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Detection of Nitrogen Atoms in the $2s(2p)^3 3s\ {}^6S_{5/2}$ Metastable Autoionizing State

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Metastable nitrogen atoms, produced by electron-impact dissociation of N_2, have been detected in time-of-flight apparatuses located at Oregon State University and at University of California at Berkeley. Two different types of metastable atoms are detected; one type resembles Rydberg hydrogen atoms, while the other does not. The measured natural lifetime for the non-Rydberg atoms is 100 ± 25 μsec. Additional information regarding the identity of both types of metastable atoms is obtained by observing their quenching in a static electric field. The field-dependent decay constant for the Rydberg atoms is essentially a step function, and these atoms are entirely quenched by a field of 1.5 kV/cm. Both results are consistent with theory if the Rydberg atoms are in levels with principal quantum number greater than 25. The field-dependent decay constant for the non-Rydberg atoms varies quadratically with the strength of the applied electric field. In a field of 4 kV/cm applied along a 6-cm length of flight path, the value of this decay constant is 0.3×10^6 sec $^{-1}$. The quenching results, together with the lifetime measurement, are a strong indication that the non-Rydberg atoms detected in the time-of-flight spectra are nitrogen atoms in the $2s(2p)^3 3s\ {}^6S_{5/2}$ metastable autoionizing state.

I. INTRODUCTION

Although several measurements involving direct observation of neutral fragments produced by dissociation of simple molecules have been reported in detail[1-9] during the past six years, studies of the dissociation of N_2 have been reported only briefly.[10, 11] In both experiments on N_2, the nitrogen molecules were dissociated by electron impact, with electron energies variable from threshold to 200 eV. The energy threshold for the detection of neutral atoms in each experiment was approximately 25 eV, indicating dissociation into a ground-state atom and a metastable partner with several eV of internal excitation energy. The highly excited partner was identified as a nitrogen atom in a Rydberg state.

The purpose of the present paper is to describe new measurements of the dissociative excitation of N_2 by electron impact. These new results indicate strongly that a significant fraction of the excited neutral dissociation fragments are atoms in non-Rydberg states. It will also be shown that the detected fragments are not nitrogen atoms in the low-lying ^2D and ^2P metastable states; therefore, it is concluded that nitrogen atoms in the $2s(2p)^3 3s\ {}^6S_{5/2}$ metastable autoionizing state are being detected in addition to atoms in Rydberg states.

II. METHOD

The products of a molecular dissociation process share a kinetic energy which is substantially larger than the ambient mean thermal kinetic energy; therefore it is often possible to isolate, and to some extent, identify dissociation fragments using the time-of-flight (TOF) technique.[1-9] The simplest configuration of the TOF apparatus used in the present experiment is shown in Fig. 1. The slits serve to define a flight path between the electron-bombardment region and the detector (labeled CEM in Fig. 1), and allow differential pumping between the two regions. The gas pressure at the detector is usually at least a factor of 10 lower than it is at the electron gun. Also the gas pressure is sufficiently low so that the gas kinetic mean free path, for neutral ground-state particles, is greater than the distance from the electron gun to the detector.

For the apparatus shown in Fig. 1, a TOF measurement is made by bombarding N_2 gas in the electron-gun region with a short burst of electrons, and simultaneously starting the charging ramp of a time-to-amplitude converter (TAC). The TAC ramp is stopped by a pulse from the detector. If this STOP pulse is initiated by a particle originally produced or excited by the electron-gun pulse, then the particle's TOF is proportional to the amplitude of the corresponding output pulse from the TAC. Output pulses from the TAC are stored in a pulse-height analyzer, and a TOF spectrum is accumulated by regular repetition of the electron-gun pulse. It is noteworthy that, in order for this spectrum to represent the velocity distribution of particles produced at the electron gun, the total count rate must be

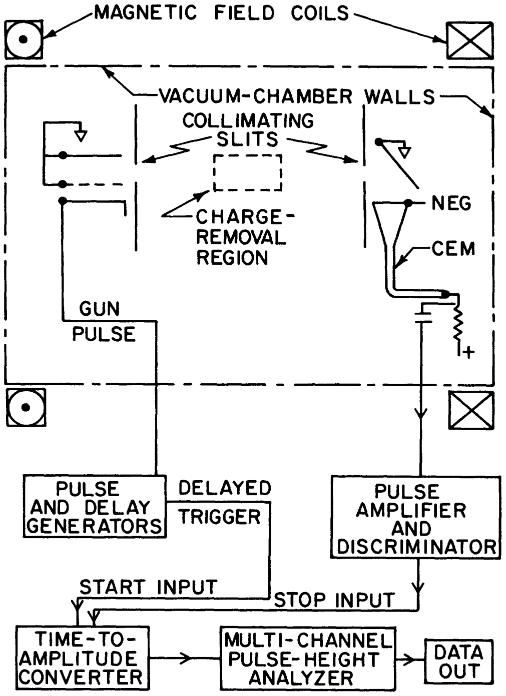

FIG. 1. Outline of the low-resolution time-of-flight apparatus.

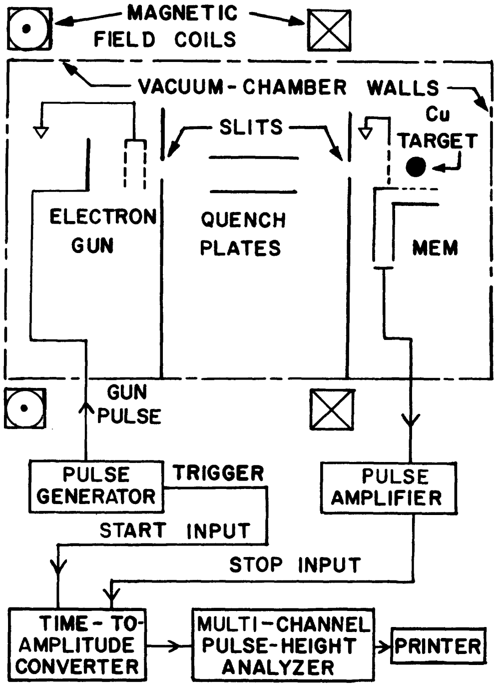

FIG. 2. Outline of the high-resolution time-of-flight apparatus.

so low that the probability for the occurrence of two or more detector pulses during one cycle of operation is negligible. The electron-gun repetition rate is typically 2000 $\sec^{-1}$, and the total count rate is always less than 200 $\sec^{-1}$. In the present experiment, slow metastable N_2 molecules require approximately 0.5 msec to reach the detector and therefore determine the maximum repetion rate.

A windowless electron multiplier used as a detector is sensitive to photons, neutral metastable atoms or molecules, and either positively or negatively charged particles. Using this type of detector biased against negatively charged particles, a typical TOF spectrum will include the following different kinds of particles arriving at the detector in order of increasing TOF:

(i) photons emitted from the excitation region during the electron-gun pulse;

(ii) positive ions, accelerated in the electron gun to several electron volts of kinetic energy during the electron-gun pulse, and accelerated again, at the end of the flight path, into the multiplier cathode;

(iii) metastable dissociation fragments;

(iv) metastable molecules of the parent gas.

Although there are often other events which appear in the TOF spectrum, the above example serves to illustrate that analysis by TOF may be used for particle identification. Additional information is obtained by varying the voltage of the electron-gun pulse to examine the threshold for the various spectra. For example, with N_2 as the parent gas, the spectra described above would appear with increasing electron-gun pulse height in the order (i), (iv), (ii), and (iii).

III. APPARATUS

A. Vacuum System

The electron gun and about three-fourths of the flight path are enclosed in a 2-l cylindrical stainless-steel chamber, which is pumped through a Sorbent A-filled pellet trap by a 2-in. stainless-steel oil diffusion pump (see Fig.1). The remaining fourth of the flight path and the detector are enclosed in a 1-l cylindrical stainless-steel chamber welded to the 2-l chamber and pumped by an 8-l/sec Vac-Ion pump. The foreline is trapped with a liquid-nitrogen trap. The base pressure of this system, which has not been baked, is about 2×10^{-7}Torr.

A large pressure differential is maintained between the electron-gun region and the detector during an experiment. For the apparatus configuration shown in Fig. 1, the differential pumping is achieved by totally enclosing the flight path between two collimating slits located at both ends of a 0.5-in.-diam stainless-steel tube. This tube is pumped out by the Vac-Ion pump at one end, and by the diffusion pump at the other end. Thus, the

N_2 target gas entering the electron gun also fills the 2-l chamber. For the configuration shown in Fig. 2, the flight path is not enclosed; instead the 2-l chamber is divided by a partition which separates the electron gun from the flight path. Also, the target gas enters the electron gun perpendicular to the flight path.

For both configurations, the pressure of N_2 target gas at the electron gun can be varied from 5×10^{-6} to 2×10^{-4}Torr, while the pressure at the detector is usually at least a factor of 10 lower than at the electron gun. Research-grade nitrogen is used, and the gas handling system, including the pressure-reduction valve, is stainless steel, except for the gaskets.

B. Electron Gun

The electron gun consists of the cathode, control grid, and screen grid removed from a glass-enclosed 6AH6 receiving tube. This tube has a rectangular cathode, 3 by 10 mm, and planar grids. The separation between cathode and control grid is about 1 mm as is that between the two grids. Thus, the excitation region is quite small. Coils external to the vacuum system furnish a magnetic field of 10–100 G at the electron gun; this field provides additional confinement of the excitation region.

The electron gun is pulsed by applying a negative voltage pulse to the cathode, with both grids at electrical ground. Commercial pulse generators are used to provide negative rectangular voltage pulses variable in both height and width. Pulse heights from 20 to 200 V and widths from 0.4 to 8.0 μsec are used in the present experiment.

C. Detectors

The detector depicted in Fig. 1 (labeled CEM) is a continuous dynode, windowless electron multiplier (Channeltron, Bendix model 450). The CEM is sensitive to photons, ions, metastables, energetic ground-state neutrals, and electrons. The entrance horn of the CEM is negatively biased to discriminate against negative ions while the position of the CEM is arranged to avoid detection of photons, emitted either at the electron gun or along the flight path. Thus, as indicated in Fig. 1, the CEM is "hidden from view" from the electron gun and most of the flight path; therefore, it is sensitive primarily to events occurring near the end of the flight path.

It is also desirable to prevent the detection of any positive ions produced in the electron gun. This is achieved by applying appropriate dc voltages to a pair of small parallel plates located midway between the electron gun and the detector. These plates are labeled CHARGE-REMOVAL REGION in Fig. 1.

The tilted straight line at the end of the flight path in Fig. 1 represents a flat 1-cm square stainless-steel plate which is placed in front of the CEM horn, at an angle of about 45° with respect to the center line of the flight path. This plate serves to effectively terminate the flight path for photons, which are mostly absorbed by the plate, and for metastable neutral atoms and molecules, which are quenched at the plate. Both photons and metastables can eject electrons from the plate, and sometimes the metastables can also be ionized at or near the surface.[12,13] If the CEM horn is positively biased, then photoelectrons and Auger electrons will be detected. However, with the CEM biased sufficiently negative, positive ions created either by surface ionization [12,13] or by electric field ionization[12,14,15] are detected.

In the present experiment, TOF spectra obtained with the CEM negatively biased are strictly attributable to positive-ion formation at the detector plate, whereas with a positive bias on the CEM the spectra appear to be contaminated by electrons created in other parts of the apparatus, e.g., at or near the ion removal region. Thus, all TOF spectra were obtained with the CEM biased at –1 kV.

Later, it became desirable to improve the resolution of the apparatus, i.e., to reduce a TOF "linewidth" due to the finite size of the excitation and detection regions. Therefore, the CEM was replaced with a small magnetic electron multiplier (Bendix model M308, labeled MEM in Fig. 2), and the stainless-steel plate was replaced by a short length of No. 12 copper wire (see Fig. 2). The entrance grid of the MEM is negatively biased at –1.6 kV so that the MEM also detects only positive ions, instead of Auger electrons.

IV. TOF SPECTRA

A TOF spectrum obtained using either configuration of the apparatus should represent primarily the flight of metastable atoms or molecules through a distance of about 15 cm. (In Fig. 1, the mean flight path is 14.7 cm and, in Fig. 2, it is 15.3 cm.) A typical TOF spectrum for nitrogen atoms, obtained with the apparatus of Fig. 1, is shown in Fig. 3. There, the most probable TOF is 22 μsec, corresponding to a speed of 0.7×10^6 cm/sec. The magnitude of the most probable velocity is somewhat smaller than this since the velocity distribution $P(v)$ is related to the TOF spectrum $P(t)$ by

$$P(v) \propto v^{-2} P(t).$$

Nonetheless, all the velocities represented in Fig. 3 are large compared with the ambient mean thermal velocity of N_2 molecules. Therefore, it is concluded that the spectrum of Fig. 3 is due entirely to metastable N atoms produced by electron-impact dissociative excitation of N_2. This spectrum has an appearance threshold, in electron bombardment energy, of approximately 26 eV, and it is essentially unaltered in shape throughout the energy range 30–50 eV. Data showing the detailed dependence of a oxygen TOF spectrum upon electron bombardment energy have been published.[4,5]

Using the higher-resolution apparatus (Fig. 2) and bombardment energies of the order of 100 eV, more complex TOF spectra are obtained, as is indicated by the spectra shown in Fig. 4. There, the peak occurring at a TOF of 21 μsec probably corresponds to the single peak in Fig. 3; but the peak at 16 μsec is either not present or is unresolved in the lower-energy spectrum. It is this fast peak which contains a large number of nitrogen atoms in non-Rydberg levels, as is shown in Secs. V and VI.

V. LIFETIME MEASUREMENT

The TOF spectra shown in Figs. 3 and 4 most likely represent nitrogen atoms in Rydberg levels,[4,5,10] *a priori,* since the low-lying ^2D and ^2P metastable states of N are probably not detected in the present apparatus, and since the $2s(2p)^3 3s\ ^6S_{5/2}$ metastable autoionizing state of N has never been observed, although its existence has been discussed.[16–20] However, in the strictest sense, the TOF spectra just described can represent atoms in any or all of the metastable states of N; therefore, a lifetime measurement was used to provide additional specification. The natural lifetimes for the ^2D and ^2P levels of N are known [21]; they are 26 h and 12 sec, respectively. Atoms in Rydberg levels should exhibit a variety of lifetimes, [14,22] dependent upon excitation conditions and the effects of collisional quenching[23]; they range from 1 to 100 μsec.

The apparatus used for the lifetime measurement is located at the University of California, Berkeley, and has been described in detail previously.[24] This apparatus, although similar to the one described above, differs in that TOF spectra are accumulated by two detectors (instead of one) located at two different positions along the flight path. At the first detector, 68 cm from the electron gun, the flight path is intersected by a 60% transparent metallic mesh, and Auger

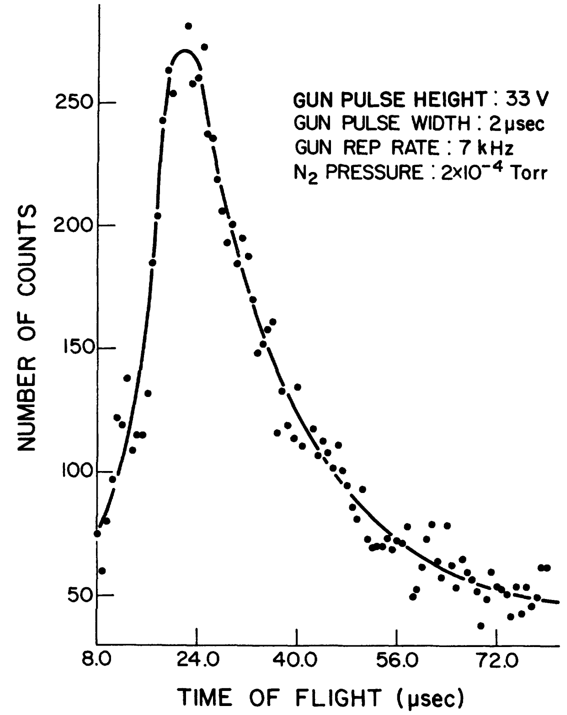

FIG. 3. Low-resolution time-of-flight spectrum. The solid line represents a qualitative interpretation of the data, which were accumulated during a 2-h period.

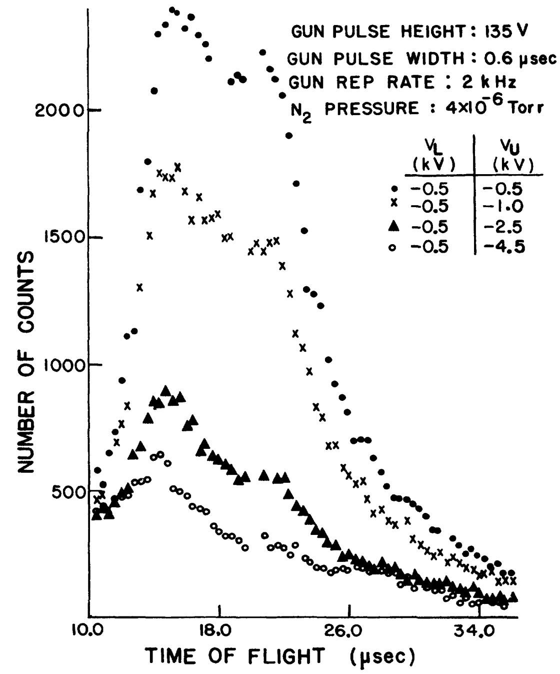

FIG. 4. High-resolution time-of-flight spectra. The four spectra were obtained using four different voltages applied to the electric-field quenching plates shown in Fig. 2, giving four different quenching fields. For example, the open circles in the figure represent an applied electric quenching field of 4 kV/cm.

electrons produced at the mesh are collected by an electron multiplier in essentially the manner indicated in Fig. 1. The flight path is then terminated by a solid metal plate and second electron multiplier, located 183 cm from the electron gun.

For atoms or molecules in relatively short-lived metastable states, the TOF spectra accumulated at the two detectors in the lifetime apparatus differ because of in-flight decay along the flight path between the two detectors.[24] If only one metastable species is present in the atomic beam, its lifetime can be obtained from a comparison of the two spectra. Specifically, the number of atoms, $N_2(t_2)$, detected at the second detector at time t_2, is related to the arrival of $N_1(t_1)$ atoms at the first detector, at the corresponding earlier time t_1 by the equation

$$N_2(t_2) = CN_1(t_1)e^{-(t_2-t_1)/\tau} ,$$

where C is a constant whose value depends upon the relative efficiencies of the two detectors,[24] and τ is the natural lifetime of the metastable state. For clarity it is noted that $t_2 - t_1 = (L_2 - L_1)/v$, where $L_2 - L_1$ is the separation of the two detectors and v is the magnitude of an initial velocity, given by $t_1 = L_1/v$, or by $t_2 = L_2/v$. The lifetime τ is obtained by evaluating, for the same velocity interval, the point-by-point ratio of the two spectra, and plotting

$$\ln(N_2/N_1) = \ln C - (t_2 - t_1)/\tau \qquad (1)$$

as a function of $(t_2 - t_1)$. The slope of the resulting straight line is $-1/\tau$.

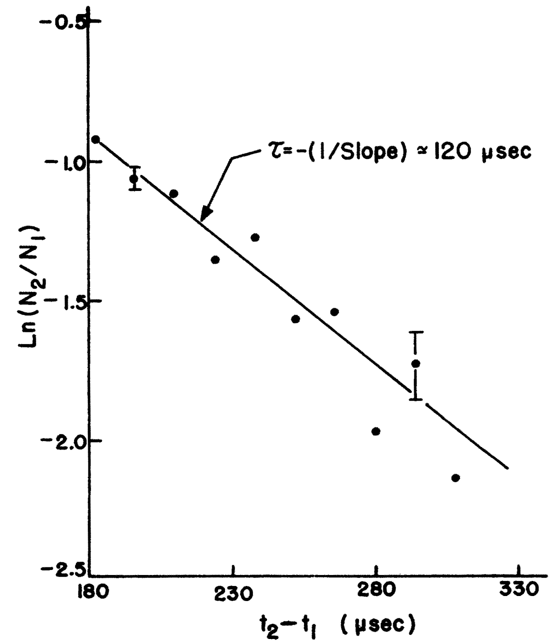

FIG. 5. Decay plot. The point-by-point ratio of the time-of-flight spectra accumulated at the two separate detectors used for a measurement of the lifetime τ. The straight line, whose slope is $-1/\tau$, is a qualitative interpretation of the data.

The data storage unit in the lifetime apparatus has a minimum channel width of 8.25 μsec; therefore, although the flight path is longer, the TOF resolving power of this apparatus is less than that of the apparatus shown in Figs. 1 and 2. Consequently, the TOF spectra for the metastable atoms produced by dissociative excitation of N_2 are not cleanly resolved from contaminant spectra.[25] However, comparison with the spectra taken at higher resolution (as in Fig. 4, for example) allows unambiguous identification.

A plot of Eq. (1), for nitrogen data, is shown in Fig. 5. Although the data are too scattered to allow specification of a single exponential decay, it is clear that the decay is dominated by components with lifetimes of the order of 100 μsec; therefore, the observed TOF spectra do not represent, to any significant extent, nitrogen atoms in the ^2D and ^2P metastable states. Further, because of the effects of collisional quenching in the excitation region, it is also highly unlikely that these spectra are due to nitrogen atoms in Rydberg levels. As has been discussed,[24] the background gas pressure along the flight path between detectors is always less than 10^{-8} Torr; however, in the excitation region, the operating pressure is typically 5×10^{-6} Torr or higher. Since the distance between the excitation region and the entrance aperture to the flight path is 10 cm, collisional quenching of Rydberg atoms can be extremely important is this region. For example, an s-state Rydberg atom with a lifetime of 100 μsec has a principal quantum number $n\simeq 35$; therefore, its gas kinetic mean free path in N_2 at 5×10^{-6} Torr is $\lambda\simeq 3\times10^{-2}$cm. Thus, if collisional-quenching cross sections are comparable with or larger than[23] geometrical cross sections, all Rydberg atoms with natural lifetimes greater than 1 μsec have a very low probability for escaping from the excitation region of the lifetime apparatus. On the other hand, metastable atoms with natural lifetimes shorter than 1 μsec must live for approximately 100 lifetimes to reach the nearest detector in this apparatus, and these atoms certainly cannot contribute significantly to the decay curve shown in Fig. 5.

VI. QUENCHING BY A STATIC ELECTRIC FIELD

A static electric field of magnitude E between the quench plates shown in Fig. 2 will decrease the lifetime of a metastable atom traveling along the flight path. The effect of this quenching field upon the TOF spectrum for a single metastable state i can be described as[26]

$$N_i(t,E) = N_{0i} \exp\{-[\gamma_{0i} + (D/L)\gamma_i(E)]t\}, \qquad (2)$$

where t is TOF, γ_{0i} is the natural decay constant for the ith state, $\gamma_i(E)$ is the field-dependent decay constant [i.e., $\gamma_i(0) = 0$], D is the length of the quench plates, L is the total length of the flight path, and N_{0i} is the number of atoms created in the ith state with a velocity of magnitude $v = L/t$. The TOF spectra obtained for two different values of quench field, E_1 and E_2, are related by the expression

$$N_i(t,E_2)/N_i(t,E_1) = \exp\{-(D/L)[\gamma_i(E_2) - \gamma_i(E_1)]t\}.$$

With $E_1 = 0$, a plot of $\ln R(t,E) = \ln[N_i(t,E)/N_i(t,0)]$, as a function of flight time, determines $\gamma_i(E)$; therefore, the functional dependence of $\gamma_i(E)$ can be determined by comparing TOF spectra for different values of the quench field. A determination of the dependence of $\gamma_i(E)$ upon E aids in the identification of metastable states, because, as is now discussed, a qualitative difference is expected between Rydberg and non-Rydberg levels.

A. Quenching Theory for Rydberg Levels

Classically, an applied electric field is expected to have a step-function effect upon the lifetime of an atom in a Rydberg level; if the electric field is strong enough to field ionize the atom, the lifetime is lowered drastically, otherwise it is essentially unaffected by the field. In this picture, the average time required for field ionization is of the same order of magnitude as the period of the unperturbed motion, which is orders of magnitude smaller than a typical TOF for any Rydberg atom observable with the present apparatus. For example, in the Rydberg levels with $n \simeq 100$ the period of a circular orbit is approximately 10^{-12} sec compared to a TOF of about 10^{-5} sec.

Similar results are obtained for hydrogen when the problem is treated quantum mechanically.[27] For example, the field ionization probability of the most readily ionized states in the $n = 25$ levels of H increases from 10^5 to 10^{11} sec $^{-1}$ as the strength of the electric field increases from 1.5 to 1.8 kV/cm. Therefore, for highly excited hydrogen levels, the field dependent decay constant $\gamma_i(E)$ in Eq. (2) can be expressed as

$$\gamma_i(E) \simeq \begin{cases} 0 & \text{if } E < E_{ci} \\ \infty & \text{if } E > E_{ci}, \end{cases}$$

where E_{ci} is a critical-electric-field strength, above which field ionization occrus. Thus, the TOF spectrum for hydrogen atoms in a single Rydberg state should either be unaffected by the electric field or completely quenched, *independent* of TOF.

The theory of field ionization of a hydrogen Rydberg is also applicable to the lowest $(2s)^2(2p)^2nl$ Rydberg levels in nitrogen if configuration mixing[28] is neglected. However, the most important contribution to configuration mixing can be taken into account in a fairly precise way, and as shown in the Appendix, field ionization in nitrogen is essentially unaffected by configuration mixing. Therefore, the field ionization of the Rydberg levels of nitrogen should be qualitatively the same as for the corresponding levels of hydrogen.

B. Quenching Theory for the $2s(2p)^3 3s\ {}^6S_{5/2}$ State of Nitrogen

The effect of an applied electric field upon the lifetime of the $2s(2p)^3 3s\ {}^6S_{5/2}$ metastable autoionizing state of N is expected to differ qualitatively from the results just described for Rydberg levels. This expectation is independent of the exact energy location of the ^6S state, but it can be assumed to be 17.2 eV above the ground state,[17] in the first ionization continuum. Then to first order, an electric field does not cause mixing of the $3s\,{}^6S$ state with the degenerate continuum states because of the selection rule forbidding two-electron jumps.[29] However, the electric field will cause a mixing of the $2s(2p)^3 3s$ and $2s(2p)^4$ configurations. Therefore, the field-dependent decay constant for the $3s\,{}^6S$ state is given approximately by[26]

$$\gamma_6(E) = \gamma_{06} + (D/L)k_6E^2,$$

where k_6 is a constant representing the details of the configuration mixing and γ_{06} is the natural decay constant.

C. Quenching Results

Electric field quenching of the nitrogen metastables is achieved by applying appropriate voltages to a pair of 3- by 6-cm parallel plates (see Fig. 2). The long dimension of the plates is parallel to the flight path. The separation d of the plates is approximately 1 cm. Typical voltages applied to the lower and upper field plates are listed in the columns labeled V_L and V_U in Fig. 4. It is necessary to use a nonsymmetric combination of V_L and V_U in order to minimize contamination[25] of the TOF spectra. The TOF data shown inFig. 4 represent quenching field strengths of 0.0, 0.5, 2.0, and 4.0 kV/cm. The lowest applied voltages do not provide a field-free flight path because a fringing field still exists. This fringing field is unimportant for

Rydberg atoms because a Rydberg atom is unaffected by the electric field until a critical field is reached, and then it is field ionized. However, for non-Rydberg atoms, the fringing field requires that the argument of the exponential function in Eq. (2) be replaced by the expression

$$-[\gamma_{0i}'(E_f)+(D'/L)\gamma_i(E)]t\ ,$$

where γ_{0i}' is independent of applied electric field, E_f is the fringing field, and $D' > D$.

The three logarithmic plots of $N(t,E)/N(t,E_f)$ for the data of Fig. 4 are shown in Fig. 6. The three straight lines in Fig. 6 represent an estimate of initial slope of the plots and correspond to the fastest atoms. The important point is that the electric field quenching is not entirely independent of TOF. Therefore, it is concluded that the observed TOF spectrum contains, in addition to a sizeable contribution from Rydberg atoms, at least one non-Rydberg component.

At the electron bombardment energy used to obtain the spectra shown in Fig. 4, it is probable that each metastable atomic state can originate from more than one dissociative molecular state. Consequently, there may be more than one initial velocity distribution and TOF spectrum for each atomic state. If the dissociative cross sections are different, there will be a distinct value of N_{0i} for each dissociative state, and the right-hand side of Eq. (2) becomes a sum of terms.

Unfortunately, the precision of the data shown in Fig. 6, representing about 40 h of collection time, is insufficient to warrant quantitative analysis. Therefore, it is regarded as fortunate that the fast atoms in the TOF spectrum appear to be dominated by a single velocity distribution representing a single metastable non-Rydberg state. (This result is consistent with the lifetime measurement discussed in Sec. V.) The slower peak appearing at 29 μsec in Fig. 6 is probably due to a second distinct velocity distribution for a non-Rydberg state. Figure 7 is a plot of the initial measured slopes $(D'/L)\gamma(E)$ of the straight lines shown in Fig. 6 as a function of E^2. Also shown is the same kind of plot for additional sets of data. These plots show that quenching of the fastest atoms varies as E^2 and indicate qualitative agreement with the theory described above for the quenching of the $2s(2p)^3 3s\ {}^6S_{5/2}$ metastable state of nitrogen. The differences in slopes and intercepts in Fig. 7 arise from different fringing fields for the various data sets.

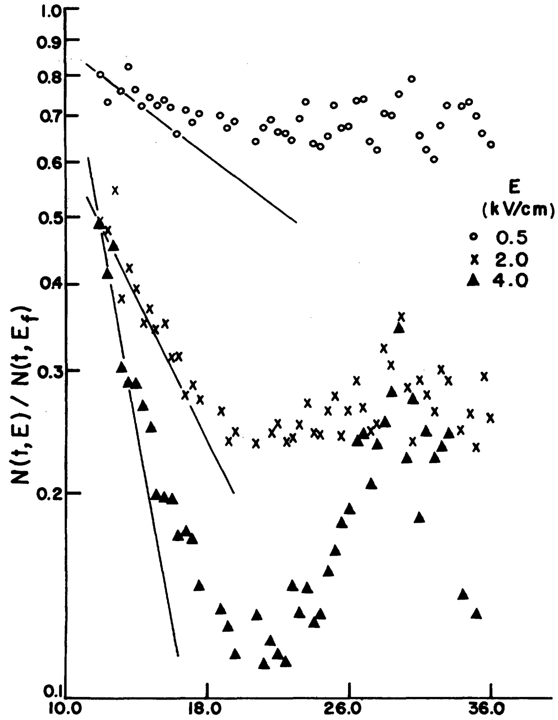

FIG. 6. Quench plots. The point-by-point ratios of the time-of-flight spectra shown in Fig. 4. The ratios plotted are those for each of the lower three spectra of Fig. 4 to the uppermost spectrum. For the uppermost spectrum, the quench plates are biased equally; therefore, only an unknown fringing field E_f remains. The straight lines in the figure represent an estimate of the initial slopes [see Eq. (2)].

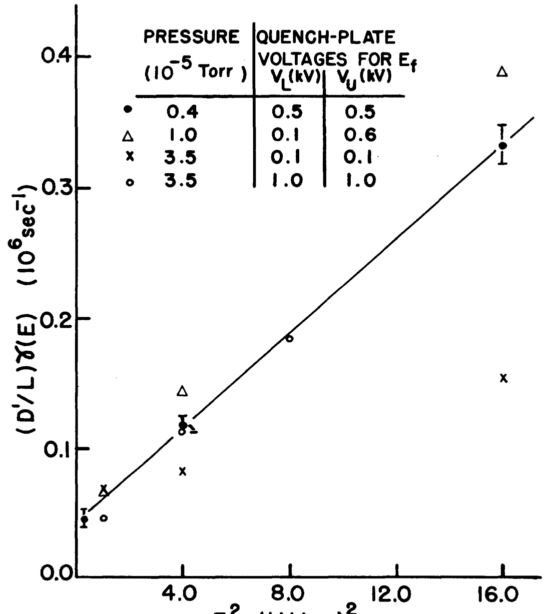

FIG. 7. Dependence of the measured decay constants on the square of the electric quenching field. The solid circles are obtained using Eq. (2) and the time-of-flight spectra of Fig. 6, while the open circles, open triangles, and crosses represent three additional sets of data, which have different residual fringing fields.

VII. CONCLUSIONS

If the lifetime measurement described here corresponds to the natural decay of nitrogen atoms in Rydberg levels, then the collisional-quenching cross sections for these atoms, in the parent N_2 gas, must be very much smaller than the geometrical cross sections. In contrast, previous work indicates the reverse to be true. The conclusion is that the measurement is the lifetime of the $2s(2p)^3 3s\,^6S_{5/2}$ metastable autoionizing state.

The electric-field-quenched TOF spectra correspond to at least two different types of metastable atomic states. For one of these the fractional quenching in a given electric field is independent of TOF. For the other, the quenched TOF spectrum decays approximately exponentially with increasing TOF, and the measured decay constant is proportional to the square of the electric field. Therefore, if the observed TOF spectra correspond only to nitrogen atoms in Rydberg levels, there are two distinct types of nitrogen Rydberg levels. For one of these, the electric field quenching is the same as for hydrogen; i.e., configuration mixing is unimportant. For the other type, the electric field quenching is due primarily to the effects of configuration mixing in violation of the general selection rules for configuration mixing.[29] In contrast, if the general selection rules for configuration mixing are *not* violated, then the field-dependent decay constant for the $2s(2p)^3 3s\,^6S_{5/2}$ state of N is predicted to vary quadratically with electric field strength, as pointed out in Sec. VI.

There is no reason to believe that Rydberg levels in atomic nitrogen are anomalous in either their collisional quenching or their electric field quenching. Therefore, it is concluded, instead, that in the range of bombardment energies where the production of Rydberg atoms by electron impact dissociation of N_2 is important, the production of nitrogen atoms in the $2s(2p)^3 3s\,^6S_{5/2}$ metastable autoionizing state is also important. Furthermore, the natural lifetime of this state is 100± 25 μsec.

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APPENDIX

For the $(2s)^2(2p)^2 nl$ Rydberg levels in nitrogen, the predominant contribution to configuration mixing should come from the four levels in the $2s(2p)^4$ configuration.[28,30] The lowest two levels (^4P, ^2P) lie approximately 10 eV above the ground state,[30] and the upper two (^2D, ^2S) are predicted to lie in the first ionization continuum.[31] Because of the selection rules for configuration mixing,[28] all of the odd-parity $(2s)^2(2p)^2 nl$ levels, and many of those with even parity do not mix, in first order, with the $2s(2p)^4$ configuration. The natural lifetimes for the remainder of these levels should be shortened by the effects of mixing, since the states in the $2s(2p)^4$ configuration decay by resonance transitions to the $(2s)^2(2p)^3$ configuration.

The situation just described is essentially unaltered by the addition to the problem of an applied electrostatic field, because, to lowest order, this perturbation does not mix states having different single-particle quantum numbers for more than one electron.[29] Therefore, the $(2s)^2(2p)^2 nl$ - $2s(2p)^4$ configuration mixing is altered very little by application of an electrostatic field. Consequently, the theory of field ionization of H should also apply fairly well to the $(2s)^2(2p)^2 nl$ Rydberg levels of N.

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Charged-Particle Scattering in the Presence of a Strong Electromagnetic Wave

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A simple and convenient approximation is obtained for the multiphoton energy-transfer processes which accompany the scattering of a charged particle by a scattering potential in the presence of a strong external electromagnetic field. It is expressed in terms of the differential elastic-scattering cross section combined with known functions, and is valid when the scattering potential is weak or when the wave frequency is small. A detailed form of the classical limit is obtained.

I. INTRODUCTION AND SUMMARY

When a charged particle, to be referred to henceforth as an electron, scatters in the presence of an electromagnetic wave, it may exchange energy with the electromagnetic field. Because, on the average, energy-absorbing encounters dominate energy-emitting encounters, the process is of central importance in the study of plasma heating by electromagnetic waves and in the study of gas breakdown. It has been extensively discussed both classically[1] and quantum mechanically,[2] the latter in the context of inverse bremsstrahlung and stimulated bremsstrahlung of single photons. When the electromagnetic field is strong, however (or when the frequency is low enough), many photons can be emitted or absorbed in a single scattering process. It is the purpose of this note to analyze these multiphoton processes and to relate them to the classical description.

The electromagnetic field will be approximated by a classical spatially homogeneous electric field throughout. The distinction between the classical and quantum treatment resides in the description of the asymptotic states of the electron before and after the collision. In the classical description, the electron follows a classical orbit, with oscillating velocity. Scattering processes are characterized by an instantaneous incident velocity determined by the phase (henceforth referred to as the scattering phase α) of the electric field at the scattering instant. While the scattering itself is assumed to be instantaneous and elastic, change in time-averaged electron energy occurs as the result of the change in electron direction. For a given scattering angle (referred to the time-averaged directions) the energy change is determined by the scattering phase α. In the quantum case, initial and final electron states are described by solutions of the Schrödinger equation. Energy