tion sehr dünner Proben besonders geeignet. Eine Probe von 50 μm Dicke, die durch Läppen ohne Schwierigkeit hergestellt werden kann, wurde auf 2,7 μm maximale Dicke geätzt, wobei Länge und Breite nur auf etwa ein Drittel reduziert wurden. Bei Beleuchtung mit weißem Licht waren das Zentrum der Probe rot und die Randbereiche gelb durchscheinend. Bei Beleuchtung mit monochromatischem Licht waren die Randzonen auch für blaues Licht durchlässig. Das Ätzverfahren liefert also die präparative Voraussetzung für Absorptionsmessungen im Bereich der Eigenabsorption des kristallinen β-rhomboedrischen Bors (Absorptionskante bei 1,56 eV, vgl. z. B. [10, 14]); entsprechende Messungen wurden eingeleitet.

Es ist mit diesem Verfahren auch möglich, größere, im Elektronenmikroskop gleichmäßig durchstrahlbare Bereiche zu erhalten. Abbildungen 6 a und 6 b zeigen elektronenmikroskopische Durchstrahlungsaufnahmen, die die große Konzentration von Baufehlern in den z. Zt. verfügbaren Einkristallen des β-rhomboedrischen Bors deutlich machen. Es handelt sich auch hier offenbar um flächenhafte Baufehler, wie sie bereits von KLEINHENZ und RUNOW [15] an polykristallinem Material nachgewiesen wurden.

Wir danken Herrn Prof. Dr. J. JAUMANN für die Förderung der vorliegenden Untersuchungen in seinem Institut, Herrn Dipl.-Phys. J. U. ARNOLD für seine Hilfe bei der Herstellung der Gefügekammeraufnahmen und Herrn cand. chem. H. FILTER für wertvolle Hinweise. — Das Elektronenmikroskop wurde von der Stiftung Volkswagenwerk zur Verfügung gestellt.

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Light Emission in the VUV by Dissociative Excitation of CO_2 with Low-Energy Electrons

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The CO_2 molecule is dissociated in a single step process by an electron into excited atoms or ions which emit a radiation in the vuv. There are 34 radiation components from C II, O II, C I, O I and CO in the wavelength region from 450 Å to 1450 Å. We have measured the excitation functions and the dependencies of the line intensity on beam current and pressure. The excitation functions of most of the O I multiplets show a pronounced structure at about 20 to 50 eV. Immediately above the first critical potential these levels are populated via partial dissociation of CO_2. About 2% of the ions (C II and O II) formed by dissociative ionization are excited and emit a radiation in the vuv.

In a series of papers it was shown that simple molecules are dissociated in a single step process by electrons into excited atoms or ions which emit a radiation in the vuv (see for example [1, 2] and the literature mentioned there). The interest in these processes has grown because this kind of excitation takes place in the upper atmosphere [3]. On the other hand the investigation of dissociative excitation and dissociative ionization excitation gives supplementary information to mass spectrometric studies, since it results in the quantum mechanical states of the ions and atoms formed by the electron molecule collision.

This work is concerned with dissociative excitation processes of CO_2 by low-energy electrons (10 to 480 eV), which can be written as follows:

$$CO_2 + e^- \rightarrow \begin{cases} C^{+*} + 2O + 2e^- \\ O^{+*} + O + C + 2e^- \\ CO + O^* + e^- \\ CO^* + O + e^- \\ C^* + 2O + e^- \end{cases}$$

Reprints request to Dr. W. SROKA, Institut für Angewandte Physik, *D-2000 Hamburg 36*, Jungiusstraße 11.

[1] J. F. M. AARTS, F. J. DE HEER, and L. VRIENS, 6th Intern. Conf. on the Physics of Electronic and Atomic Collisions, Massachusetts Institute of Technology, Cambridge (USA) 1969, p. 423 (high energies).

[2] W. SROKA, ibidem, p. 434 (low energies).

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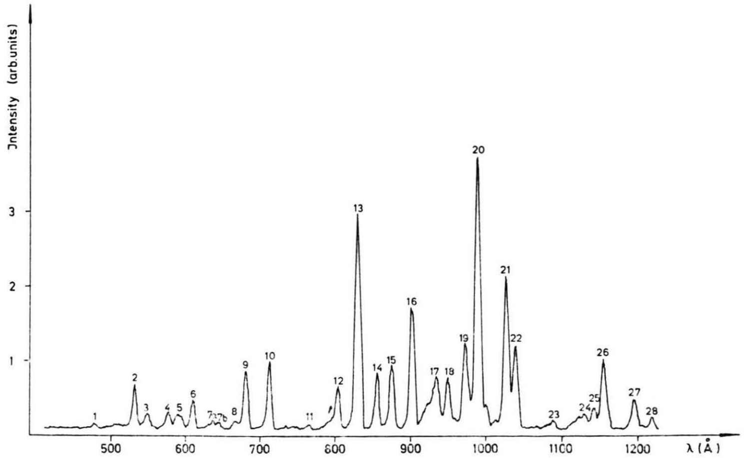
We have observed the radiation of the excited dissociation products in the wavelength region from 450 Å to 1400 Å with a vacuum monochromator and we have measured the excitation functions, the appearance potentials and the dependencies of the intensity on beam current and pressure within the collision chamber. As far as we know there exist at present no publication on dissociative excitation processes of CO_2 in the vuv.

Apparatus

An electron beam of variable energy (10 – 480 eV) and variable current (10^{-6} to $2 \cdot 10^{-3}$ A) passes the collision chamber through pressure stages. A magnetic field of 400 gauss confines the beam. The pressure within the cell (10^{-4} to 10^{-2} Torr) is measured with a capacitance manometer (MKS Baratron). During the operation the gun is heated to a temperature of 220 °C. The light emitted in the collision process is observed at right angles to the beam with a vacuum monochromator (McPherson, Model 235) and with a Bendix multiplier (M 306) as detector. A slit width of 300 μ was used because of the low photon intensity. So the wavelength resolution amounted only to about 6 Å. The energy scale of the electron beam is calibrated by measuring the appearance potential of the He line at 584 Å. If the intensity of the lines is not too low, the appearance potentials could be measured within an accuracy of ± 0.7 eV, see for illustration Fig. 6. In order to determine the absolute values of the cross sections, the excitation functions of the He-line at 584 Å and of Ly-α at 1215 Å were measured and compared with the intensity of the line in question. The excitation function of He 584 Å is sufficiently known from literature [4]. But it must be corrected for radiation imprisonment. The excitation function of Ly-α obtained by $H_2 - e^-$ collisions was recently measured [5]. Furthermore the variation of the grating reflectance (1200 lines per mm, Pt-coated, blazed for 800 Å) and the quantum yield of the multiplier cathode (tungsten) with wavelength has to be taken into account. By comparing the intensities of the CO_2 radiation with those of He 584 Å [$\sigma(100\text{ eV}) = 10^{-17}\text{ cm}^2$] or Ly-$\alpha$ [$\sigma(100\text{ eV}) = 7{,}4 \cdot 10^{-18}\text{ cm}^2$] one gets values which differ by 15% from each other. But it should be emphasized that this is not a sufficient condition for small errors. A better calibration method which uses more reference lines is prepared in our laboratory. In [6] a detailed description of the apparatus is given.

Spectra

Fig. 1 shows the spectrum obtained by electron bombardment of CO_2 at a pressure of $4 \cdot 10^{-4}$ Torr and at a beam current of 200 μA. In this experiment the electron energy amounted to 200 eV and the wavelength resolution was about 6 Å. The sensitivity of the apparatus at 500 Å is about a factor 20 larger than at 1250 Å. Fig. 2 shows the continuation of the spectrum towards longer wavelengths. The intensity ratios of the lines depend sensitively on electron energy and beam current as demonstrated in Fig. 3. This spectrum was registered at a beam current of 1.5 mA, at an energy of 50 eV and at a pressure of $7 \cdot 10^{-4}$ Torr. A striking fact is the large intensity of line No. 23 which is discussed



← Fig. 1. Spectrum of CO_2.

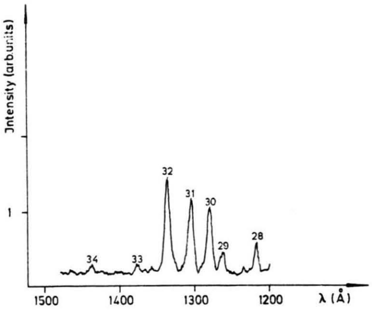


Fig. 2. Continuation of the spectrum towards longer wavelengths.

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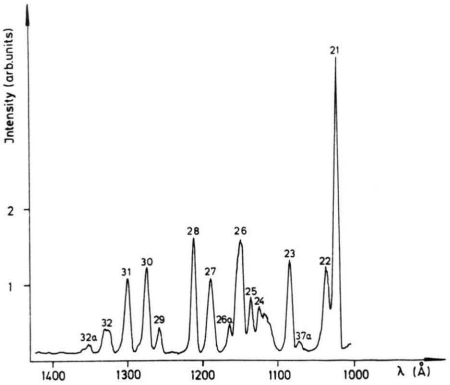
later. Unfortunately it is not possible to measure excitation functions at beam currents of the order of 1.5 mA, because secondary effects occur which result in a high energy spread of the beam.

The spectra show radiation components of C I, C II, O I, O II and CO. In Table 1 the wavelengths and the corresponding transitions are listed. Most of the lines could be identified. The intensities of the C I multiplets and of the molecular radiation of CO are extremely low in the wavelength region in question. In similar experiments in CO at the same wavelengths[7] the C I multiplets are also excited with low probability, whereas the molecular levels $B\,{}^1\Sigma^+$ and $C\,{}^1\Sigma^+$ which lead to the emission of light at 1149 Å and 1088 Å are excited with high intensity. In the case of CO_2 the molecular component of CO at 1149 Å (26) is overlapped by atomic lines and no information concerning the formation of excited CO molecules in the $B\,{}^1\Sigma^+$ state by dissociation of CO_2 can be obtained. In opposition to this line No. 23 could be identified as molecular radiation of CO. As discussed later, this radiation is only partly caused by dissociative excitation of CO_2. Every atomic line of the CO_2 spectrum also appears in the spectrum of CO because both molecules have equal dissociation products. But the cross sections for the same C II transitions are in CO larger than in CO_2 (approximately by a factor 3 to 10) and the intensity ratios are quite different. For example the most intense atomic radiation component in CO below 1050 Å is due to the C II transition $2p'\,{}^2P \rightarrow 2p\,{}^2P$ which corresponds to line No. 16 in Fig. 1. Furthermore the intensity ratio of the lines No. 21 and 22 are reversed. So the spectra of CO_2 and CO can be distinguished easily.

Excitation Processes

a) Excitation of C II

The excitation functions of the C II multiplets are shown in Fig. 4. In Table 1 the appearance potentials and the minimum energies required for the processes are listed. The minimum energy is the sum of the dissociation energies $D(\mathrm{CO}-\mathrm{O}) = 5.45$ eV[8] and $D(\mathrm{C}-\mathrm{O}) = 11.09$ eV[9], the ionisation energy of the C-atom I (C) $= 11.26$ eV[10] and the excitation energy of the carbon ion[10]. The appearance potentials are about 10 eV larger than the calculated minimum energies. Probably the excess energy is carried off by the dissociation products in form of kinetic energy. But it is also possible that a part of this energy is spend for exciting the recoiling O atoms. Immediately above threshold the energy excess is not sufficient for the process: $CO_2 + e^- \rightarrow C^{+*} + O + O^+ + 3\,e^-$ which needs an additional energy of 13.6 eV for the ionization of the



Fg. 3. Spectrum of CO_2 measured at a high beam current.

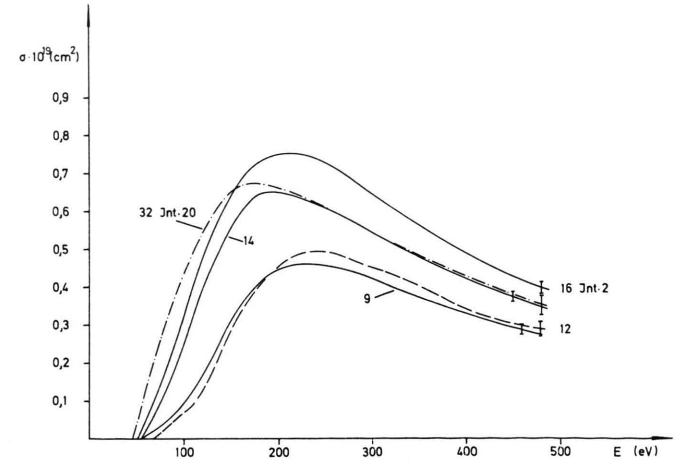


Fig. 4. Excitation functions of the C II multiplets. The intensity scale has to be multiplied with a factor 20 for No. 32 and with a factor 2 for No. 16.

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No.	Meas. Wavelength Å	Transition		Calcul. Min. Energ. eV Partial	Total	Appear. Pot. eV		(200 eV) $\cdot 10^{19}$ cm^2
1	≈ 470	O II (3d′^{2}P	$-$2p ^{2}P)	—	—	—	—	—
2	538	O II (2p′^{2}P	$-$2p ^{2}D)	—	—	—	—	—
3	≈ 549	C II (3p′^{2}P	$-$2p ^{2}P)	—	—	—	—	—
4	≈ 580	O II (2p′^{2}P	$-$2p ^{2}P)	—	—	—	—	—
		C II (5s ^{2}S	$-$2p ^{2}P)	—	—	—	—	—
5	≈ 594	C II (4d ^{2}D	$-$2p ^{2}P)	—	—	—	—	—
6	≈ 617	O II (3s ^{2}P	$-$2p ^{2}D)	—	—	—	—	—
7	644	O II (2p′^{2}S	$-$2p ^{2}P)	—	—	—	—	—
	651	C II (3d′^{4}D	$-$2p ^{4}P)	—	—	—	—	—
8	≈ 673	O II (3s ^{2}P	$-$2p ^{2}P)	—	—	—	—	—
9	687	C II (3d ^{2}D	$-$2p ^{2}P)	45.7	—	≈ 57	—	0.5
10	717	O II (2p′^{2}D	$-$2p ^{2}D)	50.7	39.7	55	≈ 85	0.7
11	≈ 770	O I (4d′^{3}P	$-$2p ^{3}P)	—	—	—	—	—
12	809	C II (3s′^{4}P	$-$2p ^{4}P)	48.5	—	—		
		C II (3d′^{2}D	$-$2p′^{2}D)	52.4	—	67	≈ 107	0.5
13	833	O II (2p′^{4}P	$-$2p ^{4}S)	45	34	48	≈ 78	2.4
14	858	C II (3s ^{2}S	$-$2p ^{2}P)	42.1	—	54.5	—	0.7
15	880	O I (3s″^{3}P	$-$2p ^{3}P)	31	19.9	23.5	≈ 50	0.8
16	904	C II (2p′^{2}P	$-$2p ^{2}P)	41.5	—	51	—	1.5
17	936	O I (4s′^{1}D	$-$2p ^{1}D)	31.7	20.7	22	—	0.7
18	953	O I (5d ^{3}D	$-$2p ^{3}P)	29.6	18.5	21.5	≈ 52	0.7
19	974	O I (4d ^{3}D	$-$2p ^{3}P)	29.3	18.2	21	≈ 44	1.3
20	990	O I (3s′^{3}D	$-$2p ^{3}P)	29.1	18	22.7	≈ 47.5	4.3
21	1027	O I (3d ^{3}D	$-$2p ^{3}P)	28.6	17.5	21.2	≈ 46	2.9
22	1040	O I (4s ^{3}S	$-$2p ^{3}P)	28.5	17.4	22.5	≈ 40	1.6
		C II (2p′^{2}S	$-$2p ^{2}P)	39.8	—			
23a	1077 ?	CO[$E\,^1\Sigma^+(0)$	$-X\,^1\Sigma^+(0)$]	—	—	—	—	—
23	1089	CO[$C\,^1\Sigma^+(0)$	$-X\,^1\Sigma^+(0)$]		16.8	15	—	—
24	≈ 1195	C I		—	—	—	—	—
25	≈ 1137	C I (6d ^{3}D	$-$2p ^{3}P)	—	—	—	—	—
26	1153	O I (3s′^{1}D	$-$2p ^{1}D)	29.2	18.2	24	47	2.7
		CO[$B\,^1\Sigma^+(0)$	$-X\,^1\Sigma^+(0)$]	—	—	—	—	—
26a	≈ 1171	?		—	—	—	—	—
27	≈ 1195	C I (4d ^{3}F	$-$2p ^{3}P)	—	—	—	—	—
28	≈ 1218	O I (3s″^{1}P	$-$2p ^{1}S)	—	—	—	—	—
29	≈ 1266	C I (3d ^{3}P	$-$2p ^{3}P)	—	—	—	—	—
30	≈ 1281	C II (4s ^{3}P	$-$2p ^{3}P)	—	—	—	—	—
31	1306	O I (3s ^{3}S	$-$2p ^{3}P)	26.1	15	21.5	38	6.4
32	1336	C II (2p′^{2}D	$-$2p ^{2}P)	37.1	—	45	—	13.3
32a	≈ 1358	?		—	—	—	—	—
33	≈ 1375	?						
34	≈ 1435	?		—	—	—	—	—

Table 1. Summary of the results.

O atom. Only in the case of No. 12 one of the O atoms may be ionized. The excitation functions are more or less curved above threshold. In the case of line No. 12 this may be partly due to the superposition of two multiplets which cannot be resolved by the monochromator. The measured excitation function include cascading from higher levels.

b) *Excitation of O II*

Fig. 5 shows the excitation functions of the O II multiplets formed by dissociative ionization excitation of CO_2. Both graphs are curved above threshold with a change in slope at about 85 eV (No. 10) and at about 78 eV (No. 13). At threshold the O^+ ion can be excited by two processes:

$$\text{a)} \quad CO_2 + e^- \rightarrow CO + O^{+*} + 2\,e^-$$

$$\text{or} \quad \text{b)} \quad CO_2 + e^- \rightarrow C + O^{+*} + O + 2\,e^-.$$

Assuming process b) the appearance potential for No. 10 is about 5 eV higher than the calculated minimum energy. This difference is equal to 3 eV for No. 13. In case a) the energy excess amounts to 16 eV (No. 10), or 14 eV for No. 13. So it cannot be determined which of the processes take place at threshold. Cascading from higher levels of the O II-ion can be excluded for No. 13. So the slight struc-

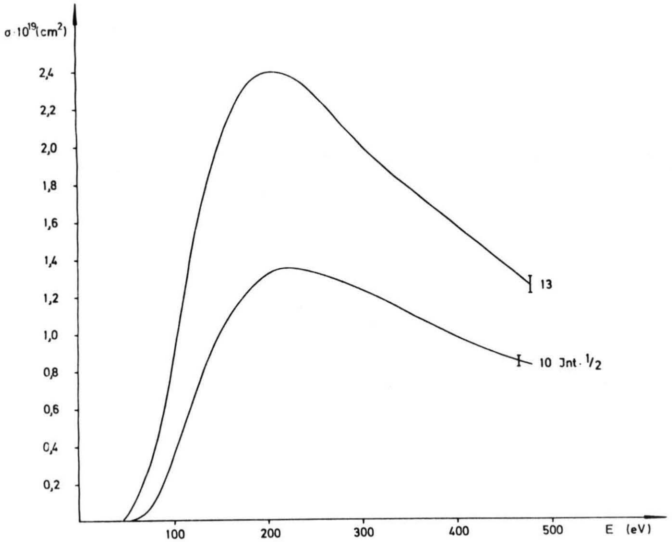

Fig. 5. Excitation functions of the O II multiplets. The intensity of No. 10 has to multiplied by a facto 1/2.

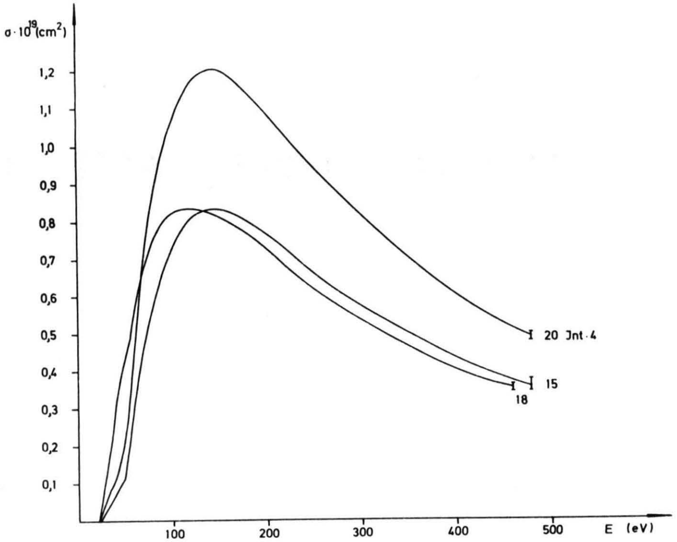

Fig. 7. Excitation functions of the O I multiplets. The intensity scale has to be multiplied by a factor 4 for No. 20.

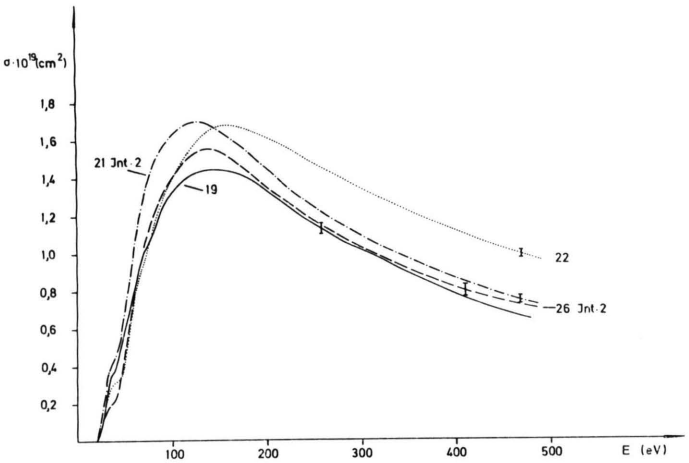

Fig. 8. Excitation functions of the O I multiplets. The intensity scale has to be multiplied by a factor given in the figure for some of the multiplets.

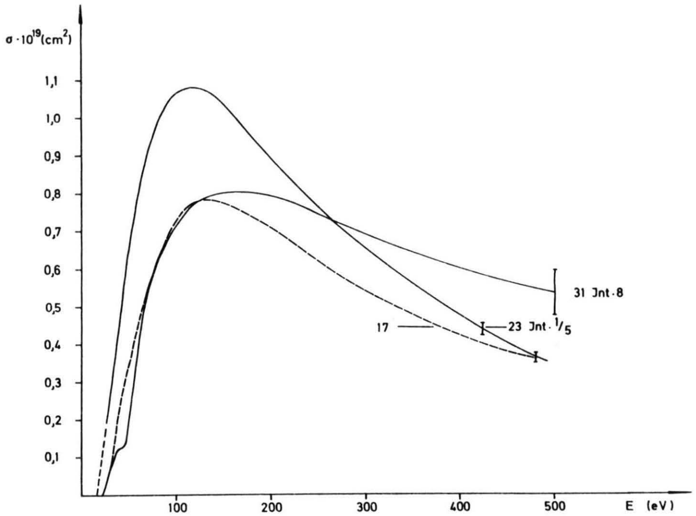

Fig. 9. Excitation functions of the O I multiplets. The intensity scale has to be multiplied by a factor 8 for No. 31 and by 1/5 for No. 23.

ture of this curve may be attributed to different excitation processes, for instance simultaneous excitation of the recoiling C- and O-atoms.

c) Excitation of O I and CO

Most of the radiation components above 870 Å are O I-multiplets. Nearly all excitation functions have a pronounced structure about 20 – 50 eV above threshold, which is demonstrated in Fig. 6. In order to control and to calibrate the energy scale of the electron beam at the beginning and at the end of each experiment the onset potential of the He 584 Å line was measured, as shown in Fig. 6.

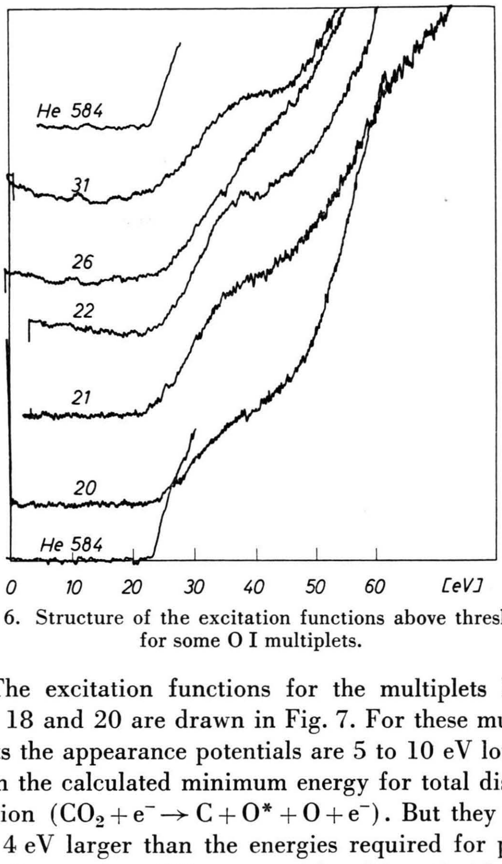

Fig. 6. Structure of the excitation functions above threshold for some O I multiplets.

The excitation functions for the multiplets No. 15, 18 and 20 are drawn in Fig. 7. For these multiplets the appearance potentials are 5 to 10 eV lower than the calculated minimum energy for total dissociation ($CO_2 + e^- \rightarrow C + O^* + O + e^-$). But they are 3 – 4 eV larger than the energies required for partial dissociation ($CO_2 + e^- \rightarrow CO + O^* + e^-$). Therefore the excited oxygen atom is formed only by this process within some eV above threshold. In this region the energy excess is not large enough for electronical excitation of the recoiling CO-molecule, because the lowest level of CO($a\,^3\Pi$) is about 6 eV above the ground state $X\,^1\Sigma^+(0)$. But it is possible that the CO-molecule is in a highly excited vibrational state. The second threshold lies about 25 to 30 eV above the first and about 10 to 20 eV above the calculated minimum energy for total dissociation. Probably the second threshold can be attributed to the onset of total dissociation. It is energetically possible, that the other dissociation products are also excited or ionized.

A similar structure was observed in CO for the same multiplets [7]. For example in CO the second onset of the excitation function for O($3s'\,^3D$), which corresponds to No. 20 in CO_2, amounts to 38 eV. This value is about 10 eV lower than that in the case of CO_2. A less pronounced structure was found for the same state of O I, excited by the process $O_2 + e^- \rightarrow O + O^* + e^-$. We did not find this structure in our first experiments in O_2, because the sensitivity of the apparatus was too low at the time [2].

Fig. 8 shows the excitation functions of some additional O I-multiplets. These components are excited in the same way as those represented in Fig. 7. Immediately above the first threshold the CO_2 molecule is dissociated into an excited O-atom and a CO molecule which may be vibrationally excited in the case of No. 19 and No. 21. Considering the uncertainty of the energy measurement ($\Delta E \approx 0.7$ eV), the CO molecule may be in the $a\,^3\Pi$ state for No. 22 and No. 26. Under this assumption the kinetic energy of the recoiling dissociation products should be less than 1 eV. Component No. 22 results from the overlapping of a O I and a C II multiplet. The minimum energy required for the population of C II ($2p'\,^2S$) is 39.8 eV. Accordingly at energies some eV above the first onset the radiation can be attributed to the transition O I ($4s\,^3S \rightarrow 2p\,^3P$). The second onset at about 40 eV is equal to the minimum energy required for the excitation of the C II multiplet and it is larger than that for the excitation of O I by total dissociation of CO_2. As already mentioned, in CO the line corresponding to No. 22 has a higher intensity than that corresponding to No. 21, in opposition to the results obtained in CO_2 and O_2. In general in CO the C II multiplets are excited with a higher intensity than the O I lines. Therefore it can be supposed that the main part of the photons of No. 22 are emitted by O I. It is possible that the level $B\,^1\Sigma^+(0)$ of CO is populated by dissociative excitation of CO_2. The transition $B\,^1\Sigma^+(0) \rightarrow X\,^1\Sigma^+(0)$ appears with high intensity in the spectrum of CO. In the case of CO_2 this radiation is overlapped by line No. 26. As demonstrat-

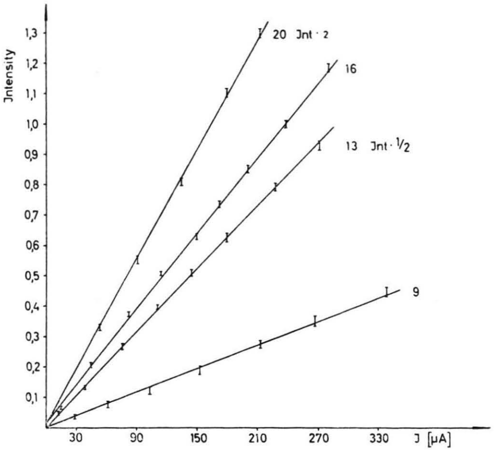

Fig. 10. Intensity versus beam current.

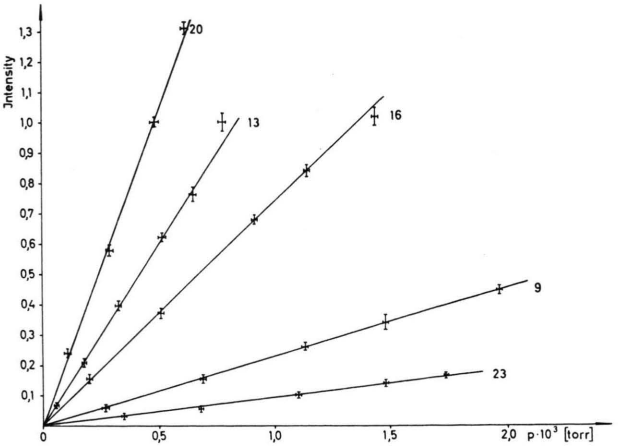

Fig. 11. Intensity versus pressure within the collision cell.

ed in Fig. 1 this line is a little bit asymmetric which may be a hint that CO ($B\,^1\Sigma^+$) is populated.

No information can be obtained whether or not at the second critical potential the other dissociation products are also excited or ionized and which part of the energy excess is carried off by them in form of kinetic energy.

Fig. 9 shows the excitation functions of two O I multiplets (No. 17 and 31) and of the transition $C\,^1\Sigma^+(0) \rightarrow X\,^1\Sigma^+(0)$ of CO (No. 23). The latter one was measured at a beam current of 400 μA, whereas in all other cases the beam current was of the order of 200 μA. Line No. 17 shows only a weak structure above threshold. For the O I multiplets of Fig. 9 the same remarks hold, which were mentioned above. It was not possible to determine the appearance potential of the CO component exactly, but it was found to be higher than 11.4 eV, which is the energy for direct excitation of CO.

In all cases discussed above the intensity of the light depends linearly on beam current and pressure. This is demonstrated in Fig. 10 and Fig. 11. Therefore the excited states are populated in a single step process. An exception was found for line No. 23, the intensity of which is linear with pressure (Fig. 11), but it depends not linearly on the beam current (Fig. 12). At constant pressure the curve can be represented by a function $I = a\,i + b\,i^2$ (I intensity; i current; a, b constants). This means that the level $C\,^1\Sigma^+(0)$ of CO is populated by the single step process

$$CO_2 + e^- \rightarrow CO^* + O + e^-$$

and simultaneously by the two step process

$$CO_2 + e^- \rightarrow CO + O + e^-, \quad CO + e^- \rightarrow CO^* + e^-.$$

Assuming a cross section of the order of 10^{-16} cm² for the dissociation of CO_2 into CO and O and a cross section of the order of 10^{-17} cm² for the excitation of CO, one easily estimates that the two step process participates in the population of $C\,^1\Sigma^+(0)$.

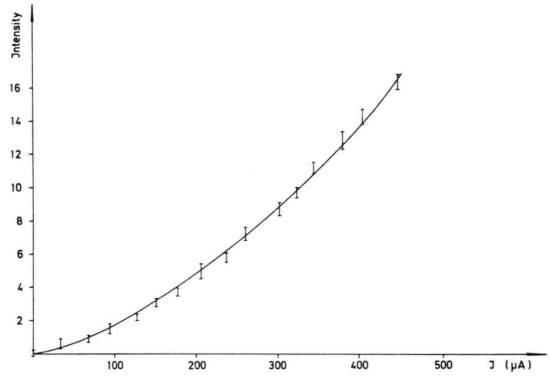

Fig. 12. Intensity of No. 23 versus beam current. The pressure amounted to 2.8×10^{-3} Torr.

The light produced by dissociative excitation may be polarized in some cases. However the intensity was not sufficient for polarization measurements.

General Remarks

The measurements show clearly the formation of excited oxygen atoms by partial dissociation of CO_2 at energies immediately above threshold. The structure of the O I multiplets is probably due to the onset of total dissociation. At present we have not enough information to decide whether or not excited O II ions are also formed by partial dissociation. Unfortunately the intensity of the C I radiation was not sufficient to measure excitation functions. The carbon ions are excited by total dissociation. We have found only an extremely small contribution of vuv radiation from highly excited CO molecules formed by dissociative excitation of CO_2. These observations are in agreement with measurements in the visible and near UV [11].

In [12] the total cross section for dissociative ionization was measured. It amounts to $10^{-16}\,cm^2$ at 200 eV. Whereas the excited ions (O II and C II) which emit light in the vuv are produced with a total cross section of about $2 \cdot 10^{-18}\,cm^2$ at 200 eV. The cross section for the formation of highly excited C and O atoms has nearly the same value as that for the excitation of the ions. In CO the fraction of the excited ions is larger.

It is very important for the understanding of dissociation processes to measure the kinetic energy of the dissociation products and to determine whether or not the other recoiling atoms and ions are also excited. We hope to get more information on these questions by using coincidence technique *.

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* *Note added in proof:* In some interesting papers (see [13] and the literature mentioned there) the dissociation of CO_2 into metastable CO* was studied. According to these works metastable CO* is formed with a high cross section, whereas in our experiments excited CO molecules (which radiate in the vuv!) are only produced with low probability, e. g. process 26 Table 1. But it follows from our experiments that a metastable CO molecule may also be formed together with an excited O atom with a cross section of the order of $10^{-19}\,cm^2$.