Stopping-power distribution for fast helium and nitrogen ions passing through metal films

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Measurements have been carried out of the stopping power distribution for helium and nitrogen ions with initial energies $E_0 \sim 330$ keV/nucleon after passing through aluminum, copper, silver, and gold films of different thickness. The maximum target thickness corresponded to $\sim 80\%$ of E_0. The widht η of the energy distributions after the target, plotted as a function of the relative energy loss $\Delta E/E_0$, has a flat maximum near $\Delta E/E_0 \sim 0.4$–0.6. Calculations based on existing theories are only in qualitative agreement with these measurements: The calculated maximum values of η are lower by a factor of 1.5–2 than the measured widths and correspond to somewhat greater values of $\Delta E/E_0$ (~ 0.6–0.8).

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1. INTRODUCTION

The energy distribution of charged particles transmitted by a layer of material is largely determined by two mechanisms. One is the statistical mechanism that is due to fluctuations in the energy losses experienced by the particles while they interact with the atoms of the medium,[1-3] whereas the second, nonstatistical, mechanism is associated with the difference between the specific stopping power dE/dt for particles with different velocities.[4] The relative importance of these two mechanisms depends on the magnitude of the mean energy loss $\overline{\Delta E}$, the thickness t of the material, the particle energy E, and the width η of their energy distribution.

The high-energy region, where the decelerating particles do not carry electrons, and the effect of electron bonds in the medium of fluctuations in the energy losses is unimportant, has by now been investigated in adequate detail. Some theories describe the spread in the energy losses in terms of a single statistical mechanism,[2,3] whilst other theories allow for the nonstatistical process.[4-7] For those values of t and $\overline{\Delta E}$ which correspond to the range of validity of each of these mechanisms, the theories are in reasonable agreement with experimental results for protons and α particles at energies between 1 and 20 MeV/nucleon.[8-11]

The low-energy region, where the slowing-down process is appreciably affected by fluctuations in the particle charge and by the ionization energies of the absorber atoms, has been investigated to a much lesser degree. According to existing theoretical ideas, the spread in the stopping power of thin targets, plotted as a function of particle energy, is a curve with a flat maximum in the region where resonance processes play an important role.[12,13] At lower energies, fluctuations in the stopping power should decrease owing to the reduction in the number of electrons effectively participating in the slowing-down process.[2,14] Experimental data obtained for ~1 MeV protons in solid and gaseous targets[9,15] are only in qualitative agreement with these theories, which means that further experimental studies in this energy band are essential.

In this paper, we report measurements of the energy spectra of $^4He_2$ and $^{14}N_7$ ions with initial energies of ~0.33 MeV/nucleon after passing through aluminum, copper, silver, and gold films of different thickness for relative energy losses $\overline{\Delta E}/E_0$ between ~0.05 and ~0.8. The root mean square charge of the helium ions transmitted by the target was found to vary between ~1.9 and ~1.0, whereas the corresponding numbers for the nitrogen ions were between ~4.5 and ~2.3. The experimental results are compared with calculations based on the Simon theory[4,5] which takes into account the velocity dependence of stopping-power fluctuations put forward by Lindhard and Scharff.[14]

2. EXPERIMENTAL METHOD

The measurements were carried out on a 72-cm cyclotron producing ions with energies up to ~0.33 MeV/nucleon. The ion beam was passed through an analyzing magnet and, after collimation by a set of slits, was intercepted by the target. The target holder had three apertures arranged on a vertical line and covered uniformly by the ion beam. The central aperture had a diameter of ~1 mm and was covered by the metal film under investigation, and the two other apertures were kept open and were used to transmit primary atoms directly to the detector.

The ion detector was a surface barrier silicon counter with a working diameter of 5.5 mm. This counter could be moved along the beam axis and was mounted at different distances from the target. Pulses from the detector were received by standard amplifiers and were examined by a 256-channel pulse-height analyzer. The overall resolution of the recording system for helium and nitrogen ions with energies in the range 0.1–0.3 MeV/nucleon was ~30 and ~70 keV, respectively. The spectrometer system was calibrated with the aid of the same helium and nitrogen ions transmitted through a variable-thickness aluminum foil placed in front of the entrance slit of the analyzing magnet. The latter thus received an ion beam with a broad energy distribution, and was used to select ions of different energy and direct them onto the detector. The value of the energy was determined from the strength of the deflecting magnetic field to within ~1%. The measured relationship between particle energy and channel number at the peak of the energy distribution was found to be linear throughout the energy range under investigation to within statistical error (1–2%). The absolute calibration of the analyzing magnet was carried out with a standard α-particle source (Ra^{226}) which was also

used for the additional control of stability and linearity of the detection system.

The targets were three films obtained by vacuum evaporation of pure metals onto the cleavage planes of NaCl crystals. The crystals were subsequently dissolved in distilled water and the film thickness t was determined by measuring the mean helium-ion energy before and after the film, and using the known energy dependence of the specific stopping power $-dE/dt$ for these ions in each particular material.[16,17] The thickness t was determined to within 1—1.5% (this ignores possible systematic errors due to undertainties in the measured values of $-dE/dt$; these errors amounted to 3—4% in[16,17]).

The energy spectra were determined 2—4 times for each film with the detector at 60, 80, and 100 mm from the target, which corresponded to the detection of particles scattered through angles less than 2.4, 1.9, and 1.5°. The position of the maximum on the spectrum of scattered ions and the distribution width were independent of the angle subtended by the detector (to within experimental error), which indicated that elastic nuclear scattering did not contribute appreciably to the slowing down of the recorded particles. All the energy distributions were practically symmetric about the maximum and, for the most part, could be described by the Gaussian distribution. For comparison with the theoretical calculations, we determined the full width at half-height (η) of the energy distributions. A correction was introduced for the finite resolving power of the detection system and for the energy spread in the primary beam, using the formula

$$\eta=(\eta_t^2-\eta_0^2)^{1/2}, \quad (1)$$

where η_t and η_0 are the experimental widths of the energy spectra of particles transmitted through the target and the primary particles, respectively. (The width η_0 is almost entirely determined by the resolving power of the recording equipment.) The values of η obtained in this way as functions of target thickness were found to vary between ~4 and 15—20%.

3. RESULTS AND ANALYSIS

The measurements were performed for aluminum, copper, silver, and gold films of thickness respectively equal to 40—720, 185—1320, 105—1100, and 105—470 $\mu g/cm^2$. The energy spectra of helium ions were obtained for 27 aluminum, 22 copper, 18 silver, and 12 gold films. The initial energy of the ions was 1200—1350 keV. For the nitrogen ions, the measurements were performed for 20 aluminum, 15 copper, 15 silver, and 4 gold films, using initial ion energies of 4550—4700 keV. Examples of ion spectra after targets of different thickness are shown in Fig. 1. Figures 2 and 3 show measured half-widths of the energy distributions as functions of the relative energy loss $\Delta E/E_0$.

Our results indicate that, as the target thickness t increases, there is at first a rapid increase in the width η of the energy spectrum of ions transmitted by the target, but this increase gradually slows down. When the energy loss reaches $\Delta E \sim 0.4E_0 - 0.6E_0$, the width reaches its maximum value and then begins to fall.

In general terms, this shape of the function $\eta = f(\Delta E/E_0)$ can be explained by the competition between the two mechanisms responsible for the energy spread. At first, when the ion velocities are roughly the same, only the statistical mechanism is effective and ensures that the spread in ion energies increases with increasing target thickness t and decreasing mean energy E. As the width of the energy spectrum increases, there is an attendant increase in the role of the nonstatistical mechanism which acts in the opposite direction. Under our conditions, the reduction in the energy of the nitrogen ions is at first accompanied by a reduction in dE/dt (for these ions, the specific stopping power reaches a maximum for $E > 5$ MeV). For helium

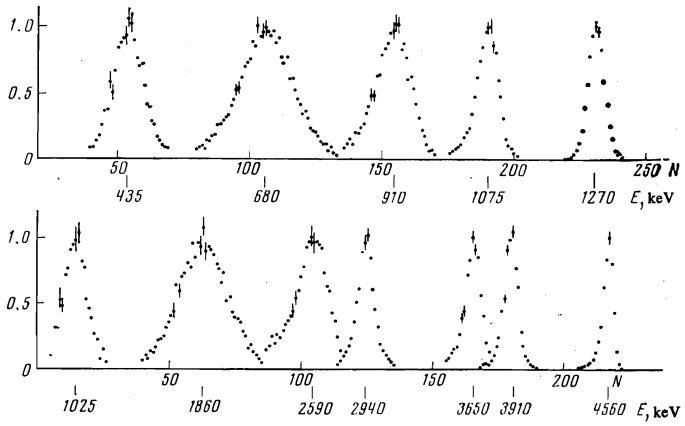

FIG. 1. Energy spectra of helium ions (top) and nitrogen ions (bottom) transmitted by a copper film of thickness t: extreme left—t = 0 (no target), extreme right—t = 1320 $\mu g/cm^2$. The ordinate axis gives the number of counts in the analyzer channel, normalized to the peak of the spectrum (N is the channel number).

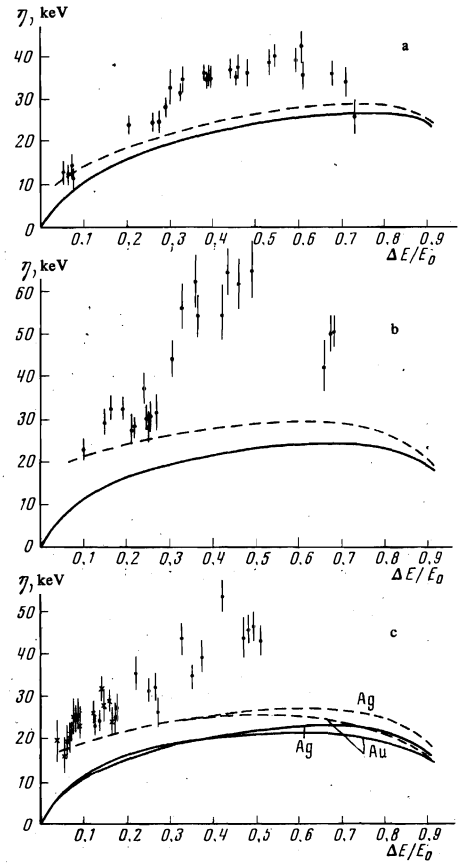

FIG. 2. Helium ions: a—Al, b—Cu, c—Ag (points) and Au (crosses). The theoretical curves were calculated for E_0 = 1300 keV.

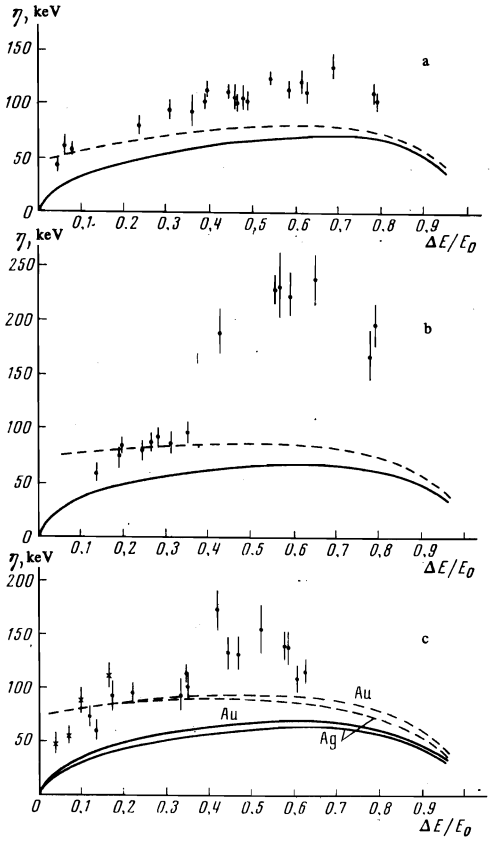

FIG. 3. Nitrogen ions: a—Al, b—Cu, c—Ag (points) and Au (crosses). The theoretical curves were calculated for $E_0 = 4600$ keV.

ions, the quantity dE/dt is a maximum for $E \lesssim 1$ MeV, so that, during the initial stages (up to $\Delta E/E_0 \sim 0.3$), it varies little but then falls appreciably with decreasing E. On the average, the slower ions lose less energy than the fast, and the result is that the width of the energy spectrum is reduced. As the ions continue to be slowed down, this reduction compensates and then exceeds the broadening of the spectrum due to the statistical mechanism.

A form of the function $\eta(\Delta E)$ similar to that shown in Figs. 2 and 3 was also obtained for α particles with initial energies $E_0 \gtrsim 5$ MeV in gases[18] and solids[19]. However, in these cases, the maximum was observed at larger values of $\Delta E/E_0$ (~0.8—0.9) and was better defined. This difference may be due to the fact that the initial energy E_0 was much greater than the value of 1 MeV at which the stopping power reached a maximum.

We used Simon's theory[4] which was given in detail by Payne[5] to analyze our results. This theory can be used to calculate the variance of the energy distribution of charged particles losing energy as a result of inelastic collisions with electrons in the absorber, subject to the condition that the mean energy loss is comparable with the initial energy. According to Simon,[4] the variance for a particle beam transmitted by an absorbing layer in which the mean energy is reduced from E_0 to E is given by

$$\Omega^2 = \left[\frac{M(E)}{M(E_0)}\right]^2 \left\{\Omega_0^2 + 2[M(E_0)]^2 \int_E^{E_0} \frac{N(T)}{[M(T)]^3} dT\right\}. \quad (2)$$

where Ω_0^2 is the initial variance, $M(E) = -dE/dt$ is the specific stopping power of the absorber, and N(E) $= \frac{1}{2} d\Omega^2/dt$ is the rate of increase of the variance due to the statistical mechanism.

Lindhard and Scharff[14] have shown that, for charged particles whose velocity v satisfies the condition $v^2/v_0^2 Z_2 \leq 3$, where $v_0 = 2.19 \times 10^8$ cm/sec is the Bohr velocity and Z_2 is the charge on the absorber atoms, the increase in the variance is related to the energy loss by the simple formula

$$d\Omega^2 = -(m_e/m_i) E dE, \quad (3)$$

where m_e and m_i are the masses of electrons and decelerated particles, respectively. Substituting (3) in (2), we obtain

$$\Omega^2 = \left[\frac{M(E)}{M(E_0)}\right]^2 \left\{\Omega_0^2 + \frac{m_e}{m_i}[M(E_0)]^2 \int_E^{E_0} \frac{T\,dT}{[M(T)]^3}\right\}. \quad (4)$$

Using the empirical function for M(E) in (4), we can calculate the values of η. The results of this calculation are shown in Figs. 2 and 3 by the solid curves. The experimental M(E) was taken from the papers by Chu and Powers[16] and Porat and Ramavataram.[17] There are no data for nitrogen ions in copper. In this case, we used the values of M(E) for silver, increased by 10%, We found experimentally that this was the ratio of losses for ~4 MeV nitrogen ions. Since the experimental spectra were nearly Gaussian, it was assumed that $\eta = 2(\ln 2)^{1/2}\Omega \approx 2.36\Omega$.

Calculations based on (4) can be simplified by replacing M(E) with a suitable approximate analytic expression.[1)] In many cases, the following expression yields satisfactory results:

$$M(E) = 2M_m (E_m E)^{1/2}/(E_m + E), \quad (5)$$

where E_m is the energy at which M(E) reaches its maximum value M_m. Substituting this in (4), and assuming that $\Omega_0^2 = 0$, we obtain the following simple expression:

$$\Omega^2 = \frac{m_e}{3m_i} E(E_m + E)\left[\left(\frac{E_m + E_0}{E_m + E}\right)^3 - 1\right]. \quad (6)$$

In particular, for helium ions in aluminum ($E_m = 500$ keV, $M_m = 1.23$ keV/μg.cm^{-2}), the expression given by (5) reproduces the experimental points reported by Chu and Powers[16] to within the experimental spread (±2%) for energies between 300 and 1900 keV, and calculations based on (4) and (6) are virtually identical.

Calculations based on (6) show that, as the energy increases, the maximum on the $\eta = f(\Delta E/E_0)$ curve shifts toward higher values of the relative energy loss. This explains the difference in the position of the maximum for low-energy helium ions (present experiment) and the high-energy ions (experiment reported in[19]).

Even when the theory does provide a correct description of the $\eta(\Delta E/E_0)$ curve, the absolute values of the energy spread found experimentally are systematically higher than those predicted by the theory. A possible reason for this may be that the target is not of uniform thickness. This is indicated by a comparison between the spread in the energy loss in gases[18] and metals.[19] In the latter case, the spread is substantially greater although the theory predicts[14] that, at low ion energies, the quantity η should not be very dependent on target material.

To estimate the effect of target nonuniformly, some of our targets were examined in a scanning electron microscope with a magnification of 10^4. Analysis of

target surfaces enabled us to conclude that the mean deviation of the surface profile from the perfect plane was 100—300 Å for target thickness between 500 and 30 000 Å. Allowance for target nonuniformity led to better agreement between calculated and experimental widths of the energy distribution (broken curves in Figs. 2 and 3), but a discrepancy has remained, especially for those values of $\Delta E/E_0$ for which the effect of the nonuniformity should have been less pronounced. It is possible that the origin of this discrepancy lies in fluctuations in the effective charge of the decelerating particles, since this charge determines the magnitude of the specific stopping power. This shows that the theory of the spread in stopping power at low energies, where it is governed by the statistical mechanism, is in need of improvement.

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[1)]Calculations of this kind can, for example, be based on the semiempirical formula reported by Brice. [20] The parameters of this formula have been determined by Lin et al. [21] for a number of materials. We note that this formula is substantially simplified and becomes suitable for analytical calculations when the universal function $f(\epsilon)$ with $\epsilon \approx v/2v_0$ is approximated by $f(\epsilon) \approx 5\pi(\epsilon + 0.11)$, which, for $\epsilon > 1$, does not differ from $f(\epsilon)$ by more than 0.5%.

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