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Electron attachment in dilute fluorine-helium mixtures[a)]

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We have made an absolute determination of the electron attachment coefficient η (cm^{-1}) in helium containing 0.1–1% fluorine covering an E/N range from $3\,Td$–$17\,Td$. At an estimated average energy of 5 eV we find a rate coefficient equal to $(7.5\pm1.5)\times10^{-10}$ cm^3/sec.

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Data on electron attachment in molecular fluorine are pivotal in understanding the physical mechanisms of HF [1–3] and rare gas–halide lasers[4–12]. It has recently been shown[13] that the electrical discharge in the latter type of laser operates in a stable mode when the attachment rate is equal to (or larger than) twice the ionization rate, thus justifying detailed studies of these processes.

The major objective of this letter is to present data on the process

$$F_2 + e^- \rightarrow F + F^- \qquad (1)$$

as it occurs in helium with a small amount (0.1–1%) of molecular fluorine additive.

The rate coefficient k_a for dissociative electron attachment in F_2[Eq. (1)] has recently been measured in two different experiments: (i) At an electron temperature of about 600° K Sides *et al.*[14] found $k_a=(4.6\pm1.2)\times10^{-9}$ cm^3/sec using a flowing afterglow technique. (ii) Results at higher electron energies (0.3–1.0 eV) have been obtained by Chen *et al.*[15] By normalizing their observations to the electron-ion recombination rate in a nitrogen plasma, they determined $k_a=(2.3\pm0.3)\times10^{-9}$ cm^3/sec at an average energy of 1 eV.

Our contribution has been to extend the measurements of k_a into an energy range of importance in practical lasers. The principle of the method,[16] which requires *no* normalization to other data, is to produce a short localized pulse of electrons at a photocathode and to observe the evolution of the external circuit current due to the motion of electrons and negative ions. The average electron energy in the gap between the plane-parallel electrodes is governed by the applied electric field E and total gas number density N.

A schematic diagram of the apparatus is shown in Fig. 1. Two plane-parallel aluminum electrodes 20 cm in diameter are separated from 0.5 to 3.0 cm by a micrometer screw. Light from a xenon ion laser[17] (2315 Å; 100 nsec half-width) is focused to a spot diameter of 0.5 mm on the cathode through a 1-mm aperture in the anode. The electrodes are situated inside a bakeable stainless-steel chamber with an end vacuum below of 10^{-7} Torr. Sapphire windows are used to admit the laser beam and to visually inspect the electrodes. The chamber is pumped by two 25 l/s ion pumps whose performance has not been impaired by the presence of fluorine. A mechanical forepump is isolated from the high-vacuum volume by charcoal and molecular sieve traps. Great care had been taken to "passivate" all surfaces with 2–5% F_2 in He overnight before any measurements were made. The partial pressure of F_2 and helium are known to within 5 and 1%, respectively, from measurements with a MKS Baratron (Model 220-2A1-10) and Texas Instruments Quartz Bourdon Tube.

The crucial feature of our procedures is to *integrate* the circuit current caused by the flow of fast electrons and slow negative ions and to observe the resultant voltage transient on an oscilloscope. If initially N_0 photoelectrons are produced at the cathode at time $t=0$, it can be shown that the voltage drop across the load resistor R (10^{10}–10^{11} Ω) at the transit time T_- of the electrons is given by

$$V_R(T_-) = (N_0 e/C\eta d)[1-\exp(-\eta d)]. \qquad (2)$$

In Eq. (2), e is the elementary charge, d is the electrode separation, C is the capacitance of the discharge circuit, and η is the attachment coefficient (in units of cm^{-1}). The observed voltage will further increase as the remaining negative ions drift out of gap and will reach a constant maximum value of

$$V_R(t \geq T_n) = N_0 e/C, \qquad (3)$$

where T_n is the negative ion transit time. It has been assumed above that the time constant $RC \gg T_n$. Typically, under our experimental conditions, we had $RC \approx 2.0$ sec and $T_n < 10^{-3}$ sec. The ratio of Eqs. (3) and (2)

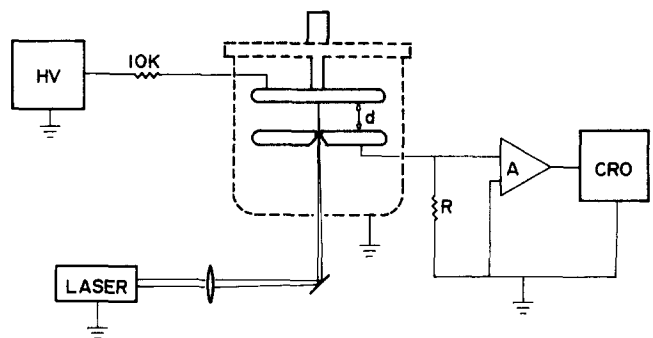

FIG. 1. Schematic diagram of the apparatus. Light from a pulsed xenon ion laser (2315 Å) is focused onto the cathode (top electrode) of a drift gap producing a burst of photoelectrons. The voltage drop across the resistor R gives information about the attachment coefficient as well as drift velocities of electrons and negative ions. The amplifier A is an unitary gain impedance converter.

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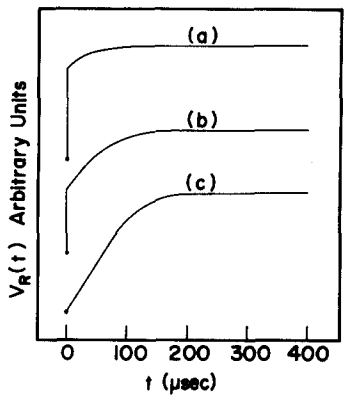

FIG. 2. Typical retraced oscillograms of integrated voltage transients (displaced in the vertical direction) chosen to demonstrate the cases of (a) weak, (b) intermediate, and (c) strong attenuation due to dissociative attachment. The fast rise in (a) and (b) is due to electrons. The slowly rising part in all curves is due to the integrated negative ion current, i.e., $\int_0^t I_n(t)\,dt = (eN_0/T_n)\{t[\exp(-\eta d)/\eta v_n][\exp(\eta v_n t)-1]\}$, where v_n is the drift velocity of the negative ions.

allows the attachment coefficient to be determined from

$$\frac{V_R(T_n)}{V_R(T_-)} = \frac{\eta d}{1-\exp(-\eta d)}. \tag{4}$$

Since η is determined from the *ratio* between two well-defined voltages, we do not have to worry about fluctuations in the initial number of photoelectrons. We determine the reaction coefficient absolutely from the relationship

$$k_a = (\eta/N')W_- \tag{5}$$

once the electron drift velocity W_- [18] and fluorine number density N' are known.

In order to give the reader an impression of the simplicity of the experimental procedure, we present examples of the integrated voltage pulse, $V_R(t)$, in Fig. 2, displaying the cases of weak [Fig. 2(a)], intermediate [Fig. 2(b)], and very strong [Fig. 2(c)] attenuation due to dissociative attachment. The ratio of $V_R(T_n)/V_R(T_-)$ as it occurs in Eq. (4) is undetermined when the attachment is so strong [Fig. 2(c)] that most of the electrons are attached to F_2 molecules close to the cathode. We also note the inherent inaccuracy in the case of weak absorption [Fig. 2(a)], and therefore adjust the experimental parameters to let $V_R(T_-)/V_R(T_n)$ fall in the range 10—90%.

The direct results for the attachment coefficient divided by the fluorine number density N' are plotted as a function of E/N in Fig. 3. We want to emphasize that this represents an absolute determination with an overall accuracy in η/N' of ± 12%.

It is of interest to convert our data for η/N' to values of the reaction rate k_a by multiplying by the electron drift velocity W_- [Eq. (5)] as shown in Fig. 4. In order to compare with Sides *et al.*[14] and Chen *et al.*[15] we have estimated the average electron energy $\bar{\varepsilon}$ in our experiment using the D/μ results of Townsend *et al.*[19] and calculated the average swarm energy by assuming a Maxwellian energy distribution, i.e., $\bar{\varepsilon} = \frac{3}{2} D/\mu$. These energies represent an *upper* bound since inelastic collisions have not been taken into account. The actual energy values can be corrected later when D/μ values become available.

Recently Nighan[20] has calculated the attachment rate in a gas mixture consisting of 0.3% fluorine in helium taking energy losses due to vibrational excitation, dissociative attachment, and dissociation into account. His results are shown as the full curve in Fig. 4 joining the low-energy data of Chen *et al.* with the present higher-energy data. The broken curve marked G & R represents the preliminary results of a calculation done by Greene and Rockwood.[21]

An additional experiment is needed to verify if k_a exhibits a maximum around 0.1—0.3 eV.

We are thankful to Professor R. W. Crompton for comments on the manuscript and to Dr. W. L. Nighan, Dr. A. E. Greene, and Dr. S. D. Rockwood for the results of their unpublished calculations.

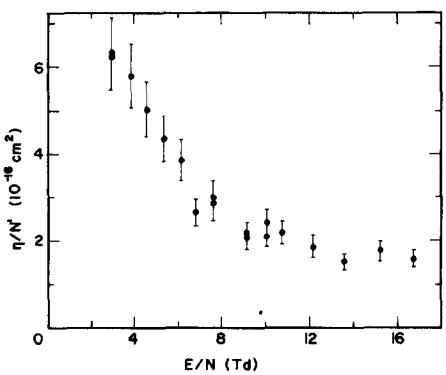

FIG. 3. Attachment coefficient divided by fluorine density as a function of E/N with a fluorine concentration of 1.0% in 10 Torr helium.

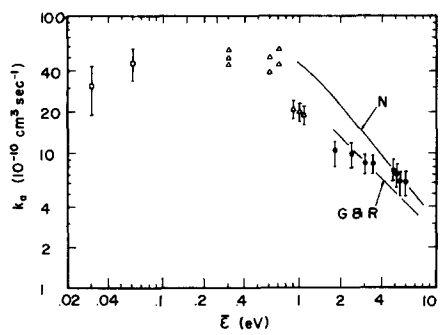

FIG. 4. Reaction rate for electron dissociative attachment in fluorine as a function of average electron energy. The broken curve shows the theoretical results of Greene and Rockwood (Ref. 21). The full curve (N) is due to Nighan (Ref. 20). □—Ref. 14; Δ—Ref. 15. The accuracy in the present data (●) is ± 20%.

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Effect of beamlet-beamlet interaction on ion optics of multiaperture sources[a)]

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Space-charge effects of beamlet-beamlet interaction in the unneutralized accelerator region of intense ion sources have been computed. If shielding effects of the electrodes are neglected, the radial space charge of a typical ion source is so great that most of the beam will hit the second electrode or will undergo nonlinear steering. Beam divergence will be an order of magnitude larger than that expected from considerations of spherical aberrations of a single beamlet caused by deviations from Pierce geometry. If shielding effects of the electrodes are sufficiently great that only nearest-neighbor beamlet-beamlet interactions are important, then the outermost beamlets (or those near a water line) will cause a total beam divergence which is the same order of magnitude as that due to single-beamlet spherical aberrations, and far more than the divergence expected from source ion temperature. A solution to this problem is proposed.

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Multibeamlet intense ion sources have been developed for plasma heating experiments.[1,2] These beams are neutralized in a gas cell after being accelerated to high energy. However, in the accelerating and electron blocking regions the beams are unneutralized and possess considerable space charge. This is utilized in the focusing of each beamlet. However, the space-charge interaction of one beamlet with the other beamlets has not been considered to our knowledge. We shall consider two situations: first, the case in which the space-charge forces are not shielded from the electrodes, so that any beamlet can interact with all other beamlets (nonlocal) and, second, the case in which the space-charge forces are shielded by the electrodes to such an extent that only nearest-neighbor beamlet interactions are important (local).

For a cylindrically symmetric array of beamlets, we take the interaction of all of the beamlets with one beamlet at a radius r, with respect to the centroid of the array, as if the ion beam were of uniform composition with a current density of the beam multiplied by the geometric transparency of the electrodes, η_g. Then we can use the standard relation for space-charge blowup of a uniform beam in a constant potential,[3,4] which is taken to be half the accelerating potential,[4]

$$r = r_0[1 + \tfrac{1}{8}\sqrt{2}\rho\eta_g(z/d)^2], \tag{1}$$

where r is the radial position of a beamlet at a position z ejected at r_0 measured with respect to the centroid of the beamlet array, d is the width of the accelerating gap, η_g is the geometric transparency of the grids, and ρ is the current density of a beamlet divided by the Child-Langmuir current density in a plasma diode[5,6] of gap d and potential equal to the accelerating potential. The divergence of the centroid of this beamlet due to

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