## Stochastic properties of Trichel-pulse corona: A non-Markovian random point process

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The stochastic properties of negative, point-to-plane, Trichel-pulse corona discharges are completely characterized in terms of a set of measured conditional and unconditional discharge pulseamplitude and pulse-time-separation distributions. The Trichel-pulse phenomenon is shown to be a clear example of a non-Markovian, marked random point process in which memory effects play an important role. Strong correlations are shown to exist among the amplitudes and time separations of successive discharge pulses that indicate how initiation and growth of a discharge pulse are affected by the presence of residual ion space charge and metastable species from previous pulses. The analysis required to assess consistency among the various measured probability distributions is discussed and used to interpret observed variations in distribution profiles. Because of the observed dependence of discharge pulse amplitude on both the amplitude of and time separation from the previous pulse, memory can propagate indefinitely back in time. The experimental limitations to verifying the extent of memory propagation are analyzed.

#### I. INTRODUCTION

The "regular" pulsating characteristic of negative point-to-plane corona discharges in air was first described in 1938 by Trichel.<sup>1</sup> The negative-corona discharge pulses that exhibit the regular relaxation oscillator behavior seen by Trichel have appropriately become known as "Trichel pulses." The phenomenon was subsequently verified by Weissler<sup>2</sup> and Loeb and co-workers<sup>3</sup> who further characterized the discharge behavior and showed that it generally occurs in all electronegative gases but not in pure nonelectronegative gases such as nitrogen and argon. In these early works the phenomenon was reported to occur over wide ranges of voltage above discharge onset and of point-to-plane electrode gap spacing. The occurrence of Trichel pulses is also not dependent on the type of electrode material. Later investigations<sup>4-8</sup> were undertaken to determine the range of conditions over which Trichel pulses appear and to measure the dependencies of their onset, amplitude, and repetition rate on such parameters as gas pressure, gap voltage, electrode spacing, and gas composition. There have also been reported measurements<sup>8-15</sup> of the discharge pulse shape as a function of different operating conditions.

Despite the abundance of phenomenological information acquired about Trichel-pulse discharges, the process has defied a complete theoretical description. Earlier explanations<sup>1,3,6,16,17</sup> of the physical mechanisms responsible for Trichel pulses were generally qualitative or semiquantitative at best. Only recently has there been some success by Morrow<sup>18,19</sup> in predicting the shapes of the *first* Trichel pulses under rather restricted conditions. Morrow's model, which is an extension of earlier work by Morrow and Lowke,<sup>20</sup> is reasonably complete in the sense that it includes all of the essential continuity equations for electron, ion, and radiation transport coupled with Poisson's equation for the electric field. It also allows for secondary-electron emission effects at the cathode.

The limitations of Morrow's model are to be found in the approximations and methods of solution required to make the calculations tractable. A onedimensional approximation which neglects diffusion is used for the charged-particle continuity equations. Because a three-dimensional treatment of Poisson's equation is required,<sup>21,22</sup> assumptions must be made about the space-charge distribution within the gap at any given time. The model has not been applied to discharge pulses that occur after the first pulse, which are often referred to in the literature as *regular* Trichel pulses. The difficulties to be encountered in using Morrow's model to describe the regular-pulse behavior should become evident from the work presented here.

It was recognized, even in the earliest investigations<sup>3,13</sup> of this phenomenon, that the amplitude of the first pulse is consistently higher than that of subsequent pulses. Cross and co-workers<sup>13</sup> have observed significant changes in pulse shape after the first pulse. A satisfactory explanation of differences between the first pulse and the subsequent regular pulses has not been given. It is evident, however, from the description of the discharge process given by Lama and Gallo,<sup>6</sup> that, after the first pulse, the gap contains moving space-charge clouds that can perturb the electric field and thereby influence the development of the regular pulses. The influence of moving space charge could, in fact, be similar to that which is known<sup>23,24</sup> to affect the recovery of Geiger counter tubes following the occurrence of a discharge pulse. Thus it can be argued from the evidence uncovered in previous investigations of Trichel pulses that the phenomenon is inherently a non-Markovian stochastic process in which "memory effects" play an important role. The memory effects are those associated with residuals such as ion space charge or metastable species from earlier pulses that influence the onset and development of subsequent pulses.

There is evidence from the recent experimental work of Steiner<sup>25</sup> that there are strong correlations among successive Trichel pulses. Steiner, however, made no attempt to give a physical explanation for these correlations. Malik and Al-rainy<sup>26</sup> measured the statistical variations of Trichel-pulse amplitudes, but failed to obtain enough information from their data to uncover correlations among successive pulses. In our previous preliminary work on this subject<sup>27-30</sup> we reported strong correlations among the amplitudes and time separations of successive Trichel pulses, but did not present a careful analysis of the possible physical mechanisms that could account for the observed correlations.

In the present work, a newly developed measurement technique<sup>27</sup> is used to obtain detailed information about the stochastic behavior of Trichel pulses in nitrogenoxygen and neon-oxygen gas mixtures. The technique allows direct measurement of a set of conditional pulseamplitude and pulse-time-separation distributions from which correlations among successive pulses can be quantitatively ascertained and related to memory effects in the discharge process. It is shown, for example, that pulse amplitude is strongly dependent on the time separation from the previous pulse and that this dependence is consistent with the influence of the moving negativeion space charge from the previous pulse in reducing the electric field at the cathode when the next discharge pulse develops. The measured conditional distributions are not independent and are shown to satisfy a set of required consistency relationships. The stochastic properties of Trichel pulses for the two gas mixtures are found to be distinctly different. This difference is attributed to the relative importance in each gas of ion space charge versus metastable states on discharge pulse initiation and growth.

The purpose of the present work is to exhibit and interpret examples of data that reveal general features of the stochastic behavior which are relevant over wide ranges of conditions. An understanding of this stochastic behavior is shown here to be essential in any attempt to formulate a successful theoretical model for regular Trichel pulses, i.e., the model must take into account the significant influence of memory uncovered here. It is not the purpose of the present work to give a complete or precise electrical characterization of Trichel pulses for well-defined conditions as has been the subject of much of the earlier work mentioned above. The results of the present work, however, indicate the difficulties that will be encountered in attempts to make precise characterizations.

#### **II. THEORETICAL CONSIDERATIONS**

The purposes of this section are (i) to define the measurable conditional and unconditional pulse-height and pulse-time-separation probability distributions, (ii) to discuss their properties, and (iii) to derive the consistency relationships that must be satisfied among these distributions for conditions that are relevant to Trichel pulses. The general theory of probabilities upon which this discussion is based can be found in numerous texts (see, for example, Refs. 31-33).

#### A. Definitions

The Trichel-pulse phenomenon can be represented by a doubly stochastic, marked random point process introduced by König and Matthes and discussed by Snyder.<sup>33</sup> The process is indicated diagrammatically in Fig. 1. The pulses occur at discrete times  $t_i$  and are marked with an amplitude  $q_i$ . The process is doubly stochastic because the evolution of the marks (pulses) and the occurrence times are influenced by an "outside" random process as well as by self-excitation from previous pulses. The outside random process in this case corresponds to ultraviolet (uv) irradiation of the cathode which, for the experimental conditions used, was required to sustain a continuous train of pulses. Although the probability for occurrence of a pulse at  $t_i$  was largely controlled by release of photoelectrons from the cathode by the uv radiation, self-excitation due to release of initiating electrons by residuals, e.g., species in metastable excited states, from previous pulses is, as shown below, significant under some conditions.

In general, both  $q_i$  and  $t_i$  depend on the set  $\{q_j, t_j\}$ , j < i, of all previous pulse amplitudes and their times of occurrence.<sup>27</sup> The Trichel-pulse phenomenon will be defined here as a stochastic process specified by a discrete, finite set  $\{q_1, q_j, \Delta t_{j-1}\}_n$ ,  $j = 2, 3, 4, \ldots, n$  (see Fig. 1), where  $\Delta t_{j-1} = t_j - t_{j-1}$ , since pulse time separations rather than actual occurrence times are the quantities either measured or specified in the experiment. The pulse amplitude was chosen as the relevant mark because, as shown in Sec. III, it is the appropriate measure of discharge *intensity* under the experimental conditions used.

A list of the probability distributions that could be measured in the present experiment are given in Table I. The distributions  $p_0$  are unconditional whereas the distributions  $p_1$  and  $p_2$  are conditional. These distributions are defined, for example, such that  $p_0(q_n)dq_n$  is the probability that the *n*th discharge pulse in the se-



FIG. 1. Diagrammatic representation of the marked random point process corresponding to the Trichel-pulse discharge phenomenon. The  $\Delta t_n$  are pulse time separations and the  $q_n$  are pulse amplitudes.

Unconditional distributions	$\begin{array}{c} p_0(q_n) \\ p_0(\Delta t_n) \end{array}$
First-order conditional distributions	$p_1(q_n q_{n-1}) p_1(q_n \Delta t_{n-j}),  j = 1, 2, 3, 4 p_1(\Delta t_n q_n) p_1(\Delta t_n \Delta t_{n-1})$
Second-order conditional distributions	$p_2(q_n \Delta t_{n-1}, q_{n-1}) \ p_2(q_n \Delta t_{n-1}, \Delta t_{n-2})$

TABLE I. Measurable pulse-amplitude and pulse-time-separation distributions.

quence has an amplitude between  $q_n$  and  $q_n + dq_n$ , independent of previous pulse amplitudes or time separations;  $p_1(q_n|\Delta t_{n-1})dq_n$  is the probability that the *n*th pulse has an amplitude in the same range if this pulse is separated from the previous pulse by a fixed time separation  $\Delta t_{n-1}$ ; and  $p_2(q_n|\Delta t_{n-1}, q_{n-1})dq_n$  is the same probability, but with both  $\Delta t_{n-1}$  and  $q_{n-1}$  fixed. The distributions apply to arbitrary *n* since *n* was not defined by the measurement process.

All conditional and unconditional distributions, by definition, satisfy the following normalization requirement:

$$\int_0^\infty p_j(x|q_{n-1}, \Delta t_{n-1}, \ldots) \, dx = 1, \tag{1}$$

which holds for arbitrary  $j \ge 0$ , where  $x = q_n$  or  $\Delta t_n$ . It is often convenient for display purposes to use distributions that are arbitrarily normalized to the maximum value, e.g.,  $p_0(q_n)/p_0(q_{n,\max})$  where  $q_{n,\max}$  corresponds to the maximum in  $p_0(q_n)$ .

#### **B.** Correlation and memory

By making a comparison of the measured conditional and unconditional pulse-height and time-separation distributions it is possible to infer immediately whether or not two random variables such as  $q_n$  and  $\Delta t_{n-1}$  are correlated and therefore dependent. If it is found, for example, that  $p_1(q_n|\Delta t_{n-1}) \neq p_0(q_n)$  for at least some allowed values of  $q_n$  and for all allowed  $\Delta t_{n-1}$  such that  $p_0(\Delta t_{n-1}) \neq 0$ , then  $q_n$  and  $\Delta t_{n-1}$  are dependent variables. Likewise, if  $p_2(q_n|\Delta t_{n-1}, q_{n-1}) \neq p_1(q_n|\Delta t_{n-1})$ 

$$R_{x,y} = \frac{\int_{x} \int_{y} p_{0}(x) p_{1}(y|x) (x - \langle x \rangle) (y - \langle y \rangle) dx dy}{\left(\int_{x} p_{0}(x) (x - \langle x \rangle)^{2} dx \int_{y} p_{0}(y) (y - \langle y \rangle)^{2} dy\right)^{1/2}},$$

which can be computed if  $p_0(x)$ ,  $p_0(y)$ , and  $p_1(y|x)$  have been measured. In Eq. (5),  $\langle x \rangle$  and  $\langle y \rangle$  are expectation (mean) values given by

$$\langle \xi \rangle = \int_{\xi} \xi p_0(\xi) \ d\xi, \ \xi = x \text{ or } y, \tag{6}$$

where the variable x must precede y in the time sequence.

for allowed  $q_{n-1}$ , then a dependence must exist between  $q_n$  and  $q_{n-1}$  at a fixed  $\Delta t_{n-1}$ .

It is found experimentally for Trichel pulses, as will be seen later, that there generally exists a significant time separation range  $\Delta t_c \leq \Delta t_{n-1} \leq \Delta t_\ell$  in which the variable  $q_n$  is strongly dependent on  $\Delta t_{n-1}$ . The variable  $q_n$ is defined here to have a *strong dependence* on  $\Delta t_{n-1}$  if there is a one-to-one correspondence between the expectation value  $\langle q_n(\Delta t_{n-1}) \rangle$  and  $\Delta t_{n-1}$ , where

$$\langle q_n \left( \Delta t_{n-1} \right) \rangle = \int_0^\infty q_n p_1 \left( q_n | \Delta t_{n-1} \right) \, dq_n, \tag{2}$$

and where the width of  $p_1(q_n|\Delta t_{n-1})$  is sufficiently small so that  $q_n(\Delta t_{n-1})$  lies within a narrow range defined by  $\langle q_n(\Delta t_{n-1}) \rangle \pm \delta q_n$ , where  $\langle q_n(\Delta t_{n-1}) \rangle \gg |\delta q_n|$  at a particular  $\Delta t_{n-1}$ . Under the strong dependence condition,  $q_n$  is approximately a function of  $\Delta t_{n-1}$ , so that

$$q_n \simeq f\left(\Delta t_{n-1}\right),\tag{3}$$

for  $\Delta t_c \leq \Delta t_{n-1} \leq \Delta t_\ell$ . Equation (3) suggests that  $p_1(q_n | \Delta t_{n-1})$  can be represented by the approximation

$$p_1(q_n | \Delta t_{n-1}) \simeq (1 - \varepsilon) \delta(q_n - f(\Delta t_{n-1})) + \varepsilon p_0(q_n),$$
(4)

where  $\varepsilon \ll 1$  and  $\delta(q_n - f(\Delta t_{n-1}))$  is the Dirac delta function.

A measure of the degree of correlation between any two random variables x and y corresponding to any two of the measured quantities in the set  $\{q_1, q_j, \Delta t_{j-1}\}_n$  is given by the correlation coefficient  $R_{x,y}$ , where

The integrals in Eqs. (5) and (6) are over all allowed values of x and y. It is seen that if x and y are uncorrelated, then  $p_1(y|x) = p_0(y)$  for all allowed x and  $R_{x,y} = 0$ ; whereas if y is strongly dependent on x over its allowed range, e.g.,  $y \propto x$ , then it can be verified that  $|R_{x,y}| \simeq 1$ . In general,  $|R_{x,y}| \le 1$ , where  $R_{x,y} = 0$  if x and y are uncorrelated.<sup>31,32</sup> If the random variable

y is positively dependent on x such that an increase in x implies an increase in  $\langle y(x) \rangle$  such as would be given by Eq. (2), then  $R_{x,y} > 0$ , and if y is negatively dependent on x such that an increase in x implies a decrease in  $\langle y(x) \rangle$ , then  $R_{x,y} < 0$ . A positive dependence will be denoted here by  $x \uparrow \Rightarrow y \uparrow$  and a negative dependence by  $x \uparrow \Rightarrow y \downarrow$ . The variable x can be any random variable that precedes y in the time sequence, e.g.,  $x = \Delta t_{n-i}$ , for  $j \geq 1$ , when  $y = \Delta t_n$ . If x does not immediately precede y in the time ordering, then the dependence of y on x may be conditioned on assumed fixed values for intermediate variables. The signs for the dependences in this case will be denoted by  $(x \uparrow \Rightarrow y \uparrow, \{\xi_i\})$  or  $(x \uparrow \Rightarrow y \downarrow, \{\xi_i\})$ , where  $\{\xi_i\}$  is a set of *fixed* intermediate variables. For example, a positive dependence of  $q_n$  on  $\Delta t_{n-2}$  for fixed  $\Delta t_{n-1}$  is denoted by  $(\Delta t_{n-2} \uparrow \Rightarrow q_n \uparrow, \{\Delta t_{n-1}\})$ .

In the case of Trichel pulses, the dependence of  $q_n$  on

$$M_{1}(t_{a},t_{b}) = \frac{\int_{0}^{\infty} \int_{0}^{t_{a,b}} \left[ p_{1}\left(q|t_{b}-\tau\right) - p_{1}\left(q|t_{a}+\tau\right) \right]^{2} d\tau dq}{\int_{0}^{\infty} \int_{t_{a}}^{t_{b}} \left[ p_{1}\left(q|\tau\right) \right]^{2} d\tau dq}$$

where

$$t_{a,b} = (t_a + t_b)/2. (9)$$

If memory does not exist for pulse time separations in the interval  $(t_a, t_b)$ , such that  $p_1(q|t_b - \tau) = p_1(q|t_a + \tau)$ for all  $0 \ge \tau \ge t_{a,b}$ , then  $M_1(t_a, t_b) = 0$ , whereas  $M_1(t_a, t_b) \ne 0$  implies the existence of some memory. It is required for a meaningful definition of  $M_1(t_a, t_b)$  that  $p_1(q|\tau) \ne 0$  in at least some subinterval  $(t'_a, t'_b) \in (t_a, t_b)$ where  $t'_a \ge t_a, t'_b \ge t_b$ . The conditional distribution  $p_1$ in Eq. (8) can be any  $p_1(q_n|\Delta t_{n-j})$  or  $p_1(\Delta t_n|\Delta t_{n-j})$  for j < n. The advantage of the memory coefficient over the correlation coefficient is that the former applies over a restricted time-separation range whereas the latter is evaluated for the entire range of allowed separations. Memory coefficients can also be defined in terms of higher-order conditional distributions  $p_i, j > 1$ .

#### C. Integral relationships among measured distributions

If memory is important in the stochastic process, then the distributions listed in Table I are not independent, e.g., the behavior of the time-interval distribution  $p_0(\Delta t_n)$  affects the shape of the pulse-amplitude distribution  $p_0(q_n)$  and vice versa. It can be shown from the law of probabilities<sup>31,32</sup> that the measured distributions must satisfy the following integral relationships:

$$p_0(q_n) = \int_0^\infty p_0(\Delta t_{n-1}) p_1(q_n | \Delta t_{n-1}) d(\Delta t_{n-1}),$$
(10)

$$p_{0}(q_{n}) = \int_{0}^{\infty} p_{0}(\Delta t_{n-j}) p_{1}(q_{n} | \Delta t_{n-j}) d(\Delta t_{n-j}),$$
$$j > 1, \quad (11)$$

 $\Delta t_{n-1}$  is positive for  $\Delta t_c \leq \Delta t_{n-1} \leq \Delta t_\ell$ , i.e.,  $\Delta t_{n-1} \uparrow \Rightarrow q_n \uparrow$ . If the pulse time separation is greater than a critical value  $\Delta t_\ell$  for which residuals from previous pulses cease to influence the onset or growth of the next pulse, a loss of memory ensues. The correlation coefficients  $R_{q_n,\Delta t_{n-1}}$  and  $R_{\Delta t_n,\Delta t_{n-1}}$  diminish increasingly from unity as the fraction of time intervals above  $\Delta t_\ell$  increases, i.e., as the probability

$$p\left(\Delta t_{n-1} > \Delta t_{\ell}\right) = \int_{\Delta t_{\ell}}^{\infty} p_0\left(\Delta t_{n-1}\right) d\left(\Delta t_{n-1}\right)$$
(7)

increases.

Another measure of the extent to which memory is important for pulse time separations in any range  $t_a \leq \Delta t_n \leq t_b$  is given by the *memory coefficient*  $M_1(t_a, t_b)$ , defined here for first-order conditional distributions by

$$p_0(q_n) = \int_0^\infty p_0(q_{n-1}) p_1(q_n | q_{n-1}) \, dq_{n-1}, \qquad (12)$$

$$p_{0}(\Delta t_{n}) = \int_{0}^{\infty} p_{0}(q_{n}) p_{1}(\Delta t_{n}|q_{n}) dq_{n}, \qquad (13)$$

$$p_{1}(q_{n}|\Delta t_{n-1}) = p_{0}(\Delta t_{n-1})^{-1} \\ \times \int_{0}^{\infty} p_{0}(q_{n-1}) p_{1}(\Delta t_{n-1}|q_{n-1}) \\ \times p_{2}(q_{n}|q_{n-1},\Delta t_{n-1}) dq_{n-1},$$
(14)

$$p_{1}(q_{n}|q_{n-1}) = \int_{0}^{\infty} p_{1}(\Delta t_{n-1}|q_{n-1}) \times p_{2}(q_{n}|q_{n-1}, \Delta t_{n-1}) d(\Delta t_{n-1}).$$
(15)

From Eqs. (10), (12), (14), and (15) it is seen that

$$p_{0}(q_{n}) = \int_{0}^{\infty} \int_{0}^{\infty} p_{0}(q_{n-1}) p_{1}(\Delta t_{n-1}|q_{n-1}) \times p_{2}(q_{n}|q_{n-1}, \Delta t_{n-1}) dq_{n-1} d(\Delta t_{n-1}).$$
(16)

The evaluation of the integrals in Eqs. (10)-(16) serves as a check on the self-consistency of the measured distributions.

In addition to the above integrals, there are other useful integral relationships that apply if the strong dependency condition between  $q_n$  and  $\Delta t_{n-1}$  is valid. Another expression for  $p_1(q_n | \Delta t_{n-1})$  from the law of probabilities is

$$p_{1}(q_{n}|\Delta t_{n-1}) = p_{0}(\Delta t_{n-1})^{-1} \int_{0}^{\infty} \int_{0}^{\infty} p_{0}(\Delta t_{n-2}) p_{1}(q_{n-1}|\Delta t_{n-2}) \times p_{2}(\Delta t_{n-1}|q_{n-1},\Delta t_{n-2}) p_{3}(q_{n}|q_{n-1},\Delta t_{n-1},\Delta t_{n-2}) d(\Delta t_{n-2}) dq_{n-1}.$$
(17)

Using the strong dependency condition, Eq. (3), and the normalization requirement, Eq. (1), it can be shown that Eq. (17) becomes

$$p_1(q_n | \Delta t_{n-1}) \simeq p_0(\Delta t_{n-1})^{-1} \int_0^\infty p_0(\Delta t_{n-2}) p_1(\Delta t_{n-1} | \Delta t_{n-2}) p_2(q_n | \Delta t_{n-1}, \Delta t_{n-2}) d(\Delta t_{n-2}).$$
(18)

From similar arguments, the following also hold for a strong  $(q_n, \Delta t_{n-1})$  dependence:

$$p_1\left(\Delta t_n | \Delta t_{n-1}\right) \simeq \int_0^\infty p_1\left(\Delta t_n | q_n\right) p_1\left(q_n | \Delta t_{n-1}\right) \, dq_n,\tag{19}$$

$$p_1(q_n | \Delta t_{n-2}) \simeq \int_0^\infty p_1(\Delta t_{n-1} | \Delta t_{n-2}) p_2(q_n | \Delta t_{n-1}, \Delta t_{n-2}) d(\Delta t_{n-1}), \qquad (20)$$

$$p_2(q_n|\Delta t_{n-1},\Delta t_{n-2}) \simeq p_1(\Delta t_{n-1}|\Delta t_{n-2})^{-1} \int_0^\infty p_1(q_{n-1}|\Delta t_{n-2}) p_1(\Delta t_{n-1}|q_{n-1}) p_2(q_n|q_{n-1},\Delta t_{n-1}) dq_{n-1}.$$
 (21)

Equations (10)-(21) are used here to understand the profiles of measured distributions and to derive consistency relationships in the signs of correlations among successive pulse amplitudes and time separations.

## D. Consistency among $p_1(q_n|\Delta t_{n-1})$ , $p_1(\Delta t_n|\Delta t_{n-1})$ , and $p_1(\Delta t_n|q_n)$

The integral relationships in the Sec. IIC imply a certain consistency of behavior among the first-order conditional distributions  $p_1(q_n|\Delta t_{n-1})$ ,  $p_1(\Delta t_n|q_n)$ , and  $p_1(\Delta t_n | \Delta t_{n-1})$  with respect to the implied signs of the dependences between the pairs of random variables  $(q_n, \Delta t_{n-1}), (\Delta t_n, q_n), \text{ and } (\Delta t_n, \Delta t_{n-1}).$  This consistency applies for monotonic dependences, i.e., dependences between random variables that do not change sign, and for the case of a strong positive dependence of  $q_n$  on  $\Delta t_{n-1}$ , thus implying that Eq. (19), which relates the above distributions, can be considered. By using Eq. (4) in Eq. (19) it can be verified that  $(\Delta t_{n-1} \uparrow$  $\Rightarrow q_n \uparrow) \text{ plus } (q_n \uparrow \Rightarrow \Delta t_n \uparrow) \text{ gives } (\Delta t_{n-1} \uparrow \Rightarrow \Delta t_n \uparrow)$ as the required behavior for  $p_1(\Delta t_n | \Delta t_{n-1})$ . According to Eq. (19) for this case, an increase in the value of  $\Delta t_{n-1}$  implies an increase in the expectation value  $\langle \Delta t_n(\Delta t_{n-1}) \rangle$ . Likewise it is found from Eq. (19) that  $(\Delta t_{n-1} \uparrow \Rightarrow q_n \uparrow)$  plus  $(q_n \uparrow \Rightarrow \Delta t_n \downarrow)$  requires  $(\Delta t_{n-1} \uparrow \Rightarrow \Delta t_n \downarrow)$ . Both cases have been observed here to occur for Trichel pulses.

It should also be noted that if there is a lack of correlation between  $\Delta t_n$  and  $q_n$ , then  $p_1(\Delta t_n|q_n) = p_0(\Delta t_n)$ for all allowed  $q_n$  and Eq. (19) gives  $p_1(\Delta t_n|\Delta t_{n-1})$  $= p_0(\Delta t_n)$ . This means that a lack of dependence of  $\Delta t_n$ on  $q_n$  requires a lack of dependence of  $\Delta t_n$  on  $\Delta t_{n-1}$  if the strong dependency condition between  $q_n$  and  $\Delta t_{n-1}$ applies.

E. Consistency among 
$$p_1(q_n|\Delta t_{n-1})$$
,  
 $p_2(q_n|\Delta t_{n-1}, q_{n-1})$ , and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$ 

As in the case of the first-order conditional timeinterval distributions discussed in Sec. II D, a strong pos-

itive dependence of  $q_n$  on  $\Delta t_{n-1}$  requires that the dependencies between the pairs  $(q_n, q_{n-1})$  and  $(q_n, \Delta t_{n-2})$  for fixed  $\Delta t_{n-1}$  be of the same sign. This can be shown using Eq. (3) in Eq. (21). Given the signs of the  $(q_n, \Delta t_{n-1})$ and  $(q_n, q_{n-1})$  dependences, the sign of the  $(q_n, \Delta t_{n-2})$ dependence can be ascertained by examining the changes in weighting of  $q_{n-1}$  and  $q_n$  when  $\Delta t_{n-2}$  is changed as implied by the first and third terms in the product forming the integrand for Eq. (21) at fixed  $\Delta t_{n-1}$ . The terms  $p_1(\Delta t_{n-1}|\Delta t_{n-2})^{-1}$  and  $p_1(\Delta t_{n-1}|q_{n-1})$  affect only the magnitude of the integral in Eq. (21) and not the weighting of  $q_{n-1}$  and  $q_n$  values. For fixed  $\Delta t_{n-1}$ ,  $(\Delta t_{n-2} \uparrow$  $\Rightarrow q_{n-1}$   $\uparrow$ ) plus  $(q_{n-1} \uparrow \Rightarrow q_n \uparrow, \{\Delta t_{n-1}\})$  requires  $(\Delta t_{n-2} \uparrow \Rightarrow q_n \uparrow, \{\Delta t_{n-1}\}).$  Likewise,  $(\Delta t_{n-2} \uparrow$  $\Rightarrow q_{n-1}$   $\uparrow$ ) plus  $(q_{n-1} \uparrow \Rightarrow q_n \downarrow, \{\Delta t_{n-1}\})$  requires  $(\Delta t_{n-2} \uparrow \Rightarrow q_n \downarrow, \{\Delta t_{n-1}\})$ . Both cases are shown here to occur for Trichel pulses. It can also be shown that a lack of dependence of  $q_n$  on  $\Delta t_{n-2}$  at fixed  $\Delta t_{n-1}$  implies a lack of dependence of  $q_n$  on  $q_{n-1}$  if the strong  $(q_{n-1}, \Delta t_{n-2})$  dependency condition holds.

It should be noted that the physical bases for the  $(q_n, q_{n-1})$  and  $(q_n, \Delta t_{n-2})$  dependences are not the same as for the  $(\Delta t_n, q_n)$  and  $(\Delta t_n, \Delta t_{n-1})$  dependences considered in Sec. II D. The dependence of  $\Delta t_n$  on  $q_n$  is related to the effect of residuals from the *n*th pulse on the *initiation* of the subsequent pulse, whereas the dependence of  $q_n$  on  $q_{n-1}$  is related to the effect of residuals from the (n-1)st pulse on the growth of the subsequent pulse.

From Eq. (20), which applies for a strong  $(q_n, \Delta t_{n-1})$ dependence, it can be seen that correlations between  $\Delta t_{n-1}$  and  $\Delta t_{n-2}$  or between  $q_n$  and  $\Delta t_{n-2}$  for fixed  $\Delta t_{n-1}$ , implied respectively by  $p_1(\Delta t_{n-1}|\Delta t_{n-2})$  and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$ , lead to a dependence of  $q_n$  on  $\Delta t_{n-2}$  independent of  $\Delta t_{n-1}$ . For uv-sustained Trichel pulses, the effects of the two correlations implied by  $p_1$ and  $p_2$  in Eq. (20) can either reinforce or counteract each other so that the sign of the  $(q_n, \Delta t_{n-2})$  dependence determined by  $p_1(q_n|\Delta t_{n-2})$  depends on the relative signs and strengths of the  $(\Delta t_{n-1}, \Delta t_{n-2})$  dependence and the  $(q_n, \Delta t_{n-2})$  dependence for fixed  $\Delta t_{n-1}$ . For a strong  $(q_n, \Delta t_{n-1})$  dependence, Eq. (15) implies that Eq. (20) is a good approximation, and therefore, the behavior of  $p_1(q_n|\Delta t_{n-2})$  should be like that for  $p_1(q_n|q_{n-1})$ , e.g.,  $(\Delta t_{n-2} \uparrow \Rightarrow q_{n-1} \uparrow)$  plus  $(q_{n-1} \uparrow \Rightarrow q_n \downarrow)$  implies  $(\Delta t_{n-2} \uparrow \Rightarrow q_n \downarrow)$ .

It should be emphasized that the above arguments about consistency of the signs apply only if the dependences are monotonic, namely that they do not change sign within the ranges of allowed values for the random variables considered. Although the signs of dependences between the random variables considered above can change, statistically significant changes were not found to occur in the data presented here.

## F. Propagation and loss of memory (deviations from Markovian behavior)

Extending the arguments in Sec. II E, it can be shown that if  $q_n$  is dependent on both  $q_{n-1}$  and  $\Delta t_{n-1}$  for all arbitrary *n*, then in general,

$$p_{j}(q_{n}|\{\Delta t_{n-i}\}_{j-1}, \Delta t_{n-j})$$

$$\neq p_{j}(q_{n}|\{\Delta t_{n-i}\}_{j-1}, \Delta t'_{n-j}), \quad i = 1, 2, \dots, j-1,$$
(22)

whenever  $\Delta t_{n-j} \neq \Delta t'_{n-j}$ , for any  $j \ge 1$ . If all the time separations in the set  $\{\Delta t_{n-i}\}_{j-1}$  have fixed values less than  $\Delta t_{\ell}$ , then the following can be shown, in the case of monotonic correlations.

(a) 
$$(\Delta t_{n-1} \uparrow \Rightarrow q_n \uparrow) + (q_{n-1} \uparrow \Rightarrow q_n \downarrow)$$
 implies  
 $(\Delta t'_{n-j} \uparrow \Rightarrow q_n \uparrow, \{\Delta t_{n-i}\}_{j-1})$  for odd  $j$   
 $(\Delta t'_{n-j} \uparrow \Rightarrow q_n \downarrow, \{\Delta t_{n-i}\}_{j-1})$  for even  $j$ .  
(b)  $(\Delta t_{n-1} \uparrow \Rightarrow q_n \uparrow) + (q_{n-1} \uparrow \Rightarrow q_n \uparrow)$  implies  
 $(\Delta t'_{n-j} \uparrow \Rightarrow q_n \uparrow, \{\Delta t_{n-i}\}_{j-1})$  for all  $j$ .

The existence of dependences between both of the pairs  $(q_j, \Delta t_{j-1})$  and  $(q_j, q_{j-1})$  therefore allows memory to propagate indefinitely back in time provided  $\Delta t_j < \Delta t_\ell$  for all values of j. A dependence of  $\Delta t_n$  on  $q_n$  can also influence memory propagation. Memory will cease beyond the first event to have a time separation from the previous event greater than the critical minimum time separation  $\Delta t_\ell$  for which  $M_1(\Delta t_\ell, \Delta t_u) = 0$ , where  $\Delta t_u$  is any time separation greater than  $\Delta t_\ell$ .

For the Trichel-pulse phenomenon, the initiation and growth of any discharge pulse is expected to depend overwhelmingly on residuals from the most recent previous pulse. From this consideration it has been erroneously suggested<sup>27</sup> that memory does not extend back beyond the most recent event. The above analysis indicates that, because of correlations between both  $(q_j, \Delta t_{j-1})$ and  $(q_j, q_{j-1})$ , memory can extend back beyond the most recent event even though there may be no residuals present from pulses prior to this event. If residuals such as metastable species are present from earlier events, then they may have second-order effects on the onset and growth of a discharge pulse. It was not possible in the present experiments to distinguish contributions to memory from such second-order effects. It is assumed here that "memory" results predominantly from the dependences of  $q_n$  (or  $\Delta t_n$ ) on  $q_{n-1}$  and  $\Delta t_{n-1}$ .

It is evident from the above discussion that the Trichel-pulse phenomenon is not a Markov process. By definition,<sup>27,33</sup> a Markov process, jointly defined for the two random variables  $q_i$  and  $\Delta t_{i-1}$ , requires

$$p_{2j+1}\left(q_n | \Delta t_{n-1}, \{q_i, \Delta t_{i-1}\}_j\right) = p_2\left(q_n | q_{n-1}, \Delta t_{n-1}\right),$$

$$i = n - 1, n - 2, \dots, n - j \text{ for all } j \le n - 1.$$
If  $q_i$  is strongly dependent on  $\Delta t_{i-1}$ , then
$$(23)$$

 $p_{2j+1}(q_n | \Delta t_{n-1}, \{q_i, \Delta t_{i-1}\}_j)$ 

$$\simeq p_{j+1} (q_n | \Delta t_{n-1}, \{ \Delta t_{i-1} \}_j).$$
 (24)

In this case, the Markov requirement becomes

$$p_{2j+1}(q_n | \Delta t_{n-1}, \{\Delta t_{i-1}\}_j) = p_1(q_n | \Delta t_{n-1}),$$
  
$$j \le n-1. \quad (25)$$

Obviously this is not consistent with Eq. (22). Also, if memory does not extend beyond the most recent event, it is required that

$$p_1\left(q_n|\Delta t_{n-j}\right) = p_0\left(q_n\right) \tag{26}$$

for all j > 1. It is shown here experimentally for Trichel pulses that, in general,

$$p_2(q_n | \Delta t_{n-1}, \Delta t_{n-2}) \neq p_1(q_n | \Delta t_{n-1})$$
(27)

and

$$p_1(q_n | \Delta t_{n-j}) \neq p_0(q_n), \ j = 2, 3, 4.$$
 (28)

#### **III. EXPERIMENTAL METHOD**

The method used to measure the distributions given in Table I has previously been described in detail by Van Brunt and Kulkarni.<sup>27</sup> A diagram of the measurement system is shown in Fig. 2. Some modifications of the system described in Ref. 27 have been made to (i) allow direct measurement of the distributions  $p_1(q_n|\Delta t_{n-j})$ , j > 2, and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$ ; (ii) eliminate intermediate pulse errors associated with measurement of the  $p_2$  distributions; and (iii) reduce background associated with accidental sampling of the tails of discharge pulses.

By appropriate positioning of the switches (S1-S6) shown in Fig. 2, it is possible to configure the system to measure any of the indicated distributions. Data from all measurement configurations are accumulated by a 256-channel, computer-controlled multichannel analyzer (MCA) which uses an analog-to-digital converter to mea-



FIG. 2. System for measuring various indicated pulse-amplitude and time-separation distributions. In this diagram, the relevant components are identified as follows: MCA, multichannel analyzer; TAC, time-to-amplitude converter; SCA, single channel analyzer; DDG, digital-delay generator; A, amplifier; G, gate; and S, switch. Additional details about the individual circuits used in this system can be found in Ref. 27.

sure pulse heights.

The corona-discharge current pulses are detected electrically using a preamplifier (A1) connected to an impedance Z in series with the discharge gap. The detected pulses are then sent to a variable gain amplifier (A2) before being routed to the required circuit path for measurement of a particular distribution as determined by the position of S1. For measurement of  $p_0(q_n)$ , the discharge pulses from A2 are fed directly to the MCA through gate G3, which is kept permanently open by the proper positioning of S4.

The distribution  $p_0(\Delta t_n)$  is measured by sending the pulses from A2 to a pulse sorter used to trigger two timeto-amplitude converters (TAC1 and TAC2) which generate output pulses of amplitude proportional to the time between the trigger pulses at the start and stop inputs. The outputs of TAC1 and TAC2 are fed to the MCA through gate G2 by appropriate positioning of S3. As shown in Ref. 27, this arrangement permits the recording of every sequential time interval, provided all time separations exceed the TAC reset time of 50  $\mu$ s.

The measurement of  $p_1(q_n|\Delta t_{n-1})$  requires the use of a logic-controlled digital-delay generator (DDG), which restricts transfer of pulses from A2 to the MCA through G3 to a narrow, preselected time range  $\Delta t_{n-1} - \delta(\Delta t_{n-1})/2$ to  $\Delta t_{n-1} + \delta(\Delta t_{n-1})/2$  for which  $\Delta t_{n-1} \gg \delta(\Delta t_{n-1})$ , provided no intermediate pulses appear before this time window. To measure  $p_2(q_n|\Delta t_{n-1}, q_{n-1})$ , a singlechannel analyzer (SCA1) is inserted between A2 and the input to the DDG time-interval control logic circuit by proper positionings of S1, S5, and S6. The SCA1 restricts triggering of the DDG to pulses having amplitudes within the selected range  $q_{n-1} - \delta(q_{n-1})/2$  to  $q_{n-1} + \delta(q_{n-1})/2$ for which  $q_{n-1} \gg \delta(q_{n-1})$ . To ensure, in the case of  $p_2$ measurements, that the time-interval control logic properly inhibits the opening of G3 if intermediate pulses occur before the selected time window, it is necessary to modify the circuit previously described<sup>27</sup> by adding the amplifier A3 with input at f. This amplifier is identical to that defined by transistor  $T_1$  in Fig. 2 of Ref. 27. In the present configuration, the input f is connected directly to the delay  $\tau_1$  of Fig. 2 (Ref. 27) which has, in turn, been disconnected from  $T_1$ .

Unlike the system previously described,<sup>27</sup> the present system allows direct measurement of  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$ . For measurement of this distribution, the switches S1 and S5 are positioned so that the input to SCA1 is derived from TAC1. The time interval measured by TAC1 is restricted by SCA1 to lie within a narrow range  $\Delta t_{n-2} - \delta(\Delta t_{n-2})/2$  to  $\Delta t_{n-2} + \delta(\Delta t_{n-2})/2$ for triggering the DDG, which, in turn, defines  $\Delta t_{n-1}$ .

By the indicated positionings of switches S1, S2, and S3, it is possible to measure the conditional pulse timeseparation distributions  $p_1(\Delta t_n | \Delta t_{n-1})$  and  $p_1(\Delta t_n | q_n)$ . For measurement of  $p_1(\Delta t_n | \Delta t_{n-1})$ , the output of TAC1 is fed to the input of the single-channel analyzer SCA2 which thus defines  $\Delta t_{n-1}$  and triggers TAC3 for the measurement of the next time interval  $\Delta t_n$ . In the case of the  $p_1(\Delta t_n | q_n)$  measurement, SCA2 is connected directly to amplifier A2 thereby used to define  $q_n$ .

The present system also differs from that previously described in that the  $p_1(q_n|\Delta t_{n-j})$  logic circuit now contains a pulse counter that can be set for any  $j \ge 2$ . By making adjustments in the delays inherent to the time-

interval control logic combined with minor adjustments in the shape of the gate pulse from this circuit which controls G3, it has been possible to largely eliminate the background problem illustrated by Fig. 5 of Ref. 27.

In the present measurement system, the pulse detection-circuit conditions are similar to those described in Ref. 7 so that the measured discharge-pulse amplitude is proportional to the net charge transported in the discharge given by

$$Q'_{n} = \int_{-\infty}^{\infty} i_{n}(t) dt, \qquad (29)$$

where  $i_n(t)$  is the instantaneous discharge current at time t for the *n*th pulse. Given the impulse response h(t - t') of the detection circuit (defined here mainly by the filter characteristics of Z and A1), the observed pulse signal is given by

$$q'_{n}(t) = \int_{-\infty}^{t} i_{n}(t') h(t-t') dt'.$$
(30)

The impulse response in the present measurement system has a width  $w_i$  of about  $1.5\,\mu$ s, whereas the intrinsic width  $w_p$  of a typical Trichel pulse is known<sup>12-15</sup> to be on the order of 10-50 ns. Therefore the condition  $w_i \gg w_p$  allows the approximation

$$i_n(t) \simeq Q'_n \delta(t - t_n), \qquad (31)$$

where  $\delta(t - t_n)$  is the Dirac delta function. Using this in Eq. (30) gives

$$q'_{n}(t) = Q'_{n}h(t - t_{n}).$$
 (32)

This means that the shapes of the observed Trichel pulses in the present experiment are governed primarily by the shape of  $h(t-t_n)$  and thus the maximum of  $q'_n(t)$ , denoted here by  $q_n$ , is directly proportional to  $Q'_n$ . Therefore  $q_n$ can be expressed in units of charge, and in the present case, it is convenient to use picocoulombs (pC). The method for calibrating the pulse-height measurements in terms of charge is the same as described previously.<sup>7</sup>

The Trichel pulses are generated by applying a dc voltage to a point-to-plane electrode gap in which the point electrode serves as the cathode. General features of the stochastic behavior reported here were found to apply over wide ranges of gap spacings, point-electrode radius, gas pressure, and gas composition. In order to achieve discharge characteristics that were constant over long periods of time (several hours) and to gain control over the rate of pulse initiation, the discharge was primarily sustained by uv irradiation from a mercury arc lamp of highly polished stainless-steel point cathodes in controlled gas mixtures. It was noted, however, that similar stochastic behavior often occurs when the discharge becomes self-sustaining or is sustained by field emission, i.e., by the Fowler-Nordheim effect,<sup>34</sup> which occurs when very sharp electrodes are used. Point-to-plane gap spacings in the range of 1.0-2.5 cm were used with points having radii of curvature at the tip in the approximate

range 0.09–0.15 mm. The gas mixtures were prepared in a metal discharge chamber after it was evacuated to a pressure of about  $1 \times 10^{-2}$  Torr (~ 1.3 Pa). The results presented here were obtained using the gas mixtures N<sub>2</sub>+20% O<sub>2</sub>, Ne+5% O<sub>2</sub>, Ne+95% O<sub>2</sub>, all at an absolute total pressure of 760 Torr (~ 100 kPa).

When measurements were made of the conditional distributions  $(p_1$ 's and  $p_2$ 's of Table I) for a fixed set of discharge conditions defined by applied gap voltage, uv intensity, electrode spacing, and gas composition, it was necessary to periodically monitor the distributions  $p_0(q_n)$ and  $p_0(\Delta t_n)$  to ensure that there was no significant drift in these conditions. As shown in Sec. IV, the profiles for these distributions are very sensitive indicators of variations in the discharge operating conditions.

It was generally possible to maintain acceptably constant discharge conditions over a period of several hours. On the other hand, due to an inability to make sufficiently precise adjustments of the operating conditions in the present experimental setup, it was not always possible to achieve exactly the same profiles for  $p_0(q_n)$  and  $p_0(\Delta t_n)$  from one day to the next. This difficulty was also noted by Malik and Al-rainy.<sup>26</sup> However, for the purposes of the present work, which are not to make a precise characterization of the phenomena but rather to investigate its stochastic properties, which, as noted above, apply over wide ranges of conditions, it was not necessary to achieve exact replications of  $p_0(q_n)$  and  $p_0(\Delta t_n)$ . The results presented here, in fact, clearly indicate that because of strong memory effects associated with the stochastic behavior of Trichel pulses, it can be expected that difficulties will be encountered in attempts to establish highly reproducible conditions.

## **IV. RESULTS AND DISCUSSION**

Data obtained for the different distributions given in Table I are presented here. Consistency among the measured distributions is analyzed and at the end of each subsection their behavior is interpreted in terms of expected physical processes that can occur in the discharge.

# A. Behavior of the unconditional distributions $p_0(q_n)$ and $p_0(\Delta t_n)$

Shown in Figs. 3 and 4 are examples of data for the unconditional pulse-amplitude and time-separation distributions at different applied gap voltages  $V_a$  for a  $N_2 + 20\% O_2$  gas mixture. The data in Fig. 3 were obtained using a higher relative uv intensity to irradiate the cathode than applies to the data in Fig. 4. The discharge conditions were otherwise the same. For convenience of display, the distributions shown in these figures are normalized to the maximum values.

At "high" uv irradiation, the  $p_0(q_n)$  distribution is seen in Fig. 3 to broaden with increasing  $V_a$  while  $p_0(\Delta t_n)$ becomes more narrow and shifts downward. The expectation values for  $q_n$  and  $\Delta t_n$  given by



FIG. 3. Measured unconditional pulse-amplitude  $p_0(q_n)$ and corresponding pulse-time-separation  $p_0(\Delta t_n)$  distributions at the different indicated gap voltages, for relatively intense uv irradiation of the cathode in a 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> gas mixture. The distributions have been normalized to the maximum values.

$$\langle q_n \rangle = \int_0^\infty q_n p_0\left(q_n\right) dq_n \tag{33}$$

and

$$\langle \Delta t_n \rangle = \int_0^\infty \Delta t_n p_0 \left( \Delta t_n \right) d\left( \Delta t_n \right) \tag{34}$$

both decrease as  $V_a$  increases. At "low" uv intensity, the data in Fig. 4 indicate that  $\langle q_n \rangle$  now increases for increasing  $V_a$  while  $\langle \Delta t_n \rangle$  again decreases. Also at low uv intensity,  $p_0(q_n)$  becomes sharply peaked at the high end of the distribution. Contrary to suggestions from earlier work,<sup>26</sup> the distributions  $p_0(q_n)$  are, in general, asymmetric and therefore distinctly non-Gaussian.

The decrease in both  $\langle q_n \rangle$  and  $\langle \Delta t_n \rangle$  with increasing  $V_a$  has been reported previously<sup>6,8,26</sup> for "self-sustained" Trichel-pulse discharges in air and oxygen. The self-sustained discharge condition (which may be at least partially sustained by field emission at the cathode) is similar to the high uv intensity case considered here. These situations are similar in the sense that electron-emission



FIG. 4. Measured unconditional pulse-amplitude  $p_0(q_n)$ and corresponding pulse-time-separation  $p_0(\Delta t_n)$  distributions at the different indicated gap voltages, for relatively weak uv irradiation of the cathode in a 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> gas mixture. The distributions have been normalized to the maximum values.

rates that govern the pulse-initiation rates are high in both cases. The values for  $\langle q_n \rangle$  implied by the  $p_0(q_n)$ distributions in Fig. 3 are consistent in magnitude with the results of Lama and Gallo for air extrapolated to the point electrode radius used in the present experiments that exceeds their largest by a factor of 2 (see Fig. 11 of Ref. 6). The pulse repetition rates implied by the  $p_0(\Delta t_n)$  distributions are also consistent in magnitude with earlier results for air.<sup>4-6</sup>

Figure 5 shows results for  $p_0(q_n)$  and  $p_0(\Delta t_n)$  obtained for different uv intensities  $I_i$  at a fixed  $V_a$ , where  $I_1 > I_2 > I_3 > \cdots$ . The gas and electrode-gap conditions are nearly identical to those that apply for the data in Figs. 3 and 4. It is seen from Fig. 5 that the profiles for  $p_0(q_n)$  and  $p_0(\Delta t_n)$  are very sensitive to uv intensity. Both  $\langle q_n \rangle$  and  $\langle \Delta t_n \rangle$  increase with decreasing  $I_i$ . When  $\langle \Delta t_n \rangle$  exceeds about 400  $\mu$ s, as implied by the  $p_0(\Delta t_n)$ data in Fig. 5 for  $I_6$ ,  $p_0(q_n)$  becomes very narrow and assumes a constant, nearly Gaussian shape for which  $\langle q_n \rangle$ approaches a limiting maximum value, denoted here by  $q_{\lim}$ .

The time-separation distributions exhibit a limiting critical minimum value  $\Delta t_c$  which is nearly independent of  $I_i$ . As seen in Fig. 6,  $\Delta t_c$  decreases with increasing voltage, consistent with results previously reported by

 $p_0$ 



FIG. 5. Measured unconditional pulse-amplitude  $p_0(q_n)$ and corresponding pulse-time-separation  $p_0(\Delta t_n)$  distributions at the different indicated relative intensities of uv irradiation  $I_i$   $(I_1 > I_2 > I_3 \cdots)$  for a gap voltage of 9.0 kV in a 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> gas mixture. The distributions have been normalized to the maximum values.



FIG. 6. Measured dependence of the critical minimum pulse separation time  $\Delta t_c$  on applied gap voltage for two different intensities of uv irradiation. The  $\Delta t_c$  were determined from data similar to those shown in Figs. 3 and 4.

Mason and Young.<sup>35</sup> This limiting time interval can be identified with the time required for restoration of the electric field near the cathode to the critical value required for initiation and growth of the next discharge pulse. This time is governed by the ambipolar diffusion of positive and negative ions away from the point.<sup>6,8,17</sup> It can be expected that  $\Delta t_c$  should decrease with increasing  $V_a$  since the rate of ion diffusion will be enhanced as the applied field increases. Provided the ion-diffusion rate does not depend significantly upon the magnitude of  $q_n$  within the range defined by the  $p_0(q_n)$ 's in Fig. 5, it can be expected that  $\Delta t_c$  will not change significantly with  $I_i$ . The present results suggest that  $\Delta t_c$  is a more well-defined characteristic of the discharge than the previously measured pulse frequency<sup>4-6</sup> given by  $1/\langle \Delta t_n \rangle$ .

The  $p_0(\Delta t_n)$  distributions shown in Fig. 5 exhibit an exponential decrease for  $\Delta t_n > \Delta t_{n,\max}$  at a rate  $d[\log_{10} p_0(\Delta t_n)]/d(\Delta t_n)$ , which is proportional<sup>36</sup> to  $I_i$ . This type of behavior is expected if the probability for discharge-pulse initiation at any time after field restoration is proportional to the rate of photoelectron release from the cathode, which is, in turn, proportional to  $I_i$ . The probability for initiation of the (n + 1)st pulse at a time separation  $\Delta t_n > \Delta t_{n,\max}$  from the previous pulse can be expressed as

$$(\Delta t_n) d(\Delta t_n) \simeq \gamma(V_a) I_i \left(1 - \int_0^{\tau_n} p_0(\Delta t'_n) d(\Delta t'_n)\right) d(\Delta t_n),$$
(35)

where  $\tau_n = \Delta t_n - \Delta t_{n,\max}$ ,  $\gamma(V_a)$  is a factor that depends here only on  $V_a$ , and the quantity in large parentheses represents the probability that a discharge pulse will not have been initiated for intervals in the range  $\Delta t_{n,\max}$  to  $\Delta t_n$ . The function that satisfies Eq. (35) is of the form

$$p_0(\Delta t_n) = \gamma(V_a) I_i \exp \left[-\gamma(V_a) I_i (\Delta t_n - \Delta t_{n,\max})\right],$$
(36)

from which it is seen that

$$d\left[\log_{10} p_0\left(\Delta t_n\right)\right] / d\left(\Delta t_n\right) \propto -I_i,\tag{37}$$

consistent with the behavior for  $p_0(\Delta t_n)$  exhibited in Fig. 5. This is identical to the Poisson distribution law for time lags in the electrical breakdown of gases first noted by von Laue.<sup>37</sup>

The factor  $\gamma(V_a)$  is included in Eq. (35) because the electric-field magnitude at the cathode is sufficiently high [often in excess of  $10^5$  V/cm as estimated using Eq. (40) of Sec. IV B] that the electron-release process is expected to be a *field-assisted* photoelectron emission, which is a well-known effect that has been observed experimentally<sup>38</sup> and is the optical analog of the Schottky effect<sup>39</sup> for thermionic emission from a cathode. The  $p_0(\Delta t_n)$  data in Fig. 3 indicate that  $\gamma(V_a)$  increases with  $V_a$  as expected. Because of this effect, perturbations of the field at the cathode due to ion space charge from pre-



FIG. 7. Measured unconditional pulse-amplitude  $p_0(q_n)$  distributions at the indicated gap voltages for very weak uv irradiation of the cathode in a 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> gas mixture (top), and corresponding  $q_{\rm lim}$  vs applied gap voltage (bottom), where the  $q_{\rm lim}$  values are associated with the maxima in the  $p_0(q_n)$  distributions shown in the top portion of the figure.

vious pulses might also have an influence on the probability of electron emission. Although such an effect appears to be relatively small for the N<sub>2</sub>-O<sub>2</sub> case shown in Fig. 5, results presented later demonstrate that it is significant in other gas mixtures. If space charge from a previous pulse influences the rate of electron emission, then  $\gamma$  will depend on  $q_n$  and  $\Delta t_{n-1}$ . In this case, positive correlations between  $\Delta t_n$  and  $q_n$  (or  $\Delta t_{n-1}$ ) become evident from the conditional distribution  $p_1(\Delta t_n | q_n)$  [or  $p_1(\Delta t_n | \Delta t_{n-1})$ ], as shown in Sec. IV C. The existence of a significant correlation between  $q_n$  and  $\Delta t_n$ , can result in a  $p_0(\Delta t_n)$ , given by Eq. (13), that may deviate in behavior from that implied by Eqs. (36) and (37).

The tendency for the  $p_0(q_n)$  distributions to approach a limiting narrow profile peaked at the high end for decreasing  $I_i$  as seen in Fig. 5 indicates that for any given set of discharge-gap conditions, there is a maximum mean amplitude  $q_{\lim}$  that cannot be exceeded. This limit undoubtedly corresponds to the condition where the gap is largely free of residuals from previous pulses. Pulses for which  $q_n \simeq q_{\lim}$  must therefore correspond to the "firstpulse" condition (n = 1) considered by Morrow.<sup>18,19</sup> Figure 7 shows results for  $p_0(q_n)$  using N<sub>2</sub>+20% O<sub>2</sub> for which the uv intensity was kept very low so that  $\langle \Delta t_n \rangle > 500 \, \mu$ s. The gap conditions were similar to those used to obtain the data for Figs. 3-5. As expected,  $q_{\text{lim}}$  increases with increasing  $V_a$ .

The widths of the distributions  $p_0(q_n)$  as  $q_{n,\max} \rightarrow q_{\lim}$ will be limited by inherent statistical fluctuations in discharge growth and gap conditions, e.g., statistical variations in the exact position on the cathode surface from which the initiating electron is released. For relatively small pulses below about 50 pC, noise in the pulsedetection circuit can limit the observable width of the pulse-height distribution. This limitation was not encountered for any of the data presented here.

The negative-corona pulses reported by Van Brunt and  $\text{Leep}^7$  in SF<sub>6</sub> that were found to be narrowly distributed in amplitude but widely distributed in time are undoubtedly Trichel pulses that satisfy the first-pulse condition. Similar pulses have recently been seen in SF<sub>6</sub>-O<sub>2</sub> gas mixtures.<sup>29</sup>

### B. Behavior of $p_1(q_n|\Delta t_{n-1})$ and the $(q_n, \Delta t_{n-1})$ correlation

The interpretation of data for  $p_0(q_n)$  and  $p_0(\Delta t_n)$  given in Sec. IV A suggests that these distributions are not independent and that there must be a significant correlation between the random variables  $q_n$  and  $\Delta t_{n-1}$ . This correlation was verified by measuring  $p_1(q_n|\Delta t_{n-1})$  at different  $\Delta t_{n-1}$ . Examples of data on  $p_1(q_n|\Delta t_{n-1})$  for  $N_2 + 20\% O_2$  are shown in Figs. 8 and 9, respectively, for high and low levels of uv irradiation. The discharge-gap



FIG. 8. Measured unconditional  $p_0(q_n)$  and corresponding conditional  $p_1(q_n | \Delta t_{n-1})$  distributions at 9.4 kV for the indicated values of  $\Delta t_{n-1}$  which apply to relatively intense uv irradiation of the cathode in a 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> gas mixture. The distributions have been normalized to the maximum values. The solid lines are Gaussian representations of the  $p_1$  data which were used together with corresponding data on  $p_0(\Delta t_n)$  in Eq. (10) to give the predicted  $p_0(q_n)$  distribution indicated by the dashed line.

conditions used to obtain the results shown in Figs. 8 and 9 are similar to those that apply to Figs. 3-7.

The inequality  $p_1(q_n | \Delta t_{n-1}) \neq p_0(q_n)$  seen from the data for allowed  $\Delta t_{n-1}$  implies a dependence of  $q_n$  on  $\Delta t_{n-1}$ . The dependence is positive, i.e.,  $\Delta t_{n-1} \uparrow \Rightarrow q_n \uparrow$ . Because the widths of the  $p_1(q_n | \Delta t_{n-1})$  are considerably narrower than the width of the corresponding unconditional distribution  $p_0(q_n)$ , the strong dependency condition defined in Sec. II B applies over at least a part of the range of allowed  $\Delta t_{n-1}$ . The strong  $(q_n, \Delta t_{n-1})$  dependence was previously noted by Steiner<sup>25</sup> and is indicated here by the computed correlation coefficients  $R_{q_n,\Delta t_{n-1}}$ shown in Fig. 10 for the case of high uv irradiation where  $R_{q_n,\Delta t_{n-1}} \to 1$ . At low uv intensity,  $R_{q_n,\Delta t_{n-1}} \to 0$  as  $V_a$  decreases due to increasing contributions from time separations above  $\Delta t_{\ell}$  (see Figs. 11 and 12) at which  $p_1(q_n | \Delta t_{n-1})$  ceases to depend upon  $\Delta t_{n-1}$ , i.e., where  $M_1(\Delta t_{\ell}, \Delta t_{n-1}) = 0 \text{ for all } \Delta t_{n-1} > \Delta t_{\ell}.$ 

The interdependence of  $p_0(q_n)$  and  $p_0(\Delta t_n)$  was verified by using the measured  $p_1(q_n|\Delta t_{n-1})$  distributions together with the measured  $p_0(\Delta t_n)$  in Eq. (10) to compute  $p_0(q_n)$ . The results of these calculations, indicated by

 $N_2 + 20\% O_2$ 

1.2



FIG. 9. Measured unconditional  $p_0(q_n)$  and corresponding conditional  $p_1(q_n|\Delta t_{n-1})$  distributions at 9.0 kV for the indicated values of  $\Delta t_{n-1}$  which apply to relatively weak uv irradiation of the cathode in a 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> gas mixture. The distributions have been normalized to the maximum values. The solid lines are Gaussian representations of the  $p_1$  data which were used together with corresponding data on  $p_0(\Delta t_n)$  in Eq. (10) to give the predicted  $p_0(q_n)$  distribution indicated by the dashed line.



FIG. 10. Computed correlation coefficients for the random variables  $q_n$  and  $\Delta t_{n-1}$  as functions of discharge gap voltage  $(V_a)$  at two different indicated levels of relative uv intensity. Data for the measured  $p_0(\Delta t_n)$ ,  $p_0(q_n)$ , and  $p_1(q_n|\Delta t_{n-1})$  distributions similar to those shown in Figs. 8 and 9 were used in Eq. (4) to compute  $R_{q_n,\Delta t_{n-1}}$ .

the dashed lines in Figs. 8 and 9, are seen to agree satisfactorily with the measured  $p_0(q_n)$ , thereby demonstrating that the measured distributions are self-consistent and the profile of  $p_0(q_n)$  is determined by the profile of  $p_0(\Delta t_n)$  and the dependence of  $q_n$  on  $\Delta t_{n-1}$ .

When computing the integral in Eq. (10), Gaussian representations of the  $p_1$  distributions shown in Figs. 9 and 10 were used as indicated by the solid lines. However, as shown by data for Ne+5% O<sub>2</sub> given in Sec. IV D, the  $p_1(q_n|\Delta t_{n-1})$  are generally non-Gaussian. In the case of Ne+5% O<sub>2</sub>, the measured  $p_1(q_n|\Delta t_{n-1})$  can often be satisfactorily represented by the sum of two Gaussians given by

$$p_{1}(q_{n}|\Delta t_{n-1}) = \left[ (2\pi)^{1/2} (\sigma_{1} + c\sigma_{2}) \right]^{-1} \\ \times \left\{ \exp \left[ -(q_{n} - Q_{1})^{2} / 2\sigma_{1}^{2} \right] \\ + c \exp \left[ -(q_{n} - Q_{2})^{2} / 2\sigma_{2}^{2} \right] \right\},$$
(38)

where  $c, \sigma_i$ , and  $Q_i$  (i = 1, 2) are dependent on  $\Delta t_{n-1}$ and obtained from fits to the data. An example of the  $\Delta t_{n-1}$  dependences of the  $Q_i$  parameters is shown in Fig. 11 for results from a Ne+5% O<sub>2</sub> gas mixture together with corresponding data on  $p_0(q_n)$  and  $p_0(\Delta t_n)$ . The result of evaluating the integral in Eq. (10) using the  $p_0(\Delta t_n)$  shown in Fig. 11 and the above representation of  $p_1(q_n | \Delta t_{n-1})$  are indicated by the dashed line and seen to be in satisfactory agreement with the measured  $p_0(q_n)$ . Indicated in this figure is the critical time  $\Delta t_\ell$ above which the parameters in Eq. (38) cease to depend on  $\Delta t_{n-1}$  and  $Q_1 = Q_2$  so that  $p_1(q_n | \Delta t_{n-1})$  becomes adequately represented by a single Gaussian which is in-



FIG. 11. Measured unconditional pulse-amplitude  $p_0(q_n)$ and corresponding pulse-time-separation  $p_0(\Delta t_n)$  distributions at 4.8 kV for relatively weak uv irradiation of the cathode in a 100-kPa Ne+5% O<sub>2</sub> gas mixture. Also shown are the  $\Delta t_n$  dependencies of the  $Q_i$ 's used in the double-Gaussian representation of the corresponding measured  $p_1(q_n|\Delta t_{n-1})$ distributions as given by Eq. (38) with the indicated  $Q_i$ 's. The values for  $q_{\lim}$  and  $\Delta t_\ell$  are marked with vertical arrows, and the dashed line represents  $p_0(q_n)$  predicted from Eq. (10).

dependent of  $\Delta t_{n-1}$ .

The dependence of  $q_n$  on  $\Delta t_{n-1}$  implied by  $p_1(q_n | \Delta t_{n-1})$  is that expected from the influence of the moving negative-ion space charge in the electrode gap as proposed independently by Lama and Gallo<sup>6</sup> and Bugge.<sup>8</sup> It is shown by these authors that several negative-ion space-charge clouds from previous discharge pulses can be present in the gap at the time a discharge pulse develops (see Fig. 14 of Ref. 6). The effect of negative-ion clouds anywhere in the gap is to reduce the electric-field magnitude at the cathode below the value it would have in the absence of such a cloud. This field reduction inhibits the growth of a discharge pulse. The greater the distance of the space-charge cloud from the cathode, the greater will be the field at the cathode and consequently the larger will be the magnitude of a discharge that develops at that time. As time elapses after the most recent discharge event, the space charge moves further from the point cathode thereby increasing the field in which the next pulse develops. The positive dependence of  $q_n$  on  $\Delta t_{n-1}$  can therefore be expected to hold until the critical time  $\Delta t_{\ell}$  is reached at which all the ions have arrived at



FIG. 12. Plot of  $q_{\max} \simeq \langle q_n(\Delta t_{n-1}) \rangle$  vs  $\Delta t_{n-1}$  from  $p_1(q_n|\Delta t_{n-1})$  data at the indicated applied gap voltages in the case of 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> gas mixture. The vertical arrows correspond to  $\Delta t_\ell$  values estimated using Eqs. (40) and (41).

the anode. For  $\Delta t_{n-1} > \Delta t_{\ell}$ , the field at the cathode is restored to its unperturbed value and  $q_n \rightarrow q_{\lim}$ .

The time  $\Delta t_{\ell}$  can be estimated using<sup>6,8</sup>

$$\Delta t_{\ell} \simeq \frac{1}{\mu} \int_0^{z_{\ell}} dz [E(z)]^{-1}, \qquad (39)$$

where  $\mu$  is the negative-ion mobility,  $z_d$  is the point-toplane gap spacing, and E(z) is the magnitude of the electric field on the point-to-plane axis for distance z measured from the cathode. A reasonable estimate<sup>6</sup> for E(z)is given by

$$E(z) \simeq \frac{2V_a z_d}{\ln\left(4z_d/r_c\right)} g(z) , \qquad (40)$$

$$g(z) = \left[z_d \left(2z + r_c\right) - z^2\right]^{-1},$$
(41)

where  $r_c$  is the cathode tip radius. This estimate neglects the ion space-charge distortion of the field, since it has been shown<sup>6</sup> that this distortion has a minor effect on ion-drift times under conditions similar to those considered here.

Figure 12 shows plots of  $q_{n,\max} [\simeq \langle q_n(\Delta t_{n-1}) \rangle]$  for  $p_1(q_n|\Delta t_{n-1})$  versus  $\Delta t_{n-1}$  for different  $V_a$  and for a  $N_2 + 20\% O_2$  mixture with gap conditions like those used to obtain the data shown in Fig. 4. The dash-dotted lines correspond to the values for  $q_{\lim}$  and the vertical arrows correspond to computed  $\Delta t_\ell$  values obtained from Eqs. (39)-(41) for the appropriate gap geometry and assuming a reasonable<sup>6,40</sup> value for  $\mu$  of  $2.7 \text{ cm}^2/\text{ Vs}$ . The computed  $\Delta t_\ell$  are seen to correspond approximately to the  $\Delta t_{n-1}$  values at which the  $q_{n,\max}$  versus  $\Delta t_{n-1}$  curves in Fig. 12 level off toward  $q_{\lim}$ . This is also consistent with the behavior of  $p_1(q_n|\Delta t_{n-1})$  implied by the data in Fig. 11 for Ne+5% O\_2. The  $\Delta t_\ell$  values agree with

the ion transit times estimated for smaller gaps in air by Lama and Gallo.<sup>6</sup> The implied dependence of  $\Delta t_{\ell}$  on  $z_d$ was evident from measurements made in air using gap spacings from 0.2 to 10.0 cm. However, due to the waning influence of ion space charge with increasing distance from the cathode,  $\Delta t_{\ell}$  becomes less clearly defined as  $z_d$ increases.

The strong dependence of  $q_n$  on  $\Delta t_{n-1}$  results from the sensitivity of discharge pulse growth to small changes in the electric field near the cathode. This sensitivity, as noted by Lama and Gallo,<sup>6</sup> is due to the exponential dependence on field of the ionization processes that control discharge growth (also see Sec. IV E). The strong positive dependence of  $q_n$  on  $\Delta t_{n-1}$  appears to be a general characteristic of the Trichel-pulse phenomenon that applies over a wide range of conditions.<sup>28-30</sup>

#### C. Behavior of $p_1(\Delta t_n | q_n)$ and $p_1(\Delta t_n | \Delta t_{n-1})$

The  $p_0(\Delta t_n)$  data for Ne+5% O<sub>2</sub> shown in Fig. 11 exhibit a behavior for  $\Delta t_n > \Delta t_{n,\max}$  that is not consistent with Eq. (36). This suggests that the initiation of a discharge pulse is influenced not only by photoelectron emission via uv irradiation of the cathode but also

by self-excitation or retardation processes whereby residuals from earlier pulses serve to either enhance or inhibit the rate of electron release at the cathode. The extent to which self-excitation or retardation processes contribute is revealed from measured conditional timeseparation distributions  $p_1(\Delta t_n|q_n)$  and  $p_1(\Delta t_n|\Delta t_{n-1})$ .

Shown in Figs. 13 and 14 are data from  $p_1(\Delta t_n|q_n)$ and  $p_1(\Delta t_n | \Delta t_{n-1})$  measurements, respectively, in  $Ne+5\%O_2$  and  $N_2 + 20\%O_2$  gas mixtures for a relatively high level of uv irradiation comparable to that for the  $I_1$  case shown in Fig. 5. The conditional distributions are compared with the corresponding unconditional distributions  $p_0(\Delta t_n)$  indicated by the dashed lines in both figures. Correlations between the pairs  $(\Delta t_n, q_n)$ and  $(\Delta t_n, \Delta t_{n-1})$  for both mixtures are indicated by the lack of equality between the conditional and unconditional distributions. The correlations are strongest in the case of Ne+5%  $O_2$  for which the dependence of  $\Delta t_n$ on  $q_n$  is positive, i.e.,  $q_n \uparrow \Rightarrow \Delta t_n \uparrow$ . Consistent with this behavior, it is seen that  $\Delta t_{n-1} \uparrow \Rightarrow \Delta t_n \uparrow$  as required by the arguments in Sec. II D. The signs of these dependences for the  $N_2 + 20\% O_2$  mixture are opposite to those for the Ne+5% O<sub>2</sub> mixture, i.e.,  $q_n \uparrow \Rightarrow \Delta t_n \downarrow$ and  $\Delta t_{n-1} \uparrow \Rightarrow \Delta t_n \downarrow$ . In both cases, the arguments



FIG. 13. Unconditional  $[p_0(\Delta t_n)]$  and conditional  $[p_1(\Delta t_n | \Delta t_{n-1})]$  and  $p_1(\Delta t_n | q_n)$  time-separation distributions at the indicated  $\Delta t_{n-1}$  and  $q_n$  values for a 100-kPa Ne+5% O<sub>2</sub> mixture ( $V_a = 4.1 \text{ kV}$ ). The distributions have been normalized to the maxima.



FIG. 14. Unconditional  $[p_0(\Delta t_n)]$  and conditional  $[p_1(\Delta t_n | \Delta t_{n-1})]$  and  $p_1(\Delta t_n | q_n)$  time-separation distributions at the indicated  $\Delta t_{n-1}$  and  $q_n$  values for a 100-kPa N<sub>2</sub> + 20% O<sub>2</sub> mixture ( $V_a = 9.1 \text{ kV}$ ). The distributions have been normalized to the maxima.

in Sec. II D hold because the relatively high level of uv irradiation ensures that there is the required strong positive dependence of  $q_n$  on  $\Delta t_{n-1}$  over the entire range of time separations defined by  $p_0(\Delta t_n)$  (see Fig. 10). Nevertheless, as shown by data in Sec. IV D, the relatively strong positive dependences between the pairs  $(\Delta t_n, q_n)$ and  $(\Delta t_n, \Delta t_{n-1})$  can persist even for weaker uv irradiation. A negative dependence of  $\Delta t_n$  on  $\Delta t_{n-1}$  was previously reported by Mason and Young<sup>35</sup> for negativecorona pulses in air which is comparable to the N<sub>2</sub>-O<sub>2</sub> case considered here.

One can only speculate at this time about the mechanisms responsible for the two different types of correlations implied by the results shown in Figs. 13 and 14. A careful investigation into the particular gap conditions that affect the observed behaviors of  $p_1(\Delta t_n | q_n)$ and  $p_1(\Delta t_n | \Delta t_{n-1})$  has not yet been undertaken. Measurements performed for varying [Ne]/[O<sub>2</sub>] ratios appear to indicate that the transition from positive to negative dependences occurs as the relative  $O_2$  content in the gas increases. In Ne+95%  $O_2$  and pure  $O_2$ , the behavior is more like that found for the  $N_2 + 20\% O_2$  case. Thus, although it appears that gas composition is relevant, it should be noted that in order to sustain the discharge, it is necessary to increase the electrode-gap voltage  $V_a$  as the  $O_2$  content increases. This corresponding increase in  $V_a$  suggests that the electric-field intensity at the cathode may also be a relevant factor.

The behavior found for the Ne+5% O<sub>2</sub> case is that expected if the presence of ion space charge from the previous pulse acts to suppress the release of electrons from the cathode. This expectation is reasonable if electron release is primarily due to a field-assisted photoelectric effect<sup>38</sup> so that reductions in the field at the cathode by moving space charge serve to inhibit electron emission. An increase in  $q_n$  corresponds necessarily to an increase in the local negative-ion space-charge density, which in turn reduces the rate of electron release and correspondingly the probability for initiating the next pulse. The behavior of  $p_1(\Delta t_n | q_n)$  indicated in Fig. 13 suggests that negative-ion space charge may reduce the cathode field enough to nearly turn off the electron release process for a significant interval of time following a "large" pulse.

The negative dependence of  $\Delta t_n$  on  $q_n$  found for  $N_2 + 20\% O_2$  indicates that the probability for initiating a discharge pulse increases as the size of the previous pulse increases. This is possible if residual positive ions or metastable species from the previous pulse contribute to the release of initiatory electrons from the cathode. Because of the relatively long minimum times between pulses (greater than 50 µs), it is likely that metastable states have a more significant effect since positive-ion clearing times near the cathode should be comparable to  $\Delta t_c$  as noted by Lama and Gallo<sup>6</sup> and Bugge.<sup>8</sup>

It was not possible to identify the role of specific metastable states that could yield initiatory electrons when quenched at the cathode surface. The  $a^1\Delta_g$  and  $b^1\Sigma_q^+$  metastable states of O<sub>2</sub> can be produced in the

discharge and are not readily quenched by collisions with either nitrogen or oxygen molecules.<sup>41-44</sup> The  $A^{3}\Sigma_{u}^{+}$ state of N<sub>2</sub> has a long radiative lifetime (1-2 s), but is more readily quenched<sup>45,46</sup> by collisions than the O<sub>2</sub> metastable states and may therefore have relatively less influence on secondary-electron release at the cathode under conditions of high gas pressure considered here. Its contribution to secondary-electron release has, nevertheless, been observed in low-pressure discharges.<sup>47,48</sup> In the Ne-O<sub>2</sub> mixtures, the role of neon metastable states should also be considered, although the rates for collisional quenching of these metastable states in O<sub>2</sub> appear to be about two orders of magnitude higher than the rate for quenching the nitrogen  $A^{3}\Sigma_{u}^{+}$  state.<sup>49</sup>

For quenching of the O<sub>2</sub>  $a^{1}\Delta_{g}$  state to be a source of secondary electrons at the cathode, a sufficiently intense electric field must be present to reduce the effective work function<sup>40</sup> of the stainless steel by at least 3 eV below that which it would have in the absence of a field.<sup>50,51</sup> This might explain why the negative dependence of  $\Delta t_n$  on  $q_n$  appears for mixtures with relatively high  $O_2$  concentrations for which higher voltages are needed to sustain the discharge. If field strengths are high enough for a field-assisted photoelectric effect, then field-assisted electron ejection by quenching of metastable states at the cathode surface would appear to be energetically possible. However, direct experimental confirmation of such a quenching process has yet to be reported. The presence of a high field could also influence the quenching of other more energetic metastable states such as the  ${}^{1}S$  and  ${}^{1}D$ metastable states of atomic oxygen. It was found that even for the Ne+5% O<sub>2</sub> mixture, the sign of the  $(\Delta t_n, q_n)$ dependence becomes negative at higher  $V_a$  at which the discharge is largely self-sustained.

It should be noted that the space-charge effect that gives a positive dependence of  $\Delta t_n$  on  $q_n$  is operative independent of whether or not metastable states contribute to discharge-pulse initiation. When the negative  $(\Delta t_n, q_n)$  dependence occurs, the effect of the metastables evidently overcomes that of space charge.

### D. Behavior of $p_2(q_n|\Delta t_{n-1}, q_{n-1})$ and $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$

From the results presented here on  $p_2(q_n|\Delta t_{n-1}, q_{n-1})$ and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$ , it will be seen that relatively strong dependences of  $q_n$  on  $q_{n-1}$  and on  $\Delta t_{n-2}$  are possible for fixed  $\Delta t_{n-1}$  which depend on the gas mixture used. Examples of data for the distributions  $p_0(q_n)$ ,  $p_1(q_n|\Delta t_{n-1})$ , and  $p_2(q_n|\Delta t_{n-1}, q_{n-1})$  obtained under the same conditions for a Ne+5% O<sub>2</sub> gas mixture are shown in Fig. 15. The gap conditions are similar to those that apply for the results shown in Fig. 11.

It is seen from these data that  $p_2(q_n | \Delta t_{n-1}, q_{n-1}) \neq p_1(q_n | \Delta t_{n-1})$ , and the expectation value  $\langle q_n(q_{n-1}, \Delta t_{n-1}) \rangle$ , given by

$$\langle q_n (q_{n-1}, \Delta t_{n-1}) \rangle = \int_0^\infty q_n p_2 (q_n | \Delta t_{n-1}, q_{n-1}) \, dq_n,$$
(42)

increases for decreasing  $q_{n-1}$  at the indicated fixed values for  $\Delta t_{n-1}$  of 177, 197, and 217  $\mu$ s. Since the widths of the  $p_2$  distributions are much smaller than the widths of the corresponding  $p_1$  distributions, it can be argued that there is a strong dependence of  $q_n$  on  $q_{n-1}$  which applies at these  $\Delta t_{n-1}$  values. Evidence for a negative  $(q_n, q_{n-1})$  dependence was seen previously in the work of Cross and co-workers for Trichel pulses in low-pressure  $O_2$  (see Fig. 12 of Ref. 13).

The dash-dotted lines in Fig. 15 represent Gaussian fits to the  $p_2$  distributions which were used together with the data shown for  $p_0(q_n)$  and a parametrized form for  $p_1(\Delta t_n|q_n)$  in Eq. (14) to obtain computed  $p_1(q_n|\Delta t_{n-1})$  indicated by the dashed lines. Under the conditions that apply for the data in Fig. 15, it was found that, in the approximate range  $150 \,\mu s \lesssim \Delta t_{n-1}$  $\lesssim 300 \,\mu s, \, p_1(\Delta t_{n-1}|q_{n-1})$  takes the form

$$p_1(\Delta t_{n-1}|q_{n-1}) = A_q \exp\left[-\omega_q \left(q_{n-1} - \beta_q\right)^2\right], \quad (43)$$

where  $\omega_q$  and  $\beta_q$  are constants that satisfy the requirements  $\omega_q > 0$  and  $\beta_q \gtrsim q_{\lim}$ . The constant  $A_q$  is necessary for proper normalization, but becomes irrelevant in Eq. (14) if  $p_1(q_n|\Delta t_{n-1})$  is normalized to the maximum value as in Fig. 15. The factor  $p_0(\Delta t_{n-1})^{-1}$  in Eq. (14) is also irrelevant for the same reason. The values for  $\omega_q$  and  $\beta_q$  that were used to obtain the dashed lines in Fig. 15

TABLE II. Parameter values for  $p_1(\Delta t_{n-1}|q_{n-1})$ [Eq. (43)] used in Eq. (14) to obtain the dashed-line representations for  $p_1(q_n|\Delta t_{n-1})$  in Fig. 15 at the indicated  $\Delta t_{n-1}$ values.

$\overline{\Delta t_{n-1}}$ (µs)	$\omega_q \ (pC^{-2})$	$\beta_q \ (pC)$
177	$0.823 \times 10^{-4}$	350
197	$1.18 \times 10^{-4}$	336
217	$1.22 \times 10^{-4}$	355

at the indicated time separations  $\Delta t_{n-1}$  are listed in Table II. In this case it is seen that the  $\beta_q$  are all slightly larger than the corresponding  $q_{\text{lim}}$  value of 320 pC given by the  $p_0(q_n)$  data in Fig. 15.

The approximate form for  $p_1(\Delta t_{n-1}|q_{n-1})$  given by Eq. (43) is consistent with experimentally observed trends, as indicated by data in Fig. 16, which apply to conditions similar but not identical to those for Fig. 15. For the conditions that apply to Fig. 16, it is evident that Eq. (43) becomes a reasonable representation for  $p_1(\Delta t_{n-1}|q_{n-1})$  when  $\Delta t_{n-1}$  is somewhat greater than  $170 \,\mu$ s. In the  $\Delta t_{n-1}$  range 150–170  $\mu$ s,  $p_1(\Delta t_{n-1}|q_{n-1})$ shows much less variation with  $q_{n-1}$  than implied by Eq. (43), and for sufficiently low  $\Delta t_{n-1}$ ,  $p_1(\Delta t_{n-1}|q_{n-1})$ 



FIG. 15. Measured  $p_0(q_n)$ ,  $p_1(q_n|\Delta t_{n-1})$ , and  $p_2(q_n|\Delta t_{n-1})$  distributions for the indicated values for  $\Delta t_{n-1}$  and  $q_{n-1}$  at  $V_a = 4.8 \,\mathrm{kV}$  in a 100-kPa Ne+5% O<sub>2</sub> mixture. The dash-dotted lines are Gaussian representations of the  $p_2$  data used together with the  $p_0(q_n)$  data and  $p_1(\Delta t_{n-1}|q_{n-1})$  given by Eq. (43) (also see Table II) in Eq. (14) to obtain the predicted  $p_1(q_n|\Delta t_{n-1})$  indicated by the dashed lines at 177, 197, and 217  $\mu$ s.



FIG. 16. Conditional time-separation distribution  $p_1(\Delta t_n|q_n)$  plotted vs  $q_n$  for the indicated  $\Delta t_{n-1}$  values (top) and vs  $\Delta t_n$  for the indicated  $q_n$  values (bottom) for conditions similar to those that apply to Fig. 15.

decreases rapidly with  $q_{n-1}$  as indicated by the 140  $\mu$ s curve in Fig. 16. The general trends implied by the data in this figure apply over a wide range of conditions. It should be noted that the variations in the shapes of the  $p_1(q_n|\Delta t_{n-1})$  distributions with  $\Delta t_{n-1}$  can be accounted for by the variations in the behavior of  $p_1(\Delta t_{n-1}|q_{n-1})$ implied by the results in Fig. 16. If  $p_1(\Delta t_{n-1}|q_{n-1})$  is assumed to be constant (independent of  $q_{n-1}$ ) as is approximately true for  $\Delta t_{n-1}$  in a narrow range around 150  $\mu$ s, then the  $p_1(q_n|\Delta t_{n-1})$  assume shapes similar to that indicated by the 147  $\mu$ s data in Fig. 15. In this case the shape is a compressed, inverse reflection of the  $p_0(q_n)$ distribution due to the negative  $(q_n, q_{n-1})$  dependence. Thus variations in the  $p_1(q_n|\Delta t_{n-1})$  profile of the type seen here imply a significant dependence of  $\Delta t_n$  on  $q_n$ .

Figure 17 shows results for the  $p_0(\Delta t_n)$ ,  $p_1(q_n|\Delta t_{n-1})$ , and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$  distributions obtained for a condition similar to that which applies to the data in Fig. 15. From the data on  $p_2$  it is seen that there is a negative dependence of  $q_n$  on  $\Delta t_{n-2}$  as required by the corresponding negative  $(q_n, q_{n-1})$  dependence and the strong positive  $(q_n, \Delta t_{n-1})$  dependence (see Sec. II E.). Moreover, the correlation between  $q_n$  and  $\Delta t_{n-2}$  at the indicated  $\Delta t_{n-1}$  values appears to satisfy the strongdependency condition.

The dashed curves at 177, 197, and  $217 \,\mu s$  are the results of performing the integral of Eq. (18) in which Gaussian representations of the  $p_2$  distributions shown



FIG. 17. Measured  $p_0(\Delta t_n)$ ,  $p_1(q_n|\Delta t_{n-1})$ , and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$  distributions for the indicated values of  $\Delta t_{n-1}$  and  $\Delta t_{n-2}$  at  $V_a = 4.8 \,\mathrm{kV}$  in a 100-kPa Ne+5% O<sub>2</sub> mixture. The dash-dotted lines on Gaussian representations of the  $p_2$  data used together with the  $p_0(\Delta t_n)$  data and  $p_1(\Delta t_{n-1}|\Delta t_n)$  given by Eqs. (44) and (45) (also see Table III) in Eq. (18) to obtain the predicted  $p_1(q_n|\Delta t_{n-1})$  indicated by the dashed lines at 177, 197, and 217  $\mu$ s.



FIG. 18. Conditional time-separation distribution  $p_1(\Delta t_n | \Delta t_{n-1})$  plotted vs  $\Delta t_{n-1}$  for the indicated  $\Delta t_n$  values (top) and vs  $\Delta t_n$  for the indicated  $\Delta t_{n-1}$  values (bottom) for conditions similar to those that apply to Fig. 16.

by the dash-dotted lines and the numerical data shown for  $p_0(\Delta t_n)$  were used together with a parametrized form for  $p_1(\Delta t_{n-1}|\Delta t_{n-2})$ . In this case the conditional timeseparation distribution was given the form

$$p_1\left(\Delta t_{n-1}|\Delta t_{n-2}\right) = A_t \exp\left[-\omega_t \left(\beta_t - \Delta t_{n-2}\right)^2\right]$$
(44)

for  $\Delta t_{n-2} \leq \beta_t$ , and

$$p_1\left(\Delta t_{n-1} | \Delta t_{n-2}\right) = A_t \tag{45}$$

for  $\Delta t_{n-2} > \beta_t$ .

This is the approximate form suggested by the  $p_1(\Delta t_n | \Delta t_{n-1})$  data shown in Fig. 18. When  $\Delta t_{n-2}$  exceeds about 180  $\mu$ s, the parameter  $\beta_t$  approaches the value of  $\Delta t_\ell$ , which for the data in Fig. 17 is about 410  $\mu$ s. The values of the parameters used in Eq. (44) to obtain the calculated  $p_1(q_n | \Delta t_{n-1})$  in Fig. 17 are given in Table III.

The observed negative dependence of  $q_n$  on  $q_{n-1}$  (and correspondingly of  $q_n$  on  $\Delta t_{n-2}$ ) in the Ne+5% O<sub>2</sub> gas mixture can be qualitatively accounted for by the effect of the negative-ion space charge from the previous discharge pulse on suppressing the *growth* of the subsequent pulse. In this case it is the *size* of the space-charge cloud

TABLE III. Parameter values for  $p_1(\Delta t_{n-1}|\Delta t_{n-2})$ [Eqs. (44) and (45)] used in Eq. (18) to obtain the dashed-line representations for  $p_1(q_n|\Delta t_{n-1})$  in Fig. 17 at the indicated  $\Delta t_{n-1}$  values.

$\overline{\Delta t_{n-1}  (\mu \mathrm{s})}$	$\omega_t \; (\mu \mathrm{s}^{-2})$	$\beta_t \ (\mu s)$
177	$1.54 \times 10^{-4}$	280
197	$0.503 \times 10^{-4}$	400
217	$0.516 \times 10^{-4}$	422

rather than its location that is relevant. The larger the previous pulse, the larger its resulting negative-ion cloud and, consequently, the greater the reduction of the field at the cathode for any given  $\Delta t_{n-1} < \Delta t_{\ell}$ . This field reduction serves to inhibit the growth of the next pulse; thus larger pulses will preferentially be followed, on the average, by smaller pulses and vice versa.

It was observed that in some gas mixtures with higher  $O_2$  content the dependence of  $q_n$  on  $q_{n-1}$  reverses sign and becomes positive. This was demonstrated by results published previously for the  $N_2 + 20\% O_2$  mixture (see Fig. 7 of Ref. 27). Shown in Fig. 19 are examples of data on  $p_2(q_n | \Delta t_{n-1}, q_{n-1})$  distributions for a Ne+95% O<sub>2</sub> mixture at different indicated gap voltages that clearly indicate a positive  $(q_n, q_{n-1})$  dependence for sufficiently small  $\Delta t_{n-1}$ . The positive dependences are weaker than the negative dependences found for Ne+5%  $O_2$  shown by the data in Fig. 15. The dependence of  $q_n$  on  $q_{n-1}$  begins to disappear for  $\Delta t_{n-1} \stackrel{>}{\sim} 75 \,\mu$ s where  $p_2(q_n | \Delta t_{n-1}, q_{n-1})$ ceases to depend on  $q_{n-1}$ . In the case of N<sub>2</sub> + 20% O<sub>2</sub> mixtures, the positive dependence of  $q_n$  on  $q_{n-1}$  was found<sup>27</sup> to persist even for  $\Delta t_{n-1}$  in excess of 100  $\mu$ s. It should be noted incidently from Fig. 19 that even the  $p_2(q_n | \Delta t_{n-1}, q_{n-1})$  distributions may exhibit distinctly non-Gaussian shapes.

The appearance of a positive  $(q_n, q_{n-1})$  dependence often coincides with a negative dependence of  $\Delta t_n$  on  $q_n$ as illustrated by the data in Fig. 14 for N<sub>2</sub> + 20% O<sub>2</sub>. It was suggested in Sec. IV C that the negative  $(\Delta t_n, q_n)$ dependence may be a manifestation of the influence of metastable species on electron release from the cathode. The occurrence of the positive  $(q_n, q_{n-1})$  dependence suggests that the metastable densities produced by earlier discharge pulses are high enough to significantly increase the effective ionization coefficient of the gas in the immediate vicinity of the cathode at the time that the next pulse develops. An increase in the local ionization coefficient enhances discharge growth.

As shown by Van Brunt<sup>52</sup> from an extension of Legler's<sup>53</sup> analysis, the probability that the initial electron avalanche consists of  $n_e$  electrons after growing a distance z from the cathode on the point-to-plane axis is given to a good approximation by

$$p_{1}(n_{e}|z) dz = \frac{1}{w(z)^{2}} \exp[u(z)] \exp[n_{e}/w(z)] dz,$$
(46)

where

$$u(z) \equiv \int_0^z \left[ \alpha_i(z') - \eta_a(z') \right] dz' \tag{47}$$

and

$$w(z) \equiv 1 + \exp\left[u(z)\right] \int_0^z \alpha_i(z') \exp\left[-u(z')\right] dz'.$$
(48)

In Eqs. (46)-(48),  $\alpha_i$  and  $\eta_a$  are, respectively, the ionization and attachment coefficients of the gas which depend on the electric-field-to-gas density ratio E(z)/N and therefore on z. The middle term in Eq. (46) indicates an exponential dependence on  $(\alpha_i - \eta_a)$ . Thus even relatively small changes in  $\alpha_i$  due to either changes in E(z)or presence of metastables could have disproportionately large effects on the electron-avalanche growth in the ini-



FIG. 19. Measured  $p_2(q_n|\Delta t_{n-1}, q_{n-1})$  distributions at the indicated  $q_{n-1}$  and  $\Delta t_{n-1}$  values for different applied gap voltages in a 100-kPa Ne+95% O<sub>2</sub> mixture. The distributions have been normalized to the maxima.

tial stages of Trichel-pulse formation. It is also this factor that comes into play in accounting for the strong dependence of  $q_n$  on  $\Delta t_{n-1}$ .

To determine the plausibility of the suggested influence of metastable species on local ionization-coefficient enhancement near the cathode, estimates were made of the resident times for metastable states in the active region of the cathode defined by the Geballe-Reeves breakdown criterion,<sup>54</sup> namely that region where  $\alpha_i > \eta_a$ . The spatial extent of this region for the case of unperturbed fields was estimated using Eqs. (40) and (41) and the recommended values<sup>55</sup> for  $\alpha_i$  and  $\eta_a$  that apply to air and therefore to the N<sub>2</sub> + 20% O<sub>2</sub> mixture. The active region was assumed to be approximately<sup>52</sup> spherical of radius  $R_{\rm crit}$ . The value for  $R_{\rm crit}$  corresponds to  $z_c/2$ , where  $E(z_c)$  is the axial field strength at which  $\alpha_i = \eta_a$ . Note that Eq. (46) requires that the initial electron-avalanche growth cease if  $z > z_c$ .

The rate equation for neutral metastable-species transport in the gas  $is^{56}$ 

$$\frac{\partial \rho(\mathbf{r},t)}{\partial t} = D\nabla^2 \rho(\mathbf{r},t) - k_m N \rho(\mathbf{r},t) - (1/\tau_m) \rho(\mathbf{r},t).$$
(49)

Here  $\rho(\mathbf{r}, t)$  is the metastable density at position  $\mathbf{r}$  and time t, D is the metastable diffusion coefficient,  $k_m$  is the rate coefficient for collisional quenching, and  $\tau_m$  is the radiative life time. Only metastable states for which  $\tau_m > \Delta t_c$  are considered here; thus the last term on the right-hand side of Eq. (49) can be neglected. Assuming spherical symmetry, Eq. (49) can be solved to give the fraction F(t) of metastable states remaining in the volume  $V_{\rm crit}$  defined by  $r \geq R_{\rm crit}$  at a time t after cessation of a discharge pulse. Consistent with the experimental conditions, the diameter of the discharge vessel is assumed to be much larger than  $R_{\rm crit}$  so that wall effects can be neglected.

If the discharge pulse leaves a uniformly distributed metastable density  $\rho_0$  that is confined to  $V_{\rm crit}$ , then it can be shown from the solution of Eq. (49) that

$$F(t) = \int_0^{R_{crit}} r^2 \rho(r,t) dr \bigg/ \int_0^\infty r^2 \rho(r,t) dr,$$
(50)

where

$$\rho(r,t) = \frac{\frac{3}{2}\rho_0 \exp\left(-k_m Nt\right)}{\left(4\pi\right)^{3/2} \left(Dt\right)^{1/2} R_{\rm crit}^3 r} \int_0^{R_{\rm crit}} r' \left[\exp\left(\frac{-r^2 - r'^2 + 2rr'}{4Dt}\right) - \exp\left(\frac{-r^2 - r'^2 - 2rr'}{4Dt}\right)\right] dr'.$$
(51)

If the metastable species are to be effective in modifying  $\alpha_i$  at the time of the next pulse, a significant fraction must remain in  $V_{\text{crit}}$  for  $\Delta t_{n-1} > \Delta t_c$ . The collisionalquenching rate must therefore satisfy the condition

$$k_m < \left(N\Delta t_c\right)^{-1}.\tag{52}$$

For  $\Delta t_c$  in the range 10-300  $\mu$ s at atmospheric pressure (see Fig. 6), Eq. (52) requires that  $k_m$  lie above the corresponding values  $4.0 \times 10^{-15}$  to  $1.4 \times 10^{-16}$  cm<sup>3</sup>/s. This condition is satisfied by the  $a^1\Delta_g$  and  $b^1\Sigma_g^+$  states of oxygen,<sup>41-44</sup> but not by the metastable states of either nitrogen<sup>45,46</sup> or neon.<sup>49</sup> In the case of the oxygen metastable states,  $k_m << (N\Delta t_c)^{-1}$  and therefore  $\exp(-k_m Nt) \simeq 1$  in Eq. (51). This approximation may also apply to long-lived, vibrationally excited N<sub>2</sub>.

Results of computing F(t) in the case where collisional quenching can be neglected are shown in Fig. 20. It is seen from this figure that, independent of the assumed value for D in the range  $0.5 \times 10^{-7}$  to  $5.0 \times 10^{-7}$  cm<sup>2</sup>/s, a significant fraction of the metastable species is present in  $V_{\rm crit}$  for times up to 100  $\mu$ s after cessation of the discharge pulse. Thus it can be concluded that metastable species generated in a discharge pulse remain near the cathode for times long enough to influence development of the next pulse.

The extent to which metastable species influence discharge growth depends on  $\rho_0$ , i.e., on the number produced in  $V_{\text{crit}}$ . It should be realized, however, that the effect of metastable species on the gas ionization coefficient can be disproportionately larger than implied by their relative densities. This is due to the fact that metastable species have a significantly lower ionization threshold than the corresponding neutral molecules in their ground state. The local ionization coefficient in the gas at any time t is given by<sup>57,58</sup>



FIG. 20. Calculated fraction of metastable states remaining in the critical volume  $V_{\rm crit}$  near the cathode at a time  $\Delta t$  following cessation of a discharge pulse for different assumed values of the diffusion coefficient.

$$\alpha_{i}(\mathbf{r},t) = (2m_{e})^{1/2} (NW_{m})^{-1} \times \left(\sum_{j} \rho_{j} K_{j} + \sum_{k} \rho_{k}(\mathbf{r},t) K_{k}\right), \quad (53)$$

where

$$K_{\ell} = \int_{\epsilon_{\ell}} \epsilon f_m(\mathbf{r}, t, \epsilon) \sigma_{I_{\ell}}(\epsilon) \, d\epsilon, \ \ell = j \text{ or } k.$$
(54)

In Eqs. (53) and (54)  $m_e$  is the electron mass,  $N(=\sum_{\ell} \rho_{\ell})$  is the total particle density,  $W_m$  is the electron drift velocity,  $\rho_j$  is the local density of the *j*th neutral species in the ground state,  $\rho_k$  is the density of the *k*th metastable species,  $\epsilon$  is the electron translational kinetic energy,  $\epsilon_{\ell}$  is the ionization threshold energy for the  $\ell$ th species,  $\sigma_{I_{\ell}}(\epsilon)$  is the electron-impact ionization cross section for the  $\ell$ th species, and  $f_m$  is the kinetic-energy distribution of the electrons. In general,  $N, W_m, \rho_j, \rho_k$ , and  $f_m$  depend on **r** and t. However, if  $N \gg \rho_k$  for any k, then the dependence of  $\alpha_i$  on t is primarily that due to  $\rho_k(\mathbf{r}, t)$ . The spatial dependence of  $\alpha_i$  is governed by the field and the density distribution of the diffusing metastable species.

Because the ionization thresholds  $\epsilon_{\ell}$  typically have values that fall in the high-energy tail<sup>57</sup> of  $f_m$  at moderate E/N for which  $\alpha_i > \eta_a$ , it can be expected that  $K_{\ell}$  will increase rapidly with decreasing  $\epsilon_{\ell}$ . Thus even relatively small metastable concentrations can be responsible for disproportionately large enhancements in  $\alpha_i$ . The influence of low metastable concentrations on gasionization rates manifests itself in the well-known optogalvanic effect,<sup>59</sup> which has been observed in both neon<sup>60</sup> and nitrogen<sup>61</sup> discharges at low pressures.

## E. Behavior of $p_1(q_n|q_{n-1})$ and $p_1(q_n|\Delta t_{n-j})$ , $j \ge 2$

If relatively strong dependences of  $q_n$  on  $\Delta t_{n-2}$  (or  $q_{n-1}$ ) occur at a fixed value of  $\Delta t_{n-1}$  as implied by the data on  $p_2(q_n | \Delta t_{n-1}, \Delta t_{n-2})$  and  $p_2(q_n | \Delta t_{n-1}, q_{n-1})$ , respectively, then it can be expected from Eqs. (20) and (15) that dependences of  $q_n$  on  $\Delta t_{n-2}$  (or  $q_{n-1}$ ) will exist independent of  $\Delta t_{n-1}$ . The sign of the  $(q_n, \Delta t_{n-2})$  dependence implied by Eq. (20) depends on the signs and relative strengths of the dependences between the pairs  $(\Delta t_{n-1}, \Delta t_{n-2})$  and  $(q_n, \Delta t_{n-2})$  for fixed  $\Delta t_{n-1}$  and should be the same as that for the  $(q_n, q_{n-1})$  dependence.

Figure 21 shows examples of measured  $p_1(q_n|\Delta t_{n-2})$ and  $p_1(q_n|q_{n-1})$  distributions for different indicated values for  $\Delta t_{n-2}$  and  $q_{n-1}$ . The results are compared with the corresponding unconditional distributions  $p_0(q_n)$ , indicated by the solid lines. The expectation values  $\langle q_n(\Delta t_{n-2}) \rangle$  and  $\langle q_n(q_{n-1}) \rangle$  decrease with increasing  $\Delta t_{n-2}$  and  $q_{n-1}$ , respectively, thus giving negative dependences, i.e.,  $\Delta t_{n-2} \uparrow \Rightarrow q_n \downarrow$  and  $q_{n-1} \uparrow \Rightarrow q_n \downarrow$ . The measured  $p_1(q_n|\Delta t_{n-2})$  distributions are consistent with the corresponding measured distributions  $p_1(\Delta t_{n-1}|\Delta t_{n-2})$  and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$ 



FIG. 21. Measured conditional pulse-amplitude distributions  $p_1(q_n|\Delta t_{n-2})$  and  $p_1(q_n|q_{n-1})$  at the indicated gap voltage and values for  $\Delta t_{n-2}$  and  $q_{n-1}$  in a 100-kPa Ne+5%O<sub>2</sub> gas mixture.

as determined by Eq. (20), which applies for a strong  $(q_n, \Delta t_{n-1})$  dependence. Because of the relatively high value of  $V_a$  and low uv intensity, both the  $(q_n, q_{n-1})$  and  $(\Delta t_{n-1}, \Delta t_{n-2})$  dependences were negative. Therefore the effect of  $p_1$  in this case was to reinforce the  $(q_n, \Delta t_{n-2})$  correlation implied by  $p_2$  in Eq. (20). Because of the strong  $(q_{n-1}, \Delta t_{n-2})$  dependence, the measured  $p_1(q_n|q_{n-1})$  distributions in Fig. 21 behave like the  $p_1(q_n|\Delta t_{n-2})$  distributions and are consistent with Eq. (15) as discussed in Sec. II E.

The measured  $p_1(q_n|\Delta t_{n-2})$  for the N<sub>2</sub> + 20% O<sub>2</sub> mixture<sup>27,30</sup> did not generally deviate significantly from  $p_0(q_n)$ . This is probably due largely to the fact that, in this case, the observed dependence of  $q_n$  on  $\Delta t_{n-2}$ for fixed  $\Delta t_{n-1}$  was considerably weaker than in the Ne+5% O<sub>2</sub> case. Note that in the limit of very weak correlations where  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2}) \simeq p_1(q_n|\Delta t_{n-1})$ and  $p_1(\Delta t_{n-1}|\Delta t_{n-2}) \simeq p_0(\Delta t_{n-1})$ , Eq. (20) reduces to Eq. (10), which simply gives  $p_0(q_n)$ . Also, in the N<sub>2</sub> + 20% O<sub>2</sub> case, the signs of the  $(q_n, \Delta t_{n-2})$  and  $(\Delta t_{n-1}, \Delta t_{n-2})$  dependences implied, respectively, by  $p_1$ and  $p_2$  in Eq. (20) tend to counteract each other.

A comparison of results obtained for  $p_1(q_n | \Delta t_{n-j})$ , j = 1, 2, 3, 4, from a Ne+5% O<sub>2</sub> mixture is shown in Fig. 22 for two different gap voltages and three different values for  $\Delta t_{n-j}$ . These results represent fits to the numerical data that have been normalized to the maxima. The lowest  $\Delta t_{n-j}$  values correspond to time separations closest to  $\Delta t_c$  in the  $p_0(\Delta t_n)$  distributions, whereas the intermediate and largest values correspond, respectively, to the maxima and high ends of these distributions. The ratios of expectation values,  $\langle q_n(\Delta t_{n-j}) \rangle / \langle q_n \rangle$ , obtained from the corresponding distributions in Fig. 22, are shown graphically in Fig. 23. Here  $\langle q_n \rangle$  corresponds to  $p_0(q_n)$  as given by Eq. (33). The bars shown in Fig. 23 represent the approximate limits for statistically significant differences between  $\langle q_n(\Delta t_{n-j}) \rangle$  and  $\langle q_n \rangle$  and provide a measure of the extent to which the  $p_1(q_n|\Delta t_{n-j})$ and  $p_0(q_n)$  distributions differ.

The previously discussed positive  $(q_n, \Delta t_{n-1})$  and negative  $(q_n, \Delta t_{n-2})$  dependences are clearly demonstrated in Fig. 23. A statistically significant positive  $(q_n, \Delta t_{n-3})$ dependence is evident from the 7.2-kV data. The 6.8-kV data suggest a nonmonotonic behavior in the  $(q_n, \Delta t_{n-3})$ dependence. Nonmonotonic behavior in the sign of the  $(q_n, \Delta t_{n-3})$  dependence occurs under some conditions. However, for the data shown in Fig. 23, it is of questionable statistical significance.

Except at the very lowest  $\Delta t_{n-4}$  values, the  $p_1(q_n|\Delta t_{n-4})$  distributions did not differ significantly from the corresponding  $p_0(q_n)$  distributions. The  $p_1(q_n|\Delta t_{n-4})$  results generally exhibit the hint of a negative  $(q_n, \Delta t_{n-4})$  dependence over at least the lower part of the  $\Delta t_{n-4}$  range. The lack of equality between  $p_1(q_n|\Delta t_{n-j})$  and  $p_0(q_n)$ , which can occur even for j = 4, unequivocally shows that memory extends back considerably beyond the preceding event. This agrees with

Δt<sub>n-j</sub>= 80 μs

lil

Δt<sub>n-i</sub>= 60 μ

∆t<sub>n-j</sub>

100 150 200 250

40 µs

50

q<sub>n</sub>(pC)

= 1

= 3

= 4

Va = 7.2 kV

 $\Delta t_{n-j} = 60 \, \mu s$ 

 $\Delta t_{n-j} = 50 \, \mu s_{j}$ 

Δt<sub>n-j</sub>= 35 μs

100 150 200 250 300

1.0

0.0

-1.0

-2.0

-1.0

-2.0

0.0

-1.0

-2.0 --3.0 -50

0.0

 $09_{10}$ [ p<sub>1</sub>(q<sub>n</sub> | $\Delta t_{n-i}$ ) / p<sub>1</sub>( q<sub>n,max</sub>|  $\Delta t_{n-i}$ )]

Ne + 5% O<sub>2</sub>

Va = 6.8 kV

recent observations of Steiner<sup>25</sup> by a completely different method and conclusively demonstrates that Trichelpulse corona is indeed a non-Markovian stochastic process. The alternating signs of the correlations with increasing j are consistent with the arguments in Sec. II F.

In the present experiment, only conditional pulseamplitude distributions up to second order could be measured (see Table I). However, the data for  $p_2(q_n|\Delta t_{n-1}, q_{n-1})$  and  $p_2(q_n|\Delta t_{n-1}, \Delta t_{n-2})$  shown in Figs. 15 and 17, respectively, indicate that it would be difficult to observe correlations between  $q_n$  and either  $q_{n-2}$  or  $\Delta t_{n-3}$  even if  $p_3$  distributions could have been measured. This is a consequence of inherent limitations in the instrumental resolution such as determined by amplifier noise and number of MCA channels. Thus, although memory may extend indefinitely back in time, there are instrumental limitations to assessing the extent of memory propagation for large values of j ( $j \gtrsim 4$  in the present case). It is shown in the Appendix that, for the present experimental situation in which second-order correlations between  $q_n$  and  $\Delta t_{n-2}$  can easily be resolved, j = 4 is the lowest value of j for which the profiles of the  $p_1(q_n | \Delta t_{n-j})$  and  $p_0(q_n)$  distributions could become indistinguishable at all allowed values for  $\Delta t_{n-i}$ .

#### V. SUMMARY AND CONCLUSIONS

The experimental results presented here demonstrate unequivocally that the Trichel-pulse discharge phe-





FIG. 23. Ratios of the expectation values  $\langle q(\Delta t_{n-j}) \rangle / \langle q_n \rangle$  computed from the  $p_1(q_n | \Delta t_{n-j})$  distributions shown in Fig. 22.

nomenon is a stochastic process in which memory effects play an important role. This phenomenon represents one of the clearest examples of a non-Markovian, marked random point process with stochastic properties that can be completely and unambiguously characterized by appropriate measurements. Any attempt to formulate a theoretical model that can account for regular Trichel-pulse behavior beyond the first pulse, i.e., for n > 1, must necessarily include effects of memory.

The stochastic properties of Trichel-pulse discharges determined here from the direct measurement of conditional and unconditional pulse-amplitude and timeseparation distributions can be attributed to the influences of ion space charge and metastable species from previous pulses on the initiation and development of subsequent pulses. The predominant and most ubiquitous memory effect is that associated with the influence of the moving negative-ion space-charge cloud from the most recent pulse on the growth of the subsequent pulse. The influence of this space-charge cloud in suppressing the rate of initiating-electron release from the cathode is also evident in data from  $Ne+5\%O_2$ . Mixtures that contain a higher relative  $O_2$  concentration, i.e.,  $N_2 + 20\% O_2$ ,  $Ne+95\%O_2$ , and  $O_2$ , show indications of the influence of metastable species from earlier pulses on both the initiation and growth of subsequent pulses. One explanation for an increasing effectiveness of metastables in releasing electrons from the cathode with increasing  $O_2$  content may be the corresponding higher fields required to initiate the discharge with consequent enhancements in fieldassisted electron ejection by quenching of metastables on or near the cathode surface.

The unconditional pulse time-separation distributions exhibit well-defined critical minimum time separations  $\Delta t_c$  below which subsequent pulses do not occur. The minimum time separation determines the maximum pulse repetition rate that can occur under any given set of gap conditions and is physically more significant and better defined than the previously measured pulse repetition rate. For uv-sustained discharge pulses,  $\Delta t_c$  is independent of uv intensity but increases exponentially with decreasing gap voltage and evidently represents the time for restoration of the field near the cathode to a value required for initiation and development of the next pulse. This time is controlled by the ambipolar diffusion of ions produced during the most recent discharge event.

When discharge initiation is controlled predominantly by field-assisted photoelectron emission at the cathode, the pulse time-separation distributions exhibit tails at the high end that decrease exponentially at a rate which increases with increasing uv intensity and are consistent with a von Laue distribution. At low uv intensity, the unconditional pulse-amplitude distributions become sharply peaked at maximum values  $q_{\rm lim}$  which increase with gap voltage and correspond to the largest pulses that can be formed in a gap free of space charge. The observed behavior of the unconditional pulse-amplitude and time-separation distributions are consistent with all previously published data on the electrical characteristics of Trichel pulses.

Because of strong correlations that occur among pulse amplitudes and time separations, the conditional and unconditional pulse-amplitude and time-separation distributions have mutual dependences that can be predicted successfully using appropriate integral relationships that connect these distributions. The measured conditional distributions also give the required consistency in the signs of dependences between the pulse-amplitude and time-separation variables. Strong correlations were not only found between  $q_n$  and  $\Delta t_{n-1}$ , but also, in some cases, between  $q_n$  and  $q_{n-1}$  and between  $q_n$  and  $\Delta t_{n-2}$  for fixed  $\Delta t_{n-1}$ . Significant dependences of  $\Delta t_n$  on  $q_n$  and  $\Delta t_{n-1}$  were also found. From the measured conditional distributions  $p_1(q_n | \Delta t_{n-j})$ , correlations were shown to exist between  $q_n$  and all  $\Delta t_{n-j}$ ,  $j \leq 4$ . Because of the dependences of  $q_n$  (or  $\Delta t_n$ ) on  $\Delta t_{n-1}$  and  $q_{n-1}$ , "memory" can extend back to a large number of earlier events in a Trichel-pulse corona. Experimental limitations to verifying the extent of memory propagation by the present method have been analyzed.

In the present work, the discharge pulse amplitude was chosen as the "mark" that gives a measure of the discharge magnitude. There are other marks associated with other characteristics of the discharge pulse that could be considered. Amplitude was selected not only because it is a measure of discharge intensity but also because it can be measured easily. Other less easily measured marks such as the pulse shape parameters might also be correlated significantly with pulse-time separation as suggested by Morrow.<sup>18</sup> Additionally, one could consider optical emission characteristics of the discharge. It should finally be noted that the measurement method applied here to investigate Trichel pulses could also be used to measure stochastic behavior in other types of pulsating or time-ordered phenomena that can be represented as random point processes for which the appropriate marks can be converted to electrical signals that increase in proportion to the magnitude of the mark.

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## APPENDIX

It was demonstrated from the results presented in Sec. IV E that for sufficiently large j, the profiles for the  $p_1(q_n|\Delta t_{n-j})$  distributions become indistinguishable from the profile for the unconditional  $p_0(q_n)$  distribution. The conditions under which this equality between  $p_1$  and  $p_0$  holds will now be examined for the case of a strong positive dependence of  $q_n$  on  $\Delta t_{n-1}$ . From the law of probabilities, the integral relationship for  $p_1(q_n|\Delta t_{n-j})$ which applies in general for arbitrary j > 1, is  $p_1\left(q_n | \Delta t_{n-j}\right)$ 

$$= \int_{\Delta t_{n-1}} \cdots \int_{\Delta t_{n-j+1}} \int_{q_{n-1}} \cdots \int_{q_{n-j+1}} p_1 \left( q_{n-j+1} | \Delta t_{n-j} \right) p_2 \left( \Delta t_{n-j+1} | q_{n-j+1}, \Delta t_{n-j} \right) \\ \times \prod_{i=2}^{j} p_{2i-1} \left( q_{n-j+i} | \left\{ \Delta t_{n-k}, q_{n-k} \right\}, \Delta t_{n-j} \right) \\ \times \prod_{i=2}^{j-1} p_{2i} \left( \Delta t_{n-j+1} | q_{n-j+1}, \left\{ \Delta t_{n-k}, q_{n-k} \right\}, \Delta t_{n-j} \right) \\ \times \prod_{i=1}^{j-1} dq_{n-i} d \left( \Delta t_{n-i} \right),$$
(A1)

where k assumes all integer values in the range

$$j - i + 1 \le k \le j - 1.$$

In the case of a strong  $(q_n, \Delta t_{n-1})$  dependence as defined in Sec. II B, Eq. (A1) reduces to

$$p_{1}(q_{n}|\Delta t_{n-j}) = \int_{\Delta t_{n-j}} \cdots \int_{\Delta t_{n-j+i}} p_{2j-2}(q_{n}|\{\Delta t_{n-\ell}\}) \prod_{i=1}^{j-1} p_{2i-1}(\Delta t_{n-j+i}|\{\Delta t_{n-k}\}) d(\Delta t_{n-i}),$$
(A2)  
where  $\ell = 1, 2, \dots, j$  and  $j-i+1 \le k \le j$ . In derive  $p_{2i-1}(\Delta t_{n-j+i}|\{\Delta t_{n-k}\}) = p_{1}(\Delta t_{n-j+i}|\Delta t_{n-j-1+i}),$ 

where  $\ell = 1, 2, ..., j$  and  $j - i + 1 \le k \le j$ . In deriving Eq. (A2) from Eq. (A1) it is necessary to use the normalization condition Eq. (1).

Consider the following four special cases such as might be imposed by instrumental resolution.

Case 1:

$$p_{2j-2}(q_n | \{ \Delta t_{n-\ell} \}) = p_2(q_n | \Delta t_{n-1}, \Delta t_{n-2}),$$
  
$$j \ge 2, \ 1 \le \ell \le j. \quad (A3)$$

Case 2:

$$p_{2j-2}(q_n|\{\Delta t_{n-\ell}\}) = p_3(q_n|\Delta t_{n-1}, \Delta t_{n-2}, \Delta t_{n-3}),$$

$$j \ge 3, \ 1 \le \ell \le j.$$
 (A4)

Case 3:

.

$$p_{2}(\Delta t_{n-\ell}|\Delta t_{n-\ell-m}, \Delta t_{n-\ell-m'}) = \begin{cases} p_{0}(\Delta t_{n-\ell}) \text{ for } m \text{ and } m' \ge 3\\ p_{1}(\Delta t_{n-\ell}|\Delta t_{n-\ell}) \text{ for } m = 1, m' \ge 3\\ p_{1}(\Delta t_{n-\ell}|\Delta t_{n-\ell-2}) \text{ for } m = 2, m' \ge 3. \end{cases}$$
(A8)

and

Cases 1 (and 2) correspond, respectively, to situations where correlations between  $q_n$  and  $\Delta t_{n-3}$  (or  $q_n$  and  $\Delta t_{n-4}$ ) either do not exist or cannot be experimentally ascertained. Likewise, cases 3 (and 4) correspond, respectively, to situations where correlations between  $\Delta t_n$  and  $\Delta t_{n-2}$  (or  $\Delta t_{n-3}$ ) either do not exist or cannot be experimentally ascertained.

Inserting the combination of cases 1 and 3 into Eq. (A2) gives

$$p_1(q_n | \Delta t_{n-j}) = \int_{\Delta t_{n-1}} \cdots \int_{\Delta t_{n-j+1}} p_2(q_n | \Delta t_{n-1}, \Delta t_{n-2}) \prod_{i=1}^{j-1} p_1(\Delta t_{n-j+i} | \Delta t_{n-j+i-1}) d(\Delta t_{n-i}).$$
(A9)

Using the integral relationships

$$\int_{\Delta t_{n-i-1}} p_1(\Delta t_{n-i}) |\Delta t_{n-i-1}| p_1(\Delta t_{n-i-1} |\Delta t_{n-i-2}) d(\Delta t_{n-i-1}) = p_1(\Delta t_{n-i} |\Delta t_{n-i-2}) = p_0(\Delta t_{n-i})$$
(A10)

and

$$p_1(\Delta t_{n-j+i} | \Delta t_{n-j+i-k}) = p_0(\Delta t_{n-j+i})$$
(A6)

for all k > 1. Case 4:

$$p_{2i-1}(\Delta t_{n-\ell}|\{\Delta t_{n-k}\}) = p_2(\Delta t_{n-\ell}|\Delta t_{n-\ell-1},\Delta t_{n-\ell-2}),$$

$$\ell = j - i \quad (A7)$$

 $1 \le i \le j - 1 \quad (A5)$ 

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and

$$\int_{\Delta t_{n-i}} p_0(\Delta t_{n-i}) p_1(\Delta t_{n-i+1} | \Delta t_{n-i}) \, d(\Delta t_{n-i}) = p_0(\Delta t_{n-i+1}), \tag{A11}$$

Eq. (A9) becomes

$$p_1(q_n | \Delta t_{n-j}) = \int_{\Delta t_{n-1}} \int_{\Delta t_{n-2}} p_0(\Delta t_{n-2}) p_1(\Delta t_{n-1} | \Delta t_{n-2}) p_2(q_n | \Delta t_{n-1}, \Delta t_{n-2}) d(\Delta t_{n-1}) d(\Delta t_{n-2}), \tag{A12}$$

which is simply

$$p_1(q_n | \Delta t_{n-j}) = p_0(q_n),$$
 (A13)

provided  $j \ge 4$ .

Using the combination of cases 1 and 4 in Eq. (A2) together with the integral relationship

$$\int_{\Delta t_{n-i}} p_1(\Delta t_{n-i} | \Delta t_{n-i-1}) p_2(\Delta t_{n-i+1} | \Delta t_{n-i}, \Delta t_{n-i-1})$$

$$\times p_2(\Delta t_{n-i+2} | \Delta t_{n-i+1}, \Delta t_{n-i}) d(\Delta t_{n-i})$$

$$= p_1(\Delta t_{n-i+2} | \Delta t_{n-i+1}) p_1(\Delta t_{n-i+1} | \Delta t_{n-i-1})$$

(A14)

gives Eq. (A13) if  $j \ge 5$ . It can also be shown that the combination of case 2 with cases 3 and 4, respectively, give Eq. (A13) if  $j \ge 5$  and 6. The above arguments can obviously be extended to cases where higher order correlations between  $q_n$  and  $\Delta t_{n-i}$  (i > 3) or between  $\Delta t_n$  and  $\Delta t_{n-i}$  ( $i \ge 3$ ) can be distinguished. The combination of cases 1 and 3 correspond to the lowest order of correlations implied by the present experimental situation. Therefore j = 4 is the lowest j for which Eq. (A13) can be expected to apply, especially for the Ne+5% O<sub>2</sub> results presented here. This appears to be consistent with the data shown in Sec. IV E. The limitations on noise level in the measurement of higher-order conditional probability distributions for a time-ordered process has previously been noted by Packard and co-workers.<sup>62</sup>

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