Note added in proof. Preliminary measurements in this laboratory to 130 keV confirm that Mapleton's calculation overestimates the $2p$ capture as suggested in the discussion of the helium results.

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PHYSICAL REVIEW A VOLUME 1, NUMBER 5 MAY 1970

Analysis of Recoil He^{+} and He^{++} Ions Produced by Fast Protons in Helium Gas*

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The cross sections for formation of He^{+} and He^{++} ions have been measured over the energy range from 0.15 to 1.00 MeV for the case of protons incident on helium gas. The individual ions were detected with an electron multiplier that was operated at near 100% efficiency. An observation of the angular distribution of the slow ions in a "field-free" environment yielded essentially an isotropic distribution in the laboratory reference frame. It was also observed that much less than 1% of the ions had energies greater than 1 eV. It was concluded that, in order for the recoil ions to have an appreciable angular distribution, there must be a substantial energy transfer in the collision, such as would be the case for heavy projectile-target combinations.

INTRODUCTION

A magnetic-deflection analyzer has been constructed for measurements differential in charge state and recoil angle of the ions produced in ionizing collisions of a beam of fast particles. The recoil He^{+} and He^{++} ions produced in helium gas by protons with energies in the range 0.15 to 1.00 MeV have been measured.

The case of incident protons, which were used in the present experiment, has been studied with fixed-angle spectrometers using a collection field by Solov'ev *et al.*[1] and by Wexler.[2] The measurements of Solov'ev *et al.* cover energies only up to 0.18 MeV, while those of Wexler ranged from 0.80 to 3.75 MeV. Both groups have studied protons on He, Ne, Ar, and Kr targets.. Although their energy ranges do not overlap, a comparison of sorts can be made by extrapolation. There is an appearance of good agreement for the low charge states of the slow ions, but this actually results from the fact that neither set of measurements was absolute. Solov'ev's group normalized to their own total ion production measurements, while Wexler normalized to previous measurements made in this laboratory on total ion production cross sections.[3,4] The apparent agreement for the ions of low charge state thus really reflects only the rather good agreement between these two sets of total ion production measurements. Significantly, the agreement does not appear to be as good for some of the higher charge states of the recoil ion; in fact, for some cross sections, the extrapolated comparisons disagree by more than a factor of 8.

Afrosimov and Fedorenko[5] have used a magnetic slow-ion analyzer, which is rotatable about a field-free collision region and has a direction-defining collimator, to study the relative production of each slow ion charge state, differential in the recoil angle. The instrument has sufficient momentum resolution to provide a low-resolution measurement of the recoil ion energy, and this was supplemented by a retarding potential feature for independent energy determinations. In studies of Ne^+ and Ar^+ ions up to 0.18 MeV in neon and argon targets, they found that quite appreciable fractions of the higher charge state recoil ions had initial energies of more than 200 eV. In fact, it was observed that virtually all of the Ar^{5+}, produced from an Ar target gas by incident Ar^+ projectiles, had energies greater than 1 keV. Further investigations revealed that earlier studies[6] made in their own laboratory of the same collision partners, with a fixed-angle analyzer and a collection field, were significantly in error for the recoil ions that were more than triply charged, particularly when the mass of the projectile was of the same order as the target mass.

Morgan and Everhart[7] have also studied the energy distribution of the recoil ions in Ar^+-on-Ar collisions, at selected recoil angles that were well forward from 90°, corresponding to very hard collisions. They did indeed find recoil particles at these angles, particularly those of the higher charge states, with energies of 1 keV and more. This particular paper gives no absolute figures on the intensities of the recoils, or on the relative contribution to the total cross section; but it does verify that there are measurable numbers of recoils at these forward angles.

The suggested conclusion is that measurements of absolute or even only relative cross sections for the production of multiply charged slow ions, using a fixed-angle spectrometer and relying on collection of the ions to the entrance slit by an electrostatic field, can be substantially in error in some circumstances. Therefore, it was considered essential to include the capability of a field-free angular measurement and yet not preclude the use of a collection field when desired. It is not indicated that either of these collection methods is individually sufficient for the general case, but rather that a combination of the two methods is necessary.

GENERAL DESCRIPTION OF APPARATUS

A detailed drawing of the apparatus is shown in Fig. 1. In this apparatus the beam was passed through a collimating cone and into the collision chamber in which it underwent ion producing collision with the target gas. Also inserted into the collision chamber was the incident beam detector and the slow ion collimator cone of the spectrometer, both of which were mounted such that they could be rotated about a fixed point in the collision region. The spectrometer, which was rigidly connected to the slow-ion collimator, employed an ion lens system for acceleration and focusing and an electromagnet for charge-to-mass analysis of the ions. An electron multiplier was used for the detection of the ions as they emerged from the analysis region.

The apertures in both the beam and slow-ion collimating cones were machined in small buttons which were inserted into the cones. The gas pressures were sufficiently low that charge-changing collisions in the collimators were unimportant in this investigation.

The design of the beam detector was determined primarily by the desire to maintain the collision region free of stray fields and still suppress secondary electron ejection from the detector. In order to implement this concept, an electrically grounded shroud was placed around the electron suppression cage which housed the beam collector. A suppression potential in excess of about − 15 V was sufficient to cause the measured beam current to saturate. The current to the entrance tube of the detector was negligible compared to the beam current.

The collision chamber was constructed from stainless-steel tubing and is attached to the beam collimator, the beam detector, and the spectrometer collimator by means of three welded stainless-steel bellows. In order to increase the range of sampling angles of the spectrometer, the collision chamber was mounted such that it would rotate about its vertical axis.

Two different types of measurements were performed with this spectrometer. The first measurements involved the use of a field-free collision region and observation of the angular distribution of the recoil ions. The second type of measurements involved the collection with an electric field of all the ions formed along a portion of the beam path, regardless of their original directions of recoil. For the latter measurement an electrode was used to repel the ions into the entrance aper-

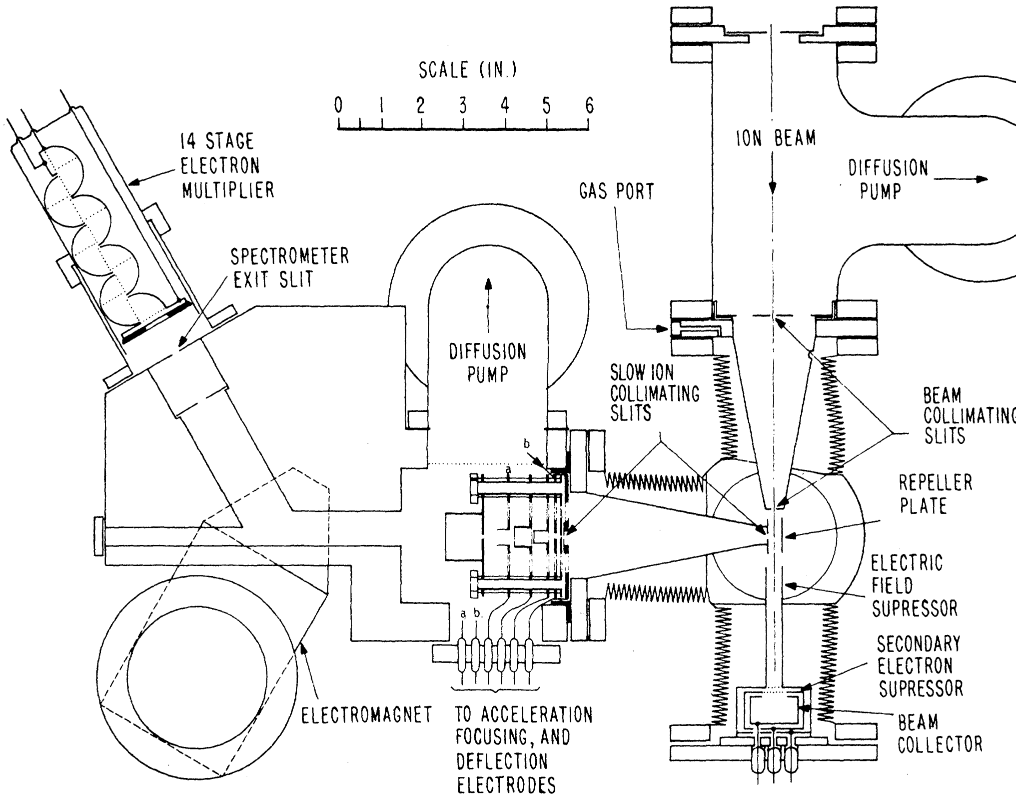

FIG. 1. Detailed drawing of collision and analysis regions.

ture of the collimator.

In both sets of measurements the ions that exited the collimator were passed through a six-element electrostatic ion lens which served to accelerate and focus the beam before entering a Nier-type[8] 60° sector magnetic field, which refocused it in the exit slit of the spectrometer. Mounted behind the exit slit was an electron multiplier detector which was operated in a pulse-counting mode.

The spectrometer was operated at rather low resolving power (about ten) because the interest in the present work was not in high resolution, but rather in the attainment of flat-topped profiles in the ion peaks.

In this apparatus, it was desirable to detect the analyzed ions with an efficiency of around 99%. The achievement of such a high efficiency required that about 99% of all incident ions eject at least one secondary electron from the first dynode of the multiplier. It was explained by Dietz[9] that the expected frequency distribution for producing n, $n = 0, 1, 2, \ldots$, secondary electrons is given by the Poisson distribution $(\gamma^n/n!)e^{-\gamma}$, where γ is the average secondary emission coefficient. Therefore, according to the above distribution, it is necessary that $\gamma = 5$ in order that 99% of all the ions eject at least one secondary electron. The relation between the mass and energy of an ion to the secondary emission coefficient for typical multiplier surfaces has been demonstrated by Akishin,[10] and, according to the figures he presented, an ion energy considerably greater than 10 keV is required to attain $\gamma = 5$ for the light helium ions. Therefore, to achieve these ion energies, a high postanalysis voltage was employed to accelerate the ions into the detector.

For the study of the recoil angles of the ionized target molecules, it was imperative that the collision and sampling environment be as free of stray fields as practicable. For this reason the collision chamber was constructed of stainless steel and sealed with metal gaskets. Liquid nitrogen-trapped mercury diffusion pumps, in lieu of oil diffusion pumps, were used in order to prevent backstreaming of insulating oils. Even with well-designed cryogenic traps, some creepage of pump oil is usually observed[11] because oil wets all trap surfaces and, therefore, can migrate along the surface into the system. However, mercury does not wet stainless steel, consequently it does not creep.[11] It should be noted, however, that even if mercury did creep into the system a thin conducting film of mercury is clearly preferred over that of oil.

To minimize the effect of contact potentials, the entire collision chamber and components were rhodium plated. Rhodium was chosen because it neither oxidizes nor amalgamates.

It is necessary to consider stray magnetic fields in the vicinity of the collision chamber, for these will also affect the ion sampling efficiency. Shielding around the collision chamber, however, served to reduce the stray magnetic fields to less than 1 G in the collision and sampling regions.

EVALUATION TESTS OF APPARATUS

Many checks on the performance of the apparatus were made for protons incident on helium targets.

An evaluation of fundamental importance in this apparatus concerned the response of the multiplier count rate to the ion accelerating voltage. It was of particular interest to demonstrate that the ion counting efficiency was near 100%. For the reasons discussed previously, it was believed that when the ion acceleration potential reached a sufficiently high value to produce an average secondary yield of five or more electrons per incident ion, then the detection efficiency would be approximately 100%. The results of this test are shown in Fig. 2. It was concluded that for potentials above about 14 kV both He^{+} and He^{++} ions are detected with equal and near 100% efficiency.

Tests were also performed to assure that thin target conditions prevailed in the collision chamber and that the ion count rate of each ion saturated when a repeller field was employed for ion collection.

In the measurements on the angular distribution of the slow ions produced in the H^{+}-He collision, it was important to evaluate the contribution of subsequent charge changing collisions in the gas. Of particular concern was the resonance charge exchange process ($He^{+} + He \rightarrow He + He^{+}$), which could destroy the He^{+} recoil angular distribution.

The following test revealed that the resonance charge exchange process was not important in this study. The He^{+} ions that are formed in the gas receive an amount of energy proportional to the distance through which they travel in the repeller field. Consequently, the ions produced through charge changing reactions have less energy than those that are produced on the beam axis. Specifically, the presence of charge changing reactions would cause the ion energy profile to be skewed toward lower energy. The energy profile could be examined by means of the spectrometer. Low repeller potentials (< 50 V) were required, however, so that the resonance cross section would still be large. It was also necessary to use a low-

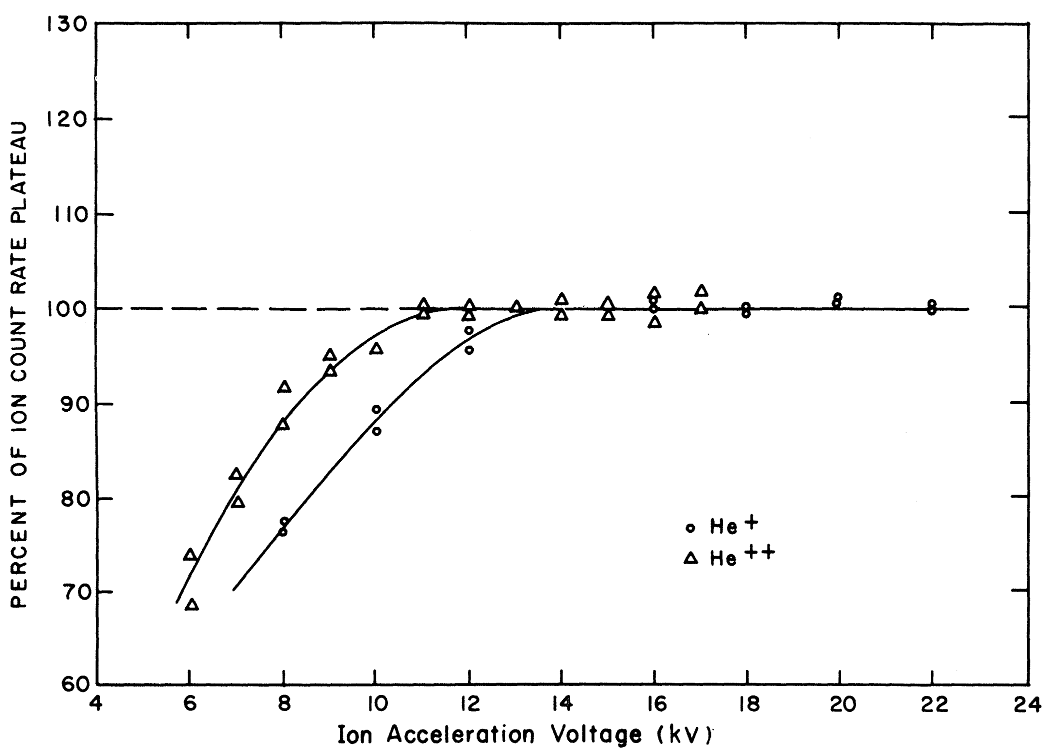

FIG. 2. Response of multiplier ion count rate to ion acceleration voltage.

ion analysis energy in order not to disguise the energy distribution acquired from the repeller field. However, when such an examination was performed over a pressure range of interest, 5×10^{-6} to 5×10^{-4} Torr, in the collision chamber, no evidence was found for secondary charge changing collisions.

Similar tests were employed to demonstrate that the effects of charge changing collisions in other parts of the apparatus were not significant.

EXPERIMENTAL RESULTS AND COMPARISONS

In the investigation using the field-free collision geometry, both the He^+ and He^{++} recoil ions were found to be isotropic over recoil angles from 60° to 95°. This finding, together with retarding potential measurements, indicated that virtually all of these recoil ions have energies less than 1 V. With ions in this energy range, the angular distribution could be substantially distorted by surface potentials and small stray magnetic fields. Furthermore, Kessel and Everhart[12] have pointed out that the effects of the thermal motion of the target atoms are sufficient to substantially broaden even a rather sharp angular distribution. Adapting the results of their analysis to the present energy range and collision pairs, it is indicated that the half-width of the peak (due to thermal motion alone) would be 10° to 20° or more for recoil angles around 90°. Considering both the low energies of the recoil ions and the effects of thermal motion of the gas, it is understandable that no angular distribution was observed.

Under these circumstances, it could not be assumed that stray field effects would not discriminate against a substantial fraction of the lowest energy ions, despite all precautions. Therefore, for total ion production measurements, the repeller electrode was installed and used to obtain the cross sections for production by protons of He^+ and He^{++} ions, irrespective of their recoil angles; these data are shown in Table I and Fig. 3.

The relative magnitudes of the two curves and the energy dependencies are absolute, but the figure has been normalized to the total ionization cross section at 1 MeV that was measured in this laboratory by Hooper.[13]

The present data give an excellent fit to a straight line on a log-log plot throughout the energy range investigated. These data, therefore, correspond to an expression of the form

$$\sigma = AE^{-c},$$

where E represents the proton energy. These two cross sections can then be represented as

$$\sigma_+ = 2.07E^{-0.75}\times10^{-17}\ \mathrm{cm^2/atom},$$

$$\sigma_{++} = 0.71E^{-1.37}\times10^{-19}\ \mathrm{cm^2/atom}.$$

Shown for comparison with the present results are the proton data of Solov'ev *et al.*[1] below 180 keV and that of Wexler[2] above 0.80 MeV. It is seen that excellent agreement is obtained with the overlapping measurements below 180 keV; in fact, the data are essentially the same. For comparison with the data above 0.80 MeV, it should be noted that Wexler also measured only relative cross sections and normalized these to the same σ_i measurements of Hooper[13] that were used in the present experiment. One can regard this as normalization of only the σ_+ cross section, because at 1-MeV energy the σ_{++} is only 0.35% of the σ_+ cross section. Therefore, the present He^+ measurements and those of Wexler are in forced agreement at the 1-MeV energy point. Consequently, the only comparison to be made between the two measurements of the σ_+ cross section is in the energy dependence, which Wexler observed to be slightly steeper (about $E^{-0.82}$) than the present value of $E^{-0.75}$.

The comparison between the present results for He^{++} and those of Wexler does *not* reflect the normalization procedure, and their absolute agreement is significant. Here also the energy dependencies are slightly different: Wexler's results demonstrate an $E^{-1.2}$ dependence as compared with the present value of $E^{-1.4}$. It is believed that this difference may be significant because the four highest energy points of the present data blend into the energy dependence observed by Wexler.

DISCUSSION

Thomas[14] has measured the cross section for the excitation of one discrete line of He II by the impact on neutral helium of protons in our energy range. From these data he was able to estimate[15]

TABLE I. Cross sections for production of He^+ and He^{++} ions in helium gas by incident protons.

Proton energy (keV)	Measured relative cross section σ_+/σ_{++}	Calculated absolute cross sections (10^{-18} cm²/atom) σ_+	σ_{++}
150	90.7	88.0	0.970
200	106	71.8	0.678
300	143	51.0	0.356
400	177	42.9	0.242
500	199	35.0	0.176
600	215	31.4	0.146
700	239	27.3	0.114
800	254	24.8	0.0977
900	274	22.7	0.0828
1000	283	20.7	0.0732

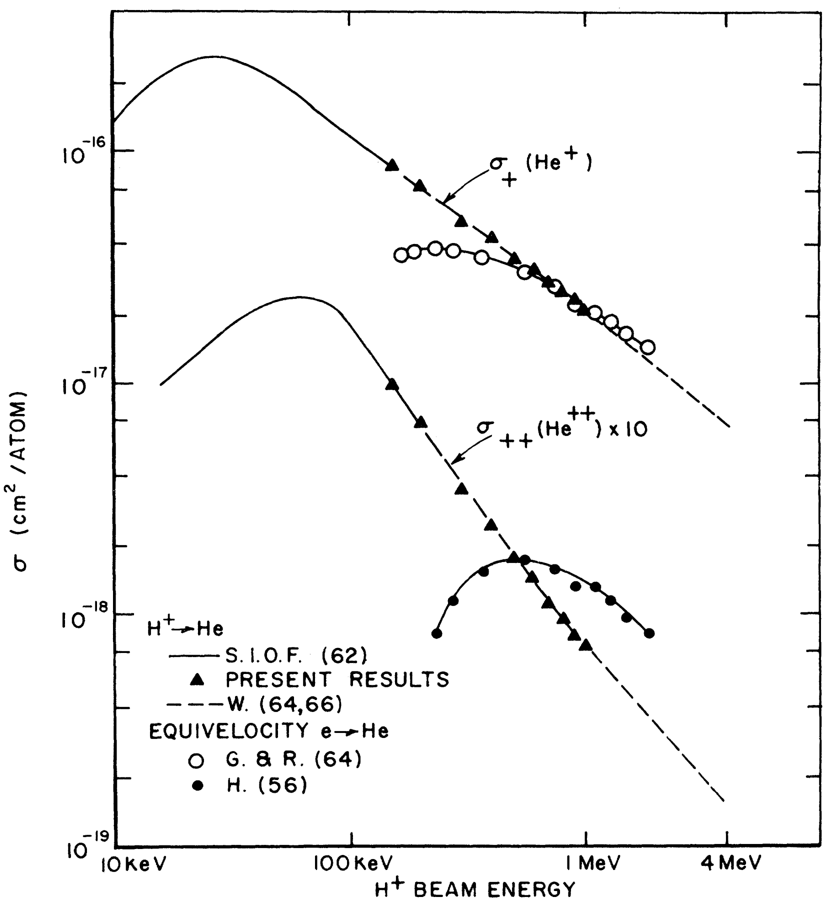

FIG. 3. Partial cross sections for production of He$^+$ and He^{++} ions by incident protons. Key to the results of other investigators: S.I.O.F. (62), Solov'ev *et al.*, (Ref. 1); W. (64, 66), Wexler (Ref. 2); G. & R. (64), D. Rapp and P. Golden [Lockheed Missiles and Space Co. Report No. 6-74-64-12, 1965 (unpublished)]. H. (56), H. Harrison, *The Experimental Determination of Ionization Cross Sections of Gases under Electron Impact* (Catholic University of America Press, Washington, D.C., 1956).

the cross section for the formation of He$^+$ with $n=4$, and he obtained an energy dependence that is strikingly similar to the present cross section for He^{++} production. This case of simultaneous excitation of one electron and the removal of the second is obviously similar to the removal of both electrons.

Mapleton has performed Born approximation calculations that are relevant to this case. He has calculated the cross section for the simultaneous ionization and excitation ($n=3$ states) of helium,[16] and also for the simultaneous capture by the proton of one electron from the helium along with excitation ($n=2$ states) of the remaining electron.[17]

Thomas[15] has scaled these calculations of Mapleton (assuming the cross sections are proportioned to n^{-3}) to estimate the total cross section for the formation of He$^+$($n=4$). These results, although somewhat lower than the estimate based on experimental data, indicated an energy dependence of about E^{-2} for the lower energies (about 0.2 MeV) where charge transfer dominated the He$^+$ formation; whereas for higher energies (above about 0.6 MeV) where ionization dominated, the energy dependence was less than E^{-1}.

Correspondingly, it is probable that in the He^{++} production there is a strong contribution from single ionization plus charge transfer in our energy range, but, that at higher energies, double ionization will dominate and will perhaps tend to an E^{-1} asymptotic dependence.

Shown for comparison purposes in Fig. 3 are the cross sections for production of these two helium ions by electrons that are of the same velocity as the protons. According to the Bethe-Born approximation,[18] which applies only to simple ionization events by point charge projectiles, the cross sections depend only on the charge and velocity of the projectile. Therefore, the simple ionization cross sections should be equal for equivelocity protons and electrons. It is seen in Fig. 3 that this prediction is fulfilled for σ_+ at proton energies above about 1 MeV. However, for the less frequent collisions that produce He^{++}, there is a substantial difference in the electron and proton cross sections even for the highest energies shown. This discrepancy is believed significant because there is considerable agreement (± 5%) on these electron cross sections by other investigators[19–21] and those that disagree[22] are usually higher and, consequently, in less agreement with the proton results.

A relevant point with regard to the experimental apparatus used by Solov'ev and Wexler is that both of these investigators employed electrometers to measure the ionization currents, in contrast to the present experiment in which an electron multiplier was used. The excellent agreement on the measured cross-section values among these several laboratories supports the conclusion that the electron multiplier was operated with equal detection efficiency for both helium ions.

The maximum error in the relative magnitudes of these two helium cross sections is not expected to be more than ± 4%. In order to assign error limits to the absolute cross sections, it is necessary to include the ± 6% error limits in the σ_i measurement of Hooper used for normalization. Thus, it is estimated that the probable error in the absolute cross sections for production of He^+ and He^{++} is less than 10%, most of which is due to the normalization procedure.

The lack of an observed angular dependence of the recoil ions in the case of protons incident on helium was previously discussed. On the basis of this observation, it was concluded that a detectable angular distribution would be obtained in the total energy spectrum of a given ion only for those collision partners for which there is substantial energy transfer (at least several eV). The Rutherford scattering expression[23] indicates that the energy transfer to the target is proportional to the square of the product of the atomic numbers of the collision partners. Therefore, in order to produce an appreciable energy transfer, a heavy projectile-target combination should be used.

In confirmation of this conclusion, observations[24] have recently been made for Ne^+ ions into argon, in the same energy range. In this case only the field-free collision region was employed, and a retarding potential within the analyzer was used to bias out all the ions below a given initial energy. The angular distributions obtained with zero retarding potential were not isotropic in this case, but they were much more diffuse than those reported by Afrosimov and Fedorenko[25] for the same condition. For small retarding potentials (< 2V) the distributions changed appreciably, and for about 2 or 3 V they were roughly similar to Afrosimov's distributions for 0 V. For at least the singly and doubly charged neon ions, there seem to be many more ions formed with energies less than about 2 V than he was able to detect.

It is evident that these large abundances of very low-energy ions will continue to be a source of considerable difficulty in attempts to perform absolute differential cross-section measurements in the future.

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Fast-Electron–Ion Coincidence Measurements of the Argon Atom-Ion Collision*

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The triple-valued structure in the inelastic energy loss in certain violent Ar^+-on-Ar collisions is commonly thought to be due to the creation of L-shell vacancies. These should deexcite through an Auger process that results in the emission of fast electrons whose energy is of the order of 180 eV. The present experiment detects such electrons in coincidence with a scattered ion appearing at a known angle θ with a known final charge state m. Large-angle scattering in the reaction $Ar^+ + Ar \rightarrow Ar^{+m} + Ar^{+n} + (m+n-1)e^-$ is studied for incident energies between 10 and 30 keV, where the Ar^{+m} is scattered to $\theta = 21°$. The electron-energy spectrum measured in coincidence shows the expected peak, though its location in energy decreases as m increases. The number of fast electrons agrees fairly well with the number expected according to a statistical model developed earlier.

I. INTRODUCTION

Large-angle Ar^+-on-Ar collisions show a striking discontinuity in the inelastic energy loss $\overline{Q}$ for collisions where the distance of closest approach is about 0.25 Å. This corresponds to 19-keV incident energy at a scattering angle of 21°, or to any reasonable combination of incident energy T_0 and scattering angle θ whose product is about 400 keV deg. In this region, the $\overline{Q}$ values have a triple structure that has been studied in many experiments: Morgan and Everhart,[1] Afrosimov, Gordeev, Panov, and Fedorenko,[2] Kessel and Everhart,[3] and Fastrup and Hermann.[4] Theoretical treatments by Fano and Lichten,[5] Lichten,[6] and Everhart and Kessel[7] have explained this structure in terms of L-shell vacancies created during the collision that are subsequently filled by Auger transitions. The three peaks in the structure are attributed to cases where such vacancies are created in neither, or one, or both of the colliding argon particles.

These Auger transitions should result in the ejection of fast electrons of about 180 eV energy. Such electrons have been seen in experiments by Rudd, Jorgensen, and Volz,[8] and by Ogurtsov, Flaks, Avakyan, and Fedorenko.[9] Further work by Rudd and Cacak[10] indicates the fast electrons are nearly isotropic in direction in a frame moving with the emitting atom. In the previous measurements of electron spectra,[8-10] the electrons studied arose from collisions where the scattering angle and impact parameter were not known.

The present experiment measures the energy spectra of only those electrons emitted from collisions of fixed T_0, θ, and final scattered-ion charge m. A coincidence technique is used to associate a known energy electron with the scattered ion from the same event. These detailed measurements should make possible a point-by-