

IONIZATION OF MOLECULAR HYDROGEN BY H^+ , H_2^+ , AND H_3^+ IONS

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We have studied the composition of the secondary ions produced by single collisions between H_2 molecules and primary H^+ , H_2^+ , and H_3^+ ions with energies from 5 to 180 kev. The total cross sections were determined: σ_0 , for electron capture by the primary ions, σ_- , for the formation of free electrons, and $\sigma_{H_2^+}$ and σ_{H^+} , for the formation of H_2^+ ions and protons. σ_{H^+} is considerably smaller than $\sigma_{H_2^+}$ over the entire range of energy investigated. The curves of $\sigma_{H^+}(v)$ for the primary ions H^+ and H_2^+ showed maxima at velocities of 1.7×10^8 cm/sec and 1.9×10^8 cm/sec respectively. The maximum value of σ_{H^+} was about 5×10^{-17} cm², approximately 100 times larger than the corresponding cross section for the case of electron bombardment.¹

INTRODUCTION

THE cross sections for ionization and charge exchange have been calculated theoretically only for the simplest cases, including the (H^+ , H) system. A number of authors²⁻⁴ have calculated the cross section for charge exchange in this system, while the cross section for ionization has been calculated by Bates and Griffing.⁵ Considerable experimental data, covering a wide range of energy,⁶⁻¹⁰ are available on the charge-exchange cross section for the (H^+ , H_2) system. However, the cross section for the ionization of hydrogen molecules by hydrogen ions has been determined only for energies below 40 kev.^{7,9,11} As to the composition of the secondary ions, we have only the very limited data by Keene.⁷

The aim of the present work was to study the ionization of hydrogen by H^+ , H_2^+ , and H_3^+ ions, and the distribution of e/m of the secondary ions over the relatively wide energy range from 5 to 180 kev.

Secondary ions can appear during collisions between protons and H_2 molecules as a result of the usual charge exchange, ionization of the molecules to form molecular ions, and dissociation of the molecular ions so formed, resulting in one or two new protons. We shall denote the cross sections for the corresponding processes by the letter σ with the upper indices c, i, and d respectively. The lower indices 01 and 02 will indicate the formation of either one or two positively-charged secondary ions from the hydrogen molecule. In addition, we must allow for possible processes in which the primary

proton captures one or two electrons, accompanied by the dissociation of the secondary molecular ion (indices cd and ccd). The process of single-electron capture plus dissociation was first studied by Lindholm,¹² who found that the cross section for this process, as in the usual types of charge exchange, depended strongly on the resonance defect ΔE . The quantity ΔE is in this case the difference between the recombination energy of the primary ion and the vertical dissociation energy of the secondary ion formed by the charge exchange.* The dissociation of H_2 molecules to form protons when bombarded with electrons and fast atomic particles should come about as the result of a transition to the ionized state in accordance with the Franck-Condon principle.

From the interaction potential-energy curves given in Massey and Burhop's monograph¹³ for various electronic states of the hydrogen molecule, we have compiled a table of possible processes by which secondary ions might be formed from the (H^+ , H_2) system.

The columns of the table list: (1) the serial number provisionally assigned to each process; (2) the formula for the process; (3) the notation for the cross section; (4) the excited state of the molecular ion corresponding to the given process; (5) is the recombination energy (or the energy of formation of a fast H^- ion from a proton) E_1 ; (6) the ver-

*By "vertical dissociation energy" we mean the energy of a transition, obeying the Franck-Condon principle, for the ground state of the molecule to the ionized state, with subsequent dissociation (See also reference 13).

1	2	3	4	5	6	7	8
N ₀	Process			E ₁ , ev	E ₂ , ev	ΔE, ev	ΔT, ev
1	<u>H</u> ⁺ + H ₂ → <u>H</u> + H ₂ ⁺	σ ₀₁ ^c	² Σg	13.6	15.6–18	2–4.4	—
2	<u>H</u> ⁺ + H ₂ → <u>H</u> + H + H ⁺	σ ₀₁ ^{cd}	² Σg	13.6	18	4.4	0
3	<u>H</u> ⁺ + H ₂ → <u>H</u> + H + H ⁺	σ ₀₁ ^{cd}	² Σu	13.6	28–32.5	14.4–18.9	5–7
4	<u>H</u> ⁺ + H ₂ → <u>H</u> ⁺ + H ₂ ⁺ + e	σ ₀₁ ⁱ	² Σg	—	15.6–18	15.6–18	—
5	<u>H</u> ⁺ + H ₂ → <u>H</u> ⁺ + H + H ⁺ + e	σ ₀₁ ⁱ	² Σg	—	18	18	0
6	<u>H</u> ⁺ + H ₂ → <u>H</u> ⁺ + H + H ⁺ + e	σ ₀₁ ⁱ	² Σu	—	28–32.5	28–32.5	5–7
7	<u>H</u> ⁺ + H ₂ → <u>H</u> ⁺ + 2H ⁺	σ ₀₂ ^{cd}	—	14.3	46–50	31.7–35.7	7.5–10
8	<u>H</u> ⁺ + H ₂ → <u>H</u> + 2H ⁺ + e	σ ₀₂ ^{cd}	—	13.6	46–50	32.4–36.4	7.5–10
9	<u>H</u> ⁺ + H ₂ → <u>H</u> ⁺ + 2H ⁺ + 2e	σ ₀₂ ^d	—	—	46–50	46–50	7.5–10

tical dissociation energy E_2 ; (7) $\Delta E = E_1 - E_2$, i.e., the energy spent in carrying out the indicated process, which is equal to the resonance defect for processes 1, 2, 3, and 8; (8) is the kinetic energy T acquired by the secondary proton formed during the process as the result of the dissociation of the molecular ion. The symbol for the primary particle is underlined in each formula.

When the primary particle is a molecular ion, the mechanism of secondary-particle formation from H_2 molecules must still be the same as in the case of the (H^+ , H_2) system, but since there is a much larger number of possible states for the primary particle, the processes are more numerous and more complex.

1. EXPERIMENTAL PROCEDURE

The apparatus in which the experiments were performed, and the methods employed, are described in detail in our previous papers.^{14,15} A beam of primary ions, uniform in composition and energy, was directed into a collision chamber where the pressure was between 1 and 1.5×10^{-4} mm Hg. This low pressure allowed only single collisions between the primary particles and the gas molecules. The pressure in other parts of the apparatus was kept below 5×10^{-6} mm Hg with the aid of differential pumps.

The secondary ions and free electrons produced in the gas were collected on the plates of a measuring capacitor located inside the collision chamber.

The total cross section σ_+ for the formation of secondary ions and the total cross section σ_- for the formation of free electrons were determined from the formulas

$$\sigma_+ = i_+ / i_1 N l, \quad (1)$$

$$\sigma_- = i_- / i_1 N l, \quad (2)$$

where i_+ and i_- are the positive and negative

saturation currents at the plates of the measuring capacitor. N is the number of gas molecules per cubic centimeter in the collision chamber, l is the length of the measuring electrodes, and i_1 is the primary ion beam current. Under the conditions of the experiments, the current i_1 was 1×10^{-7} to 1×10^{-6} amp, and the currents i_+ and i_- varied between 5×10^{-10} and 2×10^{-8} amp. The currents were measured by a mirror galvanometer with an ultimate sensitivity of 1.5×10^{-10} amp/division.

In order to analyze the e/m distribution of the secondary ions, a magnetic sector mass-spectrometer was connected to the collision chamber, in a plane perpendicular to the primary beam. The slow secondary ions were accelerated at the entrance to the analyzer, by an electric field. The analyzer was used to determine the ratio

$$\sigma_{H_2^+} / \sigma_{H^+} = \alpha_{H_2^+} / \alpha_{H^+}, \quad (3)$$

where $\alpha_{H_2^+}$ and α_{H^+} are the relative intensities of the H_2^+ and H^+ lines in the secondary ion spectrum, and $\sigma_{H_2^+}$ and σ_{H^+} are the corresponding cross sections for the formation of these secondary ions. The absolute values of the cross sections were determined from the formulas

$$\sigma_{H_2^+} = \sigma_+ \alpha_{H_2^+}, \quad (4)$$

$$\sigma_{H^+} = \sigma_+ \alpha_{H^+}. \quad (5)$$

The ion currents in the analyzer were between 2×10^{-10} and 2×10^{-13} amp, and were measured by an electrometer amplifier with a sensitivity of 2×10^{-14} amp/division.

The cross sections σ_+ and σ_- can be expressed in terms of the cross sections of the individual processes in the following way:

$$\sigma_+ = (\sigma_{01}^c + \sigma_{01}^i) + (\sigma_{01}^{cd} + \sigma_{01}^d) + 2(\sigma_{02}^d + \sigma_{02}^{cd} + \sigma_{02}^{cd}), \quad (6)$$

$$\sigma_- = \sigma_{01}^i + \sigma_{01}^d + 2\sigma_{02}^d + \sigma_{02}^{cd} + (\sigma_{12}^d + 2\sigma_{13}^d), \quad (7)$$

where the cross section σ_{12}^d accounts for the dis-

sociation of the primary molecular ion, H_2^+ or H_3^+ , with emission of an electron, while the cross section σ_{13}^d account also for the dissociation of H_3^+ ion with emission of two electrons. Subtracting (7) from (6) we get

$$\sigma_+ - \sigma_- = \sigma_{01}^c + \sigma_{01}^{cd} + \sigma_{02}^{cd} + 2\sigma_{02}^{ccd} - (\sigma_{12}^d + 2\sigma_{13}^d). \quad (8)$$

The first three terms on the right-hand side of Eq. (8) comprise the total cross section for the capture of one electron by a primary ion:

$$\sigma_0 = \sigma_{01}^c + \sigma_{01}^{cd} + \sigma_{02}^{cd}. \quad (9)$$

Since, according to the data given in reference 9, $\sigma_{02}^{ccd} \ll \sigma_+ - \sigma_-$ for protons, the total capture cross section σ_0 can be determined from the data obtained by the potential method, using the approximate formula

$$\sigma_0 = \sigma_+ - \sigma_-. \quad (10)$$

We have applied Eq. (10) also to the case of H_2^+ and H_3^+ as primary molecular ions, using the analogous approximation. The corrections necessary in this case, which must increase σ_0 slightly, can be introduced only when the cross sections σ_{12}^d and σ_{13}^d can be measured independently.

The cross section for the formation of secondary H_2^+ and H^+ can be expressed in terms of the process cross sections in the following way:

$$\sigma_{H_2^+} = \sigma_{01}^c + \sigma_{01}^i, \quad (11)$$

$$\sigma_{H^+} = \sigma_{01}^{cd} + \sigma_{01}^d + 2(\sigma_{02}^{cd} + \sigma_{02}^d + \sigma_{02}^{ccd}). \quad (12)$$

All the cross sections which we obtained were calculated "per molecule" and are expressed in cm^2 . The possible errors in determining the cross sections σ_+ , σ_- , $\sigma_{H_2^+}$, and σ_{H^+} are estimated to be $\pm 12\%$, equally caused by errors in measuring the current ($\pm 6\%$) and the gas pressure in the collision chamber ($\pm 6\%$). If the cross sections σ_0 are small compared with the corresponding σ_+ and

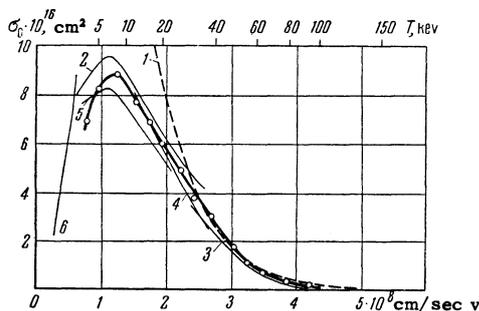


FIG. 1. Total cross section for capture of electrons by protons. \circ - data of this work, 1 - theoretical curve after Jackson and Schiff.³ Remaining curves are experimental, after the following: 2 - Keene,⁷ 3 - Ribe,⁸ 4 - Fogel,⁹ 5 and 6 - Stedeford and Hasted.¹⁰

σ_- , their measurement errors are considerably larger.

2. RESULTS OF THE MEASUREMENTS AND DISCUSSION OF THE RESULTS

1. Total Capture Cross Section σ_0 .

Figures 1, 2, and 3 show the dependence of σ_0 on v for H^+ , H_2^+ , and H_3^+ ions respectively.

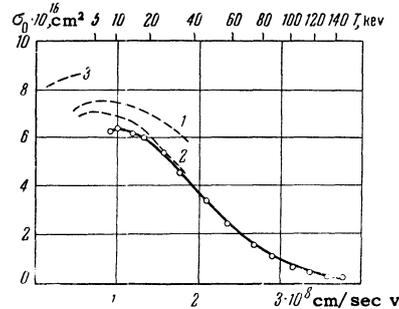


FIG. 2. Total cross section for capture of electrons by H_2^+ ions. \circ - data of this work. Experimental curves after the following: 1 - Keene,⁷ 2 and 3 - Stedeford and Hasted.¹⁰

Experimental data on charge exchange of protons in molecular hydrogen have also been obtained recently by Keene,⁷ Ribe,⁸ Forel,⁹ and Stedeford and Hasted.¹⁰ In these experimental studies, as in our work, it was really the total capture cross section that was being measured [see Eq. (9)]. All the experimental results, including our own, agree among themselves within the limits of experimental error. The curves of $\sigma_0(v)$ have a maximum at $T \approx 8$ kev, as first observed by Bartels.⁶

Figure 1 also shows the theoretical curve for the charge exchange of protons in atomic hydrogen, from the paper by Jackson and Schiff.^{3*} This curve agrees with the experimental results for the (H^+ , H_2) system in the velocity range $v > e^2/\hbar$, and gives too high a cross section for $v \leq e^2/\hbar$.

For the (H_2^+ , H_2) system (Fig. 2) we show also the data of Keene⁷ and Stedeford and Hasted.¹⁰ These agree, within experimental error, with the results of our work. The presence of a maximum in the $\sigma_0(v)$ curve is evidence that the (H_2^+ , H_2) system does not exhibit resonance. The electron capture probably occurs in an excited state of the H_2 molecule.

Electron capture by the H_3^+ ion is a complicated process, since there is no known stable state of the H_3 molecule. Apparently the capture of an elec-

*This curve practically coincides with the curves of Bates and Dalgarno² and Pradhan.⁴ In going from the (H^+ , H) system to (H^+ , H_2), the authors of references 2 and 3 double the corresponding cross sections, neglecting the molecular bonds.

tron by a H_3^+ ion leads to dissociation into an H_2 molecule and a hydrogen atom. The $\sigma_0(v)$ curve for H_3^+ ions has a maximum, which is difficult to interpret.

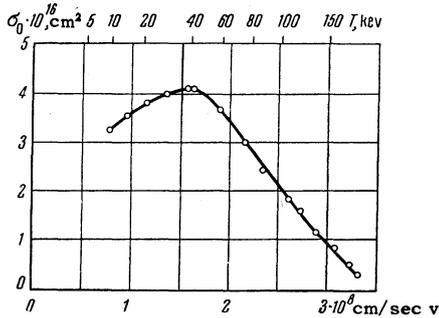


FIG. 3. Total cross section for capture of electrons by H_3^+ ions.

2. Production of Secondary H_2^+ Ions

Figure 4 shows the curves of $\sigma_{H_2^+}(v)$ which we have obtained by using primary H^+ , H_2^+ , and H_3^+ ions, and the corresponding curve for electron bombardment from reference 1. It follows from Eq. (11) that $\sigma_{H_2^+}$ is the sum of the cross sections for the usual charge exchange, σ_{01}^c , and for ionization with the emission of an electron, σ_{01}^i . A comparison of Fig. 4 with the curves in Figs. 1 to 3 shows that when the primary ion velocity is less than e^2/\hbar , the cross-section for ordinary charge exchange is the major component of σ_{H_2} . The $\sigma_{H_2^+}(v)$ and $\sigma_0(v)$ curves are therefore similar in the region where $v < e^2/\hbar$. Conversely, if $v > e^2/\hbar$, the chief contributor to the total cross section σ_{H_2} is the ionization cross section, since $\sigma_0 < 10^{-16} \text{ cm}^2$ for all primary hydrogen ions even when v is as low as $3.5 \times 10^8 \text{ cm/sec}$. From Fig. 4 it can also be seen that, in the region where $v > e^2/\hbar$, the more nuclei in the primary ions, the greater the cross section σ_{H_2} . By our assumption, the ionization processes take place mostly during collisions in which the electron shells of the ion and the molecule interpenetrate. In this case the magnitude of the ionization cross section must depend on the nuclear charge of the primary ion, and also on the number of nuclei if the primary ion is molecular. We have published analogous conclusions¹⁵ concerning the ionization of inert gases by atomic ions.

3. Production of Secondary Protons

Figure 5 shows the curves of $\sigma_{H^+}(v)$ which we have obtained for primary H^+ , H_2^+ , and H_3^+ ions, and the corresponding curve for the case of electron bombardment from reference 1. The cross section for the production of secondary protons is considerably smaller, over the whole energy region we investigated, than the cross section for the production of H_2^+ molecular ions. For pri-

mary H^+ and H_2^+ ions, the σ_{H^+} curves have a well defined peak that reaches σ_{H^+} approximately $5 \times 10^{-17} \text{ cm}^2$. The corresponding maximum cross section for electrons is only approximately 4×10^{-19} (reference 1).

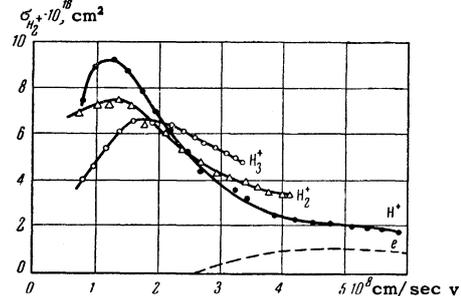


FIG. 4. Cross section for production of secondary H_2^+ ions. Corresponding primaries are marked on the curves. Dotted curve - data of reference 1.

Secondary protons can result from processes 2, 3, and 5 to 9 (cf. table). We believe that the secondary protons are due predominantly to dissociation of molecular hydrogen ions H_2^+ , which have been produced by charge exchange and by ionization with electron emission (processes 2, 3, 5, and 6). This assumption is supported by the results we have obtained by studying the kinetic energies of the secondary protons. In this study we have used a special analyzer apparatus (described in an earlier paper¹⁶) to determine, by means of a retarding electric field, the kinetic energies of secondary ions of various charges and masses, emitted at a specified angle relative to the direction of the primary beam. For primary hydrogen-ion energy of 75 kev, it was found that most secondary protons carried energies less than 7 ev, and that only an insignificant fraction had energies from 7 to 12 ev. This indicates that the cross sections of processes 7, 8, and 9 are small compared

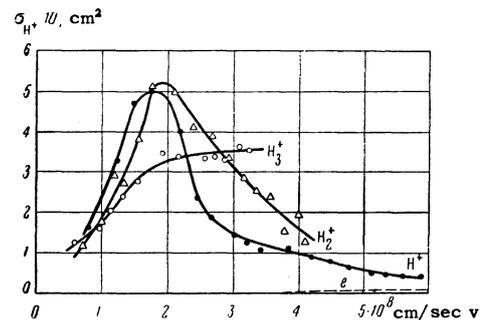


FIG. 5. Cross section for production of secondary protons. Corresponding primaries are marked on the curves. Dotted curve - data of reference 1.

with those of processes 2, 3, 5, and 6. It is interesting that the secondary-proton energy distribution is found to depend little on the angle of proton

travel. This is what can be expected if the energy is acquired by the proton not by direct transfer of momentum from the primary particle, but as a result of the dissociation of a molecular ion H_2^+ .

We believe that the maximum in the $\sigma_{H^+}(v)$ curve for H^+ and H_2^+ primaries is due to the dissociation of the molecular ions produced by charge exchange (processes 2 and 3). An argument in favor of this belief is the analogy between the $\sigma_{H^+}(v)$ curves in Fig. 5 and the curves for the total capture cross section in Figs. 1 and 2. It is easy to verify that the curves of $\sigma_{H^+}(v)$ for the primary ions H^+ and H_2^+ , like the curves of $\sigma_0(v)$, have maxima in nearly the same position and of approximately the same magnitude. It should be noted that the maxima of the $\sigma_{H^+}(v)$ curves for H^+ and H_2^+ primaries, like the maxima of the usual charge-exchange curves, are located in the region $v < e^2/\hbar$. When the primary ions are H_3^+ , the cross section σ_{H^+} for this velocity interval like the cross section σ_0 , is considerably smaller than that for the H^+ and H_2^+ ions.

In view of the fact that ΔE is larger for processes 2 and 3 than for usual charge exchange (process 1), the curve $\sigma_{01}^{cd}(v)$ can be expected to have a maximum at a higher velocity than the curve $\sigma_{01}^c(v)$. This assumption is confirmed, as can be seen from Figs. 1, 2, and 5.

With increasing energy of primary ions, as pointed out above, ionization with emission of one electron (process 4) begins to play the principal role in the production of secondary molecular ions H_2^+ . It is natural to assume that, analogously, the production of secondary protons should become more and more dependent on ionization processes 5 and 6, and then on processes 8 and 9, which involve a relatively large energy loss.

4. Production of Free Electrons

Our analysis of secondary ions by their e/m distribution gives grounds for assuming that when primary protons pass through molecular hydrogen,

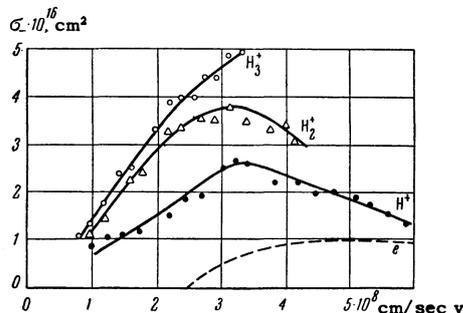


FIG. 6. Total cross section for production of free electrons. Corresponding primaries are marked on the curves. Dotted curve - data of reference 1.

the greater part of the free electrons is produced simultaneously with the H_2^+ ions. We therefore integrate the cross section σ_- in the same way as the ionization cross section σ_{01}^i . In the case of the (H_2^+, H_2) and (H_3^+, H_2) systems, the cross section σ_- contains also a contribution due to the electrons removed from the shells of the primary ions. Figure 6 shows the $\sigma_-(v)$ curves obtained for H^+ , H_2^+ , and H_3^+ ions, along with the corresponding curve for electron bombardment, taken from reference 1. The curves for H^+ and H_2^+ have a maximum somewhat above $v = e^2/\hbar$. It is easy to verify that the greater the number of protons in the given hydrogen ions, the greater the cross section σ_- (like $\sigma_{H_2^+}$). All the hydrogen ions have a large cross section σ_- even at velocities $v < e^2/\hbar$, where no ionization is observed in the case of electron bombardment.

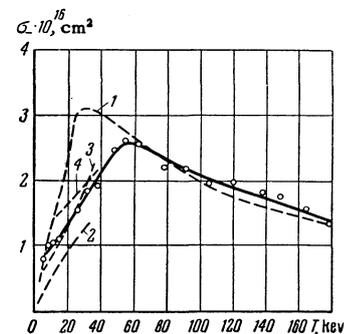


FIG. 7. Total cross section for production of free electrons by protons. \circ - data of this work. 1 - theoretical curve for ionization of H_2 by protons, calculated from data by Bates and Griffing.⁵ Experimental curves after the following: 2 - Keene,⁷ 3 - Fogel,⁹ 4 - Gilbody and Hasted.¹¹

The $\sigma_-(v)$ curve obtained for protons is compared in Fig. 7 with the analogous data from previous experimental investigations^{7,9,11} and with the theoretical curve of Bates and Griffing.⁵ These authors have calculated the cross section for the ionization of atomic and molecular hydrogen by protons, using the Born approximation. The maximum of the theoretical curve does not agree with ours. Data available from other investigations for the region in which the theoretical curve has a maximum agree better among themselves and with our data than with the theory. It must be noted, however, that the extension of the Born approximation to include the velocity region $v < e^2/\hbar$ does not lead to any discrepancy in the order of magnitude of the cross section. The theoretical curve agrees, within the limits of experimental error, with our data for the upper portion of the investigated interval.

In conclusion, the authors consider it their pleasant duty to express deep gratitude to Prof.

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288

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SOME PHOTOREACTIONS ON LIGHT NUCLEI

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The reactions $C^{12}(\gamma, 3\alpha)$, $O^{16}(\gamma, 4\alpha)$; $(\gamma, p\alpha)$ on C^{12} , N^{14} , and O^{16} ; and $C^{12}(\gamma, pt)2\alpha$ were investigated with photographic emulsions. The dependence of the γ -ray energy on the reaction cross section and the energy and angular characteristics of the disintegration products were obtained. The contribution of the reactions $C^{12}(\gamma, p\alpha)Li^7$ and $C^{12}(\gamma, pt)2\alpha$ to the total cross section for star production in the photon energy region from 30 to 80 Mev is estimated. Some possible mechanisms for the $(\gamma, p\alpha)$ processes are discussed.

THE interaction of photons with light nuclei, leading to the emission of three or more particles, has been little investigated in the region of γ -ray energy above 30 Mev. Yet observation of stars in photographic emulsions^{1,2} shows that similar "complex" reactions give, with increased γ -ray energy a substantial contribution to the total photon-absorption cross section. Investigation of such processes can, therefore, furnish information about the interaction of γ -rays with light nuclei, which becomes substantial at high energies.

In the present work we consider several types of photonuclear reactions in C^{12} , N^{14} and O^{16} . The method of photographic emulsions we used, making it possible to register charged disintegration products. The work was carried out with type Ia-2 NIKFI plates 500 μ thick, in which tracks of singly- and multiply-charged particles could be separated without difficulty. The experimental conditions were the same in the study of all reactions. Emulsions were irradiated by a bremsstrahlung beam from the synchrotron target at maximum energies