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>EXPT</td>
</tr>
<tr>
<td>Fe</td>
<td>6.52 (0.22)</td>
<td>25.1</td>
<td>27.2</td>
</tr>
<tr>
<td></td>
<td>7.87 (0.32)</td>
<td>29.9</td>
<td>29.9</td>
</tr>
<tr>
<td></td>
<td>9.01 (0.42)</td>
<td>32.5</td>
<td>32.7</td>
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<td></td>
<td>10.12 (0.53)</td>
<td>33.8</td>
<td>34.7</td>
</tr>
<tr>
<td></td>
<td>11.47 (0.68)</td>
<td>35.4</td>
<td>36.3</td>
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<tr>
<td></td>
<td>12.36 (0.79)</td>
<td>35.6</td>
<td>37.1</td>
</tr>
<tr>
<td>Cu</td>
<td>6.37 (0.21)</td>
<td>26.0</td>
<td>28.4</td>
</tr>
<tr>
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<td>7.49 (0.29)</td>
<td>28.7</td>
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<td>8.69 (0.39)</td>
<td>33.3</td>
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<td>9.74 (0.49)</td>
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<td></td>
<td>10.77 (0.60)</td>
<td>37.9</td>
<td>38.7</td>
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</table>

The experimental stopping powers *measured* (for energies close to the energies of our experiments) for *Cu* in *carbon* by *M. Abdesselam et. al.* [10] are given below (Incident energy value *(Mev/amu)* and corresponding stopping power value *(Mev·cm²/mg)* is given in brackets).

0.55 (37.63), 0.72 (40.94).
Fig 2.1: **Schematic classification** of different energy loot region*. The *curves* are qualitative and coordinates are not to *scale*. The *solid curve represents* electronic energy loss and *dotted curve represents* nuclear energy /OM.
Fig 2.2: Schematic diagram of ERDA experimental setup.
Fig 2.3: The Reduced Stopping Power \( (dE/dx) \) (in MeV cm\(^2\) / my) as a function of Reduced ion velocity \( V_{\text{red}} \). Solid line represents LSS values, filled triangles represent energy loss of \(^{27}\text{Al}\) ions, open circles show energy loss of \(^{44}\text{Ti}\) ions, filled squares represent energy loss of \(^{56}\text{Fe}\) ions and open triangles show the energy loss of \(^{64}\text{Cu}\) ion*.