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# Ionization of gases by a pulsed electron beam as studied by self-focusing. II. Polyatomic gases

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In order to analyze data on the self-focusing of a pulsed electron beam in polyatomic gases, the net current  $I_{\rm net}$  in  $H_2$ ,  $N_2$ , and  $CH_4$  was computed self-consistently as functions of time in the pressure range between 5 and 300 Torr of these gases by using swarm parameters. The computational result indicates that the larger dose  $D_{\rm obs}$ , observed by a piled dosimeter on the beam axis, is attributed to the larger  $I_{\rm net}$ , which is mainly determined by a mean ionization time  $t_1$  for secondary ionization by the electric field induced by the pulsed beam. When values of  $D_{\rm obs}$  for different gases are compared at the same pressure, the larger  $D_{\rm obs}$  is given by the larger  $t_i$ . This relationship is demonstrated for several polyatomic gases by estimating  $t_i$  from various parameters in a function of secondary electron energy or E/p such as the electron drift velocity, the first Townsend ionization coefficient, the ionization cross section, and so on. For the short pulse duration of a Febetron 706, electron—ion recombination processes scarcely affect  $I_{\rm net}$  except at high pressures of some polyatomic gases, while the effect of electron-attachment processes is appreciable in  $SF_6$ ,  $CCl_2F_2$ , and  $N_2O$ .

#### INTRODUCTION

In a preceding paper (part I),  $^1$  it has been shown that, for monatomic gases, the maximum dose  $(D_{\rm obs})$  of the depth-dose curve in a piled dosimeter given by a pulsed electron beam of a Febetron 706 (tube  $5515)^2$  can be interpreted in terms of the net current  $I_{\rm net}$ , which is mainly determined by a mean ionization time  $t_i$ . The value of  $I_{\rm net}$  has been calculated with the following equations:

$$I_{\text{net}}(t) = I_b(t) + I_{\text{back}}(t) , \qquad (1)$$

$$I_{\text{back}}(t) = \pi r_0 E_{\sigma}(t) \sigma_{\sigma}(t) , \qquad (2)$$

$$E_z(t) = -\frac{2}{c^2} \left(\frac{1}{2} + \ln \frac{R}{r_0}\right) \frac{dI_{\text{net}}(t)}{dt},$$
 (3)

$$\sigma_e(t) = \frac{e^2 n_e(t)}{n_0(2m)^{1/2} Q_m(\overline{\epsilon}) \overline{\epsilon}^{1/2}},$$
 (4)

$$\frac{dn_e(t)}{dt} = \frac{n_0\sigma_{\text{ion}}(E_b)I_b(t)}{\pi r_0^2 e} + \frac{n_e(t)}{t_i(t)},$$
 (5)

where  $I_b$  is the beam current assumed as in part I,  $I_{back}$ the plasma backward current,  $r_0$  the beam radius assumed to be 0.6 cm, R the chamber radius (6.4 cm).  $E_z$  the longitudinal electric field induced along the beam coordinate z by the pulsed beam, c the velocity of light, o, the plasma conductivity of the irradiated medium gas, m the electron rest mass, e the electron charge,  $n_0$  the number density of gas molecules,  $n_e$  the number density of secondary electrons, and  $Q_m$  the momentum transfer cross section for electrons with mean kinetic energy  $\overline{\epsilon}$ . The total ionization cross section  $\sigma_{ton}(E_b)$  for the beam electron with energy  $E_b$  is estimated for  $\overline{E}_b = 480 \text{ keV}$ from the Rieke-Prepejchal parameters. 3 The mean ionization time  $t_i$  for secondary ionization has been determined as a function of E/p for some gases by Felthenthal and Proud. For other gases,  $t_i$  can be estimated by

$$1/(pt_i) = w(\alpha/p - \eta/p) , \qquad (6)$$

where  $\alpha$  is the first Townsend ionization coefficient,  $\eta$ 

the electron-attachment coefficient, and w the electron drift velocity. It has been demonstrated from the calculated  $I_{\rm net}$  that  $D_{\rm obs}$  can be expressed approximately as

$$D_{\text{obs}} \propto \int \frac{I_b(t) I_{\text{net}}(t)}{\epsilon_b^2} dt , \qquad (7)$$

where  $\pi\epsilon_b$  is the beam emittance. Thus, the larger  $D_{\rm obs}$  corresponds to the larger  $I_{\rm net}$ , which is given by the smaller  $I_{\rm back}$  for  $dI_{\rm net}/dt > 0$  from Eqs. (1)-(3). This means that the larger  $D_{\rm obs}$  is given by a gas with the larger  $t_i$  for  $dI_{\rm net}/dt > 0$ .

The similar computation was carried out for  $H_2$ ,  $N_2$ , and  $CH_4$ . In the present paper, data on  $D_{\rm obs}$  for various polyatomic gases<sup>5-7</sup> are discussed on the basis of the computational result for these gases.

### COMPUTATION FOR H<sub>2</sub>, N<sub>2</sub>, and CH<sub>4</sub>

In the computation, the parameters  $r_0$ ,  $I_b(t)$ , and  $\sigma_{\rm ion}(E_b)$  were used on the same assumptions as made in part I. Literature values were used for the following parameters; w,  $^8$   $\alpha/p$ ,  $^9$   $D_L/\mu$ ,  $^{10}$  and  $Q_m^{-11}$  for H<sub>2</sub>; w,  $^8$   $\alpha/p$ ,  $^{12}$   $D_L/\mu$ ,  $^{13}$  and  $Q_m^{-11}$  for N<sub>2</sub>; and w,  $^{14-16}$   $\alpha/p$ ,  $^{17}$   $D_L/\mu$ ,  $^{18}$  and  $Q_m^{-19,20}$  for CH<sub>4</sub>. The value of  $\overline{\epsilon}$  was estimated from data on  $D_L/\mu$  on the assumption of the Maxwellian. For polyatomic gases, Eq. (5) was revised as

$$\frac{dn_e}{dt} = \frac{n_0 \sigma_{\text{lon}}(E_b)}{e} \cdot \frac{I_b(t)}{\pi r_0^2} + \frac{n_e(t)}{t_i(t)}$$

$$(\text{term 1}) \quad (\text{term 2})$$

$$-\alpha_r n_e(t) n_i(t) - \alpha_{rd} n_e(t) n_d(t) ,$$

$$(\text{term 3}) \quad (\text{term 4})$$
(8)

$$\frac{dn_i}{dt} = \frac{n_0 \sigma_{ion}(E_b)}{e} \cdot \frac{I_b(t)}{\pi r_0^2} + \frac{n_e}{t_i} - \alpha_r n_e n_i - k_d n_i n_0^2 , \qquad (9)$$

$$\frac{dn_d}{dt} = k_d n_i n_0^2 - \alpha_{rd} n_e n_d , \qquad (10)$$

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where  $\alpha_r$  and  $\alpha_{rd}$  are the recombination coefficients  $(\text{cm}^3/\text{s})$  between electron—ion and electron—dimer ion, respectively, and  $k_d$  is the rate constant of dimer ion formation. The number densities of ions and dimer ions are represented by  $n_i$  and  $n_d$ , respectively. We have assumed that the variation of  $\alpha_r$  and  $\alpha_{rd}$  with  $\overline{\epsilon}$  is expressed as

$$\begin{split} \alpha_r(\mathrm{H}_2^\star) &= 4.\ 7 \times 10^{-8} \,\overline{\epsilon}^{-0.40} \ \ (\mathrm{Ref.\ 21}) \ , \\ \alpha_r(\mathrm{N}_2^\star) &= 5.\ 5 \times 10^{-8} \,\overline{\epsilon}^{-0.65}, \quad \mathrm{for} \ \overline{\epsilon} > 1 \ \mathrm{eV} \ (\mathrm{Ref.\ 22}) \ , \\ \alpha_{rd}(\mathrm{N}_4^\star) &= 3.\ 7 \times 10^{-7} \,\overline{\epsilon}^{-0.65}, \quad \mathrm{for} \ \overline{\epsilon} > 1 \ \mathrm{eV} \ (\mathrm{Ref.\ 22}) \ , \\ \alpha_r(\mathrm{CH}_3^\star) &= 5.\ 5 \times 10^{-8} \,\overline{\epsilon}^{-0.93}, \quad \mathrm{for} \ \overline{\epsilon} > 0.8 \ \mathrm{eV} \ (\mathrm{Ref.\ 23}) \ , \\ \alpha_{rd}(\mathrm{C}_2\mathrm{H}_5^\star) &= 9.\ 5 \times 10^{-8} \,\overline{\epsilon}^{-0.75}, \quad \mathrm{for} \ \overline{\epsilon} > 1 \ \mathrm{eV} \ (\mathrm{Ref.\ 23}) \ , \\ \mathrm{and} \ \mathrm{for} \ \mathrm{formation} \ \mathrm{processes} \ \mathrm{of} \ \mathrm{N}_4^\star \ \mathrm{and} \ \mathrm{C}_2\mathrm{H}_5^\star \colon \\ \mathrm{N}_2^\star + 2\mathrm{N}_2 \to \mathrm{N}_4^\star + \mathrm{N}_2 \end{split}$$

$$N_2^+ + 2N_2 + N_4^+ + N_2$$
  
 $(k_d = 8 \times 10^{-29} \text{ molecule}^{-2} \text{ cm}^6 \text{ s}^{-1}) \text{ (Ref. 24)},$   
 $CH_3^+ + CH_4 + C_2H_5^+ + H_2$   
 $(k_d = 1 \times 10^{-9} \text{ cm}^3/\text{s}) \text{ (Ref. 25)}.$ 

For hydrogen, only the association reaction of  $H^*$  leading to  $H_3^*$  is known. When the However, the yield of  $H^*$  is small in comparison with that of  $H_2^*$  (about several percent). Thus, the recombination of electron— $H_3^*$  was neglected in the calculation for  $H_2$ . Only the formation of  $CH_3^*$  and  $C_2H_2^*$  was assumed in the calculation for methane.

Computational results of  $I_{\rm net}(t)$  for  $H_2$ ,  $N_2$ , and  $CH_4$  are shown in Figs. 1, 2, and 3, respectively. They show that the calculated value of  $I_{\rm net}$  at lower pressure is nearly constant after a certain time, as has been assumed previously as the  $t_B$  model. The value of  $D_{\rm obs}$  increases at first with increasing pressure in the low pressure region, and after passing the maximum it decreases abruptly at a certain pressure (usually 1-5 Torr) but increases again with further increasing pressure. The curve of  $I_{\rm net}(t)$  varies according to the  $t_B$ 

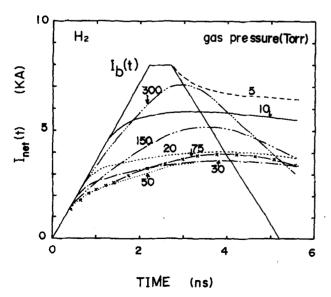


FIG. 1. The calculated net current  $I_{\text{net}}(t)$  as a function of time for various pressures of  $H_2$ . The solid straight lines indicate the beam current  $I_b(t)$ .

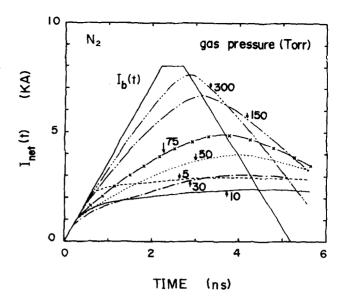


FIG. 2. The calculated net current  $I_{\text{net}}\left(t\right)$  as a function of time for various pressures of  $N_{2}$ .

model up to the pressure giving the minimum  $D_{\rm obs}$ . In this region,  $I_{\rm net}(t)$  decreases with increasing pressure.

In the present paper, we are most interested in the higher pressure region in which  $D_{\rm obs}$  increases gradually after passing the minimum. In this region, in Figs. 1-3,  $I_{\rm net}(t)$  increases appreciably with the lapse of time and also increases with increasing pressure. According to Eq. (7),  $D_{\rm obs}$  should be proportional to the value of  $\int I_b(t)I_{\rm net}(t)dt$  if the variation of  $\epsilon_{\rm b}$  is ignored. In Fig. 4, values of this integral are plotted in an arbitrary unit by solid symbols for  $H_2$ ,  $N_2$ , and  $CH_4$  by using the calculated values of  $I_{\rm net}$  in Figs. 1, 2, and 3, respectively, together with the curves of  $D_{\rm obs}$  shown by open symbols. The curve of the integral represents fairly well the aspect of the curve of  $D_{\rm obs}$ . However, when both the curves are compared at the pressure giving the minimum, the value

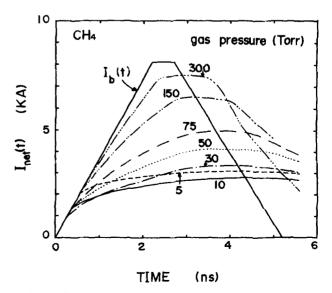


FIG. 3. The calculated net current  $I_{\rm net}(t)$  as a function of time for various pressures of  ${\rm CH_4}$ .

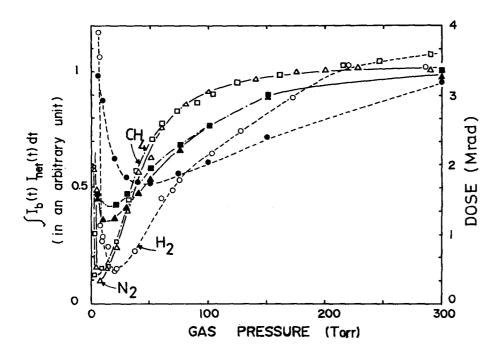


FIG. 4.  $D_{obs}$  at various pressures of  $H_2$ ,  $N_2$ , and  $CH_4$ :  $O(H_2)$ ,  $\Delta(N_2)$ , and  $CCH_4$ . Solid marks  $[\bullet(H_2), \Delta(N_2), A(N_2), A(N_2)]$ , and  $CCH_4$  indicate the values of the integral in Eq. (7) in an arbitrary unit.

of the integral is relatively larger than that of  $D_{\mathrm{obs}}$ . This can be fitted by using the correct value of  $\epsilon_b$  as has been discussed in part I. Consequently, the present computational result represents fairly well the phenomena induced by the pulsed beam in a gas chamber so that the calculated values of parameters such as  $I_{\mathrm{net}}$ ,  $n_e$ , and E/p, which are not easy to be measured, might be used semiquantitatively.

Curves of  $E_z/p$  calculated from the values of  $I_{\rm net}$  in Figs. 1, 2, and 3 are shown as functions of t in Figs. 6, 7, and 8 for  $H_2$ ,  $N_2$ , and  $CH_4$ , respectively. In the

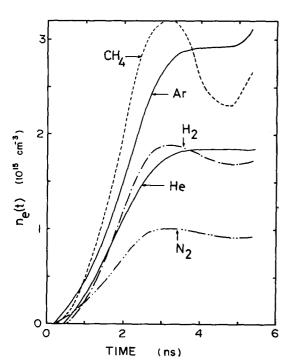


FIG. 5. The calculated number density  $n_e(t)$  of produced secondary electrons as functions of time at 50 Torr of He (—), Ar (—), H<sub>2</sub> (-·-), N<sub>2</sub> (-··-), and CH<sub>4</sub> (----).

present computation, values of  $\overline{\epsilon}$  are estimated from data on  $D_L/\mu$  on the assumption of the Maxwellian as functions of the calculated  $E_z/p$  shown in Figs. 6-8. Such values of  $\overline{\epsilon}$  at pressure of 30 Torr for three gases, as

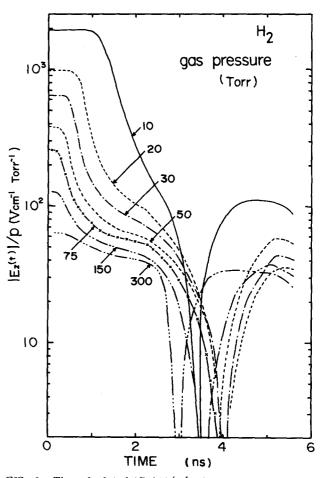


FIG. 6. The calculated  $|E_x(t)|/p$   $|E_x(t)|$  the induced longitudinal electric field] as functions of time for various pressures of  $H_2$ .

the representative, are shown as functions of t in Fig. 9, because we are interested mainly in the pressure region between 10 and 50 Torr in the present paper. Curves in Figs. 6-8 show that the value of  $E_z/p$  decreases abruptly for a short period. However, the actual value of  $\overline{\epsilon}$  for this period might not so decrease as shown in Fig. 9 so that the value of  $t_i$  for this period may not vary abruptly according to such a calculated  $E_z/p$  because of the presence of the relaxation time of  $\overline{\epsilon}$ . The present computation, however, was carried out straightforwardly according to the calculated  $E_z/p$  without any correction.

The calculated value of  $n_e(t)$  increased with increasing pressure and became the maximum at 75 Torr of  $H_2$ , 50 Torr of  $N_2$ , and 30 Torr of  $CH_4$ . The value decreased slightly with further increasing pressure. The value of  $n_e$  became the maximum at the pressure for which  $D_{\rm obs}$  is nearly equal to 1 Mrad. Such a relationship of  $n_e(t)$  between p or  $D_{\rm obs}$  is common with He and Ar, whose curves are shown in detail in part I. Then, the curve of  $n_e(t)$  becomes almost the same irrespective of pressure for  $D_{\rm obs}$  higher than 1 Mrad. Curves of  $n_e(t)$  at 50 Torr are shown as functions of t in Fig. 5.

The contribution of each term to the total  $n_e$  in Eq. (8) is as follows, although the computation was carried out for a discrete value of p: The contribution of direct ionization (term 1) is less than 10% of electron avalanching (term 2) at 20-150 Torr of  $H_2$ , 5-50 Torr of  $N_2$ , and 5-100 Torr of  $CH_4$ . Furthermore, the contribution of recombination precesses (terms 3 and 4) at the peak of

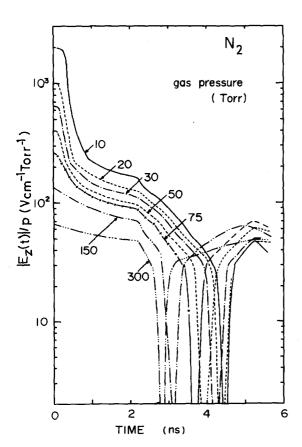


FIG. 7. The calculated  $|E_x(t)|/p$  as functions of time for various pressures of  $N_2$ .

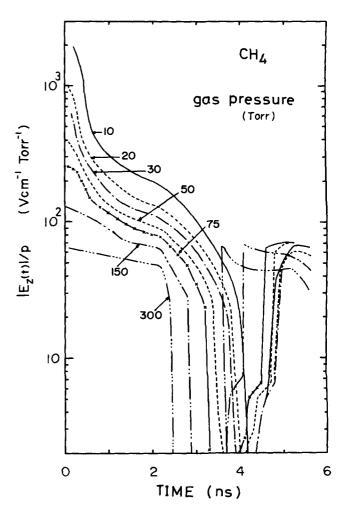


FIG. 8. The calculated  $|E_{\bf g}(t)|/p$  as functions of time for various pressures of CH<sub>4</sub>.

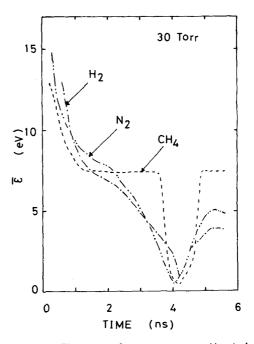


FIG. 9. The mean electron energy estimated at 30 Torr of  $H_2$  (---),  $N_2$  (----), and  $CH_4$  (----) as functions of time.

the pulsed beam (2.2 ns) is less than 10% of the total  $n_e$  at pressures below 300 Torr of the three gases. At higher pressure, however, the recombination contribution became large at 3 ns due to decreasing  $\overline{\epsilon}$  in the low-E/p region and due to the large amount of dimer-ion formation. Some results are shown in Figs. 10 and 11. The number densities of secondary electrons  $(n_e)$ ,  $N_2^*(n_i)$ , and  $N_4^*(n_d)$  at 30 and 300 Torr of  $N_2$  are shown as functions of t in Fig. 10. At 300 Torr, most of  $N_2^*$  becomes  $N_4^*$  without recombination for the pulse duration. The similar curves of  $n_e$ ,  $n_i(\text{CH}_3^*)$ , and  $n_d(\text{C}_2\text{H}_5^*)$  at 30 and 300 Torr of  $\text{CH}_4$  are shown in Fig. 11.

Generally speaking, since  $n_e$  is on the order of  $10^{15}$  cm<sup>-3</sup> as seen in Fig. 5, the half-life of secondary electrons for electron—ion recombination processes is less than 1 ns for  $\alpha$ , larger than  $10^{-6}$  cm<sup>3</sup> s<sup>-1</sup>. Such large rate constants, are reported only for  $N_4^*$ ,  $^{28}$   $O_4^*$ ,  $^{28}$  and  $CH_4^*$  or  $CH_3^*$  for thermal electrons and  $N_2O^{*\,29}$  for 0.04 eV electrons. Apparent recombination coefficients for  $N_2$ ,  $CO_2$ ,  $CH_4$ , and  $C_3H_8$  for thermal electrons<sup>30</sup> are also larger than  $10^{-6}$  cm<sup>3</sup> s<sup>-1</sup> at 50 Torr. When  $\alpha$ , is reduced according to the  $\overline{\epsilon}^{-0.5}$  law, the value may be on the order of  $10^{-7}$  cm<sup>3</sup> s<sup>-1</sup> even for these gases for the present experiments. Therefore, we can ignore the electron—ion recombination process at least for the ns pulse duration in the pressure region of interest.

#### DISCUSSION

It is concluded from Fig. 4 that the larger  $D_{\rm obs}$  is given by the larger  $I_{\rm net}$  which is due to the smaller  $I_{\rm back}$  or  $\sigma_e E_z$  from Eqs. (1), (2), and (7). From Eq. (4),  $\sigma_e E_z$  is pro-

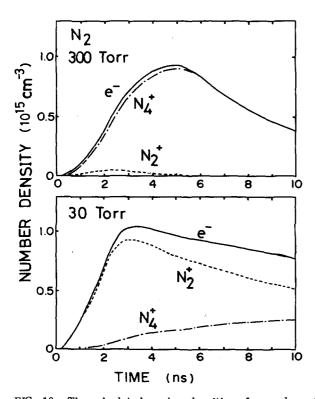


FIG. 10. The calculated number densities of secondary electrons  $e^*$ , monomer ion  $N_2^*$ , and dimer ion  $N_4^*$  as functions of time for 30 and 300 Torr of  $N_2$ .

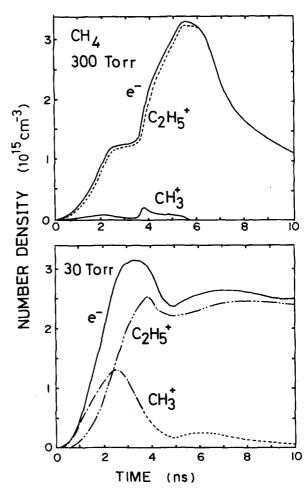


FIG. 11. The calculated number densities of secondary electrons  $e^*$ , monomer ion CH<sub>3</sub>, and dimer ion C<sub>2</sub>H<sub>5</sub> as functions of time for 30 and 300 Torr of CH<sub>4</sub>.

portional to the product of  $n_e/Q_m\overline{\epsilon}^{1/2}$  and  $E_s/p$ . The value of  $Q_m(\overline{\epsilon})\overline{\epsilon}^{1/2}$  increases with increasing  $\overline{\epsilon}$  and usually becomes nearly constant from a certain  $\overline{\epsilon}$ . Such plateau values are  $(2-3)\times 10^{-15}$  cm<sup>2</sup> eV<sup>0.5</sup> for 1-10 eV of  $\overline{\epsilon}$  (5-300 V cm<sup>-1</sup> Torr<sup>-1</sup> of E/p) for  $H_2$ ,  $(2.5-4.5)\times 10^{-15}$  cm<sup>2</sup> eV<sup>0.5</sup> for 2-10 eV of  $\overline{\epsilon}$  (1-150 V cm<sup>-1</sup> Torr<sup>-1</sup> of E/p) for  $N_2$ , and  $6.5\times 10^{-15}$  cm<sup>2</sup> eV<sup>0.5</sup> for 7.4 eV (24-136 V cm<sup>-1</sup> Torr<sup>-1</sup> of E/p) for CH<sub>4</sub>. In Figs. 6-8, these E/p regions giving the plateau value are covered by the major part of the pulse duration. The difference of  $Q_m(\overline{\epsilon})\overline{\epsilon}^{1/2}$  among gases must be noticed but the plateau values are not much different among gases except CH<sub>4</sub>. Then, the aspect of  $D_{\rm obs}$  can be analyzed mainly by E/p and  $n_e$ .

Since the larger  $I_{\rm net}$  induces the larger  $E_z$  from Eq. (3), at the same pressure, the larger  $D_{\rm obs}$  is associated with the larger  $E_z/p$  in spite of the smaller  $I_{\rm back}$  because the larger  $D_{\rm obs}$  corresponds to the smaller  $I_{\rm back}$  as pointed out above. From Eqs. (2) and (4), this means that the larger  $D_{\rm obs}$  is given by a gas with the smaller  $n_e/Q_m \overline{\epsilon}^{1/2}$  at the same pressure. The smaller  $n_e$  is given by a gas with the larger  $t_i$  because, in Eq. (8), term 2 is dominant for the major part of the pulse duration in the pressure region of interest, as described already. Therefore, if we ignore the variation of  $Q_m \overline{\epsilon}^{1/2}$  among gases,

the larger  $D_{\rm obs}$  is given by a gas with the larger  $t_i$  or  $pt_i$  at the same pressure under the larger  $E_{\rm z}/p$ , as described above. This is the principal conclusion on the relationship between  $D_{\rm obs}$  and  $t_i$ . Hereafter, data on  $D_{\rm obs}$  for various polyatomic gases will be analyzed on the basis of this conclusion. The value of  $D_{\rm obs}$  can give information on  $t_i$  for the pulse duration, which is difficult to be measured directly.

The  $E/p-pt_i$  curves for several gases have been obtained experimentally by Felsenthal and Proud<sup>4</sup> as seen in Fig. 16 of Ref. 4 in which  $pt_i$  increases with decreasing E/p. The above conclusion suggests that the  $E/p-pt_i$  curve for a gas giving the larger  $D_{\rm obs}$  at the same pressure must be placed at the upper side in such a figure. In fact, in Fig. 16 of Ref. 4, the values of E/p at the same  $pt_i$  are in the order

$$CCl_2F_2 > SF_6 > N_2 > O_2 > Ar > He$$
. (11)

This is the same order as for  $D_{\rm obs}$  at the same pressure as seen in Figs. 6 and 7 of Ref. 5. The relationship between He and Ar has been discussed in part I. <sup>1</sup>

As demonstrated by Felsenthal and Proud, <sup>4</sup> the value of  $t_i$  for a gas with no experimental value of  $t_i$  can be determined by Eq. (6) from swarm parameters  $(w, \alpha/p,$  and  $\eta/p)$ . Values of  $\eta/p$  have been measured as functions of E/p for some gases. According to references, values of  $\eta/p$  are in the order<sup>31-34</sup>

$$SF_6 \approx CCl_2F_2 \gg N_2O > O_2 \gg CO_2$$
 (12)

and ratios of  $\eta/\alpha$  are in the order

$$CCl_2F_2 \gtrsim SF_6 > N_2O > O_2 > CO_2$$
 (13)

For  $E/p > 60~\rm V~cm^{-1}~Torr^{-1}$ , the ratio of  $\eta/\alpha$  is negligibly small for  $O_2$  and  $CO_2$ . On the other hand, since the number density of gas molecules is  $3.3 \times 10^{18}~\rm cm^{-3}$  at  $20~\rm ^{\circ}C$  and  $100~\rm Torr$ , the half-life of secondary electrons for electron-attachment processes is less than 1 ns for the rate constant larger than  $2 \times 10^{-10}~\rm cm^3/s$ . Such large rate constants are reported for  $O_2$  ( $3.4 \times 10^{-10}~\rm cm^3/s$  at  $6.7~\rm eV^{35}$ ),  $N_2O$  ( $9.2 \times 10^{-10}~\rm cm^3/s$  at  $2.4~\rm eV^{35}$ ),  $SF_6$  ( $2 \times 10^{-7}~\rm cm^3/s$  at thermal energy  $^{36}$ ), and  $CCl_2F_2$  ( $5.6 \times 10^{-9}~\rm cm^3/s$  at  $1.08~\rm eV^{36}$ ). Then, the contribution of electron-attachment processes may be important in  $CCl_2F_2$ ,  $SF_6$ , and  $N_2O$ . In fact, as seen in Fig. 7 of Ref. 5, values of  $D_{\rm obs}$  in these gases are larger than in other gases; they are in the order

$$CCl_2F_2 > SF_6 > N_2O > CH_4$$
, (14)

as predicted from the ordering (12) and (13). This fact suggests that  $t_i$  is lengthened due to the large  $\eta/p$  in Eq. (6).

For the gas with no appreciable electron-attachment process, the value of  $pt_i$  can be estimated as  $p/w\alpha$ . Although E/p varies with t and p, from Figs. 6-8, we attempt to estimate the value of  $p/w\alpha$  at 100 V cm<sup>-1</sup> × Torr<sup>-1</sup> as the representative value for gas pressures between 20 and 50 Torr. From data on w and  $\alpha/p$  in Ref. 8, values of  $p/w\alpha$  are in the order

$$N_2 > CO_2 > CH_4 > O_2 > H_2$$
 (15)

This order is almost the same with the order of  $D_{obs}$  in

Fig. 7 of Ref. 5 except CH<sub>4</sub> because  $D_{\rm obs}$  for CH<sub>4</sub> is rather larger than for N<sub>2</sub>. This fact supports the present conclusion on the relationship between  $D_{\rm obs}$  and  $t_i$ .  $D_{\rm obs}$  for CH<sub>4</sub> is attributed to the large value of  $Q_m \, \overline{\epsilon}^{1/2}$  for CH<sub>4</sub> as described already.

On the other hand, for the gas with no electron-attachment process, the value of  $pt_i$  can also be estimated by the following equation instead of  $p/w\alpha$ :

$$\frac{1}{n_0 t_i} = \left(\frac{2}{m}\right)^{1/2} \int_{1.\,\mathrm{P.}}^{\infty} \epsilon^{1/2} \sigma_i(\epsilon) F(\epsilon) d\epsilon \ , \tag{16}$$

where I.P. is the ionization potential of a gas,  $\sigma_i(\epsilon)$  the total ionization cross section of a gas for electrons with energy  $\epsilon$ , and  $F(\epsilon)$  the distribution function of  $\epsilon$ . Data on  $\sigma_i$  for lower-energy electrons are given by Rapp and Englander-Golden. Equation (16) indicates that, at the same pressure, the larger  $D_{\rm obs}$  or  $t_i$  is given by the gas with the higher I.P., the slower initial slope of  $\sigma_i$ , and the larger distribution of  $\epsilon$  at the lower energy.

There are few available data on  $F(\epsilon)$  in the E/p region of interest. We can find some data on  $D_L/\mu$  as functions of E/p between 50 and 100 V cm<sup>-1</sup> Torr<sup>-1</sup> for estimation of  $\overline{\epsilon}$  for  $H_2$ , <sup>10</sup>  $N_2$ , <sup>13</sup>  $O_2$ , <sup>37</sup>  $CO_2$ , <sup>38</sup>  $CH_4$ , <sup>18</sup>  $SF_6$ , <sup>39</sup> and  $CCl_2F_2$ . <sup>40</sup> Values of  $\overline{\epsilon}$  for these gases estimated on the assumption of the Maxwellian are about 7–8 eV except  $CCl_2F_2$  (5.5 eV) at 100 V cm<sup>-1</sup> Torr<sup>-1</sup> and are in the order

$$CH_4 > H_2 \approx O_2 \approx CO_2 \approx (SF_6) > N_2 > (CCl_2F_2)$$
(17)

at 50 V cm<sup>-1</sup> Torr<sup>-1</sup>. The values for SF<sub>6</sub> and CCl<sub>2</sub>F<sub>2</sub> are extrapolated from data at E/p above 100 V cm<sup>-1</sup> Torr<sup>-1</sup>.

Let us suppose that  $\epsilon$  is lowered by  $Q_t f$ , where  $Q_t$  is the total collision cross section and f the mean fractional energy loss per collision which is given by  $^{41}$ 

$$f = 1.74 \times 10^{-14} w^2 / k_{\pi} \,, \tag{18}$$

where  $k_T$  is the Townsend energy factor  $(k_T=39.8\ D_L/\mu$  at  $20\,^{\circ}\mathrm{C}^{11}$  for the Maxwellian). Values of  $Q_t f$  from Eq. (18) are shown as functions of  $\overline{\epsilon}$  in Fig. 12. Data on  $Q_t$  are cited from references as follows;  $H_2$ ,  $^{42}$   $N_2$ ,  $^{20}$   $O_2$ ,  $^{43,44}$   $CO_2$ ,  $^{20}$   $CH_4$ ,  $^{19}$  and  $SF_6$ . When the curves in Fig. 12 are compared with the ordering (17), the high value of  $\overline{\epsilon}$  for  $CH_4$  at lower E/p is attributed to the small value of  $Q_t f$  at lower  $\overline{\epsilon}$  because of the special dependence of  $D_L/\mu$  on  $E/p^{18}$ ; the low value of  $\overline{\epsilon}$  for  $N_2$  is attributed to the large value of  $Q_t f$ , and the intermediate values for  $O_2$ ,  $CO_2$ , and  $SF_6$  are also understood by their curves of  $Q_t f$  from Eq. (18). This means that  $Q_t f$  from Eq. (18) is useful to estimate  $F(\epsilon)$  qualitatively, though there is some problem for the application of Eq. (18) to the higher E/p region.

Now, we examine the order of  $D_{\rm obs}$  as seen in Fig. 7 of Ref. 5 on the basis of Eq. (16). When data on I. P. and  $\sigma_i$  are compared for N<sub>2</sub>O and N<sub>2</sub>,  $t_i$  for N<sub>2</sub>O must be smaller than for N<sub>2</sub> because of the large  $Q_t f$  for N<sub>2</sub>. However,  $D_{\rm obs}$  for N<sub>2</sub>O is larger than for N<sub>2</sub>. This fact suggests that  $D_{\rm obs}$  or  $t_i$  for N<sub>2</sub>O may be affected appreciably by the electron-attachment process which is ignored in Eq. (16). Similarly, in the comparison between CH<sub>4</sub> and N<sub>2</sub>,  $D_{\rm obs}$  for CH<sub>4</sub> rather larger than for N<sub>2</sub> can be explained only by the large  $Q_m(\vec{\epsilon})\vec{\epsilon}^{1/2}$  for CH<sub>4</sub>. The value

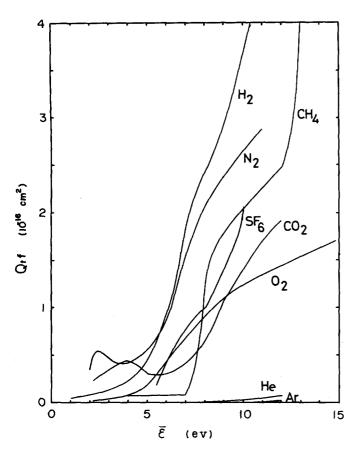


FIG. 12. The estimated values of  $Q_t f(Q_t)$ : total collision cross section for electrons; and f: mean fractional energy loss per collision) as functions of the mean electron energy.

of  $D_{\rm obs}$  for  $N_2$  rather larger than for  ${\rm CO}_2$  is attributed to the higher I. P., the smaller  $\sigma_i$ , and the larger  $Q_t f$  for  $N_2$  than for  ${\rm CO}_2$ . The value of  $D_{\rm obs}$  for  ${\rm O}_2$  smaller than for  ${\rm CO}_2$  is attributed to the lowest I. P. among gases compared in the ordering (15). On the other hand, for  ${\rm H}_2$ , the value of  $D_{\rm obs}$  is the lowest among polyatomic gases as seen in Fig. 7 of Ref. 5. This fact can be interpreted in terms of  $p/w\alpha$  as seen in the ordering (15) because of the largest  $\alpha/p$  for  ${\rm H}_2$  at 100 V cm<sup>-1</sup> Torr<sup>-1</sup>. However, the largest  $\alpha/p$  cannot be derived from Eq. (16) because  ${\rm H}_2$  has a high I. P., a small  $\sigma_i$ , and a large  $Q_t f$ .

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