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Cross-sections for CO^+ and CO^{2+} production by electron impact on CO molecule

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Abstract

Ionization and dissociative ionization of CO molecule by electron impact is relevant in astrophysics, plasma physics and environmental physics. Nevertheless there are relatively few experimental and theoretical results for the corresponding cross-sections, in particular results for CO^{2+} are scarce and in disagreement. In the present work cross-section ratios for CO^{2+} and CO^+ have been measured to provide new cross-sections values and comparison between existing results. Also the fraction of total ionization due to dissociative processes has been measured and it is pointed out that previous accurate results of total cross-sections, when corrected by dissociative ratios obtained by time-of-flight mass spectrometry, can give cross-sections values useful by theirself and for evaluation of the existing data. (Int J Mass Spectrom 219 (2002) 351–361)

Keywords: CO; Double ionization; Electron impact; Time-of-flight spectrometry

1. Introduction

Electron impact ionization of atoms and molecules has been the subject of many experimental and theoretical studies due to its fundamental importance in atmospheric science and plasma processes. In particular, related dissociative phenomena in molecules are of great interest. In general, when an electron of sufficient energy passes through a gas of diatomic molecules of components A and B can cause several ionization processes by collision, i.e., direct ionization producing a single ionized molecule (AB^+), dissociative ionization (A^+, B^+), double ionization (AB^{2+}, A^{2+} or B^{2+}), etc. Mass spectrometry is a very valuable technique to carry out these experimental studies.

As is well known CO is an important constituent of planetary atmospheres and comets and there is a continuous interest to obtain accurate values of electron impact ionization and dissociative ionization cross-sections [1–8]. As is shown later on, there are few experimental data in agreement of single, double and dissociative ionization of this molecule and neither between experimental and theoretical results. In particular for CO^{2+} there were only two previous experiments [4,8] and they are in disagreement. Also it must be noticed that according to [4,8,9] CO^{2+} ion is formed in a metastable state that decays with a lifetime of several microseconds [9]. As measurements of [4,8] were made at times of 10 and 2 μs, respectively, it seems necessary to perform measurements at different time. This work was made with the aim to review the existing data, to add new values where necessary,

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and to study the possibility of using reliable values of total ionization cross-sections as a further source of data. We made new measurements of ionization cross-sections for the processes CO → CO^+ and CO → CO^{2+} and for the fraction of total ionization coming from dissociative ionization processes. Also we reviewed the existing data on CO molecular ions and renormalized the values of [4,6] by using the more recent values for Ar^+ and Ar^{2+} cross-sections of Straub et al. [10]. The present results have an experimental uncertainties of 10% and comparison is made with the previous experimental results (1, 2, 4–8) and with the theoretical values of [11].

Among earlier studies about this molecule a relevant work was made by Rapp et al. [1] and Rapp and Englander-Golden [2]. In these works the authors obtained the fraction of total ionization coming from dissociated ions with kinetic energy grater than 0.25 eV [1] and electron impact total ionization cross-sections [2] for some atoms and molecules including CO. To obtain total cross-sections an electron beam was collimated and confined by a strong magnetic field to pass through a chamber containing the gas in study. The ions were accelerated and collected and the total cross-section were determined measuring both ion and electron currents. The absolute normalization of the relative data was accomplished taking H_2 as normalization gas. The range of electron energy was from threshold to 1000 eV.

Locht [3] studied dissociative ionization of carbon monoxide for electron impact of energy lower than 100 eV measuring the kinetic energy distribution of the produced ions. Hille and Märk [4] using a double-focusing mass spectrometer obtained absolute ionization cross-sections for the production of CO^+ and CO^{2+} in the range of energies from threshold to 180 eV. Relative measurements were normalized by using Ar ionization cross-sections.

More recently Orient and Srivastava [5] measured total and partial ionization cross-sections for CO^+, C^+ and O^+, by using a capillary array to form a molecular beam, a quadrupole mass spectrometer, a charged-particle detector and a faraday cup to measure the incident electron current. Normalization of relative values was accomplished with cross-sections values of the noble gases He to Xe. Freund et al. [6] measured ionization cross-sections to form CO^+ in the range of electron energies from 14 to 200 eV. The produced ions were focused at the entrance of a hemispherical energy analyzer, which separates parent ions from fragment ions. These authors put their results on an absolute scale by using Ar and Kr as reference gases.

The most recent experimental values to our knowledge have been obtained by Tian and Vidal [7,8]. These authors measured the cross-section for electron impact ionization and dissociative ionization of CO for electron energies from threshold to 600 eV. A continuous effusive flow molecular beam is crossed with a pulsed electron beam at right angles. The produced ions are extracted into a focusing time-of-flight mass spectrometer. The ionization signal were provided by a multichannel plate detector. Ar was used as reference gas. In [8], the cross-section of the channels corresponding to the production of ions pairs was measured by coincidence techniques providing data for cross-sections of double and triple ionization.

From a theoretical point of view, Khare et al. [12] performed a model to calculate the electron impact ionization cross-sections of molecules leading to the different ionization products. More recently Pal et al. [11] applied this model to obtain semiempirical data on direct, dissociative, double and triple ionization cross-sections of CO molecule by electron impact. They used experimental data for optical oscillator strengths of [13,14] and high energy collision experimental values of [15]. For dissociative ionization the authors found qualitative agreement with the experimental results but they noticed the need of further investigations.

In the present work a time-of-flight mass spectrometer has been used to collect the ions produced from CO molecule by electron impact. In all the mentioned works effusive gas flow have been used to introduce the gas in the ionization chamber, but in this work molecules were introduced as a supersonic jet. We pay a special attention to relate accurately the cross-section values for first a second CO ion production. For spectrometer calibration we have used Ar as reference gas.

Argon has been extensively studied as can be seen in the work of Tawara and Shevelko [16] that reports data bases about ionization processes of neutral atoms and negative and positive ions by electron impact. In the present work the mentioned data for Ar ionization by electron impact of Straub et al. [10] have been used.

2. Experimental

Fig. 1 shows the experimental set-up. It consists in three basic units: the ionization chamber, the linear time-of-flight mass spectrometer (Comstock Incorporated Model TOF 101) and the detection system. Two vacuum systems with turbomolecular pumps were operating close to the collision chamber and the detector, respectively. The ultimate pressure was 10^{-7} Torr in the flight tube. The beam of the molecules in study was formed by pulsed supersonic expansion from the gas reservoir (2.5×10^3 Torr) to the ionization chamber (10^{-7} Torr) through a nozzle of 0.1 mm of diameter. The electromagnetic pulsed valve (General Valve Corporation, Model Iota One) produced gas pulses of 400 µs at 0.5 Hz repetition rate. The velocity of the supersonic jet, proportional to the inverse of square mass root of atoms or molecules, is typically 700 m/s for CO molecules. This beam crossed at right angle with a pulsed electron beam. The electron gun was of triode structure, designed in a similar way to that used in previous experiments [17]. The electrons were extracted through a pulsed control grid. The duration of the positive extracting electron pulse was 350 ns with sharp (<10 ns) edges. The electron energy range was 0–600 eV.

The electron pulse was synchronized with the gas pulse and delayed from the aperture of the pulsed valve in order to achieve simultaneous arrival for the electron and molecular beam at the centre of the chamber to optimize the ion production and collection.

The resulting positive ions were accelerated towards the drift tube using two extraction plates. An extraction pulsed voltage of 200 V was applied to the plates at the end of the electron pulse. The pulse duration can be varied in the range 0.2–5 µs. The drift tube length was 100 cm at an operation voltage selectable in the range −1700 to −3000 V, so the positive ions

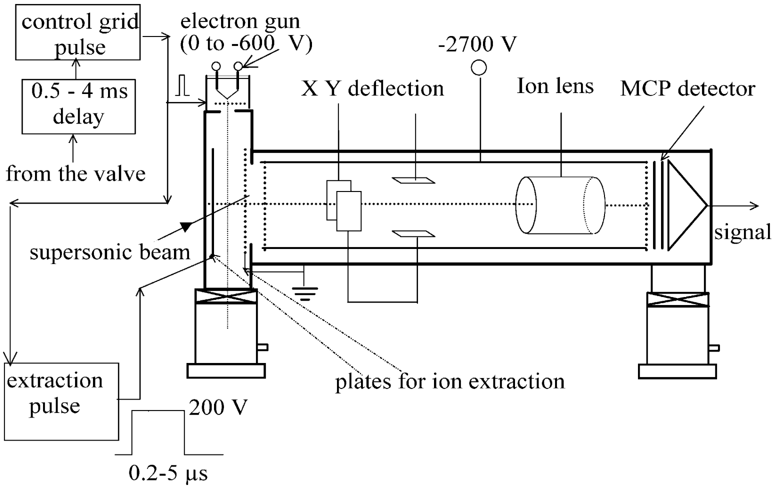

Fig. 1. Experimental set-up. A supersonic jet of the studied molecules crosses at right angle with a pulsed electron beam in a collision chamber. At once electron impact ionization is made, a 200 V pulse is applied to the extraction plates driving the positive ions to a flight tube biased at −2700 V. After a path length of 100 cm, a 4 cm diameter dual microchannel plate detects these ions. To optimize ion collection the drift tube is provided with X and Y deflection plates and a cylindrical lens. Two turbomolecular vacuum pumps operate close to the collision chamber and the detector, respectively.

extracted from the collision chamber were accelerated towards the detector. For the present work the voltage of the flight tube was −2700 V. The drift tube contained two set of deflection plates at right angle that provides a better collection of the ions. Deflection potentials V_x and V_y can be varied between −250 and +250 V. A cylindrical lens of variable bias located between the deflection plates and the detector acts as an Einzel lens. The ion lens section can be considered as the central element and the flight tube section at either end as the lateral ones.

The detector is a 4 cm diameter dual microchannel plate than can provide linear pulses up to 0.5 A. It must always maintain a positive potential of approximately 1000 V per plate to provide proper biasing for electron multiplication. In this work the voltage applied of the multichannel plates was −2000 V.

The ion detection output signals from the microchannel plates fed a preamplifier, an amplifier and a digitizer system (Phillips PM3350 A) that classified the signal corresponding to ion detection according to their time-of-flight. A personal computer stored the information for further analysis. The time-of-flight of the studied ions ranged from 3 to 10 μs.

3. Results and discussion

3.1. Processes of direct and dissociative ionization of CO

Typical mass spectra of ionization products of CO for electron energies of 150 and 450 eV are shown in Figs. 2 and 3, respectively. As can be seen from the

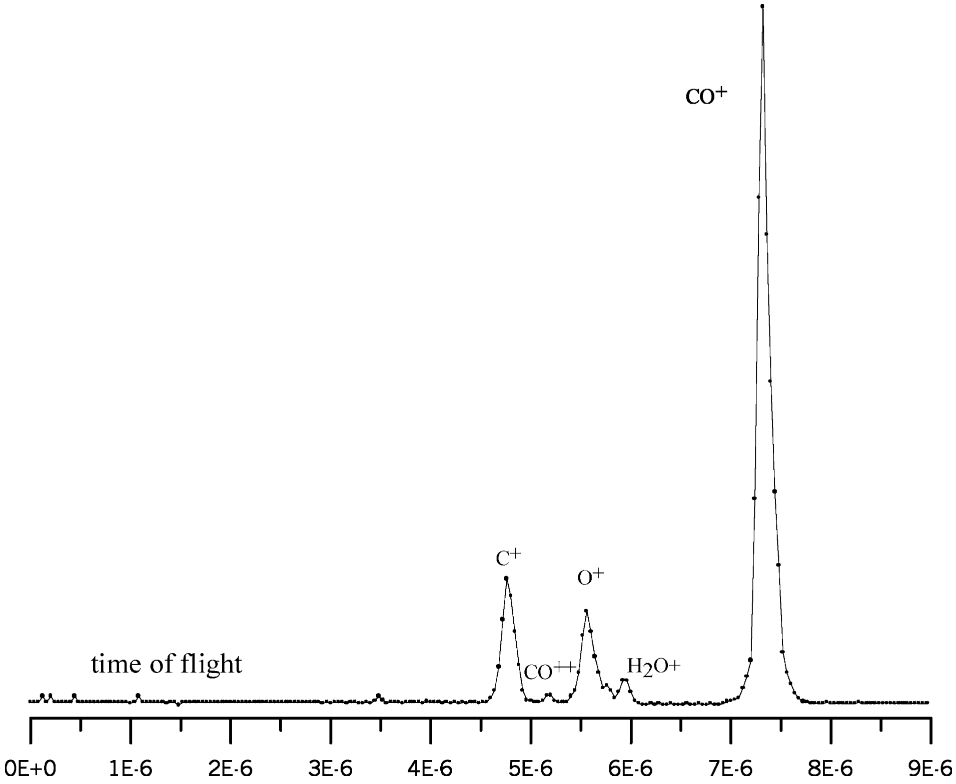

Fig. 2. Typical time-of-flight spectrum in the range 0–9 μs of ionic fragments produced by collision of CO with electrons of 150 eV energy. Also peaks due to H_2O can be seen.

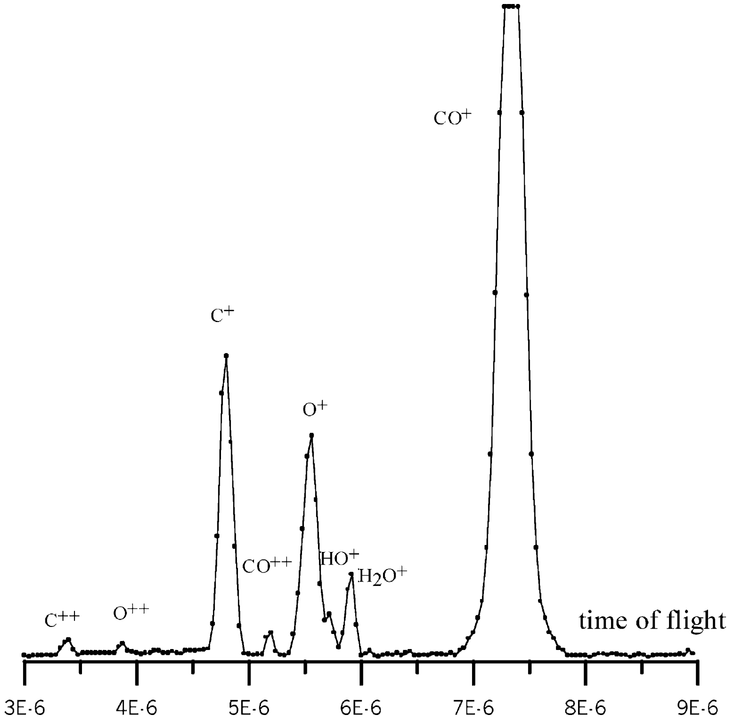

Fig. 3. Typical time-of-flight spectrum in the range 3–9 μs of ionic fragments produced by collision of CO with electrons of 450 eV energy.

figures the parent ions CO^+ and CO^{2+} and the fragments C^+, O^+, C^{2+} and O^{2+} has been detected. The spectra also contains signals from traces corresponding to water (H_2O^+, OH^+) that is the principal background gas present in our experimental system. No traces of ${N_2}^+$ were found in the apparatus. Background subtraction was made for all the present measurements.

The most probably ionization processes for CO electron impact are:

- Ionization processes leading to single ionized ions: single ionization and dissociative ionization:

  $$CO + e^- \rightarrow CO^+ + 2e^-$$

  $$CO + e^- \rightarrow C^+ + O + 2e^-$$

  $$CO + e^- \rightarrow C + O^+ + 2e^-$$

- Ionization processes leading to doubly ionized ions:

  $$CO + e^- \rightarrow CO^{2+} + 3e^-$$

When CO^{2+} molecules are formed in an excited state, $(CO^{2+})^*$, deexcitation occurs by the channels:

$$(CO^{2+})^* \rightarrow C^+ + O^+$$

$$(CO^{2+})^* \rightarrow C^{2+} + O$$

$$(CO^{2+})^* \rightarrow C + O^{2+}$$

Other channels can contribute to the total cross-section for the production of multicharged positive ions but it represents only a small fraction of the total (about 0.02%) [8].

As one can see in Fig. 3, the major contribution to the mass spectrum are the direct ionization CO^+ peak and the fragmentation C^+ and O^+ peaks. The contribution of the double ionization CO^{2+} peak is smaller. In what follows we shall use the notation $\sigma(CO^+)$, $\sigma(C^+)$, $\sigma(O^+)$, $\sigma(CO^{2+})$, $\sigma(O^{2+})$, etc. for the cross-section of production of every fragment by electron impact on CO. In this work from the ionization signals we have measured the ratios $\sigma(CO^{2+})/\sigma(CO^{+})$, $\sigma(C^{+})/\sigma(CO^{+})$, $\sigma(O^{+})/\sigma(CO^{+})$ that are proportional to the relative values of the cross-sections for the corresponding processes.

3.2. Cross-section for production of CO^{+}

The total ionization cross-section of CO by electron impact can be written as a charged-weighted sum of partial ionization cross-sections:

$$\sigma_{\text{ionization}}\,(\text{total}) = \sigma(CO^{+}) + \sigma(C^{+}) + \sigma(O^{+}) + 2\sigma(CO^{2+}) + \text{other triple ionization processes.}$$

We can write

$$\sigma(CO^{+}) = \sigma_{\text{ionization}}\,(\text{total})(1 - f)$$

where we denote by f the fraction of total ionization leading to ionization products other than CO^{+}. So it can be obtained $\sigma(CO^{+})$ measuring f and the total ionization cross-section.

As mentioned in Section 1, experiments to obtain total ionization cross-sections can be made with different experimental methods that those used to obtain fragmentation cross-sections, therefore selecting a total cross-section from the literature and measuring the fraction of total ionization it can be obtained a value for $\sigma(CO^{+})$ that allows comparison between results with different calibration references.

To evaluate the fraction f it is necessary the complete collection for ions produced by the dissociation of the molecule. In general this is not an easy task due to the fact that the fragments come off with kinetic energy [1,8]. The kinetic energy distribution of C^{+} and O^{+} have been measured by Locht [3]. At 100 eV most of C^{+} ions are produced with kinetic energy less than 1 eV and most of O^{+} ions with energy above 1 eV. In general, dependence of collection efficiency on the kind of ions in the case of conventional ions sources

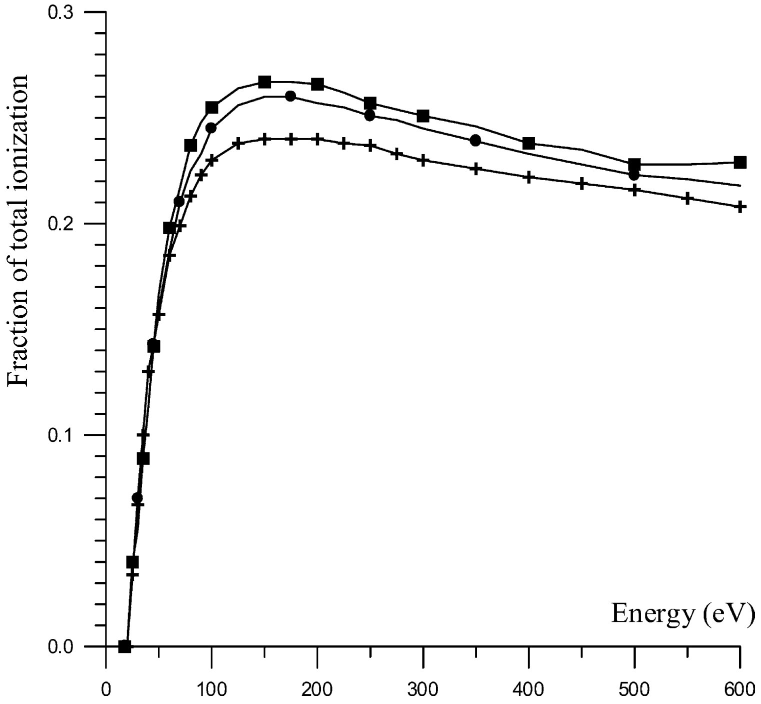

Fig. 4. Fraction of total ionization cross-section corresponding to ionization products different from CO^{+} vs. electron energy. (+) [1], (■) [8], (●) present work.

and mass spectrometers can be expected. As explained above the X and Y plates and the Einzel lens of the time-of-flight tube allow to optimize ion gathering.

Fig. 4 shows the fraction of total ionization vs. energy of electrons for the present experiment. The present experimental error is 10%. Also values of [8] and the earlier values of [1] are included for comparison. Average differences between our results and the recent values of [8] is 5% that is within the error limits.

Table 1 shows the available data for the cross-section of CO^+ production by electron impact. The first and second columns show the results deduced from the total cross-section values of Rapp and Englander-Golden [2] using the values for the fraction of total ionization of the present work, and of Tian and Vidal [8], respectively. The third column shows the values of [7]. Results of [7,8] were obtained normalizing with the cross-sections values of Straub et al. [10] for Ar^+. The error quoted for the authors are in the range 10–15%. Column four shows the experimental values of Freund et al. [6] (accuracy 10%). These values were obtained normalizing with a value for $\sigma(Ar^+)$ of 2.57×10^{-16} cm^2 at 70 eV electron energy but the values shown in the table were renormalized to the value of [10] (2.67×10^{-16} cm^2) for the same electron energy.

The two remaining columns of experimental values of Table 1 show the values of Orient and Srivastava [5], with a quoted 15% error, and of Hille and Märk [4] with experimental error in the range

Table 1
Cross-section for electron-impact single ionization of CO giving CO^+ (10^{-16} cm^2)

Electron energy (eV)	Experimental values						Theoretical values
	[2] (corrected)		[7]	[6]	[5]	[4]	[11]
	Present work	[8]					
17.5	0.21	0.21	0.14	0.23	0.21	0.14	0.14
20	0.43	0.43	0.32	0.44	0.34	0.29	0.27
25	0.82	0.81	0.74	0.78	0.72	0.65	0.50
30	1.15	1.17	1.04	1.10	1.04	0.74	0.77
35	1.39	1.41	1.33	1.31	1.28	1.10	1.10
40	1.55	1.58	1.45	1.44	1.53	1.29	1.35
45	1.69	1.69	1.53	1.56	1.63	1.42	1.55
50	1.78	1.76	1.63	1.62	1.73	1.49	1.75
60	1.90	1.87	1.72	1.74	1.88	1.56	2.00
70	1.97	1.95	1.78	1.80	1.98	1.74	2.10
80	2.00	1.97	1.81	1.81	2.03	1.81	2.16
90	2.01	1.98	1.82	1.80	2.05	1.84	2.20
100	1.99	1.97	1.82	1.79	2.06	1.87	2.22
125	1.95	1.93	1.74	1.76	2.05	1.88	2.23
150	1.90	1.88	1.69	1.69	2.00	1.85	2.22
175	1.82	1.80	1.63	1.61	1.93	1.82	2.18
200	1.76	1.74	1.54	1.44	1.85	1.74	2.12
225	1.68	1.67	1.51		1.77		2.06
250	1.62	1.60	1.43		1.70		2.00
275	1.56	1.55	1.35		1.65		1.92
300	1.50	1.49	1.30		1.59		1.84
350	1.40	1.39	1.19		1.51		1.69
400	1.32	1.31	1.12		1.45		1.57
450	1.23	1.22	1.02		1.36		1.45
500	1.16	1.15	0.98		1.32		1.35
550	1.11	1.10	0.92				1.25
600	1.05	1.04	0.86				1.20

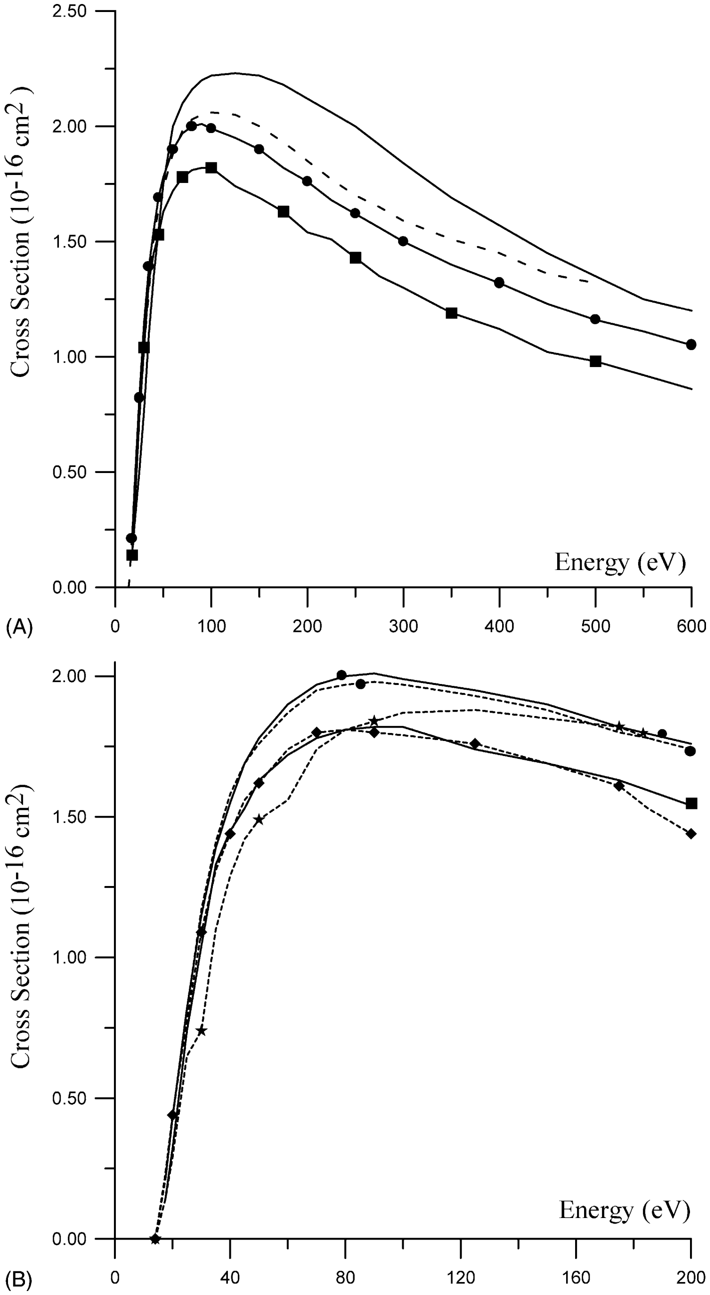

Fig. 5. (A) Cross-section for CO ionization giving CO$^+$ vs. electron energy up to 600 eV. (—) [11], (- - -) [5], (■) [7], (●) [2] correction of present work. (B) Cross-section for CO ionization giving CO$^+$ vs. electron energy up to 200 eV. (★) [4], (◆) [6], (■) [7], (- -●- -) [2] correction of [8], (–●–) [2] correction of present work.

10–20%. The latter authors normalized their results with the value for $\sigma(\mathrm{Ar}^+)$ of [18] at 136 eV electron energy (2.29×10^{-16} cm^2). Similar to [6], these values were renormalized with the corresponding value of [10] (2.47×10^{-16} cm^2). Theoretical values of Pal et al. [11] can be seen in the last column.

Figs. 5A and B show the energy dependence for $\sigma(\mathrm{CO}^+)$ vs. the electron energy obtained for the different authors. In Fig. 5A the values of the first and third columns of this table are shown. The shape of the two experimental curves are similar but the values from [2] are systematically 10% higher that those of [7], although this discrepancy is between the experimental error range. The same figure shows the values of Orient and Srivastava [5] that are higher and have a different shape for values of electron energy above 350 eV. Theoretical values of Pal et al. [11] are also included and are higher than the lowest experimental ones [7] by about 20%.

Fig. 5B shows with some more detail the curve for electron energies from the threshold to 200 eV and includes the experimental values of Freund et al. [6] and Hille and Märk [4]. It can be seen that there are small differences between the values of [2], corrected by the results of the present work, and those of [8]. It is noticeable the excellent agreement existing between the values of this last reference and the renormalized values of Freund et al. [6]. The shape of the curve corresponding to the values of Hille and Märk [4] exhibits a different energy dependence even in this energy range.

3.3. Cross-section for production of CO^{2+}

As mentioned in Section 1 we paid special attention to the measurement of the cross-section ratio for production of CO^{2+} and CO^+ vs. electron energy. The relative efficiency of the experimental system to

Table 2
Cross-sections for electron impact double ionization of CO giving CO^{2+} (10^{-18} cm^2)

Electron energy (eV)	Experimental values				Theoretical values
	Present work CO^+ values		[8]	[4]	[11]
	This work	[7]			
45	0	0	0.001	0	0
50	0.068	0.063	0.084	0.10	0.15
60	0.273	0.247	0.236	0.36	0.31
70	0.548	0.495	0.534	0.69	0.44
80	0.668	0.605	0.622	0.94	0.56
90	0.767	0.694	0.757	1.11	0.67
100	0.832	0.761	0.821	1.21	0.79
125	0.915	0.816	0.860	1.26	1.00
150	0.913	0.812	0.788	1.27	1.10
175	0.880	0.788	0.851	1.22	1.12
200	0.826	0.723	0.697		1.11
225	0.754	0.678	0.745		1.07
250	0.692	0.611	0.669		1.04
275	0.636	0.551	0.585		1.00
300	0.594	0.514	0.548		0.96
350	0.524	0.445	0.457		0.88
400	0.471	0.400	0.450		0.815
450	0.426	0.354	0.372		0.755
500	0.392	0.331	0.331		0.71
550	0.364	0.302	0.329		0.66
600	0.338	0.277	0.335		0.62

detect the first and second CO ion has been measured by comparison of the relative cross-section to produce Ar^{2+} and Ar^{+} of Straub et al. [10]. These comparisons were made along the experiment in the electron energy range from 17.5 to 600 eV. The estimated accuracy of the present results for the ratio for CO^{2+} and CO^{+} production is better than 10%.

Table 2 shows the present experimental results for the cross-section of production of CO^{2+}. Values of other authors are also shown for comparison. In the first and second columns of cross-section data the experimental values shown were obtained with the present experimental $\sigma(CO^{2+})/\sigma(CO^{+})$ ratios and the CO^{+} corrected values of Rapp and Englander-Golden [2] for CO^{+} and those of [7], respectively. The third and fourth columns show the results of Tian and Vidal (10–15% experimental error) [8] and Hille and Märk (30% experimental error) [4], respectively. In the last column the theoretical values from [11] are given.

Fig. 6 shows the results of dependence of $\sigma(CO^{2+})$ on electron energy obtained in the mentioned works. Theoretical values are higher than the experimental ones with the exception of Hille and Märk [4] results. The curves corresponding to the corrected values of [2] and the direct results of Tian and Vidal [8] have a similar shape. The first values are about 12% higher than that of [8]. Also the results of the present ratio of cross-sections applied to the $\sigma(CO^{+})$ of the [7] are shown. These last values are lower than the direct results of [8] by 6%. Although good agreement exists between these values considering the 10% experimental uncertainty of both results it must be taken into account that as reported in [8,9]. CO^{2+} molecules can be formed in a metastable excited state, $(CO^{2+})^{*}$. According to the results of [18] the decay of the state could take place either with a 16 μs decay or with a 9 μs decay with a 30% of stable component. Therefore, cross-sections values obtained at a longer time-of-flight could be lower.

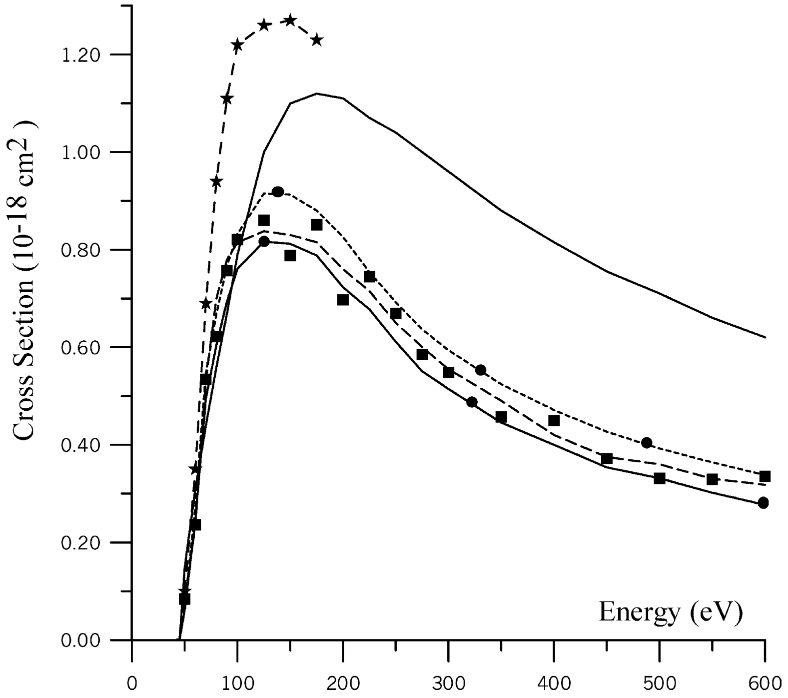

Fig. 6. Cross-section for CO ionization giving CO^{2+} vs. electron energy up to 600 eV. (★) [4], (■) [8], (—) [11], (- -●- -) present work (from previously corrected values for CO^{+}), (–●–) present work (from CO^{+} values of [7]).

Measurements of the present work and [8] have been made for a time-of-flight of 5.5 and 2 μs, respectively, whereas for [4] that reports the highest $\sigma(CO^{2+})$ values the time-of-flight used was 10 μs. In the present experiment, the potential applied in the drift tube was varied in order to produce a time-of-flight variation of ±0.5 μs but no changes in the cross-section ratio for CO^{2+} and CO^{+} was observed within our experimental error. The present results and that of [8] are compatible with a metastable lifetime of tens of microseconds, but it seems likely [9] that a stable component exists.

4. Conclusions

The present work adds new measurements for ionization by electron impact of CO molecule and its dissociation and reviews the available data from the literature. In general comparison of total ionization cross-sections with recent measurements of ionization is useful considering the fragmentation processes that can be studied by mass spectrometry techniques. In summary in the present experiment, although systematic discrepancies (6–12%) exist due the different experimental methods used, results are compatible with the most recent values within the errors quoted by the authors. The $\sigma(CO^{+})$ results obtained by correction of earlier values of Rapp and Englander-Golden [2] are 10% higher than the most recent of Tian and Vidal [7] and of Freund et al. [6] and in reasonable agreement with both, within the experimental uncertainties.

For $\sigma(CO^{2+})$ there were few previous data, so the results obtained from [2] and the present experiment provide new values that add support to the results of [8] and show that a fast decay of CO^{2+} has not been observed.

On the other hand, theoretical calculations leading to general models are very important. The available theoretical results used in the present work for comparison agree with the experimental results in the shape of cross-section vs. electron energy plot. The theoretical model used by the authors can be considered in general satisfactory. Discrepancies with experimental values can be related to details of the calculation, as the number of fragmentation channels considered and the semiempirical parameters used.

Acknowledgements

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