Stopping powers of the noble gases for (0.3–10)-MeV nitrogen ions

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Stopping powers of the noble gases for 300-keV to 10-MeV nitrogen ions were measured using a time-of-flight technique. The stopping power was directly determined from changes in ion time of flight over a fixed distance as a function of the target-gas pressure of a differentially pumped, windowless gas cell. The measured stopping-power values are compared to predictions of the modified Firsov theory of Land and Brennan (FLB) [At. Data Nucl. Data Tables **22**, 235 (1978)] and to values compiled by Ziegler *et al.* [*Stopping and Range of Ions in Solids* (Pergamon, New York, 1985)]. The measured values are close to the FLB values at $v=v_0$ for low target atomic number Z but are somewhat lower at higher Z. They are systematically smaller than those of Ziegler *et al.* at nitrogen energies below 2.5 MeV and larger from 2.5 to 10.0 MeV. For projectile velocities greater than v_0 the stopping in general is found not to be proportional to velocity.

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I. INTRODUCTION

There is significant interest in accurately determining the stopping powers of various target materials for energetic charged particles. This interest has been motivated by the increasing use of ion beams for the modification and analysis of materials. In an extensive experimental and theoretical program at the Naval Surface Warfare Center (NAVSWC), studies of range distributions and stopping powers of heavy ions have been undertaken. In previous measurements range distributions of implanted nitrogen projectiles were determined for solid elemental targets ranging from $Z=6$ to $Z=81$ [1]. From these measurements stopping powers were inferred under an assumption of velocity-proportional stopping. In related theoretical work Land and Brennan [2] have extended a model for low-velocity electronic stopping power applicable to all combinations of incident and target atoms. This work is based on a model of Firsov [3] for the energy loss in atomic excitation by ions which, in turn, was employed by Teplova *et al.* [4] for the determination of the stopping power of a Fermi-Thomas atom. These ideas were subsequently applied to include atomic shell effects for incident projectiles by Cheshire and Poate [5] in the case of channeling and by Bhalla, Bradford, and Reese [6] for amorphous carbon. The resulting stopping powers exhibited an oscillatory behavior as a function of projectile atomic number, Z_1. The model of Land and Brennan (FLB for Firsov-Land-Brennan) predicts oscillations in the stopping power as a function of the target atomic number, Z_2. In particular, minima in the stopping power were found at positions of closed d shells and minor minima at the positions of closed p shells (the noble gases).

The study reported here extends the previous NAVSWC determination of stopping powers for nitrogen ions to the noble gases and to a much higher energy range, up to 10 MeV. In order to explore the extent of the region in which velocity-proportional stopping holds, particular emphasis was placed on the velocity range of $v_0<v<2.0v_0$, where v is the projectile velocity and v_0 the Bohr velocity. This velocity range corresponds roughly to the projectile energy range for ^{14}N from 350 keV, the lowest energy readily accessible on the accelerator, to 1400 keV. Velocity-proportional dependence, by which we explicitly mean that $S(v)=c(v-v_1)$ with $v_1=0$ and $c=\text{const}$, is generally predicted by theory applicable to the low-velocity electronic stopping [4,7,8]. Results at higher energies are of interest for the determination of hydrogen depth profiling in materials using the resonance reaction $H(^{15}N,\alpha\gamma)^{12}C$.

In Sec. II we present the experimental approach used in this study which takes advantage of two techniques cited by Powers [9], a differentially pumped gas cell (for target parameter definition) and the time-of-flight spectrometer (for direct energy-loss measurement). In the next section, the stopping powers of the noble gases for nitrogen projectiles are presented. Comparison is made between the results and the predictions of the modified Firsov model of Land and Brennan at $v=v_0$ and the compilations of Ziegler, Biersack, and Littmark [10]. In addition, isotopic effects were examined by comparison of the results from ^{14}N and ^{15}N ions on He. Finally, conclusions based on these results and comparisons with theory are presented.

II. EXPERIMENTAL ARRANGEMENT

Ion beams of ^{14}N and ^{15}N were accelerated using the 2.5-MV Van de Graaff and the 3-MV positive-ion tandem accelerators at the NAVSWC White Oak Laboratory. Mass-energy selection is made by a 90° analyzing magnet. The energetic ions are injected through a windowless, differentially pumped gas cell into the time-of-flight chamber. The experimental apparatus and procedures were previously presented in more complete detail [11]. Particular care was taken to ensure that the research grade (99.999% pure) stopping gas was not contaminated especially during the transfer from the gas bottle to the gas cell. For the He stopping measurements it is observed that even small amounts of contaminants yield erroneous results since the stopping power of the contaminants (usually air) is much larger than that of helium and thus dominate the energy-loss process. The gas pressure is monitored by a differential pressure controller which regulates the pressure to better than 5% in the range from 0.01 to 1.0 torr. The time-of-flight (TOF) chamber contains start and stop microchannel plate (MCP) detectors triggered by electrons ejected from thin-carbon-foil pickoff units. Signals from these MCP detector systems are amplified and fed into a time-to-amplitude converter (TAC). The resulting time spectra are analyzed with an appropriate multichannel analyzer-computer system. With increasing pressures in the gas cell, the time intervals between start and stop pulses lengthen indicating the appropriate energy loss of the projectile. The resulting shifts of the centroids in the time spectra are computed from Gaussian fits to the spectra. Stopping powers are evaluated by correlating the centroid shifts to gas cell pressures as explained below.

III. DATA REDUCTION

To first order in $\Delta\tau/\tau_0$, the average energy loss (ΔE) in the gas cell is related to the time shift ($\Delta\tau$) of the centroid by

$$\Delta E \cong 2E_0\Delta\tau/\tau_0 = \frac{2E_0}{D}\left[2\frac{E_0}{M}\right]^{1/2}\Delta\tau\ , \qquad (1)$$

where E_0 is the initial projectile energy, D the length of the time-of-flight chamber, M the projectile mass, and τ_0 the projectile time of flight for an evacuated gas cell. Hence, the stopping cross section, ϵ, is approximated by

$$\epsilon \cong \frac{2E_0 k_B T}{DL}\left[\frac{2E_0}{M}\right]^{1/2}\left[\frac{\Delta\tau}{\Delta P}\right]\ , \qquad (2)$$

where L is the gas cell length, T the gas temperature, k_B Boltzmann's constant, and $\Delta\tau/\Delta P$ the ratio (slope) of the centroid shift for changes in gas pressure. A typical run at a particular energy E_0 in a specific gas consists of several time-of-flight measurements as a function of gas pressure. Sufficiently low gas pressures are used to ensure that the $\Delta\tau/\Delta P$ ratio remains linear. Applying a linear regression analysis to the time-pressure data, a slope ($\Delta\tau/\Delta P$) and its associated statistical uncertainty is obtained. With this procedure the problem of having to determine the absolute time of flight of the incident ion is avoided, and it is not necessary to account for the timing differences in electronic signals in start and stop detector systems. This procedure also provides a convenient means to take several $\Delta\tau$ measurements at a single ion energy and to compute a statistical average and uncertainty. This method of data analysis introduces errors based on the uncertainty of the time-pressure slope typically between 1.5% and 4% at a single energy with an occasional energy point yielding an error as high as 8%. Measurements are made with changes in $\Delta\tau$ of 1–3 nsec. Using the values of Ziegler, Biersack, and Littmark [10] as a guide, a change in $\Delta\tau$ of 3 nsec corresponds to a change in stopping power of at most 1.6% over the entire energy range studied here. This change is well within our experimental error. In fact, none of the time-pressure curves show any deviation from a linear slope which could be attributed to an appreciable change of stopping power with increased pressure.

Errors in the stopping power from other sources in this study are beam energy (< 1.0%), gas pressure (< 1.0%), length of gas cell (0.5%), length of time-of-flight cell (0.5%), and gas temperature (0.3%). Adding these errors in quadrature with the 1.6% error due to a 3-nsec change in time of flight results in an error of 2.5% which is added in quadrature to the error in the stopping power resulting from the statistical uncertainty in the $\Delta\tau/\Delta P$ slope.

Finally, we have considered corrections to the length of the gas cell due to gas flow through entrance and exit apertures resulting from the pressure differential between the gas cell and the differentially pumped region (pluming). The identical entrance and exit apertures are 0.64 mm long and 0.2 mm in diameter. Following Dushman [12], calculations of λ/d, where λ is the mean free path of the gas at the cell pressure and d the aperture diameter, indicate that the flow through the aperture is a mixture of molecular and laminar. However, calculations of conductance (see Hülskötter *et al.* [13]) show that the molecular conductance is more than an order of magnitude greater than the laminar. From calculations by Steckelmacher, Strong, and Lucas [14] for molecular flow along the axis of a cylindrical tube, the flux ratio for the system described here is down to 1% at a distance of 0.9 mm from the aperture. Thus, with a length of 46 cm the effective length of the gas cell cannot increase by more than 0.2% because of pluming. This increase would systematically reduce the values of stopping power presented here by no more than 0.2%, a value which is negligible compared to the other errors in this study.

IV. RESULTS AND DISCUSSION

The measured values of the stopping powers of the noble-gas targets for nitrogen projectiles are listed in Table I as a function of projectile energy. Singly, doubly, and triply charged ions were used to cover the entire energy range studied here. However, sufficient energy overlap and duplication ensured that no significant projectile-charge-state dependence was observed outside the errors as discussed above. Thus, the values shown in Table I and elsewhere in this paper are the results of appropriate weighted averaging.

The measurements reported here represent the total stopping powers, electronic plus nuclear. However, except at the lowest energies, the nuclear stopping is only a small percentage of the electronic and within the experimental errors of the measurement. For instance, the nu-

TABLE I. Experimental stopping powers of noble gases for energetic nitrogen-ion projectiles. Stopping powers are given in units of 10^{-13} eV cm^2 per atom.

Energy (keV) \ Gas / Proj.	Helium ^{14}N	Helium ^{15}N	Neon ^{14}N	Argon ^{15}N	Krypton ^{14}N	Xenon ^{14}N
300			0.473			
350			0.613			
400	0.312		0.635		1.28	1.80
500	0.358		0.668	1.17	1.56	
600	0.375		0.835	1.63	1.96	2.77
700	0.385	0.383	0.914	1.63	2.13	
800	0.420	0.396	0.987	1.70	2.37	2.84
850	0.452					
900	0.439	0.415		2.01	2.55	
950	0.465	0.433		2.05		
980		0.449				
1000	0.486	0.437	1.14	2.18	2.68	3.44
1020		0.460				
1050		0.466				
1100	0.500	0.469	1.18		3.07	
1200	0.528	0.518	1.21	2.57	3.08	3.88
1300	0.553		1.32		3.23	
1400	0.575	0.567	1.36	2.92	3.41	4.73
1500	0.597		1.38	2.96	3.42	
1600	0.607	0.582	1.50	3.03	3.85	4.84
1700	0.642		1.55		3.76	
1800	0.660	0.636	1.54	3.18	3.98	5.81
1900	0.662				4.18	
2000	0.677	0.671	1.69	3.43	4.33	5.66
2100	0.678				4.40	
2200	0.701		1.87	3.72	4.43	6.18
2300	0.697		1.80		4.52	
2400				3.97		6.58
2500	0.767			3.57	4.84	6.50
3000	0.790		2.11	4.14	5.23	6.81
3500	0.811		2.23	4.14	5.19	6.91
4000	0.799		2.22	4.28	5.73	7.83
4500	0.809		2.28	4.26	5.64	6.91
5000	0.904		2.23	4.23	5.49	7.19
5500	0.853		2.15	4.37	5.51	7.18
6000	0.838		2.55	4.26	5.50	7.18
6500	0.872		2.31	4.15	5.24	6.88
7000	0.876		2.20	4.13	5.40	6.87
7500	0.817		2.56	3.87	5.83	7.14
8000	0.842		2.31	3.91	5.41	7.39
8500	0.817		2.32	4.06	5.34	6.91
9000	0.794		2.39	3.79	5.32	7.17
9500	0.803		2.39	3.73	5.52	7.21
10000	0.798		2.66	3.63	5.19	7.83

clear stopping (calculated from Ziegler, Biersack, and Littmark [10]) is less than 5% of the electronic stopping at 400 keV and falls to less than 2% at 800 keV. Thus, within the experimental uncertainties the stopping powers measured in this study are the electronic stopping powers for the noble gases.

Figure 1 shows the stopping powers of the noble gases as a function of ion velocity. Generally, ^{14}N was chosen

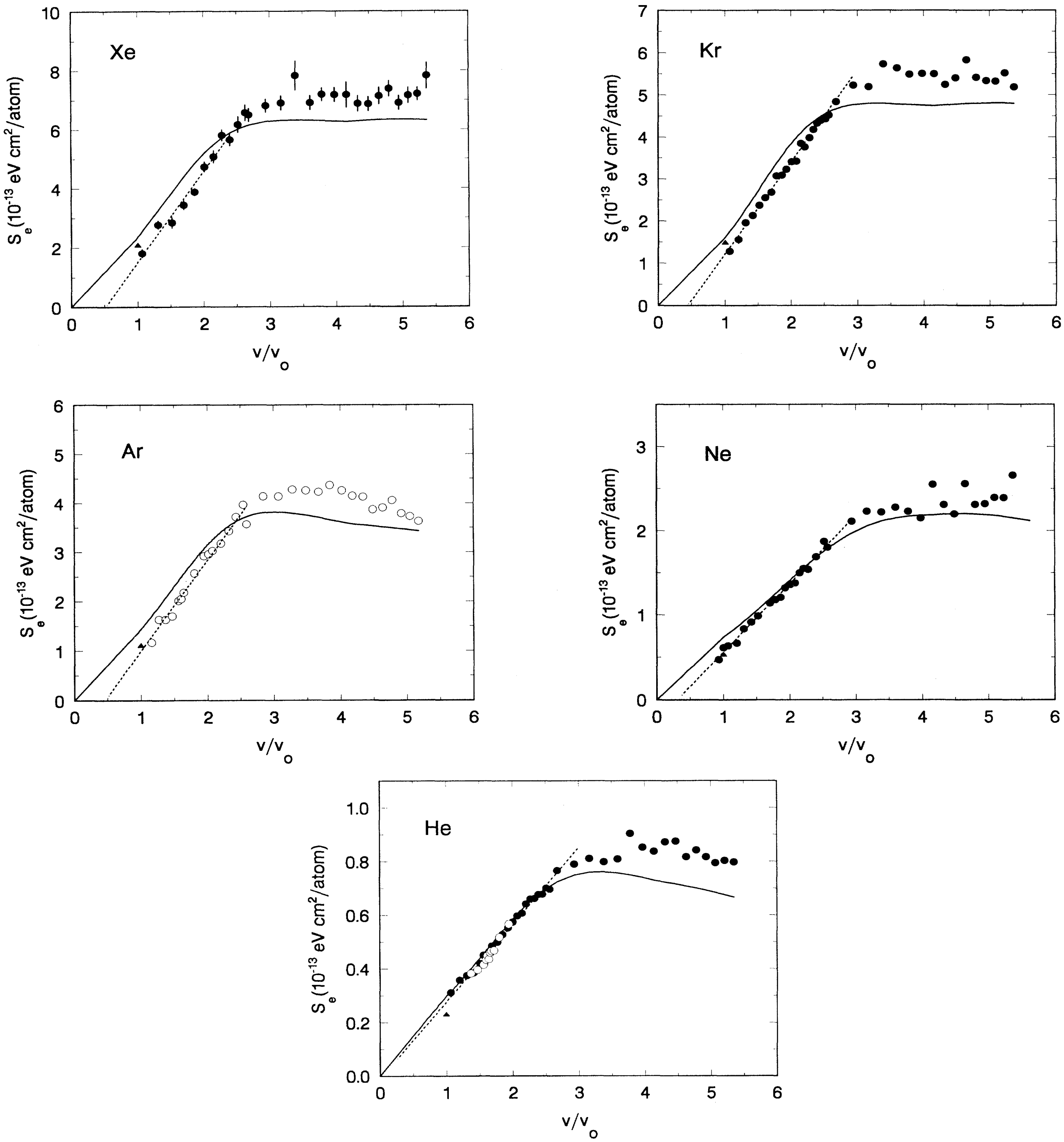

FIG. 1. Stopping powers of the noble gases for nitrogen ions as a function of reduced ion velocity (v/v_0). The values obtained for ^{14}N are shown as filled circles while those for ^{15}N are shown as open circles. While the values are generally in good agreement with the compilations of Ref. [10] (solid line), they are systematically lower than Ziegler's below $v/v_0=2.5$ and higher above $v/v_0=2.5$. The dotted curve is obtained from a linear regression of the data in the range $1<v/v_0<2.6$. The value obtained from FLB [2] is shown as a triangle at $v/v_0=1$. Shown with the data for Xe are the error bars for each experimental point which are representative for all of the experimental data of this study.

as the projectile except for the Ar target for which ^{15}N was used. The He stopping measurements were made with both ^{14}N and ^{15}N projectiles and, as expected, no difference is discernible between the two isotopes. Typical errors for this experiment are shown in the figure for xenon. Also shown are the values determined by FLB at $v/v_0=1$, the semiempirical values of Ziegler, Biersack, and Littmark [10], and a linear regression to the data below $v/v_0 \approx 2.6$ ($E \approx 2.2$ MeV). Agreement with the values of Ziegler, Biersack, and Littmark is generally good. However, the data are systematically lower for velocities below $2v_0$ and higher for velocities greater than $2.5v_0$. In the region from v_0 to $2.6v_0$ the stopping power appears to display a linear behavior in velocity. This circumstance can be exploited to examine possible velocity-proportional stopping through a linear regression to the data in this region. Except for He, the linear regression does not extrapolate to the origin but rather to a positive value of velocity, approximately $0.5v_0$. Thus, velocity-proportional behavior does not hold for these data at values of v greater than v_0. The linear regression also allows for the experimental determination of S_e at $v=v_0$ for comparison with FLB through a small extrapolation to v_0.

With respect to the results of the He stopping there appears to be a slight but consistent "dip" or depression near the velocity region of $1.6v/v_0$ where the projectile velocity matches the target electron velocity. Measurements in this region were repeated several times and the results were consistent. There is also excellent agreement between ^{14}N and ^{15}N projectiles in this region. As yet we do not have an explanation for this observation.

V. CONCLUSIONS

We have measured the stopping power of the noble gases for 300-keV–10-MeV nitrogen ions and compared these values with those of FLB and Ziegler, Biersack, and Littmark [10]. One of the stated purposes of this study was to extend the measurements of stopping power versus Z_2 for nitrogen ions to include the noble gases and to compare these values with FLB. These results are shown in Fig. 2 where the filled circles are the experimental points inferred from previous range distribution measurements in solid elemental targets [1] and the open circles are derived from the current data set. The solid curve is FLB at $v=v_0$. Table II shows a comparison at $v=v_0$ between the experimental values obtained from the linear regression fits to the data and the FLB values for the noble-gas targets.

Overall, the agreement between the experimental and theoretical values of stopping power shown in Fig. 2 is satisfactory. However, there is clearly an increasingly larger difference between the measured and calculated results for the stopping of the noble gases as Z gets larger. Two points could be noted in this regard. The stopping power for the solid targets was inferred from range distributions under the assumption of velocity-proportional stopping. In contrast to what was concluded above that the stopping power of gaseous targets for nitrogen does not exhibit velocity-proportional behavior, there is some evidence for velocity-proportional stopping for nitrogen in solid targets [15]. This assumption, however, is not necessarily exact and any deviation could account for part of this discrepancy.

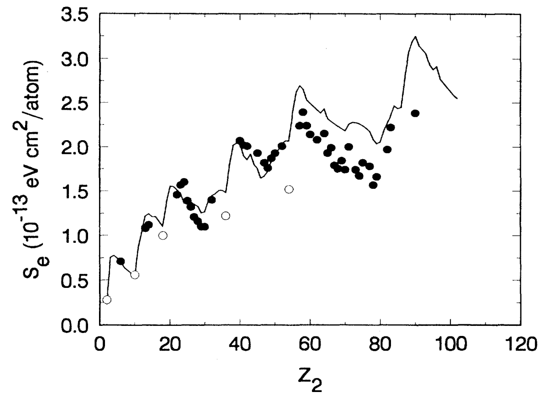

FIG. 2. Stopping power vs target atomic number for nitrogen ions. The solid curve is that obtained by FLB [2] at $v/v_0=1$. The filled circles represent stopping powers inferred from the range distributions of ^{14}N and ^{15}N implanted in solid targets [1]. The open circles represent the data points for the noble gases obtained from the linear regressions of the data from this study at $v/v_0=1$.

More interestingly, there is a physical consideration. The theoretical curve in the figure (FLB) was determined from comparisons with data for elemental metallic targets having Z between 6 and 52 and using a *single* adjustable parameter. This parameter is a distance of closest approach in the collision between projectile and target atom. It represents an effective atomic radius within which the electrons of the target atom do not contribute to the stopping through excitation or ionization because of their adiabatic response to the projectile. The difference in the measured and calculated stopping could arise from the fact that the adjustable parameter was determined by fitting the theoretical curve to the measured stopping for elemental metallic targets for which the binding of those electrons that contribute to the stopping is less than the binding of the corresponding electrons of the heavier noble gases.

A second point concerning Fig. 2, not directly related to the present work, is that the data for Z above 55 fall

TABLE II. Comparison of the values of stopping power at $v=v_0$ obtained from the linear regression of the data in the range $v_0 < v < 2.6v_0$ with the FLB values. Value of stopping power are given in units of 10^{-13} ev cm^2/atom.

Element	Expt.	FLB	Difference (%)
He	0.28	0.23	+17
Ne	0.56	0.53	+6
Ar	1.00	1.10	−11
Kr	1.22	1.48	−21
Xe	1.52	2.07	−36

below the theoretical curve. This discrepancy could be attributed to the fact that the atoms in this portion of the periodic table are larger than atoms in the region where the adjustable parameter was determined. Thus, a larger value, which would exclude an additional contribution to the stopping from inner electrons, might be needed in this region.

It would be of interest to have confirmation measurements as a function of projectile energy for solid, elemental targets using the direct time-of-flight technique similar to those reported here for gaseous targets. Such measurements are currently underway.

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