

compared to that of the nitrates. Any deviations in correspondence due to the nitrate anion should then be apparent. The results of these calculations are shown in Table VI. The degrees of constancy in  $\nu$  are about the same for both the alkali nitrates and the alkali chlorides. It would therefore appear that in spite of the complex structure of the nitrate anion, molten

alkali nitrates obey the corresponding-states laws as well as do the simpler alkali halides.

#### ACKNOWLEDGMENT

The author wishes to acknowledge the assistance of James McCormick who carried out many of the measurements.

THE JOURNAL OF CHEMICAL PHYSICS

VOLUME 44, NUMBER 10

15 MAY 1966

## Gross- and Partial-Ionization Cross Sections for Electrons on Water Vapor in the Energy Range 0.1–20 keV

J. SCHUTTEN, F. J. DE HEER, H. R. MOUSTAFA, A. J. H. BOERBOOM, AND J. KISTEMAKER

*FOM-Laboratorium voor Massascheiding, Amsterdam, The Netherlands*

(Received 23 December 1965)

Measurements have been made on the gross-ionization cross sections for 0.1–20-keV electrons on water vapor at pressures of about  $3 \times 10^{-4}$  torr. The results are compared with the cross sections of  $H_2$  and  $O_2$ . Within the experimental error of 15% we found no deviations from the additivity rule. The energy dependence of the cross section satisfies the Bethe-Fano-Platzman relation for collision-induced dipole transitions at energies exceeding 400 eV. A comparison is made with photoionization and inelastic electron-scattering cross sections. Some results are given on the partial cross sections for the production of fragment ions for 0.02–2-keV electrons on water vapor.

### 1. INTRODUCTION

AS there is much interest for ionization of water vapor by electrons in radiation physics and amazingly little has been done in this field, measurements on this reaction have been carried out. This work is an extension of the research on gross- and partial-ionization cross sections for electrons on noble and diatomic gases<sup>1–4</sup> and on hydrocarbons.<sup>5</sup> We discuss our measurements on gross ionization in Secs. 2 and 3 and on partial ionization in Sec. 4.

### 2. EXPERIMENTAL PROCEDURE FOR THE GROSS-IONIZATION CROSS SECTION

#### Apparatus

The apparatus used (Fig. 1), is in principle the same as described before.<sup>1</sup> The ionization is determined by the conventional condenser technique. The electrons produced by an electron gun, are accelerated to energies between 0.1 and 20 keV. The beam is aligned by an axial magnetic field and shot into a collision chamber containing  $H_2O$  gas. The positive ions produced by the

electron beam along a well-defined path length are collected on one of the side electrodes by means of a transverse electric field.

The current of the primary electron beam is measured at the Faraday cage. The collision chamber has a wide-open pipe connection with a container with ice which is kept at a constant temperature of about  $-80^\circ C$ . This is done by means of a surrounding Dewar filled with mixtures of ether and dry ice or acetone and dry ice. From the temperature of these mixtures the pressure of the water vapor in the collision chamber is derived. The gross-ionization cross section can be calculated from

$$\sigma = (I_+/I_-)(lp)^{-1}[T/(273 \times 3.535 \times 10^{16})] \text{ cm}^2, \quad (1)$$

where  $I_+$  is the current of positive ions to the measuring electrode;  $I_-$  is the electron current to the Faraday cage;  $l$  is the length from which ions are extracted in centimeters;  $p$  is the gas pressure in torricelli;  $T$  is the temperature of the gas in degrees Kelvin (room temperature). Details on the current measurements and the suppression of secondary emission of electrons and the correction for fast scattered electrons have been extensively discussed in Ref. 1.

#### Determination of the Pressure of the Water Vapor

The pressure of the water vapor depends on the temperature of the bath around the container with ice. With mixtures of ether and dry ice and of acetone and dry ice we got temperatures of  $-81.0^\circ$  and  $-84.3^\circ C$ ,

<sup>1</sup> B. L. Schram, F. J. de Heer, M. J. v. d. Wiel, and J. Kistemaker, *Physica* **31**, 94 (1965).

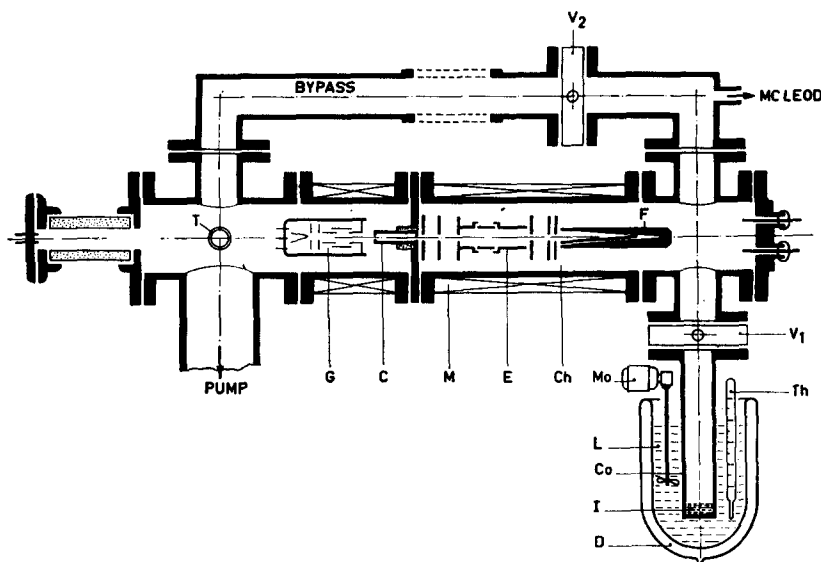
<sup>2</sup> B. L. Schram, H. R. Moustafa, J. Schutten, and F. J. de Heer, *Physica* (to be published).

<sup>3</sup> B. L. Schram, A. J. H. Boerboom, and J. Kistemaker, *Physica* **32**, 185 (1966).

<sup>4</sup> B. L. Schram, *Physica* **32**, 197 (1966).

<sup>5</sup> B. L. Schram, M. J. v. d. Wiel, F. J. de Heer, and H. R. Moustafa, *J. Chem. Phys.* **44**, 49 (1966).

FIG. 1. Schematic view of the apparatus used for measuring ionization cross sections. G is the electron gun and accelerating system; C is the collimator; T is the cold trap; Ch is the collision chamber; E is the electrode system; F is the Faraday cup; M is the magnetic coil; Co is the container; I is the ice; D is the Dewar; L is the liquid at about  $-80^{\circ}\text{C}$ ; Mo is the stirring motor; Th is the thermometer;  $V_1$  and  $V_2$  are the valves.



respectively. As is seen from Fig. 2 this, respectively, corresponds with vapor pressures of  $3.4 \times 10^{-4}$  and  $1.9 \times 10^{-4}$  torr. The temperature of the bath has been determined with a thermometer which was calibrated to an accuracy of  $0.5^{\circ}\text{C}$ . The variation of the temperature in the bath of constant temperature and in the container with dry ice is estimated to be less than  $0.5^{\circ}\text{C}$ . The total possible error in the determination of the temperature of the ice thus is about  $1^{\circ}\text{C}$ , corresponding with a possible error in the measurement of the  $\text{H}_2\text{O}$  gas pressure of 13% (see Fig. 2).

In order to avoid the poisoning effect of the water on the oxyd cathode in the electron source, the bypass was closed during the measurements. The collimator has such a large pumping resistance that a pressure of  $10^{-6}$  torr is obtained in the pumping chamber, while operating with a target pressure of about  $3 \times 10^{-4}$  torr. Because of the low pumping speed through the collimator the gas pressure in the collision chamber could be taken equal to the vapor pressure above the ice in the container. A Veeco ionization gauge was used as a monitor for the vapor pressure during the ionization

measurements and gave a control on variations in the pressure. During a complete set of measurements the fluctuations in the pressure could be kept lower than 3%.

As the apparatus could not be baked out and in the case of water vapor no cold trap could be used as in Ref. 1, the influence of the restgas was larger than usual. The corresponding contribution to the slow ion current to the side electrode had to be subtracted. The background pressure could be measured with a McLeod gauge and was equal to about  $5 \times 10^{-5}$  torr.

#### Additional Experimental Details and Estimation of Errors

The electrical measurement on condenser and Faraday cage were performed with a digital voltmeter, precision resistors, and a quick-acting switch. In this way the ratio  $I_+/I_-$  [see Eq. (1)] could be determined with an accuracy of better than 1%, except at energies above 10 keV, where leakage currents and zero offset had some influence.

The ion currents due to the restgas amounted to 30% of the total value in some cases which causes an extra possible error of about 6% in the value of  $I_+/I_-$ .

The total uncertainty in the cross section calculated with Eq. (1) is mainly due to the determination of the pressure of the water vapor and the correction for the restgas and is estimated to be about 15% in total. The reproducibility of the measurements over a period of several days seemed to be better than 4% at both pressures used.

### 3. RESULTS ON GROSS IONIZATION AND COMPARISON TO PHOTOIONIZATION CROSS SECTIONS

In Table I the gross ionization cross sections are given which have been determined at different impact

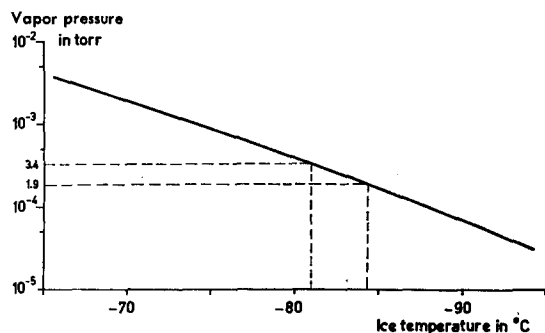


FIG. 2. Water vapor pressure as a function of temperature [values from *Handbook of Chemistry and Physics*, C. D. Hodgman *et al.*, Eds. (Chemical Rubber Publ. Co., Cleveland, Ohio, 1957), 45th ed., p. D-91].

TABLE I. Ionization cross sections for electrons on H<sub>2</sub>O gas, compared with the sum of the cross sections of H<sub>2</sub> and ½O<sub>2</sub> from Refs. 1 and 2.

$E_{e1}$ (in keV)	$\sigma_{H_2O}$ (in units of $10^{-16}$ cm <sup>2</sup> /molecule)	$\sigma_{H_2} + \frac{1}{2}\sigma_{O_2}$
0.08	1.98	...
0.09	2.01	...
0.1	1.98	2.10
0.12	1.96	2.05
0.16	1.83	1.89
0.2	1.71	1.75
0.3	1.46	1.34
0.4	1.26	1.22
0.5	1.12	1.06
0.6	1.00	0.94
0.8	0.77	0.75
1	0.622	0.649
2	0.375	0.374
3	0.261	0.271
4	0.220	0.214
5	0.183	0.182
6	0.160	0.153
8	0.123	0.121
10	0.101	0.101
12	0.088	0.086
14	0.078	0.076
16	0.064	0.069
18	0.058	0.062
20	0.054	0.058

energies of the electrons incident on water vapor. For comparison we computed  $(\sigma_{H_2} + \frac{1}{2}\sigma_{O_2})$  from values of Refs. 1 and 2 in order to check the additivity. Within the experimental uncertainty of about 15% for water vapor there is no deviation from the additivity rule.

Theoretical calculations of ionization cross sections for charged particles have been made on the basis of Born approximation. Starting from the equations originally derived by Bethe,<sup>6</sup> it was shown by Miller and Platzman<sup>7</sup> that the ionization cross section of high-energy electrons can be represented by

$$\sigma = (4\pi a_0^2 R / E_{e1}) M_i^2 \ln c_i E_{e1}, \quad (2)$$

where  $a_0$  is the first Bohr radius of atomic hydrogen,  $R$  is the Rydberg energy,  $E_{e1}$  is the electron energy, and  $c_i$  is a constant. The quantity  $M_i^2$  is related to the optical oscillator strength  $(df/dE)$ , where  $E$  is the excitation energy of the molecule in its continuum. We have

$$M_i^2 = \int_{I.P.}^{\infty} \frac{df}{dE} \frac{R}{E} \eta(E) dE, \quad (3)$$

where  $\eta(E)$  is the efficiency of ionization when the molecule has acquired an excitation energy  $E$ . Due to the fact that superexcited states (states with excitation energy exceeding the ionization potential) will decay either by ionization or dissociation,  $\eta(E)$  will be smaller than unity.

<sup>6</sup> H. Bethe, Ann. Phys. 5, 325 (1930).

<sup>7</sup> W. F. Miller and R. L. Platzman, Proc. Phys. Soc. (London) A70, 299 (1957).

In Formula (2) a correction has to be made for relativistic effects at electron energies exceeding 1 keV (see Ref. 1). We have to replace  $E_{e1}$  by  $E'_{e1}$ ; in our energy range we obtain in good approximation

$$E'_{e1} = \frac{1}{2} m_0 v^2 = \frac{1}{2} m_0 c^2 \left\{ 1 - \frac{1}{(1 + E_{e1}/m_0 c^2)^2} \right\}, \quad (4)$$

where  $m_0$  is the rest mass of the electron.

In order to verify whether our cross sections satisfy the theoretical relation, a plot of  $\sigma E'_{e1}/4\pi a_0^2 R$  versus  $\ln E'_{e1}$  was made both for H<sub>2</sub>O and for H<sub>2</sub> + ½O<sub>2</sub> (see Fig. 3). A linear relation is present between about 0.5 and 10 keV. Above 10 keV the water points deviate from the straight line, which is probably due to enlargement of inaccuracy of the measurements at the highest energies. We find by means of a least-square analysis  $M_i^2 = 3.14 \pm 0.047$  and  $c_i = 0.075 \pm 0.008$  eV<sup>-1</sup> for H<sub>2</sub>O, and  $M_i^2 = 3.19 \pm 0.045$  and  $c_i = 0.065 \pm 0.007$  eV<sup>-1</sup> for H<sub>2</sub> + ½O<sub>2</sub>.

By using the relation between the oscillator strength and the photoionization cross section we can write (3) as

$$M_i^2 = \frac{mcR}{\pi e^2 h} \int_{I.P.}^{\infty} \frac{\sigma_{ph}(E)}{E} dE = 0.124 \int_{985}^{\infty} \frac{\sigma_{ph}(\lambda)}{\lambda} d\lambda, \quad (5)$$

where  $\sigma_{ph}(\lambda)$  is the photoionization cross section given in megabarns at wavelength  $\lambda$ , corresponding to the excitation energy  $E$ . The photoionization results  $\sigma_{ph}(\lambda)$  of Wainfan, Walker, and Weissler<sup>8</sup> extending between 985 and 473 Å were extrapolated down to zero wavelength. By integrating  $[\sigma_{ph}(\lambda)/\lambda]$  numerically between the limits 985 and 0 Å we could roughly estimate the value for  $M_i^2$ . This value may contain an extra error of about 10% as a consequence of the procedure of extrapolation. Wainfan *et al.* estimate a probable error of 30% in their measurements. The value of  $M_i^2$  derived by this method is  $M_i^2 = 2.59$ , being about 25% lower than the value obtained from our electron ionization experiments (see Table II). However, when we analyze the

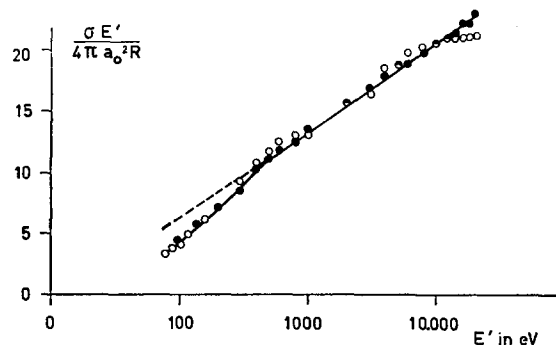


Fig. 3. Ionization of H<sub>2</sub>O by electrons, compared with similar measurements of H<sub>2</sub> + ½O<sub>2</sub> (see Refs. 1 and 2). ○ is H<sub>2</sub>O; ● is H<sub>2</sub> + ½O<sub>2</sub>.

<sup>8</sup> N. Wainfan, W. C. Walker, and G. L. Weissler, Phys. Rev. 99, 542 (1955).

data of Wainfan *et al.*<sup>8</sup> and calculate  $M_i^2$  for  $N_2$  and  $O_2$  from their photoionization curves, we get mostly values which are lower than our values obtained from electron ionization<sup>1,2</sup> (see Table II). The ratio between our  $M_i^2$  values and those of Wainfan *et al.*<sup>8</sup> for  $N_2$ ,  $O_2$ , and  $H_2O$  is almost a constant. As our measurements for  $N_2$  and  $O_2$  are much more accurate than for  $H_2O$ , it seems even possible that our value of  $M_i^2$  for  $H_2O$  is more accurate than the estimated 15%.

The same procedure applied to the photoionization results on water vapor of Metzger and Cook<sup>9</sup> leads to a value of  $M_i^2$ , which is about 5% smaller than in the case of Wainfan *et al.*<sup>8</sup> However, because the measurements of Metzger and Cook extend only down to 600 Å, the extrapolation procedure down to zero wavelength is rather arbitrary. Photoionization measurements on water vapor of others as Watanabe and Jursa<sup>10</sup> do not extend to sufficiently low wavelengths to be compared with our electron measurements.

The value of  $M_i^2$  can also be obtained from inelastic scattering experiments at small angles. For water vapor such measurements have been carried out by Lassettre and White.<sup>11</sup> This has been done by measuring the electron loss of the scattered electrons in a high-resolution electron spectrometer and their intensity at several angles as a function of energy loss.<sup>12</sup> The optical oscillator strength ( $df/dE$ ), which is proportional to the differential-scattering cross section at zero angle for electrons with energy loss  $E$ , has been obtained for different  $E$  values by Lassettre and White.<sup>11</sup> They extrapolated their experimentally found results from Ref. 8 through 0.58 deg to zero angle. We have used their ( $df/dE$ ) values to calculate again  $M_i^2$  by means of Eq. (3). The value of  $\eta(E)$  in this equation has been taken from the work of Wainfan *et al.*<sup>8</sup> who measured both the photoabsorption and photoionization cross sections for water vapor. They found little variation in  $\eta(E)$  as a function of excitation energy over a large range of  $E$ , and therefore we took  $\eta(E) = 0.70$  independent of  $E_0$ . Combining this value with the ( $df/dE$ ) data of Lassettre and White,<sup>11</sup> we find by numerical integra-

TABLE II. Values of  $M_i^2$ .

Gas	Electron ionization	Photoionization <sup>a</sup>	Ratio
$N_2$	3.85	3.23	1.19
$O_2$	4.75	4.00	1.19
$H_2O$	3.14	2.59	1.21

<sup>a</sup> From Ref. 8.

<sup>9</sup> P. H. Metzger and G. R. Cook, J. Chem. Phys. **41**, 642 (1964).

<sup>10</sup> K. Watanabe and A. S. Jursa, J. Chem. Phys. **41**, 1659 (1964).

<sup>11</sup> E. N. Lassettre and E. R. White, Sci. Rept. 12 OSURF, Contract No. AF 19 (122)-642, Ohio State University, 1958.

<sup>12</sup> E. N. Lassettre, J. Chem. Phys. **40**, 1222 (1964).

TABLE III. Relative ion abundance with 100-eV electron energy.

Ion species	Mann <i>et al.</i> <sup>a</sup>	This work
$H_2O^+$	100	100
$OH^+$	23.2	25.8
$O^+$	2.0	2.65
$H_2^+$	0.07	0.30
$H^+$	5.0	29.6

<sup>a</sup> See Ref. 11.

tion  $M_i^2 = 2.94$ , which is in reasonable agreement with our result.

#### 4. PARTIAL-IONIZATION CROSS SECTIONS

The analysis of the cracking pattern of the  $H_2O$  molecule caused by electron bombardment can be done in principle with a mass spectrometer. However it is well known that fragment ions may possess appreciable amounts of initial kinetic energy. The lighter the mass the higher the energy. Therefore the abundance of  $H^+$  and  $H_2^+$  ions in the case of cracking of a  $H_2O$  molecule is always measured far too low if a normal Nier-type mass-spectrometer ion source has been used. Apparently this has been a reason why not much work on  $H_2O$  has been done. The only publication which really treats the determination of the cracking pattern is by Mann *et al.*<sup>13</sup> However it gives only one bombardment energy of 100 eV. Moreover we see in Table III that the results for the light masses are dubious, due to mass discrimination. For the influence of the isotope effect (H or D) on the fragmentation of the water molecule due to electron impact we refer to the work done by Fiquet-Fayard.<sup>14</sup> Knewstubb and Tickner<sup>15</sup> measure the relative abundance of  $H_3O^+(H_2O)_n$  with  $n=0,1, 2, \dots$ , in ionized water vapor at pressures of 0.3 to 0.6 torr. These ions originate from secondary processes. No primary ions like  $H_2O^+$ ,  $H^+$ ,  $OH^+$ ,  $O^+$ , or  $H_2^+$  have been found.

To avoid the mass discrimination effect we developed a special cycloidal mass spectrometer with an extremely high-ion extraction efficiency, using an open ion source construction in  $\vec{E} \times \vec{B}$  configuration. The distance between source and Faraday collector was 126 mm and the mass resolution was 40.

We measured the fragments  $H_2O^+$ ,  $OH^+$ ,  $O^+$ ,  $O^{2+}$ ,  $H_2^+$ , and  $H^+$ . In order to check the completeness of collection of each ion species, for each ion the peak height was measured as a function of electric or magnetic field at a constant value of  $B^2/E$ . The first four ions showed a saturation effect at about 100-V/cm electric-field strength. Below this value the collection efficiency is apparently less than one which is caused by

<sup>13</sup> M. M. Mann, A. Hustrulid, and J. T. Tate, Phys. Rev. **58**, 340 (1940).

<sup>14</sup> Fl. Fiquet-Fayard, Bull. Soc. Chim. Belges **73**, 373 (1964).

<sup>15</sup> P. F. Knewstubb and A. W. Tickner, J. Chem. Phys. **38**, 1031 (1963).

TABLE IV. Partial-ionization cross sections for electrons on water vapor in units of  $10^{-18}$  cm<sup>2</sup>/molecule.

$E_{el}$ (in keV)	H <sub>2</sub> O <sup>+</sup>	OH <sup>+</sup>	O <sup>+</sup>	O <sup>2+</sup>	H <sub>2</sub> <sup>+</sup>	H <sup>+</sup>	Total
0.02	0.42	0.03	0.0009	...	0.0018	0.007	0.46
0.03	0.76	0.13	0.0037	...	0.0022	0.078	0.97
0.05	1.07	0.24	0.017	...	0.0030	0.24	1.57
0.10	1.32	0.34	0.035	...	0.0039	0.39	2.10
0.15	1.16	0.31	0.036	0.0006	0.0036	0.37	1.89
0.20	1.08	0.28	0.035	0.0014	0.0029	0.36	1.75
0.50	0.64	0.17	0.019	0.0013	0.0018	0.23	1.06
0.70	0.50	0.14	0.014	0.0009	0.0016	0.17	0.83
1.00	0.41	0.11	0.011	0.0006	0.0008	0.13	0.65
1.50	0.29	0.08	0.008	...	...	0.11	0.49
2.00	0.23	0.06	0.006	...	...	0.08	0.37

the kinetic energy which the ions obtain at dissociation of the parent molecule. For H<sup>+</sup>, saturation could not be obtained, even at the maximum obtainable field strength of 1000 V/cm. But from the shape of the curve we have the impression that the extracted H<sup>+</sup> current is 5% from saturation. Some control measurements were done on He and Ne gas. The relative abundances of He<sup>2+</sup> to He<sup>+</sup> as well as of Ne<sup>2+</sup> to Ne<sup>+</sup> fitted with the results by Schram *et al.*<sup>3</sup> within 20%. Better results could not be expected from such a simple ion source, where no precautions against secondary electrons were taken.

In Table III we see the big relative difference between our results and those of Mann *et al.*<sup>13</sup> for H<sup>+</sup> and H<sub>2</sub><sup>+</sup>. The results of the partial-ionization cross sections for electrons on water vapor are given in Table IV. They have been obtained by a determination of the relative amounts of the fragment ions and normalization of their sum on the gross-ionization cross sections in Table I.

Below 500-eV impact energy, the normalization factor appeared to be constant. Consequently, for the measurements below 100 eV, where no gross-ionization cross sections were available, we have used the same normalization factor for calculating partial-ionization cross sections. From 500 to 2000 eV the normalization factor decreased gradually about 20% which might be due to insufficient precaution in this mass-spectrometer ion source against secondary electrons. As the results on the fractionizing pattern of the H<sub>2</sub>O molecule seem to be unique at this moment, we still present them above 500 eV although the uncertainty is probably 25%.

From Table IV we see the main part of the fragment ions is composed of H<sub>2</sub>O<sup>+</sup>, OH<sup>+</sup>, and H<sup>+</sup>. The formation of O<sup>+</sup>, O<sup>2+</sup>, and H<sub>2</sub><sup>+</sup> seems to be very improbable. The ratio H<sub>2</sub>O<sup>+</sup>/OH<sup>+</sup>/H<sup>+</sup> seems to be rather constant over the whole energy range between 100 and 2000 eV. This might indicate that these ions all originate from

the same type of collision-induced dipole interaction<sup>16</sup> as indicated by Formula 2 and by Fig. 3.

The relatively large fraction of H<sup>+</sup> and OH<sup>+</sup> ions in Table IV shows that there is a big chance for a valence electron to be ejected. H<sub>2</sub>O contains two OH bindings. If one of these bindings is broken, this gives rise to either an OH radical plus an H<sup>+</sup> ion or to an OH<sup>+</sup> ion plus a neutral H atom, both formed with equal chance. We observe, indeed, that the cross sections for the formation of OH<sup>+</sup> and H<sup>+</sup> ions are about equal.

If the ejected electron is no valence electron, H<sub>2</sub>O<sup>+</sup> can be formed. The cross section for H<sub>2</sub>O<sup>+</sup> is about 1.6 to 2 times larger than for H<sup>+</sup> and OH<sup>+</sup> together at electron energies higher than 50 eV. Because less energy is needed to eject a nonvalence electron this seems reasonable.

The smaller abundance of O<sup>+</sup>, O<sup>2+</sup>, and H<sub>2</sub><sup>+</sup> follows from the fact that in the case of their formation two bindings have to be broken or two valence electrons have to be ejected. It is trivial that the fraction of O<sup>2+</sup> is smaller than that of O<sup>+</sup>.

Auger processes due to inner-shell excitation of oxygen (1s ejection) start at about 0.54-keV impact energy. Above that energy they may contribute to formation of O<sup>2+</sup> and H<sub>2</sub><sup>+</sup>. If we make a rough estimation of the cross section of 1s excitation of oxygen by the Bethe relation at 1-keV impact energy (see Durup and Platzman<sup>17</sup>), we find a value of the order of  $10^{-19}$  cm<sup>2</sup>, which is the same order of magnitude as the cross sections for O<sup>2+</sup> and H<sub>2</sub><sup>+</sup> formation in Table IV.

The ratio of H<sub>2</sub>O<sup>+</sup>, OH<sup>+</sup>, and O<sup>+</sup> fragments in the case of photoionization measurements is reported to be 100:40:5 at 600 Å, uncorrected for detector efficiency.<sup>9,18</sup> The ratios of these fragments show the same order of magnitude in our electron experiments above 50-eV impact energy, as seen from Table IV.

#### ACKNOWLEDGMENTS

The authors wish to thank Professor R. L. Platzman for suggesting the experiment and to Professor A. Ore and Dr. M. Inokuti for their valuable comments. They also wish to express their thanks to B. Adamczyk, M.J. v. d. Wiel, and B. L. Schram for helpful cooperation.

This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (Foundation for Fundamental Research on Matter) and was made possible by financial support from the Nederlandse Organisatie voor Zuiver-Wetenschappelijk Onderzoek [Netherlands Organization for the advancement of Pure Research (ZWO)].

<sup>16</sup> R. L. Platzman, Intern. J. Appl. Radiation Isotopes **10**, 16 (1961).

<sup>17</sup> J. Durup and R. L. Platzman, Discussions Faraday Soc. **31**, 56 (1961).

<sup>18</sup> J. A. R. Samson and G. R. Cook, Bull. Am. Phys. Soc. **4**, 454 (1959).