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Measurement of the Total Cross Section for Single-Electron Transfer in Collisions of He^+ with He in the Energy Range 2–22 keV*

W. N. Shelton and P. A. Stoycheff
Department of Physics, The Florida State University, Tallahassee, Florida 32306
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Measurement of the total cross section for electron transfer between a ground-state singly ionized helium atom and a ground-state helium atom has been made in the energy range 2–22 keV. The method used was that of collection of the slow ions produced by the charge transfer process. The collection was made via parallel plates with an electric field imposed perpendicular to the ion beam. The results are estimated to be accurate to 5%. The data are compared with the theories of Rapp and Francis; Sural, Mukherjee, and Sil; and Moiseiwitsch.

I. INTRODUCTION

During the past two decades many measurements have been made of the total cross sections for single-electron transfer between atoms and ions. Despite the relative straightforwardness of the experimental apparatus required to make these measurements, large discrepancies exist between the results of various experimenters; large discrepancies also exist between the various theories of the electron transfer process. The following single-electron transfer processes may occur when an ion A^+ collides with an atom A:

(i) $A^+ + A \rightarrow A + A^+$,

(ii) $A^+ + A \rightarrow A^* + A^+$,

(iii) $A^+ + A \rightarrow A + (A^+)^*$,

(iv) $A^+ + A \rightarrow A^* + (A^+)^*$.

Here the asterisk designates an excited state. The process of resonant or symmetric charge transfer (i) is the simplest process from the theoretical viewpoint, since the two atomic systems resulting from the charge transfer are the same as the ones before the collision. The cross section for this process is usually much larger than those for the other three processes. This resonant process may be viewed as not involving an electronic transition, but simply resulting in a phase change of the total wave function of the system. Although the collision of a proton with a hydrogen atom is the simplest such process to treat theoretically, it must be carried out experimentally with the difficult crossed-beam technique because of the chemical instability of the hydrogen atom. The next simplest process is the collision between a ground-state singly ionized helium atom and a neutral ground-state helium atom. Since the helium atom is chemically stable, no special preparation of the target gas is needed. The helium atom does not readily form a negative ion, so that double-electron transfer may be ignored. The high ionization potential of ground-state singly ionized helium (54.4 eV) makes the cross section for ionization of the projectile ion negligibly small compared to the charge transfer cross section. The large energy required to excite ground-state singly ionized helium (40.8 eV) makes the cross section for excitation of the projectile negligibly small compared to the charge transfer cross section, so that processes (iii) and (iv) need not be considered. Ionization of the target-gas atoms by the projectile may be ignored insofar as the experiment may be easily arranged in such a way that this process is not registered. Thus our measurements give the cross section for the sum of processes (i) and (ii), with (i) predominating.

The experimental method chosen for the present measurement is the direct method, as opposed to the indirect or charge-equilibrium method. In the direct method the experimental conditions must be such that the projectile makes only a single collision within the collision chamber. If the number density of target-gas atoms in the collision volume is known, and if the length of the collision volume is known, then the charge-transfer cross section may be obtained either by measuring the fractional neutralization of the ion beam, or by measuring the number of electrons removed from the collision volume by a known number of ions passing through the collision volume. Of these two measurement methods, the latter is preferable because it allows a clear definition of the collision volume, whereas the former does not. To see how the first method fails in this respect, consider an enclosure containing the target gas and having one opening through which the ion beam enters, and another opening through which the partially neutralized ion beam emerges. Because the target-gas atoms effuse from the openings in the enclosure, charge transfer will occur outside

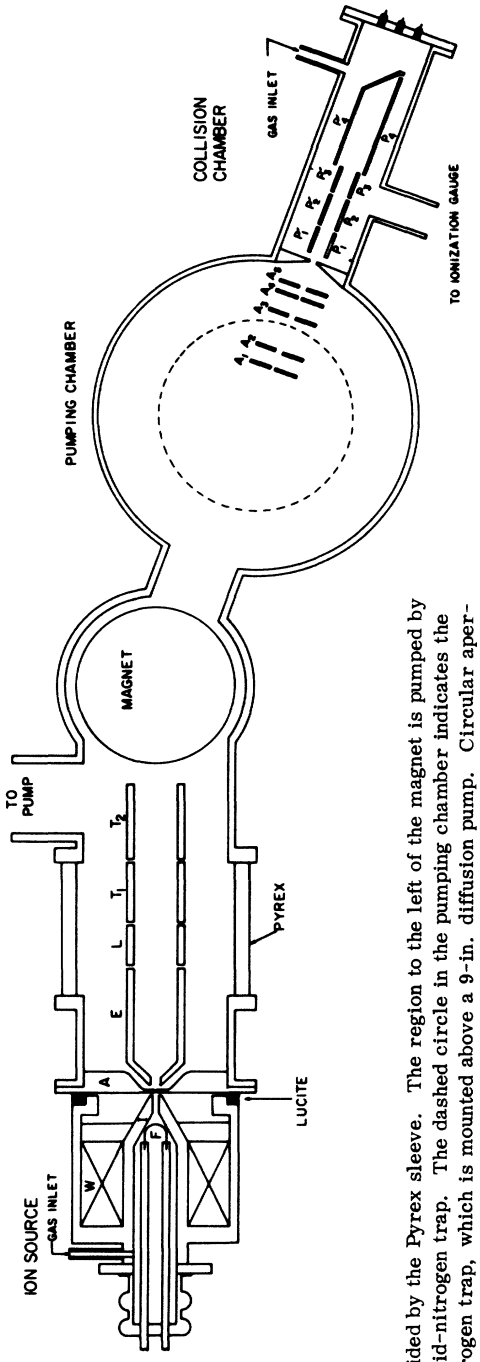

FIG. 1. Experimental apparatus. Ion source is a duoplasmatron. F is the filament; I the intermediate electrode; A the anode; E the extractor; L the einzel lens; T_1 the tube at same potential as E; and T_2 the grounded tube. Typical operating conditions of the ion source are − 200 V on F and − 100 V on I, with respect to A. Acceleration of the ion beam is attained by raising the ion source above ground by means of the insulation provided by the Pyrex sleeve. The region to the left of the magnet is pumped by a 4-in. diffusion pump with liquid-nitrogen trap. The dashed circle in the pumping chamber indicates the entrance to the 9-in. liquid-nitrogen trap, which is mounted above a 9-in. diffusion pump. Circular apertures A_1 through A_5 collimate the ion beam before it enters the cylindrical collision chamber. The vacuum envelope is made of aluminum, except for the pyrex sleeve and the exterior envelope of the ion source, which is made of soft iron. The elements I and A are also made of soft iron in order to form the magnetic circuit necessary to produce a strong magnetic field in the plasma region between I and A.

the chamber, and corrections of questionable reliability must be made. The second measurement method is free of this defect, since positive ions resulting from the charge transfer to the beam may be collected along a well-defined length of the ion beam within the enclosure where the target-gas density is constant. In using this latter method, great care must be taken in order to avoid measuring spurious currents. This will be discussed in Sec. II C.

II. EXPERIMENTAL METHOD

A. Beam Preparation

Figure 1 is a schematic diagram of the experimental apparatus. The ion source is a duoplasmatron.[1] Electrons emitted from the filament F are accelerated to the intermediate electrode I and pass through a hole to enter the plasma region between I and the anode A. The plasma is maintained by a strong magnetic field produced by the windings W in the region between the tip of I and the anode A. Ions are extracted through a 0.005-in. hole in A by the extractor E. The ions are focused by the einzel lens L, drift through the tube T_1, are further accelerated or deaccelerated at the junction of T_1 and T_2, and are mass selected by the 20° magnet. The ion beam then passes into the 15-in.-diam pumping chamber which is pumped by a 9-in. diffusion pump through a 9-in. liquid-nitrogen trap. The beam is then collimated by the 0.075-in. apertures A_1 and A_4 such that no ions may strike the 0.150-in. entrance aperture of the collision chamber. The apertures A_1 and A_4 are operated at +300 V in order to recollect electrons ejected from them by the beam. The grounded apertures A_2 and A_3 are 0.120 in. in diameter and give additional shielding against electrons ejected from the aperture A_1. The grounded aperture A_5 prevents electric field lines which leave the aperture A_4 from penetrating into the collision chamber through the entrance aperture.

B. Collision Chamber

Upon entering the collision chamber, the ion beam passes between three pairs of ion collection plates before entering the Faraday cage composed of the electrode P_4' and the plate P_4. Electrode P_4' is operated at − 55.5 V (see Fig. 2 for electrical connections), while electrode P_4 is operated at +55.5 V. Electrons ejected from P_4' by the beam are collected by electrode P_4 because of the electric field between P_4' and P_4. The sides of the Faraday cage are closed by plates connected to P_4' although this cannot be seen from the drawing. Only the end facing the incoming beam is open. The first pair of plates seen by the beam upon en-

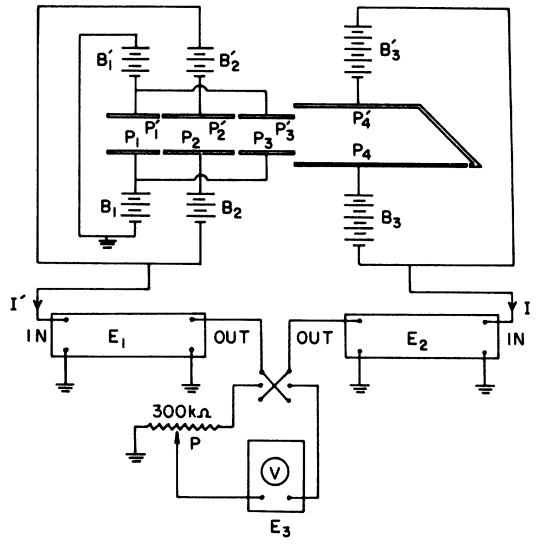

FIG. 2. Schematic diagram of electrical circuitry used in the measurements. E_1 and E_2 are Keithley No. 610B electrometers used in the unity gain mode. E_3 is a Keithley No. 600A battery-operated electrometer which is operated as a voltmeter and serves as a null indicator. The reversing switch allows a fraction of the output of the electrometer having the greater output to be balanced against the output of the other electrometer by adjusting the ten-turn potentiometer P.

tering the collision chamber P_1-P_1' and the third pair P_3-P_3' act as sets of guard plates which establish uniform conditions along the actual collection region defined by the pair of plates P_2-P_2'. The plates P_1', P_2', and P_3' are operated at -32.19 V, while the plates P_1, P_2, and P_3 are operated at $+32.19$ V. The separation between the plates is 0.450 in. The first set of guard plates extends along the beam a distance $L_1 = 0.750$ in., the collection plates extend a distance $L_2 = 1.000$ in., and the second set of guard plates extends a distance $L_3 = 0.800$ in. The Faraday cage is 2.500 in. long. The potential across the Faraday-cage electrodes is chosen so that the equipotential planes from the Faraday cage extend directly into the second set of guard plates P_3-P_3'.

The plates P_1, P_2, and P_3 were fabricated from a standard 1-in.-wide glass microscope slide. A layer of gold was deposited on one side of a slide by vacuum evaporation. The slide was then placed in a specially machined jig and two scratches were made across the slide with a diamond needle to divide the gold layer into three regions. A second slide was prepared in the same jig to produce the plates P_1', P_2', and P_3'. The lengths of the plates were checked with a traveling microscope. The Faraday cage was fabricated from 0.020-in.-thick nickel.

C. Theory of Experiment

The helium ions which become neutralized by the charge transfer process are deflected less than a degree[2] on the average, while the slow ions recoil with low energy approximately perpendicular to the beam, and are easily collected on the collection and guard plates. The determination that the potential between the plates was adequate to collect all ions produced was made by measuring the collected current as a function of this potential. Target atoms are ionized by the ion beam in the collision region, but this does not produce a current to ground through the electrometer E_1 shown in Fig. 2, but only a circulating current between the two plates P_2 and P_2'. Electrons ejected from the collection plates by the impact of ions or electrons do not produce a current through E_1 for the same reason. The Faraday-cage current passing through electrometer E_2 represents the unneutralized fraction of the ion beam leaving the second set of guard plates. The ratio of the ion current I' (to the collection plates) to the Faraday-cage current I is obtained by balancing the output of one electrometer against a fraction of the output of the other electrometer by the precision potentiometer P. That this ratio $R = I'/I$ yields the total charge transfer cross section is seen as follows. (The effect of the residual impurity gas atoms is momentarily ignored.) Designate by I_0 the part of the beam remaining unneutralized upon passage from the first set of guard plates to the collection region. At the end of the collection region it will be attenuated to $I_0 e^{-n\sigma L_2}$. At the end of the second set of guard plates it will be attenuated to $I_0 e^{-n\sigma(L_2+L_3)}$, and this is the current I measured in the Faraday cage. The measured ratio R is thus independent of I_0, and is given by

$$R = \frac{I'}{I} = \frac{1 - \exp(-n\sigma L_2)}{\exp[-n\sigma(L_2 + L_3)]} \quad . \tag{1}$$

This equation is easily solved by numerical methods for σ, since the lengths L_2 and L_3 are known, and n (the number of target gas atoms per cm^3) can be inferred from measurements of the pressure and

TABLE I. Values of the total electron transfer cross section for ground-state He^+ on ground-state He at an ion energy of 8 keV, and determined at various pressures. The average of these values is 6.12×10^{-16} cm^2.

Pressure (10^{-4} Torr)	Cross section (10^{-16} cm^2)
1.096	6.144
2.466	6.090
7.947	6.112
10.69	6.183
17.98	6.095

temperature in the collision chamber. It is worth noting that the result is independent of the amount of charge transfer taking place in the vicinity of the entrance aperture where the pressure is unknown. The nonuniformity of the electric field at the entrance of the first set of guard plates also does not affect the results.

The effect of residual gas in the collision chamber was ignored in deriving Eq. (1), which thus gives the result $R = 0$ for $n = 0$. But since there is always some residual background gas in the chamber due to outgassing and leakage, a nonvanishing value of I' was observed with no target gas admitted to the collision chamber. The corrected form of Eq. (1) is

$$R = \frac{I'}{I} = \frac{1 - \exp(-n\sigma L_2 - n_b\sigma_b L_2)}{\exp[(-n\sigma - n_b\sigma_b)(L_2 + L_3)]} \quad , \tag{2}$$

where n_b is the number density of background atoms and molecules and σ_b is the average effective cross section of the background gas. Since we are mainly interested in the product of these latter two quantities, we define $c = n_b\sigma_b$, which is a constant for a given energy (see Sec. II E). If the value of R is observed when no target gas is admitted to the chamber, the value of c is easily obtained from Eq. (2), which reduces to $R_0 = e^{c(L_2+L_3)} - e^{cL_3}$. When the value of c so determined is inserted in Eq. (2), the final form of our working equation is obtained:

$$R = \frac{1 - \exp[-(n\sigma + c)L_2]}{\exp[-(n\sigma + c)(L_2 + L_3)]} \quad . \tag{3}$$

The cross section is determined by admitting target gas at a known pressure to the chamber and observing R. The number density of target atoms is obtained from the observed pressure, after subtraction of the background gas pressure.

Measurements were made at 1-keV intervals in the energy range 2–22 keV. At each energy, measurements were made at five pressures in the range 1.0×10^{-4} to 1.8×10^{-3} Torr. The results at 8 keV are given in Table I. It is seen that the values obtained for the cross section are nearly independent of the target-gas pressure used, as they should be. The value of the cross section at 8 keV is obtained by averaging these five results.

A more sophisticated way to handle the data is to consider σ to be an adjustable parameter in Eq. (3), which is to be fitted to the data (consisting of observed values of the current ratio at various pressures). In this way, a value of σ is found which minimizes the sums of the squares of the fractional deviations $\Delta R/R$ of the data points from a curve of the form of Eq. (3). Since Eq. (3) does not give a straight line, the minimization must be carried out with a nonlinear least-squares routine. The reason that the *fractional deviations* from the fitted curve of the observed values of R were used, rather than the deviations themselves, was that the percentage error of R was approximately independent of R in the pressure range used. Minimization of the squares of the deviations themselves would have resulted in undue emphasis on the high-pressure points at the expense of the low-pressure points. The fit to the data at 8 keV is shown in Fig. 3, and the resulting value of σ was thus determined to be 6.13×10^{-16} cm^2. The percentage deviation of the fitted curve from the data points was typically 1% or less at all energies. The results of Table II were obtained by this method.

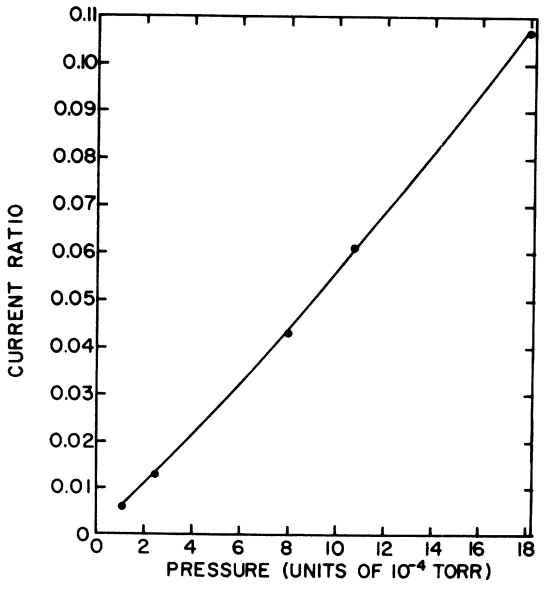

FIG. 3. Observed ratio of the collected ion current I' to the Faraday-cage current I is plotted against the pressure in the collision chamber. Cross section is obtained by fitting Eq. (3) to these data while treating σ as an adjustable parameter. Fitting was carried out so as to minimize the sum of the squares of the percentage deviations. Curve shown is the fit to the data, which were taken at 8 keV.

D. Pressure Measurement

The pressure was measured continuously with a National Research Corporation ionization gauge (type No. 518) operated at an emission of 2 mA. The gauge was connected to the system by a 24-in.-long section of glass tubing bent into a corkscrew shape and painted flat black on the exterior. This was found necessary in order to completely eliminate measurable effects due to photons and electrons from the gauge entering the collision chamber This tubing undoubtedly resulted in a pressure drop between the entrance of the glass tube and the gauge bulb, due to ion pumping in the gauge. This did not result in an error, however, since the glass tubing

TABLE II. Total electron transfer cross section for ground-state He^+ on ground-state He. The accuracy is estimated to be ±5%.

Energy (keV)	Cross Section (10^{-16} cm^2)
2	8.46
3	7.78
4	7.29
5	6.94
6	6.58
7	6.33
8	6.13
9	5.96
10	5.81
11	5.69
12	5.56
13	5.45
14	5.36
15	5.26
16	5.15
17	5.05
18	4.97
19	4.89
20	4.81
21	4.74
22	4.63

was permanently attached to the gauge, and was used during calibration, as described below. The gauge was calibrated immediately after the measurements were completed. The calibration was carried out against a Consolidated Vacuum Corporation McLeod gauge (type No. GM 110) which was operated in an upright freezer at a temperature of − 20 °C (see Fig. 4). The refrigeration was necessary to eliminate the error associated with the pumping effect[3−11] due to streaming of mercury vapor. Corrections were made for thermal transpiration due to the reduced temperature at the McLeod gauge, and for the change in the density of mercury arising from the cooling. The calibration was made at seven pressure points in the range 1.0×10^{-4} to 2.7×10^{-3} Torr. The heights of the mercury columns were read with a cathetometer through a glass window inserted in the door of the freezer. Provision was made for tapping the mercury columns without opening the door of the freezer. At each of the seven pressure points, 20 measurements were made, 10 with the mercury rising in the capillaries, and 10 with the mercury falling. These measurements were made with the gas compressed by various amounts in the capillary tube, in order to average out possible variations in the diameters of the capillary tubes, and variations of surface conditions within the capillary tubes. The McLeod gauge was initially baked at 300 °C. The results at each pressure point were averaged, and the seven pressure points were fitted to a straight line using a weighted least-squares code. The slope of this line gave the calibration constant. The standard deviation of the resultant calibration constant was 0.5% of its value, which is below the 1% volumetric error of the McLeod gauge quoted by the manufacturer.

E. Error Discussion

The possibility of excited ions existing in the beam was traced by variation of the voltage in the source between the filament and anode. Variation between 100 and 500 V produced no measurable variation in the cross section. This is to be expected since the metastable $^2S_{1/2}$ state is separated from the allowed $^2P_{1/2}$ state only by the energy of the Lamb shift. Even the small electric field seen in the rest frame of the ion as it passes through the magnetic field of the mass-selector magnet is adequate to produce sufficient mixing of these two states to quench any metastable ions in the beam.

The intensity of the ion beam used for this experiment varied between 1 and 3 μA depending on the energy. The background pressure in the collision chamber was about 10^{-6} Torr after several days of baking in preparation for a run. A background-pressure rise amounting to as much as 4×10^{-8} Torr was observed when the beam was passed into the collision chamber, and was found to depend on

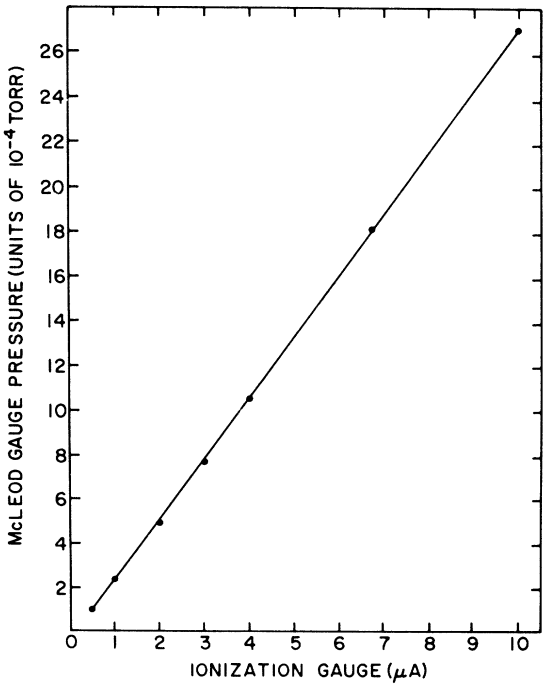

FIG. 4. Calibration of the ionization gauge against the McLeod gauge. Each point was obtained by averaging twenty measurements taken with varying conditions in the McLeod gauge.

the amount of energy delivered to the Faraday cage by the beam. This was ascribed to outgassing of the Faraday cage due to heating by the beam. This effect makes the measured cross sections slightly dependent on the beam intensity, if not properly compensated. One way to compensate for it is to measure the constant c in Eq. (3) as a function of the beam intensity. Such a set of measurements must be made at each energy, however, and this is very inconvenient. A better way to compensate for this effect is simply to maintain the beam intensity constant for all measurements at a given energy, so that the value of c does not change. When this was done, the measured cross sections were found to be unchanged when the entire procedure was carried out at a different beam intensity.

The background pressure in the pumping chamber was about 2×10^{-7} Torr, and did not rise above 10^{-6} Torr at the highest pressures used in the collision chamber.

In order to ascertain the voltage between the collection plates required to collect the electrons and slow ions, a saturation curve measurement was carried out. The apparatus used for this consisted of a set of six batteries fitted with voltage-dividing potentiometers and mounted on a Lucite plate. This allowed the values of the six batteries shown in Fig. 2 to be varied. Saturation was found to be essentially complete when the voltage between the collection plates was set at 40 V. This test apparatus was not used during the actual cross-section measurements, since it was found that a 90 V dry

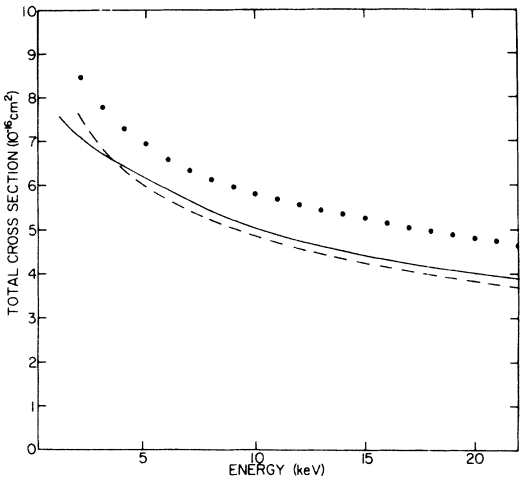

FIG. 5. Total electron transfer cross sections between ground-state He^+ and ground-state He. The measured values are given by the points. The broken curve was calculated from the theory of Rapp and Francis, (Ref. 12), while the solid curve is the calculation of Sural *et al.* (Ref. 13).

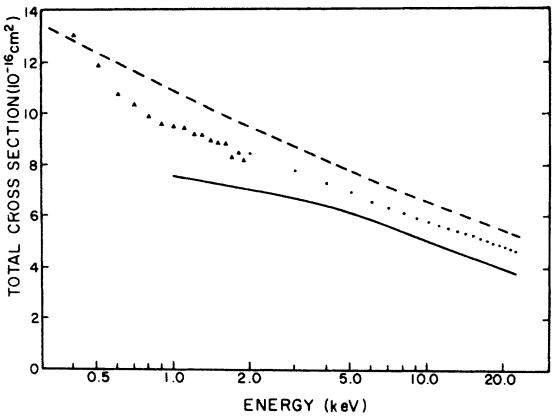

FIG. 6. Total electron transfer cross sections between ground-state He^+ and ground-state He. Points give the measured values of the present experiment. Triangles are the data points of Nagy *et al.* (Ref. 2) normalized to the value obtained in the present experiment at 2 keV. Broken curve is the calculation of Moiseiwitsch (Ref. 14). Solid curve is the calculation of Sural *et al.* (Ref. 13).

cell under a load of 1 mA has a voltage which is constantly varying with time by as much as 0.1 V. The batteries used for the actual measurements were operated without load, and were found to maintain a potential constant to within a few mV over a period of several days. The choice of 64.38 V for the potential between the collecting plates resulted from the use of 30-V batteries matched to within 10 mV of 32.19 V, so that the collection and guard plates were maintained at the same potential to within this 10 mV. A test of the sensitivity of the collected current to the difference between the guard and collection fields showed that a mismatch of 30 mV would produce an error of less than 0.1%.

The electrometers E_1 and E_2 were operated in the 100% inverse feedback mode, so that they introduced potential variations of only about 1 mV on the collection plates and Faraday cage. The outputs of the electrometers were taken directly from their internal feedback resistors so that loading effects due to the introduction of the precision potentiometer P was automatically compensated to within 0.01%. The only appreciable error in the electrical measurements was that due to the error in the high-megohm resistors of the input circuits of the electrometers. Since only the ratio of currents is of interest in deriving the cross section, it was possible to reduce this error to about 1% by making comparisons of the resistors in the two electrometers. The precision potentiometer P had a resistance of 300 kΩ and a linearity of 0.04%.

A second set of collection and guard plates was constructed to test for possible dependence of the measured cross section on the length of the collec-

tion region. The collection region for this second set was 0.5 in. long, and was found to give the same result as the first set to within 0.5%.

In order to test for asymmetrical scattering of the ion beam onto the collection plates, or for any other asymmetry effects, the roles of the electron and slow-ion collection plates were electrically interchanged (along with the corresponding guard and Faraday-cage elements). Any such effects present were too small to be observed.

The possible loss of electrons or ions from the region between the plates was checked. A grounded grid was mounted along the open region between the plates. Outside this grid was mounted an insulated collecting electrode. With an ion-beam intensity of 1.45 μA at an energy of 15 keV, and with a target-gas pressure of 1.8×10^{-3} Torr, a positive current of 2×10^{-10} A was observed on this electrode. The loss of twice this current (allowing for the opening on the opposite side) from the collection region is to be compared to the collected current of 0.17 μA, and is seen to give an error of approximately 0.2%.

The helium-gas purity was better than 99.99%. It is believed that the over-all accuracy of the measurements is 5% provided unknown systematic errors[7] in the McLeod gauge do not exceed 3%.

III. RESULTS AND DISCUSSION

The measured cross sections are given in Table I. These cross sections are for electron capture by the helium ion to form all possible excited states as well as the ground state. The resonant transfer to the ground state of the ion gives the major contribution, however. Figure 5 shows a comparison of the present results with the crude theory of Rapp and Francis[12] which includes only the resonant capture to the ground state, and which assumes a hydrogenlike wave function. This theoretical curve falls below our measured points. Also shown is the recent and elaborate calculation of Sural, Mukherjee, and Sil[13] which includes the possibility of capture into the $2\,^1S$ and $2\,^3S$ states of helium, as well as resonant capture into the ground state. This calculation also included the possibility of excitation of the target atom to the $2\,^1S$ or $2\,^3S$ states without electron transfer taking place. This curve also falls below our data, but agrees well with the observed energy dependence.

Recent measurements in the energy range 400–2000 eV have been made by Nagy, Savola, and Pollack[2] using the beam-attenuation method. Since their scattering volume was not well defined, the absolute magnitude of their results may be off by as much as 12%, while they believe their relative results are accurate to $\pm 6\%$. Since we believe the present results have a better absolute accuracy, we have renormalized their results to ours at the 2-keV point where the two experiments overlap. This resulted in a reduction of the magnitude of their results by 10.6%. In Fig. 6 the renormalized data of Nagy *et al.* are shown along with the present measurements plotted on a logarithmic energy scale to cover the extended energy range. The theoretical curve of Sural *et al.* is also shown. Another theoretical curve calculated by Moiseiwitsch[14] using the two-state approximation and a two-parameter helium wave function [this is the calculation Moiseiwitsch designates by (iii) in his paper] is shown. There is a considerable disagreement between the two theoretical curves. The data points fall between the two theoretical curves, except for the 400-eV point of Nagy *et al.* At energies above about 6 keV both theoretical curves depend on energy in approximately the same way as the present data. Below 6 keV the calculation of Moiseiwitsch agrees better with the energy dependence of the present measurements. In the energy range 1–2 keV the Moiseiwitsch curve has approximately the same energy dependence as the data points of Nagy *et al.* Below 1 keV the Moiseiwitsch curve deviates from the energy dependence of the data of Nagy *et al.*

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