Geometrical effect on the measurement of stopping powers: Angle-dependent energy loss of 7-MeV protons in Be, Al, Cu, Ag, and Ta

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The geometrical effect on the measurement of stopping powers for 7-MeV protons, which was a hitherto-not-noticed effect until our previous experiment, has been studied more systematically with 7-MeV protons by changing the target thickness for Be, Al, Cu, Ag, and Ta. An improved experimental arrangement has been used to prevent the edge-scattered protons from affecting the energy-loss measurement. For all targets, the observed energy losses have been confirmed to increase with increasing emergence angle. It has also been found that this effect has a strong dependence on the thickness of the target. The dependence of this effect on the target atomic number cannot be judged properly from the present experiment. Angular distributions of protons due to multiple scattering agree fairly well with the predictions of Molière's theory. The comparison between our geometrical effect and the angular dependence of the energy loss observed for low-energy heavy ions has been discussed.

I. INTRODUCTION

When well-collimated protons of several MeV energy pass through a thin solid target of random matter, the protons lose their energy according to the thickness of the target, and the direction of the protons diverges due to multiple scattering with atomic nuclei in the target. In the case of several-MeV protons, it had been firmly believed[1-5] that all protons lose essentially the same energy in the target independent of the emergence angle. In a previous experiment,[6] it was found that if we collimate the proton beam extremely sharply and make the angular resolution of the detector extremely high, we can observe that the energy loss of protons in the solid target increases by several percent as the emergence angle increases from 0° to 1.67°.

It has been noted that this increase is not due to (1) the effective increase of the target thickness by oblique emergence, i.e., $1/\cos\theta$, where θ is the emergence angle; (2) the increase of the actual path length of protons due to multiple scattering; or (3) the increase of the energy loss due to energy transfer to recoil nuclei. It has also been confirmed by x-ray-diffraction examination that the increase of the energy loss is not due to target texture. It was concluded that the observed increase of the energy loss with increasing emergence angle is due to a hitherto-not-noticed effect.

It was supposed that this effect is very likely the dependence of the energy loss on the average impact parameter of individual collisions which protons make with atomic nuclei within the solid target. We have named this effect the "geometrical effect."

In the case of heavy ions at low energies, it has also been known[7-10] that the energy loss depends on the emergence angle when the ions pass through a solid target. However, this fact is not surprising because in the case of low-energy heavy ions, nuclear stopping (elastic energy loss) plays an important role during the penetration through the target. The comparison between our geometrical effect and the phenomenon observed for low-energy heavy ions will be discussed later.

In this experiment, in order to investigate the nature of the geometrical effect more systematically, the increase of the energy loss with increasing emergence angle has been measured with an improved experimental arrangement and by changing the target thickness for Be, Al, Cu, Ag, and Ta. A part of this data has been used for the computer simulation[11] of this effect.

II. EXPERIMENTAL PROCEDURE

The schematic diagram of the experimental setup is shown in Fig. 1. The proton beam of 7 MeV from the cy-

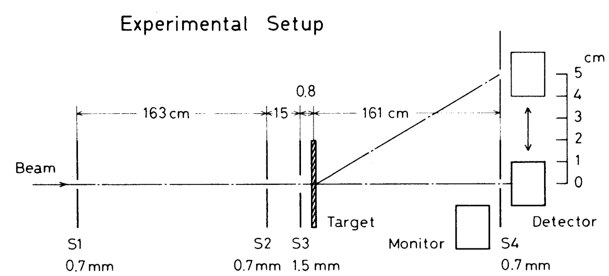

FIG. 1. Schematic diagram of experimental setup. The detector system is movable in the horizontal plane perpendicular to the direction of the incident protons in a range of 5 cm. The diameters of diaphragms $S1$, $S2$, and $S4$ are 0.7 mm and that of the baffle $S3$ is 1.5 mm.

clotron of Kyoto University was collimated by a double diaphragm system of $S1$ and $S2$; the diameter of each was 0.7 mm and they were 163 cm apart. The divergence of the incident beam was less than 0.05° before hitting the thin target. In order to prevent the edge-scattered protons that originated at diaphragms $S1$ and $S2$ from affecting the energy-loss measurement, a baffle $S3$ of 1.5 mm diameter was placed 15 cm behind diaphragm $S2$. The target was placed 8 mm behind the baffle. The detector system consisted of a diaphragm $S4$ of 0.7 mm diameter and a surface-barrier silicon detector, which was placed 161 cm behind the target. The detector subtended a solid angle of 1.5×10^{-7} sr as seen from the target. The detector system was movable perpendicular to the direction of the incident beam in a range of 5 cm. At the displacement of 5 cm the emergence angle was 1.78°. In this arrangement the energy-loss measurement was quite free from the edge-scattered protons at 1 cm displacement.

The pulses from the detector were amplified with a low-noise amplifier and fed into a 4096-channel pulse-height analyzer. Another silicon detector was used to monitor the angular distribution due to multiple scattering. The energy of the incident proton was measured by the analyzing magnet with an accuracy of $\pm0.01\%$.

In order to monitor the gain of the detector-amplifier system, the pulses of protons which were scattered by a thin Au foil of 180 $\mu g/cm^2$ were measured. Figure 2 shows the device for mounting the target and the thin Au foil. The part labeled A is essentially an ammeter. When ac power is supplied, the hand labeled B makes a metronomic motion. Double frames labeled C are fixed to hand B. To the left frame the sample target is fixed and to the right frame the thin Au foil is fixed. When ac power is supplied and hand B makes a metronomic motion, the incident beam traverses the target and the thin Au foil alternately. This procedure minimizes the effect of possible nonuniformity of the target thickness. The pulses of protons which pass through the target and those which are scattered by the thin Au foil hit the detector alternately.

Thus the two pulse heights were recorded on the 4096-channel pulse-height analyzer simultaneously in one exposure. Because the energy of the incident protons was very well stabilized, the energy of the protons scattered by the thin Au foil was also very stable. Thus we could monitor the gain of the detector-amplifier system. The stability of the detector-amplifier system was also cross-checked by a very-high-precision pulse generator (Ortec 448).

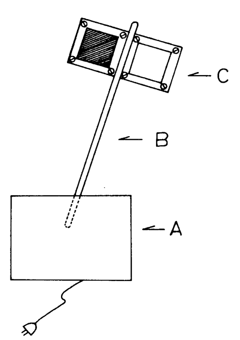

FIG. 2. Schematic diagram of the device to mount the target and the thin gold foil. A is essentially an ammeter. The double frames labeled C are fixed to the hand labeled B. The target and the thin Au foil were fixed to each frame. When ac power is supplied, hand B makes a metronomic motion. Then, the proton beam traverses the target and the thin Au foil alternately.

In principle, in our method the energy loss should be determined by measuring the difference between the pulse height of protons that passed through the target and the pulse height of the incident protons. The pulse height of the incident protons has been determined from the pulse height of the protons scattered by the Au foil. The energy loss of 7-MeV protons in the Au foil was estimated to be 4 keV by using our previous stopping-power data.[12] The decrease of the energy due to elastic scattering was negligible. The geometrical effect of energy loss in the thin Au foil is quite negligible because the energy loss in the Au foil is very small as compared with the energy loss in the sample target. The energy calibration of the pulse-height spectrum was performed with the high-precision pulse generator. The ionization defect of the surface-barrier silicon detector has been investigated in the previous experiment[12] and has turned out to be substantially zero for 7-MeV protons.

The above-mentioned metronomic method could not be applied in the case of zero emergence angle because the counting rate of protons that passed through the thin Au foil was too high. Therefore, when the measurement at zero emergence angle was made, the sample foil was stopped. The pulse height of the incident protons was determined by measuring the protons scattered from the Au foil at 1 cm displacement in a separate measurement. Because there would be nonuniformity of the target thickness, the measurement was made at the same stopped position of the target for 1 cm displacement of the detector. Then the metronomic measurements were performed for 1—5-cm displacements. The value of the energy loss for zero emergence angle was normalized by the measurement made at 1 cm displacement.

All targets were commercially obtained. The thicknesses of the targets were determined by measuring the weight and the area. The thicknesses of the targets were 2.198, 4.244, and 6.441 mg/cm^2 for Be; 2.502, 4.143, and 5.427 mg/cm^2 for Al; 3.743, 7.576, and 11.311 mg/cm^2 for Cu; 4.333, 8.712, and 13.027 mg/cm^2 for Ag; and 7.291 and 10.444 mg/cm^2 for Ta. The measurements of the energy loss and the angular distribution due to multiple scattering have been repeated four times for each target at seven emergence angles between 0° and 1.78°.

III. RESULTS

The energy loss as a function of emergence angle and the angular distribution due to multiple scattering are shown in Fig. 3. The upper half of each figure shows the angular distribution of protons and the lower half shows the relative increase of the energy loss as a function of emergence angle. The theoretical prediction of the angular distribution is calculated using Molière's theory[13] and is also shown in Fig. 3.

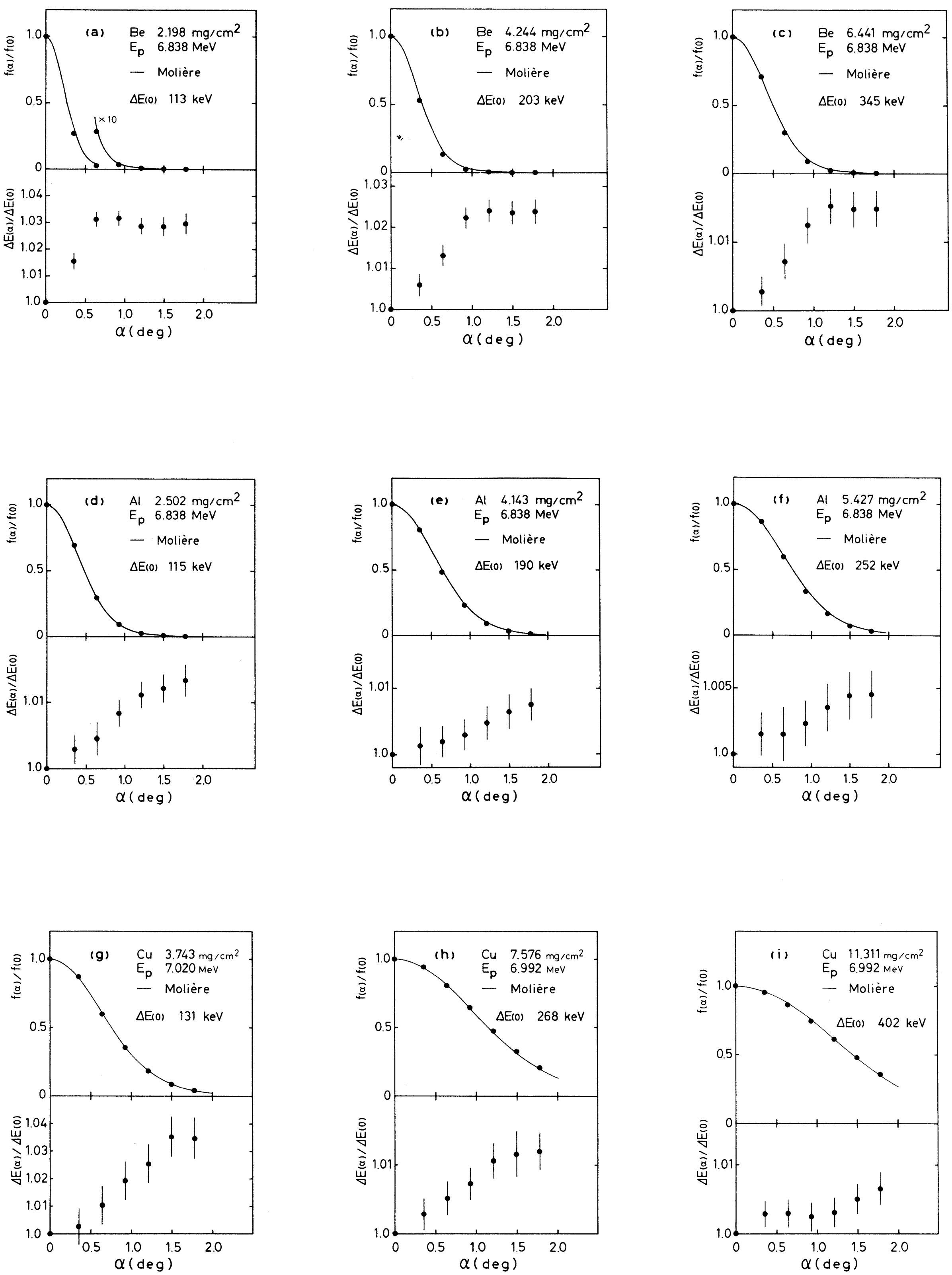

FIG. 3. Angular distribution due to multiple scattering and energy loss as a function of emergence angle, (a)—(c) for Be; (d)—(f) for Al; (g)—(i) for Cu; (j)—(l) for Ag; and (m) and (n) for Ta. Solid circles indicate the experimental results. Solid curves are the predictions of Molière's theory.

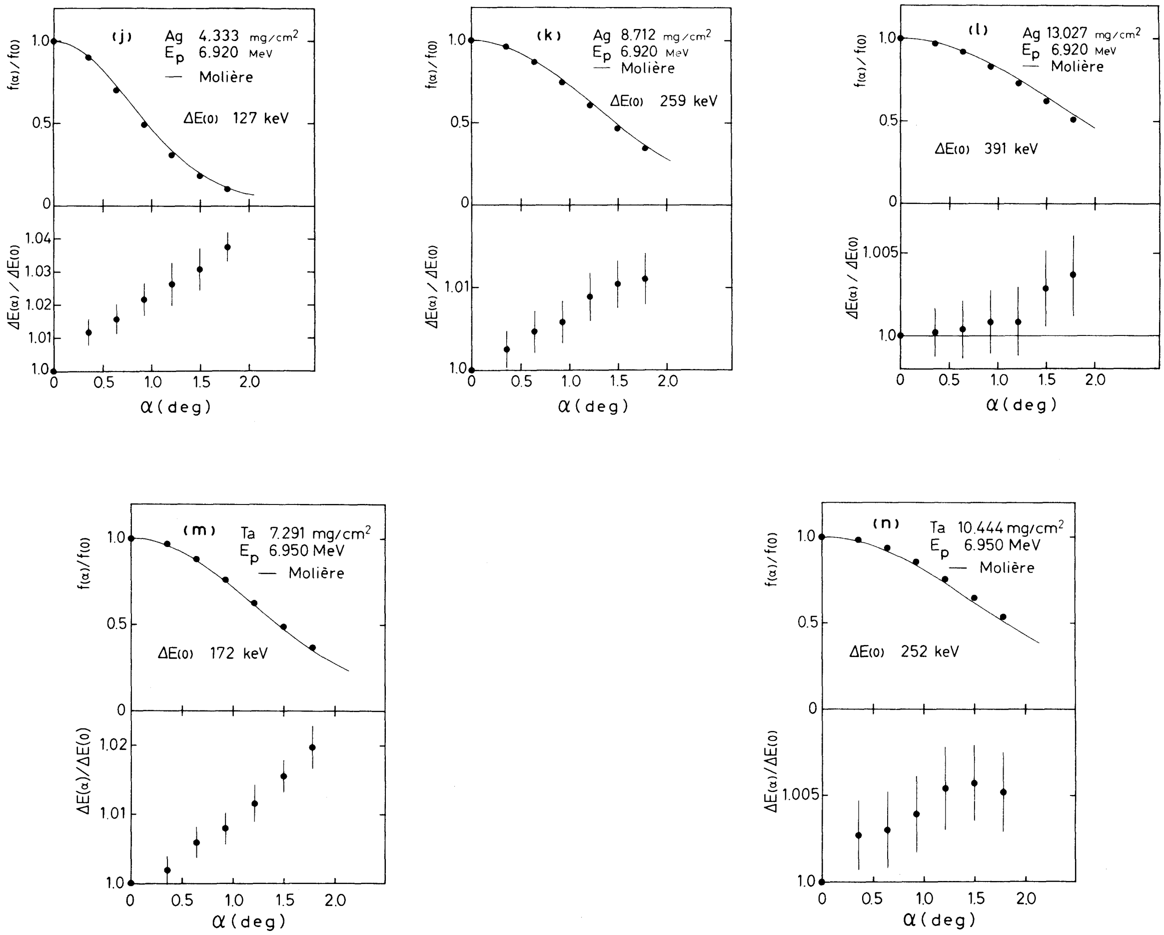

FIG. 3. (*Continued*).

As already seen in a previous experiment,[6] the agreement of the observed angular distribution with Molière's theory is fairly good. The energy loss is found to increase for all targets with increasing emergence angle also in the present experiment. As clearly seen from the figures, the increase of the energy loss shows a strong dependence on the target thickness. The increase is larger for thinner targets. It appears that there is a trend toward saturation of the increase of the energy loss as the emergence angle increases. The data for Cu of 3.743 and 7.576 mg/cm^2 have already been published with the computer simulation of this effect.[11]

IV. DISCUSSION

The increase of the energy loss with increasing emergence angle has been confirmed again in the present experiment, in which measurements of energy losses were quite free from the edge-scattered protons. The increases of the energy losses for Be, Al, and Ag are slightly smaller than those observed in a previous experiment.[6] This might be due to the elimination of the edge-scattered protons by the baffle. As already discussed in detail in a previous paper,[6] the increase of the energy loss cannot be explained by the known effects.

The most noticeable feature of this effect found in the present experiment is the strong dependence of the effect on the target thickness. The increase of the energy loss with increasing emergence angle becomes strikingly large as the thickness of the target decreases. This remarkable feature of the effect can be seen for all target elements investigated.

Our computer simulations,[11] in which we assumed the energy loss of one proton by a collision with one atom depends on the impact parameter, could reproduce fairly well the observed increase of the energy loss with increasing emergence angle, the dependence of the effect on the target thickness, and the angular distribution. It has also been shown by the computer simulation that the average value of the individual scattering angles of the protons with atomic nucleus increases with the increase of the emergence angle and the average value of individual impact parameters decreases with increasing emergence angle.

Thus, it has become certain that the essence of this effect is the dependence of the energy loss on the average value of the individual impact parameters of protons with atomic nuclei. It is supposed that this effect will depend upon the target atomic number. However, because this effect shows strong dependence on the target thickness, at the present stage it is difficult to compare the strength of the effect for different targets quantitatively.

If we tentatively compare the saturation value of the increase of the energy loss when the energy loss is 122 ± 9 eV, it appears that the effect becomes large as the target atomic number increases from Al through Ag. In the case of Ta, the energy loss is 172 keV and the increase does not reach the saturation. It cannot be inferred to what extent the increase of the energy loss approaches. It should be noted that in the case of Be the effect is much larger than in the case of Al. This feature has been observed also in a previous experiment.[6] Therefore the effect for Be does not obey the trend observed from Al to Ag.

As already mentioned in previous papers,[6,11] if we determine the stopping power by measuring the protons that emerge from the target at zero angle, the resultant stopping power will be smaller than the stopping power of usual definition. In usual stopping-power measurement, energy losses of all protons that pass through the target are measured. For example, the deviation is about 1.5% in the case of Cu of 3.743 mg/cm^2. Therefore, in the case when stopping power is measured by the time-of-flight method, the resultant stopping power will be systematically small. In order to decrease this deviation, the sample target should be as thick as possible to the extent that average energy of protons in the target has good physical meaning.

Here we shall discuss the comparison between our geometrical effect and the dependence of the energy loss on the emergence angle which is observed for heavy ions at low energies.[7–10] As is well known, in the case of heavy ions at low energies nuclear stopping plays an important role besides electronic stopping.[14] At first it had been believed[7–10,15] that dependence of the energy loss on the emergence angle is entirely due to nuclear stopping, and electronic stopping is essentially independent of scattering angle.

Beauchemin and Drouin[10] made detailed measurements on dependence of the energy loss on the emergence angle in the case of neon on carbon and argon on carbon. They have analyzed their data from the standpoint that dependence of the energy loss on the emergence angle is entirely due to nuclear stopping.

On the other hand, Meyer, Klein, and Wedell[16] (MKW theory) assumed that electronic stopping also depends on the scattering angle and developed a phenomenological theory on the energy-angle distribution of heavy ions at low energies. They put the electronic energy loss (electronic stopping) $q_e(\chi)$ in a single collision with a solid target atom,

$$q_e(\chi)=C_{e0}+C_{e2}\eta^2 , \tag{1}$$

where χ is the scattering angle in the laboratory system, η is the reduced scattering angle proportional to χ, C_{e0} is the angle-independent part of electronic stopping, and C_{e2} is the angle-dependent part of electronic stopping.

The nuclear energy loss (nuclear stopping) in a single collision was

$$q_n(\chi)=C_n\eta^2 , \tag{2}$$

where C_n was calculated analytically. Thus the total energy loss in a single collision was taken as

$$Q(\chi)=q_e(\chi)+q_n(\chi)=C_{e0}+(C_{e2}+C_n)\eta^2 , \tag{3}$$

where C_{e0} and C_{e2} were treated as adjustable parameters which should be determined by comparison with experiment. Thus the MKW theory does not start from first principles.

Beauchemin and Drouin[17] have analyzed their experimental results with the MKW theory. In their conclusion (c), they state that the basic hypothesis of the MKW theory seems to be verified. Actually, however, contrary to their above statement, their analyses of their data lead to very irrational results. That is, in the case of neon (40–120 keV) on carbon, the angle-dependent part of electronic stopping, C_{e2}, has turned out to be the same order of magnitude as C_n around 100 keV and increases with decreasing energy; while in the case of argon (40–240 keV) on carbon, C_{e2} is of the same order of magnitude as C_n around 200 keV, and decreases with decreasing energy and becomes negligibly small as compared with C_n around 50 keV. The behavior of C_{e2} is quite different for neon and argon. This means that their data are not self-consistent or the MKW theory is erroneous. Therefore, considered impartially, the experimental data of Beauchemin and Drouin[10,17] do not corroborate the existence of the angle-dependent part of electronic stopping, C_{e2}.

Recently, Ellmer and Wedell[18] have modified the MKW theory. They have pointed out that its basic assumption, i.e., Eq. (1), is inadequate and does not agree with the angular dependence of q_e derived from the Firsov theory.[19] They made numerical calculations of q_e by the Firsov theory and q_n for a special case and compared them with each other. They concluded that in the scattering angle range of interest C_{e2} is negligibly small as compared with C_n. They have developed their modified theory of energy-angle distribution by neglecting C_{e2} as compared with C_n. That is, they assumed that the dependence of the energy loss on the emergence angle is entirely due to nuclear stopping. The distributions of the total energy loss calculated by their modified theory show essentially better agreement with experimental results than the MKW theory.

It is not possible to separate electronic stopping from nuclear stopping by experiment; the help of theory is necessary to separate them. The recent theory of Ellmer and Wedell[18] for heavy ions at low energies neglects C_{e2} as compared with C_n.

Until now, to the best of our knowledge there exists no evidence that the angle-dependent part of electronic stopping, C_{e2}, can be extracted from the measurements for low-energy heavy ions passing through a solid target. A small fraction of the angle dependence due to electronic stopping might exist conceptually, but even if it really exists the effect will be covered by the overwhelming fraction of the angle dependence due to nuclear stopping and will not be separated from nuclear stopping. Consequently, it is natural to consider that dependence of the energy loss on the emergence angle for heavy ions at low energies is substantially due to nuclear stopping only. Actually, in the case of heavy ions at low energies, for instance, 80-keV neon on carbon, the interval of the emergence angle is from 0° to 30°, the angular resolution of the measurement is $\sim 1°$, and the increase of the energy loss amounts to 70%. Also, the increase of the energy loss does not saturate.

In this experiment we have detected the increase of the energy loss in the small interval of the emergence angle from 0° to 1.78°. The angular resolution of the measurements is smaller than 0.025° and the increase of the energy loss is at most $\sim 4\%$. The angular interval, the angular resolution of the measurement, and the magnitude of the increase of the energy loss are quite small as compared with the case of heavy ions at low energies. Furthermore, the increase of the energy loss saturates in our case. In our geometrical effect, as discussed in detail in previous papers,[6,11] nuclear stopping is completely negligible. Therefore, our geometrical effect is without doubt a manifestation of angle dependence of electronic stopping, i.e., impact parameter dependence of electronic stopping.

In the case of low-energy protons ($50 \sim 400$ keV), where electronic stopping prevails, to our knowledge there are two recent experiments[20,21] which have observed dependence of the energy loss on the emergence angle after passing through a solid target. Iferov and Zhukova[20] have measured the energy losses of 100-, 200-, and 400-keV protons in gold foils. They have observed that the energy loss increases as the emergence angle increases from 0° to 12°. They have analyzed their data by assuming that their targets are sufficiently thin so that a proton emerging at an angle θ has made a single collision with an atom in the target, and treated the energy loss as follows:

$$Q_e(\theta)=\Delta E(\theta)-\Delta E(0)-\Delta E_n(\theta)-\frac{\theta^2}{4}\Delta E \; , \tag{4}$$

where $\Delta E(\theta)$ and $\Delta E(0)$ are the energy losses at the angles θ and 0°, $\Delta E_n(\theta)$ is the nuclear stopping at θ, $(\theta^2/4)\times\Delta E$ is the correction to the enlargement of path length, and $Q_e(\theta)$ is the electronic energy loss of the proton corresponding to a scattering angle θ for a single collision in the target.

Jakas *et al.*[22] have pointed out that the thicknesses of the targets of Iferov and Zhukova are not sufficiently thin and that their procedure of analyzing the data with Eq. (4) is erroneous. On the other hand, by using transport theory, Jakas *et al.*[23] have derived a general expression which connects $\Delta E(\theta)$ and $Q(\phi)$ and shown that $\Delta E(\theta)$ is not related straightforwardly with $Q(\phi)$, where ϕ is the scattering angle in a single collision and $Q(\phi)$ is the sum of electronic and nuclear stopping in a single collision:

$$Q(\phi)=Q_e(\phi)+Q_n(\phi) \; . \tag{5}$$

They have made measurements[21] of the dependence of the energy loss on the emergence angle for 50-, 100-, and 200-keV protons and 200-keV He ions on C and Al in the angle interval from 0° to 2°. They have observed that the increase of the energy loss as a function of θ increases remarkably as the proton energy increases for the same target.

A most remarkable dependence of the emergence angle dependence of the energy loss on the proton energy has been observed in the case of protons on Al. The saturation values of the increase of the energy loss, $[\Delta E(\theta)-\Delta E(0)]/\Delta E(0)$, are 1.7% for 50 keV, 8.2% for 100 keV, and 15% for 200 keV.

Because at the proton energies of Jakas *et al.* there is no theory to express $Q_e(\phi)$, they have tried to obtain $Q_e(\phi)$ from the observed $\Delta E(\theta)$. In the present experiment there are theoretical means[24,25] to express $Q_e(\phi)$. Therefore, the theory of Jakas *et al.* may be applicable to the explanation of our geometrical effect. In conclusion, more systematic experimental studies such as those on dependences on proton energy and on target atomic number and theoretical explanation are needed for the proper understanding of the geometrical effect.

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