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Electron Swarm Study
-An Effective Approach to Electron-Atom/Molecule Collision Study-

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Abstract

Electron swarm studies to derive electron collision cross sections of atoms and molecules from measurements of electron swarm data are reviewed. Stress is placed on those using molecular gas-rare gas mixtures.

1. Introduction

In order to understand the properties of weakly ionized plasmas in terms of electron collisions with atoms and molecules and of subsequent reactions, it is important to construct detailed models of the plasmas. And if simulations based on these models are to have any reality we will need a complete set of reliable cross sections for the elastic and inelastic collisions for atoms and molecules in the gas mixture. Unfortunately many of the cross sections for molecules used in the current plasma processing and laser technologies are not known and the need for a quantitative understanding of these plasmas put increasing demands on the availability and reliability of related cross sections in the range fo low energies. Electron collision cross sections for various atoms and molecules are determined by electron beam techniques and electron swarm techniques experimentally. And it is well known that the advantages and disadvantages of the two techniques are completely complementary(Huxley and Crompton 1974), and close cooperation between groups using the alternative techniques has been urged for better cross sections.

Although the electron swarm studies have been carried out in pure gases mostly, we started a new type of electron swarm study using molecular gas-rare gas mixtures in order to determine low-lying inelastic collision cross sections for the molecule. So far we have been successful in applying the new swarm technique for variety of molecules, and determined a number of molecules including SiH_4, CF_4, GeH_4, C_2H_6, and CO_2.

The swarm behavior of electrons in gases is described in the following, and stress will be placed on determinations of the atomic and molecular cross sections for low energy electrons from measured swarm parameters.

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2. Basic principles of electron swarm study

J.S. Townsend and his collaborators(1921) found the energy dependence of the mean free paths of the electrons. The discovery means the energy dependence of the total electron collision cross section, and it was the first electron swarm study. Frost and Phelps(1962) first applied numerical approach in solving the Boltzman equation and in evaluation of the integrals for transport parameters. The introduction of the numerical technique increased the scope and the accuracy of the results derived from swarm experiments revolutionary.

Energies of the electrons in a swarm are not confined in a narrow range as electrons in a beam have and this may be the obvious disadvantage of swarm experiments to beam experiments. In spite of this disadvantage, however, the electron swarm studies have unique features which can complement the results of beam experiments. Firstly, they can derive very low energy cross section data which are more reliable than those from beam studies. Secondary, swarm experiments are usually carried out in relatively high gas pressure and, therefore, the number density of the gas is determined accurately, on which the absolute magnitude of the cross section depends. The determination of the cross section depends. The determination of the absolute value is a major difficulty intrinsic to the beam experiments.

(a) Elastic momentum transfer cross sections for atoms

The basic procedure of electron swarm study is summarized in Fig. 1. Determination of the cross sections involves repeated calculation of swarm parameters and modification of related cross section(s) over a certain energy range until errors between the calculated and the experimental swarm data come with the experimental error limits.

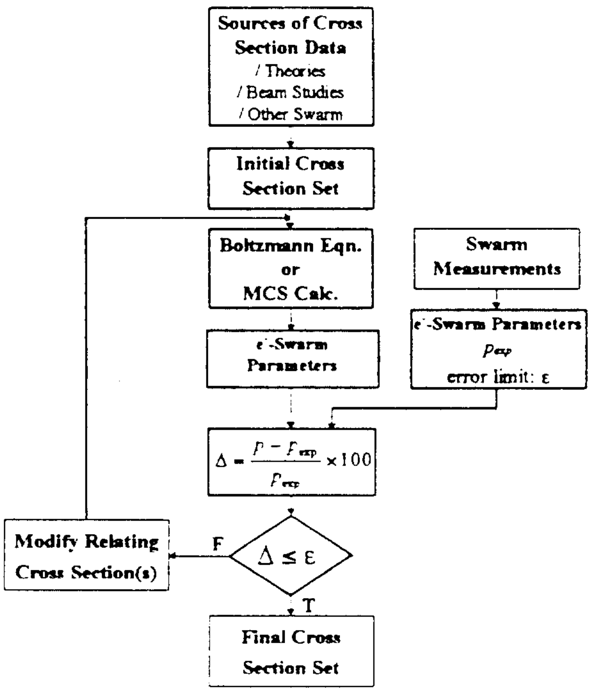

Fig. 1. The basic procedure of electron swarm study.

In the absence of inelastic scattering, for example, in the monatomic gases at low values of E/N, the magnitude and the energy dependence of the derived elastic momentum transfer cross section depend solely on the accuracy of the experimental swarm data. Crompton et. al.(1967) determined a elastic momemtum transfer cross section of helium atom from measured electron drift velocity over the energy range from 0.009 to 3 eV with claimed error limit of $\pm 2\%$(Fig.2). The electron swarm study is apparently an indirect method to obtain electron collision cross sections and, in spite of this indirectness, this method can supply the only data currently available below 0.1eV.

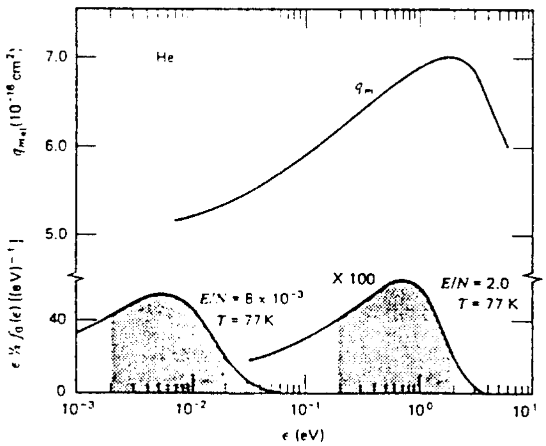

Fig. 2. The momentum transfer cross section for He and electron energy distribution functions at 77K and at E/N=8 mTd and 2Td. Approximately 80% of the electrons in the swarm have energies in the region shown shaded(Crompton *et al* 1967).

We also measured the drift velocity and the longitudinal diffusion coefficient in pure argon (Figs. 3-5) and in pure xenon and determined the elastic momentum transfer cross section for each atom.

(2) Electron collision cross sections for molecules by electron swarm study

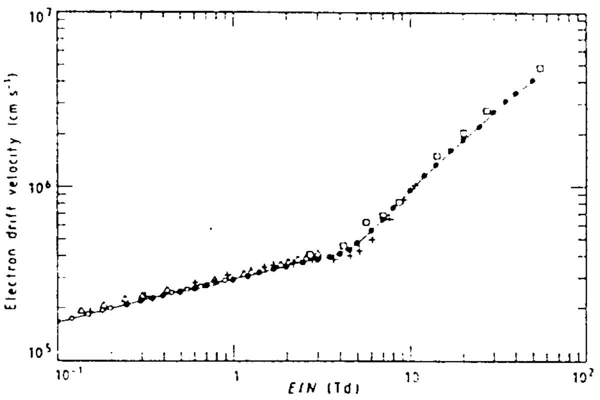

Fig. 3. The electron drift velocity as a function of E/N in Ar. The full curve shows the drift velocity calculated with the cross section set in Fig.5.
Symbols: ●, Nakamura(1988);
△,Wagner *et al*(1967);
○, Robertson(1977);
□, Kücükarpaci and Lucas(1981);
+, Christophorou et al(1979).

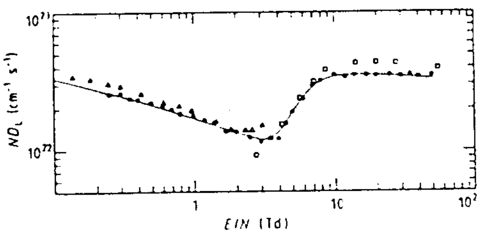

Fig. 4. The value of the longitudinal diffusion coefficient times the gas number density, NDL, as a function of E/N. The full curve again shows the result using the set shown in Fig.5.
Symbols: ●, Nakamura(1988);
△, Wagner *et al*(1967)
□, Kücükarpaci and Lucas(1981).

In recent plasma-enhanced chemical processing technologies, such molecules as SiH_4, GeH_4 and CF_4 are frequently used, and accurate knowledge of the electron collision cross sections for these molecules is needed to understand and to design plasmas for better processing.

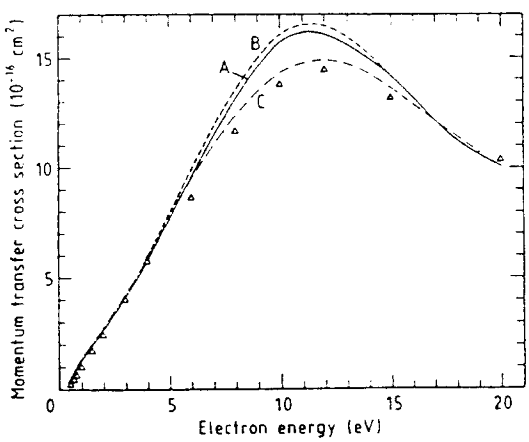

Fig. 5. Momentum transfer cross sections for the argon atom. Curves: A, Nakamura (1988); B, Fon et al(1983); C, Bell et al(1984). △, Frost and Phelps(1964).

Molecules usually have a number of competing collision processes (elastic and inelastic) with low energy electrons and it was believed to be difficult, in principle, to determine each cross section separately by usual electron swarm study. In order to avoid this difficulty and to use full advantage of swarm study we use electron swarm parameters both in dilute molecule-rare gas mixtures and in pure molecular gas. Electron swarm parameters in dilute molecule-rare gas mixtures (mixing ratio of molecular gas is usually less than a few percent and argon is used as a buffer gas) are almost insensitive to elastic cross section of the molecule because the mixing ratio of the molecule is low, but they depend mostly on the elastic momentum transfer cross section of argon and also on the vibrational excitation cross sections of the molecule. The drift velocity and longitudinal diffusion coefficient in these mixtures actually very sensitive to changes in the vibrational excitation cross sections for the molecule and, therefore, sensitive determination of them is possible. The vibrational cross sections determined by using swarm data measured in the mixtures are then used to analyze swarm data in pure molecular gas and to derive the momentum transfer cross section for the molecule. The momentum transfer cross section and the vibrational cross sections can also be used to analyze the ionization coefficient and to derive the total inelastic cross section with threshold energy immediately below the ionization threshold. The ionization cross section for most gases are usually known with sufficient accuracy.

As explained above, the vibrational excitation cross sections and the elastic momentum transfer cross section can be determined separately by using electron swarm parameters measured in molecular gas-rare gas mixtures and in the pure molecular gas.

This procedure can actually be applied to a wide variety of molecules and metal atoms and we have so far determined a number of molecules including SiH_4(Kurachi and Nakamura 1988, 1989, 1991), GeH_4(Soejima and Nakamura 1993), CO_2(Nakamura 1994) and so on. A few examples will be shown below.

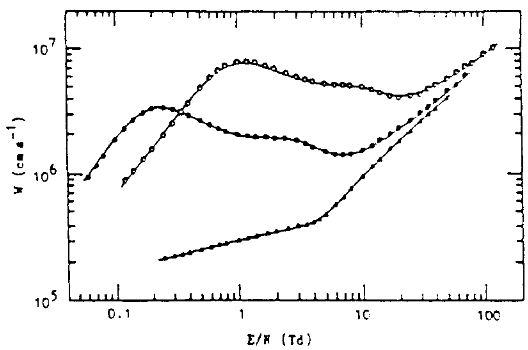

Fig. 6. The electron drift velocity in SiH_4-Ar mixtures as a function of E/N.

In Fig.6 and 7 show, respectively, the electron drift velocity and the ratio of the longitudinal diffusion coefficient to the electron mobility in SiH_4-Ar mixtures. Solid circles and open circles show the measurements in 0.501% and 5.04% SiH_4-Ar mixtures and solid triangles show the results in pure argon. Solid lines show the calculation using the cross section set shown in Fig.8 for SiH_4 molecule. Main feature seen in swarm parameters in the mixtures depends solely on the vibrational excitation cross sections of the SiH_4 molecule and the elastic momentum transfer cross section of argon atom. Fig.8 shows the set of cross sections for SiH_4

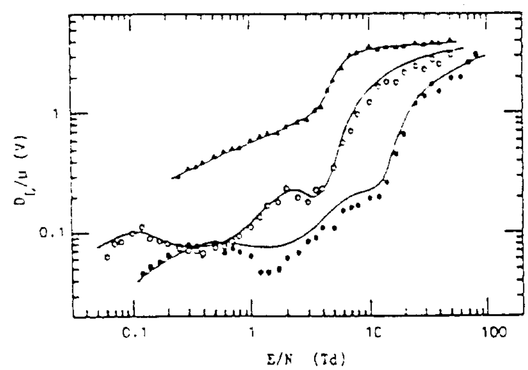

Fig. 7. The ratio of the longitudinal diffusion coefficient to the electron mobility in SiH_4-Ar mixtures.

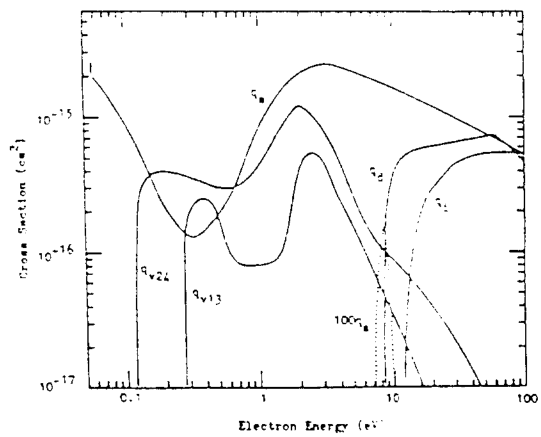

Fig. 8. The derived electron collision cross sections for SiH_{15} molecules: qm-momentum transfer cross section; q^{v24} and q^{v13}-vibrational excitation cross sections; qa-attachment cross section; qd-dissociation cross section; and qi-ionization cross section.

molecule determined so that the measured swarm parameters can be reproduced by the Boltzmann equation analysis. The elastic momentum transfer cross section and the dissociation cross section in Fig.8 are determined from the electron drift velocity (Cottrell and Walker 1965) and the ionization coefficient (Shimozuma and Tagashira 1986), respectively, measured in pure SiH_4.

3. Conclusions and the future scope of electron swarm studies

The application of high speed computers and the sophistication of numerical methods have improved the accuracy and the scope of the electron swarm study revolutionary. The analysis of electron swarm data can produce cross section data for very low energy electrons. The uniqueness of the derived cross section is strengthened by using additional swarm parameters, for example the longitudinal diffusion coefficient, other than the electron drift velocity.

The alternative use of the electron swarm parameters measured in a pure molecular gas and its dilute mixtures with argon has made the electron swarm study possible to determine elastic and inelastic collision cross sections of the molecule separately, which is not possible otherwise. New resonance in vibrational excitation cross sections for several molecules have actually been discovered by this procedure and, accordingly, new elastic momentum transfer cross sections were determined for the molecules.

The swarm study can produce cross section data for electrons with much lower energies than beam experiments can and the data from the study are expected to serve as a critical test of current theories of low energy electron scattering by atoms and molecules, in particular, as well as crucial information for quantitative simulations of weakly ionized plasmas.

4. References

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