N90-26779

ELECTRON-IMPACT SPECTROSCOPY

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ABSTRACT

The methods of electron impact spectroscopy and cross section measurements are discussed and compared to optical spectroscopy. A brief summary of the status of this field and the available data is given.

INTRODUCTION

The first studies of electron collision phenomena in the early 1930's contributed significantly to the understanding of the quantum states of matter by demonstrating that electrons can lose only well defined amounts of energy in collision with atoms. These first activities were followed by a long dormant period up to the 1960's when the field became very active due to need of spectroscopic and cross section data for a large variety of systems (planetary ionospheres, astrophysics, lasers and various plasma devices). The aim of this paper is to briefly describe the status of electron collision field and the methods used to generate spectroscopic and cross section data. A comparison between optical and electron-impact spectroscopy is made and elastic, excitation, dissociation, ionization, momentum transfer and total electron scattering measurements are described. The discussion is limited to electron interactions with neutral molecular (atomic) species.

ELECTRON-IMPACT VERSUS OPTICAL SPECTROSCOPY

In a typical electron scattering experiment (see Fig. 1) an electron beam of well defined energy (E_o) crosses a target gas beam and the scattered electron signal at a direction θ with respect to the incoming beam is measured as a function of energy lost by the electrons under single collision conditions. The resulting curve is an energy-loss spectrum. Fig. 2 shows such spectrum for He. It is very similar to an optical absorption spectrum. The locations of the energy-loss features contain the spectroscopic information (energy level scheme of the target species) and the scattering intensities are related to the corresponding cross sections. Indeed, it can be shown that at high impact energies and small scattering angles (small momentum transfer conditions) the optical absorption and electron-impact energy-loss spectra are equivalent and the same cross sections can be deduced from either of them. In other words, the same (dipole) selection rules apply for both cases and optical absorption

and photoionization cross sections can be obtained by electron scattering techniques. The sharp (in time) electric field experienced by the target in high-energy electron collision, may be Fourier-transformed to represent a wide range of frequencies. The electron, therefore, corresponds to an ideal continuum light source.
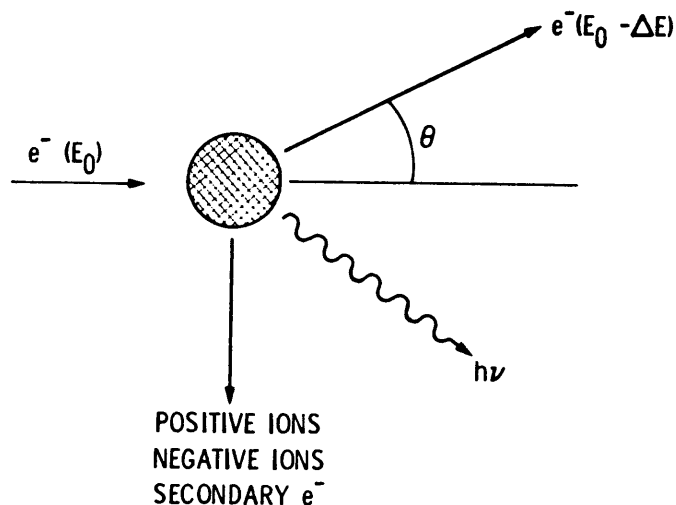

Fig. 1: Schematic representation of electron-beam/molecular-beam scattering experiment. The incoming and scattered electron beam as well as secondary products (electrons, photons, ions, dissociation fragments) are indicated.

At low impact energies and large scattering angles (small momentum transfer conditions) the situation is quite different. Electron collision processes do not obey optical selection rules and all types of symmetry and/or spin forbidden transitions may take place yielding information which is not available from optical spectroscopy. E.g. in Fig. 2 the ($^1S \rightarrow ^3P$) spin forbidden excitation peak is quite prominent, while in optical spectra it is 10^7 times weaker than the optically allowed ($^1S \rightarrow ^1P$) excitation. An additional important difference between the two methods is that in electron-impact spectroscopy the full spectrum ranging from the microwave to the x-ray region can be obtained in a single scan with the same instrument while in optical spectroscopy very different techniques are required for various spectral regions.

Now turning to the quantitative aspects of electron-impact spectroscopy, it should be mentioned that the cross section data can be obtained either from the measurement of the scattered electron signal or from the measurement of the secondary electron, photon, ion or fragment signals associated with the scattering process in question (see Fig. 1). More detailed information concerning the electron molecule interactions (multiply differential cross sections, scattering amplitudes and various coherences among these amplitudes) can be obtained from coincidence measurements of two (or more) particles.

DEFINITION OF CROSS SECTIONS

The parameter which quantitatively characterizes a scattering process is the cross section. It represents the time-independent probability for the occurrence of a particular scattering process. The notion of constant collision cross section, used in gas kinetic collisions, is not appropriate for electron scattering since the interaction is velocity and angle dependent.

Beam-beam scattering measurements yield the differential scattering cross section (DCS) for a given process (n).

$$DCS_n(E_o, \theta) = \overline{\frac{d\sigma_n(E_o, \Omega)}{d\Omega}}, \tag{1}$$

where the averaging over azimuthal angles (ϕ), instrumental energy and angular resolution and experimentally indistinguishable processes is indicated by the horizontal bar. Integration over all solid angles then yields the integral cross section

$$\sigma_n(E_o) = 2\pi \int_o^\pi DCS_n(E_o, \theta) \sin\theta d\theta \tag{2}$$

and in case of elastic scattering, the momentum transfer cross section

$$\sigma^M(E_o) = 2\pi \int_o^\pi DCS_o(E_o, \theta)(1 - \cos\theta) \sin\theta d\theta. \tag{3}$$

The total electron scattering cross section represents the sum of all integral cross sections:

$$\sigma_{TOT}(E_o) = \sum_n \sigma_n(E_o) \tag{4}$$

Where the summation includes the elastic, all inelastic and ionization channels.

The most basic quantity, of course, is the complex scattering amplitude and its square modulus represents the differential scattering cross section.

MEASUREMENT OF CROSS SECTIONS

Differential Cross Sections

The experimental techniques used to obtain electron collision cross sections of interest to us here are briefly summarized below. More detailed description of these techniques have been given by Trajmar and Register (1984) and Nickel *et al.* (1989).

Most of the electron collision cross section measurements are carried out with an electron beam-molecular beam configuration (see Fig. 1.) The resulting energy-loss spectra

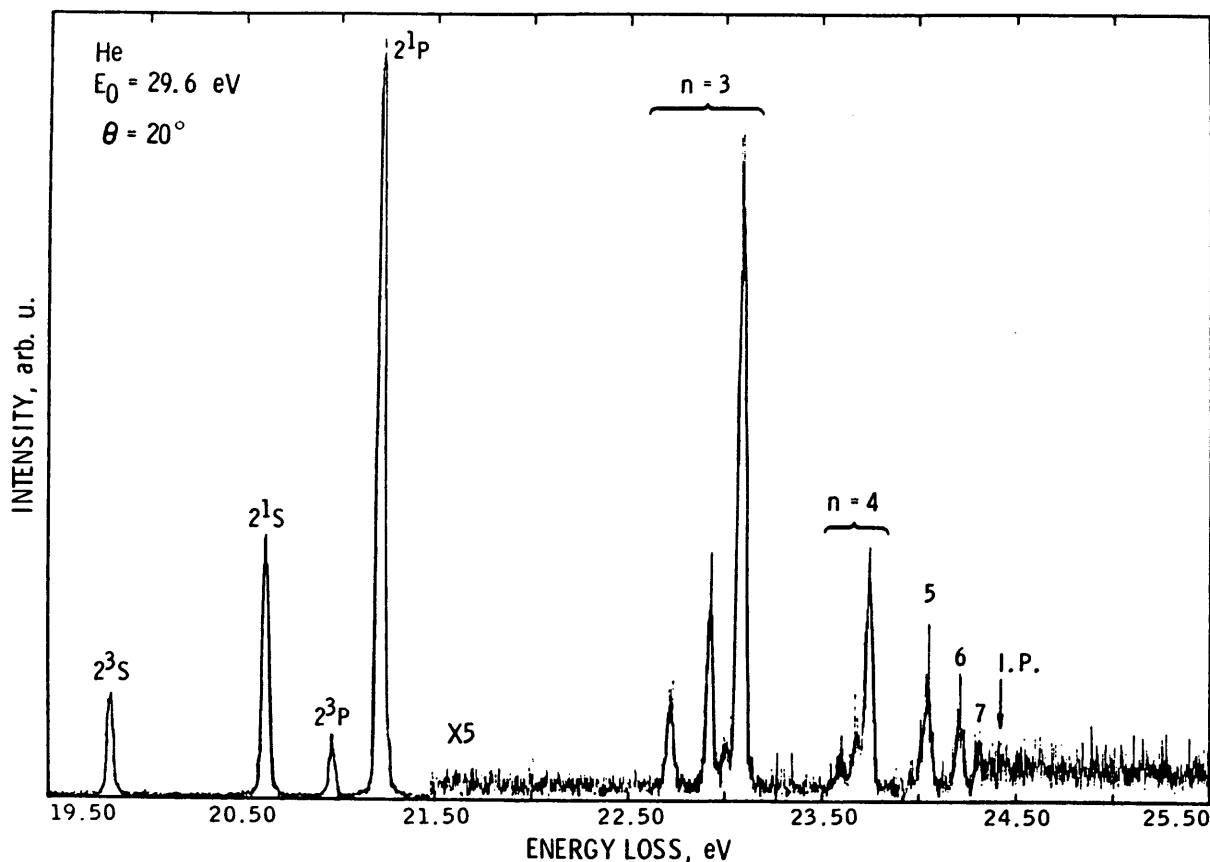yield scattering intensities associated with a given process at fixed E_o and θ values. (See Fig. 2).

Fig. 2 Energy-loss spectrum of He. The region corresponding to the $n = 2, 3...$ manifold excitations and to the ionization continuum is shown under medium momentum transfer conditions.

The scattering intensity is related to the differential cross section as

$$I_n(E_o, \Omega) = \int_{\vec{r}} \int_{E_o'} \int_{\Omega'(\vec{r})} \rho(\vec{r}) f(E_o', \vec{r}) F(E_o', \vec{r}) \tag{5}$$

$$\times \frac{\partial \sigma_n(E_o', \Omega')}{\partial \Omega} d\Omega' dE_o' d\vec{r} \ ,$$

where $I_n(E_o, \Omega)$ is the scattering signal (counts $\sec^{-1}$) measured at a nominal impact energy E_o and scattering polar angles $\Omega(\theta, \phi)$, $\rho(\vec{r})$ is the spatial density distribution of the target atoms (molecules cm^{-3}), $f(E_o', \vec{r})$ is the distribution function (both energy and spatial) of the electron flux (electrons $\sec^{-1} cm^{-2}$), $F(E_o', \vec{r})$ is the response function of the detector, $d\Omega' = \sin[\theta'(\vec{r})] d\theta'(\vec{r}) d\phi(\vec{r})$ and $\vec{r}$ is the position vector for scattering points which contribute to the signal.

The rigorous deconvolution of the cross section from Eq. 5 is complicated but for most practical purposes we can define the differential cross section, which is averaged over the instrumental energy and angular resolution (and azimuthal angles), as defined by Eq. 1

and thus obtain the simple equation

$$I_n(E_o, \theta) = DCS_n(E_o, \theta) V_{eff}. \tag{6}$$

The problem then boils down to the determination, calibration or elimination of V_{eff} (the effective scattering volume) in Eq. 6. It is a lengthy procedure and described in details e. g. by Trajmar and Register (1984) and Nickel *et al.* (1989). Basically, the procedure relies on relative intensity measurements and normalization to He elastic scattering for which accurate cross section values are available.

Integral Cross Sections

In general, the direct measurement of integral cross sections is not feasible for elastic and excitation processes. Integral cross sections then can be obtained from DCS using Eq. 2.

One can, in certain cases, deduce integral excitation cross sections from optical excitation functions which are obtained by measuring the optical radiation intensity caused by electron impact excitation and subsequent decay of the excited level. This procedure requires the knowledge of cascade contributions and branching ratios and is applicable only to optically allowed excitation processes in general. Note that the measurement of the optical excitation functions and electron impact cross sections are complementary to each other. Going the other way around, one can deduce from the measured electron impact excitation cross sections the optical excitation functions. For this purpose the dipole transition probability between the excited and ground levels and knowledge of cascade contributions are needed. The best philosophy is to measure cross sections if cross sections are needed and measure optical excitation functions if photoemission cross sections are needed.

Electron impact excitation to continuum states of molecules results in dissociation. If the dissociation products are ions (dissociative ionization, polar dissociation or dissociative attachment) or if the neutral disociation products are in excited states which consequently decay by radiation, the total dissociation cross sections can be determined by measuring charged particle or photon yields. Similarly direct ionization cross sections of molecules (atoms) can be conveniently determined by measuring the ion signal. One faces, however, a much more difficult task when the concern is dissociation to neutral ground state species. Only recently have these problems successfully been addressed by fast beam coincidence techniques by Helm and Cosby (1987).

At very low electron energies, when only a few inelastic channels are open, integral excitation cross sections can also be deduced from electron swarm measurements. In the swarm method a stream of electrons enter a diffusion chamber containing the target gas and proceed through the gas under the influence of an electric field. The stream proceeds to the anode, on its way colliding with the gas and spreading by diffusion. The behavior of the electrons is governed by the Boltzmann equation. The collisions with the gas

manifest themselves by affecting the Boltzman collision integral and as a consequence the macroscopic transport properties. Treating the applicable cross sections as free parameters, their value can be determined by fitting the measured transport coefficients to those defined by the Boltzmann equation. Obviously when too many parameters are required to characterize the system, the fitting procedure becomes ambiguous.

Momentum Transfer Cross Sections

In the case of elastic scattering Eq. (3) can be utilized to obtain $\sigma^M(E_o)$ from DCS_o (E_o, θ). These cross sections can also be obtained very accurately at low electron energies from swarm measurements. This is a fortunate situation since electron beam techniques become very difficult and uncertain at low electron energies. Therefore, the two methods complement each other in an ideal way and in the energy region of overlap cross checkings are possible.

Total Electron Scattering Cross Sections

Total electron scattering cross sections are measured by various transmission measurements. The attenuation of the electron beam through a gas beam (or static target) or the deflection of the molecular beam crossed by an electron beam (recoil measurements) as a function of electron energy is related to σ_{TOT} (E_o). In some variations, time-of-flight techniques are utilized to build up the full $\sigma_{TOT}(E_o)$ curve by observing the attenuation of electrons distributed over a wide energy.

At very low impact energies again swarm measurements yield reliable total scattering cross sections.

Resonances in Electron Scattering

Low-energy electron scattering processes are frequently dominated by resonances which may cause the cross sections to change drastically with impact energy. The velocity of the incoming electron at these energies is comparable to the velocities of the outer valence electrons of the atom or molecule, and it is appropriate to treat the incident electron as if it were temporarily attached, giving rise to a negative atomic or molecular ion. The negative ion state exists for a time longer than the orbital period, and it has identifiable properties. Although the concept of compound state formation has been widely utilized in nuclear physics, it was discovered in electron scattering only about 15 years ago, and became the subject of extensive studies only recently. (In a quantum mechanical picture, an interference between the direct and resonance scattering amplitudes is responsible for the drastic change of the cross section.)

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Resonant electron capture by molecules and subsequent dissociation of the molecules into neutral or ionic species have been the subject of extensive studies in recent years. If the resonance state is repulsive, dissociation may take place in competition with electron detachment. If one atom (or fragment) has a positive electron affinity, a stable negative ion fragment is formed (dissociative attachment), otherwise the products are neutral fragments plus an electron.

Brief Summary of Cross Section Data

Recent reviews and summaries of available cross section data were published by Brandsen and McDowell (1978), Hayashi (1981), Trajmar *et al.* (1983), Mark and Dunn (1984), Christophorou (1984), Shimamura and Takayanagi (1984), Srivastava and Nguyen (1987), McConkey *et al.* (1988), Heddle and Gallagher (1989), Shimamura (1989), Itikawa *et al.* (1986), Itikawa and Ichimura (1989), Itikawa *et al.* (1989), Tawara *et al.* (1989), King *et al.* (1989). The situation can be briefly summarized as follows:

Atomic Species

For H and the rare gas atoms extensive sets of measured differential and integral cross sections are available for elastic scattering and for excitation of the lowest states. Accurate momentum transfer and total electron collision cross sections are also available from a few tenths of an eV to few thousand eV impact energies. Surveys on these subjects have been published by Brandsen and McDowell (1978), Hayashi (1981), Fabricant and Shpenic (1988), Heddle and Gallagher (1989), Shimamura (1989) and King *et al.* (1989). The very recent paper by Shimamura (1989) gives an up-to-date summary of total electron scattering, momentum transfer, integral elastic, total excitation and metastable production cross sections for H and the rare gas atoms.

Extensive measurements have now been reported for atomic oxygen by Shyn and Sharp (1986), Shyn *et al.* (1986), Germany *et al.* (1987), Doering and Gulcicek (1989a and b) and Williams and Allen (1989). The cross section data was reviewed by Itikawa and Ichimura (1989). Measurements on C, N and S have been initiated (Doering and Dagdigian, 1989; Doering, 1989).

Experimental data for the metal atoms are scarce. Most of the information is available in the form of optical excitation functions and this subject was recently reviewed by Heddle and Gallagher (1989). Electron collision measurements are mostly restricted to the alkali atoms.

Ionization cross sections are available for all atomic species to a satisfactory degree for modelling purposes and they are summarized by Mark and Dunn (1984), Bell *et al.* (1983) and Lennon *et al.* (1989a), and Freund *et al.* (1989).

A summary of available experimental cross section data is given in Table I.

Table I. Summary of Experimental Atomic Excitation Cross Section Data

Species	Elastic	Excitation	σ^M	σ_{TOT}	Opt.	σ_{ION}
H	h	h	h	h	h	h
He	h	h	h	h	h	h
Ne, Ar, Kr, Xe	h	m	h	h	m	h
O	m	m	–	–	l	h
C, N, S,	–	–	–	–	–	h
Li, Na, K	m	m	m	h	m	h

h, m and l refer to high, medium and low degree of data availability. Opt. designates optical excitation functions.

Molecular Species

The molecules of principal interest for planetary atmospheres are: H_2, CO, N_2, O_2, H_2O, CO_2, SO_2, CH_4, NH_3.

Elastic Scattering cross sections have been measured for all molecular species of interest to us over a wide range of impact energies. It should be noted, however, that the rotational excitation is not separated from elastic scattering in most of the works and elastic scattering cross sections then represent the sum for these two processes.

Rotational excitation can be resolved with presently available techniques only for H_2. Unfolding of experimentally unresolved rotational structures was carried out for N_2, CO, H_2O and CO_2. At very low impact energies, integral rotational excitation cross sections can be obtained from swarm measurements. For homonuclear diatomic molecules the direct excitation is due to the polarizability or electric quadrupole moment of the molecule and the $\Delta J = \pm 2 (\pm 4..)$ selection rule applies. For molecules with permanent dipole the $\Delta J = \pm 1$ process dominates over many other processes. At low electron energies resonance processes can greatly increase the rotational excitation cross sections.

Vibrational excitation is effectively achieved by low-energy electronsespecially through resonance processes. Cross sections have been measured for all the diatomic molecules of interest to us but gaps in the energy range persist and the coverage for O_2 is rather limited. In the case of the polatomic molecules, only CO_2 is reasonably well covered and only very fragmentary or no information is available for other species.

Electronic state excitation cross sections for discrete states have been reported for the diatomic molecules but only for a few states and over limited energy ranges. In most

cases the low energy cross sections are scarce. N_2 has been the most extensively studied molecule. Some cross section data exists for SO_2 and CH_4 but practically nothing for H_2O and CO_2. Efforts are now in progress to extend the measurements to low energies but progress is slow because of the difficulties encountered in handling low-energy electrons, the break down of the Franck-Condon principle and the presence of resonance processes. Excitations to continuum states lead to a large variety of dissociation processes (neutral or charged fragments). When the dissociation products are charged or excited they can be conveniently detected and integral cross sections can be directly measured. Most of the experimental data falls in this category. For a review on optical measurements see McConkey (1983). A more recent review on this subject would be timely. Measurements on dissociation to neutral fragments are difficult. Winters (1966) and Winters et al. (1964) applied a technique based on measurement of pressure changes (N_2). Recently by Helm and Cosby (1987) devised a fast neutral beam technique which was applied to CO, N_2, O_2 and CO_2 by Cosby (1989).

Momentum transfer cross sections are available for all molecular species with good accuracy over a very wide energy range from combined swarm and beam data and various consistency checks. A comprehensive summary is given by Hayashi (1981).

Total electron scattering cross sections are also available with high accuracy over a wide energy range for all molecular species (see Hayashi, 1981). These are the most reliable cross sections and can serve for consistency checks on integral cross sections.

Total Ionization cross sections are available for all molecular species but the energy range is much more limited and the accuracy of the data is much lower than in the case of atoms. For summary of available data see Mark and Dunn (1984), Ehrhardt and Langer (1987) and Lennon et al. (1989b). The situation is complicated by the fact that a large number of ionization channels leading to various charged fragments, besides the parent molecule ionization, are available. Information on partial and multiply differential cross sections are fragmentary.

The status of molecular cross section data is summarized in Table II.

Table II. Summary of Experimental Molecular Excitation Cross Section Data

| Species | Elastic | Excitation | | | | σ^M | σ_{TOT} | Opt. | σ_{ION} |
		Rot.	Vibr.	El. D.	El. C.				
H_2	h	m	h	m	m	h	h	m	h
CO	h	(m)	h	l	l	h	h	m	h
N_2	h	(m)	h	h	m	h	h	m	h
O_2	h	–	m	h	l	h	h	m	h
H_2O	h	(m)	m	–	l	h	h	m	m
CO_2	h	(m)	h	–	l	h	h	m	m

h, m and l have the same meanings as in Table I. El. D. and El. C. refer to electronic discrete and continuum state excitations, respectively. Opt. refers to optical excitation functions.

For SO_2, CH_4 and NH_3 the cross section information is very fragmentary.

Summary and Future Prospects

The large body of electron collision data which is required for reliable modelling of ionospheric behavior is not available at the present time and can not be expected to be available from laboratory measurements alone in the foreseeable future. This is partly due to experimental difficulties and partly to time and budgetary constraints.

The best approach to generate the necessary information is a joint experimental/theoretical attack. Laboratory measurements should supply benchmark data. Theory checked against experimental data should be able to extend the energy ranges covered by measurements and supply data in cases when measurements are very difficult or impossible with present techniques. This approach should work well for atomic species. For molecules, the situation becomes much more difficult as far as theoretical calculations are concerned and even in the case of simple molecules theoretical methods and approximation will have to be extensively checked against experiment before they can be trusted.

For completeness, it should be mentioned that the introduction of coincidence techniques and the application of lasers (both for preparation of target species and for analyzing collision products) opened up new areas of electron collision physics. Investigation of the fine details of the collision process is now possible, information about the complex scattering amplitudes and their coherences can be obtained, and the measurements can be extended to excited target species.

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