A NOVEL TECHNIQUE FOR THE MEASUREMENT OF

PHOTOENHANCED ELECTRON ATTACHMENT: IMPLICATIONS

FOR AN OPTICALLY–CONTROLLED DIFFUSE DISCHARGE

OPENING SWITCH[1]

LAL A. PINNADUWAGE

LOUCAS G. CHRISTOPHOROU

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STORAGE INDUCTOR L_S

SWITCH S_2

$t = 0$

E-BEAM WINDOW

DIFFUSE DISCHARGE OPENING SWITCH S_1

LOAD Z_L

CURRENT SOURCE

E-BEAM SOURCE

LASER

LASER WINDOW

ELECTRODES

CURRENT I_0

V_1

LASER PULSE

2

3

TO
DETECTION
CIRCUIT

$|V_1| > |V_2|$

V_2

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DATA ANALYSIS

NUMBER DENSITY OF THE NEGATIVE IONS,

$$N_I = \int_0^{\tau_L} N_e(t) \, N^*(t) \, k_a \, dt$$

NUMBER DENSITY OF ALL NEGATIVE CHARGES,

$$N_T = N_e(t = \tau_L) = N_E + N_I$$

MEASURED VOLTAGE RATIO,

$$R_v = \frac{V_I}{V_T} = \frac{N_I}{N_T} = R_v(k_a, I, N_a, \text{other})$$

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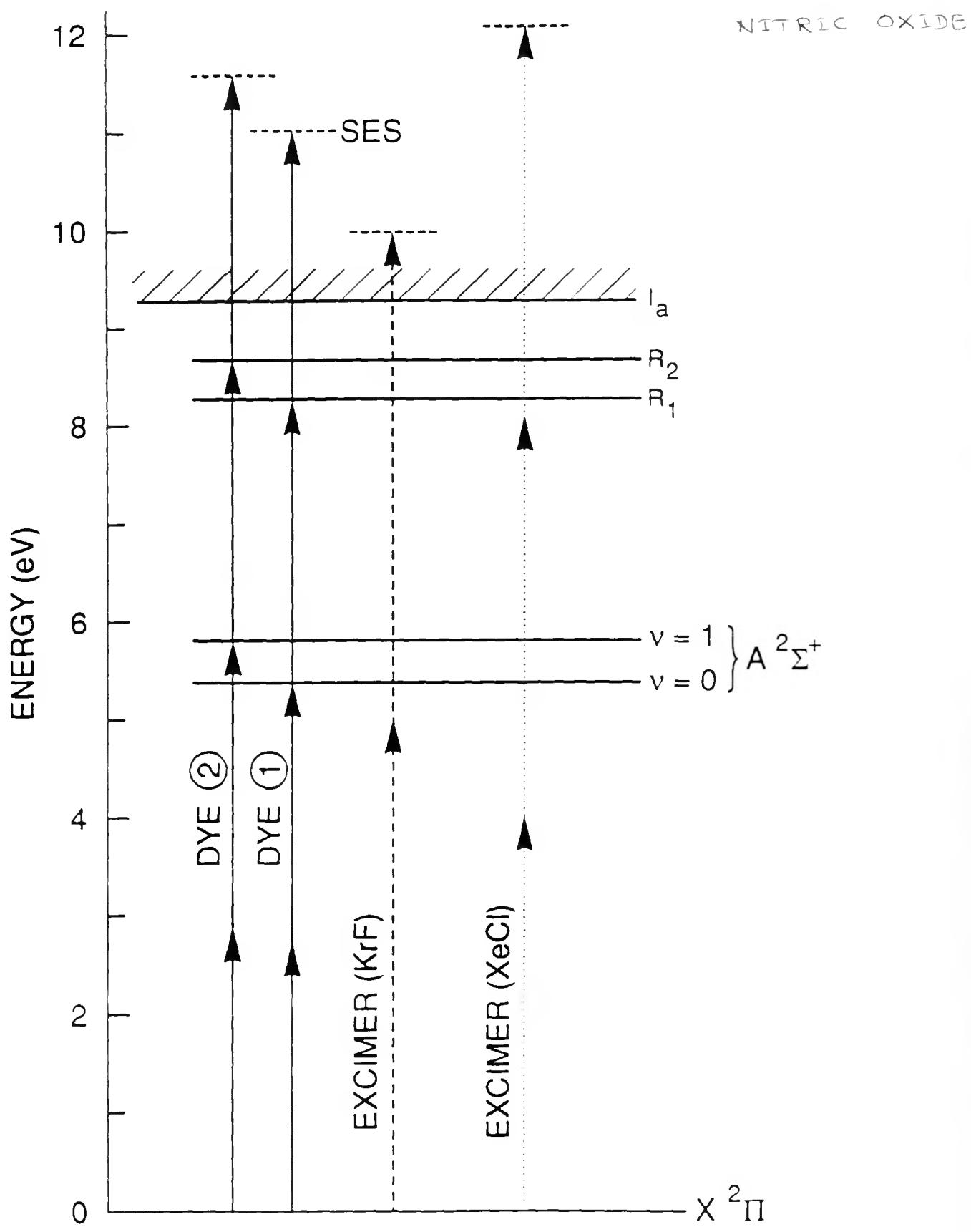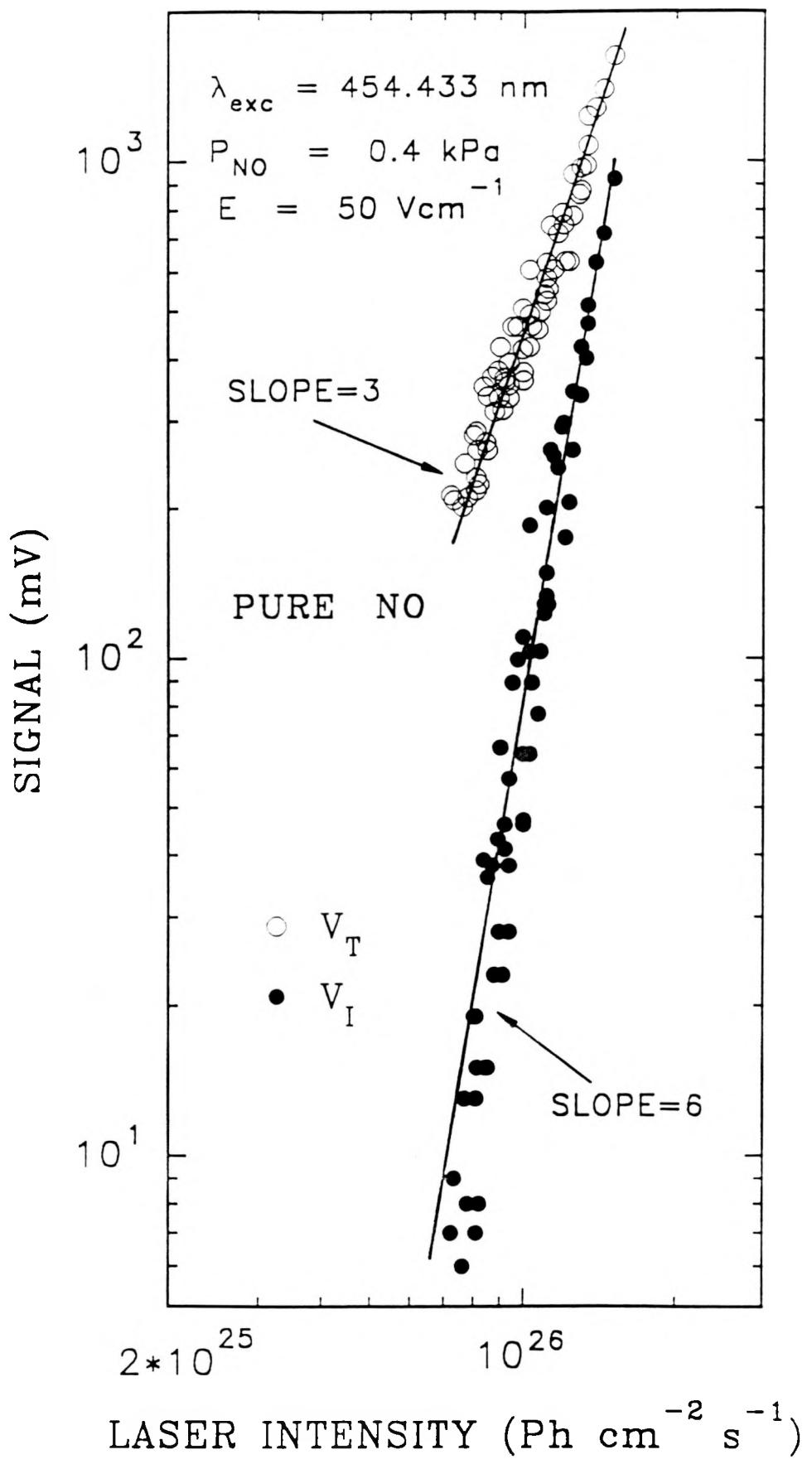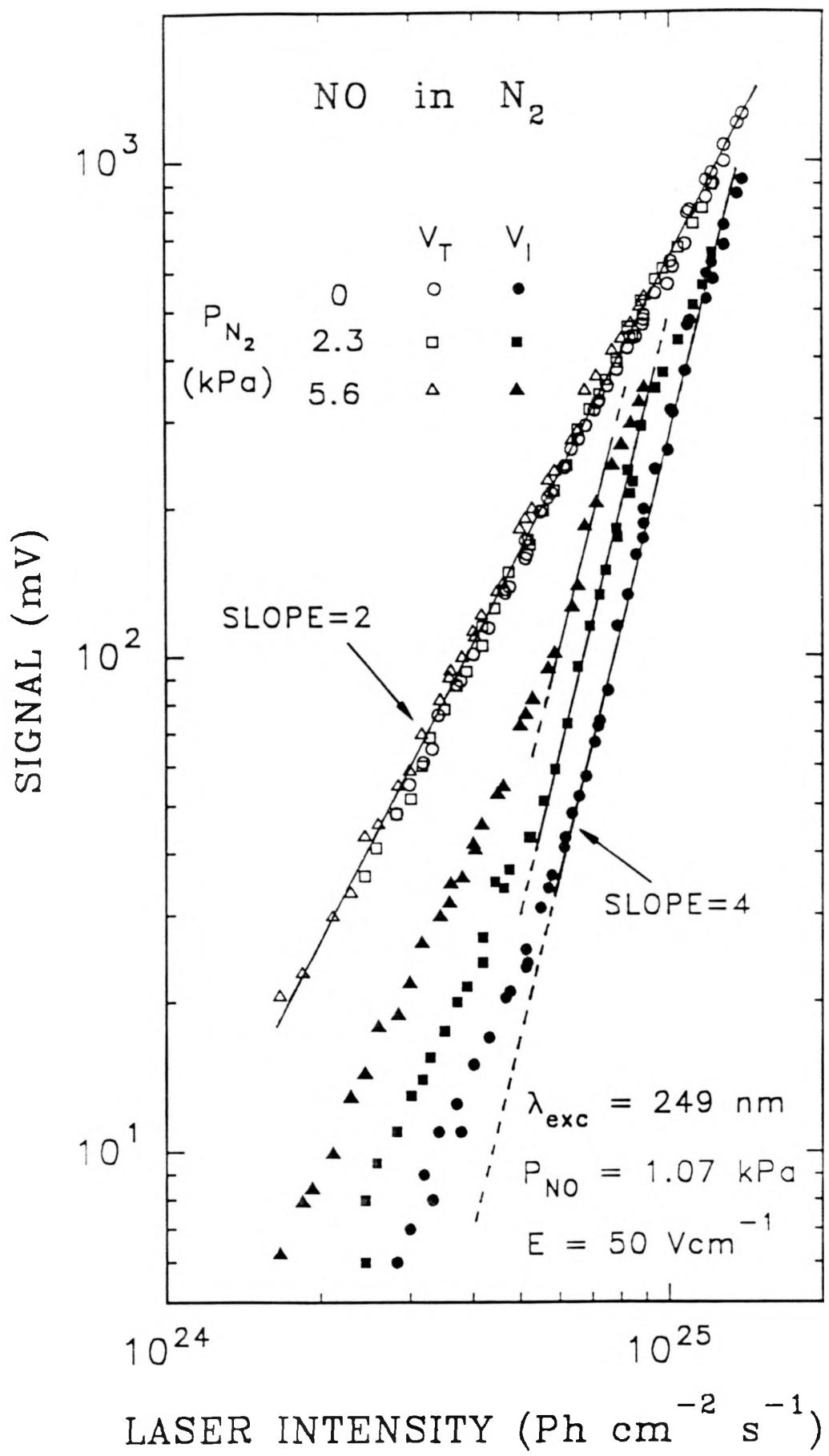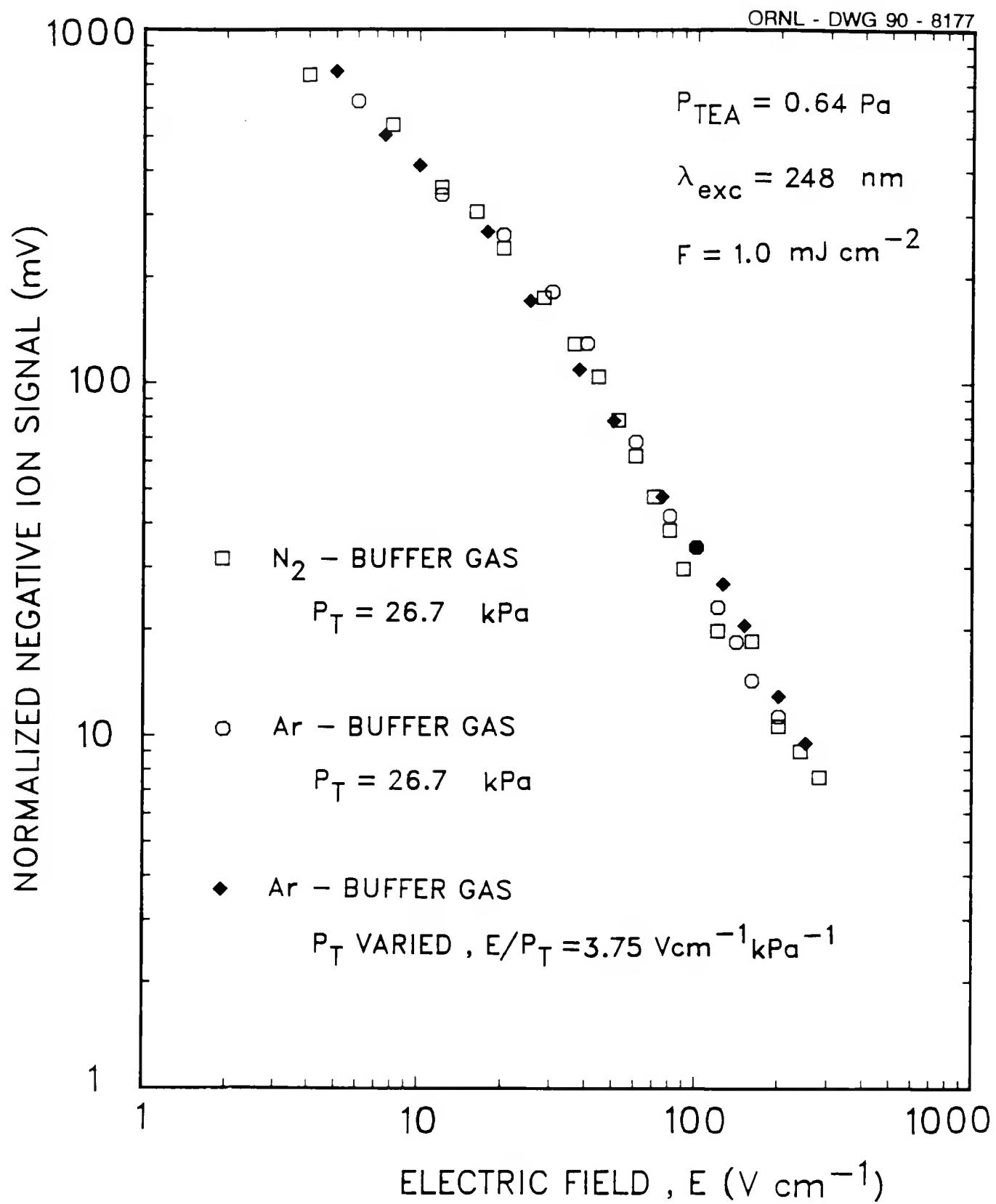
SUPEREXCITED STATES OF NO

$$NO^{**} + e \longrightarrow NO^{**-} \longrightarrow N + O^-$$

$$\tau_a k_a \sim 10^{-9} \text{ cm}^3 \quad @ \; \lambda_{exc} = 454.433$$

$$\tau_s < 10^{-8} \text{ s} \Longrightarrow k_a > 10^{-1} \text{ cm}^3 \text{ s}^{-1}$$

GROUND STATE NO

$$k_a < 10^{-13} \text{ cm}^3 \text{ s}^{-1}$$

$$\text{ENHANCEMENT} > 10^{12}$$

IMPLICATIONS FOR AN OPENING SWITCH

1. PROVIDES REALISTIC ELECTRON ATTACHMENT DATA

2. SWITCH OPENING TIME α ELECTRON ATTACHMENT TIME

$$= \frac{1}{N^* k_a}$$

$N^* > 10^{12} \text{ cm}^{-3}, \; k_a > 10^{-3} \text{ cm}^3 \text{ s}^{-1}$

$\Longrightarrow$ SUBNANOSECOND OPENING TIMES

3. SWITCH REPETITION RATE $\alpha \; \dfrac{1}{\text{LIFETIME}}$ OF EXCITED SPECIES

LIFETIME OF SES: $10^{-12} \text{ s} - 10^{-8} \text{ s}$

$\Longrightarrow$ HIGH REPETITION RATES

A NOVEL TECHNIQUE FOR THE MEASUREMENT OF PHOTOENHANCED ELECTRON ATTACHMENT: IMPLICATIONS FOR AN OPTICALLY—CONTROLLED DIFFUSE DISCHARGE OPENING SWITCH[1]

Lal A. Pinnaduwage and Loucas G. Christophorou

Atomic, Molecular, and High Voltage Physics Group
Health and Safety Research Division
Oak Ridge National Laboratory
P. O. Box 2008
Oak Ridge, Tennessee 37831—6122

and

Department of Physics, The University of Tennessee
Knoxville, Tennessee 37996

Abstract

Enhanced electron attachment to transient electron attaching species, such as electronically—excited molecules produced via laser irradiation, could be employed to switch conduction/insulation properties of a gaseous medium and has potential applications in diffuse—discharge opening switches. In this paper, we discuss a new technique that is capable of measurement of enhanced electron attachment to very short—lived (lifetime $< 10^{-8}$ s) electron attaching species. We also report measurements carried out using this technique on superexcited states (electronically—excited states lying above the first ionization threshold) of triethylamine and nitric oxide; these exhibit several orders of magnitude enhancement in electron attachment compared to the corresponding ground electronic states. Implications of such measurements for an optically—controlled diffuse discharge opening switch are indicated.

Introduction

The ability of a gaseous medium to switch large externally—sustained currents appears to be well—suited for fast, high—power, repetitive switching. Many studies have been conducted to understand and to develop diffuse gas discharge opening and closing switches for pulsed power applications [1—3]. The diffuse discharge opening switch in particular, may play a significant role in pulsed power technology employing inductive energy storage [2]. Diffuse discharge switches for inductive storage are characterized by two distinct stages: (i) the switch conduction stage where the storage inductor is being charged by the current flowing through the switch and, (ii) the non—conducting (transferring) stage, where the current through the switch is turned off and the stored energy in the inductor is transferred to the load. To optimize switch characteristics, the electrons supplied by the external source (e—beam or laser) must remain free in the conduction stage, but must be removed as fast as possible once the external source is turned off, i.e., the transition from conducting stage to the non—conducting stage must be rapid. To obtain fast opening of the switch after the external source is turned off, electrons in the switch gap must be removed by attachment since diffusion and recombination processes are too slow [2]. However, since electron attachment during the conducting stage must be low, what is needed is the rapid enhancement of electron attachment properties of the switch medium concurrently with the turning off of the external electron source. *This can be accomplished by optically—enhanced electron attachment.*

Among several possible laser—controlled mechanisms [4], optically—enhanced electron attachment to short—lived (lifetime $< 10^{-8}$ s) electronically—excited states appears to be particularly attractive since the switch could be made to operate at high repetition rates. However, the short lifetime poses problems for basic studies since electron attachment measurements have to be made during this short time. We recently developed [5] a new technique for carrying out such measurements. The details of this technique, together with examples of measurements carried out using that technique on electron attachment to superexcited states of triethylamine and nitric oxide, are discussed below.

The Technique

The basic idea behind this technique is to produce the attaching electrons volumetrically over the laser—irradiated region (via multiphoton ionization) concomitantly with the production of excited species by a single laser pulse, see Fig. 1. Since the excited species and the electrons are produced in close proximity, it is possible for electron attachment to the excited species to occur even when the lifetime of the excited species is $< 10^{-8}$ s. The attaching electrons may be produced by photoionization of the same gas that produces the electron attaching species, or a suitable additive gas.


Fig. 1. A schematic diagram of the experimental apparatus.

·Since positive ions are also produced via photo—ionization, an inherent requirement of this method is the ability to separate and detect negative charges (electrons and negative ions) unambiguously. As shown in Fig. 1, this is accomplished by separating the detection region (located between electrodes 2 and 3) from the interaction region (located between electrodes 1 and 2) via a three—electrode arrangement. Charge transmission between the two regions is through a fine grid which also electrically shields the two regions from each other. A laser pulse propagating parallel to the electrodes in the interaction region produces photoexcitation and photoionization. The same electric field is maintained in both regions and — depending on the direction of the field — either negative or positive charges produced in the interaction region are extracted into the detection region.

A pulsed—Townsend technique [6] is employed to record the voltage signal induced by the motion of the charged particles in the detection region located between electrodes 2 and 3 (Fig. 1). The signals due to the negative ions and the unattached electrons can be distinguished due to the difference in their (drift) velocities. This is accomplished automatically if the attaching gas pressure exceeds ~ 0.1 kPa; for smaller attaching gas pressures, a "buffer" gas (such as N_2 or Ar) can be added to slow down the ions compared to electrons.

Consider an electron attaching excited species of number density N^* produced by a laser pulse of duration τ_L. The electron attachment time, τ_a, is given by,

$$\tau_a = \frac{1}{k_a N^*} \tag{1}$$

where, k_a is the electron attachment rate constant of the excited species. Then, for electron attachment to occur,

$$\tau_a < \max\left\{\tau, \tau_L\right\} \tag{2}$$

where, τ is the lifetime of the excited species. Electron attachment has to occur prior to the decay of the excited species; for $\tau_L > \tau$, this upper limit is dictated by the duration of the laser pulse, since excited species are continually being produced within the duration of the laser pulse.

For the excimer lasers that were employed in the present studies, $\tau_L \sim 10^{-8}$ s and the τ for electronically—excited singlet states of molecules, is normally $\lesssim 10^{-8}$ s. It follows, then, that in our studies involving electronically—excited states of molecules, τ_a must be $< 10^{-8}$ s in order for electron attachment to occur. In the present studies, the attaching electrons are produced concomitantly with the production of the excited species; therefore, the energy of the attaching electron is close to that imparted on it upon laser photoionization [5].

The number density of negative ions formed within the laser irradiated region is given by,

$$N_I = \int_0^{T_L} N_e(t)\, N^*(t)\, k_a\, dt \tag{3}$$

where, N_e is the number density of electrons produced via photoionization. The number density of all negative charges, N_T, (sum of negative ions, N_I, and unattached electrons, N_E) would be the number density of electrons produced via photoionization if there was no electron attachment, viz.,

$$N_T = N_I + N_E = N_e(t = \tau_L, k_a = 0) \tag{4}$$

If we can disregard possible electron attachment to ground state molecules during the drift to and within the detection region (as is true for the two species discussed in this paper) then, the measured voltage ratio, R_v, is given by

$$R_v = \frac{V_I}{V_T} = \frac{N_I}{N_T} \tag{5}$$

The R_v of (5) is a function of the laser fluence (intensity), the partial pressure of the gas which yields the attaching species, and k_a, among others. Measurement of R_v versus laser fluence would yield the value of, or a quantity related to, k_a.

It should be mentioned that this technique can also be used to measure electron attachment to long—lived species (such as ground state, molecules). The data analysis for that case together with the measurements on electron attachment to ground state SF_6 (which was carried out to verify the technique) is described in detail in reference 5.

Experimental

Apparatus

A schematic diagram of the experimental apparatus is shown in Fig. 1. The gap distance in the interaction region located between the electrodes 1 and 2 was normally kept at 2 cm. Charge transmission to the detection region was through a fine grid (~90%, optical transparency) in electrode 2; the detection region gap separation was kept at ~ 0.3 cm. The electrode system was enclosed in a stainless—steel vacuum chamber which had a base pressure of ~ 1 x 10^{-6} Pa. The gas handling system was capable of introducing calibrated amounts of three different gases into the chamber.

The unfocused uniform laser pulse propagating horizontally through the interaction region was 0.1 − 0.2 cm away from the grid in electrode 2. In order to minimize losses of laser pulse energy via absorption by the gas(es) in the chamber two "light pipes" were employed as shown in Fig. 1. The transmitted pulse energy was measured by a Molectron J—25 pulse energy meter and was corrected for window losses and losses inside the chamber due to gas absorption to estimate the value at the middle of the interaction region.

Three laser sources were employed: (i) a Lambda Physik FL2002E dye laser pumped by a Lumonics 510 Excimer laser operating at the XeCl (308 nm) line; (ii) a Lumonics TE—860—4 excimer laser operating at KrF (248 nm) and ArF (193 nm) lines; (iii) Lumonics 510 excimer laser operating at XeCl and KrCl (222 nm) lines. All three laser outputs had pulse durations (FWHM) of ~ 10 ns. The dye laser was equipped with an intracavity etalon and had a bandwidth of ~ 0.04 cm^{-1}.

Results for Nitric Oxide

A schematic energy level diagram of NO and excitation schemes for the main laser lines employed are shown in Fig. 2. The location of the superexcited states (SES) reached in the experiments (see Fig. 2) varied from ~ 0.7 eV at the KrF line to ~ 2.9 eV at the XeCl line. The attaching electrons were produced via photoionization which occurs concomitantly with the excitation of SES. It has been shown that in the case of dye laser radiation, abundant amounts of near—zero—energy electrons are produced [7] via photoionization. In the case of KrF and XeCl lines no near—zero—energy electrons are produced [7] and the electrons are expected to have energies of ~ 0.7 eV and ~ 2.9 eV respectively. As we see below the energy of the attaching electron plays a crucial role in the electron attachment process.
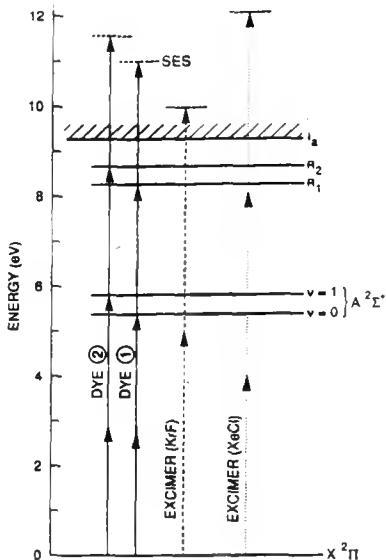
Fig. 2. A schematic energy level diagram of NO indicating the excitation schemes employed in the present study. At the 454.433 and 428.428 nm dye laser wavelengths (labelled as DYE 1 and DYE 2) the excitation was 2–photon resonant, respectively, with the v = 0 and v = 1 vibrational states of the A $^2\Sigma^+$ state; they were also resonant with higher–lying Rydberg states, denoted by R_1 and R_2. No real intermediate states were involved in the case of KrF and XeCl laser lines for which the transitions to the continuum involved 2 and 3 photons, respectively.

Data for the 454.433 nm dye laser radiation for pure NO are shown in Fig. 3; the total signal, V_T, (which is proportional to the initial number density of electrons produced via photoionization) and the negative ion signal, V_I, are plotted versus the laser intensity, I. The power dependence observed, $V_T \alpha I^3$ and $V_I \alpha I^6$, can be explained [8] in terms of the production of attaching electrons via photoionization and the subsequent attachment of those electrons to the superexcited states (SES).

A rate equation analysis [5] carried out for the data of Fig. 3 yielded $k_a \tau_s \sim 10^{-9}$ cm^3, where k_a and τ_s are the electron attachment rate constant and the lifetime of the SES involved. The SES lifetimes are not known, but are expected to be in the range $10^{-12} < \tau_s < 10^{-8}$ s; thus $k_a > 10^{-1}$ cm^3 s^{-1}, which is a very large value. The production of abundant amounts of near–zero–energy electrons [7], via photoionization of NO at the dye laser wavelengths involved, has been suggested to be responsible for such a large rate constant [8].

In the case of the KrF excimer laser radiation (see Fig. 2), the electrons produced via photoionization have energies of ~ 0.7 eV and no near–zero–energy electrons are expected [7]. Figure 4 shows the data for the KrF line for pure NO and NO/N$_2$ mixtures (N$_2$ is not affected by the laser radiation at the intensities involved and acts as a "buffer" gas [5,8]).
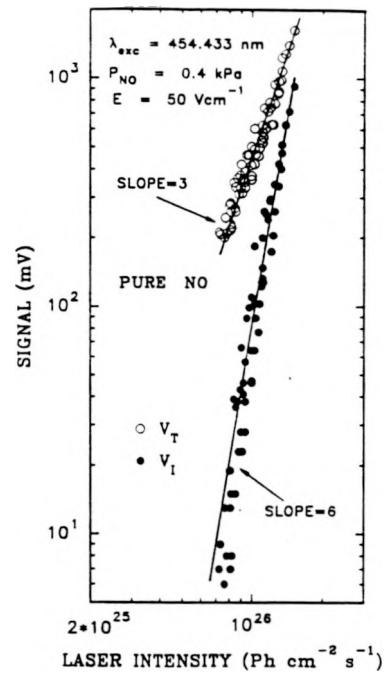
Fig. 3. Laser intensity, I, dependence of the measured total, V_T, and negative ion, V_I, signals for the dye laser line at 454.433 nm (see the text).
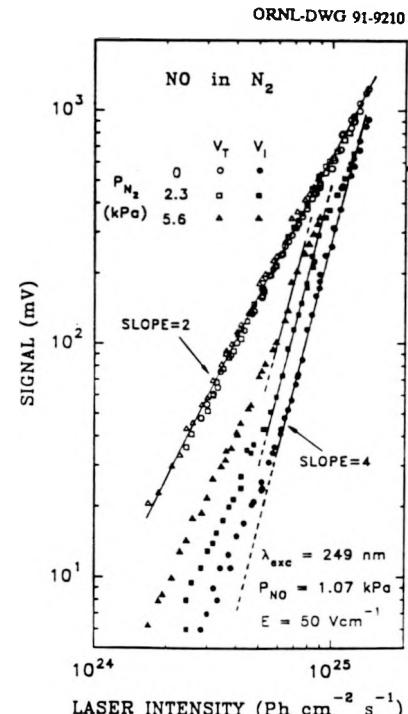
Fig. 4. Laser intensity, I, dependence of the measured total, V_T, and negative ion, V_I, signal for the KrF excimer laser line for pure NO and NO/N$_2$ mixtures (at the laser intensities involved, N$_2$ was not affected significantly by the laser radiation).

Compared to the data for the dye laser lines, the data for the KrF line display two major differences; (i) negative ion formation is enhanced in the presence of N_2, and (ii) below a certain laser intensity, I_c, (which depended on the NO pressure and is $\sim 1.5 \times 10^{26}$ ph cm^{-2} s^{-1} for the data of Fig. 4), V_I deviates from the I^4 dependence. These two observations are related and will be discussed below. First, we point out that the observed dependencies, $V_T \, \alpha \, I^2$ and $V_I \, \alpha \, I^4$ (for $I > I_c$), are consistent with the two-photon excitation to the continuum and the subsequent attachment of resulting electrons to concomitantly produced SES.

A rate equation analysis [5] for the data of Fig. 4 yielded $k_a \sim 10^{-3}$ cm^3 s^{-1} for the case of pure NO; k_a was $\sim$ 2.3 times larger for the NO/N_2 mixture at the N_2 pressure of 5.6 kPa (in both these cases data in the laser intensity ranges where $V_I \, \alpha \, I^4$ were used). These values are more than two orders of magnitude smaller than the value estimated for the dye laser radiation. This is understandable, since the attaching electrons in this case are expected to have energies of ~ 0.7 eV compared to the case of dye laser irradiation when an abundance of near–zero–energy electrons was available.

The enhancement in electron attachment in the presence of a buffer gas such as N_2 (Fig. 4) or Ar can be explained in terms of the slowing down of the attaching electrons via collisions with the buffer gas prior to attachment; at low laser intensities, the electron attachment time is larger and even in the case of pure NO, the electron can undergo such energy loss collisions with ground state NO prior to attachment; this explains the "additional enhancement" in electron attachment at low laser intensities for pure NO at the KrF line (Fig. 4).

The observations at the XeCl line were qualitatively similar to those for the KrF line. In all three cases (dye laser, KrF, XeCl) the negative ion yield decreased rapidly with the increasing applied electric field.

Results for Triethylamine (TEA)

A detailed description of the studies on electron attachment to the superexcited states of TEA is given in Ref. 5. Briefly, the excitation of TEA to the continuum was carried out via two–photon absorption at the KrF, KrCl (222 nm), and ArF (193 nm) excimer laser lines. Rate constants for electron attachment to these superexcited states were measured and were found to be of the same order of magnitude as for the case of NO.

In Fig. 5, we show the applied field dependence on negative ion formation for TEA in TEA/N_2 and TEA/Ar mixtures which is similar to the case of NO; the negative ion yield decreases rapidly with the increasing applied electric field. Figure 5 also illustrates that the negative ion formation is not affected by the presence of a buffer gas as in the case of NO irradiated with the dye laser radiation. Therefore it appears that abundant amounts of near–zero–energy electrons are produced when TEA is photoionized at these wavelengths.
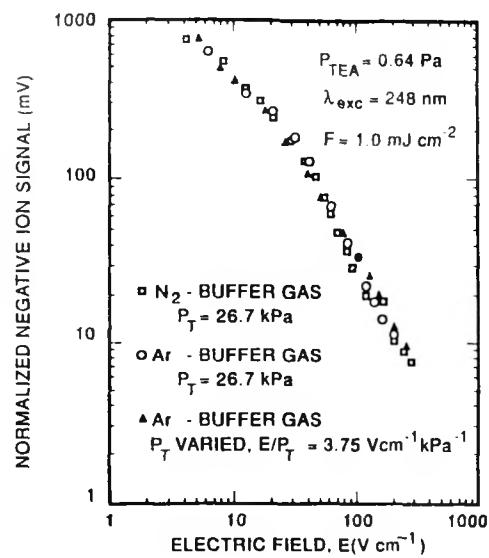

Fig. 5. Field dependence of the normalized negative ion signal obtained with Ar and N_2 buffer gases for KrF laser line (see the text).

## Implications for an Optically–Controlled Diffuse Discharge Opening Switch

In an optically–controlled diffuse discharge opening switch (OCDDOS), the fast opening of the switch is accomplished by producing strongly–electron–attaching species (via irradiation with a laser pulse) concomitantly with the turning off of the external e–beam. The time, t_r, taken for removal of the remaining electrons in the discharge, via electron attachment, will determine the switch opening time. Since optical excitation occurs within $\sim 10^{-15}$ s, which is much smaller than the electron attachment time, t_a, $t_r \sim t_a = \dfrac{1}{N^* k_a}$, where, N^*, and k_a are the number density and the electron attachment rate of the electron attaching excited species. The switch repetition rate depends on the relaxation time (lifetime) of the excited species. With excited species of short lifetimes, the switch can be made to operate at high repetition rates as far as electron attachment is concerned.

As in a OCDDOS, in the present technique the electron attaching excited species are produced via laser irradiation of the inter–electrode gap, and electron attachment can occur immediately after the formation of the excited species. Therefore, this technique can provide realistic basic electron attachment data for a OCDDOS. Furthermore, the two excited species discussed here (the superexcited states of NO and TEA) seem to have ideal properties for a OCDDOS, i.e., large electron attachment rate constants ($k_a > 10^{-3}$ cm^3 s^{-1}) and short lifetimes (lifetimes of the SES are expected to be in the range $10^{-12} - 10^{-8}$ s). Since $N^* > 10^{12}$ cm^{-3} can easily be obtained, switch opening times of $< 10^{-9}$ s can be achieved.

In both cases (SES of NO and TEA), the k_a decreased with the increase of, (i) the energy of the attaching electron and, (ii) the applied electric field. During switch opening, the electric field across the switch will increase rapidly and the energy of the electrons will increase as well. Therefore it is important to synchronize the turning off of the e–beam and the turning on of the laser pulse so as to "catch" the electrons during the very early stages of the switch opening. Addition of a buffer gas such as N_2 will help, since it can slow down the attaching electrons thereby enhancing electron attachment (see Fig. 4).

Finally, there is also a possibility that an external laser pulse may not be needed to produce the superexcited states. As the switch opens, the field across the gap increases and the electrons are accelerated to higher energies; thus such an electron can excite a gas molecule (losing its energy in the process) and can be captured concomitantly by the same excited molecule; viz.,

$$AB + e_f \longrightarrow AB^{**} + e_s \longrightarrow A + B^- \qquad (6)$$
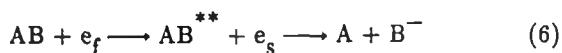
where, AB and AB^{**} denote ground and superexcited states of the molecule and e_f and e_s denote fast and slow electrons, respectively.

Conclusions

The technique described in this paper can be used to obtain electron attachment data on molecules suitable for an optically–controlled diffuse discharge opening switch. The two molecules discussed (NO and TEA) seem to fulfill many desired properties for such switches.

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