Single- and double-electron loss of H^- in collisions with SF_6, CH_4, and CO

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Measurement of absolute, total cross sections for single- and double-electron detachment of H^- projectiles in sulfur hexafluoride, methane, and carbon oxide has been made. The relative energy range under investigation was from 1 to 5 keV. The resulting cross sections for the single-electron loss of H^- in SF_6 exhibit a flat behavior, whereas for methane and carbon monoxide the single detachment cross section increases with the energy. A free-collision model for electron loss was applied, and very good agreement was found for SF_6, while for the other targets there is a departure of the trend of the experimental data. A comparison with the results from other research laboratories is given. The measured double-electron detachment cross sections are roughly one order of magnitude smaller than the single detachment cross sections. [S1050-2947(96)03207-6]

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I. INTRODUCTION

Collisional electron detachment of negative ions is known to be one of the most fundamental processes occurring in negative-ion scattering. Their large cross sections are an indication of the important role of this process in plasma physics, ionospheric and flame chemistry, and astrophysics. From a fundamental point of view, negative-ion collisional detachment can serve as a probe of the subtle effect of electron correlation.

Most of the experimental data and theoretical calculations for negative-ion detachment have been performed for H^- in noble gases as a target. A review and recent data for these processes was published recently by Kvale *et al.* ([1], and references therein), in the energy range of 5–50 keV, using He, Ne, and Ar as targets. These same authors [2] published data for double-electron detachment in the same energy range and for noble gas targets. Electron detachment data for H^- in molecules are very scarce. A review of these collision experiments can be found in Ref. [3]. From the theoretical point of view Meron and Johnson [4] pointed out that there is no exact theory capable of predicting electron loss cross sections over a broad energy range and for all possible targets; they also noticed that the most complex energy range for the theories lies when $v_p \approx v_e$, where v_p is the projectile velocity and v_e is the equivalent bound electron velocity. These authors provide a semiempirical formulation, based on the free-collision model, that can fit over a wide range of energies for electron loss.

This paper reports the absolute single- and double-electron detachment cross sections for collisions of H^- in SF_6, CH_4, and CO molecules over the 1–5-keV energy range. To the best of our knowledge there are no data in the literature over this range of energies. The free-collision model [4] was applied to H^- electron detachment in the molecular gases.

SF_6 was selected as a target because of its ability to attach electrons. Its electron affinity ranges in the literature from 0.3 to 1.0 eV, the latter value being the widely accepted value, and is comparable to the 0.75-eV value for H^-. The other molecular targets have no electron affinity, but exhibit a shape resonance that can modify the trend of the electron detachment cross sections. CO is a linear molecule, exhibiting a shape resonance at about 2 eV above the ground state of the neutral molecule as stated by Tuan and Esaulov [5], who also notice that the shape resonance cross section reaches its maximum at collision energies of about 3 keV for the H^-+CO process. This reaction has been studied by Huq, Doverspike, and Champion [3] at relative collision energies between 2 and 300 eV, and by Pilipenko, Gusev, and Fogel [6] over the range of energies from 3 to 30 keV.

CH_4 is a spherical top molecule, and has a resonant region at about 5 eV above its first ionization potential [7]. The detachment cross sections have been studied by Huq, Doverspike, and Champion [3] at relative collision energies in the energy range 2–300 eV.

To the best of our knowledge there are no absolute cross sections reported for double-electron detachment of H^- in SF_6, CO, and CH_4. In this paper we report the measured cross sections for these processes, together with the results from other laboratories and those for SF_6 with the semiempirical calculations from Ref. [4].

II. EXPERIMENTAL METHOD

A schematic diagram of the experimental apparatus is shown in Fig. 1. The H^- beam was electrostatically accelerated, momentum analyzed by means of a magnetic mass spectrometer, and passed through a series of collimators before entering the gas cell. The detachment cross sections were measured by passing the H^- beam through the gas cell. The beam interacted with the gas in the cell and emerged as a multiple-charge-state beam. This beam was charge analyzed by applying a voltage to a set of condenser plates and the intensity of each charge state was measured. The rate of increase of a charge state as the gas cell pressure was increased gave a measure of the corresponding cross section. The data from which the cross sections were calculated were obtained under ''single collision condition'' by the well-known expression

$$F_f = \alpha \sigma_{if} P,$$

where F_f is the ratio of the intensity of the charge-changed component of charge f to the intensity of the initial beam

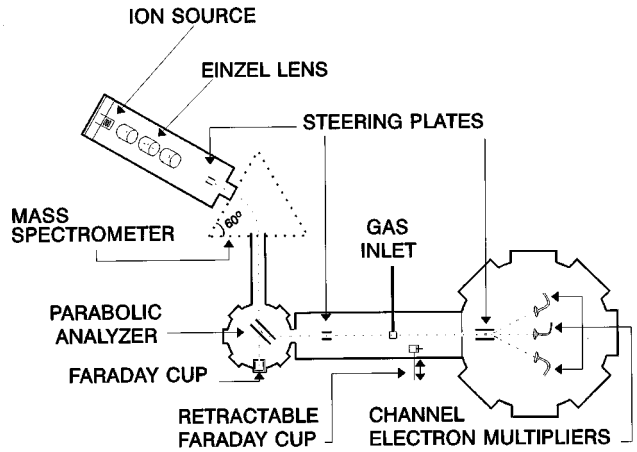

FIG. 1. Scheme of the experimental setup. Not to scale.

with single charge i, σ_{if} is the cross section for a charge changing collision, where i is the charge state of the beam particle before interaction, f is the charge state after the interaction, P is the gas cell pressure, and α is given by the expression

$$\alpha = N_L l/RT,$$

where N_L is Loschmidt's number, R is the gas constant, T is the absolute temperature, and l is the effective length of the gas cell.

The interaction cell contained the molecular gas targets (SF_6, CO, or CH_4) at a constant pressure of less than 0.1 mTorr, thereby guaranteeing single collision conditions. The interaction cell was located inside a differentially pumped vacuum chamber, where neutral and charged particles formed in the collision cell were charge separated by a perpendicular electric field produced by a pair of parallel plates. The parent H^- beam and its H^+ product were separated and guided by the field toward their corresponding detectors, a couple of channel electron multipliers (CEM) located sideways at $\pm 10°$ from the undeflected beam direction, and 17.4 cm away from the edge of the plates. A third CEM was located at $0°$, so that the neutrals could be detected. The detectors were shielded to prevent unwanted events from being registered. This geometry allowed the measurement of the intensity of the neutrals (single-electron detachment process) and positive products (double-electron detachment) formed in the target cell and also made it possible to determine the intensity of the H^- parent beam. Extreme care was taken not to exceed the allowed current on the channel electron multipliers in order to keep their gain at optimum conditions. Interchanging CEMs ensured the same gain for all detectors.

Figure 2 displays the relative intensities of the fragments formed in the gas cell as a function of SF_6 gas pressure for 1.5-keV energy of the incoming H^- beam.

III. RESULTS AND DISCUSSION

The variety of different channels operative in the interaction of a negative projectile and molecular target are described in detail in Refs. [3,8]. In addition to the direct electron detachment, in which the loosely bound electron is promoted to the continuum, or a curve crossing mechanism, other important channels are present. Over the present range of energies one of the electron detachment mechanisms involves the charge transfer to a shape resonance of the molecular target followed by a rapid decay of the negative molecular ion, leaving the molecular target vibrationally excited. Other processes such as reactive scattering, with the possibility of nuclear rearrangement and associative detachment, are important at lower energies than those used in this research.

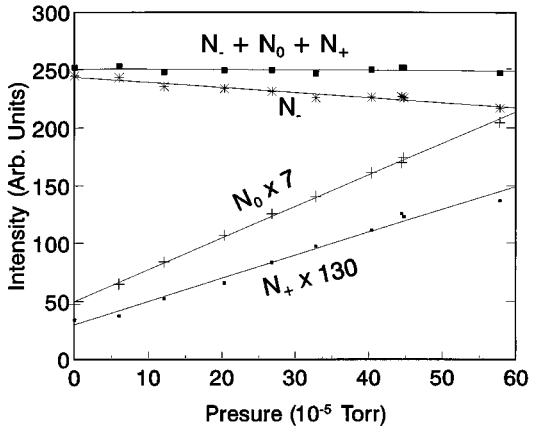

FIG. 2. Raised or decreased number of charge-changed component in an initial pure H^- beam on SF_6 at 1.5 keV.

Figure 3 displays the absolute total detachment cross section for H^- on SF_6. The cross section shows almost no variation over the energy range under study. The ''free-collision model'' [4] has been applied to this case and the results are also displayed in the same figure. This model offers four different ways to calculate cross sections, all of them showing small differences at the highest energies under

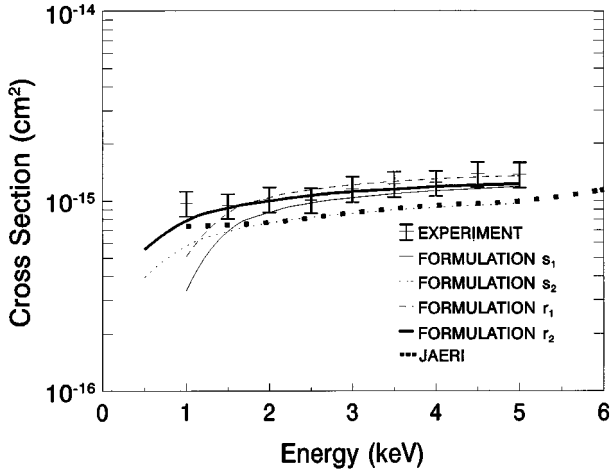

FIG. 3. Total electron detachment cross sections for H^- on SF_6 is presented with error bars. The solid line is representative of the results of the so-called r_2 formulation in the Meron and Johnson analytical expressions for electron detachment. Other formulations from the same authors are dotted, dashed line, or fine line outlined. The squares (■) are the average of the electron detachment cross sections for H^- impact on Ar, taken from Ref. [9].

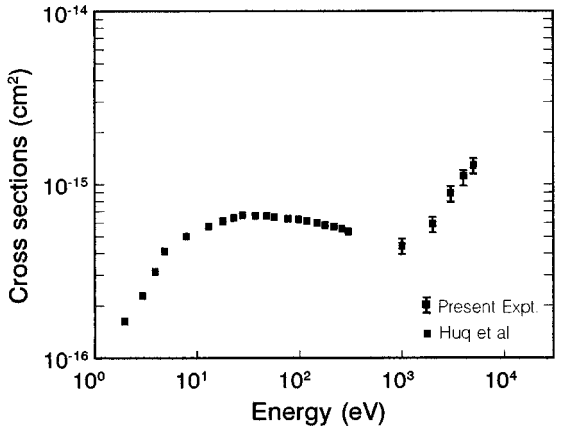

FIG. 4. Total electron detachment cross sections for H^- on CH_4. The squares with error bars represent the present results (■). The squares without error bars are from Huq, Doverspike, and Champion (■).

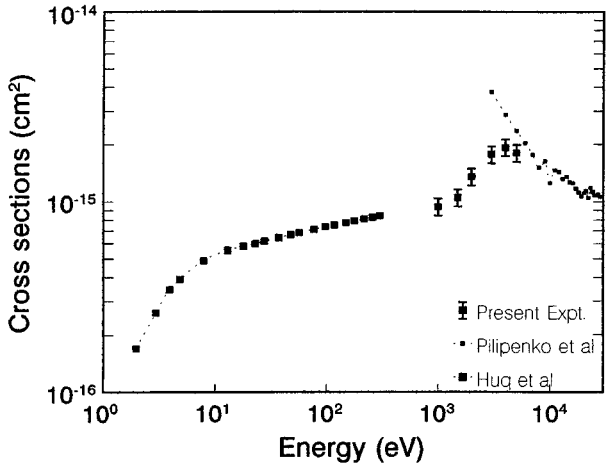

FIG. 5. Total electron detachment cross sections for H^- on CO. The squares with error bars represent the present results (■). The squares without error bars are from Huq, Doverspike, and Champion (■). The small squares are from Pilipenko, Gusev, and Fogel (■).

investigation, but a departure from their trend is observed at lower energies. The formulation labeled $r_2$in Ref. [4], based on a target with regular electronic density and gradual variation in the ionization probability, fits remarkably well with the experimental data. The agreement with this model suggests a process involving the outer electron of H^- and the target, just as it occurs in collisions of H^- with noble gases. It is worth noticing, for instance, that the detachment cross section values for H^- on SF_6 are very close to those for H^- on Ar [9]; both targets have the same ionization potential and zero dipole moment. The only difference besides its masses is the electronegativity of SF_6, this fact being revealed by its slightly higher cross sections in comparison with those of Ar, as is shown in Fig. 3.

Total electron detachment cross sections σ_{-10} for H^- incident on methane are given in Fig. 4 as a function of the relative collision energy. Displayed on the same graph are also the available experimental results from Ref. [3]. From the plot, it is observed that a soft curve can be drawn between the maximum cross-section values from Ref. [3] and the minimum of the present ones, although there is an abrupt change in the trend, with the magnitude of the cross sections increasing steadily over the energy range under investigation. This change of behavior can be explained as follows. All the experimental data from Ref. [3] have been taken over the collision relative energy range between 2 and 300 eV. In this energy range the main process is the direct detachment, dealing mainly with one electron interaction. The electron-electron interaction for H^- begins to take place at about 1.5 keV according to Refs. [10,8], opening more reaction channels in the interaction between the projectile and the target. This fact, together with the evidence of the presence of negative-ion resonances for CH_4, [7] modify the low-energy trend of the data in the detachment process. These data were tested by the free-collision model but the calculation did not fit the experimental data, either in magnitude because the calculations based on the free-collision model are on average a factor of 2 lower, or in their trend, since it was different.

The experimental results for the total electron detachment cross sections σ_{-10} for collisions of H^- with CO have been measured by Huq, Doverspike, and Champion [3], over the relative energy range from 2 to 300 eV, and by Pilipenko, Gusev, and Fogel at projectile energies from 3 to 30 keV. These measurements are displayed in Fig. 5, together with the present ones. A smooth connection line can be drawn between the present data and those from Ref. [3] at the lower energies and also with those of Ref. [6] for energies higher than 5 keV. The rapid increase of the cross sections has the same explanation as for the H^--CH_4 system. Tuan and Esaulov [5] studied this molecular system in a time-of-flight tube and observed a significant excitation of the CO molecule. They also found some similarities with the H^--N_2 system [11], where the detached electron energy spectrum is more prominent at about 3 keV, thus indicating that it is at these energies that a charge-exchange shape resonance increases the reaction. It is noted also that it is at energies of $\sim$1.5 keV and over where the electron-electron interaction begins to be important [10]. The occurrence of both physical

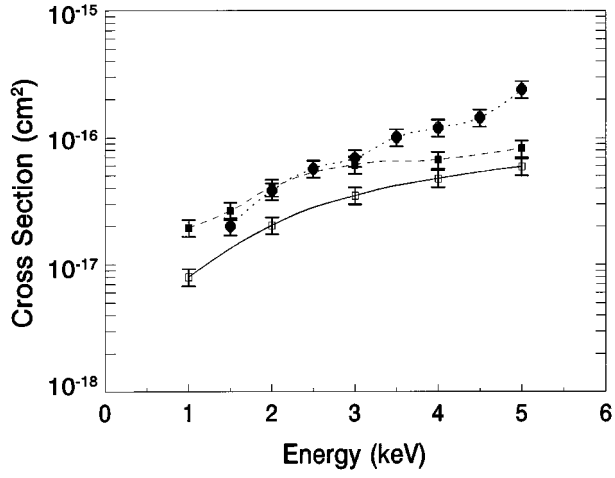

FIG. 6. Double-electron detachment cross sections as a function of the incident H^- ion for impact on (a) SF_6, solid circles (●); (b) CO, solid squares (■); and (c) CH_4, open squares (□).

phenomena modifies the trend of the electron detachment cross section. The free-collision model was again applied, but it did not fit the experimental data, its fate being similar to that of the H^--CH_4 process discussed above.

Double-electron detachment has been studied also mainly on noble gases ([2], and references therein) and one theoretical approach to calculate the cross sections based on the semiclassical free-collision model is presented in Ref. [12]. There are no double-electron detachment cross sections of H^- on SF_6, CO, and CH_4 available with which to compare our data, and no attempt was made to expand the theory at lower energies for the molecules under study.

Finally, Fig. 6 displays the absolute total cross sections for double-electron detachment of H^- in SF_6, CO, and CH_4. It is seen that the three cross-section sets increase their values with the energy, and that they follow approximately the same trend.

IV. SUMMARY

The absolute values of the total cross sections for single- and double-electron detachment of H^- on SF_6, CH_4, and CO have been measured. The single-electron detachment cross sections for H^- on SF_6 show almost no variation, whereas those for the other two targets indicate the influence of two processes, namely, the electron-electron interaction and the negative-ion resonances which produce a rise on the cross-section values. The semiempirical theoretical model has been applied, and very good agreement was found in the case of H^- on SF_6. Very good agreement was found between the present data and those of Ref. [3].

ACKNOWLEDGMENTS

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