

## Measurements of the Total Cross Section for the Scattering of Low-Energy Electrons by Metastable Argon†

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Measurements are presented of the total cross section for scattering of low-energy electrons by a mixture of  $^3P_0$  and  $^3P_2$  metastable argon. The atom-beam-recoil method was used to make an absolute measurement. The cross section was measured from 0.35 to 6.75 eV with an angular resolution of  $16.1^\circ$  in the electron polar scattering angle at an electron energy of 1 eV. The velocity distribution of the atom beam was measured by time-of-flight spectroscopy. Electron beams with typical energy distributions of 0.3-eV full width at half-maximum were used. The over-all error in the cross section is estimated at  $\pm 13\%$  and the electron-energy determination is believed accurate to  $\pm 0.150$  eV.

### I. INTRODUCTION

The atom-beam-recoil technique<sup>1</sup> has been used in recent years to investigate low-energy electron scattering from atomic and molecular targets.<sup>2-4</sup> All of these experiments, with the exception of that of Neynaber *et al.*<sup>4</sup> for which only preliminary data were reported, were on ground-state systems. The metastable rare gases afford the opportunity of making a scattering measurement on an excited state of an atomic system.

The  $^3P_{0,2}$  metastable states of argon are the subject of this investigation. The  $^3P_0$  state lies 0.175 eV above the  $^3P_2$  state which in turn lies 11.57 eV above the ground state. The electron configuration is  $1s^2 2s^2 2p^6 3s^2 3p^5 4s$ . The role of the polarization of the atomic electrons in the scattering process will be emphasized here because of the very large electric dipole polarizability<sup>5</sup> ( $\approx 330a_0^3$ ), which is attributable almost entirely to the valence electron.

Although there has been a surge of interest lately in the calculation of elastic scattering cross sections for electrons on the metastable states of the rare gases,<sup>6-9</sup> most of the calculations have been made on metastable helium. To our knowledge the only published calculation of the elastic scattering cross section for electrons on metastable argon has been made by Robinson.<sup>8</sup> The results of this calculation will be discussed in Sec. V C.

### II. EXPERIMENTAL METHOD

An outline of the apparatus used in this experiment is shown in Fig. 1. A beam of ground-state argon atoms is allowed to effuse from an oven at thermal velocities. It is then cross fired with an electron beam and a small fraction of the atoms is placed into the  $^3P_2$  and  $^3P_0$  metastable states. The beam then enters the scattering region where a second electron gun is alternately turned on and off. Those metastable atoms elastically scattered on the average by more than a critical angle, signifying

the angular resolution of the apparatus, are not detected. The cross section can be calculated through knowledge of the electron current, the average velocity of the atom beam, the geometry of the interaction region, and the ratio of the scattered-out atom beam to the full (unscattered) beam.

Since scattering via other channels, including direct deexcitation inelastic and ionization events, will contribute to the scattering signal as well, the cross-section measurements reported here are total cross sections. However, the polarization interaction is so strong in the metastable rare gases that, particularly at low energies, the contribution of elastic scattering to the total cross section is expected to be dominant.

As shown in Fig. 2 the atom beam has cross-sectional area  $h_a w$  while the electron beam threads through the atom beam with cross-sectional area  $h_e l$ . The probability that a single electron will be scattered is simply

$$P = N\sigma/h_e l, \quad (1)$$

where  $N$  is the total number of target atoms present in the interaction volume,  $\sigma$  is the effective cross section (taking into account the angular resolution), and  $h_e l$  is the total area an electron can pass through.  $N$  can be calculated from

$$N = \int [J_a(x, y)/\gamma\langle v \rangle] d\tau, \quad (2)$$

where  $J_a(x, y)$  is the detected atom-beam current density, as measured by a detector whose efficiency is  $\gamma$ ,  $\langle v \rangle$  is the average speed of the particles found in a volume element of the beam, and  $d\tau$  is a volume element in the interaction region. With the assumption that  $J_a(x, y)$  is a constant over the interaction volume, we obtain

$$P = I_a \sigma / \gamma h_a \langle v \rangle, \quad (3)$$

where  $I_a = J_a h_a w / \gamma$  is the atom-beam current passing through the interaction region and hitting the detector. If  $P$  is multiplied by the electron current

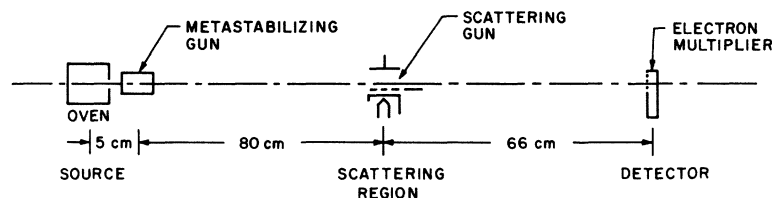


FIG. 1. Schematic view of apparatus.

$I_e$  (electrons/sec) passing through the interaction volume, the scattered current  $I_s/\gamma$  (events/sec) is obtained, where  $I_s$  is the observed scattered current, again as measured by the same detector which determined  $J_a$ .

The cross section is thus given by

$$\sigma = (I_s h_a / I_e J_a) \langle v \rangle. \quad (4)$$

The ratio  $I_s/I_e$  is the ratio of the decrease in atom-beam current detected when the electrons are on to the entire atom-beam current detected. Note that the detection efficiency does not appear in Eq. (4) so long as  $I_s$  and  $I_e$  are measured with the same detector. A more complete discussion of the validity of the assumptions made in deriving Eq. (4) is contained in a recent article by Collins, Beder-son, and Goldstein,<sup>10</sup> in which analogous measurements on potassium are presented.

The value of  $\langle v \rangle$  can be determined either by assuming a specific speed distribution, as in previous electron scattering experiments,<sup>2,3</sup> or by direct measurement. Since the production of the metastable beam by electron bombardment will necessarily affect the source speed distribution, a measurement of the metastable atom speed distribution is clearly desirable. Such a determination was made in the present experiment by a time-of-flight technique.<sup>11</sup> The source gun is pulsed briefly and the metastables arriving at the detector are counted as a function of the elapsed time from the pulse. This transit time distribution is then converted to a speed distribution and the average velocity can be computed numerically.

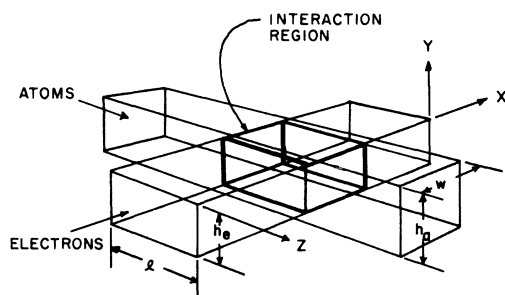


FIG. 2. Interaction-region geometry.

### III. APPARATUS

#### A. Source

Argon gas is fed into the source well through a leak valve, and leaves through a 6-mm-high by 1-mm-wide slit. An electron beam intersects this ground-state atom beam along the vertical direction. It is produced by a Philips type-A dispenser cathode. A magnetic field of approximately 250 G is used to collimate the electron flow. The source gun is operated well above the threshold for metastable production so that some photons are produced as well. These must be corrected for, although they also perform the useful function of supplying a zero-time reference for the time-of-flight analysis.

#### B. Scattering Gun

Both electron guns used in this experiment are of the same mechanical construction and reflect the same design considerations. They are of modular type, using interchangeable parts that are adapted for each particular use<sup>12</sup> (see Fig. 3). The electrons move as a rectangular beam through parallel grids perpendicular to the electron beam. The grids are photoetched and accurately positioned so that all grid meshes are aligned. The scattering region is formed by two molybdenum blocks and an entrance grid. The anode is formed in two pieces, a slitted anode which collects 95% of the electrons and an additional electrode which collects the electrons that pass through the slit. This arrangement is useful when making retarding potential measurements since reflecting 5% of the electrons does not significantly disturb the space-charge distribution further back in the gun. The gun uses a collimating magnetic field of about 400 G.

The scattering gun uses an oxide-coated cathode, Raytheon type 4D32. Currents of 10–4000  $\mu\text{A}$  are produced with typical energy distributions of 0.3-eV full width at half-maximum (FWHM), as measured by the retarding potential technique. The background pressure in the scattering chamber is normally about  $5 \times 10^{-8}$  Torr.

The absolute energy of the electrons, independent of work-function difference, was also determined by the use of retarding potentials. The energy distribution of the electron beam is plotted for a series of cathode-interaction region potentials (see Fig. 4). For high values of this potential the dis-

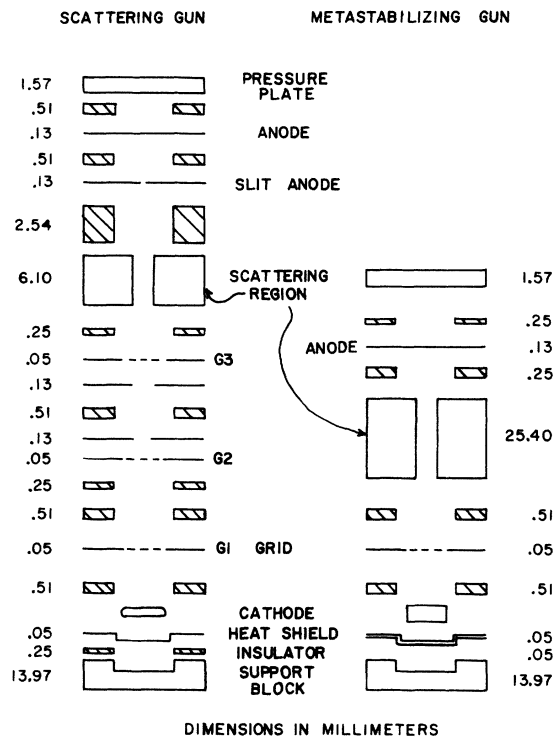


FIG. 3. Schematic side views of scattering and metastabilizing guns.

tribution is reasonably symmetric and exhibits 0.2–0.4-eV FWHM. As the cathode-interaction region potential is decreased the distribution becomes highly skewed exhibiting a sharp cutoff on the low-energy side. This is due to electrons being retarded and turned around at the interaction region. This cutoff determines the zero of energy in the interaction region. This energy determination is accurate to  $\pm 0.150$  eV.<sup>13</sup>

#### C. Detector and Electronics

The detector consists of a Bendix model 306 magnetic electron multiplier with a suitable mask. The metastable beam falls directly on the tungsten cathode of the multiplier and is detected by emission of Auger electrons. Multiplier currents of the order of  $3 \times 10^{-8}$  A are typically observed, with a gain of about  $10^6$ .

Time-of-flight spectra are accumulated by connecting the multiplier output to a preamplifier, amplifier, and discriminator in series, and feeding the discriminator output to a multichannel analyzer operating in its multiscale mode.

For scattering measurements an electrometer is used. Its output is digitized with a voltage-to-frequency converter and fed to the input of the analyzer again operating in a multiscale mode. A programming sequence is employed in which the scattering

electrons are cut off completely for one part of the sweep, and the electron energy is increased linearly through the remainder of the sweep. The analyzer is triggered to run continuously with cross-section measurements made over the entire spectrum of electron energies every 5 sec. About 10 000 such sweeps constituted a typical run.

## IV. RESULTS

### A. Time-of-Flight Data

Figure 5 shows data taken to measure the transit-time distribution in the beam. Each of the 400 channels corresponds to a 25- $\mu$ sec interval in the transit time. The pulse which activates the metastabilizing gun is also of 25- $\mu$ sec duration and is delayed with respect to the start of each sweep. The photons generated by this pulse appear in channel 18. This provides us with a precise indication of a zero-time base line. Also, since the photons are always counted in the unscattered beam, it is necessary to correct the beam signal for the photon background. The photon current is measured in the time-of-flight mode by lengthening the metastabilizing pulse to approximately 20 channels and comparing the area under the curve due to photons to the total area. The correction applied as a consequence of this background was usually of the order of 3.0%.

### B. Scattering Data

Most of the data accumulated in this experiment were obtained by the technique described in Sec. III C. As a check, additional data were taken at 30 values of the energy. For these determinations the electron energy was preset and the electrons were modulated on and off. Each of these points required a separate electron-energy determination. Data

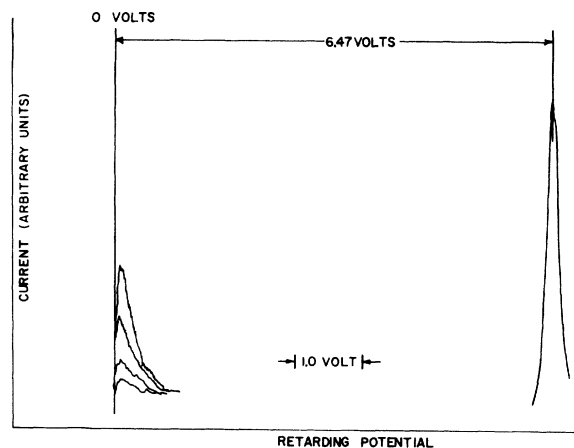


FIG. 4. Retarding-potential technique for absolute electron-energy determination.

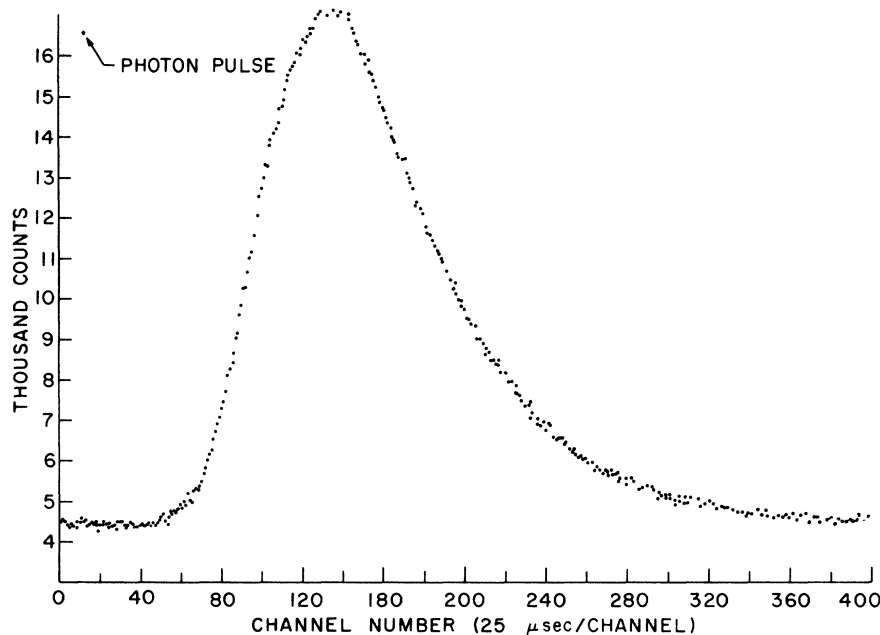


FIG. 5. Typical arrival-time spectrum for metastable argon. Photon pulse determines  $t = 0$ .

taken in this manner were in excellent agreement with the sweep data. The results are presented in Fig. 6.

## V. ERRORS AND COMPARISON WITH THEORY

### A. Error Estimates

The random statistical error present in the cross-section data is relatively small. It ranges from  $\pm 2$  to  $\pm 8\%$ .

The most serious sources of systematic error occur within the electron gun. Electrons hitting gun surfaces may produce low-energy secondary electrons which participate strongly in the scattering process. In order to minimize the occurrence of such events two steps are taken. First, the in-

teraction region is designed so that no grid is present as the electrons leave the region. Even though this grid mesh would be aligned with the others to minimize collisions, a serious problem would still exist. Second, the anode potential is kept at the relatively high value of +35.0 V with respect to the interaction volume, to trap low-energy electrons produced there.<sup>14</sup> The effectiveness of these procedures in eliminating scattering by electrons going the wrong way is tested by observing the "scattering-in" signal when the detector is placed off the atom-beam center to the cathode side of the electron gun. From momentum-conservation considerations, no "scattering-in" signal should appear here when there are no reflected electrons. Proper

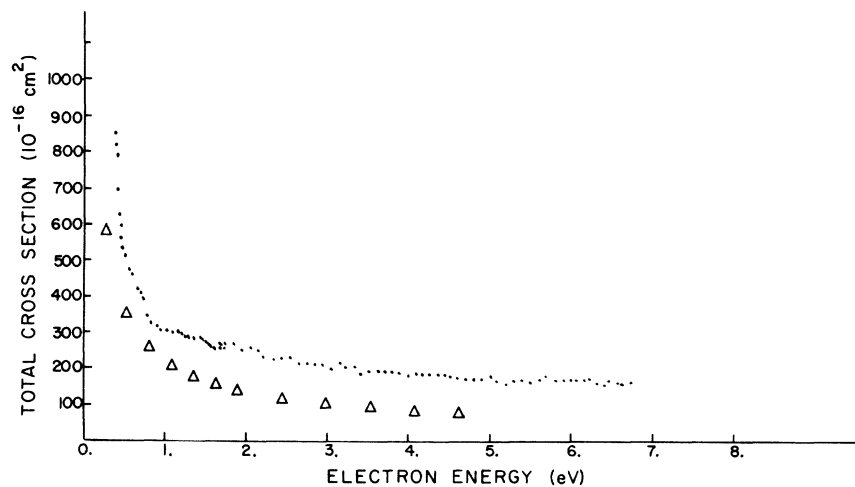


FIG. 6. Comparison of experimental results (●) and theoretical results of Robinson (▽) for total cross sections for the scattering of  $e$  electrons by metastable  $^3P_{0,2}$  argon. The Robinson results are corrected to correspond to the angular resolution of the present experiment.

choice of gun parameters eliminated this problem.

The electron-energy determination provides another possibility for systematic error. The retarding-potential procedure described above cannot correct for the space-charge potential depression that occurs in the scattering region. In order to correct for this the experimental electron-energy determination, taken under conditions where the space-charge potential depression was negligible, was used to fix the boundary conditions of the interaction volume. Then, using these boundary conditions, a solution was found numerically for the space-charge equation

$$\nabla^2\phi = K/\sqrt{\phi}, \quad (5)$$

where

$$K = 4\pi J\sqrt{m/2e}. \quad (6)$$

Here  $\phi$  is the potential,  $J$  is the electron current density,  $m$  and  $e$  are the mass and charge of the electron, respectively. This equation is valid with the assumption that the collimating magnetic field is strong enough that the space-charge depression acts only to slow electrons but not to significantly defocus them. The solution of this equation, averaged over the area of the atom beam, provides the actual electron energy that the beam sees, including space-charge effects and contact-potential difference. A typical space-charge correction is  $-0.18$  eV for a nominal energy of  $1.56$  eV when the current is  $0.166$  mA. Since the region over which the calculation was made included the anode, the effect of field penetration from the anode into the scattering region is also included. The over-all accuracy of the absolute electron-energy determination is estimated at  $\pm 0.150$  eV.<sup>13</sup>

Miscellaneous errors in the cross-section determination, including the error in the determination of  $\langle v \rangle$ , the error in calculating the photon background current, and the effect of metastables which are not normally detected being scattered into the detector, all contribute to an error estimate of  $\pm 13\%$  for the value of the cross section.

#### B. Angular Resolution

The term angular resolution, as applied here, refers to the ability of the apparatus to distinguish between atoms scattered by electrons and those that experience no interaction. Since both the atom beam and the detector have a finite height and width it is possible for an atom to be scattered and still be counted at the detector, i. e., not be registered as a collision. In order to simplify the angular-resolution calculation we think of the detector slit as being constructed from the overlap of a slit of finite width and infinite height and a slit of finite height and infinite width. For each of these slits we define a detection efficiency for the observa-

tion of a scattering event. We define  $\eta_w(\theta)$  and  $\eta_h(\theta)$  as the probabilities that a scattering event will be observed for a given electron polar scattering angle  $\theta$ , due to the finite width or height of the detector, respectively. These efficiencies are calculated by integrating over the azimuthal electron-scattering angle  $\phi$  in the expressions for the fraction of the atom beam scattered out of the detector. This is done for fixed values of the electron energy and average atom-beam velocity and as a function of electron polar scattering angle.<sup>15</sup> The over-all efficiency for the detection of a scattering event is obtained from the separate efficiencies by

$$\eta(\theta) = \eta_w(\theta) + \eta_h(\theta) - \eta_w(\theta)\eta_h(\theta). \quad (7)$$

Figure 7 shows these efficiencies for argon and 1.0- and 5.0-eV electrons. The stated angular resolution at 1.0 eV,  $16.1^\circ$ , is the point at which  $\eta(\theta)$  reaches 0.5. This analysis applies only to the elastic part of the cross section since all inelastic events will be detected with unit efficiency.

#### C. Comparison with Theory

Robinson<sup>6</sup> has performed an adiabatic calculation based upon an interaction potential of the form

$$V = V_{\text{HFS}} + \frac{e^2}{r} (1 - e^{-r/a}) - \frac{1}{2} \frac{\alpha e^2}{(r^2 + r_p^2)^2}, \quad (8)$$

where the first two terms are the Hartree-Fock-Slater potential of the atom modified by a factor

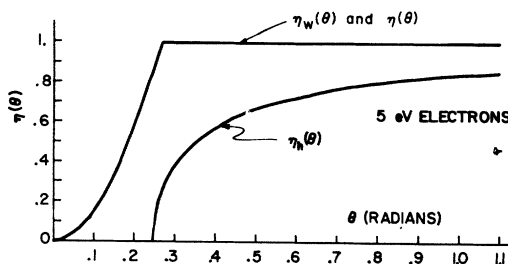
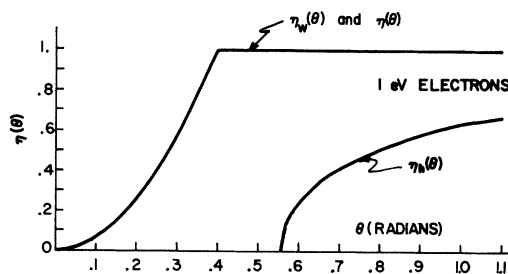


FIG. 7. Efficiencies for detection of a scattering event as a function of the electron polar scattering angle  $\theta$ .

which removes the Coulomb tail of  $V_{HFS}$ , and the third term represents the static electric dipole interaction.  $\alpha$  is the electric dipole polarizability taken from experiment,  $r_p^2$  is chosen to approximate the mean-square radius of the valence-electron orbital, and  $a$  is adjusted to yield the experimentally observed electron affinity deduced from measurements on electron resonances. The actual value of  $r_p$  employed was taken from the experimental polarizability by means of the variational formula  $\alpha = \frac{4}{3} \langle r_p^2 \rangle_{av}^2$ . Exchange is indirectly taken into account because the empirical electron affinity is influenced by exchange. However, no attempt was made to obtain properly symmetrized scattered wave functions.

It is known that the phase shifts, particularly at very low energies, are quite sensitive to variations in the parameters  $\alpha$ ,  $a$ , and  $r_p^2$ , particularly the last of these. Exchange, also at very low energies, contributes quite significantly to the scattering (as observed in the analogous experiments in the alkalis<sup>10</sup>). Thus it would be quite difficult to estimate the error in the Robinson calculation, although the principal feature of the scattering problem, which is the influence of the large polarizability, is well illustrated and should give qualitatively correct results. These observations are indeed borne out by comparison of theory and experiment.

It should also be noted that the experimental atom beam was composed of a mixture of  $^3P_0$  and  $^3P_2$  metastable argon while Robinson's calculation considers only the  $^3P_2$  state. Also, as mentioned in Sec. II, the experiment measures the total cross section while the calculation considers elastic scattering only.

In order to properly compare the present results to Robinson's prediction a correction must also be made for the angular resolution of the apparatus. This is particularly true because of the large number of higher partial waves in the theoretical cross section.

The effective elastic cross section that would be measured by the experiment is

$$\sigma = 2\pi \int_0^\pi \eta(\theta) \sigma(\theta) \sin\theta \, d\theta, \quad (9)$$

where  $\sigma(\theta)$  is the differential cross section. The value of  $\sigma$  has been calculated from Robinson's phase shifts and plotted along with the experimental results in Fig. 6. The correction was of order 10% at 1 eV. As expected, the experimental data lie above the predicted elastic cross section. There is no fine structure present in either curve. At higher energy the discrepancy becomes greater than a factor of 2, implying that the difference cannot be accounted for by inelastic events alone. This is not unexpected, in view of the considerations outlined above.

## VI. CONCLUSION

The atom-beam-recoil technique has been utilized to perform an absolute measurement of the total cross section for electrons on metastable argon. Innovations in this experiment are the use of direct measurements of the velocity distribution in the atom beam and the calculation of the potentials within the scattering gun to a high degree of accuracy. We have demonstrated the feasibility of performing accurate collision experiments employing metastable states indicating the need for a more complete theoretical attack on the problem. Work is continuing in our laboratory on further measurements on the other metastable rare gases. Both fine-structure and magnetic-substate selection is being employed in these experiments.

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PHYSICAL REVIEW A

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## Angular and Energy Distribution of Cross Sections for Electron Production by 50-300-keV-Proton Impacts on N<sub>2</sub>, O<sub>2</sub>, Ne, and Ar<sup>†</sup>

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Cross sections differential in angle and ejection energy for electron production by proton impact on nitrogen, oxygen, neon, and argon have been measured using electrostatic analysis and counting of individual electrons. The range of proton energies was 50–300 keV, the angles ranged from 10° to 160°, and the electron energies were measured from 1.5 to 1057 eV. Integrations over angle and/or electron energy yielded singly differential and total electron production cross sections. Our total cross sections for oxygen fall halfway between previous data of deHeer *et al.* and Hooper *et al.*, but our argon cross sections agree better with deHeer *et al.* Cross sections for electron ejection in the backward hemisphere are much greater for these multishell targets than for hydrogen and helium. The momentum-energy conservation hump which was prominent in hydrogen is less conspicuous for these gases.

### INTRODUCTION

The production of electrons in fast ion-atom and ion-molecule collisions is of basic interest in a number of areas. The energy distribution of electrons from such collisions is useful in understanding stopping power,<sup>1</sup> energy deposition phenomena,<sup>2</sup> and auroral and upper atmospheric processes.<sup>3</sup> The angular distribution is of considerable importance in testing various theoretical descriptions of the ionization process. Some of the theoretical aspects are discussed in the following paper.<sup>4</sup>

Cross sections for electron ejection which are differential in both angle and energy have become available only in the last few years. Kuyatt and Jorgensen<sup>5</sup> made the first such measurements for protons of 50–100 keV on hydrogen. Rudd and Jorgensen<sup>6</sup> made measurements on helium from 50 to 150 keV. The energy range was extended for both gases to 300 keV in the work of Rudd, Sautter, and Bailey.<sup>7</sup> This enters the region where Born-approximation calculations yield accurate results for total ionization cross sections, but it was shown that the angular distributions were still off by large factors. Cacak and Jorgensen<sup>8</sup> recently studied Ne<sup>+</sup>-Ne and Ar<sup>+</sup>-Ar collisions from 50 to 300 keV. Torburen<sup>9</sup> recently measured doubly differential cross sections

for electrons from H<sup>+</sup>-N<sub>2</sub> collisions from 300 to 1700 keV.

In the present work, we have measured angular and energy distributions of electrons from N<sub>2</sub>, O<sub>2</sub>, Ne, and Ar bombarded by protons from 50 to 300 keV. Comparison of these data with those on hydrogen and helium is made to determine the effect of inner shells on the ionization process. The data on atmospheric gases should be useful in determining the interaction of the protons in the solar wind with the upper atmosphere. Also since oxygen and nitrogen are important constituents of protoplasm, these results can be applied to the problem of energy deposition in cells and tissues.

### EXPERIMENTAL METHOD

The apparatus used in this experiment is the same as that used by Rudd and Jorgensen<sup>6</sup> and modified in the work of Cacak and Jorgensen.<sup>8</sup> A collimated, magnetically analyzed proton beam entered a chamber containing the target gas at pressures of about  $4 \times 10^{-4}$  Torr. The beam was collected in a shielded, positively biased Faraday cup and integrated. Electrons from a short length of the beam went to a 127° electrostatic analyzer placed at any of eight angles from 10° to 160° from the beam. Before entering the analyzer they were accelerated by