(12)

OFFICE OF NAVAL RESEARCH

(15) Contract N00014-77-C-0100

Project No. NR 395-573/11-12-76(421)

ARPA Order No. 2840

(9) ANNUAL REPORT

TECHNICAL REPORT NO. 1

1 December 1976 - 30 November 1977

(6) ATTACHMENT RATES IN RARE GAS-HALIDE LASERS.

BY

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(11) 31 December 1977

(12) 26p.

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PREFACE

Data on electron transport coefficients are needed to conduct parametric optimization studies of rare gas-halide lasers.

This document represents the First Annual Technical Report for progress made under contract ONR N00014-77-0100 for the period of December 1, 1976 through November 30, 1977. Contents:

Part A entitled "Electron attachment in Dilute Fluorine-Helium Mixtures" has been accepted for publication in Applied Physics Letters.

Part B entitled "Electron Drift Velocities in Helium-Fluorine Gas Mixtures" has been submitted to Applied Physics Letters.

Part C describes a point-to-plane electrical discharge that has been developed to produce a burst of electrons of 10 nsec duration.

Each part is independent with its own set of illustrations and references.

LIST OF CONTENTS

Part A

Electron Attachment in Dilute Fluorine-Helium Mixtures*

by

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We have made an absolute determination of the electron attachment coefficient $\eta(cm^{-1})$ in helium containing 0.1 - 1% fluorine covering an E/N range from 3 - 17 Td. At an estimated average energy of 5eV we find a rate coefficient equal to $(7.5 \pm 1.5) \times 10^{-10}$ cm^3/sec.

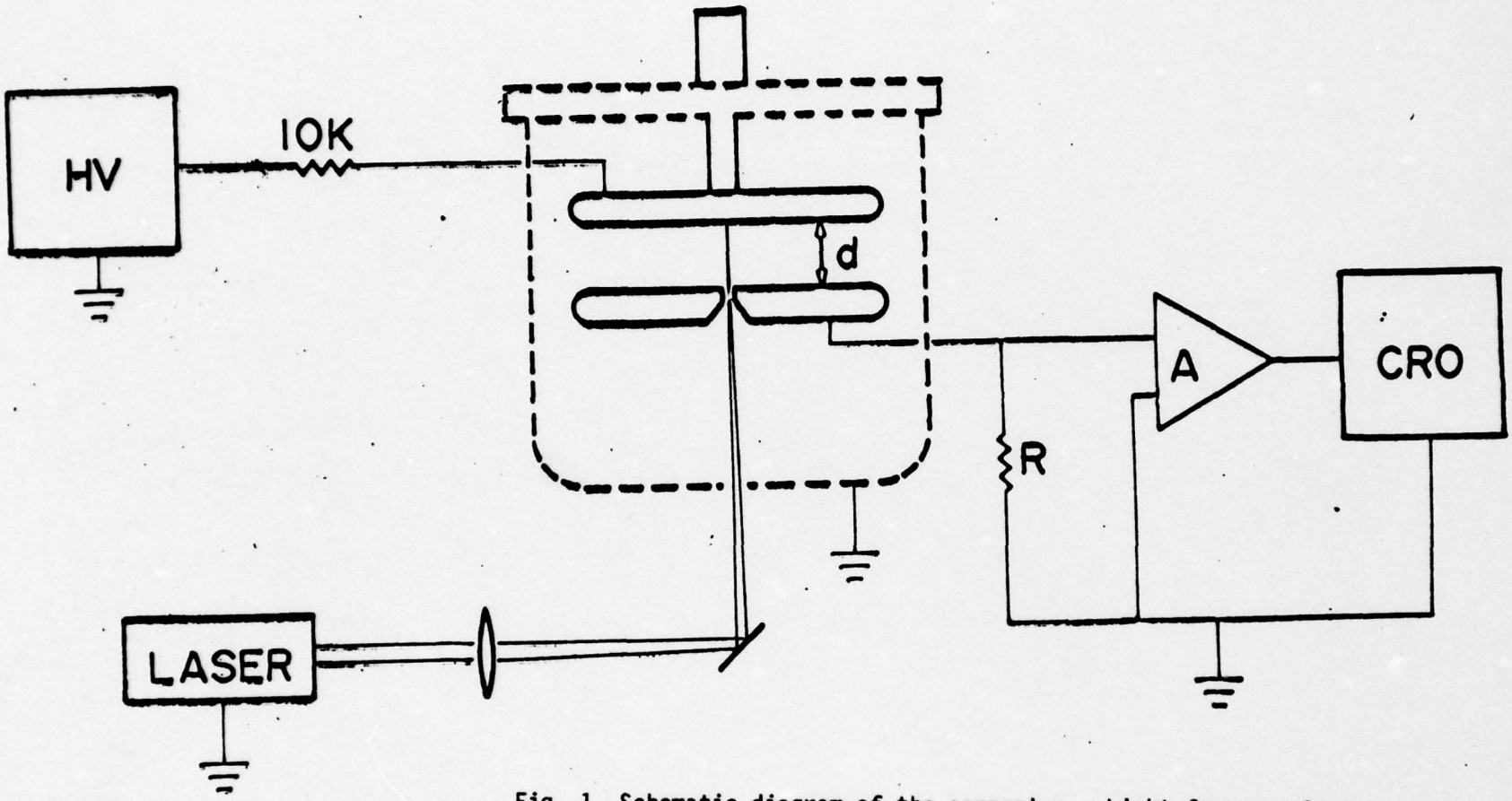


Fig. 1. Schematic diagram of the apparatus. Light from a pulsed xenon ion laser (2315 Å) is focussed onto the cathode (top electrode) of a drift gap producing a burst of photoelectrons. The voltage drop across the resistor R gives information about the attachment coefficient as well as drift velocities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

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 600K Sides *et al.*[14] found $k_a = (4.6 \pm 1.2) \times 10^{-9}$ 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 $k_a = (2.3 \pm 0.3) \times 10^{-9}$ 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 of 20 cm diameter are separated from 0.5 - 3.0 cm by a micrometer screw. Light from a xenon ion laser[17] (2315 Å; 100 nsec half-width) is focussed 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_o photoelectrons are produced at the cathode at time t = 0, it can be shown that the voltage drop across the load resistor $R(10^{10} - 10^{11}\Omega)$ at the transit time T_- of the electrons is given by

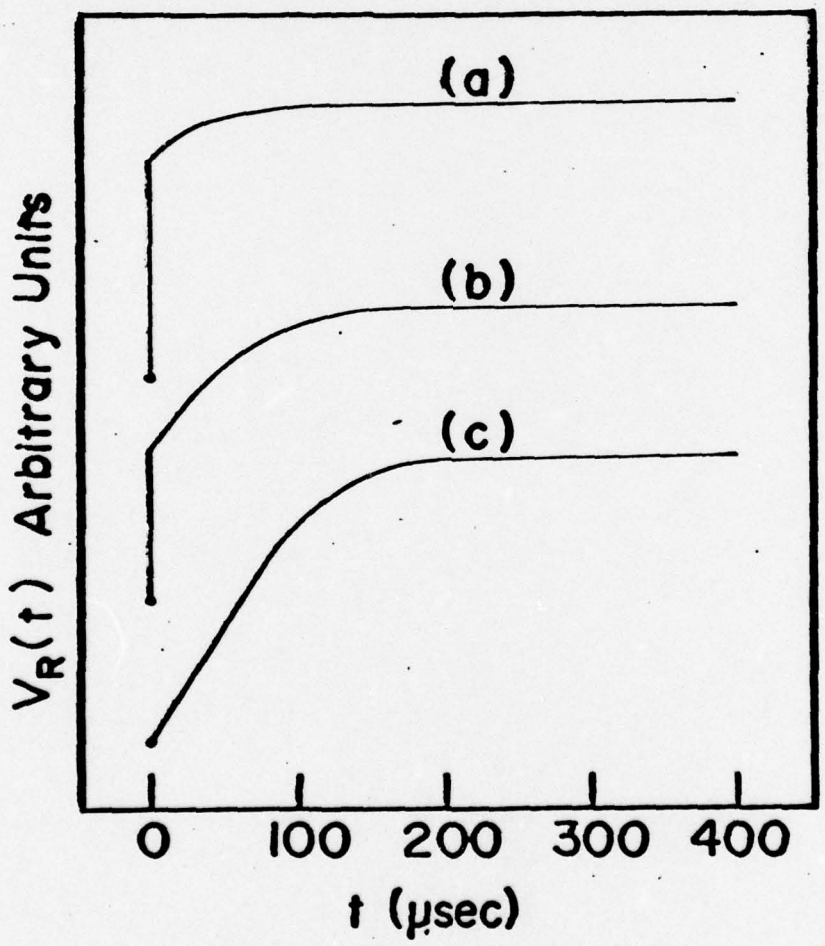
$$V_R(T_-) = \frac{N_o e}{C\eta d}\,[1 - \exp(-\eta d)]\ . \qquad (2)$$

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

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.,

$$\int_0^t I_n(t)dt = \frac{eN_o}{T_n}\left[t - \frac{e^{-\eta d}}{\eta v_n}\left(e^{\eta v_n t} - 1\right)\right],$$

where v_n is the drift velocity of the negative ions.



$$V_R(t \geq T_n) = \frac{N_o e}{C} , \tag{3}$$

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 $RC \simeq 2.0$ sec and $T_n < 10^{-3}$ sec. The ratio of Eqs. (3) and (2) 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 (a) weak, (b) intermediate, and (c) very strong 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 (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 (a), and therefore adjust the experimental parameters to let $V_R(T_-)/V_R(T_n)$ fall in the range from 10 - 90%.

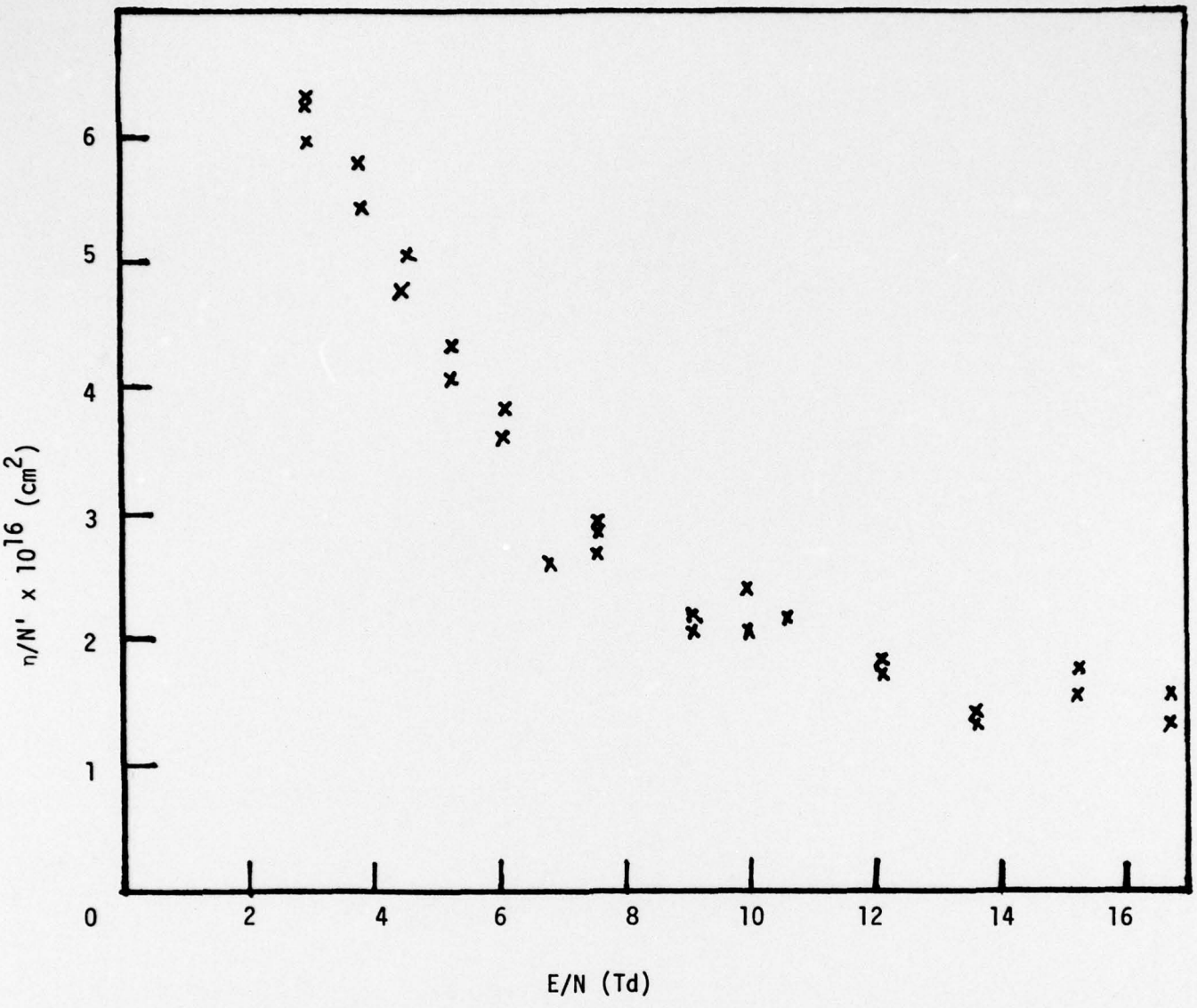


Fig. 3. Attachment coefficient divided by fluorine density as a function of E/N with a fluorine concentration of 1.0% in 10 Torr helium.

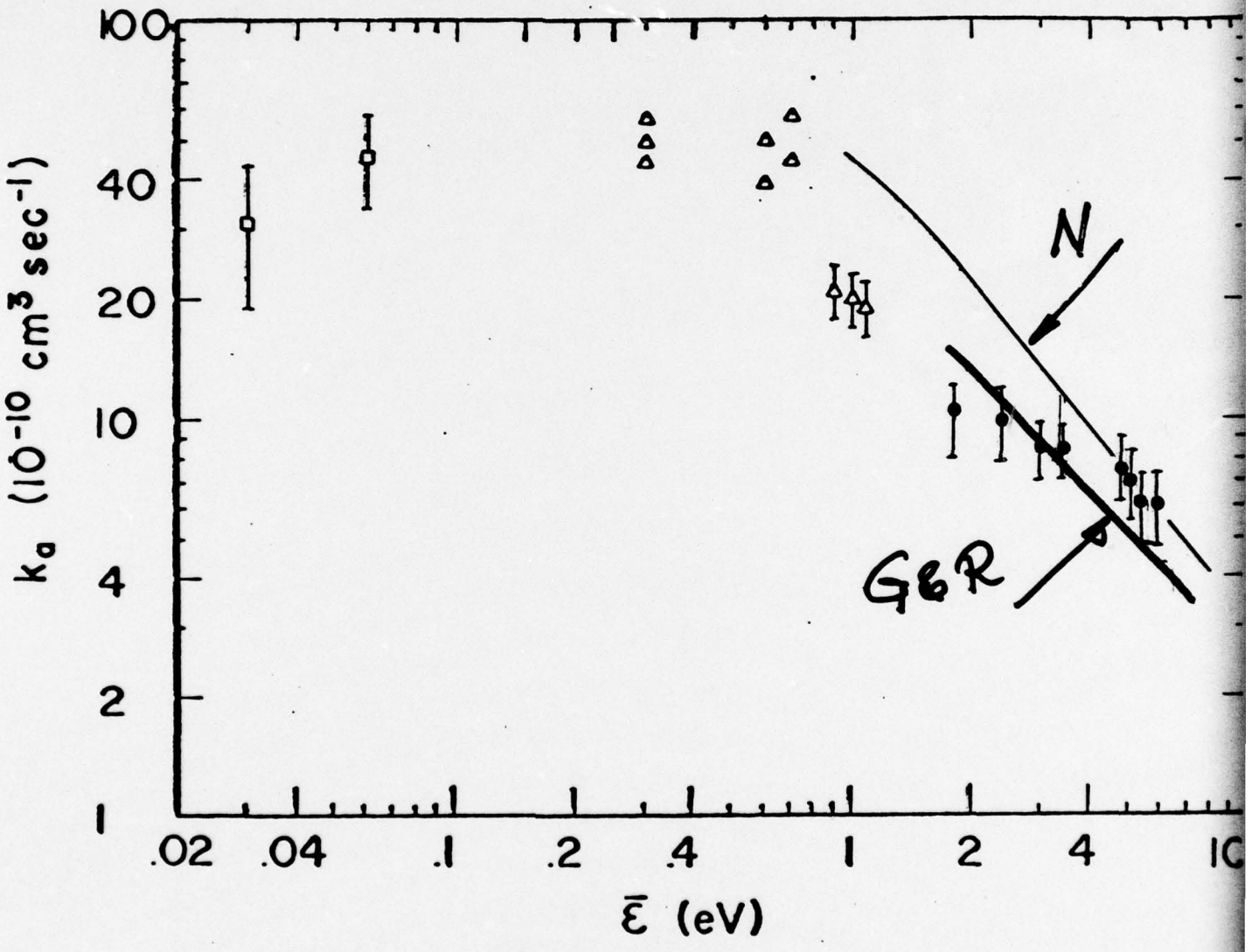


Fig. 4. Reaction rate for electron dissociative attachment in fluorine as a function of average electron energy. The broken curve shows the theoretical results of Greene and Rockwood (Ref. 21). The full curve (N) is due to Nighan (Ref. 20). □ - (Ref. 14). Δ - (Ref. 15). The accuracy in the present date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

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 $\pm$ 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 $\frac{D}{\mu}$ results of Townsend et al.[19] and calculated the average swarm energy by assuming a Maxwellian energy distribution, i.e., $\bar{\varepsilon} = \frac{3}{2}\frac{D}{\mu}$. These energies represent an upper bound since inelastic collisions have not been taken into account. The actual energy values can be corrected later when $\frac{D}{\mu}$ 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 Drs. W.L. Nighan, A.E. Greene and S.D. Rockwood, for the results of their unpublished calculations.

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18. In a separate experiment (unpublished) we have measured the dependence of W_- on fluorine concentration and found that for an F_2 concentration of less than 0.1% we may use electron drift velocities in pure helium and still be within 5% of the actual drift velocity. (See H. B. Milloy and R. W. Crompton, Phys. Rev. A 15, 1847 (1977)). For higher fluorine concentrations we rely on our own measured values.

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ELECTRON DRIFT VELOCITIES IN HELIUM-FLUORINE GAS MIXTURES*

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Using a simple time-of-flight technique, we have measured the electron drift velocity in helium with 0.1 - 1% fluorine additive. Our results in a 0.1% mixture are in close agreement with data in pure helium. The measured drift velocity increases with increasing fluorine concentration.

Modeling of HF[1] and rare gas halide lasers[2-4] requires accurate data on electron transport coefficients in gas mixtures containing fluorine. We are presently engaged in a general program for direct experimental determination of these parameters, and have previously measured the rate coefficient for electron dissociative attachment[5] in low concentrations (0.1 - 1.0%) of F_2 in He. The objective of this letter is to report measurements of the electron drift velocity v_d under similar experimental conditions.

In general, the electron drift velocity is related to the velocity distribution $f_o(v)$ by the relation

$$v_d = -\frac{4\pi}{3}\frac{e}{m}\frac{E}{N}\int_0^\infty \frac{v^2}{q_m(v)}\frac{df_o(v)}{dv}\,dv \quad , \tag{1}$$

where conventional gaseous electronics notation has been used. Note that $f_o(v)$ is normalized according to

$$4\pi\int_0^\infty v^2 f_o(v)\,dv = 1$$

and is defined in such a way that it represents the distribution function for a steady stream of electrons at a given E/N, but with allowance for inelastic collisions. A direct measurement of the electron drift velocity can therefore be used to estimate the mean energy if the effective cross section for momentum transfer, $q_m(v)$, is known.

The principle of our method is to produce a short and well localized pulse of photoelectrons at time t = 0 and observe their motion between two plane-parallel electrodes. The number density, $n(\vec{r},t)$, within the resulting traveling group of electrons, taking both elastic and inelastic collisions into account, is governed by[6]

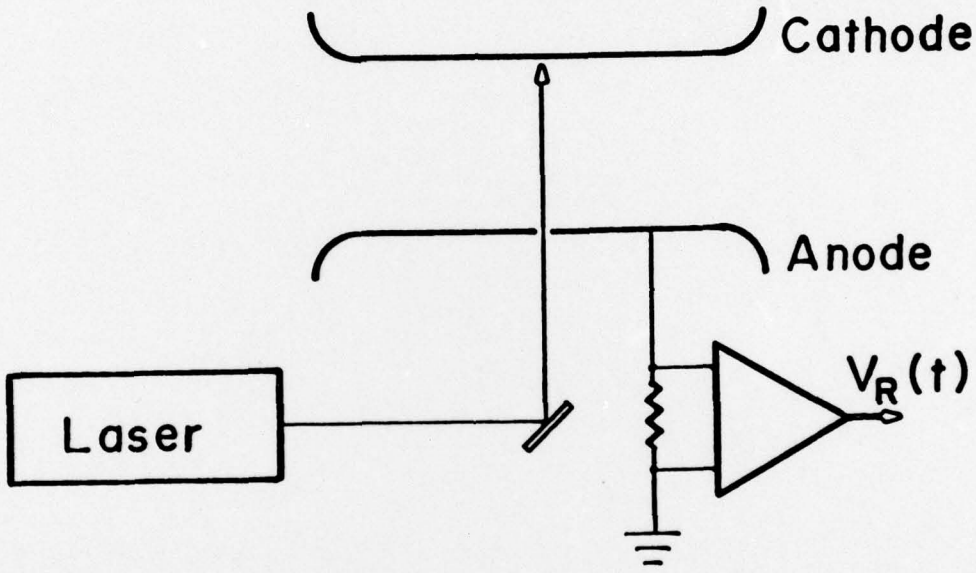

Fig. 1. Principle of Experiment. Light from a Xe-ion laser (2315 Å; 100 nsec half-width) passes through a 1 mm aperture in the anode and produces photoelectrons within a 0.2 mm dia. spot on the cathode. The resultant electron current transient is integrated and displayed on an oscilloscope.

$$-\frac{\partial n}{\partial t} + (\bar{\nu}_i - \bar{\nu}_a)n + D_\perp\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)n + D_{||}\frac{\partial^2 n}{\partial z^2} - v_d\frac{\partial n}{\partial z} = 0. \qquad (2)$$

In Eq. (2), $\bar{\nu}_i$ and $\bar{\nu}_a$ are the average ionization and the attachment frequencies, respectively. Furthermore, $D_{||}$ and $D_\perp$ represent the diffusion coefficients perpendicular and parallel to the z-direction. The distance travelled by the centroid of the diffusing electron group over a time t is given by

$$\bar{z} = v_d t. \qquad (3)$$

Assuming that an initial number of electrons $n_o(0)$ starts drifting from the cathode (z = 0) at time t = 0, the solution to Eq.(2) may be written as

$$n(\vec{r},t) = \frac{n_o(0)\exp[(\alpha-\eta)\bar{z}]}{(4\pi D_\perp \bar{z}/v_d)(4\pi D_{||}\bar{z}/v_d)^{1/2}} \exp\left[-\frac{(x^2+y^2)v_d}{4D_\perp \bar{z}}\right] \exp\left[-\frac{(z-\bar{z})^2 v_d}{4D_{||}\bar{z}}\right], \qquad (4)$$

where α is a Townsend's first ionization coefficient and η is the attachment coeffient. We note from Eq. (4) that, to the order of approximation to which it is valid to use Eq. (2), the position of the electron centroid has not been modified by the presence of weak ionization and/or attachment, and we are therefore justified in employing a time-of-flight method to determine the electron drift velocity.

The details of the apparatus have been described elsewhere[5]. With light from a pulsed Xe-ion laser[7] we generate $n_o(0)$ photoelectrons at the cathode at time t = 0. Mathematically, $n_o(0)$ has been corrected for back-diffusion to the cathode and represents the number of electrons that start drifting through the gas against the electric field $\vec{E}$. If there were no electron loss or gain processes, we would measure a <u>constant</u> current in the external circuit during the electron transit time T_-. In order to increase the sensitivity of the method

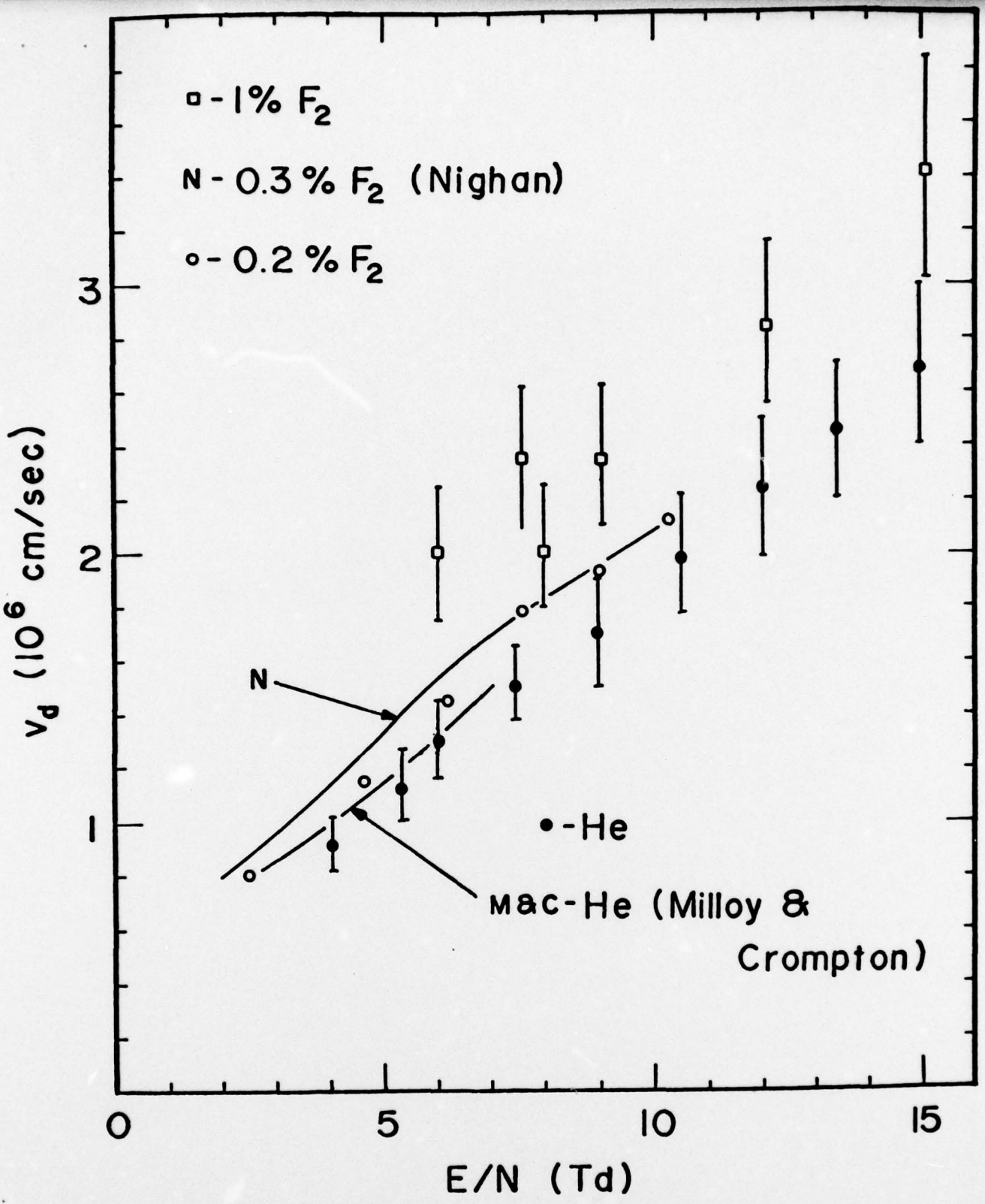

Fig. 2. Electron drift velocities in He-F_2 mixtures as a function of E/N. All data points are present results. The fulldrawn curve marked N is due to Nighan (Ref. 15) and pertains to a calculation in 0.3% F_2 + 99.7% He. The curve marked M & C depicts measurements in pure helium by Milloy and Crompton (Ref. 8).

we integrate the current pulse and then observe a linearly rising voltage pulse at the output of the amplifier in Fig. 1. In the presence of attachment, the integrated electron current rises as $[1 - \exp(-\eta v_d t)]$. It exhibits a discontinuity at $t = T_-$ since $v_d >> v_n$ (negative ion drift velocity), allowing the electron drift velocity to be determined from $v_d = d/T_-$. (d is the electrode separation, which may be changed from 0.3 - 3 cm.)

First, to check out the method, we have measured the electron drift velocity in pure helium (Fig. 2) and compared our results with those of Milloy and Crompton[8]. In the overlapping E/N region there is agreement within the experimental errors. (Milloy and Crompton claim an accuracy of better than $\pm$1.5%; the present accuracy is $\pm$10%.)

Second, in a gas mixture consisting of 1% F_2 + 99% He, there is a definite increase in the drift velocity, as shown by the open squares. (It was practically impossible to get any data at higher fluorine concentrations for the reason that most of the electrons would be lost due to attachment taking place close to the cathode.) At a lower fluorine concentration(0.2%), there is a discernable difference in the measured v_d - values as compared to our own results in pure He. At a concentration of 0.1% the observed v_d - values fall within 10% of the pure He data. (Our experimental errors in v_d are increased to 12-15% when F_2 is admitted to the system).

The systematic trend showing an increase in electron drift velocity with increasing fluorine concentration has been supported by Nighan[9] in a recent calculation for a 0.3% F_2 + 99.7% He mixture (See curve marked N in Fig. 2). His model includes vibrational excitation, dissociative attachment, and dissociation of the F_2 molecule, and the results are consistent with the experimental data.

The increase in drift velocity that results from the addition of F_2 is similar to that which has been observed previously when a molecular gas has been added to,[10,11] or has been present in[12,13], a monatomic gas. The increased mobility is due to the introduction of numerous low energy inelastic energy exchange processes that are absent in a pure monatomic gas. The enhanced energy exchange between the electron swarm and the gas reduces the average energy of the electrons in the swarm and hence increases the mobility.

In the case of fluorine, there is an additional process which acts to decrease the mobility. Since the attachment rate for dissociative attachment in F_2 decreases with increasing electron energy[5,14], the low energy fraction of the swarm tends to be depleted preferentially. The remaining electrons will thus have a mean energy that is higher than would otherwise be the case with a consequent reduction in their mobility. We are unable to assess the significance of this latter process, which might be termed *attachment heating*, due to lack of cross section data, but it is evident that it is less significant than the former process.

Our observations confirm that, in rare gas - fluorine lasers, the electron velocity distribution, and consequently the electron drift velocity, is strongly dependent on fluorine concentration. However, for a fluorine concentration of 0.1% our measurements show no detectable effect on the electron drift velocity, thus supporting recent model calculations.[2-4,15]

We want to thank Professor R.W. Crompton for comments on the manuscript and Dr. W.L. Nighan for the results of his unpublished calculation.

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*Work supported in part by ARPA/ONR under contract No. N00014-77-C-0100 and by Los Alamos Scientific Laboratory under Order No. N47-86053-1.

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AUXILIARY DISCHARGE ELECTRON SOURCE

A number of problems have been encountered with the pulsed xenon ion laser photoelectron source used in the present experiments. The power output of the laser diminished with time, possibly due to contamination of the laser mirrors. Further problems with the photoelectron source were that the photoelectric conversion efficiency of the aluminum cathode was considerably reduced at the higher concentrations of fluorine used in this work. The source was also only able to be used over a relatively low pressure range, being from approximately 5 torr to 20 torr.

For these reasons, it was decided to modify the system to enable a pulsed gas discharge source to be used as the electron source while waiting for the new ArF laser to be completed. A schematic of the system is shown in Fig. 1. A 1 mm hole was drilled in the cathode and a molybdenum pin source placed immediately behind the hole. The apparatus was modified to allow the gap separation between the pin source and the electrode to be varied. This allowed the optimum pulse shape to be obtained at each operating pressure.

A typical discharge pulse is shown in Fig. 2, which indicates that pulse widths of less than 10 nsecs half width are possible. The source has been shown to work over the pressure range of 100 to 1000 torr, which is considerably nearer the operating pressure range of present rare gas-halide lasers. It shows great promise for future electron and ion mobility experiments.

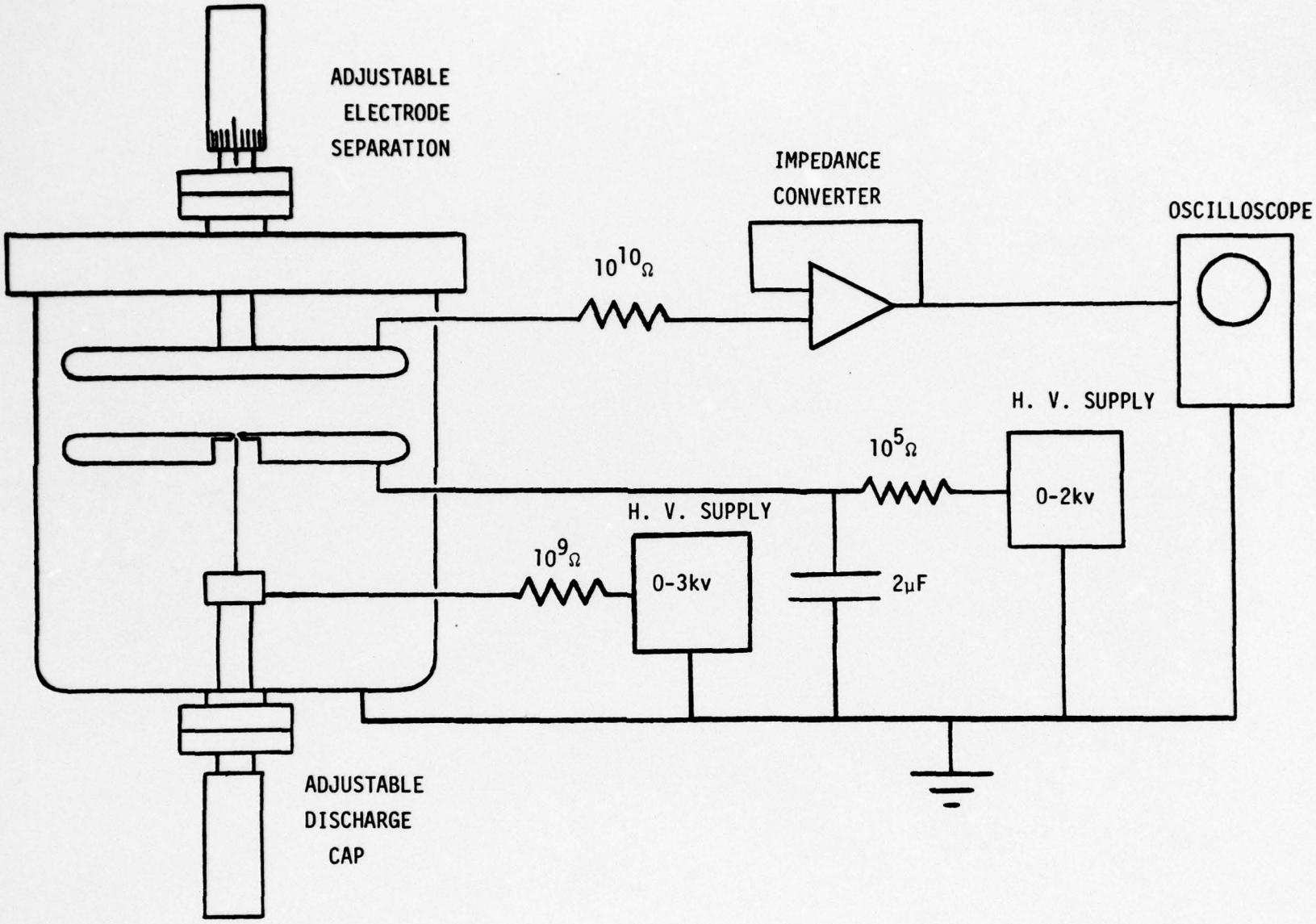
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Figure 1

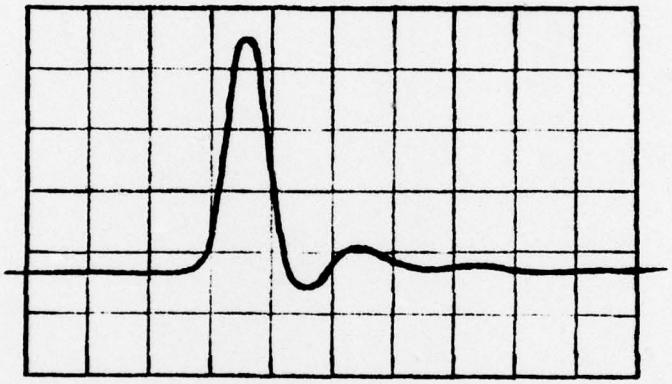

Figure 2. Tracing of photograph showing the discharge pulse.

Horizontal scale = 10 nsec/cm

Vertical scale = 5 volts/cm

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8. CONTRACT OR GRANT NUMBER(s)
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18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)
F_2 dissociative attachment, rare gas-halide laser kinetics, electron drift velocities.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)
We have made an absolute determination of the electron attachment coefficient $\eta(cm^{-1})$ in helium containing 0.1-1% fluorine covering an E/N range from 3-17 Td. At an estimated average energy of 5eV we find a rate coefficient equal to $(7.5 \pm 1.5) \times 10^{-10}$ cm^3/sec. Using a simple time-of-flight technique, we have measured the electron drfft velocity in helium with 0.1-1% fluorine additive. Our results in a 0.1% mixture are in close agreement with data in pure helium. The measured drift velocity increases with increasing fluorine concentration.