Total Electron–Atom Collision Cross Sections at Low Energies—A Critical Review*

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Experiments relating to measurements of total and momentum-transfer cross sections for the scattering of low-energy electrons by atoms and diatomic molecules are critically reviewed. Principal emphasis is placed upon the Ramsauer method, dc swarms, and crossed-beams experiments, which account for the bulk of the reliable data in the literature although other techniques including differential measurements are also discussed. The theories of the various methods and possible sources of error are discussed. The case of low-energy electron scattering by helium is exhaustively reviewed since this system has been most intensively studied experimentally and is particularly amenable as well to theoretical calculations. The best available cross section values, along with comments on individual experiments, are presented in several tables.

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1. INTRODUCTION

1.1 Historical Background

Since the discovery of the electron at the close of the nineteenth century, the study of electron–atom collisions has played a central role in the development of our present understanding of quantum mechanics and of many-body systems. This article is chiefly concerned with one particular category of such processes, the determination of total and, to a lesser extent, momentum-transfer electron–atom (and electron–molecule) cross sections. Historically, these were the first areas in the field of electron–atom collisions to achieve status as a quantitative field, stimulated largely by the pioneering efforts of Ramsauer and Townsend and their students and collaborators. The experimental approaches of these two schools were quite different, the first developing what could now be characterized as "transmission" experiments, the latter what are now called "swarm" experiments. Both methods had already been extensively exploited before World War II. A third method, that of crossed beams, which relies heavily upon modern technology, began its development only in post-World War II days and has really only recently begun to yield results.

The period of greatest activity of Ramsauer and his co-workers extended from 1921 at the Radiological Institute at Heidelburg through 1930 at the Research Institute of the AEG, Berlin.[1] During this time, total cross section measurements were made on essentially all atomic and simple molecular systems which were sufficiently stable and noncorrosive to survive in a room-temperature apparatus. These measurements were made over a very wide range of electron energies, going down to less than 1 eV in some cases. Some angular distribution measurements were performed as well.

In the early days the most fruitful transmission

* The writing of this review was supported by the National Bureau of Standards, Office of Standard Reference Data, as part of the National Standard Reference Data Program.

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[1] C. Ramsauer's (1914) first paper concerned the investigation of the velocity distribution of photoelectrons emitted by a zinc surface. He later decided to add gas to the system and perform attenuation measurements. The first paper in this series (Ramsauer, 1921a) employed the original two-circle apparatus. This was soon replaced by the one-circle apparatus, whose principles continued to be exploited in all subsequent Ramsauer work. In the 1920's when vacuum techniques were not highly developed, it was nevertheless possible, by taking *difference* measurements, to avoid difficulties with background gas impurities. By assuming that these impurities remained constant with and without scattering gas present, the contribution to the signal of the impurities could be subtracted off. Such a procedure made for a lively experiment in the early Ramsauer work since only a few minutes were available after gas filling before the background impurities began to grow to serious proportions. Also, of course, this subtraction procedure is highly vulnerable when the magnitude of the scattering gas cross section is very much smaller than the cross sections of the impurity gases (e.g., helium and neon in air and mercury).

method for measuring total cross sections turned out to be what is now known as the "Ramsauer technique." This is a transmission experiment performed in the presence of a magnetic field oriented perpendicular to the direction of motion of the electron beam. The field serves the double purposes, first, of assisting in the energy selection of the incoming electrons, and, second, of providing energy and momentum discrimination of the elastically and inelastically scattered electrons. A scattering event changes the magnitude of the radius of curvature of the electron for inelastic scattering or for elastic scattering out of the plane of the apparatus. Elastic scattering in the plane changes the position of the center of curvature (guiding center) of the rotating electron, without changing its radius significantly. Ramsauer (1921a) very early recognized that this type of measurement should detect scattering events with a high degree of sensitivity, i.e., with "excellent" angular and energy resolution. A quantitative discussion of these resolution problems follows in later sections.

Ramsauer's first apparatus did not contain provisions for applying accelerating potentials to the electron beam. He nevertheless managed to produce beams at two energies (0.8 and 1.1 eV) by using different arc sources for the photoelectrons and observed a precipitous decrease in the argon cross section (roughly a factor of 2) in going from the higher to the lower energy. This was a precursor of the later verification by Ramsauer and Kollath (1929), and Rusch (1925), of the existence of a cross section minimum at very low energies in the heavy rare gases (Ar, Kr, Xe). This minimum had already been postulated by Townsend and Bailey (1921, 1922a, 1922b, 1923) as a result of their swarm measurements in argon. This "Ramsauer–Townsend" effect has played a historic role in the early application of quantum mechanics to scattering theory.

Brode, first at California Institute of Technology and then at Berkeley, constructed a modified Ramsauer apparatus in the early 1920's. This device had the unique capability, for that time, of being operable at elevated temperatures (to 400°C). This feature allowed for considerably improved vacuum techniques and also permitted the study of certain metal vapors (the alkalis, Hg, Cd, Tl, Zn). To this day, Brode's data remain the only available information on certain of these elements above thermal energies (where some swarm data exist). Somewhat later Bruche (1926a, 1926b, 1927a, 1927b; Bruche, Lilienthal, and Schrodter, 1927) first using transmission in a longitudinal magnetic field, and then using the Ramsauer technique, performed an independent set of measurements on many atomic and simple molecular systems, in some cases repeating Ramsauer's measurements. In general, reasonable agreement with Ramsauer was obtained. These experiments were done with considerable care and skill and have survived the test of time quite well. Recently, several new Ramsauer-type apparatuses have been constructed, with improvements in technique appropriate to present day technology. Notably, Golden and coworkers at Lockheed Missile and Space Company, Palo Alto, California, have remeasured many of the systems previously studied, using improved vacuum and gas-handling techniques. Thus, this method, which has not changed drastically over almost half a century, continues to be actively exploited.

Angular distributions were also studied in pre-World War II days,[2] by a variety of techniques, including the use of rotating collectors or guns (Arnot, 1931; Bullard and Massey, 1931) and fixed zone plates (Ramsauer and Kollath, 1931a, 1932). Relative angular distributions of elastic and total cross sections from energies as low as 1 eV to several hundred electron volts were obtained in many gases. In cases where studies by different experimental groups, using differing techniques, overlapped, agreement of the relative shapes of the differential curves was quite good. Chief among the early workers in this area were Arnot, Bullard and Massey, Dymond, Hughes, McMillen, and Ramsauer and Kollath. Recent angular distributions studies are discussed in Sec. 2.2.

Atomic-beam techniques now play an essential role in collision studies because they permit the preparation of the target into specific species (and in some cases, into specific excited states or magnetic substates) even though the species is chemically unstable, or may normally occur in a mixture with other species, or may possess an inconveniently low vapor pressure at convenient temperatures.[3] Nevertheless the development of atomic beam technology preceded its application to collision studies by two decades. Beginning in the 1920's, with Stern and his students, among them Rabi (see Ramsey, 1956), a series of fundamental experiments were performed, starting with direct verifications of kinetic theory predictions in low-density gases, through the historic Stern–Gerlach experiment, followed by studies of magnetic properties of atomic and nuclear systems (and of induced electric dipole moments in the alkalis).

Although the potential use of atomic beams in the study of atomic collisions was recognized early, and in fact several important beam–gas experiments were performed in the 1930's and 1940's,[4] it was not until the mid-fifties that experiments using bona-fide crossed-beam techniques were successfully performed. Fite and coworkers (Fite and Brackmann, 1958a, 1958b, 1959; Brackmann and Fite, 1958) performed a series of electron scattering experiments on atomic hydrogen, dramatically demonstrating the power of the crossed-beam method. Groups at New York University, at the Convair Division of the General Dynamics Corporation

[2] Angular distribution work performed before 1950 is fully referenced in Massey and Burhop (1952).

[3] Some recent general surveys of atomic and molecular beam techniques include: Ramsey, 1956; Kusch and Hughes, 1959; King and Zacharias, 1956; Ross, 1966; Hughes and Schultz, 1967; Bederson and Fite, 1968.

[4] See, for example, Massey and Burhop (1952), pp. 385–397.

in San Diego, and at the University of Bonn also began beam work at about the same time. Many of the techniques developed originally for magnetic resonance work were taken over bodily and applied to collision work, among these being the use of magnetic state selectors and atomic-beam source and detector technology. It appears certain that we have thus far witnessed only the beginnings of the application of this method to collision studies. We shall see, however, that up to the present time there has not been an overwhelming amount of data forthcoming, at least in measurements within the purvue of this article.

At approximately the same time that Ramsauer was developing the concepts and techniques for performing electron transmission experiments, J. S. Townsend, in the Cavendish Laboratory at Cambridge, was attacking a related problem, namely, the behavior of charged particles in a gaseous medium under the influence of electric and magnetic fields (Townsend and Bailey, 1921, 1922a, 1923). This work stemmed from the famous studies of Thomson on the free electron and was the precursor of an entire field of physics, presently referred to as "gaseous electronics." More specifically, Townsend was concerned with the passage of electrons through gases under steady-state conditions, i.e., assuming the electrons suffer large numbers of collisions with the neutral gas medium. Such experiments, which assume that the charged-particle densities are sufficiently low so that space-charge effects can be neglected, are called "swarm" experiments.

The behavior of the charged particle in the presence of fields is governed by the Boltzmann equation with appropriate collision terms. From the point of view of this article, which is mainly concerned with electron–atom collision processes, such swarm experiments are of importance because they provide an alternative method of determining collision cross sections as a result of their effect on the Boltzmann collision integral, and, as a consequence, on the macroscopic transport properties (Morse, Allis, and Lamar, 1935; Margenau, 1946; see also the classic review article by Allis, 1956, which summarizes the application of first-order perturbation theory to the solution of the binary Boltzmann collision integral, in the presence of a weak electric field).

Perhaps the most significant developments in swarm experiments in post-World War II years have been the refinement of the dc (Townsend) techniques by Phelps, Crompton, and coworkers and the development of ac (microwave) techniques, particularly to determine electron densities by the measurement of the ac conductivity of weakly ionized gases. The microwave experiments were pioneered by Brown and his students at M.I.T. (Phelps, Fundingsland, and Brown, 1951), and much of the subsequent development of this subject has been pursued by his students, particularly Biondi and Phelps. It will be seen, however, that it is principally the dc work with which we will be concerned in this article since the microwave techniques have not been extensively exploited in the quantitative determination of momentum-transfer cross sections.

Very recently a technique new to the atomic physics scene, time of flight, has appeared. This method, discussed briefly in Sec. 2.8, is a variant of the straight transmission experiment. It has thus far yielded published results only for helium and argon; however, it offers interesting possibilities for other systems. There is also work in progress using this technique to study positron scattering.

1.2 Bibliographic Material

Several early reviews contain discussions of the work performed before World War II. Perhaps the most complete of these is by Ramsauer and Kollath (1933) (see also Kollath, 1958). This article discusses essentially all of the total electron cross section measurements made before 1933 and includes discussions of most of the angular distribution work as well. (Ion–atom collisions also are reviewed in this article.) An earlier review article by Kollath (1930) is also quite valuable in that it offers criticisms of the various experimental techniques and also attempts to make value judgments on all of the total cross section data published up until that time. Review articles in English include those of Brode (1933) and McMillan (1939), which is a summary of the theoretical and experimental state of electron–atom collisions as of that time. More recently the comprehensive monographs of Massey and Burhop (1952, 1969) contain much of the existing atomic collision data as of 1950 (First Edition), and as of 1968 (Second Edition). Little, if any, in the way of critical appraisal is contained, however, in these volumes. Recent books by McDaniel (1964) and Hasted (1964) on atomic collision processes include substantial sections on electron–atom collision cross sections. Other valuable reference sources for this field include the books of abstracts of papers presented at the biennial meetings of the International Conference on the Physics of Electronic and Atomic Collisions. For convenience this conference series will be referred to in the bibliography as ICPEAC. The review article by Kieffer and Dunn (1966) on ionization cross sections contains discussions of possible sources of systematic errors in cross section measurements. Many of the errors discussed there are equally applicable to total cross section work.

Bibliographic material and cross section curves are presented in a recent compendium by Brown (1967). A complete bibliography of low-energy cross section data through 1966 is contained in the U. S. Department of Commerce National Bureau of Standards Miscellaneous Publication 289 (Kieffer, 1967), which lists all published electron–atom collision work, both theoretical and experimental, and which is revised periodically (Chamberlain and Kieffer, 1970). Experimental techniques are discussed in Volumes 4 and 7 of the series, "Methods of Experimental Physics"

(Hughes and Schulz, 1967; Bederson and Fite, 1968). Volume 4 deals with primarily atomic beam sources and detectors, while Volume 7 covers techniques generally employed in atomic collisions work.

The journal *Atomic Data* (Katharine Way, Editor, Academic Press, New York) has published two compilations of low-energy electron collision cross section data (Kieffer, 1969a, 1969b, 1971).

1.3 Scope of the Present Article

The principal goal of this article is to critically review the currently available techniques for measuring total cross sections for the scattering of low energy electrons by atoms and molecules, and to attempt appraisal, where possible, of the accuracy of published data. This necessarily emphasizes, therefore, beam-type experiments, where these quantities are most directly investigated. To the extent, however, that swarm experiments can be utilized to yield total cross sections, these will also be considered. Such swarm measurements cannot be applied to molecules as readily as to atoms because of the presence of low-lying rotational and vibrational excited states, and so our swarm discussions will be primarily restricted to atomic systems. The rather limited available data involving purely elastic cross sections and measurements performed upon excited states will also be briefly discussed. The energy range to be covered is from the order of several hundred electron volts to the lowest energies which have been studied (fractions of 1 eV).

The theories of the various beam–gas, beam–beam, and swarm experiments will be discussed since these are necessary for a critical evaluation of the data. General discussions regarding possible sources of systematic errors in these various methods will be included. Detailed and comprehensive curves and tabulations of data are contained in the JILA Data Compilation in *Atomic Data* (Kieffer, 1971).

We employ here the definition of cross section as follows: $\sigma_{ij}(\theta, \phi)\, d\Omega$ is the flux of particles scattered into the range of solid angle $d\Omega$ per unit incident flux, per unit target density, per unit length of scattering region. The subscripts i, j refer to the initial and final states of the target particle, respectively. The quantity

$$\sigma_{ij} = \int \sigma_{ij}(\theta, \phi)\, d\Omega, \tag{1}$$

where the integration is performed over all angles, is referred to as the "total cross section for the excitation $i \rightarrow j$." $\sigma_{ii}(\theta, \phi)$ and σ_{ii} are the differential elastic cross section and the total elastic cross section for the state i. $\sigma_i(\theta, \phi)$ and σ_i are the differential and total cross sections, respectively, for reactions into all possible final states from the initial state i. When the subscript is omitted it is usually understood that the system originates in the ground state. It is primarily the quantity σ which is of interest in this article since the total cross section experiments performed to date usually refer to ground-state targets and have not included energy analysis of the final state.

The momentum-transfer cross section σ_{MT} is defined as

$$\sigma_{MT} = \int \sigma_{ii}(\theta, \phi)\,(1 - \cos\theta)\, d\Omega \tag{2}$$

with the integration performed over all angles, and with σ_{ii} referring to the ground state, that is, σ_{MT} refers to elastic collisions only, as employed in this article.

Cross sections will generally be given in units of square centimeters. Very often, particularly in the early literature, cross sections were presented as P_c, the number of collisions suffered by an electron per centimeter of travel at 1 torr at 0°C. We have converted these to standard cross sections using the relation $\sigma = 2.83 \times 10^{-17} P_c$ cm^2. Occasionally experimental cross sections are given in the literature in atomic units, or in units of a_0^2, where a_0 is the first Bohr radius. These are converted to standard cross sections using the relations $\sigma(\text{cm}^2) = \sigma(\pi a_0^2) \times 0.880 \times 10^{-16}$ and $\sigma(\text{cm}^2) = \sigma(a_0^2) \times 0.283 \times 10^{-16}$. The normal unit of energy is the electron volt, and energy scales are plotted in these units. Occasionally electron velocity has been employed as abscissa rather than energy, and in most of these cases the abscissa has been given in square root of volts. The relation between electron speed v and the square-root scale is $v = V^{1/2} \times 5.93 \times 10^7$ cm/sec. The abscissa, particularly in theoretical papers, is sometimes given in terms of k, the wave number, or k^2, usually in units of a_0, the first Bohr radius. These are related to energy in electron volts by $k^2 = (\text{eV}/13.6)$. Energy is also sometimes given in atomic units, where 1 a.u. = (eV/27.2).

2. DESCRIPTION OF EXPERIMENTAL METHODS

Most of the work described in this article employs what can perhaps be described as "conventional" electron gun design, a typical gun assembly usually consisting of either a photocathode, a tungsten filament or an indirectly heated cathode, acceleration and control electrodes, a field-free interaction region, perhaps some additional collimation, and finally a collecting anode. Little use has been made of energy selection devices, with the notable exception of the Ramsauer device, which uses magnetic velocity selection and is, in fact, capable of achieving resolutions which are quite comparable to those obtained with the currently favored electrostatic selectors.

It is interesting to note that most of the early work performed by the German school used photoemission, rather than thermionic emission, for electron sources. None of the early experiments used differential pumping, so that the electron source was exposed to the full gas pressure of the scattering volume. It was known that the emission properties of the photocathode (zinc was most commonly used) were less subject to the influence of changes in pressure, and by reactions with the gas,

than a hot cathode. The heat source at the cathode could also cause temperature and density gradients, as well as thermal transpiration, in the scattering volume. The resulting gas currents are undesirable and also serve to further cool the cathode and alter the emission. Of course, the photoemission technique possesses its own difficulties. Chief among these is the fact that the uv (usually obtained from a high-intensity mercury arc discharge) could reflect off the cathode and produce photoelectrons throughout the apparatus. Considerable care had to be taken to minimize this problem. Photoionization of the target gas could also produce serious systematic errors.

The energy spreads obtainable from thermionic and photoelectric sources are comparable, with the former being perhaps narrower under proper operating conditions. These spreads, as well as the absolute values of the electron energy, were usually measured using retarding potentials although in the Ramsauer device it is also possible to achieve this by means of the magnetic field. The use of retarding potentials, its limitations and its difficulties, are discussed by Simpson (1961), by Schulz (1959), and by Kuyatt (1968). This subject will not be discussed in detail here, other than to point out some of the obvious possible sources of systematic error. There is foremost the problem of establishing an absolute energy scale, which can be in serious error because of focusing effects and local contact potential differences in the vicinities of the interaction chamber and the retarding electrodes, temperature gradients and the like.[5] Particularly in the early work, energy scales as determined by retarding potential methods alone must be considered uncertain by as much as 1 or 2 eV. A particular problem arises in attempting retarding potential measurements in the presence of magnetic fields since retarding fields cause changes in the curved electron trajectories as the electron slows down, rather than the trajectories normal to the surface which are actually required. (It should be noted, however, that the principal method of determining absolute eelctron energies in the Ramsauer experiments involved use of the magnetic field, rather than retarding potentials.) Most of the work discussed in this article employs very small currents (10^{-7} A) and space-charge effects in the vicinity of the retarding field are usually not significant.

Among the other commonly experienced difficulties, particularly at low energies, is the problem of electron reflection. This subject is also discussed at length by various authors (McGowan, 1967; Golden and Bandel, 1965) although there is by no means universal agreement over the best means of minimizing it. It is, however, generally recognized that polished metal surfaces are particularly efficient reflectors of electrons, and that some sort of coating [electron velvet (Marmet and Kerwin, 1960), "aquadag," "alcadag" (McGowan, 1967)] is required to reduce the reflectivity to manageable proportions. In addition, it is desirable to use baffles and shields wherever possible, to keep the reflected electrons from entering neighboring chambers.[6]

[5] Unless otherwise stated in the original articles, it must be assumed that the apparatus contained the usual assortment of experimentalists' materials, including brass, copper, nickel, etc.

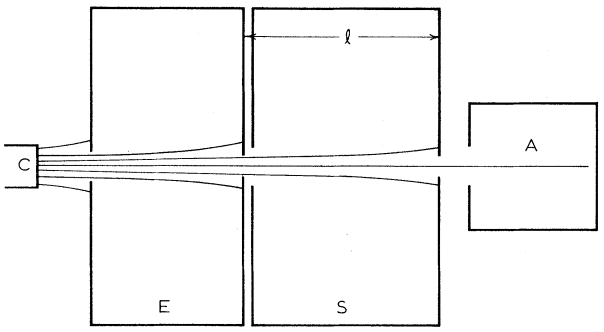

FIG. 1. The basic elements of a total electron–atom cross section measurement performed in transmission. C, electron gun; E, energy selector; S, scattering region; A, anode or collector.

Analysis of the resolution of a magnetic energy selector is similar to that of an electrostatic analyzer since the problem is basically one of choosing allowed circular trajectories through the several collimating slits. Assuming three equally spaced collimators of equal widths, a 180° selector possesses an energy resolution $\Delta E/E$, where ΔE is the full width of the energy E, which is given approximately by the relation (Golden and Bandel, 1965)

$$\Delta E/E \approx 2(\Delta R/R),$$

where R is the mean radius of curvature of the circular path of the electrons and ΔR is the collimating slit-width, and effects caused by the component of velocity along the direction of the magnetic field are neglected. For the dimensions of the Golden–Bandel (GB) apparatus, for example, $\Delta E/E$ is stated to be 6% or 7%, to give a spread at half-maximum of about 3.5%. For energies below 3 eV, GB measured a width at half-maximum of about 3%, using retarding potentials. At higher energies the resolution improves and ultimately becomes independent of energy, apart from a 1% dependence attributed to field penetration, since the limiting value should depend only upon the spread of energies leaving the cathode (about 0.1 eV at half-maximum).

2.1 Transmission Techniques

The basic elements of a total electron–atom cross section measurement in transmission are shown in Fig. 1. The analysis presented here also applies to the

[6] Such elements complicate contact potential problems, even if great care is taken to ensure that baffles, collimators and the like are fabricated out of identical materials to the remainder of the chamber. However, colloidal coating effectively eliminates contact potential differences. Metal vapors (e.g., cesium and mercury) are also quite effective for this purpose.

Ramsauer experiment, provided magnetic affects are ignored. (See discussion later in this section.)

The apparatus contains an electron source C, an energy selector E, a scattering region S, of length l, and an anode A. The scattering region and anode are normally maintained at the same potential (usually ground), and the energy of the electron is therefore determined by the potential V established on the cathode. Retarding potential measurements are usually made at, or behind, the anode.

The measured quantities are $I(s)$ and $I(a)$, the currents to the scattering chamber and anode, respectively. Neglecting end effects and the generation of secondary electrons in the gas or at surfaces, the sum

$$I=I(s)+I(a)$$

is constant. Differential quantities will refer to current in the energy range dE at E. We define $T(E)$ as the beam-shape form factor normalized to unity (i.e., $T(E)$ describes the two-dimensional electron beam shape), so that

$$\int T(E)\ dS=1,$$

where the integration is performed over a reference plane which for convenience could be the plane containing the aperture connecting the scattering chamber and the anode. Thus $T(E)\ dS$ is the fraction of the full current passing through an area dS normal to the direction of the electron velocity (assumed for simplicity to be parallel to the apparatus axis). $T(E)$ depends parametically upon details of the electron optics geometry and applied voltages, cathode effects, and upon space-charge effects, but does not depend upon effects directly attributable to the scattering gas. For example, $T(E)$ may depend upon the magnitude of the current leaving the cathode through space-charge interactions. It will also be affected indirectly by the presence of the target gas through cathode interactions, altered conditions in the energy selector, background gas ionization and excitation, and so on.[7] Thus we have

$$dI_0(a)=dI_0\int_{\text{entrance aperture}} T(E)\ dS,$$

where the integration is performed over the entrance aperture to the anode. The zero subscript refers to currents measured in the absence of gas in the scattering region, that is, under vacuum conditions.

In the presence of a scattering gas, $dI(a)$ is altered both by *scattering out* of $dI_0(a)$ and by *scattering in* of $dI_0(s)$. Again we must introduce a new quantity which takes these effects into account, and define $\eta(\theta,\phi;G)$ as the fractional number of electrons scattered from a point in the interaction region, denoted symbolically by G, into (θ,ϕ) which are scattered out of (or into) the anode region, i.e., which are registered as scattering events at the anode. The total transmitted current in the presence of scattering gas is then

$$dI(a)=dI_0\exp\left[-n\int T(E)\eta(\theta,\phi;G)\sigma(\theta,\phi)\ d\tau\ d\Omega\right], \tag{3}$$

where the integration is performed over the entire region in the interaction volume where there are beam electrons, as well as in the region outside the interaction volume where scattering can affect $dI(a)$. The dependence of $T(E)$ upon scattering gas and multiple scattering effects are ignored. n is the target gas density assumed constant. In terms of the electron energy distribution function $f(E)$, normalized to unity,

$$dI_0(a)=I_0(a)f(E)\ dE,$$

we obtain the full anode current

$$I(a)=I_0\int f(E)\times\left\{\exp\left[-n\int T(E)\eta(\theta,\phi;G)\sigma(\theta,\phi)\ d\tau\ d\Omega\right]\right\}\ dE. \tag{4}$$

Equation (4) is the basic equation connecting the observable in a transmission experiment, $I(a)$, to the desired quantity, which is the total cross section

$$\sigma=\int\sigma(\theta,\phi)\ d\Omega.$$

It is seen that this connection is not nearly so close as one might expect. In fact, Eq. (4) looks uncomfortably like the Boltzmann binary collision integral, which of course in a sense it actually is. Only under ideal experimental conditions can σ be directly determined from Eq. (4); that is, if a pencil beam of infinitesimal width and energy distribution is assumed to be traveling through an apparatus defined by infinitesimally narrow collimating slits, then we have $\eta=1,0$ for scattering out and in, respectively, $T(E)=1,0$ for $I_0(a)$ and $I_0(s)$, respectively, and Eq. (4) reduces to

$$I(a)=I_0(a)\ \exp\ (-nl\sigma), \tag{5}$$

which is the usual starting point of the standard transmission experiment. The cross section actually measured in a nonideal experiment of course must take into account the averages over T and f and is then an "effective" cross section defined as follows:

$$\sigma_{\text{eff}}=(1/l)\int T(E)\eta(\theta,\phi;G)\sigma(\theta,\phi)\ d\tau\ d\Omega, \tag{6}$$

which is simply the analytic way of stating that one must always properly account for the electron spatial distribution and apparatus geometry in a transmission experiment. Equation (6) explicitly demonstrates that the quantity σ is *never* measured in a transmission experiment; it is rather σ_{eff}. Finally, an average over the energy distribution also occurs, through Eq. (4).

Aside from the requirements relating to knowledge of the details of the experimental setup, i.e., to $T(E)$ and $f(E)$, one encounters here a central problem of this type of measurement. This is the fact that the connection between σ_{eff} and σ can only be made if one has

[7] For example, f may thus depend upon pressure in such a way that the slope of the ln I/I_0 versus p curve will be constant, but the cross section as calculated from the slope will be erroneous.

knowledge of the differential cross section $\sigma(\theta, \phi)$ throughout the region where η differs from unity.

In practice only qualitative considerations of these difficulties are made.[8] It is assumed that Eq. (5) applies. A specific experimental problem arises because $I_0(a)$ cannot be directly measured since the electron optics does not necessarily remain constant as the pressure is varied during an attenuation measurement. Thus one cannot simply plot $\ln I(a)/I_0(a)$ to obtain σ. It is instead assumed that $I_0(a)$ is proportional to I_0 [a rather dubious assumption in the light of the above discussion of $T(E)$], so that one can write

$$\sigma = (1/nl)\{\ln [I_0/I(a)] + \text{const}\}. \quad (7)$$

A plot of n versus $\ln I_0/I(a)$ should then be a straight line whose slope should yield the cross section.

Despite the many drastic assumptions made in the derivation of Eq. (7), it is the basic equation used in virtually all published transmission experiments, including those using the Ramsauer technique (see below).

The Ramsauer experiment utilizes the basic elements discussed above, but in addition possesses a uniform magnetic field oriented transverse to the plane of motion of the electron beam. The elements of Fig. 1 are arranged in a circle, and typically the electrons travel through an arc of approximately 270° from cathode to anode, with the scattering chamber occupying the last 90° of the trajectory (see Fig. 2). The dual purpose of this field is to supply both energy selection and enhanced angular resolution for both elastic and inelastic scattering, as discussed in the introduction. This device is quite compact and uncomplicated in construction, which helps account for its appeal to workers in this field. However, the coupling of the energy selection and scattering processes together introduces some special problems, which are discussed later in this section.

Three variations of this device have been used in obtaining the data to be cited in this article. The first, or "two cage" method, used by the Ramsauer school, is the device described in the preceding paragraph. The second, or "one cage" method, is a modification of the Ramsauer apparatus used by Brode and his students. The Brode apparatus uses no separate energy selector, but rather the scattering cage occupies 180° of the electron's trajectory and doubles as an energy selector. The third variation is a contemporary two cage device used by Golden and his colleagues. It is very similar to the original Ramsauer design, but employs differential pumping, so that the target gas, insofar as possible, is pumped out of the cathode region.

Before indicating the specific problems which arise because of the addition of the magnetic field, it should first be noted that the Ramsauer device has accounted

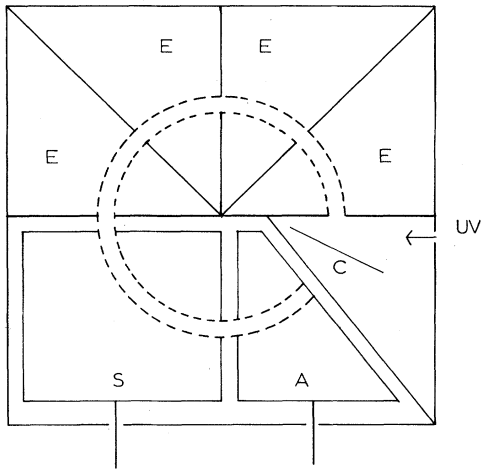

FIG. 2. A schematic diagram of Ramsauer's apparatus. Uniform magnetic field is oriented perpendicular to plane of motion of electrons. C, cathode, illuminated by uv light source; E, 180° energy selector; S, scattering region; A, anode.

for a vast majority of all total cross section measurements performed in transmission. The success of this technique is attributable to the combination of experimental simplicity and compactness, good energy selection ability, and good angular and energy-loss discrimination for elastic and inelastic collisions, respectively. It is remarkable that of the more than 40 transmission measurements listed in Table I, which summarizes all relatively reliable measurements reported on atoms and simple molecules, only two did *not* employ a variant of the Ramsauer method!

The principal difficulties inherent in the Ramsauer device include the following: (1) The need to simultaneously adjust both applied voltage and magnetic field at each energy. This makes it inconvenient to perform fast runs and also introduces uncertainties relating to systematic changes in the electron optics. (2) If the energy is obtained from a retarding potential measurement, the transverse magnetic field introduces an uncertainty whose effect has never been quantitatively analyzed. (3) The inability to measure T makes a complete analysis impossible, even were f and η known. (4) Even for purely elastic collisions, an analysis of η in the presence of a magnetic field is too difficult to perform analytically; a numerical analysis involving point-by-point ray tracing has never been attempted.

Additional difficulties are associated with the Brode modification because of the use of a single cage. A well-defined beam is not formed, so that there exists a substantial scattering-in contribution in Eq. (4) whose magnitude is impossible to determine. Furthermore, since electron focusing properties may change as the energy is varied, both T and f are unknown functions of the energy. Spurious structure in the cross section may result from such focusing effects. This could be especially pronounced at low energies, where the

[8] An exception to this is the Golden and Bandel (1965) experiments, where the angular resolution was calculated, with some simplifying assumptions (e.g., effect of magnetic field and of $I_0(S)$ were neglected).

velocity distribution is presumably most sensitive to gas density. Finally, the electron energy distribution leaving the cathode may depend strongly upon background gas pressure, which is varied during the course of a run. This effect would likely be particularly important when dealing with metal vapors, such as the alkali elements, which were extensively studied by Brode. It is for these reasons that the Brode device is particularly subject to systematic errors, and this is noted in presenting the Brode data in Table I.

In *all* attenuation experiments the linear relation between n and $\ln I_0/I(a)$ is a necessary condition on the data. It must be emphasized, however, that it is not a *sufficient* condition. This is made evident by the fact that a substantial disagreement has often occurred between experiments which do indeed all satisfy this linearity requirement.

The angular resolution of a collision experiment performed in transmission refers to the range of electron polar scattering angles for which events are observed with "reasonable" efficiency by the apparatus. Clearly there is no single such angular range in a given experiment since finite collimation, beam shapes, interaction path lengths and the like result in there being a distribution of resolving powers for different portions of the electron beam and of the scattering region.

Equally significant, scattering-in contributions, as indicated earlier, must also be taken into account, and when these are substantial, the angular resolution concept is not very meaningful. Thus, in the early Ramsauer experiments and particularly in the single cage device, where there is little or no information concerning the beam shapes [that is, $T(E)$ in Eqs. (3)–(6)], it is quite futile to attempt an angular resolution analysis.

Ideally the angular resolution θ_0 would be defined as the smallest polar scattering angle which is observable in a scattering experiment; that is, events which scatter elastically into angles smaller than θ_0 are not distinguishable from the unscattered beam (of course, inelastic events, or more generally, events resulting in a change of the internal states of the reactants, can be distinguished in principle for all scattering angles). A similar quantity can be defined for the maximum observable scattering angle.

More generally one refers to an "effective" resolution $\bar{\theta}_0$, defined so that a certain fraction of all elastic collisions occurring in the beam are observed as scattering events, averaged over the apparatus. This can be done as follows: An average efficiency $\bar{\eta}(\theta)$ is defined by performing the volume and ϕ integrals in Eq. (4),

$$\bar{\eta}(\theta) = \int T\eta(\theta, \phi; G)\, d\tau\, d\phi/2\pi \int T\, d\tau. \qquad (8)$$

$\bar{\theta}_0$ is then defined as being that angle for which $\bar{\eta}(\bar{\theta}_0)$ is a predetermined fraction. This evaluation can be effected only in the simplest experimental setups, specifically, for crossed beams. Kusch (1964)[9] has discussed the case of a rectangular beam possessing finite width and height, interacting with a scattering target possessing negligibly small dimensions.

Because the electron de Broglie wavelength in the electron volt region is comparable to or larger than atomic dimensions, electron elastic scattering is not strongly peaked in the forward direction, that is, into angles which are small compared to typical values of θ for which $\bar{\eta}$ differs substantially from unity. It is for this reason that uncertainties in the determination of θ_0 do not generally result in gross errors in the measurement of σ. This is not the situation in heavy-particle collisions, where scattering is indeed generally strongly peaked, and inadequate angular resolution can result in order-of-magnitude errors. Nevertheless, a quantitative electron–atom cross section determination requires reliable estimates of $\bar{\eta}$. Knowledge of $\sigma(\theta, \phi)$ of course is also required if one is to make the claim that the measured σ_{eff} differs from the total cross section σ by no more than some predetermined fraction.

Thus we see that a properly reported cross section measurement should not only discuss the "effective" angular resolution of the experiment, but should also describe the manner in which this quantity was obtained, including estimates of contributions due to scattering in, to effects caused by the beam shape, etc.

The angular resolution problem is also related to multiple scattering as well as end effects (scattering occurring in the neighborhood of entrance and exit orifices). These problems are not discussed here. In practice one invariably employs the linearity criterion to ensure lack of multiple scattering. That is, one operates at pressures sufficiently low so that scattering signal is a linear function of target gas density. End effects are never taken into account quantitatively.

2.2 Angular Distribution Techniques

The early angular distribution measurements have been discussed in a number of books and review articles (see Footnote 2) and will be only briefly described here. The design of an experiment to measure angular distribution, either elastic or inelastic, encounters little conceptual difficulty. One must allow for some means of rotating either source or detector about a reasonably well-defined interaction region. Alternatively, a number of separate, fixed collectors, each subtending a fixed range of solid angles about the scattering region, can be employed. Currents to each of these "zones" are measured in turn. Only Ramsauer and Kollath (1931a, 1931b, 1932) have used such a device for electron scattering, having studied the rare gases down to quite low energies, albeit with rather minimal angular resolution.[10] Their experiments, incidentally, possess the distinction of being among the very few attempts at *absolute* distribution measurements to date.

Angular distributions are generally relative, so that

[9] A more comprehensive discussion of the angular resolution problem as applied to molecular beam atom–atom scattering is given by von Busch (1966). See also Footnote 8.

[10] Recently McGowan (McGowan, Vroom, and Comeaux, 1969) has used a zone-plate device for observation of the angular distribution of photoelectrons from rare gases.

the peculiar difficulties associated with absolute determinations are avoided. In general, reasonably good agreement has been achieved by different groups where comparisons can be made. However, it should be pointed out that such comparisons have mostly been made at energies considerably higher than those at which comparison is possible in the total cross section work. It is at very low electron energies that some of the characteristic difficulties associated with electron scattering enter with full force.

A schematic diagram of Arnot's (1931) apparatus is shown in Fig. 3 in order to illustrate a characteristic experimental setup for the measurement of elastic angular distributions. The electron gun was capable of rotation through $\pm 120°$ by means of a glass joint. Suitable potentials on the various collector apertures discriminated against inelastically scattered electrons, ions, secondaries and reflected electrons from the collector. Multiple scattering could be minimized by insisting upon a linear pressure dependence of the scattered current. The slight changes in the dimensions of the effective interaction volume as a function of scattering angle were estimated and corrected for. The apparatus was bakeable (except for the rotating glass joint) to 500°C. Bullard and Massey (1931) used a similar device, however, with fixed electron gun and rotatable detector.

The lowest-energy data were obtained by Ramsauer and Kollath (RK) using the zone-plate apparatus. Recently a number of excellent angular distribution studies have been performed by Ehrhardt and colleagues (Andrick and Ehrhardt, 1966; Ehrhardt, Langhans, and Linder, 1968; Ehrhardt and Willmann, 1967), particularly at resonances (which do not directly concern us in this article). Of particular relevance to this paper, which is concerned mainly with total cross sections but not specifically with resonance structure, is the recent experiment by Gibson and Dolder (1969). In this work, measurements are reported of differential cross sections for energies between 3.1 and 19.1 eV.

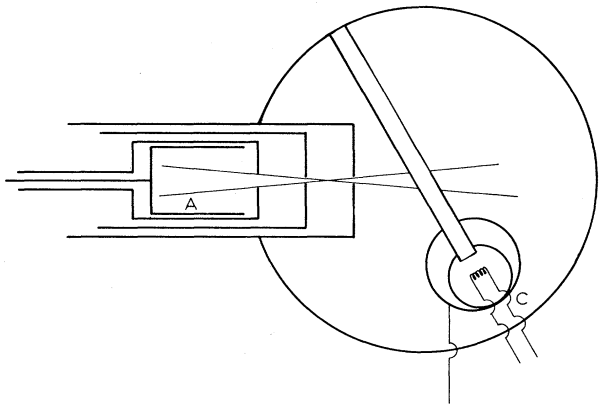

FIG. 3. A schematic diagram of Arnot's (1931) apparatus for measuring angular distributions. Electron gun (C) was capable of rotation through $\pm 120°$.

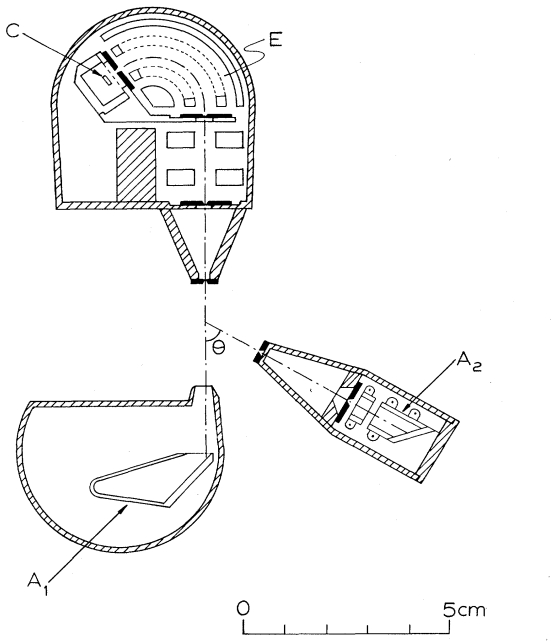

FIG. 4. A schematic diagram of angular distribution apparatus of Gibson and Dolder (1969), employing 127° electron monochromator.

An absolute calibration was made by means of a phase shift determination at the 19.3 eV resonance. These measurements, employing monochromatic electrons (65 meV energy spread), are probably the most definitive low-energy differential determinations available at this time. They are discussed more fully in the section on helium (Sec. 3.2). A diagram of this apparatus is shown in Fig. 4.

Angular distribution measurements are of course of central importance in achieving a better understanding of the scattering process. From the point of view of this article, differential cross sections play a special role. This relates to the angular resolution problem in total cross section determinations, as already discussed in Sec. 2.1. In order to properly correct for the finite angular resolving power of any collision experiment, the angular dependence, down to zero scattering angle, of the relevant reactions must be known. In electron–atom collisions at low energies, theory usually cannot supply a reliable guide in this matter, and one must therefore depend upon experimental determinations.[11] This is discussed more fully in Sec. 3.2, where the problem of the total elastic electron helium cross section is considered in detail.

Among the most important work in differential collision studies in recent years has been that of Lassettre and co-workers at Carnegie–Mellon University (Lassettre and Francis, 1964; Lassettre and

[11] An important exception involves the use of modified effective range theory, which uniquely determines the first several phase shifts from the low-energy behavior of the total cross section (see Sec. 3.2).

Jones, 1964; Lassettre, Skerbele, Dillon, and Ross, 1968; Lassettre, Skerbele, and Dillon, 1969). This work, performed with high energy and angular resolution, has yielded elastic and inelastic cross sections for a large number of reactions in many atomic and molecular gases. The bulk of this work has been performed at energies above 100 eV, and we do not, therefore, include it in our discussion.

2.3 Crossed Beams

Atomic-beam techniques possess great potential versatility in regard to the preparation of selected species, velocities, and states. Although conceptually simple, such experiments are often very difficult (and expensive) to perform in practice. Particularly when state selection, unstable species, or some other form of energy or state discrimination is employed, the signal-to-noise ratio, and in some cases the absolute signal itself, is quite unfavorable. Thus elaborate data-averaging procedures must be employed simply to overcome the statistics. As a consequence, very limited use has been made of these crossed-beam capabilities in the past although this situation is now improving.

We briefly review here the ways crossed beams have already been used in electron–atom scattering experiments involving total cross sections and also discuss the relative merits and drawbacks of the techniques employed.

The simplest means of producing an unstable species in a beam is by thermal dissociation of a molecular gas. The concentration of an atomic vapor in equilibrium with its (diatomic) molecule is given by a form of the law of mass action

$$[P(\mathrm{A})]^2/P(\mathrm{A}_2)=K(T), \tag{9}$$

where $K(T)$, the equilibrium constant, depends upon the dissociation energy of the molecule A_2 into two ground-state atoms A. $P(\mathrm{A})$ and $P(\mathrm{A}_2)$ are the equilibrium partial pressures of A and A_2. Thermal dissociation has been extensively used to produce atomic hydrogen (Lamb and Retherford, 1950; Fite and Brackmann, 1958a, 1958b) (the dissociation energy of H_2 is 4.48 eV) and some attempts have been made to produce atomic oxygen (Fite and Brackmann, 1963) (dissociation energy of O_2 is 5.1 eV). One significant advantage of this source is the fact that the atoms are formed virtually exclusively in the ground state, a very desirable feature in many cases. Atoms produced by this means are fast ($\sim$2500°K), which could be a handicap.

A more universal source for the generation of unstable species (that is, species in their ground or metastable states, which normally form stable compounds at laboratory temperatures) is the radio-frequency or microwave discharge. At least in principle these devices can produce usable quantities of many different unstable systems, including atomic systems such as O, N, H, Cl, etc., and free radicals such as OH. The discharge source is generally a fairly complex device; it usually operates under nonequilibrium conditions (i.e., velocity distributions are not known), and produces excited states, ions, electrons, as well as possibly other unstable compounds, by means of complex discharge chemistry, besides the ones desired.

Techniques for producing mechanically velocity-selected beams are well developed (Pauly and Toennies, 1968). Still, they have not been used in electron–atom beam experiments. Stern–Gerlach velocity selectors (Rubin, Bederson, Goldstein, and Collins, 1969; Fluendy, 1965) and time-of-flight velocity analyzers (Freund and Klemperer, 1967; Celotta, Brown, Molof, and Bederson, 1969) have been used in recoil experiments. The former device spin-polarizes the atom beam at the same time as velocity-selecting it, and therefore is a convenient and very simple tool in the performance of scattering experiments with spin analysis.

When used in conjunction with an ionizing detector followed by a mass analyzer, the effective temperature (mean speed) of a beam can also be estimated by the use of a seeded impurity. A low concentration of a rare gas, for example, is introduced into the discharge, and the partial beam intensity attributable to it is monitored with and without the discharge operating. Assuming constant throughput of gas through the source slit, the rare-gas beam intensity is inversely proportional to the mean speed. The uncertainties associated with this method will be discussed later in this section. A far better method for determining beam speed distribution is the use of a direct time-of-flight analysis, employing a high-speed beam chopper, and considerable progress using this technique has been achieved (Celotta, Brown, Molof, and Bederson, 1969).

The Stern–Gerlach magnet is also useful as a species selector since unstable atomic systems normally possess unpaired spins, and therefore magnetic moments of the order of a Bohr magneton, while the parent molecule possesses a magnetic moment which is only of the order of nuclear magnetons (the principal exception being O_2). The atom is therefore deflected (either focused or discarded) while the molecule is unaffected by the Stern–Gerlach field. More elaborate methods involving multipole electric and magnetic fields[12] have been used to state-select. Once again, these techniques have not yet been extensively applied to electron–atom beam collision experiments.

The details of crossed electron–atom beam experiments are discussed in review articles by Fite (1962) and by Bederson (1968). Only features of these analyses relevant to total and differential cross section work will be summarized here. In a completely general analysis it would be necessary to allow for variations in both the densities and the directions of motion of the two beams in the interaction volume. In all electron–atom beam work to date, however, it has been assumed that the

[12] Such state selection schemes are summarized in Table II, p. 260, of Pauly and Toennies, 1968.

beams travel in parallel, straight lines, and that they intersect at right angles in the interaction volume. (At least one of the beams possesses a uniform density throughout the interaction volume.) The interaction is illustrated in Fig. 5, in which electron and atom beams are traveling in the x, z directions, respectively.

We define the following quantities:

I_i^a, $J_i^a(x, y)$ are the total atom beam particle flux and flux density in the vicinity of the interaction volume, of a particular species and/or state i (particles/second, particles/centimeter^2·second).

I^e, $J^e(y, z)$ are the total electron beam particle flux and flux density in the vicinity of the interaction volume (electrons/second, electrons/centimeter^2·second).

$I_{ij}'(\theta, \phi)$ is the differential (per unit solid angle) scattered electron flux due to the reaction $i \rightarrow j$, where (θ, ϕ) are the polar and azimuthal angular coordinates of the detector measured with respect to a coordinate system oriented as shown in Fig. 5. The corresponding differential *atom* flux is $I_{ij}'(\psi, \chi)$, where ψ, χ are the atomic recoil scattering angles. This quantity, which may in fact not be constant across the atom beam, is normalized to the local density $n(x, z)$, that is,

$$\int f_i(x, y; V)\, dV = n(x, y).$$

If all speeds which contribute to the atom speed distribution are negligibly small compared to the electron speed, and if multiple scattering is neglected, then the number of electrons which are scattered into the element of solid angle $d\Omega$, measured with respect to the initial direction of motion of the scattered electron, is

$$I_{ij}'(\theta, \phi)\, d\Omega = \sigma_{ij}(\theta, \phi)\, d\Omega \int \eta(\theta, \phi; xyz) n(x, y) J^e(y, z)\, d\tau, \tag{10}$$

where τ is the interaction volume.

This is the differential equivalent of Eq. (4). Here η is the collection efficiency for electrons scattered into (θ, ϕ) from (x, y, z). The optics of the collector, i.e., its finite aperture and varying efficiency across the aperture, must be taken into account. In the shadow of the dc electron beam $I_{ij}'(\theta, \phi)$ includes both *scattering-in* and *scattering-out* contributions, as in the transmission case.

The total scattered current for the reaction $i \rightarrow j$, I_{ij}', is obtained by integrating Eq. (10) over all angles. In performing a crossed-beam experiment in transmission by observation of either the electron or the atom beam, the quantity $I_i - I_{ij}'$ is determined. We define the overlap integral

$$F(\theta, \phi) = \int_\tau \eta(\theta, \phi; xyz) n(xy) J^e(yz)\, d\tau. \tag{11}$$

Since the atom flux density J^a is

$$J^a = \int f_i V\, dV = n\langle V \rangle,$$

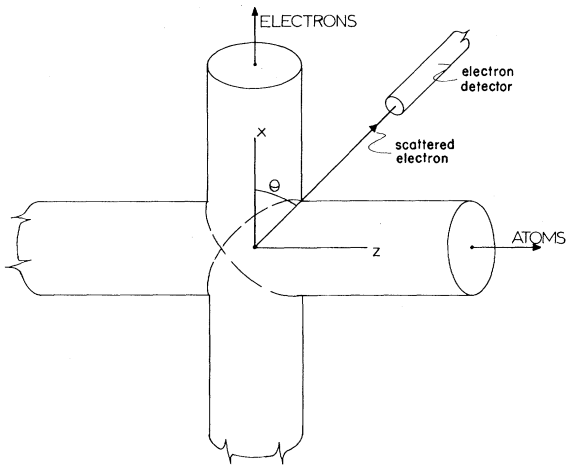

FIG. 5. Geometry of a crossed-beam experiment.

where $\langle V \rangle$ is the mean speed of the atom beam, F can be written symmetrically as

$$F(\theta, \phi) = \int_\tau \eta(\theta, \phi; xyz) J^a(xy) J^e(yz)\, dx\, dy\, dz / \langle V(xy) \rangle. \tag{12}$$

The appropriate form of F is chosen by convenience. If one uses a perfectly effusive source in thermodynamic equilibrium, Eq. (11) can be used; f_i is then precisely equal to the speed distribution function in the source, which is presumably known from the source temperature. In all other cases, the velocity distribution must be measured, and then the form to be taken for F depends upon the type of detector employed. Detectors based upon surface effects, for example, surface ionization, metastable atom detection by Auger electron ejection, and surface deposition detectors, are flux detectors and the quantity J^a is observed. An ionizing-type detector ideally measures $f_i\, dV$ although the ion collection system may in fact introduce serious distortions.

The source can itself introduce serious distortions in the velocity distribution which makes a direct velocity distribution determination essential. An example is the production of a metastable beam by electron bombardment where the atomic recoil can introduce significant distortion in the speed distribution (Celotta, 1971; Pearl, Donnelly, and Zorn, 1969).

Equation (10) is the basic equation of crossed electron–atom beam work. One of course recognizes that only an approximate knowledge is possessed of the various beam and geometry parameters appearing explicitly or implicitly in this relation. Specifically, Eqs. (11) and (12) directly reveal one of the central problems associated with obtaining absolute cross sections from crossed-beam experiments. This concerns the evaluation of the overlap integral F. Relative total cross section measurements avoid some of the more

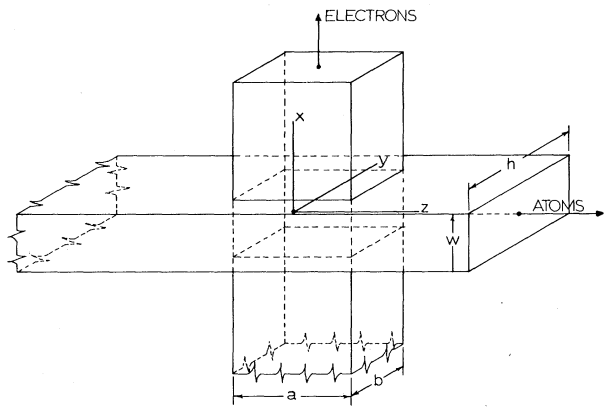

FIG. 6. Geometry of a crossed-beam recoil experiment assuming rectangular beams (See discussion in Sec. 2.3).

serious difficulties since F can perhaps be maintained constant throughout the measurement. However, varying electron optics with energy can cause serious errors even here.

In a measurement of relative angular distributions, Eq. (10) can be used directly, provided F is kept constant as the scattering angle is varied. Care must of course be taken to ensure that the solid angle element $d\Omega$ subtended by the detector is known as a function of scattering angle.

Absolute values may be obtained by brute-force evaluations of F at at least one value of either energy or angle, to supply suitable normalization of the entire cross section curve. A more common method is to normalize to a known cross section, using "identical" geometries and with velocity distributions which differ at most, hopefully, by a mass factor. Such a procedure is best accomplished by normalization to the corresponding parent molecular cross section, when studying an unstable atomic reaction, bearing in mind that the angular resolution may cause a serious normalization error.

There have been rather few measurements performed to date of total and differential cross sections using crossed-beam techniques, primarily because of the experimental difficulties and of the generally unfavorable signal-to-noise ratios obtained in these measurements. In a typical total cross section measurement, performed in transmission, the atom beam is mechanically modulated, and a coherent, properly phased electron signal is detected. Only a small fraction of the electron current is scattered, so that there is usually a substantial noise signal as well, even with synchronous detection and long integration times. When crossed-beam techniques are used to study unstable, i.e., atomic systems, the parent molecule must either be completely removed from the beam, or its effects must somehow be taken into account. In fact, the presence of the molecule can be turned into an advantage since the atom cross section can be directly normalized to that of the molecule, whose cross section is presumably known. Of course, it is necessary to make certain assumptions when doing this, including assuming that the atomic and molecular constituents possess identical beam geometries, and that both velocity distributions and the degree of dissociation are known. Angular distributions can be obtained by rotating the electron detector about the interaction volume, as discussed in the previous section.

There have been no experiments of this type which contain complete analysis of the angular resolution problem, as outlined above.

In the recoil method, observation of collision events is made on the scattered atom beam, rather than on the electrons (Bederson, 1968; Rubin, Bederson, Goldstein, and Collins, 1969). The method is especially useful in the performance of spin-state selected experiments since the spin state of the scattered atom is far more readily analyzed than is that of the electron.

The method is capable of yielding angular distributions and total cross sections as well. To measure the total cross section, the atom beam detector is set on the beam axis, and the decrease in atom current noted when the cross-fired electron beam is turned on. The total cross section is then obtained from the integral form of Eq. (10), in which I_{ij}' is now the total scattered-out atom current. To obtain F it is assumed that the atom beam is rectangular in cross section (of width w and height h; see Fig. 6). The overlap integral [Eq. (12)] can, of course, be evaluated only if η as well as both current densities and the mean speed are known throughout the interaction volume. To illustrate its determination in a recoil experiment we derive it here using several simplifying assumptions.

The geometry is shown in Fig. 6, and we specifically assume the following: (1) J^a is a function of x only, i.e., the atom beam profile varies *across* the beam, but not *along* it (this is a reasonable assumption for the case of a long, rectangular beam); (2) $\langle V\rangle$ is constant across the beam; (3) the entire electron beam crosses through the atom beam (i.e., $b<h$ in Fig. 6); it need not possess uniform density; (4) η is independent of z. This is a reasonable assumption if $a\ll L$, where L is the distance between the interaction volume and the detector plane.

We need to evaluate

$$T_i(\theta,\phi)=\int\eta(\theta,\phi;xy)J^a(x)J^e(yz)\,dx\,dy\,dz/\langle V\rangle. \quad (13)$$

Performing the z integration first, we replace $\int J^e(y,z)\,dz$ by the electron current per unit length $I^e(y)$. We now define an average $\bar{\eta}(\theta\phi)$ by the relation

$$\bar{\eta}(\theta,\phi)\int J^a(x)I^e(y)\,dx\,dy=\int\eta(\theta,\phi;xy)J^a(x)I^e(y)\,dx\,dy. \quad (14)$$

The simplified integral on the left side of Eq. (14) is

easily evaluated, to yield for σ_i

$$\sigma_i = I'h\langle V\rangle / I^a I^e \bar{\eta}, \tag{15}$$

where $\bar{\eta}$ has been averaged over (θ, ϕ).

If I' and I^a are measured using the same detection and data processing systems, then the determination of σ is absolute, in the sense that all quantities in Eq. (15) are either observable or are apparatus parameters, which can presumably be evaluated. Another important feature of this method is the fact that when a discriminating-type detector is used (e.g., an ionizing gun followed by a mass spectrometer), other beam constituents do not contribute to the scattering signal.

Angular distributions are obtained by moving the detector off-axis, and collecting the atoms which were scattered out of the beam. A transformation must then be made to relate the observed atom scattering angles to the electron scattering angles. Of course it is necessary in this case to velocity-select the atom beam, in order for the transformation to be unique.

In principle one would expect that a crossed-beam experiment would present a relatively clean-cut situation for an angular resolution analysis of the type discussed in the previous section. This would be the case if the interaction volume subtended a small, well-defined solid angle at the detector, and the other criteria discussed previously were met. Similar considerations of course apply to observations on the scattered electron. In practice such conditions have not always been even approximately satisfied. As a result, a realistic angular resolution analysis can be as difficult and uncertain as in a beam–gas experiment.

When observation is made on the scattered electrons in transmission, the electron collector is usually quite close to the interaction volume, so that the solid angle subtended by various portions of the interaction volume at the detector can vary over a substantial range. Variations in J^e across the beam, which seriously influence the overlap integral, are never taken into account. Quantitative analyses relating to this question have not been made in the few published experiments using this technique.

In recoil experiments the interaction volume does subtend a very small angle at the detector. A rough approximation to the average angular resolution θ_0, referred to the electron polar scattering angle θ_0 in radians, is given by

$$\theta_0 \approx (w/\beta L)^{1/2},$$

where w is the detector width, assumed equal to the beam width, and β is the ratio of electron momentum to atom momentum. L is the distance from the interaction region to the detector, and it is assumed that $a \ll L$, where a is the length of the interaction region. For a typical case, using a thermal atom beam and assuming $w = 0.1$ cm, $a = 2$ cm, and $L = 100$ cm, θ_0 is in the range of 1°–15°.

The variation of θ_0 across the interaction region is

$$\Delta\theta_0/\theta_0 = \tfrac{1}{2}(a/L)$$

which is quite small. A reasonably straightforward analysis can therefore be made on the scattered atom beam, to obtain the resolution in the atom scattering angle; this can then be transformed to obtain a well-defined value for resolution in the electron polar scattering angle.

2.4 Swarm Experiments

The relation of "fundamental" atomic processes, i.e., cross sections, to the macroscopically observable transport properties of a gas has, of course, long been recognized. As described briefly in the introduction, once the theory governing this relation has been developed and placed on a firm footing, one can work backwards from the laboratory observables, i.e., the transport coefficients, to the relevant cross sections, employing the appropriate kinetic theory to make the connection.

The general philosophy is at present perhaps best exemplified in the case of dc swarm experiments by the work of Phelps and co-workers[13] and Crompton and co-workers.[14] Their method is derivative from the techniques originally developed by Townsend and Bailey and can be described briefly as follows. Certain observables representing independent transport properties of electrons passing through gases, governed by the momentum-transfer cross section σ_{MT}, are measured under carefully controlled laboratory conditions. These observables are the electron drift velocity along a uniform electric field E, $W_{||}$, the ratio $D_\perp/\mu$, where $D_\perp$ is the diffusion coefficient perpendicular to the electric field and μ is the mobility (the ratio of the drift velocity to E), and the ratio $W_\perp/W_{||}$, where $W_\perp$ is the electron drift velocity at right angles to crossed, uniform electric and magnetic fields.

$W_{||}$ is obtained from direct time-of-flight measurements. A schematic of an actual drift tube, employed by Crompton *et al.*, is shown in Fig. 7. $D_\perp/\mu$ is obtained using the Townsend–Huxley method (Huxley and Crompton, 1962), which consists of a measurement of the ratio of total current traversing a region of uniform field to a fraction of that current which has drifted off-axis due to diffusion, as shown in Fig. 8. $W_\perp/W_{||}$ is obtained by splitting the annulus and outer collecting rings in Fig. 8 to observe the asymmetry in current collected in the plane of the anode caused by the $\mathbf{E}\times\mathbf{B}$ drift. Each of these observables is related to σ_{MT} by a collision integral that contains both σ_{MT} and $f(\mathbf{v})$, the distribution function, inside an integral. $f(\mathbf{v})$

[13] A recent review of dc swarm techniques particularly as applied to molecular systems is given by Phelps (1968).

[14] A general review of dc swarm techniques is given by Huxley and Crompton (1962). See also the recent review article by Crompton (1969).

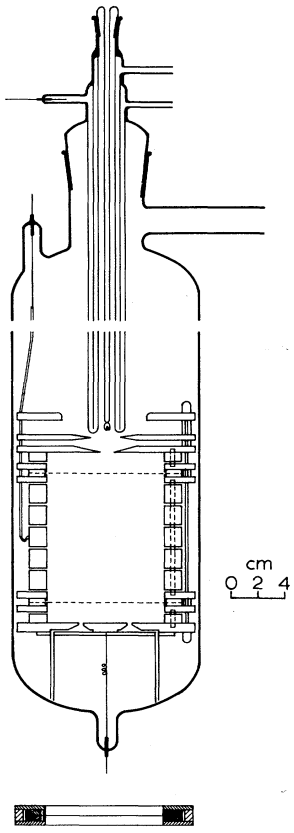

FIG. 7. Schematic diagram of a drift tube employed by Crompton and co-workers (Crompton, Elford, and Jory, 1967).

itself is obtained from a solution of the Boltzmann equation, which of course also depends upon σ_{MT}.

Obviously some sort of unfolding procedure must be employed. Where more than one type of interaction is involved, e.g., inelastic cross sections, this procedure must necessarily be fairly complicated and need not be unique. The situation is certainly simpler when one has reason to expect that only a single reaction can occur, that is, when only the (elastic) momentum-transfer cross section contributes to the transport properties. Uniqueness is then circumvented basically by the use of trial and error. This is discussed in more detail in Sec. 3.2.

We shall not attempt to discuss all aspects of the subject of swarms. For practical reasons we shall confine our considerations to experiments where only elastic collisions occur, so that it is easier to compare results with those obtained in low-energy beam experiments. This necessarily restricts this discussion to swarm experiments in atomic gases, primarily the rare gases, since even at the lowest temperatures attainable rotational excitations contribute to transport properties in molecular gases.

A basic gas parameter in these measurements is the quantity E/N, the ratio of applied (uniform) electric field E to the (uniform) gas density N. Under steady-state conditions the mean increment in kinetic energy acquired by an electron between collisions as a result of the applied electric field is a function of E/N, and so any macroscopic property of the gas which is controlled by this quantity will also be a function of E/N, that is, will be independent of E and N, provided the ratio is kept constant. In this paper we will then be concerned with the "low" E/N range, where only a negligible fraction of electrons possess energy sufficient to undergo inelastic collisions.

The environment is assumed to be a very weakly ionized gas, with electrons being the sole carriers of negative charge. The kinetic theory analysis, i.e., the solution for the electron velocity distribution function, is based upon a first-order perturbation solution to the binary Boltzmann collision integral, assuming elastic collisions only between electrons and the neutral gas (Allis, 1956). The perturbation is caused by the applied field and results in an anisotropic distribution function, $f=f_0+f_1$, where f_0, f_1 are the isotropic and anisotropic parts of f, and it is assumed that $f_1 \ll f_0$. However, f_0 need not be Maxwellian, and in fact will not be even approximately Maxwellian when E/N is large. For example, the well-known Druyvesteyn distribution results when one assumes that the momentum-transfer cross section is velocity independent.

Use of this kinetic-theory analysis and measurements of transport properties to determine momentum-transfer cross sections is well illustrated by the recent paper of Crompton, Elford, and Jory (1967) (hereafter referred to as Crompton *et al.*), in which the momentum-transfer cross sections of helium in the energy range of 0.02 to 3 eV were obtained by this means. This paper is particularly significant since it represents a refinement of measurement and analysis to the point where the authors claim an accuracy of better than $\pm 2\%$ over this entire energy range. After making allowance for the difference between σ_{MT} and the total cross section σ, this accuracy is even higher than the best accuracy

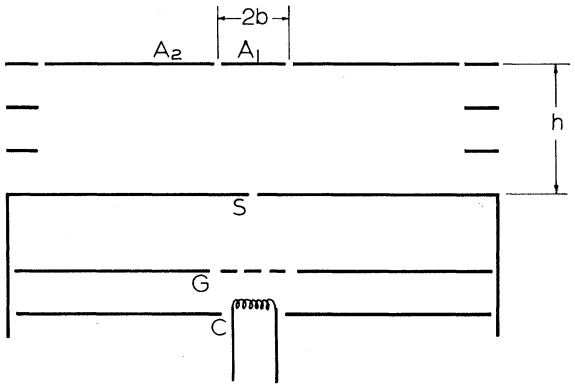

FIG. 8. Schematic of apparatus for measuring $D_\perp/\mu$ (Huxley and Crompton, 1962). C is the electron source; G is the control grid; S is the entrance aperture to the drift chamber; A_1 and A_2 are the concentric anodes. h is the length of the drift space and 2b is the diameter of the central anode.

currently claimed in a beam experiment, that of Golden and Bandel (1965), also for helium. As will be seen, however, the results of Crompton *et al.* and Golden and Bandel do not agree to within the combined quoted errors although the difference is relatively small.

For low E/N in helium it is reasonable to assume that only elastic collisions can occur. The procedure used by Crompton *et al.* can then be described as follows. First, the set of observables $W_{||}$, $D_\perp/\mu$, and $W_\perp/W_{||}$ are measured over the appropriate range of E/N. These quantities are then calculated from transport theory, using a trial set of σ_{MT}'s, and the results of the calculation compared with the measurements. The calculations are then repeated using an adjusted set of σ_{MT}'s. This iteration process is continued until the agreement between the calculated and measured values of $W_{||}$ lie within the experimental error, for the entire range of E/N studied. Comparison of calculated and measured values of $D_\perp/\mu$ and $W_\perp/W_{||}$ then serve as consistency checks on the entire procedure. $W_{||}$ is used as the primary observable since it is felt that it can be determined with better precision than $D_\perp/\mu$. The experimental and theoretical reliability in the determination of $W_\perp/W_{||}$ is considered by Crompton *et al.* to be less reliable than in the other two comparisons.

In the calculation of $W_{||}$, use is made of a first-order perturbation solution to the Boltzmann equation (Lorentz approximation), which is assumed to be of the form

$$(eE_i/m)(\partial f/\partial v_i) = \partial f/\partial t_{coll}. \tag{16}$$

Thus, it is assumed to be time independent, and the spatial gradient term is neglected [the error introduced by neglecting this term is discussed by Parker and Lowke (1969), Lowke and Parker (1969), Parker (1963), and will be considered later]. The first-order solution for $f_0(v)$ from Eq. (16) is

$$f_0\left(v; \frac{E}{N}\right) = A \exp\left(-\int_0^v \frac{3mv\,dv}{3kT + M(e/m)^2\{v\sigma_{MT}\}^{-2}(E/N)^2}\right) \tag{17}$$

where m, M are the electron and atom mass, e the electronic charge, A a normalization constant, and f_0 the isotropic part of f. Note that f_0 reduces to a Maxwellian distribution for E/N equal to zero. $f_0(v; E/N)$ is first calculated for each E/N measured using Eq. (17). Using the set of trial σ_{MT}'s, $W_{||}$ is then calculated from the equation

$$W_{||} = -\frac{4\pi}{3}\left(\frac{eE}{mN}\int_0^\infty \frac{v^2}{\sigma_{MT}}\frac{\partial f_0}{\partial v}\,dv\right). \tag{18}$$

The $D_\perp/\mu$ ratio is given by

$$\frac{D_\perp}{\mu} = -\left(\frac{m}{e}\right)\int_0^\infty \frac{v^3}{\sigma_{MT}} f_0\,dv \Big/ \int_0^\infty \frac{v^2}{\sigma_{MT}}\frac{\partial f_0}{\partial v}\,dv. \tag{19}$$

The expression for $W_\perp/W_{||}$ is

$$\frac{W_\perp}{W_{||}} = \left(\frac{eB}{mN}\right) \times \int_0^\infty \left[\left(\frac{v}{\sigma_{MT}^2}\right)\frac{\partial f_0}{\partial v}\right]dv \Big/ \int_0^\infty \left[\left(\frac{v^2}{\sigma_{MT}}\right)\frac{\partial f_0}{\partial v}\right]dv. \tag{20}$$

For example, in the helium paper, Crompton *et al.* use as their trial set of σ_{MT}'s the values calculated by Frost and Phelps (1964) from earlier mobility measurements of Pack and Phelps (1961). The choice of such a set, however, is quite arbitrary; the final results are independent of these. A thorough analysis of all theoretical and experimental assumptions involved in these swarm experiments is beyond the scope of the present paper. Certainly these experiments do not possess the more direct relation between observable and cross section which is considered to be one of the virtues of beam experiments. Still, there are several very compelling arguments in favor of the use of dc swarm data in the determination of collision cross sections. Among these are, first, the fact that such measurements can be and have been made down to energies which are not yet accessible in beam experiments; second, the experimental method and the magnitudes of the signals involved are such that data can be obtained which possess good statistics, being of the order of 1%, for example, in mobility measurements at low E/N.[15] Third, the dc swarm experiment is in a sense more complete than the transmission experiment, where knowledge (usually unobtainable) of beam shape collection efficiencies is required. In the swarm experiment, σ_{MT} appears without any modifying factors in the determining relations [Eqs. (18)–(20)]. Finally, it can be convincingly argued that the quantity which is obtained in these experiments, the momentum-transfer cross section, is precisely what is needed in calculating other transport coefficients, and that beam measurements without complete angular distribution determinations simply cannot be used for this purpose.

The modern analysis of swarm data, i.e., the procedures employed by the Phelps and Crompton groups, relies heavily upon computers and would not have been feasible otherwise. The only feasible techniques prior to the use of computers was to assume σ_{MT} to be so slowly varying over the range of electron speeds represented in the distribution function that it could be placed outside

[15] This quality of good statistics, i.e., good internal consistency of the raw data, compared to the relatively poor statistical error in crossed-beam experiments, is of course a characteristic feature of the difference between bulk-type data and those obtained in directly observing individual events. The price usually paid in return for the relative ease of obtaining the raw data in bulk experiments is a correspondingly more complicated theory of the experiment.

the integral in Eqs. (17)–(20), or alternatively to assume an analytic form for σ_{MT} with a few adjustable parameters. The earlier data did not in general possess adequate accuracy to warrant such an analysis. For these reasons, earlier data will be considered to be primarily of qualitative value.

The reliability of cross section determinations obtained through measurement of transport properties clearly rests directly on the corresponding reliability of both the measurements of the transport properties themselves and on the theoretical analysis which connects these to the cross sections. Piecemeal considerations of the associated errors exist throughout the swarm literature (Crompton and Jory, 1962; Crompton, Elford, and Gascoigne, 1965; Burch and Huxley, 1967). Of specific interest to us are the errors associated with time-of-flight measurements of electron beams in gases as a function of E/N, since this is the basic data from which drift velocities, and consequently, momentum transfer cross sections are derived. The question of uniqueness will be discussed in Sec. 3.2 in connection with dc swarm experiments. We will not discuss the more general aspects of the uniqueness problem in swarm experiments, as these are largely irrelevant to the limited scope of this article.

Two particular types of possible errors have been analyzed in detail, relevant to time-of-flight electron swarm measurements. These are, first, the "shutter problem," that is, systematic errors resulting from various ways in which the electric shutters can distort the electron time-arrival spectrum, and second, the errors resulting from possible oversimplifications in the theory. An example of these is the neglect of the density-gradient term in the solution to the Boltzmann equation.

The shutter problem is discussed by Lowke (1962), particularly for the Bradbury–Nielson shutter, which is employed in most swarm work, including the recent work of Crompton. This device consists of two parallel networks of grid wires, separated by the drift space which contains the longitudinal, uniform electric field. Identical sinusoidal voltages placed across contiguous pairs of wires in the grid networks of both shutters results in a transmission current through the second shutter which depends upon the applied frequency f and the drift velocity W. The maximum in transmission current is reached when the transit time of the electron possessing the mean drift speed approximately equals the reciprocal of the fundamental frequency. The analysis is based upon the assumption that the shutters are opaque to electrons except during a very small time interval about the times when the sinusoidal voltage passes through zero. At other times the electrons are collected by either one set of the grid pairs or the other.

The current I passing through the second shutter is then approximately given by

$$I=C\left(\frac{f}{D}\right)^{1/2}\exp\left(-\frac{(h-W/f)^2 f}{4D}\right), \tag{21}$$

where only a single burst of electrons (i.e., no harmonics) has been assumed. Equation (21) is simply the standard solution to the one-dimensional diffusion equation with the boundary condition that electron density at the second shutter is assumed to be zero during the time the shutter is closed. This boundary condition is dealt with mathematically by allowing a negative image term of the correct strength, originating at $2h$ and traveling with a velocity W.

The most obvious correction to be applied to the analysis is due to the fact that the maximum I does not occur for $f=W/h$, but rather for

$$f=W/h+D/h^2. \tag{22}$$

This correction is easily taken into account.

Other correction factors considered by Lowke include backdiffusion (slow electrons caught by the first shutter as it closes), boundary condition errors, the changing "open" time of the shutter as the frequency changes, the change in the mean speed due to shutter effects, and the error caused by the density gradient. This latter effect will be discussed more fully below. For all of these effects, the errors are of the order of D/hW, where h is the length of the drift region. The errors can be made small by employing either sufficiently large h or large gas pressure.

A related problem is that of obtaining a correct mathematical formulation and solution for the distribution function, considered by Parker (1963) in connection with an analysis of D/μ measurements. In the usual solution to the Boltzmann equation which is employed in swarm work, transport is assumed to result purely from the interplay between energy acquired by electrons from the applied electric field and energy dispersed by collisions with the neutral gas. Parker goes beyond this formulation by including the "diffusion current," the transport resulting from the electron density gradients. The solution is not a complete one since some additional assumptions are made, and an analytic solution is obtained only for the case of an elastic cross section which possesses a $1/v$ velocity dependence (constant collision frequency). For this type of cross section, Parker obtains an analytic expression for the error estimate in D/μ caused by neglect of the density gradient which is

$$\frac{\delta(D/\mu)}{D/\mu}\approx\frac{1}{2}\frac{D}{\mu Ez}-\frac{1}{4}\left(\frac{\rho}{z}\right)^2, \tag{23}$$

where ρ and z are the radial distance from the beam axis and the axial distance from the beam source. A less complete analysis for the constant cross section case is made for which the error in the calculated electron density for limiting cases of low and high applied fields is obtained. Application of this calculation to momentum-transfer cross sections in the rare gases is discussed in Sec. 3.2. This problem has also been recently discussed in detail by Francey (1969).

Parker and Lowke (1969) have analyzed the validity

of the assumption usually made in time-of-flight measurements that the electron energy distribution in the electron pulse possesses no spatial dependence. This assumption is only exactly correct when the electron density is uniform. In practice the nonuniformity of the electron density results in difference values for longitudinal and transverse diffusion coefficients, with the difference depending quantitatively upon the energy dependence of the momentum-transfer cross section (in the case where only elastic collisions occur), and the strength of the applied electric field. This important result does not generally affect the swarm data discussed in this paper since these relate primarily to $D_\perp$ which is not influenced by this effect.

2.5 Microwave Experiments

In the late 1940's and early 1950's, Brown and his students at the M.I.T. Radiation Laboratory began to exploit the microwave techniques as a diagnostic to study electron processes in weakly ionized gases (Phelps, Fundingsland, and Brown, 1951). It was realized that an afterglow technique is particularly suited to the study of transient atomic processes in gases, an afterglow being defined as the state of a partly ionized system after the ionizing agent has been removed. Provided one waits an appropriate length of time for short-lived and metastable excited states to decay, and for the excited electrons to undergo many collisions with the heavy constituents of the gas so that higher modes to the solution of transport properties have decayed, the afterglow plasma relaxes into a quiescent state in which the electrons are assumed to be in equilibrium with the heavy constituents. This afterglow plasma of course undergoes change with time, and in particular the charge density decays as a result of various electron and ion removal processes. By passing microwave radiation through the afterglow plasma, the electron density and the conductivity can be measured as a function of time.

Performing a straightforward first-order perturbation solution to the Boltzmann equation, under the assumptions of (a) uniform densities of charged and neutral constituents, (b) a sinusoidal time-varying electric field as the only forcing term, (c) binary electron–atom collisions only (i.e., no plasma effects), Margenau (1946) derived expressions for the real and imaginary parts of the electric conductivity $\eta_r+i\eta_i$. When employing a microwave cavity or waveguide, these quantities must be averaged over the spatially varying electric field distribution (Gould and Brown, 1954) to obtain

$$\langle\eta_r\rangle=-\frac{4\pi}{3}\frac{e^2}{m\omega}\times\int_\tau\int_0^\infty\left(\frac{(\nu/\omega)v^3(\partial f_0/\partial v)\,dv}{1+(\nu/\omega)^2}\right)nE^2\,d\tau\Big/\int E^2\,d\tau \quad (24)$$

and

$$\langle\eta_i\rangle=\frac{4\pi}{3}\frac{e^2}{m\omega}\int_\tau\int_0^\infty\left(\frac{v^3(\partial f_0/\partial v)\,dv}{1+(\nu/\omega)^2}\right)nE^2\,d\tau\Big/\int E^2\,d\tau, \quad (25)$$

where n is the electron density, ω is the radian frequency of the applied field, f_0 is the isotropic part of the electron velocity distribution function, and E is the electric field strength in the cavity. ν is the velocity-dependent collision frequency for momentum transfer, defined as

$$\nu(v)=n_0v\int_0^\pi\int_0^{2\pi}\sigma(\theta,\phi)(1-\cos\theta)\sin\theta\,d\theta\,d\phi=n_0v\sigma_{MT}, \quad (26)$$

where n_0 is the neutral gas density and σ_{MT} is the momentum-transfer cross section. It is assumed that the gas background is stationary ("Lorentzian" gas).

If f_0 is independent of E, as is the case for weak electric fields, and if the effects of spatial gradients on f_0 are small, then the integral over velocity can be taken outside the integral over position to give

$$\frac{\langle\eta_r\rangle}{\langle\eta_i\rangle}=\frac{\eta_r}{\eta_i}=\int_0^\infty\left(\frac{(\nu/\omega)v^3(\partial f_0/\partial v)}{1+(\nu/\omega)^2}\right)dv\Big/\int_0^\infty\left(\frac{v^3(\partial f_0/\partial v)}{1+(\nu/\omega)^2}\right)dv. \quad (27)$$

Note that this result is independent of electron density and of electric field strength for small E. Thus, a phase measurement using either a wave guide or a cavity could be used to infer values of ν averaged over f_0. Again here, as in the dc swarm experiments, it is necessary to know f_0. Usually f_0 is assumed to be Maxwellian, corresponding to the temperature of the background gas, thereby eliminating the volume integrals in Eq. (27). The energy dependence of ν can be studied by varying the gas temperature and/or the strength of the probing field although attempts have also been made to study the energy dependence of ν by applying a second, heating pulse to the electrons to elevate their temperature above ambient (Anderson and Goldstein, 1956).

In principle the mean energy of the swarm can be varied by varying the applied field strength, f_0 being Maxwellian with

$$\langle v^2\rangle=2kT_g/em+2MeE^2/3m^3\omega^2,$$

where T_g is the gas temperature and it is assumed that $\omega^2\gg\nu^2$ or $\eta_r\ll\eta_i$. Actually it is very difficult to obtain quantitative energy dependence of σ_{MT} by this means because even in the fundamental mode E varies over the cavity (Gould and Brown, 1954). Some simple analytic form for the velocity dependence of ν is generally assumed in order to unfold the integral of Eq. (27). A better method of varying the energy, over a limited

range, is simply to heat or cool the gas volume. Gould and Brown (1954), for example, performed measurements on helium over the temperature range 77–700°K, as well as over a range of electric field strengths.

Electron loss mechanisms, especially wall diffusion, which is particularly significant for helium, cause the electron density in the afterglow to be a function of position in the cavity, so that the density does not cancel out in the ratio of Eq. (27). Resort must then be made to theory, for example, ambipolar diffusion theory, to calculate n. Even so, the σ_{MT} values reported using this technique by Phelps *et al.* and another by Gould and Brown for helium (see Table III) are in excellent agreement with the dc measurements of Crompton *et al.*

Many variations of the microwave method have been used. In all cases, however, the analysis and the experimental techniques are far more complex than in the relatively straightforward dc method. It is more difficult in ac swarms to employ the systematic iteration procedures developed by Phelps and Crompton when using field heating since one must take into account variations of the electric field strength, thermal conduction, etc. In addition, the use of an electric discharge, so commonly employed in afterglow work to establish the initial electron density distributions, possesses many features, particularly the role of metastable atomic states in governing electron relaxation, which are not clearly understood.

A considerable literature relating to cross section determinations by microwave techniques has accumulated during the past two decades.[16] Microwave measurements have also been made in active, rather than afterglow, plasmas, particularly in cesium,[17] that is, with a steady glow discharge operative. However, such measurements are prone to additional uncertainties, associated with the presence of excited states, etc. (see Sec. 2.7).

We mention here several specific problems with the microwave method. First, in almost all published work to date a simple analytic (power-law) form for $\sigma_{MT}(v)$ has been assumed, which of course may cause difficulties if there are rapid and strong variations of σ_{MT}. This is not a fundamental drawback, however, since the iterative techniques of Phelps could certainly be extended to the ac case. There are obvious difficulties concerning lack of knowledge of f_0 and the spatial variation of E^2 when electrons are nonthermal. Finally, we point out one additional problem, as an indication of the difficulties associated with an attempt to critically evaluate this type of data (Nighan, 1967). In order to obtain usable signals, the afterglow electron densities generally need to be larger than 10^8 electron/cm^3 (in some cases 10^{10} electron/cm^3). Under such conditions, what is the contribution of electron–ion collisions to η_T? An approximate estimate of the ratio $\sigma_{MT}(ei)$ to $\sigma_{MT}(en)$ is given by

$$\frac{\sigma_{MT}(ei)}{\sigma_{MT}(en)} = \frac{\ln \Lambda}{(kT/e^2)^2} \frac{f}{\sigma_{MT}(en)},$$

where $\Lambda = (3/2)e^3(k^3T^3/ne)^{1/2}$, n is the number density of the charged constituents, and f is the fractional ionization of the gas. In the above relation, $\sigma_{MT}(ei)$ is the effective electron–ion cross section for a 90° deflection (many small collisions). For helium at room temperature, assuming $\sigma_{MT} = 6\times10^{-16}$ cm^2,

$$\sigma_{MT}(ei)/\sigma_{MT}(en) \simeq 6\times10^4 f \ln \Lambda.$$

For example, if $f = 10^{-6}$, a not unusual fractional ionization, $\ln \Lambda$ is approximately 6 and the above ratio is about 36%. Thus, under normal operating conditions, the role of ion collisions may be significant and must somehow be corrected for. It then becomes necessary to determine the electron density as a function of both position in the cavity and of time, and this is, of course, not easy to do. Such an analysis has in fact been performed by Nighan (1967), for σ_{MT} measurements in weakly ionized cesium; he also attempted to bring earlier cesium swarm experiments into better harmony by applying similar corrections to them. (See also Chen, 1964.)

One is faced, in fact, with the more general problem of lack of knowledge of the exact nature of the afterglow constituents. Afterglow chemistry could produce large effects, for example, by adding low-level impurities with very large momentum-transfer cross sections.

It should be noted that the recent microwave measurements of Hoffmann and Skarsgard (1969) do in fact take account of many of the difficulties discussed above, particularly by, first, using an iterative technique involving a set of trial σ_{MT}'s rather than assuming a simple analytic form for σ_{MT}, and, second, by taking electron–ion (though not electron–electron) collisions into account. In addition, in this experiment the mean electron temperature was directly determined using a microwave radiometer. They have reported results on helium, neon, krypton, and xenon.

It is quite apparent that one cannot readily establish criteria for evaluation of cross sections obtained by this method. The consistency of σ_{MT} determinations obtained in different microwave experiments of course can give some indication of whether any gross features of the method are misbehaving. This is illustrated in Table III, which presents a summary of recent measurements in helium at 300°K, where one could reasonably expect laboratory conditions to be most amenable to reliable measurement. The six determinations were made by three groups and it is seen that the values have a 38% spread, which cannot be considered as too unreasonable.

A summary of the other principal microwave experi-

[16] Recent reviews of theory and experimental techniques include Goldstein (1955, p. 473) Brown (1959), Golant (1961), and Ginsburg and Gurevich (1960a, 1960b).

[17] See references for Table III.

ments is also presented in Table III, along with appropriate comments.

The microwave analysis can readily be extended to allow for the presence of a uniform, constant external magnetic field, usually applied in the axial direction of a cylindrical cavity. Adding this field introduces an additional characteristic frequency into the afterglow environment, namely, the cyclotron frequency $\omega_b = eB/mc$. The complex conductivity η then becomes a tensor, the transverse component of which is

$$\eta_T = \frac{4\pi ne^2}{3m}\int_0^\infty \frac{\nu(\nu^2+\omega^2+\omega_b^2)-j\omega(\nu^2+\omega^2-\omega_b^2)}{[\nu^2+\omega^2+\omega_b^2][\nu^2+(\omega-\omega_b)^2]} \times\left(v^3\frac{\partial f_0}{\partial v}\,dv\right), \quad (28)$$

assuming uniform electron density in the cavity. Several different uses can be made of this relation. One method is to measure the pressure dependence of the cyclotron absorption resonance frequency (see review article by Allis, 1956; also, Shkarofsky, 1961; Fehsenfeld, 1963; Kelly, Margenau and Brown, 1957), which is a function of ν^2. Another method, used by Hirshfield and Brown (1958), involves the observation that, according to Eq. (28) the imaginary part of η_T can be made zero by establishing a suitable value of ω_b, which can be varied by the experimenter. In any case, an analytic form of the velocity dependence of ν is usually assumed, as well as a form for f_0 (usually Maxwellian). The velocity dependence is traditionally assumed to be of the form

$$\nu = Cv^h. \quad (29)$$

For example, if h is assumed zero (velocity-independent ν), the linewidth is Lorentzian and equal to ν^2. η_T is then real when $\nu^2=\omega_b^2-\omega^2$. Problems similar to those discussed in the previous section arise here as well, plus some others. For example, as Hirshfield and Brown point out, it is necessary to ensure that only those modes get set up for which E is precisely perpendicular to B throughout the cavity since otherwise the measured η would not correspond to the transverse component at η. This is usually accomplished by using a TE mode in a cylindrical cavity. The use of the simple Eq. (29) for the velocity dependence of ν may be quite unrealistic at low energies, as already mentioned.

The most commonly used cyclotron method, however, is that of relating the cyclotron absorption resonance half-width to the collision frequency. Kelly, Margenau, and Brown (1957) showed that the half-width at resonance is equal to the collision frequency, provided it is velocity independent. The proof is based upon use of the elastic binary collision integral, that is, inelastic collisions are not considered in the analysis. Since inelastic energy loss, particularly through rotational excitation, at low electron energies, may be significant and can, in fact, dominate relaxation phenomena in gases, this technique can only be applied in this simple form to atomic systems. A more complicated relation exists between half-width and ν for other velocity dependencies.

Recently Tice and Kivelson (1967a, 1967b) have used this technique to measure collision frequencies in many simple molecules and in the rare gases. In these experiments a flowing afterglow technique (Fite, 1968) was used in which the carrier gas, which is N_2 with a small additive of O_2, is broken down to produce a plasma, which flows downstream and thermalizes, presumably, as it travels. The gas to be studied is added to the carrier downstream. The collisional half-width due to N_2 is first measured, and subtracted off the full half-width to obtain the contribution due to the additive. The effects of the N_2, unfortunately, are not fully understood since there are residual metastables and atomic N in the test sample. In addition, the effects of inelastic collisions in N_2 may then affect the ν determinations of the atomic additive (see notes in Table III).

2.6 Optical Line Shift (Fermi Method)

Of the various mechanisms responsible for the broadening and displacement of spectral lines, there is one which can be attributed directly to low-energy electron elastic scattering by ground-state atoms. Amaldi and Segre (1934) and Fermi (1934) noted that when an atom in a highly excited state, where the orbiting electron is very weakly bound, collides with a ground-state atom, the interaction can be considered to be an elastic collision between a quasifree electron and an atom. If the range of the interaction is small compared to the size of the circumferential electron orbit, the net effect on the electron is to introduce a phase shift in its unperturbed wave function after being scattered which is precisely equal to the elastic scattering phase shift. For the electron to remain in a stationary state it must therefore adjust its orbit slightly, thereby resulting in a small energy change of the excited state. The effect can be either positive or negative, depending upon whether the phase shift is positive or negative, i.e., whether the interaction is effectively attractive or repulsive.

If one assumes a pure s-wave elastic interaction (i.e., sufficiently highly excited states) and neglects the polarization of the excited state by the perturbing atom, the frequency shift is

$$\Delta\nu = \pm(\hbar N/2m)(\pi\sigma)^{1/2}, \quad (30)$$

where N is the number density of the perturbing gas and σ the total elastic cross section, at the speed appropriate to the specific excited state of the bound electron.

In practice the excited states have usually been produced in alkali vapors and the perturbing systems have been high concentrations of rare gases, so that such measurements have mainly yielded low-energy cross sections for rare-gas scattering. Fuchtbauer and

his co-workers (Fuchtbauer, Schulz, and Brandt, 1934; Fuchtbauer and Gossler, 1935; Fuchtbauer and Reimers, 1935) used this technique to study Ar, Ne, He, N_2, Hg, Xe. Recently interest in the "Fermi method" has been revived in the USSR with theoretical analyses by Firsov (1951a, 1951b) and Alekseev and Sobel'man (1966), and experimental work by Mazing and Vrublevskaya (1966), and by Alekseev, Mazing, Serapinas, Sobel'man, and Vainshtein (1967), who studied Ar and Cs. The theory has been extended to take into account contributions by higher partial waves, and to make more quantitative estimates of the polarization effects.

This very interesting subject obviously possesses considerable relevance to the low-energy collision problem and deserves further study (Mittleman, 1967; Choudhury, 1969; Presnyakov, 1970; Bederson, 1970). However, for the purposes of this review the numerical values thus far obtained are primarily of qualitative value, and the method must await more detailed theoretical and experimental investigation.

2.7 Measurements in Active Systems

The diagnostic methods employed in swarm experiments do not require the medium to be in a passive state; that is, measurements can be made while an ionizing agent is present rather than in the afterglow. It is obvious that during this active phase, with a discharge, a shock, or other drastic means of producing a plasma operative, a very complicated situation exists. An attempt to study a single reaction such as that attributable to elastic electron–atom collisions under such conditions requires massive application of theoretical analysis and experimental ingenuity. And in the end, one is also invariably required to make certain assumptions which cannot be fully documented, for example, regarding the roles played by excited states, and the velocity distribution functions of the various neutral and charged constituents.

Because of experimental difficulties associated with the employment of ac techniques under active discharge conditions, most such measurements have been done using dc techniques. Several measurements have been made using alkali metal vapors, but the most interesting application of this technique has been to study unstable atomic systems, in particular O and H which are normally not present in the parent molecular gases. Conditions are generally such that very high effective temperatures prevail in the active medium. Four measurements on atomic hydrogen have been made (Maecker, Peters, and Schenk, 1955; Drawin, 1956; Kolesnikov and Obukhov-Denisov, 1962; Wiese, 1963), all using dc arcs, and two on atomic oxygen (Maecker, Peters, and Schenk, 1955; Lin and Kivel, 1959), using shock-induced plasmas, with varying results.

We will briefly discuss the atomic hydrogen measurements here as a group, as representative of this type of experiment. In these works, the dc conductivity is assumed to be

$$\eta=[e^2N_e/(3mkT)^{1/2}][K/(\sum N_a\langle Q_a\rangle+\sum N_i\langle Q_i\rangle)]. \tag{31}$$

This equation is a variation of Eq. (24) with ω set equal to zero. N_e, N_a, N_i are the electron, atom, and ion densities, with the sum performed over all neutral and ionic species, including molecular constituents and excited states, and K is a calculated coefficient of the order of unity. The cross sections $\langle Q_a\rangle$ and $\langle Q_i\rangle$ are the appropriate effective momentum-transfer cross sections averaged over the electron energy distribution. In Eq. (31) it is assumed that an effective (and unique) temperature can be attributed to the active medium, and that the electrons possess this same temperature.

Because dc cylindrical arcs were used, where densities and temperature are presumed to vary in the radial (r) direction, but not in the azimuthal direction, Eq. (31) must actually be averaged over r, i.e., is used in the form

$$\eta_{\text{eff}}=\frac{2\pi e^2}{(3mk)^{1/2}}\int_0^R\frac{N_eKr\,dr}{T^{1/2}[\sum N_a\langle Q_a\rangle+\sum N_i\langle Q_i\rangle]}, \tag{32}$$

where all quantities underneath the integral are functions of r. Here $\eta_{\text{eff}}=J/E$, i.e., the arc current density divided by the electric field strength, which is assumed constant throughout the plasma volume. Thus the measurement of $\langle Q_a\rangle$ requires determinations of the neutral and ionic densities and electron temperature as functions of r, as well as the total arc current and electric field strength. Wiese (1963), for example, used the values measured in an earlier experiment using a similar arc. The effective electron–ion total cross sections were obtained from standard plasma theory (Spitzer, 1962). In all four experiments, the distribution functions were assumed to be Maxwellian, the contributions of excited states can be orders of magnitude higher than that of the ground state, so that a very small concentration of such states will contribute a significant, and perhaps even a dominant, amount to an effective momentum-transfer cross section. In view of the large currents and high power dissipation of these arcs, such states must certainly be present in copious amounts and must be taken into account in a complete analysis. It should also be noted that Eqs. (31) and (32) do not take into account electron–electron collisions, which do indeed play a significant role in such experiments (Johnson, 1962; Shkarofsky, Bernstein, and Robinson, 1963; Schweitzer and Mitchner, 1966).

Again one is forced to the conclusion that values derived from such experiments do not satisfy minimum criteria of reliability.

Brief mention should also be made of the active measurements in cesium, usually in a low-intensity glow discharge or a plasma diode. These environments are

rather more quiescent than those of arc discharges. However, there is even here adequate reason to suspect serious difficulties. Published cross section values in about 10 independent investigations yield cross sections ranging from 1.5 to 10×10^{-14} cm^2 in the neighborhood of 0.05 to 0.15 eV, with no convincing means available for selecting one over the others. Nighan (1967) has shown that these results can be brought into better agreement by attributing the differences to small (between 10^{-1} and 10^{-6}) but varying fractional ion concentrations (see Sec. 2.5). These contribute importantly to the measured conductivity, but have not been corrected for in the original papers.

2.8 Time of Flight

In this method a pulse of electrons at a fixed energy is introduced into the scattering region at $t=0$. The arrival-time spectrum is observed with and without gas present in the scattering chamber, using a fast detector and suitable data processing. The difference in the spectra with and without gas is attributable to scattering out, so that this technique measures the total effective cross section, similar to that defined by Eq. (6). This technique, routinely exploited in nuclear physics, has only begun to be explored by atomic physicists (Nakai, LaBar, Harter, and Birkhoff, 1967).

The most complete study using time of flight has been made by Baldwin (Baldwin and Friedman, 1967) for helium. This work is in a continuing state of development. A similar experiment (using, however, a longitudinal confining magnetic field) is in progress using low-energy positrons, by a group at Gulf General Atomic (Groce, Costello, McGowan, and Herring, 1969). Several other groups have also begun similar studies, but are only in the very early stages of their experiments. Thus, quantitative results from this interesting technique are not yet available; critical comment at this time would be premature.

3. DISCUSSION OF TOTAL CROSS SECTION DATA

3.1 General Considerations

In attempting to critically evaluate total cross section data, one is faced with a difficult task indeed. Even were one for the moment to forego the "conventional" evaluation criteria (e.g., adequacy of experimental techniques, gas purity, pressure measurement problems), one is still faced with a fundamental question. This relates to the fact that, perhaps with the exception of crossed-beam experiments (and the actual available data obtained using this method are still relatively scarce), and the dc swarm experiment, the theories of the experimental methods used in this field have not really been thoroughly explored.

As an illustration of this difficulty, consider the Ramsauer method. One does possess an intuitive feeling that this type of transmission experiment should be adequate to its task of measurements of a total collision cross section. The narrow collimation of the electron beam, to begin with, results in a relatively tolerable angular resolution which, however, is not really specified. In addition, the transverse magnetic field serves as a momentum selector, in the sense that elastic scattering out of the plane of the trajectory of the electron beam, and all inelastic collisions, result in a reduced radius of gyration as well as a change in position of the gyrating center of the scattered electron. As a consequence, it is reasonable to suppose that relatively few scattered electrons reach the collector, i.e., that the efficiency of the Ramsauer experiment for the counting of scattering events is relatively high.

The difficulty enters in attempting to replace these qualitative considerations by a quantitative analysis of the collection efficiency as a function of scattering angle for elastic and inelastic collisions. Such an angular resolution analysis has not been made for the Ramsauer experiment, despite the half-century history of the method. Golden and Bandel have made a partial analysis of a simplified transmission experiment, in which the effect of the magnetic field was not considered. Again, one expects intuitively that adding the magnetic field should improve the angular resolution. Still it can readily be shown that the magnetic field can cause a decrease in angular resolution under some (perhaps extreme) conditions.[18] Furthermore, it is also apparent that at sufficiently high energies where the energy loss of inelastically scattered electrons is small compared to its original energy, and where most of the scattering is small angle, the Ramsauer method must fail completely. Thus at very high energies one would expect the measured cross section to approach zero too rapidly, i.e., to yield erroneous results. Since Ramsauer experiments have been performed at energies of up to 400 eV, there is reason to suspect that the angular resolution is not in fact always adequate to the task.

Until very recently the situation regarding low-energy swarm experiments was equally unsettling. The critical element in the theoretical analysis is the calculation of the velocity distribution of the electron swarm. Once this is obtained, transport properties can be calculated and compared with experiment. However, there is no convenient method available to directly measure this distribution. Thus the principal check that the experimentalist has on his cross section determination is consistency, that is, the degree of agreement obtained over as wide a range of energies and experimental parameters as possible, between calculated and measured transport properties (and as large a number of these as possible as well). The direct measurement of

[18] An electron will have both the magnitude and orientation of its guidance center altered by an inelastic collision. The combination of these two changes could cause an electron, which would otherwise miss it, to pass through the aperture into the anode region, i.e., to not be counted as a scattering event.

the electron velocity distribution remains an elusive goal because of the formidable experimental difficulties such a determination entails.

One consequence of such indirect procedures is the generally vague estimates of experimental error associated with the cross section determinations in swarm experiments. Firm claims of error estimates are rarely encountered in the swarm literature, and indeed such claims would in most cases be unrealistic and difficult to justify.

Nevertheless in recent years the low-energy swarm technique has made substantial progress in one particular area, that of the Townsend-type drift experiment, in which mobility and diffusion measurements are made in the presence of weak electric, and sometimes magnetic, fields. In addition to the improved accuracy in the measurements of the transport properties themselves, the most significant advance has been in the greatly increased sophistication of the analytic methods developed by groups at Westinghouse (Phelps, 1968) and the Australian National University (Huxley and Crompton, 1962; Crompton, 1969). The Westinghouse group has been interested in simple molecular gases (e.g., N_2, H_2), as well as the rare gases. The group at the Australian National Laboratory has been particularly interested in obtaining highly accurate transport coefficients, particularly in the light rare gases, in order to determine momentum-transfer cross sections with high accuracy. The molecular gas analysis is far more complicated than the corresponding rare-gas case because of the need to include inelastic (rotational, vibrational, and electronic) excitation even at very low energies and cannot at this time be asserted to yield the type of precision obtainable in the rare gases, where only elastic scattering obtains in most of the low-energy domain.

From the point of view of this article, which is concerned principally with total collision cross sections, only those swarm measurements which can be directly related to these through knowledge of the angular distribution will be discussed here. These can be known either by direct measurement or by theory, particularly in those cases where scattering occurs at energies sufficiently low so that only two or three partial waves at most contribute to the elastic scattering. Thus we restrict ourselves in this article to only those swarm measurements which yield elastic-momentum-transfer cross sections, where this condition is generally best (though by no means completely) satisfied. The low-energy rare-gas studies using the swarm technique satisfy this criterion and will be discussed in the next section.

Table I contains a list of total cross section experiments for all atomic species and homonuclear diatomic molecules that have been measured (Kieffer, 1967; Chamberlain and Kieffer, 1970). The experiments listed were selected on the basis of our conclusions about the measurement techniques (see Conclusions and Appendix). In cases, for the atomic species, where only one or two measurements have been reported, they are included in spite of any reservations about the techniques used.

3.2 Atomic Hydrogen and Helium

The most obvious system deserving in-depth study is atomic hydrogen, because of the basic nature of the three-body interaction which it represents, as well as the relative simplicity of three-body theory compared to the many-body theory required for other atomic and molecular systems. In fact an impressive body of literature specifically concerned with elastic scattering of atomic hydrogen by low-energy electrons has developed during the past two decades. No fewer than 130 papers on this subject are listed in the National Bureau of Standards Publication No. 289, "Bibliography of Low Energy Electron Collision Cross Section Data" (Kieffer, 1967)! Of these, a number of recent calculations of elastic phase shifts are particularly relevant to this discussion. Schwartz (1961) calculated S-wave phase shifts using a variational principle technique. Combined with the P-wave phase shifts of Armstead (1968), these calculations yield total elastic cross sections for hydrogen which are in substantial agreement with other more recent calculations, performed using a variety of methods. These include a variational calculation of Gailitis (1965), a "polarized orbital" calculation of Temkin and Sullivan (1963), a recent determination of upper *and* lower bounds by Madan (1968), and many variants of close-coupling-type calculations (Ormonde, Whitaker, Heubner, and Burke, 1969; Burke and Schey, 1962; Burke, Gallaher, and Geltman, 1969). All of these calculations are in agreement to better than 10%, when compared properly, i.e., when singlet and triplet contributions and higher angular momentum states (particularly the P state) are taken into account. Because of the consistency of the theoretical results, particularly in view of the relative simplicity of this particular problem, it is generally agreed among theorists that the present state of knowledge of the total elastic cross section for electrons on atomic hydrogen, between 0 and 10 eV, certainly possesses an accuracy of better than $\pm 10\%$ (Eisner, 1969).

Experimentally, however, atomic hydrogen is so difficult to study that only a handful of measurements have been made on it, and none of these possess an accuracy comparable to that of the better recent calculations (see Table I). The principal problem is the need to dissociate H_2 in order to produce H, either by means of a very high temperature oven or by a discharge. Thus, electron beam–gas and simple swarm experiments are precluded. A summary of the data from the atomic hydrogen experiments quoted in Table I is presented in Fig. 9.

The theoretically most simple atom which at the same time can be employed in static volume experi-

TABLE I. Summary of best available total and momentum-transfer cross sections for atoms and diatomic molecules.

System	Method	Type of cross section measured	Energy range (eV)	Authors	Footnotes
One-electron atoms					
H	Crossed beam scattering in	Total	1–10	Brackmann and Fite (1958)	a
	Crossed beam, scattering out (transmission)	Total	3.1–12.3	Neynaber, Marino, Rothe, and Trujillo (1961b)	b
	Crossed beam recoil, scattering out	Total	0.7–10.5	Eisner (1969)	c
Li	Crossed beam recoil	Total	0.5–10	Perel, Englander, and Bederson (1962)	d
Na	Ramsauer–Brode	Total	1–400	Brode (1929a)	e
	Crossed beam recoil	Total	0.5–10	Perel, Englander, and Bederson (1962)	f
K	Ramsauer–Brode	Total	1–400	Brode (1929a)	e
	Crossed beam recoil	Total	0.5–10	Perel, Englander, and Bederson (1962)	f
	Crossed beam recoil	Total	0.4–9	Collins, Bederson and Goldstein (1971)	ad
	Crossed beam recoil	Total	0.3–9	Visconti, Slevin, and Rubin (1971)	ad
Rb	Ramsauer–Brode	Total	1–400	Brode (1929a)	e
	Crossed beam recoil	Total	0.3–9	Visconti, Slevin, and Rubin (1971)	ad
Cs	Ramsauer–Brode	Total	1–400	Brode (1929a)	e
	Crossed beam recoil	Total	0.3–9	Visconti, Slevin, and Rubin (1971)	ad
Rare gases					
He	Ramsauer	Total	0.6–40	Ramsauer (1921b)	g
	Ramsauer	Total	1.2–49	Bruche, Lilienthal, and Schrodter (1927)	h
	Ramsauer	Total	0.15–2.3	Ramsauer and Kollath (1929)	i
	dc swarm	Momentum transfer	0.003–30	Frost and Phelps (1964)	j
	Ramsauer	Total	0.3–28	Golden and Bandel (1965)	k
	dc swarm	Momentum transfer	0.025–6	Crompton, Elford, and Jory (1967)	l
He*($1s2s^3S_1$)	Crossed beam recoil	Total	0.87–8.3	Neynaber, Rothe, Trujillo, and Marino (1964)	m
Ne	Ramsauer	Total	1.5–40	Ramsauer (1921b)	g
	Ramsauer	Total	1.2–49	Bruche, Lilienthal, and Schrodter (1927)	h
	Ramsauer	Total	0.15–1.4	Ramsauer and Kollath (1929)	i
	Ramsauer	Total	0.37–20	Salop and Nakano (1970)	k
Ar	Ramsauer	Total	0.6–36	Ramsauer (1921b)	g
	Ramsauer	Total	0.8–64	Ramsauer (1923)	n
	Ramsauer	Total	1.2–49	Bruche, Lilienthal, and Schrodter (1927)	h
	Ramsauer	Total	0.15–1.0	Ramsauer and Kollath (1929)	i
	Crossed beam recoil	Total	1–25	Aberth, Bederson, and Sunshine (1964)	o
	dc swarm	Momentum transfer	0.003–30	Frost and Phelps (1964)	j
	Ramsauer	Total	0.1–21.6	Golden and Bandel (1966)	p
Ar*	Crossed beam recoil	Total	1.0–7.0	Celotta, Brown, Molof, and Bederson (1971)	q
Kr	Ramsauer	Total	0.8–100	Ramsauer (1923)	n
	Ramsauer	Total	0.17–1.6	Ramsauer and Kollath (1929)	i
	dc swarm	Momentum transfer	0.003–30	Frost and Phelps (1964)	j
Xe	Ramsauer	Total	0.8–100	Ramsauer (1923)	n
	Ramsauer	Total	0.15–1.2	Ramsauer and Kollath (1929)	i
	dc swarm	Momentum transfer	0.003–30	Frost and Phelps (1964)	j

TABLE I (*Continued*)

System	Method	Type of cross section measured	Energy range (eV)	Authors	Footnotes
Metals					
Zn	Ramsauer–Brode	Total	1–400	Brode (1930)	r
Cd	Ramsauer–Brode	Total	0.64–400	Brode (1930)	r
Hg	Ramsauer–Brode	Total	0.5–400	Jones (1928)	s
	Ramsauer–Brode	Total	0.64–400	Brode (1929b)	r
Tl	Ramsauer–Brode	Total	0.8–100	Brode (1931)	r
Other atoms					
N	Crossed beam	Total	1.6–10	Neynaber, Marino, Rothe, and Trujillo (1963)	t
O	Crossed beam	Total	2.3–11.6	Neynaber, Marino, Rothe, and Trujillo (1961a)	u
	Crossed beam recoil	Total	0.5–100	Sunshine, Aubrey, and Bederson (1967)	v
Homonuclear diatomic molecules					
H_2	Ramsauer	Total	2.25–49	Bruche (1927a)	w
	Ramsauer	Total	0.15–1.0	Ramsauer and Kollath (1930)	x
	dc swarm	Momentum transfer	0.003–2	Frost and Phelps (1962)	y
	dc swarm	Momentum transfer	0.007–30	Englehardt and Phelps (1963)	z
	Ramsauer	Total	0.25–15	Golden, Bandel, and Salerno (1966)	aa
D_2	dc swarm	Momentum transfer	0.007–30	Englehardt and Phelps (1963)	z
	Ramsauer	Total	0.25–15	Golden, Bandel, and Salerno (1966)	aa
N_2	Ramsauer	Total	2.25–49	Bruche (1927a)	w
	Ramsauer	Total	0.15–1.5	Ramsauer and Kollath (1930)	x
	dc swarm	Momentum transfer	0.003–2	Frost and Phelps (1962)	y
	dc swarm	Momentum transfer	0.007–30	Englehardt and Phelps (1963)	z
	Crossed beam recoil	Total	1–25	Aberth, Bederson, and Sunshine (1964)	o
	Ramsauer	Total	0.3–5	Golden (1966)	ab
O_2	Ramsauer	Total	1–48	Bruche (1927b)	ac
	Ramsauer	Total	0.15–1.4	Ramsauer and Kollath (1930)	x
	Crossed beam recoil	Total	1–25	Aberth, Bederson, and Sunshine (1964)	o
	Ramsauer	Total	2.35–21	Salop and Nakano (1970)	k

[a] A thermally dissociated atomic hydrogen beam was cross fired by an electron beam. Dissociation fractions were 90% to 96%. Observations were made of electrons scattered into a cone of 45° half-apex angle with its axis at 90° to the electron beam. Normalized cross sections were obtained by making use of the relation

$$\sigma(\mathrm{H}) = \sigma(\mathrm{H_2})\,(1/\sqrt{2}D)\,[(S/S_r)\,(T/T_r)^{1/2} + D - 1],$$

where $\sigma(\mathrm{H})$ and $\sigma(\mathrm{H_2})$ are the cross sections of the hydrogen atom and molecule, respectively, for scattering into the cone of observation, D is the dissociation fraction, S is the total scattering-in signal at the temperature T, and S_r is the reference scattering signal due to H_2, only, at the temperature T_r. In order to obtain $\sigma(\mathrm{H})$, the differential cross sections of Ramsauer and Kollath (1932) were integrated over the angles appropriate to the observing cone. $\sigma(\mathrm{H})$ was then converted to the absolute total elastic cross section in two ways: (1) assuming only s wave, i.e., isotropic scattering, (2) assuming a P-wave contribution to the scattering as calculated by Bransden, Dalgarno, John, and Seaton (1958). Statistical error in the experiment amounted to 20%. See Fig. 9.

[b] An rf-discharge atomic-hydrogen source was used, with the temperature of the source measured by scattering from a krypton additive to the beam, as well as a thermocouple. The experiment was performed in transmission (scattering out) with absolute values obtained by normalization to the total H_2 cross section [an average of Bruche (1927a) and Normand (1930)] using the relation shown in Footnote a. A small correction was applied because Knudsen (effusive flow) conditions were not completely satisfied at the source exit. Actual data points were not shown in paper, but statistical deviations were as follows: half of experiments' points lie within ±9% of the average curve below 7.24 eV, and within ±6% above 7.25 eV. Results are consistent with Brackmann and Fite (see Footnote a), with the mean curve lying perhaps 10% below the mean of the Brackmann–Fite results. The angular resolution was calculated to be 25%. No direct correction was made for this error but since the ratio of the scattered current due to H_2 and that due to H were taken as the ratio of the total cross sections and independent total cross sections for H_2 were used to obtain the normalized atomic hydrogen cross sections, this error was at least partially corrected. See Fig. 9.

[c] A microwave, cooled discharge source was used. The temperature was measured using an argon-tracer technique and a thermocouple. Absolute values were obtained without normalization (see Sec. 2.3). Angular resolution was quoted as <6°, and the absolute accuracy was estimated to be ±25%, including statistical and systematic errors. See Fig. 9.

[d] A double oven was used, employing Li and K. The Li cross sections were normalized by obtaining Li cross sections relative to K and using Brode (1929a) absolute total cross sections. Because of the present uncertainty regarding the Brode data (see Footnote c and Sec. 3.4 of the text), a renormalization of the Li data downward by a factor of about 2 is probably required.

[e] The Brode alkali experiments are discussed in Sec. 3.4, while the Brode modification of the Ramsauer technique, and difficulties therewith, are discussed in Sec. 2.1. Present indications are that the Brode absolute values may be too high by about a factor of 2, and that the pronounced structure in the vicinity of the first excitation energy is at least partly spurious.

[f] This is a relative recoil crossed-beam experiment performed in the scattering-out mode. The absolute value measurement at 13 eV is probably too high by a factor of 2 due to reflected electrons from the anode.

[g] This is the classic Ramsauer paper. It is the only Ramsauer work on He and Ne apart from the later extension to very low energies by Ramsauer and Kollath (see Sec. 3.2 of the text).

[h] The Bruche experiments, using a conventional two cage Ramsauer apparatus, were performed with considerable care and skill. The Bruche

TABLE I (*Continued*)

data in general show good statistics; some points were measured with extreme accuracy in order to supply absolute "check points." However, not all the Bruche data were obtained using a straightforward transmission technique. At low energies, in some cases, he used an indirect method which relied upon an analysis of the change in shape of the electron energy distribution caused by the scattering gas. Bruche characterized these data as "qualitative"; usually, however, no distinction is made between the "qualitative" and "quantitative" Bruche data when they are presented in the literature.

[i] These are the famous Ramsauer–Kollath experiments—a true tour de force, considering the available technology. A modification of the Ramsauer method was used to extend the measurements down to several tenths of an eV, with good energy resolution. The method was similar to that employed by Bruche (see Footnote h). The Ramsauer effect was exhaustively explored in Ar, Kr, and Xe. This work can be judged to be of qualitative value only, because of the extreme difficulties associated with low-energy electron beams, particularly considering the relatively crude vacuum and metallurgical techniques available at the time. The "oscillating structure" observed in helium is very likely spurious (see Sec. 3.2).

[j] This paper employs previously obtained transport data, particularly drift velocity and D/μ coefficients measured by Pack and Phelps (1961) to obtain momentum transfer cross sections for the rare gases, excluding neon (see discussion in Secs. 2.4 and 3.1).

[k] The Golden and Bandel experiment is the first in a series of measurements using a modern version Ramsauer apparatus by Golden and coworkers at Lockheed. This experiment is discussed in detail in Sec. 3.2. The Salop experiment was performed using the same apparatus.

[l] This work is discussed in detail in Sec. 3.2. It is the most precise dc transport measurement of the He momentum transfer cross section performed to date.

[m] This experiment employed state selection, using a Stern–Gerlach magnet, to distinguish the metastable $1s2s^3S_1$ from the $1s2s^1S_1$ state in helium. The experiment was done using the recoil technique in transmission (scattering out). The results presented were only preliminary ones, with extremely large statistical spread. The experiment was discontinued before final results could be obtained.

[n] This is a continuation of the work reported in Ramsauer (1921b). The deep minimum and position of the peak of the Ar cross section were reconfirmed and the measurements were extended to Kr and Xe.

[o] This recoil experiment was done in the scattering-out mode. There is the possibility of reflected electrons from anode re-entering interaction region, causing measured cross-section to be somewhat high (by perhaps 10%–20%). Angular resolution average ranges from about 28° at 1 eV to about 12° at 25 eV.

[p] See Footnote k. These data are in very good agreement with Bruche, Lilienthal, and Schrodter (1927) (within 5%). A thorough investigation of the Ramsauer minimum region yielded a sharper and somewhat deeper Ramsauer minimum than previous work.

[q] Total cross section for metastable $3p^54s^3P_{2,0}$ argon (no state selection), using recoil technique in scattering-out mode; with direct determination of argon velocity distribution using time-of-flight. Very small statistical spread (5%), with over-all error claim of ±35% in absolute magnitude cross section and ±0.250 eV in energy scale.

[r] The Brode experiments on cadmium, mercury, zinc, and thallium were performed in essentially the same manner as the alkali experiments. Similar uncertainties are expected as a consequence (see Footnote e).

[s] Jones used both a Ramsauer–Brode technique and a transmission method. The results are in good relative agreement, but the transmission results are consistently about 20% higher than the Ramsauer results. Agreement with Brode is fair; general shapes are not dissimilar.

[t] This crossed-beam transmission experiment was similar to previous work on H (Footnote b) and O (Footnote u). Because of the difficulty with dissociating N_2, a high-powered, pulsed dc discharge was used, with the beam being formed outside the active discharge region. About 20% average dissociation in the beam was observed. A Pierce-type scattering gun, using relatively high currents, was used because of the generally unfavorable signal to noise in this experiment. The angular resolution was estimated to be about 16°. Results were normalized to Normand (not included in this table), but normalization to Bruche (1927a, Footnote w) would not have changed results significantly. The results vary from about $4\pi a_0^2$ at 2.5 eV to $6.5\pi a_0^2$ at 10 eV. Qualitative values only were obtained between 1.6 and 2.2 eV, with an upper bound quoted at $4.5\pi a_0^2$. The random error is about ±15%–25%; inclusion of systematic errors would probably increase these somewhat.

[u] This experiment is basically the same as the crossed-beam atomic hydrogen experiment of Neynaber *et al.* (see Footnote b), with differences relating primarily to the discharge characteristics of the rf atomic beam source and the normalization procedure. Using a small H_2 additive (~2%–3%), and a rf power of about 30 W, between 25%–35% dissociation was obtained. The experiment was done in transmission (scattering out); the angular resolution was estimated to be about 25°. Absolute values were obtained by applying the formula in footnote a to relative cross sections of O and O_2, and by normalization of the relative O_2 curve to Bruche (1927b; see Footnote ac) at one energy (11.5 eV). Excited states were not deemed to contribute significantly to measured cross sections. Constant (energy-independent) cross section of $6.2\pi a_0^2$ was obtained between 2.3 and 11.6 eV with a claimed error of ±8%. This error is purely statistical and corresponds to the band which includes half of the 64 experimental points (the data points are not presented in the paper). Inclusion of systematic errors would probably increase this error.

[v] This was a crossed-beam recoil experiment in transmission (scattering out) using a rf discharge source, with about 25% dissociation obtained. Absolute data for O and O_2 are presented; the O_2 data below 12 eV agree in relative shape to that of Bruche (1927b), but lie between 10%–20% higher in absolute values. Between 12 and 100 eV, the absolute values are in somewhat better agreement. The O cross sections vary from 5.3×10^{-16} cm² at 0.5 eV to 8.3×10^{-16} cm² at 7.1 eV. Below 6 eV the ratios of $\sigma(O)/\sigma(O_2)$ for the Sunshine *et al.* and the Neynaber *et al.* (Footnote u) agree to well within the combined errors; above 6 eV, the former group's value lies somewhat higher (about equal to the combined estimated errors). The claimed errors including systematic effects are ±20% for the absolute data, except for the 0.5 eV point, which possesses an error of ±30%. The error in the ratio data is quoted at ±13%. Angular resolution for O ranges from 15.6° at 1 eV to 9.6° at 12 eV, and for O_2 from 18.5° at 1 eV to 11.4° at 12 eV.

[w] This is an extremely elaborate and thorough study of H_2 and N_2 total cross sections using the Ramsauer technique. The experiment involves direct attenuation, as well as some indirect determinations at lower energies, involving observation of modifications of the electron energy distribution caused by the scattering gas. These, along with corresponding measurements on the rare gases (Footnote h) and on O_2 (Bruche, 1927b, Footnote w) constitute the most reliable of the prewar Ramsauer-type experiments.

[x] Extension of low-energy technique formerly applied to rare gases, to H_2, O_2, and N_2 (see Footnote i).

[y] Employs previously obtained swarm data, particularly the drift velocity and D/μ coefficient measurements of Pack and Phelps (1961), as well as theoretically calculated cross sections for rotational excitation. See Sec. 2.4.

[z] The swarm analysis is extended to higher energies to include contributions of elastic scattering, rotational, vibrational and electronic excitation and ionization. See Footnote y and Sec. 2.4. Phelps (1968) has indicated a need for revision of the H_2 analysis at the higher energies.

[aa] Same technique as that discussed in Footnotes k and p.

[ab] Substantial vibrational excitation structure was observed in the vicinity of the large peak in the total cross section around 2–3 eV; the energies and number of the observed oscillations are in excellent agreement with those observed directly in inelastic experiments. Similar and much smaller oscillations observed at lower energies, which cannot be attributed to inelastic channels, have not been confirmed by other observers.

[ac] An extension of the work described in Footnote w, to O_2, CH_4, CO, CO_2, N_2O, and NO.

[ad] See discussion in Sec. 3.4.

ments is helium, and it is mainly for this reason that helium has been so extensively studied experimentally. While the theoretical literature for helium is not as exhaustive as for atomic hydrogen, in recent years a number of excellent calculations have been made, with claimed accuracy also within the 10% (and even, in some of the most recent work, of better than 5%) range.[19] It therefore develops that of all possible atomic systems, helium is the one which has received the most attention, considering theory and experiment combined. This will be discussed further in Sec. 4. Here we discuss the relevant helium work and attempt to arrive

[19] See Table II for a summary of one aspect of some recent *e*–He calculations.

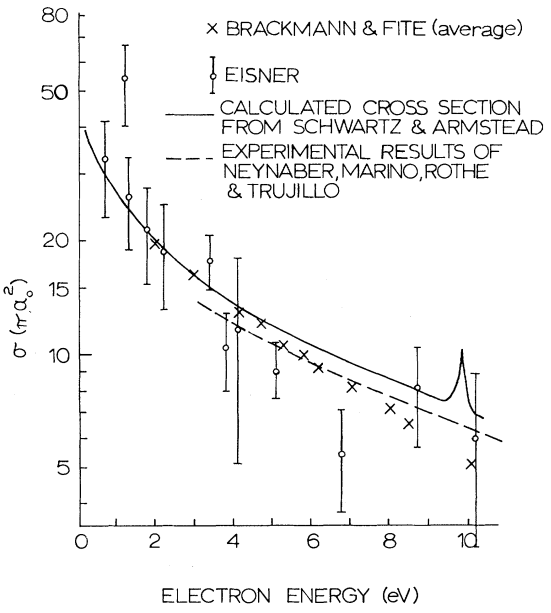

FIG. 9. Summary of recent crossed-beam experimental determinations of total electron-atomic hydrogen cross sections. Compared to S-wave calculation of Schwartz (1961) combined with P-wave calculation of Armstead (1968).

at a conclusion regarding the precision to which the elastic cross section is in fact known at this time.

Reference to Table I reveals that, aside from microwave measurements which are not included, seven quantitative studies of helium have been made, five of them by the Ramsauer method, and two by swarms. One of these is the Ramsauer and Kollath study at very low energy (from 0.15 to 2.3 eV), supplementing the original Ramsauer experiment which covered the range of 0.6 to 40 eV. In comparing the Ramsauer-type measurements to the swarm measurements it must of course be borne in mind that unless the total cross section is isotropic in polar scattering angle, σ and σ_{MT} will not be equal since the definition of these quantities differ, i.e.,

$$\sigma = \int_{\text{all angles}} \eta(\theta, \phi)\sigma(\theta, \phi)\, d\Omega,$$

$$\sigma_{MT} = \int_{\text{all angles}} \sigma(\theta)\,(1 - \cos\theta)\, d\Omega,$$

where $\eta(\theta, \phi)$ is a factor which takes into account the relative efficiency of the detector in collecting particles scattered into different combinations of θ, ϕ (and actually also depends upon other geometry factors as well) (see Sec. 2.1). Clearly several assumptions must be made in effecting a direct comparison of σ and σ_{MT}, particularly concerning knowledge of η and the relative angular dependence of $\sigma(\theta)$. (Of course, one encounters the same difficulty with respect to η when comparing different measurements of σ.)

Of the five Ramsauer-type experiments, we will consider here principally those of Ramsauer (1921b); Bruche, Lilienthal, and Schrodter (1927, hereafter referred to as Bruche *et al.*); and Golden and Bandel (1965). The Ramsauer and Kollath (1929) experiment, which is an extension of the earlier Ramsauer measurements to very low energies, presents a special problem and will be discussed separately below. We have not included the Brode (1925) and Normand (1930) measurements. [See Footnote c of Table I. See also Golden and Bandel (1965) for a detailed comparison of the Brode and Normand results with those of Golden and Bandel.] A direct comparison of the Ramsauer, Ramsauer and Kollath, Bruche *et al.*, and Golden and Bandel results is shown in Fig. 10.

Comparison of the transmission experiments can be summarized as follows. Above 4 eV the shapes of all three measurements are approximately the same, with those of Ramsauer lying highest, those of Bruche *et al.* lying perhaps 15% below Ramsauer, and those of Golden and Bandel lying an additional 5% below Bruche *et al.* The Ramsauer curve peaks at about 2 eV; the Bruche peak, at about 2.75 eV; and that of Golden and Bandel, at about 1.2 eV. The Bruche *et al.* curve possesses quite a sharp peak, while the Golden and Bandel curve is the flattest. A critical appraisal of the operational aspects of the Ramsauer and Bruche experiments would be virtually impossible since details of the pressure measurements, gas purity, vacuum and current measurement techniques are lacking for these experiments, which were conducted over 40 years ago. It is possible, on the other hand, to form a more quantitative judgment of the operational aspects of the Golden and Bandel experiment. The discussion of systematic errors contained in the original Golden and Bandel paper has been supplemented, during the course of preparation of this review, by a thorough exploration of the Lockheed experiment and of the techniques

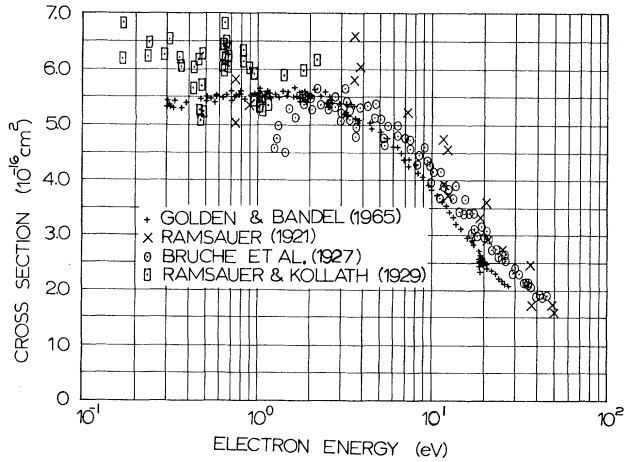

FIG. 10. Summary of "best" total cross section data for electron–helium scattering, all performed using the Ramsauer technique.

employed.[20] A schematic drawing of the Golden and Bandel apparatus is shown in Fig. 11. Some of its features, which represent advances over the original Ramsauer apparatus, are as follows: First, the apparatus itself was machined out of a *single* block of metal, thereby eliminating gross contact potential differences which result from the use of dissimilar metals, although not eliminating local variations which result from differences in surface conditions. These were minimized by coating the apparatus with colloidal graphite. Second, differential pumping was employed; that is, the cathode region was continually pumped, while sustaining reasonably high gas densities in the scattering and collecting regions. Thus, one of the principal uncertainties of the original Ramsauer work, the possible change in emission characteristics of the emitting surface as the gas pressure was changed, was minimized. A typical run in a Ramsauer experiment involves making a transmitted-current-versus-gas-pressure run at constant energy, so that systematic gas–surface effects could be playing an important role which is difficult, if not impossible, to evaluate. Third, modern ultrahigh vacuum techniques, including baking of the apparatus and gas-handling equipment, were utilized. Fourth, pressure measurement was accomplished using a Schulz–Phelps ionization gauge calibrated by a capacitor manometer, a method which avoids the contamination and possible pumping effects encountered using a directly coupled McLeod gauge (Kieffer and Dunn, 1966). Some of these advances were partially mitigated by certain practical considerations. Thus, it was found necessary to install additional collimating slits to minimize effects due to reflected electrons, thereby introducing dissimilar metals after all although use of graphite coating should suppress contact potentials; the use of iron pole faces made it impossible to calibrate the magnetic field under operating conditions, making it necessary to rely solely upon retarding potential measurements to determine the electron energy and energy spread. It should be noted, however, that the helium resonance at 19.3 eV was observed at the correct energy, thereby lending confidence to the reliability of the retarding potential technique in the presence of a magnetic field. The introduction of the gas in the channel located at the entrance to the scattering region makes it very difficult to accurately determine the effective electron path length. Measurement of the pressure a considerable distance from the interaction region introduces an uncertainty in the absolute pressure determination.

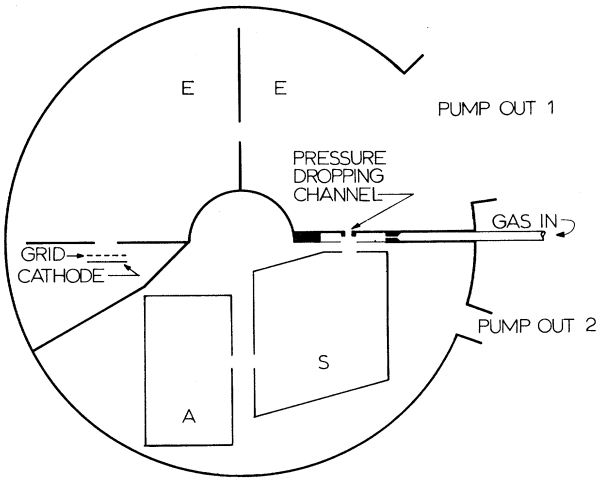

FIG. 11. Schematic diagram of the Golden and Bandel (1965) apparatus. (See Fig. 1 for definition of symbols.)

In the Golden and Bandel paper, it was concluded that the pressure measurement introduced by far the largest error (2%), while all other errors, each contributing 1% or less, were assumed to add randomly to produce a total error of 3%. We believe that the general quality of the Golden and Bandel results, as well as the subsequent papers of the Lockheed group, is quite high. The over-all error estimate we believe to be somewhat understated, and would prefer that larger brackets be used. It appears more realistic to assume (Kieffer and Dunn, 1966) a 5%–10% error in the absolute pressure determination, with an over-all systematic error estimate somewhat higher than this figure. Note that apart from the energy range between 1.5 and 2 eV, where the Bruche *et al.* data appear to be falling very rapidly with decreasing energy, the agreement between Golden and Bandel and Bruche *et al.* is extremely good.

Concerning swarms, as already discussed, the Crompton *et al.* experiment on helium represents the most intensive attempt to date to determine momentum-transfer cross sections from transport properties of electrons in a gas and we will therefore restrict our discussion here to their experiment. We would like to establish an estimate of the reliability of the momentum-transfer cross section transformation to total cross sections, which can then be compared with the transmission measurements. To effect this transformation it is necessary to employ differential cross sections. This will be discussed further later in this section.

We will first briefly discuss the Crompton *et al.* experiment, which measured drift velocities by a time-of-flight technique (Bradbury and Nielsen, 1936; Lowke, 1963), the ratio $D_\perp/\mu$, the diffusion coefficient to the mobility, and $W_\perp/W_{||}$ (see Sec. 2.4) velocity in the presence of a magnetic field. All these quantities were measured at room temperature.[21] The three experiments were not considered to be of comparable preci-

[20] We are greatly indebted to Dr. David Golden for his cooperation, during the preparation of this article, in discussing with us the laboratory routines employed and in showing us in detail the experimental setup at Lockheed.

[21] More recent measurements by the Crompton group, have been performed on He and Ne at liquid nitrogen temperatures. See note 22.

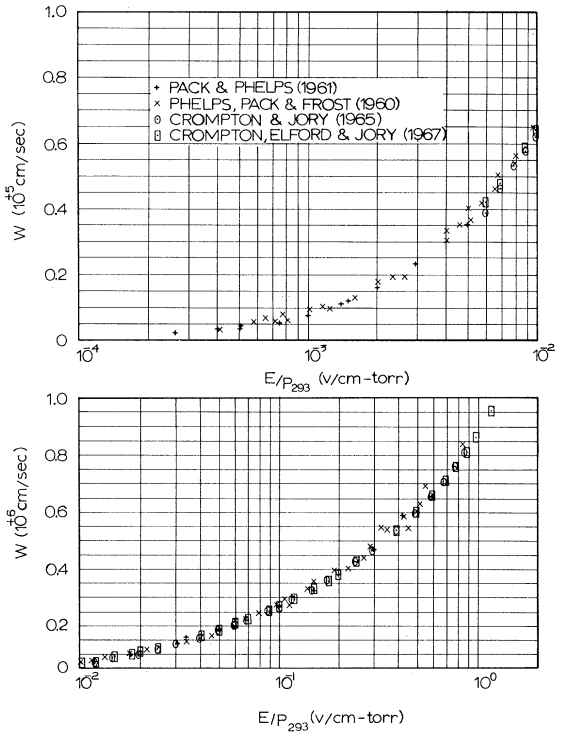

FIG. 12. Summary of recent measurements of drift velocity w vs ratio of electric field to pressure (E/P) by Crompton and Phelps groups.

sion, with the drift velocity determinations being the most precise.

The claimed precision of the drift velocity measurements is $\pm 0.5\%$[22] for σ_{MT} independent of energy, which transforms into an error of $\pm 1\%$ in the cross section (Frost and Phelps, 1964).

Figure 12 shows some recent measurements of drift velocity versus E/P, where P is the pressure, in helium, at or near room temperature. These were taken by the Phelps and Crompton groups, using refined time-of-flight techniques. The spread in the data, over more than three decades of E/P values, is generally not greater than 5%. For the purpose of this review we shall accept the measurement error quoted by Crompton *et al.*, bearing in mind that the error in the derived cross section is roughly double that in the drift velocity.

Aside from the measurement error, the over-all error claimed in the Crompton *et al.* cross section determinations is based upon several interrelated factors. These can be briefly summarized as follows: first, there is the convergence of the iteration procedure; that is, the fact that starting with an arbitrary set of cross sections one iterates until agreement is obtained to within the experimental error with the observed drift velocities. The sensitivity of the trial cross section agreement with the drift velocity data is illustrated in Figs. 13–15.[23] Figure 13 shows the Crompton *et al.* "correct" momentum-transfer cross sections (curve 1), one (curve 2) which is consistently 2% lower than the Crompton *et al.* cross sections, and one (curve 3) which starts 2% lower at 2×10^{-2} eV and increases to become 2% higher at 2 eV. The calculated drift velocity assuming these three cross sections are plotted in Fig. 14 (curves 1 and 2) and Fig. 15 (curve 3). The sensitivity to the "correct" choice of σ_{MT} is clearly apparent, the mismatch to the experimental data lying well outside the experimental scatter when the "incorrect" σ_{MT} are used.

The other main factor which enters into the final error claim is the agreement of the measured $D_\perp/\mu$ and $W_\perp/W_{||}$ values with the corresponding calculated values, obtained using the Crompton *et al.* cross sections and the same distribution functions used in the σ_{MT} determinations. Such a comparison constitutes an essentially independent verification of the correctness of the calculated velocity distribution. Agreement to better than 1% throughout the entire energy range (10^{-2}–2 eV) was obtained for the $D_\perp/\mu$ values, and to better than about 2% in the $W_\perp/W_{||}$ values. Both sets of comparisons agree to within the claimed errors in the measurements.

The fact that the same set of momentum transfer cross sections can be used to predict three independent sets of transport coefficients, measured on two separate apparatuses, is indeed the most convincing argument in favor of this entire experimental and analytic procedure. It appears reasonably certain therefore that the Crompton *et al.* helium results are reliable and quite accurate.[22]

Regarding structure in the momentum-transfer cross section curve,[24] the sensitivity of the Crompton *et al.* measurements is indeed quite high. The Crompton *et al.* claim that a 10% oscillation in the cross section at about 0.5 eV, necessary to be compatible with the structure in the early Ramsauer and Kollath and Normand measurements, would be easily observable, is a reasonable one. The lack of any observed structure in the Crompton *et al.* cross section therefore serves as a convincing confirmation that such structure is spurious.[25]

The remaining problem is that of connecting σ_{MT} to

[22] Recent work by the Crompton group has extended the helium measurements to low temperatures (76.8°K) [R. W. Crompton, M. T. Elford, and A. G. Robertson 1970]. This group has performed drift velocity measurements on neon at room temperature and at 77°K, with comparable precision obtained for σ_{MT}; i.e., the measurements are extended down to 0.008 eV. The He results for σ_{MT} agree to within 1% with those of Crompton *et al.*, and also permit a direct determination of the scattering length ($1.19a_0$) obtained without employing an extrapolation technique.

[23] We are indebted to Dr. Malcom Elford for performing the necessary calculations and for plotting these results.

[24] See, for example, Ramsauer and Kollath (1929), Normand (1930), Golden and Bandel (1965), Schulz (1965), Bullis, Churchill, Wiegand, and Schubert (1967).

[25] Several explanations for the undulations in the Ramsauer and Kollath and Normand curves have been offered, chief among them being, first, impurities, particularly N_2, and second, electron optics effects. In this connection, it should be noted that many of Normand's curves exhibited anomalous peaking at 1 or 2 eV.

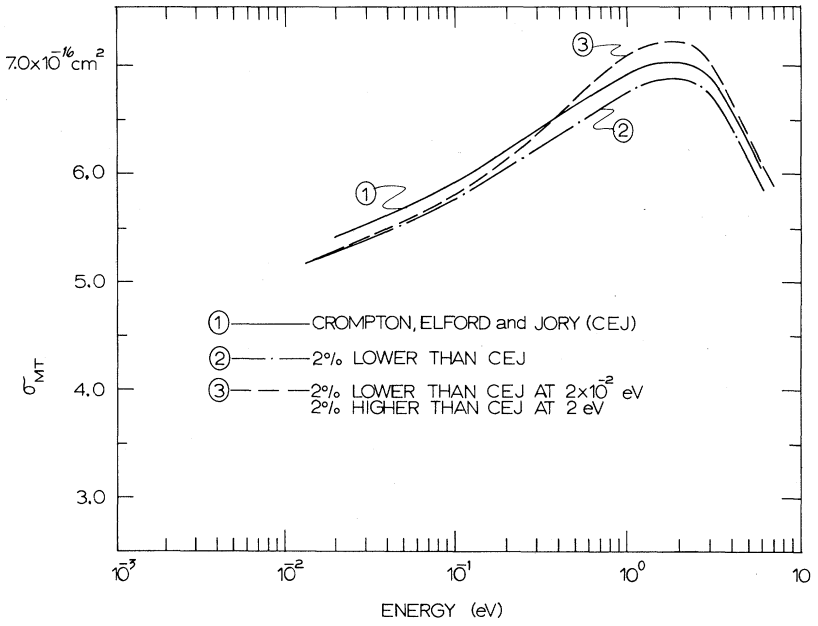

FIG. 13. Figures 13–15 demonstrate sensitivity of calculated drift velocity to choice of σ_{MT}. Figure 13 plots the "correct" σ_{MT} [as determined by Crompton, Elford and Jory (1967)], and two other choices differing from these values as stated in the figure. These figures were supplied by Dr. M. Elford.

σ. This may be done using measured values of the differential cross sections, using the relation

$$\frac{\sigma_{MT}}{\sigma} = 1 + \int_{-1}^{1} \sigma(x)\, x\, dx \Big/ \int_{-1}^{1} \sigma(x)\, dx, \qquad (33)$$

where $x = \cos\theta$. Here the relative angular dependence of $\sigma(x)$ need only be known since a constant scaling factor in $\sigma(x)$ would cancel out. A serious difficulty in using experimental values arises since very small and very large angle measurements are usually not available. For helium at low energies, until very recently the only available data were those of Ramsauer and Kollath, obtained using the zone-plate method. This is a rather qualitative experiment because of the coarse angular-mesh size as well as problems associated with the method, e.g., reflected electrons from the collection plate.

The recent experiment of Gibson and Dolder (1969) is certainly a more reliable experiment although the lowest energy employed was 3.1 eV, compared to 1.8 eV in Ramsauer and Kollath. In this experiment, an electrostatic analyzer is employed to produce a monochromatic (65 meV energy spread) electron beam. The entire chamber is filled with gas; suitable collimators and a rotating collector are used to obtain relative angular distributions. Because of geometric problems,

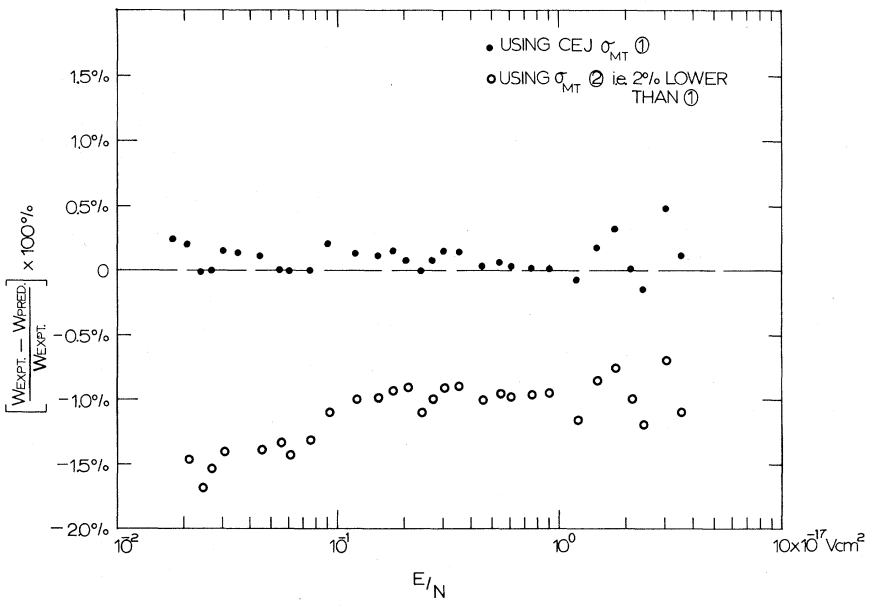

FIG. 14. The relative deviation, in percent, of the calculated drift velocity vs E/N, using the "correct" σ_{MT} from Crompton, Elford, and Jory (1967) and the values of σ_{MT} shown in curve 2 of Fig. 13.

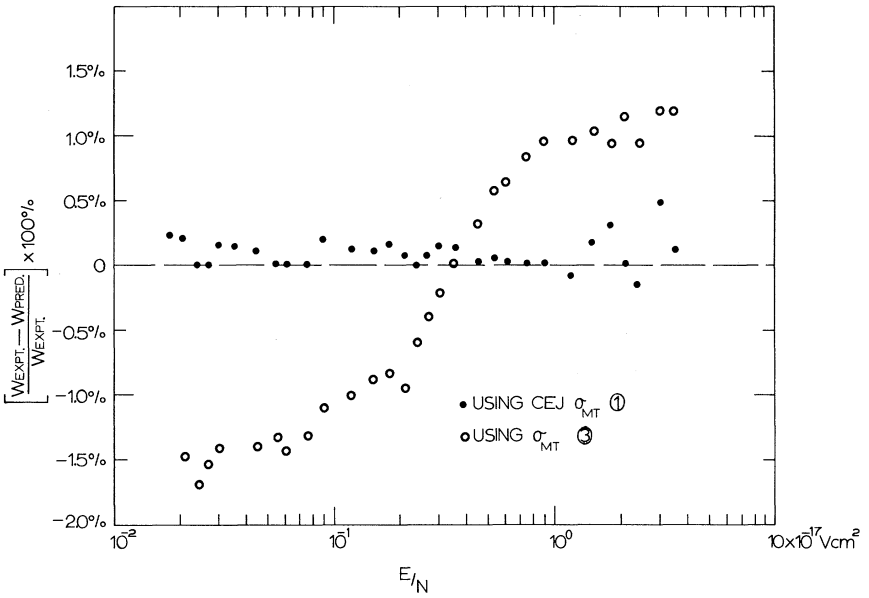

FIG. 15. Similar to Fig. 14, but employing the values of σ_{MT} shown in curve 3 of Fig. 14.

it is always difficult to obtain absolute values in a differential cross section experiment. Gibson and Dolder therefore normalized their relative values to an absolute determination obtained from a phase shift analysis of the observed angular distribution at the 19.3 eV resonance. Their results are in fair agreement with Ramsauer and Kollath for angles above about 60°. Below 60° the Ramsauer and Kollath cross sections increase rapidly with decreasing angle, while the Gibson and Dolder results decrease with decreasing angle, in agreement with the theoretical values of Callaway, LaBahn, Pu, and Duxler (1968), as well as with the values obtained from the Bransden and McDowell (1969) phase shifts.

In effecting the σ_{MT}–σ comparison, therefore, we have employed the Callaway *et al.* phase shifts. No better procedure seems available at the present time.

In terms of computed elastic phase shifts η_l, we have

$$\frac{\sigma_{MT}}{\sigma}=\sum_{l=0}^{\infty}(l+1)\sin^2(\eta_l-\eta_{l+1})/\sum_{l=0}^{\infty}(2l+1)\sin^2\eta_l. \quad (34)$$

Again, computed errors in this ratio will tend to be smaller than the corresponding error in σ_{MT} or σ computed separately. Equation (34) can be employed, using calculated or measured phase shifts, to effect a comparison of momentum-transfer (Crompton *et al.*) and total cross sections.

A form of comparison can also be obtained by the use of modified effective range theory (MERT).[26] In the low-energy limit, σ and σ_{MT} a are given by

$$\sigma=4\pi[A^2+(2/3)aE^{1/2}+O(E\ln E)+O'(E)] \quad (35)$$

and

$$\sigma_{MT}=4\pi[A^2+(4/5)aE^{1/2}+O(E\ln E)+O''(E)], \quad (36)$$

where A is the scattering length, and a is a constant determined from atomic structure parameters. At sufficiently low energies one obtains from Eqs. (35) and (36) a unique expression for the ratio of the slopes of the two cross sections,

$$(\sigma_{MT}'/\sigma')(\lim E\to 0)=1.20. \quad (37)$$

This relation can be used as an additional check on the compatability of the swarm and transmission experiments. From theory, or from a differential measurement, where the phase shifts η_0, η_1 are obtained, the above ratio can be obtained from the relation

$$\sigma_{MT}'/\sigma'=1+\eta_1'/(A-\eta_0'), \quad (38)$$

where η_0', η_1' are the derivatives of η_0, η_1 with respect to k evaluated at zero energy. For example, using the calculated phase shifts of Callaway *et al.* (1968), at $k=0.01$ (0.00136 eV), one obtains $\sigma_{MT}'/\sigma'=1.226$. The scattering length itself can be obtained by extrapolating either σ_{MT} or σ to zero energy. Table II shows a list of recent scattering length determinations for helium, obtained from theory, as well as those obtained by extrapolation to zero energy of the measurements of Ramsauer and Kollath, Golden and Bandel, and Crompton *et al.* The table reveals a clustering of the theory about the two mean values 1.4 a_0 and about 1.15 a_0, with the experimental scattering lengths all falling near the latter value.

Equation (37) can be applied by, first, using the extrapolated σ values obtained by Golden (1966) by a least-squares fit to the Golden and Bandel data via MERT, and second, by calculating σ_{MT}' directly from the very low-energy data of Crompton *et al.* The ratio thereby obtained is 1.27, about 6% higher than the theoretical value. This small difference is possibly

[26] The use of effective range theory in atomic collision work, where the long-range polarization force plays such an important role, was developed in a series of important papers by Spruch and his students. These papers include: Spruch, O'Malley, and Rosenberg (1960), O'Malley, Spruch, and Rosenberg (1961), O'Malley, Rosenberg, and Spruch (1962), and O'Malley (1963).

TABLE II. Recent calculated and "measured" scattering lengths in helium.

Reference	Scattering length (units of a_0)	Comments
		Theoretical
Moiseiwitsch (1960)	1.442	Exact numerical integration including exchange, neglecting polarization, using Hartree wave function for helium
Hashino and Matsuda (1963a, 1963b)	1.50	Same result obtained using Kohn and Hulthen variational methods
Kestner, Jortner, Cohen, and Rice (1965)	1.193	Uses appropriately chosen pseudopotential and Kohn-type variational procedure
Houston and Moiseiwitsch (1966)	1.483	
Pu and Chang (1966)	1.18	Modified optical potential in a variational calculation of phase shifts, extrapolated to zero energy
Lawson, Massey, Wallace, and Wilkinson (1966)	1.15	Dispersion relation
	1.20	Adiabatic exchange calculation
LaBahn and Callaway (1966)	1.132	Adiabatic exchange, dipole polarization potential
	1.097	Adiabatic exchange, total polarization potential
	1.151	Extended polarization potential
Peterkop (1968)	1.483	Static-exchange approximation
	1.282	Includes many correlation terms
Callaway, LaBahn, Pu, and Duxler (1968)	1.151	"Extended" polarization potential, including distortion effects
Houston (1968)	1.10	Includes correlation and exchange; Kohn variational method
Michels, Harris, and Scolsky (1969)	1.145	Adiabatic potential including distortion, polarization and exchange with accurate Slater-type orbitals
Sheorey (1969)	1.195	Quantum-defect method; isoelectronic sequence extrapolation from negative energies, using spectroscopic data. Includes quadrupole potential term
		Experimental
Ramsauer and Kollath [O'Malley (1963)]	1.19	Extrapolation of Ramsauer–Kollath data by O'Malley using modified effective range theory (MERT)
Golden (1966)	1.15	Extrapolation of Golden–Bandel data, using MERT
Crompton, Elford, and Jory (1967)	1.18	dc swarm experiment extrapolation to zero energy
Crompton, Elford, and Robertson (1971)	1.19	dc swarm experiment at 76.8°K, extrapolated to zero energy from 0.008 eV

fortuitous, considering the small changes in σ and σ_{MT} which are used in this calculation.

A quantitative comparison of the momentum-transfer and transmission data can be obtained via Eq. (34). We have used the phase shifts of Callaway *et al.* to obtain σ_{MT}/σ in order to convert the Crompton *et al.* cross sections to total cross sections. The results of this comparison are shown in Fig. 16; plotted are the Ramsauer, Bruche, and Golden and Bandel total cross sections, the Crompton *et al.* cross sections modified, as well as the total cross sections calculated directly from the Callaway *et al.* phase shifts and those of Michels, Harris, and Scolsky (1969). The Ramsauer and Kollath cross sections are not included. These would lie some 15%–20% above the theoretical curve.

An alternative method of comparison of σ_{MT} and σ involves the use of MERT, as discussed above. At sufficiently low energies, a best fit to Eqs. (35) and (36) may be made by varying the parameters A, a. Once these are obtained, σ_{MT} may be calculated from σ, or vice versa. Golden has used this approach to calculate σ_{MT} from the Golden and Bandel data, and thereby effect a comparison with Crompton *et al.* and Pack and Phelps (1961). Again it is seen that the Golden and Bandel data lie perhaps 10% below the Crompton *et al.* data; this bias is consistent with that obtained from use of the Callaway *et al.* phase shifts. Of course it should be noted that MERT itself is not an exact formalism, being based on the assumption of adiabaticity. There is no rigorous analysis of the precision to which this assumption is valid at low energies; small errors arising from the use of MERT cannot be completely discounted.

We summarize the results of this comparison below:

(1) In the very low-energy limit, Crompton *et al.* and Golden and Bandel extrapolated using MERT and the most recent calculations using adiabatic potential approximations agree to within about 4%.

(2) In the limit of zero energy the slope ratio σ_{MT}'/σ', which should yield 1.20, gives 1.27, which is not considered to be a significant discrepancy.

(3) At very low energies the Golden and Bandel relative energy dependence is in considerably better agreement with theory than Ramsauer and Bruche *et al.* Above 2 eV, Bruche *et al.* lies very close to theory, with Golden and Bandel lying perhaps 8% below, and Ramsauer about the same amount above theory.

(4) The Crompton *et al.* momentum-transfer cross sections are in excellent agreement (within 6%) with theory.

(5) On the average, the Golden and Bandel and

Crompton *et al.* curves differ by about 9% in absolute values when compared using the Callaway *et al.* phase shifts. The Golden and Bandel and Crompton *et al.* curves differ by as much as 12% at the peak and by about 4% at the lowest energy where comparison is possible (0.03 eV), when compared by Golden's transformation from σ to σ_{MT} using MERT. The Crompton *et al.* curve lies consistently higher.

We have excluded the Ramsauer and Kollath and Bruche *et al.* data below 2 eV from consideration because of the difficulties associated with the evaluation of data obtained at such low energies before the advent of "clean" vacuum practice.

By excluding these values it is possible to bracket the four helium experiments discussed here to within about 20%. The calculations of Callaway *et al.* and Michels *et al.* fall at about the center of these brackets. We believe that the errors associated with all of the experiments are not inconsistent with these brackets. Excluding the Ramsauer data reduces the size of brackets substantially, to perhaps 12%–15%. It is reasonable to assign a smaller weight to the Ramsauer data than to that of Golden and Bandel, considering the improved techniques employed in the latter experiment, although there is no compelling argument to eliminate these altogether. The data discussed here and the results of the calculations of Callaway *et al.* are presented in Fig. 16.[27]

Bransden and McDowell (1969) have recently performed a very complete phase shift analysis of much of the experimental data available at the time of their work. For this purpose they used the Golden and Bandel total cross sections, the differential cross sections of Gibson and Dolder, and the momentum-transfer cross sections of Frost and Phelps. These data were all weighted equally, i.e., they were all assigned an error of $\pm 10\%$. The phase shifts obtained thereby were compared with recent calculations. In addition, the real part of the scattering amplitude $\mathrm{Re}\, f(0, k^2)$, obtained using a dispersion relation, was compared with the *total* cross sections, obtained by a combination of methods up to 1 keV. The final consistency of their results with the available data, as well as with the more elaborate recent calculations (particularly of Callaway *et al.*), is of the order of 10%. They did not use the Crompton *et al.* cross sections, however.[28] We conclude that the helium total cross section, as measured in transmission by beams techniques, has not been determined with certainty to better than perhaps 10%–15%. Further work in more definitive transmission and crossed-beam experiments is clearly desirable. The swarm experiment has been carried out to a precision which is limited only by the current experimental and theoretical state of the art.

3.3 Other Rare Gases

The remaining rare gases (Ne, Ar, Kr, Xe) have been studied by various investigators over the years, as tabulated in Table I. Argon has been the most thoroughly studied of the heavier rare gases, among other reasons because of the existence of the Ramsauer effect, which makes argon an interesting system theoretically.

Regarding argon, as well as the other heavy rare gases, the Ramsauer–Townsend minimum is by now well substantiated. The rather rapid rise to large values at higher energies is also well documented. The detailed shape of the Ramsauer minima are, on the other hand, a matter of some controversy.

Because of the lack of accurate angular distribution data at very low energies, it is not possible to make a direct comparison of σ from Golden and Bandel, Ramsauer, and Ramsauer and Kollath with the best available swarm data, that of Frost and Phelps. Golden has made a comparison by use of MERT (see discussion in Sec. 3.2) for argon and finds that the Golden and Bandel–Ramsauer minimum, when transformed into a momentum-transfer cross section, lies far deeper than that of Frost and Phelps. He attributes this to the basic inability of a swarm experiment to resolve structure in the cross section which is substantially smaller than the electron energy distribution of the swarm. On the other hand, as Phelps has pointed out, since σ_{MT} occurs in the denominator in the integral of Eq. (18), the swarm experiment is in fact more sensitive to a cross section minimum and should tend to exaggerate its appearance.

The only significant difference in the heavy-rare-gas σ_{MT} determination of Frost and Phelps and the helium σ_{MT} determination of Crompton *et al.* lies in the pre-

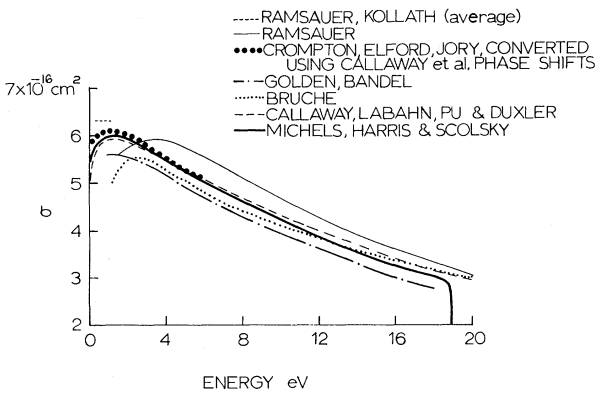

FIG. 16. Summary of "best" electron–helium total cross section data, including swarm data of Crompton, Elford, and Jory (1967), transformed using phase shifts of Callaway *et al.* (1968), compared to several recent calculations.

[27] We do not include the data obtained using the promising time-of-flight technique (Baldwin and Friedman, 1967). See Sec. 2.6.

[28] Even more recently McDowell has reported results of a refined phase-shift analysis at the Second International Conference on Atomic Physics (held at Oxford, 1970). He concludes that the swarm data of Pack and Phelps are more consistent with their error analysis than are those of Crompton *et al.* Thus, his analysis does not appear to help resolve the helium situation and, if anything, adds further uncertainty to the subject.

cision of the drift velocity data. The σ_{MT} values for the heavier rare gases do not possess precision comparable to that for helium.

3.4 Alkali Metals

The only other atomic systems which can be produced in a stable vapor at reasonably low temperatures are the alkali metals, excepting lithium, and cadmium, zinc, and mercury. The alkalis in particular are of special interest because of the relative simplicity of their electronic structure ($ns^2S_{1/2}$). Collision theory can thereby be formulated in terms of a single valence electron model, similar to atomic hydrogen except for core effects. The alkalis are characterized by their large electric dipole polarizabilities, so that the distinctive problem associated with low-energy collisions, which is the effect of the interaction of the atomic wave function, i.e., polarization, plays a central role in the collision process. Thus, considerable theoretical effort has been expended in recent years on the electron–alkali problem, and it would be expected that here exists an excellent common meeting ground for elastic collision theory and experiment.

Until recently, this has not turned out to be the case. The Brode experiments, discussed in Sec. 2.1, constitute the most complete alkali study to date. These are in fact the only beam–gas transmission experiments in the published literature. A considerable number of swarm experiments, performed in alkali vapor cells undergoing some type of weak or strong discharge, exist, as summarized in Table III. These experiments do not satisfy the basic criteria discussed earlier with regard to the use of transport data to extract momentum-transfer cross sections. Thus, in all of these measurements there exist the possibilities that there are contributions due to the existence of excited states, of plasma effects (electron–electron and electron–ion collisions), inelastic and ionization collisions, and radiation effects as well. Afterglow swarm experiments have been performed by Chen and Raether (1962) with cesium but not for other alkalis. Some dc swarm-type experiments of the Townsend-Crompton variety have been performed by Chanin and Steen (1964) but the experimental conditions were far from ideal.

Several crossed-beam experiments have been reported, using the recoil technique. Perel, Englander, and Bederson (1962) performed scattering-out measurements on lithium and sodium. These were relative determinations, normalized to Brode's potassium data. Several attempts to obtain absolute values of the total cross sections at 13 eV were also made although it is now believed that these values are in error due to inadequate shielding from secondary and reflected electrons. Recently Collins and co-workers (Collins, Goldstein, Bederson, and Rubin, 1967; Collins, 1968; Collins, Bederson, and Goldstein, 1971), in connection with a program to use recoil to measure differential and differential-exchange cross sections in potassium, also performed a scattering-out measurement to obtain an absolute total cross section. In both the Perel and the Collins experiments the observed structure near the first excitation threshold, which is so pronounced in the Brode curves, is either much smaller or in fact barely perceptible at all. In the Collins work the absolute values for potassium are about a factor of 2 lower than Brode's. Renormalization of the Perel cross sections to Collins would also reduce the Li and Na values by a corresponding amount. The revised values are in far better agreement with recent close-coupling-type calculations (Karule, 1965; Bederson, 1969; Collins, Bederson, and Goldstein, 1971).

The total cross section results for potassium have been confirmed in an independent recoil experiment performed by Visconti, Slevin, and Rubin (1971), whose results are in excellent agreement with those of Collins *et al.* Figure 17 shows the results of the two recoil experiments, along with those of Brode and of Karule and Karule and Peterkop. Visconti *et al.* also report on total cross sections in rubidium and cesium. Again the results are in substantial disagreement with the Brode cross sections, lying well below these, and rising essentially monotonically with decreasing energy without significant structure. The results for cesium, however, do not agree as well with the close-coupling results of Karule and of Karule and Peterkop.[29]

Taking into account the fundamental difficulties associated with the single cage Ramsauer device (see Sec. 2.1), which could be particularly serious when operating in an alkali vapor atmosphere, we are led to the conclusion that the Brode data for the alkalis (and, by inference, for Cd, Zn, and Hg) are in serious question. In particular, the alkali data appear to be too high by a factor of about 2, and the structure shown in the vicinity of the first excitation energy appears greatly exaggerated.

Swarm measurements in the alkalis have been mainly confined to cesium, partly because of recent interest in the use of cesium in thermoelectric energy conversion. Some of these measurements are summarized in Table III.[30]

Clearly, further work is called for in the alkalis in all three areas of transmission, crossed beams, and swarms, and until such work is forthcoming, the situation here remains unresolved.

3.5 Other Systems

The remaining atomic systems which have been studied require molecular dissociation, and either

[29] It should also be noted that Shpenik, Zavilopulo, Aleksakhin, and Zapesochny (1969) have published preliminary results of a crossed-beam experiment for cesium in transmission which possesses a very large peak at about 2.2 eV. Absolute values are not quoted. Details of this experiment are lacking, and further evaluation must await publication of final results.

[30] A recent report by Dayton (1969) presents a comprehensive summary of momentum-transfer cross sections from theory and experiment in cesium.

TABLE III. Summary of recent dc swarm, ac swarm, and cyclotron resonance measurements of σ_{MT} in rare gases and cesium.[a]

System	Method	σ_{MT} ($\times 10^{-16}$ cm^2)	Mean energy or energy range[b] (eV)	Authors	Footnotes
He	ac swarm	5.4	0.04	Phelps, Fundingsland, and Brown (1951)	c
	ac swarm	5.2	0–0.75	Gould and Brown (1954)	d
		5.4	2.2		
	ac swarm	6.8	0.04	Anderson and Goldstein (1955)	e
	ac swarm	6.6	0.04	Anderson and Goldstein (1956)	f
		7.3	0.4		
	ac swarm	5.7	0.04	Hirshfield and Brown (1958)	g
	dc swarm	6.9	0.13–4.0	Bowe (1960)	h
	ac swarm	5.3	0.026–0.039	Chen (1963)	i
	dc swarm	6.0	0.23	Harris (1963)	j
	dc swarm	···	0.003–30	Frost and Phelps (1964)	v
	dc swarm	···	0.025–6	Crompton, Elford, and Jory (1967)	w
	Cyclotron resonance	5.5	0.04	Tice and Kivelson (1967a)	k
	ac swarm	5.2	0.04	Hoffmann and Skarsgard (1969)	x
		6.2	1.0		
Ne	ac swarm	0.93	0.04	Phelps, Fundingsland, and Brown (1951)	c
	ac swarm	0.54	0.053	Gilardini and Brown (1957)	l
		1.7	2.1		
	dc swarm	1.5	0.38	Bowe (1960)	h
		2.3	8.0		
	dc swarm	1.6	0.23	Harris (1963)	j
	ac swarm	0.45	0.026[b]	Chen (1964)	m
		0.95	0.22[b]		
	ac swarm	0.48	0.04	Hoffmann and Skarsgard (1969)	y
		1.15	1.0		
Ar	ac swarm	0.59	0.04	Phelps, Fundingsland, and Brown (1951)	c
	dc swarm	2.0	1.6	Bowe (1960)	h
		20	11		
	ad swarm	0.4	0.23	Harris (1963)	j
	dc swarm	···	0.003–30	Frost and Phelps (1964)	v
	Cyclotron resonance	5.5	0.005	Tice and Kivelson (1967a)	k
		0.4	0.06		
Kr	ac swarm	15.2	0.04	Phelps, Fundingsland, and Brown (1951)	c
	dc swarm	6.3	1.6	Bowe (1960)	h
		12	3.0		
	ac swarm	22	0.026	Chen (1963)	i
		16	0.039		
	dc swarm	···	0.003–30	Frost and Phelps (1964)	v
	ac swarm	···	0.04–1.0	Hoffmann and Skarsgard (1969)	z
Xe	ac swarm	50.9	0.04	Phelps, Fundingsland, and Brown (1951)	c
	dc swarm	7.41	1.0	Bowe (1960)	h
		18	2.4		
	ac swarm	62	0.026	Chen (1963)	i
		43	0.039		
	dc swarm	···	0.003–30	Frost and Phelps (1964)	v
	ac swarm	···	0.04–1.0	Hoffmann and Skarsgard (1969)	z
Cs	dc swarm	36	0.15	Mullaney and Dibelius (1961)	n
	dc swarm	200	0.20	Mirlin, Pikus and Yurev (1962)	o
	ac swarm	1080	0.059	Chen and Raether (1962)	p
		880	0.071		
	dc swarm	50	0.65	Morgulis and Korchevoi (1963)	q
	Cyclotron resonance	47–51	0.5–0.6	Flavin and Meyerand (1963)	r
	dc swarm	70	0.14	Roehling (1963)	s
		150	0.24		
	dc swarm	300	0.23	Harris (1963)	j
	dc swarm	200	0.39–0.65	Polushkin and Dudko (1966)	t
	ac swarm	1000	0.589	Nighan (1967)	u

TABLE III (*Continued*)

[a] Estimates of error for these types of experiments are not usually presented in the original papers; no attempt is made here, with certain exceptions, to derive such estimates. All ac experiments presented here were performed in the afterglow (see also Table I). The original papers should be consulted to determine, in each case, the precise method used in obtaining σ_{MT} from the data.

[b] In many cases the original papers quote temperature ranges. These have been converted to energy in electron volts, using the relation energy (eV) =temperature (°K) $\times 1.3\times 10^{-4}$ (eV/°K) [i.e., assuming the mean energy of the swarm $=3/2kT$].

[c] This is the original paper on use of microwave complex conductivity measurements in afterglow to obtain momentum-transfer cross sections.

[d] A continuation of work of Phelps *et al.* (Footnote c). Energy varied by varying both ambient gas temperature and applied rf electric field strength.

[e] A cross modulation technique was used. This is an indirect method which cannot be considered to be as reliable as the simpler conductivity results (Footnotes c and d).

[f] This is a continuation of work of Anderson and Goldstein (1955). The energy was varied by using exciting microwave pulses in the afterglow (see Footnote e).

[g] A magnetic field was used in a resonant cavity to obtain the reactive part of the conductivity in the afterglow. The error was quoted as $\pm 5\%$.

[h] This is a dc swarm experiment. Drift-velocity data and assumed power law energy dependence of σ are used to infer σ from the transport equations.

[i] The microwave conductivity of the afterglow was measured. Helium was used as a buffer gas for the heavier rare gases.

[j] This dc conductivity experiment was performed at elevated temperature. The gas sample was seeded with cesium.

[k] This experiment was performed in a flowing afterglow with N_2 as the buffer gas. The cross section was deduced from an analysis of the linewidth of the electron cyclotron resonance.

[l] The technique of Gould and Brown (1954; Footnote d) was used in these measurements.

[m] The microwave conductivity of an afterglow in pure neon was measured. The temperature of the gas was varied from 200 to 600°K.

[n] The conductivity of a quiescent plasma in cesium with crossed electric and magnetic field was measured. The cross section was deduced from the measured conductivity. The cesium gas temperature was 1125°K.

[o] A plasma diode was used to measure the dc conductivity of a cesium plasma in thermodynamic equilibrium.

[p] The microwave measurement of complex conductivity in afterglow, in pure Cs and in He–Cs mixtures was used to determine the collision cross section.

[q] The dc conductivity in an arc discharge was measured. Ion densities were estimated to be 10^{11}–10^{12} cm^3.

[r] The momentum transfer cross sections were determined from measurements of the microwave cyclotron resonance absorption and radiation spectra of a thermalized cesium plasma in a dc magnetic field.

[s] The dc resistivity of a cesium plasma in thermal equilibrium was measured.

[t] The measurements were carried out in a dc glow discharge. A rare gas was seeded with cesium. Both the dc and microwave conductivity were measured.

[u] The dc conductivity of an arc discharge was measured. Corrections for the effect of electron ion collisions were made.

[v] See Footnote j of Table I.

[w] See Footnote l of Table I.

[x] Microwave method similar to Gould and Brown (Footnote d), except that mean electron temperature directly determined using microwave radiometer, and iterative technique using trial set of momentum-transfer cross sections used rather than assuming analytic form for σ_{MT}. Excellent agreement with σ_{MT} results obtained by Golden (1966) from Golden and Bandel (1965) using modified effective range theory.

[y] See Footnote x. Reasonably good agreement with Gilardini and Brown.

[z] Fair agreement with Frost and Phelps although position of the Ramsauer minimum is shifted and is somewhat deeper.

crossed-beam techniques, or a swarm-type measurement in an active system. The results, where deemed of reasonable reliability, relative to the difficulties associated with such experiments, are listed in Table I, and appropriate comments are offered there.

In general, it can be stated with some emphasis that while data involving unstable systems exist, there has as yet been no precision experiment performed, and that probable errors involving factors of 2 and more are to be expected in this work.

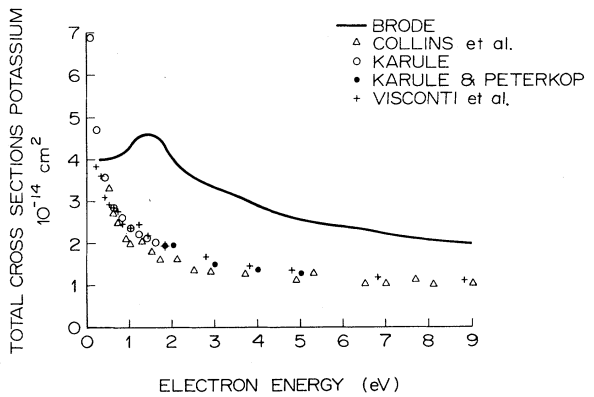

FIG. 17. Summary of total electron–potassium cross section measurements, compared to close-coupling calculations of Karule and Karule and Peterkop. Taken from Visconti *et al.* (1971).

The diatomic homonuclear molecules studied by both the Ramsauer and crossed-beam techniques are also listed in Table I. We present no swarm data on molecules in Table I since it is not generally possible to make a direct comparison between σ_{MT} and σ, due to the existence of inelastic channels, particularly of rotational excitation, at very low energies.[31]

4. CONCLUSIONS

The Appendix briefly summarizes the essential features (including the principal drawbacks) of the three basic methods employed in most total cross section work (Ramsauer apparatus, dc swarms, and crossed beams). The experimental difficulties peculiar to the determination of total cross sections have been, we trust, clearly emphasized in this article. To underscore this remark, we can state here that for *no* experiment yet performed in either transmission or beams has a complete analysis of the experiment, including a complete error analysis, been presented in the literature. In the crossed-beams experiment, where such an analysis is in principle simplest (though by no means trivial), signal-to-noise problems often make for

[31] A notable exception to this statement is the recent experiment of Crompton, Gibson, and McIntosh (1969) on parahydrogen at 77°K. In this experiment only a single rotational state is populated, and the $J=0\rightarrow J=2$ excitation is the only inelastic channel available in an appropriate range of E/N.

statistical errors so large as to be, in some cases, rather uninteresting.

With regard to swarm experiments, we believe that Crompton and his co-workers have demonstrated the feasibility of using low energy dc swarm experiments both to measure drift velocities and other transport properties to high precision and to use these to determine momentum-transfer cross sections. It is to be expected that this work will continue to yield improved results, particularly for the remaining rare gases and for other select systems at an increased range of temperatures, including cryogenic temperatures. One could particularly hope that such measurements will be extended to the alkali metals.

It appears reasonably certain that the total electron–helium cross section is the best known. Even so, further work remains to be done before this can be accepted as a suitable cross section standard.

The ac swarm experiment, that is, the use of microwave radiation to infer momentum-transfer cross sections, has not achieved the same degree of refinement as has the dc experiment. Because more elaborate experimental techniques must be used and a more complicated analytic procedure is needed to unfold the Boltzmann collision integral, results from ac swarm experiments are considerably less satisfactory, from the point of view of this article.[32] All variants of the ac method, including measurements of the complex conductivity and of half-widths of cyclotron absorption lines, suffer from the same basic difficulties when one attempts to convert qualitatively valid results to quantitative ones. We have therefore tabulated the better ac swarm results separately and have suggested that they represent qualitative determinations only, with possible errors of factors of 2 or more. Notable exceptions to this general *caveat* include the results of Phelps *et al.* and Gould and Brown on helium and, to a lesser extent, the remaining rare gases. Measurements made in active discharge systems are especially difficult to interpret.

Perhaps surprisingly, the Ramsauer-type experiments performed by Ramsauer, Bruche, and others have stood the test of time rather well. One must very likely disregard the "fine structure" seen in some of their data, particularly in helium. The question of absolute values remains unresolved, by and large. No actual error estimates are given for these early measurements, nor could such estimates be now placed upon them, other than perhaps a general feeling one possesses, based upon agreement with other techniques where comparison is possible, that the typical Ramsauer experiment is reliable to better than say 25%.

The work of Golden and co-workers, while representing among the best transmission-type experiments thus far produced, was unfortunately cut short before it could acquire its full effectiveness. This phenomenon of an experimental group developing promising techniques but terminating their program prematurely for reasons not connected with scientific merit is a recurrent phenomenon in this field (and, doubtless, in others as well!). It would be highly desirable were the Ramsauer technique to now receive the complete treatment it deserves, using modern technology to its fullest capability.[33]

Crossed-beam work, really in its infancy, will ultimately yield to the experimentalist's ingenuity and produce total cross sections of stable and metastable systems which will be competitive with other methods. The recoil-type experiment seems particularly well suited for *total* cross section determinations not only because of its selectivity in observing beam-species constituents but also because there is no need to know either the neutral beam density or detection efficiency. Meanwhile, many crossed-beam problems remain to be solved, including difficulties associated with excited state populations in unstable beams, and the inherently poor statistics in many crossed-beam experiments not involving ion or photon detection. On the other hand, the crossed-beam experiment, and particularly the recoil technique, offers the best hope for performance of such measurements upon excited states (both metastable and nonmetastable), and upon spin-state selected or spatially oriented beams.

ACKNOWLEDGMENTS

We would like to thank the many individuals who assisted us in the preparation of this paper. Specifically we wish to thank Dr. A. V. Phelps, Dr. G. C. Baldwin, Dr. E. Rothe, Dr. R. Neynaber, Dr. T. M. Miller, Dr. H. H. Brown, Dr. G. H. Dunn, Dr. E. C. Beaty, Dr. W. L. Nighan, Dr. R. H. Bullis, and Dr. M. T. Elford for discussions on specific experiments and related matters. We especially thank Dr. R. W. Crompton and Dr. D. E. Golden for their generous assistance and cooperation in connection with the analyses of their experiments, and Dr. A. V. Phelps for a critical reading of the manuscript.

A substantial part of this paper was prepared while one of us (B.B.) was a Visiting Fellow at the Joint Institute for Laboratory Astrophysics of the National Bureau of Standards and the University of Colorado.

[32] That is, for the determinations of momentum-transfer cross sections. On the other hand, the use of microwave techniques to probe electron *densities* in the plasma afterglow has proved to be a very fruitful technique for studying electron loss mechanisms (e.g., ambipolar diffusion and electron–ion recombination), as exemplified by the continuing work of Biondi and coworkers at Westinghouse and the University of Pittsburgh.

[33] Such a Ramsauer experiment, in which electron beams of energies down to 0.050 eV have been obtained, is currently in progress at United Aircraft Corporation. Preliminary data on helium has been published (Bullis, Churchill, Wiegand, and Schubert, 1967).

We would also like to thank the girls of the JILA Information Center for the unstinting, unflagging, cheerful, capable, and invaluable assistance in the preparation of the manuscript, and for their patience in dealing with delays, changes, and other peculiarities of the authors.

APPENDIX: SUMMARY OF TECHNIQUES FOR MAKING TOTAL CROSS SECTION MEASUREMENTS

Analysis of Techniques

Ramsauer The technique consists of confining a low-energy electron beam to a circular path by means of a magnetic field perpendicular to the electron path. Energy selection of the beam is accomplished by a series of apertures placed on a circular path. Cross sections are determined by measuring the current to the scattering chamber and the collector (See Fig. 2) which follows the scattering chamber as a function of gas pressure. This is the so called "two cage" method. This technique when applied above 1 eV and at energies where inelastic processes are not important has given consistent cross section measurements when used by different groups. In addition, comparison with other techniques, where possible, indicates consistency of $\pm 10\%$. Recent measurements by Golden using this technique but with modern vacuum practices still shows differences with measurements using other techniques of the order of 10%. The basic defect of this technique is the lack of any analysis of its theoretical resolution, i.e., what scattering events have been detected. Analogies with straight transmission experiments indicate only 2%–3% of the scattering events are not detected, but the introduction of the magnetic field in the real problem causes considerable uncertainty in that estimate. The 10% uncertainty quoted above is our present best estimate of the accuracy of this technique and one would not attribute all of that to the uncertainty of the resolution.

Ramsauer as modified by Brode The Brode modification to the Ramsauer technique consisted of eliminating the scattering chamber. The current to the final collecting electrode was taken as the transmitted current and the initial current (i.e., scattered+transmitted) was taken proportional to the total cathode emission. The serious objection to this modification first noted by Ramsauer was that gas–cathode interactions had been demonstrated to significantly affect the energy distribution of the electrons emitted. This problem is particularly severe for heated cathodes. The basic assumption of this technique is that the initial distribution of electrons is not affected by the gas pressure or the total emission. Independent measurements using this technique have been inconsistent and comparisons with other techniques have shown serious disagreements. Modern measurements using crossed beam techniques imply errors in the alkali measurements made using the Brode modification of a factor of 2. Because of these considerations, this technique cannot be considered a reliable alternative to the Ramsauer or two cage method. In some cases, as noted in Table I, the only data available for some atomic species were taken using this technique. Errors of a factor of 2 may exist in these data, not all of which can be attributed to the basic defect noted.

dc swarms Analysis of the behavior of electron swarms moving in a gas under the influence of a uniform electric field was first used by Townsend to deduce the qualitative shapes of cross sections. Particularly notable was the prediction of the deep minimum in the argon cross section at low energies. More recently, with the advent of more precise measurements of transport coefficients and fast digital computers, a very sophisticated analysis due to Phelps and Crompton yields precise and quantitative momentum transfer cross sections. The basic technique is to use an assumed cross section to deduce an electron velocity distribution which then is used to calculate the transport coefficients (drift velocities, diffusion coefficients, etc.). The input cross section is then adjusted until agreement with the transport coefficients is achieved within the accuracy of the original transport data. The resulting "best fit" cross section is regarded as the measured cross section. If inelastic processes are possible, the analysis is more complicated and less unambiguous. The procedure is extremely sensitive to small cross section changes when the change occurs slowly with energy. Comparison with the latest theoretical elastic scattering cross sections indicates very good agreement. The basic defect of this technique is that the calculated electron velocity distribution is not an observable. Errors in the calculated velocity distribution will result in errors in the "best fit" cross section. Evidence that no serious errors exist in this calculation is deduced from the fact that transport coefficients other than the drift velocity, which is the most precise data, can also be accurately predicted. Barring any indication that the velocity distribution calculation is seriously in error, the momentum transfer cross sections due to elastic collision would appear to be accurate to a few percent when the cross section varies slowly with energy, as in helium. The accuracy of the original transport data does not appear to be a limiting factor.

Crossed beams This technique consists of crossing an electron beam with a beam of the atom or molecule under consideration and observing the electrons scattered or the scattered atoms or molecules. This technique has the great advantage that the geometry of the intersecting beams can be accurately probed and therefore the resolution can be calculated. In addition, many unstable species can be prepared in a beam which could not be observed using other techniques. The two

serious defects this technique has are the following: (a) The low atom (or molecular) beam densities result in small scattering probabilities and therefore small signal-to-noise ratios. (b) Preparation of unstable targets may result in considerable uncertainty in the state of the target. Because of the large statistical variations, a general statement about the accuracy of this technique is difficult. It does appear that the most modern measurements using atom detection are becoming comparable to the Ramsauer technique in reliability.

Others All other techniques, of which there are many, have not succeeded in producing consistent qualitative or quantitative cross section measurements, which is certainly a minimum criterion of reliability. The three techniques which have met this test are the Ramsauer, or two cage; Crompton–Phelps analysis of dc swarm data; and crossed beams.

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