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ELECTRON-HYDROGENSULPHIDE TOTAL IONIZATION AND ELEC-TRON ATTACHMENT CROSS SECTIONS*

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Cross sections for absolute total ionization between 10 eV and 100 eV, and dissociative attachment between 0 eV and 14 eV of the hydrogensulphide molecule have been measured using a parallel plate interaction chamber. The maximum of the ionization curve was found to be 6.28×10^{-20} m² at an electron energy of 65 eV. The total attachment curve shows four maxima at energies of 2.1 eV (HS⁻), 5.3 eV (H⁻), 7.6 eV (H⁻), and 9.5 eV (S⁻), and a shoulder at energies higher than 12 eV. Using available relative mass-spectrometric measurements the values of partial attachment cross sections for various processes are proposed.

1. Introduction

In this paper we present an electron beam study of ionization and dissociative attachment cross sections in hydrogensulphide molecule. Preliminary results have been reported by Belić and Kurepa^{1, 2)}.

The interest in the hydrogensulphide molecule has various sources. It is one of the specific pollutants to be found around industrial plants, and it was discovered in astronomical objects. One the other hand it has a structure similar to that

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of the water molecule so that a behaviour of the same kind could be expected, specially in electron attachment processes.

The first ionization threshold of the H_2S molecule was determined by Rydberg series analysis³, photoionization without⁴) and with mass analysis⁵, and by electron impact⁶⁻⁸, the agreement being satisfactory.

Relative abundances of ions formed by electron collision were determined by Dibeler and Rosenstock⁸, and Frost and McDowell⁷. Ionization cross sections for single low electron energies were measured by Lampe et al.⁹, and Otvos and Stevenson¹⁰ and at high energies by Fiquet-Fayard et al.¹¹. First differential ionization efficiency curves were investigated by Morrison and Traeger¹².

Recently the H_2S molecule has been investigated using a low energy electron spectrometer¹³ and by a threshold electron spectrometer¹⁴, as well as by a coincidence electron spectrometer¹⁵. The photoelectron spectrometer technique was used by Al-Joboury and Turner¹⁶, Al-Joboury¹⁷ and Baker et al.¹⁸.

Dissociative electron attachment processes were investigated, mainly with mass spectrometers, by Kraus¹⁹, Jäger und Henglein²⁰, Fiquet-Fayard et al.²¹, Azria et al.²², Tronc et al.²³, and by a combined electron spectrometer — mass spectrometer technique by Azria et al.²⁴ and Azria and Aboauf²⁵.

2. Experimental procedure

Full details of the apparatus used in this experiment are given elsewhere²⁶, so only a brief description will be presented here. For the absolute cross section determination the equation.

$$\sigma = \frac{I_i}{I_e} \frac{1}{nL}$$

was used, where I_i is the total positive or negative ion current formed by the passage of the electron beam of current I_e along a collector of length L, inside the target-gas atmosphere of particle density n (=p/kT).

The interaction chamber in our experiment was a paralel plate condenser system introduced by Smith²⁷⁾, and Tate and Smith²⁸⁾, and used thereafter by many authors. The electron beam from an electron gun was energy selected by a trochoidal electron monochromator²⁸⁾. In the present measurements the electron beam half-width was 120 meV and its intensity in the range of 0.1 μ A.

The pressure of the molecular gaseous target in the range 10^{-3} to 10^{-1} Pa was measured by two different methods. In most of the experiments a gas-flow method^{30,31}, directly coupled to the vacuum chamber containing a parallel plate condenser was used.

Final results were obtained using a capacitance manometer (Baratron type) for target gas pressure measurements with relative error of ± 0.02 . Through the measurements the gas was introduced through a leak valve from the gas handling system, and it flowed towards the pumping system so that an equilibrium pressure was maintained in the interaction chamber region.

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3. Experimental results

With the parallel-plate type interaction chamber it is not possible to distinguish and measure separately the various ionic species formed in electron-molecule collisions. The measurements were done by separately collecting all the positive or negative ions formed along the electron beam path.

The collection of positive ions gives absolute values for the total ionization cross section. Positive ions are formed in a great number of processes, the most important being molecular ionization

$$H_2S + e \rightarrow H_2S^+ + e + e$$

various dissociative ionizations

$$H_{2}S + e \rightarrow \begin{cases} HS + H^{+} + e + e \\ S + H_{2}^{+} + e + e \\ H_{2} + S^{+} + e + e \\ H + HS^{+} + e + e \end{cases}$$

and ion pair formation processes

$$H_{2}S \rightarrow \begin{cases} HS^{+} + H^{-} + e^{\frac{1}{2}} \\ HS^{-} + H^{+} + e^{\frac{1}{2}} \\ S^{-} + H^{+}_{2} + e^{\frac{1}{2}} \\ S^{+} + H + H^{-} + e^{\frac{1}{2}} \\ S^{-} + H^{+} + H^{+} + e^{\frac{1}{2}} \end{cases}$$

All other processes with molecular ion, radicals, atoms or ions in excited states are possible as well and contribute to the total positive ion signal.

Negative ions can be formed in processes of dissociative attachment

$$H_{2}S \rightarrow \begin{cases} HS^{-} + H \\ S^{-} + H_{2} \\ S^{-} + H + H \\ H^{-} + HS \\ H^{-} + H + S, \end{cases}$$

and ion pair formation. Thus the collection of negative ions gives the cross section for total negative ion formation.

3.1. Total ionization

Total ionization cross sections were measured in the energy interval between 10 and 100 eV. The energy scale was calibrated within 0.05 eV by linear extrapo-

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lation of the cross section curve to the 10.46 ± 0.01 eV value for the ionization of hydrogensulphide obtained by photoionization^{4,5)}. Numerical values of total ionization cross sections are listed in Table 1.

E/ m eV	Cross section	E/eV	Cross section
10.0	0.000	27.5	4.74
10.5	0.021	28.0	4.83
11.0	0.104	28.5	4.88
11.5	0.299	29.0	4.95
12.0	0.376	29.5	5.0)
12.5	0.502	30	5.06
13.0	0.669	31	5.16
13.5	0.794	32	5.25
14.0	0.878	33	5.31
14.5	1.08	34	5.39
15.0	1.21	35	5.47
15.5	1.37	36	5.52
16.0	1.58	37	5.58
16.5	1.79	38	5.62
17.0	1.96	39	5.68
17.5	2.13	40	5.71
18.0	2.29	42	5.81
18.5	2.48	44	5.89
19.0	2.63	46	5.95
19.5	2.84	48	6.06
20.0	3.01	50	6.08
20.5	3.17	52	6.12
21.0	3.30	54	6.14
21.5	3.47	56	6.17
22.0	3.73	58	6.20
22.5	3.72	60	6.22
23.0	3.84	62	6.25
23.5	3.96	65	6.28
24.0	4.14	70	6.22
24.5	4.26	75	6.20
25.0	4.35	80	6.18
25.5	4.45	85	6.14
26.0	4.52	90	6.08
26.5	4.64	95	6.02
27.0	4.68	100	5.98

TABLE 1.

Total absolute cross sections for ionization of hydrogensulphide (In units of 10^{-20} m²).

To the authors' knowledge there are no other measurements of the total ionization cross section in this energy range with which our data could be compared. There are only two different values of the cross section for the incident electron energy of 75 eV. Both Otvos and Stevenson¹⁰⁾ and Lampe et al.⁹⁾ used a mass spectrometer to analyze the ionization products of the H₂S molecule, and added the relative intensities of various ionic species to obtain the total positive ion in-

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tensities. These have been then normalized to the argon atom total ionization cross section of Tate and Smith²⁸⁾ at 75 eV of 3.52×10^{-20} m². There are more recent and more reliable measurements of total ionization cross sections in argon. Rapp and Englander-Golden³²⁾ obtained at 75 eV energy electrons a value of 2.82×10^{-20} m², while Kurepa et al.²⁶⁾ got the value of 2.90×10^{-20} m². They are much lower than the value of Tate and Smith²⁸⁾, so that a renormalization of data by Otvos and Stevenson¹⁰⁾ and Lampe et al.⁹⁾ for H₂S had to be done and the value of Rapp and Englander-Golden was used for that. The result of this renormalization is presented in Table 2, together with the result of our total ionization cross section measurement at 75 eV.

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Authors	$\begin{array}{c} \text{Cross} \\ \text{section} \\ \times 10^{-20} \\ \text{m}^2 \end{array}$	Remarks
Otvos and Stevenson ¹⁰⁾	3.39	$I^{+}(H_{2}S)/I^{+}(Ar) = 1.2018$
Lampe et al. ⁹⁾	5.14	$I^{+}(H_2S)/I^{+}(Ar) = 1.8239$
Present	6.20	Absolute measurement

Total ionization cross sections of hydrogensulphide at 75 eV incident electron energy.

The differences are great and well outside the experimental error of our experiment. Mass-spectrometric experiment for the measurement of relative intensities of various ionic species in the whole electron energy range are needed in order to determine the partial ionization cross sections.

3.2. Dissociative attachment

The dissociative attachment processes have been investigated in the incident electron energy range from zero to 14 eV, and the experimental cross section curve is presented in Fig. 1. The energy scale was calibrated within 0.05 eV to the appearance potential of positive ions at 10.46 eV.

On the cross section curve four maxima can be distinguished, the first two well separated, and an additional shoulder of low intensity at the electron energies higher than 12 eV. An overwiev of known dissociative electron attachment processes, dissociation products, appearance potentials, peak positions and cross section values is given in Table 3.

The largest in cross section value and lowest in energy is the peak at around 2.2 eV. Fiquet-Fayard et al.²¹⁾ were the first to derive cross section values. In their experiment the ratio of negative ion current intensity in the peak at 2.2. eV to the total positive ion current intensity at 75 eV electron energy was determined to be 3.45×10^{-3} . In our experiment the total ionization and total attachment cross sections were determined by the measurement and absolute calibration of all re-

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lated parameters. The ratio of the 2.2 eV peak cross section and 75 eV total ionization cross section is $(3.5 \pm 0.21) \times 10^{-3}$, this being in very good agreement with data of Fiquet-Fayard et al.²¹⁾. The difference in attachment cross sections, as seen



Fig. 1. Dissociative electron attachment cross section of the H_2S molecule: \bigcirc and heavy full curve — present experimental results for total cross section; light full curves: GA — gaussian approximation of the present results, partial attachment cross sections obtained by normalizing relative measurements of Jäger und Henglein²⁰ — JH to the 2.2 eV maximum, and those of Kraus¹⁹ — K to the 9.6 eV maximum, respectively.

in Table 2, comes presumably from the normalization procedure applied by Fiquet-Fayard et al. They used the ratio of relative total ionization cross sections at 75 eV electron energy of hydrogensulphide and argon, normalized to the argon atom total ionization cross section at 75 eV of Rapp and Englander-Golden³²⁾. Lampe et al.⁹⁾ determined this ratio to be equal to 1.82 (= 6.42/3.52), while Otvos and Stevenson¹⁰⁾ gave quite a different value of 1.20. The former was used for this normalization procedure with the assumption that this particular measurement is free of discrimintation effects in the mass spectrometer.

As stated by Azria et al.²⁾ the 2.2 eV peak is dominated by HS⁻ ions with a small contribution of S⁻ ions, but the value of the ratio of negative ion current intensities was not cited. Jäger and Henglein²⁰⁾ have also investigated the mass spectrum of negative ions in this energy range, giving to the HS⁻ peak at 2.27 eV a relative intensity of 234, while for the S⁻ peak at 2.67 eV the relative intensity was found to be equal to 3. Since the S⁻ intensity is lower than the HS⁻ one by two orders of magnitude, it does not influence appreciably the HS⁻ peak shape.

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The second well resolved peak is at 5.3 eV. The investigation of Tronc et al.²³⁾ shows that it is due mainly to the H⁻ ions. The cross section values in the peak of our experiment and that of Tronc et al. are in fairly good agreement. A small contribution to this peak, at the higher electron energy wing, comes from the S⁻ ions. Jäger and Henglein²⁰⁾ gave a relative intensity of 4 for the S⁻ peak at 5.9 eV as compared with the relative intensity of 234 for the 2.27 eV peak of HS⁻. Since 2.1 eV and 5.3 eV attachment cross section peaks are of the same order of magnitude, the contribution of the S⁻ ion intensity to the 5.3 eV peak shape is small. The other source of information, the measurements of Kraus¹⁹⁾, gives a ratio of 0.122 of the S⁻ intensity in the peak at 6.25 eV to the 9.6 eV peak of S⁻, leading to slightly larger value of the cross section than those from normalization of data by Jäger and Henglein²⁰⁾, but of the same order of magnitude.

The position and cross section values of the process peaking at 7.5 eV were determined from experimentally measured total attachment cross section curve by extrapolating the 5.3 eV and 9.5 eV maxima by gaussian distribution and substracting the so calculated values from the experimental ones. The cross section in the peak of 5.5×10^{-23} m² is in good agreement with the value of Tronc et al.²³⁾. According to Tronc et al. this maximum is due to H⁻ ions only.

The fourth peak on the total attachment cross section curve, at an energy of 9.5 eV, has a cross section higher that the value given by Tronc et al.²³ for 50%, which is much outside the experimental error in both our and their measurements. According to Kraus¹⁹ and Tronc et al.²³ this maximum is due to S⁻ ions only.

Finally at electron energies higher that 11 eV the cross section curve shows a shoulder. If the 9.5 eV peak is approximated by a gaussian curve, as in Fig. 1, and substracted from the measured total attachment cross section curve, the remaining signal could be interpreted as a new dissociative attachment process. Its peak is at energy around 12.0 eV. In this energy range Azria and Abouaf²⁵ discovered H⁻ ions with zero kinetic energy, and suggested that the HS fragment is excited into the A² state (E₀₀ = 3.801 eV, Herzberg³³).

Numerical values of the measured total electron attachment cross sections are listed in the second column of Table 4. Partial cross sections for different dissociative attachment channels are listed, too. They have been derived from the measured values by gaussian deconvolution as a first approximation. The gaussian chape cross section curve for the process having a maximum at 2.2 eV is close to the measured, so that the original data have been taken as partial cross sections in the energy range from 0.5 eV to 3.3 eV, while above that the approximated data are listed. Cross sections for the peak at 5.3 eV in the energy interval from 4.0 eV to 6.0 eV are listed as they were measured. The higher energy side of this partial cross section was approximated by a gaussian curve, fitted to the peak position and width. The peak at 9.5 eV was also fitted by a gaussian curve to its higher energy side. The signal remaining after substraction of the processes at 5.3 eV and 9.5 eV maxima was also approximated by a gaussian curve and it corresponds to the process with the maximum at 7.5 eV.

Partial attachment cross section for dissociative processes giving S⁻ ions with maxima at 2.67 eV, 5.9 eV and 9.5 eV were calculated by normalizing the relative negative ion intensity curve of Kraus¹⁹⁾ to the 9.5 eV peak cross section value as measured in our experiment.

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Negative ion	Neutral fragment	Appearance potential eV	Peak position eV	Cross section	Authors	Remarks
HS-	H	$\begin{vmatrix} 2.19 \pm 0.1 \\ 1.56 \pm 0.1 \\ 1.45 \pm 0.1 \\ \end{vmatrix}$	$\begin{array}{c} 2.75 \\ 2.27 \pm 0.1 \\ 2.20 \pm 0.1 \\ 2.10 \end{array}$	234 rel. u. $1.7 \times 10^{-22}m^2$ $2.18 \times 10^{-22}m^2$	a b c present	$\sigma_{-}(2.2)/\sigma_{+}(75) = 3.45 \times 10^{-3}$ $\sigma_{-}(2.2)/\sigma_{+}(75) = 3.51 \times 10^{-3}$
<u>s-</u>	H ₂	$\boxed{2.03\pm0.1}$	$ 2.8 2.67 \pm 0.1 $	$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	a 🤗 👌 👌	S ⁻ signal at 9.6 eV normalized to present HS ⁻ at 2.20 eV peak normali- zed to present
	H ₂ or 2H	$-$ 5.2 \pm 0.1	~6.25 5.9 ± 0.1	22 rel.u. $(=8.55 \times 10^{-24} \text{ m}^2)$ 4 rel.u. $(=3.73 \times 10^{-24} \text{ m}^2)$	a b	S ⁻ peak at 9.6 eV normalized to present HS ⁻ at 2.2 eV peak normalized to present
	H + H or H_2^*	7.8	$\begin{array}{c} \sim 9.6 \\ 9.2 \pm 0.1 \\ 9.5 \\ \sim 9.6 \\ 9.5 \pm 0.1 \end{array}$	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	a b d e present	used for normalization HS ⁻ peak at 2.2 eV normalized to present
-	H + H		\sim 12.0 \pm 0.2	$\sim 4.0 \times 10^{-24} \text{m}^2$	present	
H-	НS (²П)	4.25	$5.35 \\ 5.30 \pm 0.1$	$\frac{1.2 \times 10^{-22} \text{m}^2}{1.40 \times 10^{-22} \text{m}^2}$	e present	
	HS (2Π)		$7.65 \\ 7.50 \pm 0.2$	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	e present	
-	$\mathrm{HS}\left(\mathrm{A}^{2}\Sigma\right)$		~ 11.50 ~ 12.00	$4 \times 10^{-24} \text{ m}^2$	f present	

Characteristics of dissociative electron attachment processes in hydrogen sulphide.

a — Kraus¹⁹⁾ b — Jäger and Henglein²⁰⁾ c — Fiquet-Fayard et al.²¹⁾

d — Azria et al.²²)
e — Tronc et al.²³)
f — Azria and Abouaf²⁵)

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ELEC- TRON	TOTAL ATTAC- HMENT CROSS	PARTIAL ATTACHMENT CROSS SECTIONS In units of $\times 10^{-22}m^2$						
$ \begin{array}{c c} ENERGY \\ (eV) \\ & \\ & \\ & \\ & \\ & \\ & \\ & \\ & \\ & \\ $	HS- + H	H- + HS	$H^- + HS$	H-+HS*	S-+H2	$S^- + H_2$ or 2H	$S^- + 2H$ or H_2^*	
0 0.2 0.4 0.6 0.8	0.0 0.01 0.02 0.03 0.06	0.01 0.02 0.03 0.06		9-25 0-10 0-10 0-10			5,0 52,0 00,0	F.8 5.8 8.8
1.0 1.2 1.4 1.6 1.8	0.14 0.54 0.88 1.48 1.86	0.14 0.54 0.88 1.48 1.86					25.0 P3.0 Sand 0 Sand 0	8.8 9.9 8.9 9.0 10.0
2.0 2.2 2.4 2.6 2.8	2.16 2.12 1.73 1.03 0.50	2.16 2.12 1.69 1.08 0.55	800,1 910,8 2810	-		0.015 0.024 0.039 0.052 0.052	-9,72 0.26 0.22 0.17	2.01 0.01 3.01 3.01
3.0 3.2 3.4 3.6 3.8	0.26 0.12 0.07 0.04 0.04	0.23 0.02	 0.0015 0.0068	Carrier Constant La constantes Segon Philip The		0.048 0.037 0.023 0.016 0.011	0.10 0.10 0.07 0.03 0.04	4.11 6.11
4.0 4.2 4.4 4.6 4.8	0.06 0.11 0.21 0.46 0.76		0.025 0.079 0.20 0.44 0.77				6.84 9.659 9.659 9.625	12.4 12.5 12.4 13.0
5.0 5.2 5.4 5.6 5.8	1.18 1.37 1.38 1.05 0.72	in a second	1.13 1.36 1.36 1.13 0.77	0.031		interior	0.013 0.025 0.040	a bos isto
6.0 6.2 6.4 6.6 6.8	0.47 0.35 0.29 0.31 0.31		0.44 0.20 0.079 0.025 0.007	0.059 0.103 0.167 0.249 0.343	CT X M		0.075 0.081 0.072 0.046 0.025	
7.0 7.2 7.4 7.6 7.8	0.47 0.55 0.59 0.61 0.62		0.001	0.436 0.51 0.55 0.56 0.51			0.017	0.009 0.0178 0.0327 0.057 0.094

TABLE 4.

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ELEC- TRON	TOTAL ATTAC- HMENT CROSS	PARTIAL ATTACHMENT CROSS SECTIONS In units of $\times \ 10^{-22} m^2$						
ENERGY (eV) SECTI- ONS × 10 ⁻²² m ²	$HS^- + H$	H- + HS	H- + HS	H-+HS*	S-+H ₂	$S^- + H_2$ or 2H	$S^- + 2H$ or H_2^*	
8.0 8.2 8.4 8.6 8.8	0.61 0.60 0.58 0.58 0.60			0.43 0.34 0.25 0.167 0.10		16.0	10 102 2.0 0.0 0.0 0.0 0.0	0.146 0.21 0.30 0.398 0.498
9.0 9.2 9.4 9.6 9.8	0.63 0.67 0.69 0.69 0.66			0.06 0.03 		41.8 41.8 41.9 41.9	6.65 6.56 6.55	0.588 0.657 0.69 0.69 0.657
10.0 10.2 10.4 10.6 10.8	0.59 0.48 0.39 0.26 0.22				0.004 0.006 0.010 0.0185		2.12 2.12	0.588 0.489 0.398 0.302 0.216
11.0 11.2 11.4 11.6 11.8	0.17 0.13 0.10 0.07 0.05				0.020 0.025 0.031 0.036 0.039	233) 	64 30.0 31.0 50.0	0.146 0.094 0.057 0.032 0.0178
12.0 12.2 12.4 12.6 12.8	0.04 0.04 0.04 0.035 0.030				0.040 0.039 0.036 0.031 0.025		200 0 200 0 1 1 0 1 5 0 5	0.009
13.0 13.2 13.4 13.6 13.8	0.025 0.020 0.010 0.008 0.005				0.020 0.015 0.010 0.006 0.004			0.2 0.2 1.2 8.3

Total and partial electron attachment cross sections of the H₂S molecule.

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TOTALNI PRESECI ZA JONIZACIJU I ZAHVAT ELEKTRONA U VODONIKSULFIDU

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Apsolutni preseci za jonizaciju u domenu 10 eV do 100 eV, te za disocijativni zahvat u domenu 0 eV do 14 eV molekula vodoniksulfida mereni su primenom interakcione komore sa paralelnim pločama. Maksimum krive preseka za jonizaciju od $6.2 \times 10^{-20} \text{ m}^2$ nalazi se na energiji elektrona od 65 eV. Kriva za proces zahvata ima četiri maksimuma na energijama od 2,1 eV (HS⁻); 5,3 eV (H⁻), 7,6 eV (H^-) i na 9,5 eV (S⁻), te pregib na energijama višim od 12 eV. Korišćenjem raspoloživih relativnih merenja sa masenim spektrometrima za parcijalne procese zahvata predložene su vrednosti preseka za te pojedinačne procese.