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DISSOCIATIVE EXCITATION OF CH_3Cl BY ELECTRON IMPACT

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Dissociative excitation of CH_3Cl molecules is studied by determination of absolute emission cross sections of excited H and Cl fragments. They are investigated as a function of the electron impact energy in the interval of 80—700 eV. The He benchmark procedure was adopted for absolute cross section values determination. The measured data are corrected with light collection efficiency factor for the loss of optical signal due to nonthermal energies of H excited fragments. The obtained cross section values are analysed applying the Bethe theory in order to get some information on dissociative states of CH_3Cl molecules. The power-low dependence for the Balmer series is determined.

1. Introduction

Dissociation processes of highly excited molecular states, due to electron impact in binary collision mode, have attracted considerable attention. Alkanes, alkenes and some of their fluoro, chloro and iodine derivatives have been specially investigated[1-4]. The aim of these investigations was to obtain information on the primary excitation and dissociative excitation processes of molecules. The purpose of these was to reach better understanding of electron molecular collisional phenomena, processes in astrophysics, mass spectrometry and specially in radiation chemistry and excimer laser physics.

Molecular excitation with electron beams of sufficiently high energy results in the formation of excited, so-called *superexcited* molecular states. They are unstable when defined as molecular states whose excitation energy E is higher than the first ionization potential of the relevant molecules. Since they are unstable, they degrade their energy undergoing different dissociation and ionization processes leading to the formation of neutral and excited atomic, ionic and molecular fragments[5-6].

This work is concerned with the investigation of optical radiation obtained in electron — CH_3Cl excitation processes. Our aim is to measure the emitted radiation in 320 nm — 900 nm spectral range due to binary collisions with electrons in medium energy range of 80 eV — 700 eV. These measurements are to show which excited molecular fragments are formed, and for some of them to determine the light emission quantitatively.

Some studies have been done on the dissociative excitation of CH_3Cl molecules and some other chlorinated methanes by electron impact. K. Hirota et al.[3] and M. Toyoda et al.[7] have investigated dissociative excitation processes in 280 nm — 450 nm spectral range with low energy electrons. They have concentrated their attention only to the excited fragment identification and the proof that the obtained HCl^+ excited fragments are produced in secondary processes. V. T. Koppe et al.[8] and N. P. Danilevskii et al.[9] have also investigated the CH_3Cl molecule but in high electron energy interval (0.4 — 6 KeV) measuring the line emission cross sections for some Cl^+ excited fragments in 380 nm — 600 nm wavelength range.

However, our main purpose is the systematic investigation of CH_3Cl atomic fragments emission for electron energies where the Bethe[10] approximation could be valid. The results are presented as absolute line emission cross sections for atomic fragments, i. e. excited H and Cl fragments. Special attention was paid to the analysis of the influence of the escaping possibility of the so-called *hot* H excited fragments on the accuracy of the obtained cross section data.

The comparison with the Bethe theory has been performed using Fano plot representation[10] from which some indication about the primary electron — molecular excitation processes could be obtained. The power-low dependence of CH_3Cl dissociation to excited H fragments on the principal quantum number n is likewise determined.

For the absolute line emission cross section determination we have adopted the recently proposed He benchmark normalization procedure[11] using n^1S — -2^1P transitions as benchmark cross sections.

2. *Experimental method and results*

2.1. *Apparatus*

Detailed description of the used apparatus has been presented by Kurepa and Tasić[12] and here it will be discussed only briefly. It consists of two differentially pumped vacuum chambers, i. e. the electron gun chamber and the interaction chamber, pumped to background pressure of 10^{-4} Pa. They are mutually separated by a real aperture used for electron beam transport from one chamber to

the other. The stray magnetic fields and the earth magnetic field in both chambers have been eliminated by a pair of Helmholz coils to a value smaller than 3 mG. The electron beam transport through the interaction chamber has been achieved using a four-element cylindrical lens-system[13] where the fourth cylindrical electrode has been lengthened to serve as a real interaction chamber of the apparatus. As electron beam collector, a specially designed Faraday cage has been used[14] with back scattered electron beam supression. The main advantage of the used electrode system is that it provides the electron beam image position in the interaction chamber to be constant. However, the electron beam diameter and the electron beam intersity are also constant due to permanent electrode lens magnification[13].

The optical emission has been obtained from the interaction chamber and was observed at 90° with respect to the electron beam. It was detected through a quartz window, a set of optical lenses, and optical monochromator (Carl Zeiss, SPM-2) by RCA 31034 C, cooled photomultiplier. The ORTEC single pulse counting system was used for photomultiplier signal measurements.

The working gas pressure was measured by a capacitance manometer (MKS, Baratron, Mod. 77 H — 1), and electron beam current by galvanometer (Kipp and Zonen, Microva A 14). The gas used in the experiment was of 99.99% purity (Baker Chemicalien).

The relative wavelength sensitivity for the used detection system has been obtained by the use of OSRAM tungsten strip lamp (Mod Wi 17/G) for two regions of interest: 320 nm — 400 nm and 400 nm — 900 nm, thus utilizing two different difraction gratings.

2.2. *Experimental procedure and results*

To investigate the electron excitation processes of CH_3Cl molecules in binary collision mode, we have used in the interaction region the working gas pressure of 10^{-2} Pa and the electron beam current of 20 — 100 μA. To measure the obtained light emission in the range 320 mn — 400 nm the used optical grating in the monohcromator had reciprocial dispersion of 4 nm/mm, whereas this parameter of the used grating in the 400 nm — 900 nm range has the value 2 nm/mm. We have detected the optical emission in UV, visible and near the IR region that are presented in Fig. 1. The obtained spectra were identified. They originate primarily from the excited H fragments, from CH, Cl, CH^+ and Cl^+ fragments. Some traces of HCl^+ excited spacies from the spectra in the UV region were detected as well.

The most dominant emission originated from the excited H fragments, as Balmer lines emission ($n = 3, 4, 5, 6, 7, 9 \rightarrow n = 2$). CH (0,0) vibrational band emission from the $A^2\Delta \rightarrow X^2\Pi$ transition and CH (0,0) and (1,0) vibrational bands from the $B^2\Sigma \rightarrow X^2\Pi$ transitions have been identified. CH^+ (0,0) vibrational band from $^3\Sigma - {}^3\Pi$ transition and some lines which could originate from the excited HCl^+ fragments from the $A^2\Sigma \rightarrow X^2\Pi$ transition are also detected. Cl fragments emission in the near IR region was obtained where among[13] identified spectral lines five were well resolved and suitable for quantitative measurements. They all originate from the $4s^4P - 4p^4S^0$ and $4s^4P - 4p^4D$ transitions.

Our main concern was to investigate the fragments light emission primarily well resolved and of interest for quantitative measurements using the He benchmark procedure[11]. The emission from H and Cl fragments has fulfilled these criteria. So, the Balmer lines emission and five lines from Cl fragments originating from the $4s^4P$ level have been measured and analysed. The obtained results are presented as the absolute line emission cross sections, $Q_{em,x}(E)$, for the relevant spectral lines $x(\lambda)$ in the energy interval of 80—700 eV.

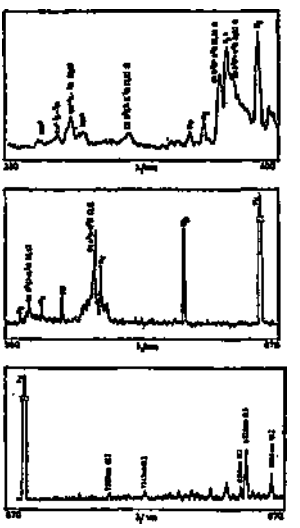

Fig. 1. Emission spectra of CH_3Cl in the 330 nm — 870 nm region: electron energy 100 eV, electron-beam current 50μA, $p = 4 \cdot 10^{-2}$ Pa a) in 330—400 nm region, b) in 380—670 nm region, c) in 670—870 nm region.

The first application of the He benchmark procedure based on the preliminary determined He absolute cross sections for the $n^1S - 2^1P$ transitions has been demonstrated by Kurepa and Tasić[12]. It was applied for the measurement of the Balmer β emission originating from some dissociated hydrocarbons. In Ref. 12 the proposal for the use of this method and some analysis of its advantages are discussed.

Briefly, in this method instead of using the well known equation for $Q_{em,x}(E)$ determination from experimental parameters[1,2,4], the following equation should be used for the basicaly same purposes:

$$Q_{em,x}(E_n) = Q_{em,\mathrm{He}}(E_n) \frac{|S(\omega, E)_{E_n}(e/I)|_x}{|S(\omega, E)_{E_n}(e/I)|_{\mathrm{He}}} \cdot \frac{P(E_n)_x \cdot K(\lambda')_{\mathrm{He}}}{P(E_n)_{\mathrm{He}} \cdot K(\lambda)_x}. \qquad (1)$$

The above relation is used for the nominalization purposes of the measured optical excitation functions. Namely, this method includes two stages: the measurements of the optical excitation function in a desired electron energy range for the spectral line of interest at $x(\lambda)$, i. e. $S(\omega, E)/(I/e)$ values, and the measurement of the same quantity at normalization energy E_n (eV) for the chosen He benchmark. This is all done at the equal gas concentration in both gases, CH_3Cl and He.

The quantitaties $|S(\omega, E)_{E_n}|_x$ and $|S(\omega, E)_{E_n}|_{He}$ are the light signals (obtained under the same conditions for CH_3Cl fragment emission and He benchmark transitions, respectively), whereas I/e is the number of incident electrons per second in the interaction chamber measured during the relevant light signal determination. $P(E_n)_x$ and $P(E_n)_{He}$ are the correction factors for anisotropic angular distribution of radiation due to polarization, and $K(\lambda)_x$, $K(\lambda')_{He}$, are the absolute sensitivities of the used detection system at λ_{nm} (wavelength of the measured fragment emission) and λ'_{nm} (wavelength for the utilized He benchmark). Actually, $P(E_n)_x$ and $P(E_n)_{He}$ in the whole investigated energy range, for the emissions both from CH_3Cl fragments and He benchmarks, should be set to 1 in Eq. (1) according to Refs. 11, 15, 16, 17. The ratio $K(\lambda')_{He}/K(\lambda)_x$ has been determined from the relative sensitivity curves (Fig. 2) for our detection system at the wavelengths λ_{nm} and λ'_{nm}.

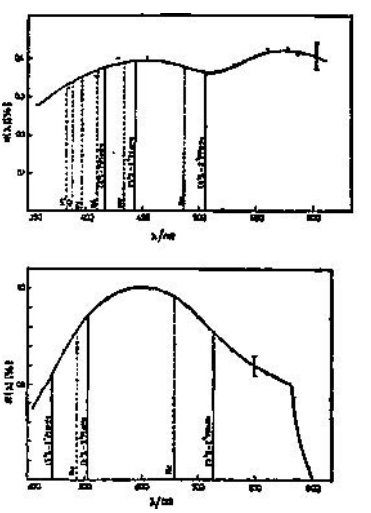

Fig. 2. Relative quantum sensitivity curves $K(\lambda)$ (%) for the detection system used.

The determined absolute line emission cross sections data are given on Table 1 for H_α, H_β, H_γ, H_δ, and H_ε lines. H_ξ line could not be measured being blended by CH fragments emission. The absolute line emission cross sections for the Cl fragments at 725.7 nm, 754.7 nm. 833.3 nm, 837.6 nm and 858.6 nm are also tabulated (Table 2). The first two lines of Cl fragments are due to the $4s^4\,P - 4p^4\,S^0$ (J values being 5/2 — 3/2 and 3/2 — 3/2) transitions, and the last three to the $4s^4\,P - 4p^4\,D$ (J values being 3/2 — 3/2, 5/2 — 7/2, 3/2 — 5/2) transitions[18]. The measured excitation functions for all spectral lines of interest have been obtained with a reproducibility of $\pm$ 3%. The chosen He benchmarks were spectral lines from: $3^1\,S - 2^1\,P$, $4^1\,S - 2^1\,P$ and $5^1\,S - 2^1\,P$ transitions which at normalization energy $E_n = 100\,\text{eV} - Q_{em,He}$ have the values: $24.9 \pm 2.8 \cdot 10^{-24}$ m^2, $5.61 \pm 0.25 \cdot 10^{-24}$ m^2 and $2.30 \pm 0.12 \cdot 10^{-24}$ m^2, respectively. These values have been obtained[11] with high confidence level of $\pm$ 7%. Measuring in our experiment the gas concentrations by capacitance manometer with a pre-

TABLE 1

E/eV	H_α	H_β	H_γ	H_δ	H_ε
80	5.2	2.6	1.6	0.86	0.65
90	12	4.8	2.7	1.4	0.87
100	17	5.9	3.0	1.6	1.0
120	18	6.8	3.3	1.9	1.2
140	18	6.7	3.2	1.9	1.2
160	17	6.1	3.0	1.8	1.1
200	15	5.2	2.5	1.6	0.97
250	12	4.3	2.0	1.3	0.82
300	10	3.5	1.7	1.1	0.70
400	7.6	2.5	1.3	0.94	0.57
500	5.7	2.0	1.1	0,79	0.49
600	4.6	1.6	0.91	0.69	0.42
700	4.2	1.4	0.81	0.61	0.35

Balmer line cross sections data for CH_3Cl molecule (units of 10^{-23} m^2).

TABLE 2

	$4s^4P \rightarrow 4p^4S^0$	$4s^4P \rightarrow {}^4p\,{}^4S^0$	$4s^4P \rightarrow 4p^4D^0$	$4s^4P \rightarrow 4p^4D^0$	$4s^4P \rightarrow 4p^4D^0$
E/eV	725.7 nm	754.7 nm	833.3 nm	837.8 nm	858.6 nm
80	3.7	3.1	6.0	41	20
90	3.9	3.4	6.9	43	22
100	4.1	3.8	7.4	44	23
120	4.1	3.7	7.2	42	22
140	3.9	3.6	6.9	39	21
160	3.4	3.2	6.5	37	20
200	2.8	2.7	5.1	31	16
250	2.4	2.3	4.2	25	13
300	2.2	1.9	3.2	19	11
400	1.7	1.4	2.6	15	7.5
500	1.5	1.2	2.1	12	6.2
600	1.4	1.1	1.7	9.7	5.0
700	1.2	1.0	1.5	8.9	4.8

Absolute emission cross sections data for Cl ($4s^4P - 4p^4S^0$) and Cl ($4s^4P - 4p^4D^0$) transitions (units of 10^{-24} m^2).

cision of $\pm$ 1%, the electron current with galvanometer, having precision better than $\pm$ 1%, the obtained absolute cross section data have the expected accuracy of $\pm$ 15%. This was achieved by the use of the He benchmark procedure which allows the elimination of some possible errors as: errors due to unprecisely determined geometrical parameters in the experiment, errors in absolute gas concentration determinations, as well as the errors due to the absolute $K(\lambda)$ measurements.

Only the last two sources of errors could contribute to the total uncertainty of the measured $Q_{em,x}(E)$ data up to the order of $\pm 15\%$[11,12,23,24].

In Figs. 3 and 4 the obtained absolute line emission cross sections data given on Tables 1 and 2 are presented.

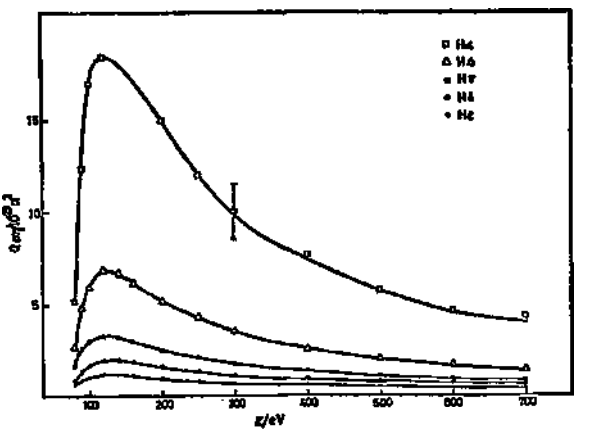

Fig. 3. Balmer line cross sections for CH_3Cl.

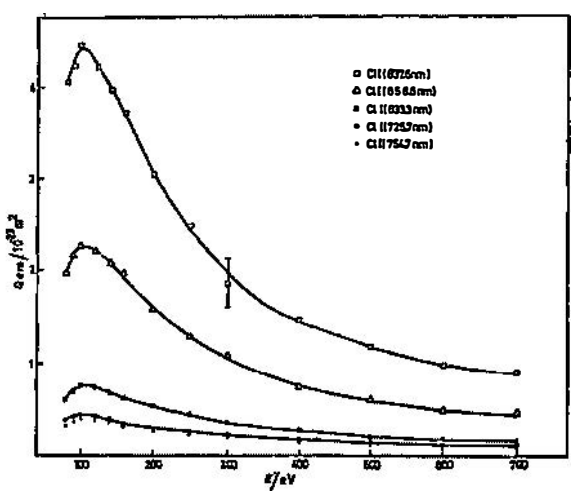

Fig. 4. Some ClI absolute cross sections for CH_3Cl

3. *Discussion*

In all experiments dealing with the electron-molecular dissociative excitation processes, when the quantitative light emission measurements are required, it is necessary that all the investigated excited dissociated fragments deexcite in

the detection observation region of the used experimental set-up[11,16,18]. The reason for this established by the transitional spectroscopy experiments[17,19-21], is the fact that from these dissociative processes the excited H fragments, for example, have nonthermal kinetic energies i. e. energies ranging from 2 eV to 10 eV. depending on the electron energy range and on the type of the molecules. Independent of the normalization procedure used in different experiments, this problem appearing during quantitative light emission determination, must be taken into account and possible signal loss of *hot* (nonthermal) H fragments must be somehow compensated. Analysing this problem in general and for our experimental set-up[18], we got Eq. (2) for the light collection efficiency factor F, for such nonthermal H excited fragments:

$$F = 1 - \frac{\operatorname{erf}\left(\frac{a}{2\bar{V}\cdot\tau} + \frac{w}{a}\right) - \operatorname{erf}\left(\frac{a}{2\bar{V}\cdot\tau} - \frac{w}{a}\right)}{2\left|\operatorname{erf}\left(\frac{a}{2\bar{V}\cdot\tau} + \frac{w}{a}\right)\cdot\exp\left(\frac{w}{\bar{V}\cdot\tau}\right) - \operatorname{erf}\left(\frac{a}{2\bar{V}\cdot\tau}\right)\right|} \cdot e^{-\frac{\Delta}{\bar{V}\cdot\tau}} \cdot \qquad (2)$$

The quantity F is defined as the ratio of the number of detected excited fragments to the total number of produced excited fragments.

In our experiment where the electron beam density is of the Gaussian shape, $2w$ is the electron beam diametar in the interaction chamber, 2Δ is the width of the observation region defined by the optical monochromator slit height, while a is the numerical factor determined from the electron beam distribution. The quantity $\bar{V}\cdot\tau$, so-called decay length, incorporates $\bar{V}$, the average kinetic energy of the excited H fragments (expressed in mm/s), and τ the mean lifetime of the upper level of the relevant spectral line.

CH_3Cl molecules have not been investigated in the translational spectroscopy experiments i. e. $\bar{V}$ for their H excited fragments are not measured. However, regarding the results for $\bar{V}$ for CH_4 molecules and other hydrocarbons[17,20] and the results from $CHCl_3$ molecules[21], it is possible to calculate F values for the Balmer emission in our experiment from CH_3Cl.

According to Refs. 17 and 20, $\bar{V}$ for H excited fragments is 4 eV, but in Ref. 21 it is claimed that above the electron energies of 100 eV these H excited fragments to the states of $n = 3$, $n = 4$, $n = 5$, $n = 6$ $\bar{V}$ are 3.9 ± 0.6 eV. Adopting the $\bar{V} = 4$ eV, we have calculated the F values fot the Balmer lines originating from dissociated CH_3Cl molecules. On Table 3 the obtained F values are given. On Table 3 also the corrected values for the Balmer lines cross sections are presented. Absolute line emission cross sections for Cl fragments have not been corrected for the same effect regarding the mass difference of H and Cl fragments, this being expected and supported by some experimental evidence for heavy fragments[25].

The absolute line emission cross sections data for the H and Cl fragments have been analysed according to the Bethe theory revised by Inokuty[10].

According to Ref. 10 the cross sections for optically allowed excitation processes by electron impact at sufficiently high energies, which lead to the formation of a particular excited fragments can be expressed as:

$$Q_{em,x} = (4a_0^2\, R/E)\, M_{em}^2 \ln (C_{em}\, E/R), \tag{3}$$

TABLE 3

E/eV	H_α	H_β	H_γ	H_δ	H_ε
80	5.2	2.6	1.6	0.94	0.92
90	12	4.8	2.7	1.5	1.2
100	17	5.9	3.0	1.8	1.4
120	18	6.8	3.3	2.1	1.7
140	18	6.7	3.2	2.1	1.7
160	17	6.1	3.0	2.0	1.6
200	15	5.2	2.5	1.8	1.4
250	12	4.3	2.0	1.4	1.2
300	10	3.5	1.7	1.2	1.0
400	7.6	2.5	1.3	1.0	0.82
500	5.7	2.0	1.1	0.86	0,70
600	4.6	1.6	0.91	0.76	0.60
700	4.2	1.4	0.81	0.67	0.50
F	1.0	1.00	0.995	0.911	0.699

Corrected Balmer line cross sections data for CH_3Cl molecule (units of 10^{-23} m^2).

a_0 being the first Bohr radius, R the Rydberg energy, E the incident electron energy, while C_{em} is a constant dependent on the properties of the primary excited molecular state. The quantity M_{em}^2 is related to the sum of the optical oscillator strengths for all dipole transitions to primary excited molecular states responsible for excited fragments formation.

Plotting $Q_{em,x}\, E/4\, a_0^2\, R$ versus $\ln E$ — the so-called Fano plots[10], the values for M_{em}^2 and C_{em} are determined for all Balmer lines as well as for Cl lines. It is done by the least square fitting procedure for our corrected (H cross sections) and uncorrected (Cl cross sections) data. M_{em}^2 — values are presented in Table 4, being specific for two energy intervals.

In Figs. 5 and 6 the Fano plots for the measured Balmer lines and Cl lines absolute cross sections are given. From the analysis of the obtained graphs (Figs. 5 and 6) and relevant M_{em} and C_{em} values, and from the predictions of the used theoretical model[10,1,2,4], it could be suggested that the primary excited states of CH_3Cl molecules, in energy range of 80—160 eV, are formed by optically allowed electron transitions. Above 200 eV they are formed by preferably forbidden transitions, probably via two electron transitions to high Rydberg or *superexcited* molecules states.

TABLE 4

a) Balmer lines

E(eV)	H_α	H_β	H_γ	H_δ	H_ε
80—140	78	27	13	8.2	6.2
160—700;	−2.3	−0.86	1.2	−1.0	0.51

b) CII lines

E(eV)	725.7 nm	754.7 nm	833.3 nm	837.7 nm	858.6 nm
80—140	0.93	0.94	1.8	8.8	5.0
160—700;	0.46	0.28	0.014	0.081	−0.030

M^2_{em} values for Balmer series and CII lines × 10^3.

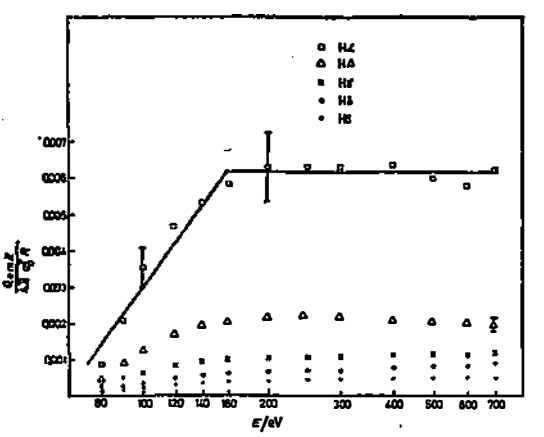

Fig. 5. Balmer line cross sections for CH_3Cl plotted as $Q_{em} \cdot E/4\pi a_0^2 R$ versus ln E.

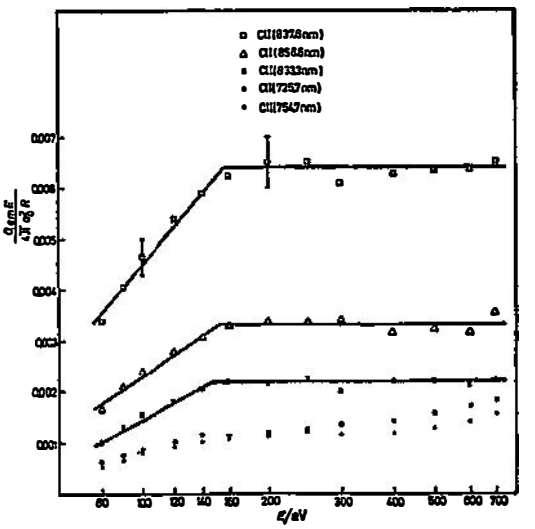

Fig. 6. Some CII line cross sections for CH_3Cl plotted as $Q_{em} \cdot E/4\pi a_0^2 R$ versus ln E.

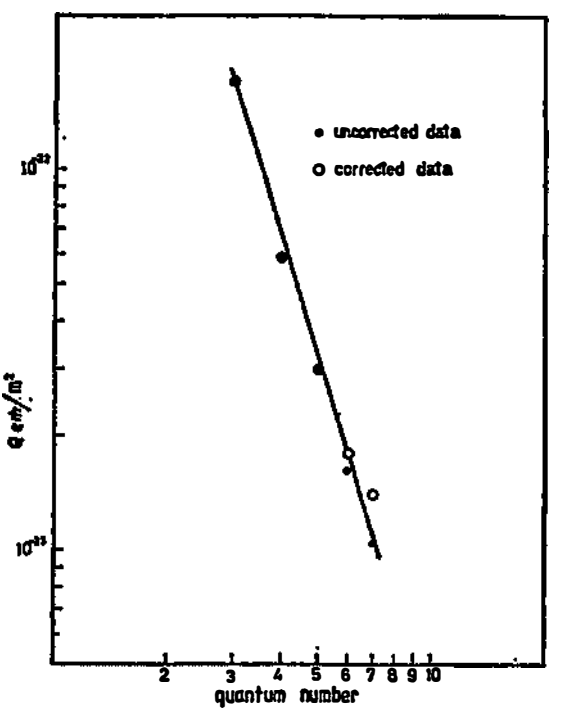

Fig. 7. Double logarithmic plot of Q_{em} $(n \to 2)$ versus n for CH_3Cl in the case of 100 eV electrons.

Obtaining the absolute line emission cross sections data for H excited fragments i. e. for Balmer series, it was interesting to verify or reject Ochkur's and Petrunken's prediction based on classical theory and Born approximation[22]. Namely, according to Ref. 22 excitation cross sections of one series, if n (the principal quantum numbers of upper atomic excited states) are large enough, the power-low dependence of $Q_{em,x}$ versus n, could be expressed as.

$$Q_{em} = a \cdot n^{-b}. \tag{4}$$

In this equation, if it is applied to the molecules, a represents the characteristics of the molecule, and b is the power-low dependence. It is predicted, for n large enough, to be $b = 3$.

We have performed this analysis for $Q_{em,x}$ values of H excited atoms originated from CH_3Cl molecules for electron energy of $E = 100$ eV. Using the least square fitting procedure for obtained cross sections data presented as $\log Q_{em,x}$ versus $\log n$ (Fig. 7) we obtained one value for this parameter, b_1 value. However, we have adopted as the second method, the Khayarallah's proposal[16] for the power-low dependence, obtaining another value, b_2 value, for the same parameter. The two values for the quantity b are: $b_1 = 3.0 \pm 0.4$ and $b_2 = 2.5 \pm 0.4$, from these two independent methods, respectively.

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DISOCIJATIVNA EKSCITACIJA CH_3Cl MOLEKULA ELEKTRONSKIM UDAROM

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Ispitivana je disocijativna ekscitacija CH_3Cl molecula u interakciji sa elektronima merenjem apsolutnih emisionih preseka ekscitovanih H i Cl fragmenata, a u energetskom domenu 80—700 eV. Za dobijanje vrednosti apsolutnih preseka korišćen je metod He standarda. Mereni podaci su korigovani na gubitak netermalnih ekscitovanih H fragmenata. Dobijeni rezultati su analizirani primenom Bethe-ove teorije sa ciljem dobijanja informacija o disocijativnim stanjima CH_3Cl molekula. Određena je i funkcionalna zavisnost apsolutnih emisionih preseka Balmer-ove serije od glavnog kvantnog broja n, gornjeg nivoa ove serije.