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FAST HEAVY IONS IN MATTER
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AND P. ARMBRUSTER

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ABSTRACT

A detailed study of the stopping power and energy loss straggling is presented for heavy ions ($18 \leq Z_1 \leq 92$) in the energy region of (0.5 - 10.0) MeV/u using different solids and gases ($2 \leq Z_2 \leq 92$) as absorbers. The Z_2 -oscillation and the density effect in the stopping power and the contribution of charge fluctuations induced by atomic collision processes to the energy loss straggling are demonstrated and compared to theoretical and semiempirical predictions.

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1. INTRODUCTION

The energy loss and energy loss straggling of energetic projectiles while interacting with a stopping medium have been widely investigated theoretically and experimentally in the last seven decades. The experimental results are usually compared to the Bohr [1], Bethe [2], and LSS [3] stopping theories which were developed for different ranges of projectile velocities.

From a bibliography search [4] it is seen that most of the experimental results are for projectiles with atomic numbers $Z_1 < 20$ at relative low velocities ($E/M < 1$ MeV/u). For these mass and energy regions, the dependence of the stopping power S on Z_1 , Z_2 and velocity is known reasonably well. Especially the fine structure in the dependence of S on Z_1 (Z_1 -oscillations) [5] and on the atomic number of the stopping medium Z_2 (Z_2 -oscillations) [6] should be mentioned. For heavier projectiles and higher energies, around the maximum of the stopping power where the ions are partially stripped, none of the existing stopping theories is valid and only few experiments have been published. The data available do not give a clear picture for the detailed dependence of S on Z_1 , Z_2 , and velocity. It was found recently [7] that the Z_2 -oscillations still exist for 1.3 MeV/u U ions whereas for α -particles at this energy they are washed out.

In order to overcome the problem of lack in experimental stopping power data of heavy ions at higher energies, scaling laws, and the concept of the effective charge were used to create several stopping power tables as the widely used Northcliffe and Schilling [8] and the recently published Ziegler [9] and Hubert et al. [10] tables.

There is almost no experimental data for the energy loss straggling δE of heavy ions in matter. The basic theories [1,11,12] deal with the contribution of the nuclear collisions and the electronic collisions to the straggling by assuming a constant charge for the projectile. However, large discrepancies exist between the experimental and calculated energy loss straggling for heavy ions, suggesting that also charge fluctuations induced by atomic collision processes during the slowing down in the target contribute significantly to the δE of heavy ions [12,13,14].

We have done systematic measurements of S and δE in the region of $18 \leq Z_1 \leq 92$, $2 \leq Z_2 \leq 92$, and $0.2 \leq E/M \leq 10.0$ MeV/u. Representative results are given here together with a comparison to the published results of lighter ions and to theoretical and semiempirical predictions.

2. EXPERIMENTAL PROCEDURE

2.1 Stopping Power Measurements

The experimental system for energy loss measurements of projectiles in the energy region up to 1.4 MeV/u in solids has been described already earlier [7] and, therefore, will be given here in a condensed form. The UNILAC accelerator provides heavy ion beams of $18 \leq Z \leq 92$ in the energy interval (1.4 - 10.0) MeV/u. The energy could be further degraded by inserting homogeneous plastic stacks into the beam line. In the energy region up to 1.4 MeV/u the measurements were carried out with a 1.4 m time of flight (TOF) spectrometer. The start signal was derived from secondary electrons released from a thin carbon foil and amplified in a channel plate. A cooled, totally depleted, silicon surface barrier detector was used as the stop detector. The intrinsic resolution of the TOF spectrometer was 100 ps fwhm. Its total resolution was 300 ps including 280 ps contribution from the incoming beam. For higher energies a 4.0 m TOF spectrometer was used. At these energies the energy resolution of the beam is much better and a total resolution of 80 ps at 10 MeV/u has been reached.

Energy losses in gases were measured by introducing a closed gas cell into the beam line. The plastic windows of the cell had a diameter of 3 mm and a thickness of about 50 $\mu\text{g}/\text{cm}^2$ each. The beam path length in the cell was 14.5 cm and gas pressures of (10 - 100) torr were applied. A closed copper volume in contact with the gas cell was used as a reference volume in order to eliminate temperature effects. A commercial gas controlling unit combined with a capacitance manometer regulated the pressure difference between the reference volume

and the gas cell. The pressure was measured with an absolute pressure sensor. The accuracy and the stability of the system during a measurement was better than 0.1 %. The temperature was measured in the cell with an integrated circuit temperature transducer which produced an output current proportional to the absolute temperature. The intensity of the primary beam could be adjusted continuously with the aid of specially designed attenuators. Both, the solid targets and the gas cell were mounted on vacuum feedthroughs and could be removed out of the beam line during the measurements. Therefore, the energy of the incident beam E_0 could be checked periodically during the measurements. The distance between the stop and start detectors could be changed by 20 cm, and therefore an absolute calibration of the system was obtained by measuring the time of flight in two different path lengths. The position and width of all the measured peaks were obtained by a least square fitting of a Gaussian with two exponential tails. The experimental set-up and the data handling procedure would allow energy loss measurements with an accuracy better than 1 %, unless the targets would determine the final accuracy.

2.1 Energy Loss Straggling Measurements

Energy loss straggling values δE (fwhm) are typically less than 1 % of E_0 . Therefore, small energy fluctuations may cause severe problems in the δE measurements. In order to overcome this difficulty, we added to the previously described TOF system an additional 5.9 m TOF system before the projectiles interact with the stopping medium. A schematic representation of this double time of flight (DTOF) system is given in Fig. 1. The start detector of the first TOF unit is a combination of a photomultiplier with a thin plastic scintillator film [15] and the stop detector is again a secondary electron detector [16]. With this DTOF set-up the energies of a given pro-

jectile before and after interacting with the stopping medium were determined simultaneously. By setting energy windows (typically 0.1 % of E_0) on the first TOF spectrum, one eliminates the energy fluctuations of the incoming beam. Furthermore, with the DTOF system, also energy degraded beams with a broad energy distribution could be used for δE measurements. Also here the absolute calibration of the system was obtained by changing the path length of the second TOF spectrometer.

2.3 Range Measurements

We have measured the ranges of ^{238}U projectiles in solids [17]. About 5×10^{13} uranium ions were implanted into a stack of foils having a total thickness larger than the range of the projectiles in the given solid. The number of the ^{238}U projectiles stucked in the individual foils was obtained by thermal neutron activation and then by measuring the γ -radiation following the β -decay of ^{239}Np into ^{239}Pu .

2.4 Target Properties

The thicknesses of the solid targets used in the experiments varied from 0.03 to 1.5 mg/cm². In order to get high uniformity the targets were produced by evaporation [18,19]. The thicknesses of the targets were obtained by α -spectrometry using a thin α -source and a cooled Si-surface barrier detector (resolution ~ 10 keV fwhm). The position and the width of the α -lines were obtained by least square fitting. The mean thickness and its standard deviation for each foil were obtained by using tabulated stopping power values for α -particles [20]. Targets that produced large tails in the α energy loss distribution were discarded. After the measurement the target

was removed from its frame and its average thickness was determined by weighing. The thickness obtained by these two procedures may deviate if impurities exist in the target with a considerable different stopping power, than that of the target material. Furthermore, oxidation on the target surfaces will have a larger effect on the thinner targets. Therefore, for a given Z_1 , targets with different thicknesses were used. Moreover, using different target thicknesses the influence of the multiple scattering process on the stopping power result could be checked. Fluorescence analysis was done on a limited number of targets. Inhomogeneity of the foils, surface roughness and target oxidation were found to be the main contributions to the scattering of the S values at a given Z_1 , Z_2 and E_0 . This scattering amounts up to 5 % from the total S and is assumed in the following as the representative uncertainty of the measured S results.

δE measurements are much more sensitive to target nonuniformities than S measurements, therefore, targets that were found adequate for S determinations may still have too large nonuniformities for δE measurements. Assume a projectile loses an energy ΔE while interacting with a uniform foil of thickness x . The energy loss straggling for this case is δE_{st} . If the target has a thickness nonuniformity (δx) fwhm, the additional energy loss straggling δE_x in this case will be :

$$\delta E_x = \left(\frac{\delta x}{x} \cdot \Delta E \right)$$

and

$$\delta E^2 = \delta E_{st}^2 + \delta E_x^2$$

For α -particles at 1.4 MeV/u a 1 mg/cm² carbon target of $\frac{\delta x}{x} = 2.7$ % contributes 10 % to δE , whereas for uranium projectiles at the same velocity already a value as small as $\frac{\delta x}{x} = 0.6$ % gives the same contribution to δE .

In spite of the careful measures taken in the preparation procedures of the foils, one should not be surprised that discrepancies up to factors of 2 were found in the δE values of heavy ions in single foils with the same Z_2 . Tests to measure δx values of some of our evaporated foils by surface roughness measurements with a Talystep [18] instrument, optical multiple beam interference, and electron microscopy were found to be not sensitive enough for $\delta x/x$ measurements below 1 %. It was therefore concluded that only selected plastic foils could be used for δE measurements in solids. Therefore, most of the δE data represented here were obtained for gases having a purity better than 99.99 %.

3. STOPPING POWERS

3.1 Z_2 Dependence

The dependence of S on Z_2 is given in Fig. 2 for Pb and Xe projectiles at 1 MeV/u. The experimental results are also compared to the tabulated values of Ziegler [9] and of Northcliffe and Schilling (NS) [8]. A comparison is made also for values obtained by scaling the proton stopping powers [21] with the effective charges calculated from Forster et al. [22]. The Z_2 -oscillations of the experimental stopping powers are clearly seen. The tabulated values as well as the scaled ones deviate considerably from the experimental results at the maxima of the oscillations. At 1 MeV/u the Z_2 oscillations of light particles as protons and helium ions are already washed out. Therefore, scaling procedures with effective charges (q_{eff}) not depending on Z_2 can reproduce qualitatively the Z_2 -oscillations of heavy ions only at energies smaller than 1 MeV/u. For higher energies and for heavy projectiles the oscillations are still pronounced whereas smooth dependence of S on Z_2 would be obtained by the scaling.

In a previous paper [7] it was shown that for energies up to 1.3 MeV/u the amplitudes of the Z_2 -oscillation decrease with increasing velocity and decreasing Z_1 . A detailed picture of these oscillations for uranium projectiles in solids for energies up to 10 MeV/u is given in Fig. 3. The decrease of the amplitude with velocity can be followed up to energies as high as 3.6 MeV/u at $Z_2 \approx 30$ and 40.

In Fig. 4 the measured ranges of $^{1.4} \text{MeV/u } ^{238}\text{U}$ projectiles in solids are compared with the NS tabulated values. At $Z_2 \approx 22$

and 30 a maximum and minimum exists in the Z_1 -oscillation of S which causes Z_1 -oscillations in the ranges but with opposite amplitudes as seen in the figure. The NS values for electronic S are underestimated and one would expect higher range values from these tables but this effect is partially reduced by the too high values taken for the nuclear stopping [23].

3.2 Density Effect

According Bohr and Lindhard [24], the ionic charges of projectiles in solids should be higher than those in gases. This density effect would cause higher stopping powers in solids than in gases. However, in most of the published experimental results no evidence was found for this effect [25] and it is generally accepted that the higher ionic charges found for projectiles after a solid target were caused by Auger cascades after the projectile left the target as suggested by Betz and Grodzins [26]. In Fig. 5 we represent the stopping powers of 3.6 and 7.9 MeV/u uranium projectiles in solids and gases. It is clearly seen that the S values in gases are lower by up to 20 % than those in solids. The effective charge of a heavy ion $q_{eff,HI}$ is defined by

$$\frac{1}{q_{eff,p}} \cdot S_p = \frac{1}{q_{eff,HI}} S_{HI}$$

where S_p is the stopping power of protons having the same velocity as the heavy ion.

The q_{eff} of uranium at 3.6 and 7.9 MeV/u in solids and gases are shown in Fig. 6. The q_{eff} in gases are up to 10 % lower than those in solids. This result suggests that only a part of the higher ionic charges found after a solid target results from losing electrons by Auger cascades after the target. The density

effect contributes to the difference by up to 10 % in the charges of an ion in a solid compared to those in a gas target. A higher charge in solids than in gases has also been found by analyzing the projectile M x-ray spectra [27]. It was shown that the projectiles in the average carry more than one vacancy in the M-shell and therefore they may be taken as representative for the average of all projectiles, as investigated by the q_{eff} from stopping power measurements.

3.3 Z_1 Dependence

Fig. 7 shows the experimental stopping powers of nine projectiles in C, Al, Zr, and Au at 1.0 MeV/u. It is seen that for a given target the stopping power increases monotonously with Z_1 . In the work of Ward et al. [5] and of Hvelplund and Fastrup [28] Z_1 -oscillations were reported for projectile velocities of $v \leq v_0$ ($v_0 = 2.18 \times 10^8$ cm/sec). Here, at $v = 6.3 v_0$ the maximum at $Z_1 = 22$ and the minimum at $Z_1 = 29$ that were found at the low velocities, are not seen.

The minimum beyond $Z_1 > 56$ in the S -values indicated in measurements with fission products [29] and heavy ion reaction products [30,31] could not be investigated as beams in this Z_1 -range have not been available for our measurements. The question of Z_1 -oscillations needs further detailed investigations.

3.4 Velocity Dependence

The experimental stopping powers of ^{235}U projectiles in the energy region (0.5 - 10) MeV/u in C, Ti, and Au are shown in Fig. 8. The experimental results are also compared to the

tabulated values of Hubert et al. [10], Ziegler [9], and NS [8]. Generally, the Hubert tables give the best predictions for the S values. The NS tables underestimate the values by up to 15 % in C while for Ziegler the discrepancies are up to 20 %. For larger Z_1 values all the tabulated values give better estimates of S. The NS values are considerably better than the Ziegler ones for energies up to 6 MeV/u. It should be also noted that a wrong location of the maximum in S causes large errors in the tabulated values.

4. ENERGY LOSS STRAGGLING

δE results for 1.4 MeV/u Pb ions attenuated in different thicknesses of N₂, Ar, Kr, and Xe are given in Fig. 9. It is seen as expected that δE increases with increasing Z_2 . The δE values for large energy losses show a maximum. This can be explained by the fact that at the energy region where S is proportional to v , the low energy side of the energy peak has lower S values than the high energy side and, therefore, will cause a reduction effect on the δE while the statistics of the collisions in the slowing down process cause an increase in δE [32]. In Fig. 10 the δE of 1.4 MeV/u Kr, Xe, and Pb ions is shown for different energy losses in Kr. It can be seen that δE increases with Z_1 . For Kr projectiles the δE reached the maximum while for the heavier projectiles the maximum seems to be reached at higher energy losses.

In Fig. 11 we compare the δE of 1.4 MeV/u ions in gas (methane) and in solid (plastic) which have about the same average atomic composition. It can be seen that within the accuracy of the measurements no significant difference is seen at these energies.

In order to demonstrate the contribution of charge fluctuations induced by atomic collision processes to δE for heavy ions, we represent in Fig. 12 δE of 1.4 MeV/u He (from ^{41}Am) and Pb ions in argon at different energy losses. The experimental results are compared to the calculated ones obtained from the theory of Tschalär [11] and corrected for shell effects according to Livingston [33]. It is seen that the theory predicts with good accuracy the δE results for fully stripped He ions, whereas for the partly stripped Pb ions the agreement is poor. By the use of a Monte Carlo Code [34], δE was calculated by taking into account

scattering and energy loss as well as charge fluctuations introduced into the calculation by experimental charge exchange cross sections [35]. The results are shown in Fig. 12, suggesting that charge fluctuations are contributing significantly to the overall energy straggling. This result is in agreement with the results obtained recently for carbon and oxygen ions in Al [36].

The discrepancy between the Monte Carlo calculation and the measured values probably is due to the difference between the charge exchange cross sections determined long after deexcitation of the ions used in the calculation and the actual cross sections within the gas. A larger value of the ionic charge distribution within the gas than after passage through the gas would account qualitatively for the discrepancy.

5. CONCLUSIONS

From the results presented here and in a previous paper [7] we conclude that the maxima and minima in the Z_1 -oscillations of heavy ions are roughly at the same Z_1 values as found for α -particles. The amplitudes of these oscillations decrease with increasing velocity and disappear for light projectiles at lower velocities than for the heavier ones. For example, the Z_1 -oscillations at $Z_1 \approx 22$ disappear for α -particles at ~ 1 MeV/u while for uranium projectiles at ~ 3.6 MeV/u. Furthermore, the amplitudes of the oscillations at higher Z_1 values are washed out at higher velocities than at low Z_1 values.

The stopping powers of heavy ions in gases are smaller by up to 20 % than those measured in solids. This density effect is pronounced at energies of several MeV/u where the Z_1 -oscillations of S are negligible. The ionic charges in gases at these velocities are therefore smaller by up to 10 % compared to the values in solids. This phenomenon, together with the Auger electrons lost by the projectiles after leaving a solid target, contribute to the difference in the ionic charges of a given projectile after traversing a solid and a gaseous target.

For energies smaller than 2.5 MeV/u the NS [8] tables were found to underestimate the S values especially at Z_1 values near the maxima of the Z_1 oscillations. This is explained by the fact that in these tables no structure was introduced for the dependence of S on Z_1 and the experimental S data used were at roughly the minima of the Z_1 -oscillations.

Although the Z_1 -oscillations were introduced in the Ziegler [9] tables, they are still underestimating the S values at the maxima of the Z_1 -oscillations.

For energies higher than 2.5 MeV/u where the Z_2 -oscillations for most of the projectiles are negligible, the Hubert et al. [10] tables give the best predictions of S. Also here the NS [8] and Ziegler [9] tables underestimate the S values, especially for light stopping media.

It was found that the energy loss straggling measurements with heavy ions are extremely sensitive to nonuniformities of the target. Although the special measures taken in order to get uniform solid targets, it was found that most of these targets were not adequate for δE measurements. However, it was found that the δE for high uniform plastic targets is roughly the same as for methane.

It was shown that the energy loss straggling of completely stripped He projectiles in gases is predicted by the collision theory with a reasonable accuracy, while for partly stripped heavy projectiles, at the same velocity, serious discrepancies were found. By introducing to the δE calculations of heavy ions charge fluctuations induced by the collision processes a significant improvement was obtained, suggesting that such processes contribute dominantly to δE .

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FIGURE CAPTIONS

- Fig. 1 Experimental set-up for the double time of flight (DIOF) system.
- Fig. 2 The experimental stopping powers in different stopping media for Pb and Xe ions at 1 MeV/u compared to semi-empirical prediction as explained in the text.
- Fig. 3 Stopping cross sections for uranium ions in Z₂ in the energy region (0.5 - 10.0) MeV/u.
- Fig. 4 Experimental ranges of 1.4 MeV/u ²³⁸U projectiles in different solids, compared to NS [8] tables.
- Fig. 5 Uranium stopping powers in solids and gases at 3.6 and 7.9 MeV/u.
- Fig. 6 Effective charges of uranium ions in gases and solids at 3.6 and 7.9 MeV/u.
- Fig. 7 Stopping powers of different projectiles in C, Al, Zr, and Au at 1 MeV/u.
- Fig. 8 Uranium experimental stopping power data in C, Ti, and Au in the energy region (0.5 - 10) MeV/u compared to the tabulated values of NS [8], Ziegler [9], and Hubert et al. [10].
- Fig. 9 Energy loss straggling of 1.4 MeV/u Pb ions in N₂, Ar, Kr, and Xe vs. the energy loss in the gas.

Fig. 10 Energy loss straggling of 1.4 MeV/u Kr, Xe, and Pb ions in Kr vs. the energy loss in Kr.

Fig. 11 Energy loss straggling of 1.4 MeV/u Pb ions in plastic and in methane vs. the energy loss in these media.

Fig. 12 Experimental energy loss straggling of 1.4 MeV/u He and Pb ions in Ar vs. energy loss in Ar, compared to predictions according collision theory [11,33] and results from a Monte Carlo code [34].

FIG.1

FIG. 2

FIG. 3

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FIG.12