

## Electron-helium absolute total scattering cross sections from 0.5 to 50 eV

R. E. Kennerly\* and R. A. Bonham

Chemistry Department, Indiana University, Bloomington, Indiana 47401

(Received 4 November 1977)

Absolute total cross sections for electron scattering from He have been measured over the energy range from 1 to 50 eV with an accuracy of 3% or better. A novel method employing time-of-flight monochromatization and energy determination of electrons from a pulsed secondary-emission source in transmission through a gas cell has been used. This method is contrasted with conventional ones and a comprehensive error discussion is given. The reported results are then compared to existing experimental and theoretical results. Discrepancies in existing experimental data are resolved and good agreement is obtained with some recent theoretical results.

### I. INTRODUCTION

Electron-helium scattering phenomena have played a central role in the development of the quantum theory of scattering and of many-body systems in general because the helium atom is the simplest which can be easily handled experimentally. The total cross section (TCS), i.e., the total (elastic plus inelastic) differential cross section integrated over all angles of scattering, was the first cross section to be measured quantitatively due to its practical importance and the relative simplicity of transmission-type measurements. These absolute values are extensively used to assess the accuracy of various theoretical treatments since they have been generally acknowledged to be the most accurate available. Electron-helium cross sections also continue to be heavily used experimentally for calibration and normalization purposes. Below the excitation threshold (19.8 eV) the TCS is especially important because it is purely elastic.

Despite the importance of absolute electron-helium TCS, there have been few direct measurements. The only published results since 1932 are those of Golden and Bandel<sup>1</sup> from 0.3 to 28 eV using an improved Ramsauer method,<sup>2</sup> and those of Blaauw *et al.*<sup>3</sup> from 16 to 750 eV. These measurements disagree by about 15% in the overlapping energy range. The earlier measurements,<sup>2,4-8</sup> all of which employed the Ramsauer method, are in serious disagreement and it is impossible to adequately assess their reliability. The situation up to 1971 has been discussed in detail by Bederson and Kieffer<sup>9</sup> in their excellent review. Their conclusion, briefly stated, was that the TCS as derived from single-scattering transmission experiments (which all employed the basic Ramsauer method) was known only to about (10-15)% in the energy range to 30 eV, much larger than the 3% error limits originally stated in Ref. 1.

There are several relatively recent experimental results which bear indirectly on the electron-helium total cross section. The momentum-transfer cross section (MCS) of Crompton *et al.*<sup>10</sup> from 0.008 to 6 eV inferred from the measurement of electron drift velocities in a swarm was judged by Bederson and Kieffer<sup>9</sup> to be accurate to within the 2% (5% above 3 eV) stated uncertainty limits. This work has recently been extended to 12 eV by Milloy and Crompton.<sup>11</sup> Conversions between TCS and MCS can be made only with a knowledge of the relative differential cross section or the scattering phase shifts. These can be gotten from recent theoretical calculations (discussed below) and such comparisons between TCS and MCS are suggestive and informative, if not conclusive. Extensive measurements have recently been made of the relative differential cross sections from 2 to 19 eV by Andrick and Bitsch.<sup>12</sup> Absolute TCS's were inferred by first fitting the observed relative differential cross sections with the partial wave formula to determine phase shifts, then computing the TCS from the phase shifts. The results of both of these indirect methods are in general disagreement by about 10% or more with the results of Golden and Bandel.<sup>1</sup> The Andrick and Bitsch results, however, are in good agreement with those of Blaauw *et al.*<sup>3</sup> in the narrow overlapping range (16-19 eV).

The theoretical situation regarding low-energy electron-atom scattering has been reviewed recently by Nesbet.<sup>13</sup> Recent calculations<sup>14-17</sup> are in good agreement, except in the range below several eV and above about 15 eV, where these TCS's differ by up to 10%. These theoretical TCS's are in general smaller than those from Andrick and Bitsch<sup>12</sup> and larger than those of Golden and Bandel.<sup>1</sup>

The available direct experimental TCS's are thus inadequate for discriminating among the results of recent theoretical advances over large portions of the low-energy range. This situation

is particularly lamentable in the case of He, the traditional proving ground for both experimental and theoretical methods in electron scattering. The purpose of this paper is to present new results for the electron-He TCS from 0.5 to 50 eV measured using a new transmission method involving time-of-flight monochromatization of electrons from a pulsed secondary-emission source in free flight through a gas cell. In the following sections, the method will be described and its advantages over other methods will be discussed. Finally, the present results will be presented and compared to previously existing results with the aim of resolving the discrepancies mentioned above.

## II. APPARATUS

A schematic representation of the apparatus is shown in Fig. 1. The basis of the capability of this apparatus for measuring TCS's is its utility for measuring the energy distributions of electrons ejected from gaseous or solid targets by electron impact. This aspect has been described in detail elsewhere<sup>18-20</sup> and only an abbreviated description will be given here. The modifications required for TCS measurement are few and simple, being basically the placement of a suitable solid in the target position, an aperture in front of the detector to convert the free-flight tube to a gas cell, and the addition of equipment for gas handling and

pressure measurement.

In order to determine the time-of-flight (TOF) of electrons ejected from the target a pulsed incident electron beam must be used (in the absence of coincidence methods). The pulses were produced by sweeping a dc beam across a 1 mm aperture. In order to produce secondary electrons with a wide range of energies, a beam energy of several keV was used. The beam was swept by applying a rectangular voltage pulse of about 10 V and 30 nsec duration to one plate of a pair of deflection plates. The dc beam was swept across the aperture by the voltage-pulse rise; in order to prevent the beam from recrossing the aperture with the fall, the pulse was delayed by 15 nsec and applied to one plate of another pair, orthogonal to the first pair. The beam thus traced out a rectangle with one side centered on the aperture. This aperture system, shown in Ref. 19, was designed to be an efficient trap for the stationary dc beam between pulses. The collected current ( $\sim 1 \mu\text{A}$ ) was passed to an external current measuring device. The duration of the emitted electron pulses can be varied by changing the risetime of the voltage pulses; durations from 0.1 to 1 nsec were routinely achieved using voltage-pulse risetimes from 0.7 to 7 nsec. Repetition rates of several hundred kHz were used.

The electron pulses impinge on a solid target to generate the desired secondary emission. The

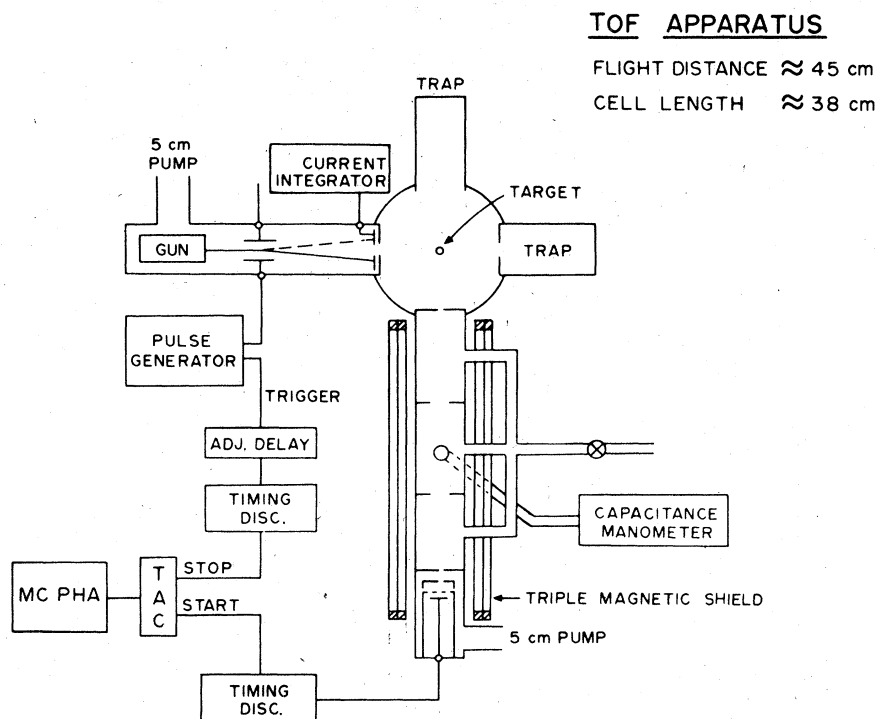


FIG. 1. Schematic representation of the electron time-of-flight apparatus as used for the measurement of total scattering cross sections.

target was a 0.4-mm-diameter platinum tube (perpendicular to the plane of Fig. 1) coated with either colloidal graphite or CsI. The unscattered beam was trapped. In addition there was a trap opposite the free-flight tube to minimize the effect of backscattering and secondary emission from chamber surfaces in view of the detector.

Secondary electrons entered the 40-mm-i.d. flight tube through a 1 or 2 mm circular entrance aperture 38 mm from the source. At the detector end of the tube was a pair of circular apertures to be called the exit and the skimmer apertures. The exit aperture, the one nearest the source, was 380 mm from the entrance aperture and terminated the absorption cell. Openings from 2 to 5 mm were used here. The purpose of the skimmer aperture, 20 mm from the exit aperture with an opening about 25% larger, was to provide better discrimination against small-angle scattering in the gas cell. There were also two large apertures within the gas cell to serve as baffles to restrict the paths of electrons scattered within the cell. The inner diameters were 11 and 19 mm and they were equally spaced between the entrance and exit apertures. Free flight was terminated 445 mm from the source and 6 mm from the skimmer by a fine wire mesh (20- $\mu$ m wire, 81% optically transparent) behind which was the positively biased detector.

All parts of the apparatus in view of the electrons were made from aluminum. All interior surfaces were coated with colloidal graphite applied as an aerosol to suppress surface potential variations, secondary emission, and backscattering. Three orthogonal pairs of Helmholtz coils 2 m in diameter were used to reduce the ambient magnetic field in the region of the target chamber to about 1 mG. The drift and detector regions were enclosed by a three-layered magnetic shield (120 mm i.d.), reducing the field there to <1 mG.

A dual-chevron channel-electron-multiplier array with 18-mm-diameter active area was used as the detector, its advantages being excellent timing characteristics and a flat active area, which when placed perpendicular to the electron trajectories introduced no path length uncertainty in the time-to-energy conversion. The fast-timing electronics were conventional. A time-to-amplitude converter (TAC) was started by an amplified detector pulse and stopped by a delayed trigger pulse from the pulse generator. "Constant-fraction" timing discriminators were used in both lines. The TAC produced an output pulse with amplitude proportional to the time difference between start and stop pulses. A multichannel pulse-height analyser (MCPHA) then accumulated a spectrum representing the number of detected events versus the TOF.

Overall timing resolution of 170 psec has been achieved as measured by the time width of the elastic peak for 2-keV electron scattering from Ar (introduced from a nozzle at the target location). Over a period of hours various instabilities broaden this to about 300 psec.

Although high resolution was not the aim of the present study, the capabilities are quite good at low energies, as shown in Ref. 19. Note that in the present case the path-length uncertainty due to the target radius is  $\frac{1}{3}$  that for gas-beam targets<sup>19</sup> and is not limiting at any energy. A possible limitation at low energy is that due to the effect of potential variations along the flight path due to surface-potential variations. For an arrangement like the present case, this contribution is probably less than 5 meV.<sup>21-23</sup>

Gas was admitted to the cell at three places along its length through aluminum tubes (4 mm i.d.) passing through the magnetic shields. Flow was controlled by a precision metering valve. Cell pressures used ranged from 1.3 to 7 mTorr and were measured by a precision capacitance manometer<sup>24</sup> connected to the cell by another aluminum tube located 90° from the middle admittance tube. The capacitance-manometer output was measured with a precision differential voltmeter. Gas escaping the cell through the entrance and exit apertures was removed by diffusion-pumping systems of 500 l/sec (air) at the target region and 25 l/sec at the detector region. Base pressures are about 2  $\mu$ Torr. With gas in the cell, pressures ranged from 3 to 5  $\mu$ Torr in the target chamber and 10 to 30  $\mu$ Torr at the detector. That the gas pressure within the cell was uniform throughout can be argued on the basis of the large ratios of the cross-sectional area of the cell and gas entrance ports to the aperture area. A convincing demonstration was that the observed TCS's showed no systematic change as the entrance and exit aperture area was changed by a factor of 6.

### III. PROCEDURE

The basis of total-cross-section measurement by transmission techniques is the Beer-Lambert relation expressing the attenuation of a particle flux by a scattering medium,

$$I(x) = I_0 e^{-\sigma_T n x} \quad \text{or} \quad \sigma_T = \frac{1}{n x} \ln \left( \frac{I_0}{I(x)} \right), \quad (1)$$

where  $I(x)$  is the intensity remaining after traversing a length  $x$  of medium whose number density is  $n$ , and  $I_0$  is the intensity in the absence of the scattering medium. The application of this expression to the accurate determination of scattering cross sections has been complicated in previous low-en-

ergy methods, i.e., the Ramsauer method and its variants, by the impossibility of determining  $I_0$  in the expression above. (This problem has been treated in some detail in Refs. 1 and 9.) As a result, TCS's have been inferred instead from the slope of a plot of the logarithm of the ratios of the experimentally available currents best approximating  $I_0$  and  $I(x)$  versus the number density of the scattering gas. (Linearity of this function is a criterion for data acceptability.) The  $y$  intercept of such a plot is not zero, demonstrating that these approximations to the ideal  $I_0$  and  $I(x)$  in the above expressions are not equal in the limit  $n \rightarrow 0$ . This requirement of performing the measurement over a wide range of  $n$  has rendered the Ramsauer method rather tedious, as is indicated by the infrequency of the attempts outlined in Sec. I. This is amplified by the fact that the electron energy is not continuously variable, but must be changed by careful adjustment of the accelerating voltage and the transverse magnetic field.<sup>1</sup> A more serious problem, as it relates to the accuracy of the inferred TCS, is the possible pressure dependence of the residual "scattered" current which is not due to the direct scattering within the scattering chamber. At  $n=0$ , this residual is simply the intercept of the above-described plot. If this residual is not constant, then the TCS inferred from the slope will be in error. Such a pressure dependence could result from the presence of gas at the hot-cathode electron source due to lack of or inadequate differential pumping of the cathode region, or space-charge and surface-charging effects with consequent electron-optical effects. That such effects do occur to some extent would appear certain, as is indicated by the serious disagreement among measurements of this type, to all of which the linearity criterion was applied.

The present TOF method, on the other hand, is to a great extent free from these problems. There are several reasons for this advantage. The secondary-electron source is at room temperature and is much less sensitive to the presence of gas than are thermionic cathodes. The target chamber remained at very low pressure even with gas in the flight tube. The pressure change with and without gas was approximately equal to the background pressure, about  $2 \mu\text{Torr}$ . This degree of differential pumping is much larger than that used in the experiments of Golden and Bandel<sup>1</sup> and in earlier versions of the Ramsauer method, which employed no differential pumping at all. The electron current on the flight path was at most only about  $10^{-14}$  A, corresponding to an average of much less than one electron in flight at any time. Space-charge and surface-charging effects were consequently negligible. In addition to the above, there are sev-

eral other advantages of the present TOF method which will be discussed in later sections.

The experimental procedure followed was to record in half the MCPHA memory the TOF distribution of the secondary electrons transmitted to the detector without gas, then in the other half with gas in the cell. These two segments, hereafter called "reference" and "attenuation," were scaled using the product of the data-collection times and the average primary-beam currents delivered to the pulsing aperture during each segment. The time used was the MCPHA's internally generated and stored "live" time, that is, the actual clock time elapsed minus the time the analyzer was engaged and not accepting input pulses from the TAC. This was the true system "dead time" because the MCPHA dead time was much larger than that of any of the other components. The average beam current delivered was determined by collection of the total current, digitization, and counting during the course of the segment. As added insurance, the primary-beam current was stabilized manually to within about 2%. The scale factors derived in this way differed from those derived using "live" time only typically by about 0.15%, indicating that the averaged current was very stable. The positional stability of the primary beam was excellent. The drift in position over a period of about 5 h (the time required for the TCS measurement) was such that the transmitted fraction of the beam (about 95%) usually had changed by several percent or less.

Rather than use one long reference segment and one long attenuation segment, the 5-h measurement period was divided into short (10–15 min) periods in which the attenuation and reference segments were alternated. The advantage of this is that potentially deleterious "drifts" are greatly reduced. For example, over a period of several hours the energy distribution of the secondary emission was observed to change by up to 20% at low energies under the influence of electron impact from the primary beam pulses. Another problem for extended segments would have been zero drift in the capacitance manometer. By using short alternating segments such problems are eliminated.

The pressure reading and "zero" reading were recorded at the beginning and end of each attenuation segment, and the pressure for that segment was assumed to be the zero-corrected average. The changes in the zero and in the pressure during the segment were typically less than 1%, as was the change in the pressure during the individual segments over the course of the entire measurement. The average of the pressures for the individual attenuation segments weighted by the seg-

ment "live" time and average beam current (as above) was the final value used for data reduction.

Pressure was converted to number density using the expression

$$n \text{ (cm}^{-3}\text{)} = [9.656 \times 10^{-13} P \text{ (Torr)} / T \text{ (}^\circ\text{K)}] 0.989.$$

The expression in brackets is derived from the ideal-gas law, deviations from which are negligible at the pressures used. The remaining factor is a correction for the effect of thermal transpiration<sup>25</sup> on the observed pressure readings. The head of the capacitance manometer was regulated at 49 °C, whereas the gas cell was at room temperature. Two measurements of this effect have been published<sup>26,27</sup> for capacitance-manometer heads similar to the one used here. Both imply that for pressures less than about 50 mTorr a small pressure-dependent correction factor is required, but that at about 10 mTorr and below the correction has reached a maximum and is constant. However, different values of this constant correction factor were found, 1.1%<sup>26</sup> and 2.6%,<sup>27</sup> contrasting with the 4.1% from  $(T/T_0)^{1/2}$  derived from simple kinetic theory for simplified systems.<sup>25</sup> These findings imply that the correction required depends on the geometrical details of the region over which the temperature change occurs. Here the former value was chosen because the arrangement was most similar to the present one, i.e., the head was installed as supplied from the manufacturer in the open air, whereas the latter value was derived using a head (the same model) enclosed in foam insulation which shifted and extended the region of temperature drop.

The TAC and MCPHA combination was calibrated using a precision time-difference generator. The 16.84-nsec TOF of the 2-keV elastic peak was used to establish the zero of the TOF scale for each experiment. Since the elastic peak was not resolved for scattering from the solid target, an Ar jet from a nozzle at the normal position of the solid target was used. This was done before and after each complete TCS measurement to check for shifts of the TOF scale. In the conversion by computer of TOF to energy, the accelerated flight from screen to detector was accounted for and relativistic kinematics were used.

The method detailed here is similar in concept to several previous ones which also employed TOF electron-velocity analysis. Baldwin and co-workers<sup>26,29</sup> used photoemission as a source of electrons for a transmission measurement aimed at TCS determination. This choice, however, limited their method to a small energy range (about 0.4–1.8 eV), poor energy and angular resolution, and very low count rates. Preliminary results were published for He and Ar,<sup>28</sup> as well as a more thor-

ough study of N<sub>2</sub>.<sup>29</sup> The work of Land and Raith<sup>22</sup> was not directed toward absolute cross-section measurements but rather to high-resolution resonance spectroscopy below about 0.5 eV. The electron intensity was concentrated in this range by directing the primary-beam pulse directly into the transmission cell which was very near cathode potential. Electrons in the cell thus had very low energy with a distribution dictated by the thermionic emission from the hot cathode of the gun, and were steered and focused with very weak magnetic fields. Resonance structure in O<sub>2</sub> and H<sub>2</sub> was studied, and work on other molecules is currently under way. A comprehensive review by Raith<sup>23</sup> of TOF scattering spectroscopy covering electron impact as well as other aspects may be consulted for a more complete bibliography of TOF methods in general.

#### IV. ERROR DISCUSSION

It is useful for the present discussion to distinguish the different types of possible error in this type of measurement. Broadly speaking, these are constant multiplicative errors (scale factors), constant additive errors (scale shifts), and non-constant errors or "shape" errors.

The first class is due principally to errors in either the density or the absorption length. Regarding the first, several studies<sup>24,26,27,30</sup> have found this type of capacitance manometer to be linear to better than 0.5% over the pressure range of this study. The "absolute" head was calibrated at low pressure to better than 0.2% by the manufacturer using deadweight testers. Combined errors from sources such as temperature changes, nonlinearity, and zero shifts were less than about 1%. Errors due to fluctuations were effectively reduced by the alternation of reference and attenuation segments, which ensured that the final pressure value used was the average of 30 to 40 individual determinations. The potential error associated with the choice of thermal transpiration correction is about +2%, -0.5% in the TCS. (Increasing the correction would increase the inferred cross sections.) Errors from nonequilibrium and related lack of knowledge of the attenuation length were judged less significant. The distance between the entrance and exit aperture planes was used as the absorption length. This is believed to be a very good approximation because of the large pressure ratio across the apertures (~100×), the relatively small size of the apertures, and large mean free path in the cell. Under these conditions, the end effects are small anyway, but, in addition, the depletion near the apertures inside the cell is equivalent to the enhancement outside, there being no net effect.<sup>31</sup> The ap-

erture separation was measured to better than 0.5 mm or 0.1%.

Constant additive errors result from inaccurate scaling of the reference and attenuation intensities. Such an error would appear in Eq. (1) as a constant attenuation factor, and thus as an additive constant to the TCS. Typically, scaling errors were less than 1% of the computed factor. This corresponds at 2 eV, for example, to a 0.2% TCS error at the highest pressure used and 0.9% at the lowest, and, at 50 eV, to 0.7% and 2.4%, respectively.

Variations in the secondary-electron distribution during a measurement, a possible source of "shape" errors, were minimized by the alternation of short reference and attenuation segments, as described above. To test the effectiveness of this procedure, "dry-run" measurements were performed in which all aspects of the normal procedure were followed, but without gas in the cell during the attenuation segments. Deviations from an "attenuation" of unity then indicated any residual noncancellation of source variations. The results can be summarized by saying that the rms value of the absolute error was fairly constant throughout the energy range, except for an increase below 1 eV. The resulting relative error in a TCS measurement was less than 0.5% for cell pressures around 2 mTorr, except at the extremes of the energy range, where it increased to about 1% at 0.5 and 2.5% at 50 eV. For higher pressure, the error is reduced proportionately.

As a check on gas-source effects, the measurement procedure was performed as above (with no gas in the cell) but with helium in the target region during the "attenuation" segments, at a pressure twice the highest during any of the actual measurements. Deviations from zero "attenuation" (above the level of those described in the previous paragraph) then indicated any modification of the secondary emission from the target caused by interaction of the effusing helium and the source. None whatsoever was observed.

Another potential source of "shape" error is that due to background subtraction. This was significant only at the lowest and the highest reported energies for each measurement, as can be seen from Fig. 2 which shows a typical unscaled TOF distribution for the reference intensity. Below 1 eV the TOF intensities can be seen to drop precipitously into the background. Background correction was by subtraction of the averaged intensity prior to TOF=0. This background decreased as the cell pressure increased, demonstrating that most scattered electrons were collected by the cell surfaces. Those undergoing many wall collisions were effectively uncorrelated with the primary beam pulse and contributed a constant background,

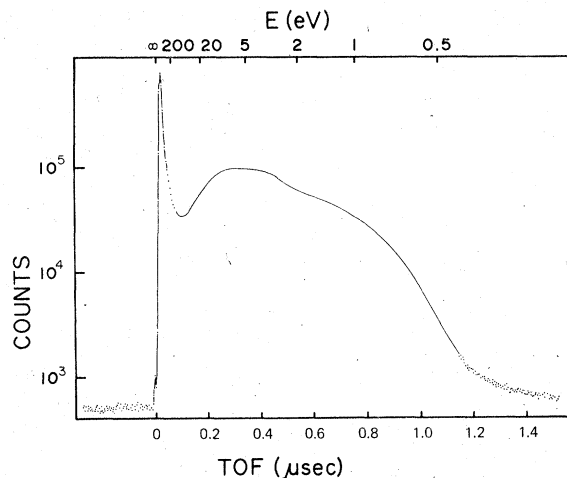


FIG. 2. Typical time-of-flight distribution (3.92 nsec/channel) of secondary emission from the graphite source. Energy in eV is given at the top. This particular distribution is the "reference" from measurement d in Table I and Fig. 3. The small peak near TOF=0 is due to electron-impact-induced photoemission.

which is correctly accounted for. The ratio of cell-wall area to exit-aperture area ( $\sim 10^4$ ) indicates that the probability of time-correlated electrons escaping was very small. A background uncertainty of 10% was used to infer TCS errors from this source. TCS values at energies for which the constant background exceeded 10% of the total intensity were not used. At this cutoff point (which was at somewhat different energies for each measurement), TCS error from this source was less than about 3%, decreasing precipitously with increasing energy to a roughly constant level of from 0.2% to 0.7% throughout the energy range, depending on the particular measurement.

A potential source of error at the highest energies reported here was that due to imperfect discrimination against very small angle scattering. A convenient quantity to characterize the small-angle discrimination of a transmission-type experiment is the percentage loss of total scattering for an isotropic angular distribution of scattering. No full treatment of this effect has been published for the Ramsauer method, but Golden and Bandel<sup>1</sup> have given the results of an analysis for a simplified situation with straight trajectories and no magnetic field, using the actual slit dimensions of their apparatus, as well as those of Ramsauer and Kollath,<sup>6</sup> and of Brode<sup>4</sup> and Normand<sup>7</sup> (who used the same apparatus). This purely geometric evaluation gave as percentages of forward and backward scattering not detected 3.6% and 1.6%, 3.9% and 3.9%, and 2.2% and 0%, respectively, for the above apparatuses. For the present case, the

TABLE I. Experimental conditions for the four measurements.

Curve	Secondary-emission source	Apertures (mm): entrance, exit, skimmer	Cell pressure (mTorr)	Helium pressure at source ( $\mu$ Torr)	Attenuation		Count rate (kHz)		Number of segments
					50 eV	2 eV	Reference	Attenuation	
a	CsI	2, 3, 4	2.0	4.0	1.5	4.5	3.5	1.9	23
b	graphite	2, 3, 4	1.4	2.8	1.3	3.0	4.3	2.7	19
c	graphite	1, 2, 3	7.1	3.5	4.3	180	1.7	0.67	13
d	graphite	2, 5, 6	1.3	2.5	1.3	2.6	3.3	1.8	19

above simplified situation actually corresponds to the real one, and a similar evaluation gives as percentages of forward scattering not counted 0.013%, 0.024%, and 0.055% for the three aperture sets given in Table I. (All backscattering is, of course, accounted for.) The tremendous improvement in the present case is due to the small aperture size relative to the cell length and the use of the skimmer aperture between the exit aperture and the detector. These numbers serve as a useful parametrization of forward-scattering sensitivity from a purely geometrical viewpoint, but for a meaningful evaluation of possible TCS error at higher energies, a more realistic differential cross section (DCS) is required. For this purpose, the first-Born-approximation values for He for elastic and inelastic scattering was used to estimate the missed forward scattering at 200 eV and above. Estimates for lower energies were then obtained by interpolation with the isotropic values valid at very low energies, giving at 50 eV from 0.2% to 0.8% for the three aperture sets, or about 15 times the isotropic values. (Similar increases for the Ramsauer-type apparatuses would appear to render them largely useless for reliable TCS at higher energy.) The accuracy of the Born total DCS at 200 eV is questionable but served to indicate the magnitude of the effect. These estimates were doubled for use in the total error estimation.

Error limits from the above sources were combined quadratically to give total uncertainties at selected electron energies for each of the four measurements reported in Sec. V.

The energy error is due principally to two sources, the potential difference between the target surface and the interior of the gas cell, and calibration error and nonlinearity in the time measurement. Due to the direct measurement of average electron velocity, the error from the former source is much less than the potential difference itself and was found to be 20 meV or less. Relative energy error from calibration was estimated to be about 1% or less at low energy, decreasing at

higher energy as  $1 - [E/(2 \text{ keV})]^{1/2}$  since the elastic TOF was used as a reference time.

The helium, Matheson "Ultra High Purity" grade with maximum impurities of 0.001% was used directly from the cylinder. Care was taken in the construction and operation of the gas-inlet system to maintain this purity.

## V. RESULTS

The TCS results from the four measurements (labeled a-d) are shown in Fig. 3. The experimental parameters for each are summarized in Table I. The energy range for each is from 50 eV to the energy at which the background exceeded

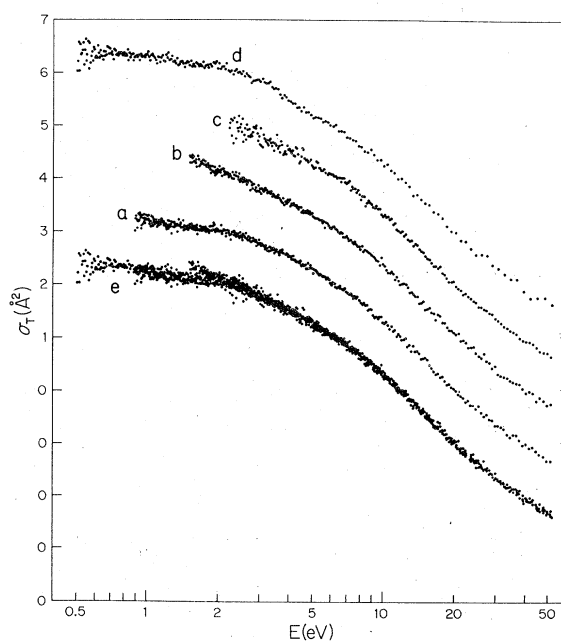


FIG. 3. The total cross sections from measurements a-d of Table I, plus a composite superimposing all four results e. The scale shown for  $\sigma_T$  applies to result d and the progressively lower zeros are those for results c, b, a, and e, respectively.

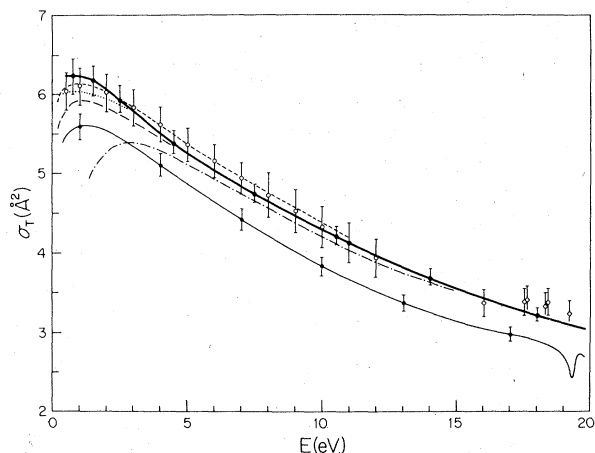


FIG. 4. The present results for the electron-He total cross section (heavy unbroken curve) compared to the best previous results from 0–20 eV. Note that the zero is suppressed on the ordinate. Other experimental results: Brüche *et al.* (Ref. 5) (dot-dashed curve), Golden and Bandel (Ref. 1) (thin unbroken curve), Blaauw *et al.* (Ref. 3) (diamonds), and Crompton *et al.* (Ref. 10) and Milloy and Crompton (Ref. 11) momentum-transfer cross section converted using phase shifts from Refs. 14 and 15 (open circles). Theoretical results: Sinfailam and Nesbet (Ref. 14) (long-dashed curve), Yarlagadda *et al.* (Ref. 15) (dotted curve), and Berrington and O'Malley (Ref. 17) (short-dashed curve).

10% of the total signal in the TOF distributions. Of particular interest in Table I are the wide ranges of the various experimental parameters in the different measurements. These results are compared in curve e of Fig. 3, which is the superposition of the four measurements. The excellent agreement among these results is good evidence that the present method is free of significant pressure-dependent systematic errors. The large range of exit-aperture solid angles (from  $2.2 \times 10^{-5}$  sr to  $1.4 \times 10^{-4}$  sr with respect to the entrance aperture) indicates that there is no significant error due to lack of sensitivity to small-angle scattering. This has been a significant but highly uncertain error source, even at relatively low energies (20 eV), in Ramsauer-type measurements.<sup>32</sup> The agreement between a, which employed a CsI secondary-emission source, and the others, which employed carbon, demonstrates the lack of dependence on the energy distribution of the electron source. Generally, the very solid overlap suggested that no significant errors from unknown sources were present.

The present *e*-He TCS is compared with other experimental and theoretical results in Figs. 4 and 5. For this purpose, the separate results (a–d) in Fig. 3 were smoothed and then averaged, each weighted by the inverse of its uncertainty at

TABLE II. Present results for the electron-He total cross section.

$E$ (eV)	$\sigma_T$ (Å <sup>2</sup> )	Error (%)	$E$ (eV)	$\sigma_T$ (Å <sup>2</sup> )	Error (%)
0.5	6.24	±5	12	3.96	+3, -2
1	6.23	±3	14	3.69	+3, -2
1.5	6.18	±3	16	3.43	+3, -2
2	6.06	+3, -2	18	3.22	+3, -2
2.5	5.92	+3, -2	20	3.03	+3, -2
3	5.78	+3, -2	22	2.86	+3, -2
4	5.50	+3, -2	24	2.71	+3, -2
5	5.25	+3, -2	26	2.57	+3, -2
6	5.04	+3, -2	28	2.44	+3, -2
7	4.83	+3, -2	30	2.36	+3, -2
8	4.64	+3, -2	35	2.14	+3, -2
9	4.46	+3, -2	40	1.95	+3, -2
10	4.30	+3, -2	45	1.81	+3, -2
			50	1.68	+3, -2

that energy, to yield a composite result. The composite error estimate was a similarly weighted average. This TCS is given in Table II for a sufficient number of energy values that the full curve can be reproduced with sufficient precision by simple interpolation.

TCS's in the 0–20 eV range are shown in Fig. 4. It should be pointed out that, with the presence of errors that are energy dependent, the shape of the actual TCS can only be inferred from the present results to lie within the error limits and to be smoothly varying. E.g., these results do not imply that the TCS is constant or nondecreasing below 1 eV.

Of the older work employing the Ramsauer meth-

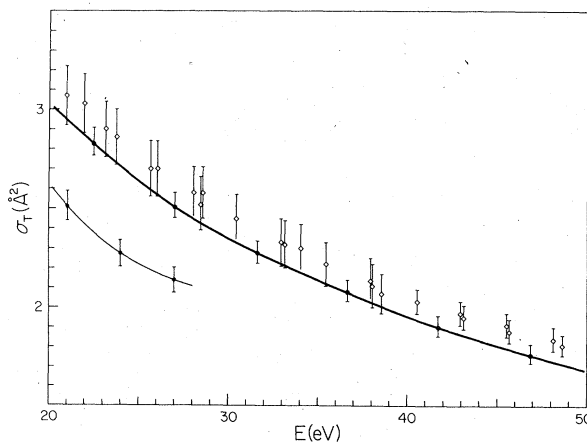


FIG. 5. The present results for the electron-He total cross section (heavy unbroken curve) from 20 to 50 eV compared to the best previous results. Note that the zero is suppressed on the ordinate. Golden and Bandel (Ref. 1) (thin unbroken curve), and Blaauw *et al.* (Ref. 3) (diamonds).



od, that of Brüche *et al.*<sup>5</sup> from 1 to 49 eV is the best and is shown in Fig. 4. Above about 6 eV, these results are in good agreement with the present values. The reason for the large discrepancy at lower energy is not known. The Ramsauer-method measurement of Golden and Bandel<sup>1</sup> from 0.3 to 28 eV appears to be in serious error at all energies, as was judged to be the case by Bederson and Kieffer. The error is perhaps one of scale and it has been suggested<sup>9</sup> that the problem was nonequilibrium within the scattering cell. The larger relative error at large energy suggests other types of errors; in fact an increase of the TCS by  $0.4 \text{ \AA}^2$  at all energies would bring this result into good agreement with the present results. Whatever the nature of the error, the Golden-Bandel error estimate was unrealistic and several percent more than the  $\pm 10\%$  suggested by Bederson and Kieffer<sup>9</sup> would place these results in good agreement with the present ones. No attempt was made in the present work to resolve the resonance at 19.4 eV (as was done by Golden and Bandel). Best instrumental resolution here was about 70 meV, but data-point density for these measurements was only 1 to 2 per eV.

The recent indirect determination of the TCS (now shown) by Andrick and Bitsch<sup>12</sup> from a phase-shift analysis of relative differential cross sections agrees (considering the combined error limits) with the TOF results, the former being in general several percent larger. Errors associated with this analysis are such that the relative error in the TCS increases with decreasing energy so that below 5 eV the uncertainty encompasses all the results shown in Fig. 4.

The direct measurement of Blaauw *et al.*<sup>3</sup> using a new linear-transmission method extends from 16 to 750 eV. The values below 20 eV (with an estimated uncertainty of 5%) are shown in Fig. 4 and are about 4% larger than the present results. A discussion of this discrepancy is given later.

There have been many calculations of the elastic scattering below threshold using a variety of methods. Some of the earliest to yield reasonably reliable results were the "polarized-orbital" methods. Varying results have been obtained due to the approximate character of the treatment<sup>33</sup> however and the only one within the error limits of the present results at all energies is that of LaBahn and Callaway,<sup>34</sup> one of the earliest. Another approximate method yielding good results is that of Yarlagadda *et al.*<sup>15</sup> (see Fig. 4) with many-body perturbation theory. Above 3 eV very good agreement is obtained with the present results, and below 2 eV the calculated values are at the lower experimental-error limits. [See note added in proof (i).]

There have recently been several *ab initio* or "close-coupling"-type calculations of the low-energy elastic phase shifts. The results of Sinfailam and Nesbet,<sup>14</sup> using a "matrix-variational" method, from 0.136 to 16.5 eV are in excellent agreement with the present results above 5 eV. At lower energies, there is an increasing discrepancy which reaches about 5% at 1 eV (2% below the lower error limit). The accurate MCS of Crompton and co-workers<sup>10</sup> supports the conclusion that these theoretical results are too small in this energy range. The difference in the MCS is about 4%, which is twice the experimental error limit up to 3 eV. Sinfailam and Nesbet suggest<sup>14</sup> that the discrepancy may be due to neglect of target-atom electronic correlation in their calculation. A similar method was employed by Wichmann and Heiss<sup>16</sup> from 2 to 20 eV. Agreement with the TOF results is within error limits to 10 eV, above which the calculated values of the TCS are too small. A very recent calculation from 0 to 16.5 eV by Berrington and O'Malley<sup>17</sup> using the "R-matrix" method is in very good agreement with the present values up to 11 eV. This work is very similar to that of Burke *et al.*,<sup>35</sup> but employed superior wave functions. The result at 16.5 eV (not shown in Fig. 4) is too small but, at the time of this writing, the calculations for the higher partial waves for this energy were not yet complete. The author's judgment that the final result will be larger and hence in better agreement with this experiment.

A direct comparison between an MCS and a TCS can be made with the aid of the scattering phase shifts, from which the ratio as a function of energy can be calculated. Of course, with accurate knowledge of the phase shifts the answer is known anyway, but the motivation for attempting this comparison using imperfect phase shifts is the reasonable assumption that the errors in the ratio will be much less than the errors in the total. Such a treatment of the Crompton *et al.*<sup>10</sup> and Milloy-Crompton<sup>11</sup> MCS's extending to 12 eV using the theoretical ratios (which agree to within 1% above 0.5 eV for the phase shifts from the various calculations discussed above) is indicated in Fig. 4. This result is within the TOF error limits at all energies at which they overlap. Considering the combined error limits and the uncertainties of the conversion this is excellent agreement.

There has been relatively little work on the TCS in the 20–50 eV region shown in Fig. 5. The TCS here is the sum of the cross sections for many channels and consequently gives less information about each, unless the others are well known. Recent interest has been due in large part to the controversy concerning the validity of the forward dispersion relation for electron scattering.<sup>36</sup> Of

the older measurements,<sup>4,5,7</sup> only that of Brüche *et al.*<sup>5</sup> is in reasonable agreement with the TOF values, the others being much too small. Those of Golden and Bandel<sup>1</sup> extending to 28 eV are about 0.4 Å<sup>2</sup> less than the present results (as was found for the lower energy region), the relative difference being up to 15%. The comparison with the results of Blaauw *et al.*<sup>3</sup> is also consistent with that at lower energy. There is a very close correspondence in the shape, a difference of about 4% in magnitude, and overlapping error limits. As discussed in Sec. IV, this type of discrepancy is probably due to an error in the pressure or the cell length. With regard to pressure, Blaauw *et al.* used the simple  $(T/T_0)^{1/2}$  formula for correction of thermal transpiration effects, whereas measured values less than those implied by the formula were used in the present case. The use of a more realistic value in the former case would result in lowering that TCS uniformly by about 1%, while use of the higher of the measured values (see Sec. IV for discussion) in the TOF case would increase the present results by 1.5%. With regard to effective cell length, Blaauw *et al.* judged that there was an ambiguity in their determination which could have as an effect the reduction of their reported TCS by up to 5%. Detailed consideration of the dynamics of gas flow from such a cell suggests that 5% is too large, but (1-2)% is certainly not ruled out. [See note added in proof (ii).] Due to the much longer cell length and the lower pressures used, there is no such uncertainty in the present case.

## VI. CONCLUSIONS

With the results presented above, the total electron scattering cross section is known to within 3% from 1 to 50 eV. The unreliability of the Ramsauer method, especially for low energies, has been demonstrated. The present measurement overlaps those of Crompton and co-workers<sup>10,11</sup> and Blaauw *et al.*<sup>3</sup> to a sufficient extent to confirm the basic reliability of precision-swarm methods for smoothly varying elastic momentum-transfer cross sections at low energy, and for the latter, to confirm the accuracy of their method for total cross sections at higher energies, excepting perhaps an ambiguity at the level of a few percent in

the choice of effective cell length. In conjunction with these results, experimentally determined integrated electron scattering cross sections with good accuracy are now available from 0.008 to 750 eV, a range of almost 10<sup>5</sup>. This accuracy and range will provide valuable tests for future developments in the theory of electron scattering, especially for approximate methods for which application to more complex atoms or molecules is likely. In this regard, accurate measurements for some of these more complex targets are clearly called for. It has been shown that for He feasible calculations of elastic scattering in the low-energy region, with accuracy similar to that of the present results, can be made with some existing methods.

The advantages of the present time-of-flight method are (i) the lack of pressure-dependents effects *outside* the scattering cell, (ii) no need for measurement of the attenuation as a function of pressure, (iii) the continuous electron-energy distribution, (iv) the direct and highly accurate energy determination, (v) the good energy resolution below about 20 eV, (vi) the excellent discrimination against small-angle scattering, and (vii) the basic simplicity of the method and its error evaluation.

*Note added in proof.* (i) Good agreement is also found with the random-phase-approximation (with exchange) results of M. Ya. Amusia and N. A. Cherepkov [Case Stud. At. Phys. 5, 47 (1975)]. (ii) A reevaluation of the effective cell length by the authors of Ref. 3 has indicated that their reported values should, in fact, be reduced by 5% (private communication).

## ACKNOWLEDGMENT

This is Contribution Number 3113 from the Chemical Laboratories of Indiana University. This research was supported in part by the National Science Foundation, Grant No. CHE 76-83683, and also by the Petroleum Research Fund, administered by the American Chemical Society. The authors wish to thank Drs. R. W. Crompton and C. E. Kuyatt for careful readings of, and helpful comments on, the manuscript.

\*Present address: Joint Institute for Laboratory Astrophysics, Univ. of Colorado, Boulder, Colo. 80309.

<sup>1</sup>D. E. Golden and H. W. Bandel, Phys. Rev. 138, A14 (1965).

<sup>2</sup>C. Ramsauer, Ann. Phys. (Leipz.) 66, 546 (1921).

<sup>3</sup>H. J. Blaauw, F. J. de Heer, R. W. Wagenaar, and

D. H. Barends, J. Phys. B 10, L299 (1977).

<sup>4</sup>R. B. Brode, Phys. Rev. 25, 363 (1925).

<sup>5</sup>E. Brüche, D. Lillenthal, and K. Schröder, Ann. Phys. (Leipz.) 84, 279 (1927).

<sup>6</sup>C. Ramsauer and R. Kollath, Ann. Phys. (Leipz.) 3, 536 (1929).

C. E. Normand, Phys. Rev. 35, 1217 (1930).

<sup>7</sup>C. Ramsauer and R. Kollath, Ann. Phys. (Leipz.) 12,

- 529 (1932).
- <sup>9</sup>B. Bederson and L. J. Kieffer, *Rev. Mod. Phys.* **43**, 601 (1971).
- <sup>10</sup>R. W. Crompton, M. T. Elford, and R. L. Jory, *Aust. J. Phys.* **20**, 369 (1967); R. W. Crompton, M. T. Elford and A. G. Robertson, *ibid.* **23**, 667 (1970).
- <sup>11</sup>H. B. Milloy and R. W. Crompton, *Phys. Rev. A* **15**, 1847 (1977).
- <sup>12</sup>D. Andrick and A. Bitsch, *J. Phys. B* **8**, 393 (1975).
- <sup>13</sup>R. K. Nesbet, *Adv. Quantum Chem.* **9**, 215 (1975).
- <sup>14</sup>A. L. Sinfailam and R. K. Nesbet, *Phys. Rev. A* **6**, 2118 (1972).
- <sup>15</sup>B. S. Yarlagadda, G. Csanak, H. S. Taylor, B. Schneider, and R. Yaris, *Phys. Rev. A* **7**, 146 (1973).
- <sup>16</sup>E. Wichmann and P. Heiss, *J. Phys. B* **7**, 1043 (1974).
- <sup>17</sup>K. A. Berrington and T. F. O'Malley, in *Proceedings of the Tenth International Conference on the Physics of Electronic and Atomic Collisions, Paris, 1977*, edited by G. Watel (Commissariat a l'Energy Atomique, Paris, 1977).
- <sup>18</sup>R. E. Kennerly and R. A. Bonham, *Chem. Phys. Lett.* **43**, 245 (1976).
- <sup>19</sup>R. E. Kennerly, *Rev. Sci. Instrum.* **48**, 1682 (1977).
- <sup>20</sup>R. E. Kennerly and R. A. Bonham (unpublished) (available from the authors).
- <sup>21</sup>F. C. Witteborn and W. M. Faribank, *Rev. Sci. Instrum.* **48**, 1 (1977).
- <sup>22</sup>J. E. Land and W. Raith, *Phys. Rev. Lett.* **30**, 193 (1973); *Atomic Physics*, edited by S. J. Smith and G. K. Walters (Plenum, New York, 1973), Vol. 3, p. 553; *Phys. Rev. A* **9**, 1592 (1974).
- <sup>23</sup>W. Raith, *Adv. At. Mol. Phys.* **12**, 127 (1970).
- <sup>24</sup>Model 310BHS-1 from MKS Instruments, Inc., 22 Third Ave., Burlington, Mass. 01803.
- <sup>25</sup>E. H. Kennard, *Kinetic Theory of Gases* (McGraw-Hill, New York, 1938), p. 66.
- <sup>26</sup>J. P. Bromberg, *J. Vac. Sci. Technol.* **6**, 801 (1969).
- <sup>27</sup>G. C. Baldwin and M. R. Gaertner, *J. Vac. Sci. Technol.* **10**, 215 (1973).
- <sup>28</sup>G. C. Baldwin and S. I. Friedman, *Rev. Sci. Instrum.* **38**, 519 (1967).
- <sup>29</sup>G. C. Baldwin, *Phys. Rev. A* **9**, 1225 (1974).
- <sup>30</sup>Flow and Pressure Group, Physical Standards Branch, U. S. Army Metrology and Calibration Center, Redstone Arsenal, Ala. 35809 (unpublished), communicated to R. W. Crompton.
- <sup>31</sup>B. P. Mathur, J. E. Field, and S. O. Colgate, *Phys. Rev. A* **11**, 830 (1975).
- <sup>32</sup>A. Salop and H. H. Nakano, *Phys. Rev. A* **2**, 127 (1970).
- <sup>33</sup>W. M. Duxler, R. T. Poe, and R. W. LaBahn, *Phys. Rev. A* **4**, 1935 (1971).
- <sup>34</sup>R. W. LaBahn and J. Callaway, *Phys. Rev.* **147**, 28 (1966).
- <sup>35</sup>P. G. Burke, J. W. Cooper, and S. Ormonde, *Phys. Rev.* **183**, 245 (1969).
- <sup>36</sup>See, e.g., P. K. Hutt, M. M. Islam, A. Rabheru, and M. R. C. McDowell, *J. Phys. B* **9**, 2447 (1976); and F. J. deHeer, M. R. C. McDowell, and R. W. Wagenaar, *ibid.* **10**, 1945 (1977).