

The role of dissociative electron capture in collisions of slow He^{2+} ions with CO

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Abstract

Translational energy spectroscopy (TES) has been used to study one-electron capture by $0.2\text{--}1\text{ keV amu}^{-1}$ He^{2+} ions in CO and identify, for the first time, the main excited product channels and determine cross sections for both dissociative and non-dissociative processes. Cross sections are shown to be strongly energy dependent over the energy range considered with the contribution from exothermic dissociative electron capture channels associated with $\text{He}^+(1s)$ formation accounting for about 99% of the total cross section at the lowest energy considered. Endothermic channels arising from non-dissociative one-electron capture have been observed which increase in relative importance as the impact energy increases. These lead to either $\text{He}^+(n = 2)$ or $\text{He}^+(n = 3)$ formation and various states of CO^+ with some evidence of vibrational excitation. Of these, the $\text{He}^+(n = 2) + \text{CO}^+[\text{X } ^2\Sigma^+]$ product channel (with an energy defect $\Delta E = -0.42\text{ eV}$) is dominant and this accounts for about 40% of the total cross section at 1 keV amu^{-1} .

1. Introduction

There is strong current interest in slow electron capture collision processes between ionic components of the solar wind and cometary atmospheres (cf Cravens 1997) in which CO is a primary constituent. Slow He^{2+} ions form an important part of the solar wind (cf Johnson 1990) and, in this work, we have used the well established technique of translational energy spectroscopy (TES) to study one-electron capture in collisions of $0.2\text{--}1\text{ keV amu}^{-1}$ He^{2+} ions with CO. Within the limitations of the available energy resolution, the TES technique allows the main excited product channels resulting from non-dissociative electron capture to be identified and their relative importance established. In addition, the role of dissociative electron capture processes can be assessed quantitatively. The effectiveness of this approach was shown in previous TES studies in this laboratory (Hodgkinson *et al* 1995) of one-electron capture in slow $\text{He}^{2+}\text{--H}_2$ collisions and we have used the same basic apparatus in this paper.

An early study of He^{2+} -CO collisions in this laboratory (Graham *et al* 1974) in the range 3.2–28 keV amu⁻¹, employed a simple mass spectrometric technique to analyse the slow ionic products arising from both electron capture and ionization. Although these measurements were unable to provide any data on individual collision channels, they did indicate that dissociative processes become dominant at the lowest impact energies considered. In subsequent work in this laboratory, Shah and Gilbody (1990) used time-of-flight analysis and coincidence counting of the slow ionic products in 6.7–65 keV amu⁻¹ He^{2+} -CO collisions. They were able to obtain cross sections for the formation of non-dissociative and dissociative products arising from specific one-electron capture and ionization processes but no information on the final excited states. At the relatively high impact energies used in their work, non-dissociative electron capture was dominant. More recently Folkerts *et al* (1997) have also used a time-of-flight coincidence counting technique to study the fragmentation and kinetic energy release spectra during the dissociation of CO by 4 keV amu⁻¹ He^{2+} ions. In addition, total cross sections for one-electron capture by He^{2+} ions in CO in the range 0.3–1500 eV amu⁻¹ have been measured by Ishii *et al* (1999) and Okuno (2001) has provided additional information about these data. In this paper we have made use of these total cross sections to normalize the measured relative excited and dissociative product yields identified by TES and, in this way, obtained cross sections for the main observed collision channels.

2. Experimental approach

The present TES studies are based on measuring the difference ΔT between the kinetic energy T_1 of the primary He^{2+} ion and the kinetic energy T_2 of the He^+ product ion in states n, l arising from specific channels in



where $\text{CO}^+(\Sigma)$ includes all final bound or dissociative states and ΔE is the energy defect for each individual product channel. The measured energy change $\Delta T = (T_2 - T_1) = \Delta E - \Delta K$, where ΔK is a small target recoil correction. Provided that $\Delta E/T_1$ and the scattering is confined to small angles (McCullough *et al* 1984), $\Delta T \approx \Delta E$. Thus, measurement of the yields $\text{He}^+(n, l)$ in the observed energy change spectra allows the relative cross sections for specific product channels characterized by energy defects ΔE to be determined.

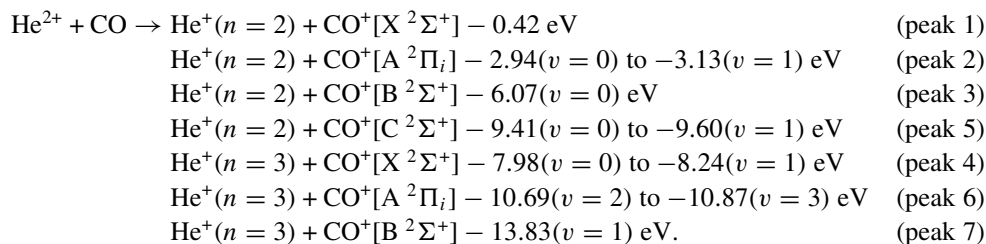
The translational energy spectrometer and the measurement procedure have been described in our previous work (cf Hodgkinson *et al* 1995 and references therein) and only the main features need to be summarized here. A $^4\text{He}^{2+}$ primary ion beam, obtained from an ECR ion source and momentum analysed with a resolution high enough to discriminate against any H_2^+ impurity ions, was focused and decelerated to 120 eV before passage through two hemispherical electrostatic analysers which reduced the energy spread to below 1 eV FWHM. After further focusing and acceleration to the required final energy within the range 0.2–1 keV amu⁻¹, the beam was then passed through the target gas cell into which CO flowed at a constant rate. The fast forward-scattered He^+ products of one-electron capture (within a mean half angle of 0.5°) emerging from the cell were then energy-analysed by a third hemispherical analyser and then counted as pulses by a computer controlled position-sensitive detector. As in previous work, great care was taken to ensure that the CO gas pressure was at all times low enough to ensure single-collision conditions.

The energy scale on our measured energy change spectra was calibrated by reference to our previous data for He^{2+} -O₂ collisions (McCullough *et al* 1992) when O₂ was substituted for CO in the target cell. The energy defects ΔE for individual product channels were then

obtained by reference to the data on the electronic and vibrational states of CO⁺ (Carlsson-Göthe 1990, Carlsson-Göthe *et al* 1991, Karlsson 2001) based on both calculations and on measurements using photoelectron spectroscopy.

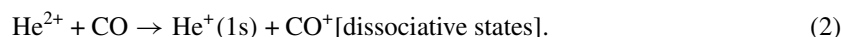
3. Results and discussion

Figure 1 shows our observed energy change spectra for one-electron capture by 0.2, 0.25, 0.5 and 1.0 keV amu⁻¹ He²⁺ ions in CO. The main endothermic collision channels corresponding to peaks 1–7 in these spectra can be identified with non-dissociative electron capture as follows:



The contributions to these peaks can be seen to decrease in relative importance with increasing endothermicity. Peaks 1, 2, 3 and 5 involve electron capture into the $n = 2$ states of He⁺ but peak 1 associated with CO⁺[X ²Σ⁺] formation and an energy defect of only −0.42 eV can be seen to make by far the greatest contribution to non-dissociative electron capture. Peaks 4, 6 and 7, which correspond to channels with quite large energy defects and involve electron capture into the $n = 3$ states of He⁺, can be seen to become large enough to be identifiable only at the highest energies considered.

The broad peaks in figure 1 observed for energy changes ΔE between 0 and about 20 eV contain all the channels involving dissociative one-electron capture into the He⁺(1s) state through the process



As suggested by Graham *et al* (1974), these fragmentation processes leading to C⁺ or O⁺ formation may proceed via a two-step process involving first the creation of a vacancy in the lowest molecular orbital of CO⁺ followed by predissociation of the ion. While it is evident that the present measurements are unable to resolve individual contributions to the broad peak observed in figure 1, the sum of dissociative electron capture processes can be seen to rapidly increase in relative importance as the energy decreases. The previous work of Shah and Gilbody (1990) in this laboratory did show that, at 10.7 keV amu⁻¹, the lowest energy they considered, dissociative one-electron capture channels account for a total cross section of $5.05 \times 10^{-16} \text{ cm}^2$ compared with a total of $11.9 \times 10^{-16} \text{ cm}^2$ from non-dissociative one-electron capture. They also find that dissociative electron capture is dominated by the process:



although the sum of the following channels leading to C²⁺ formation:



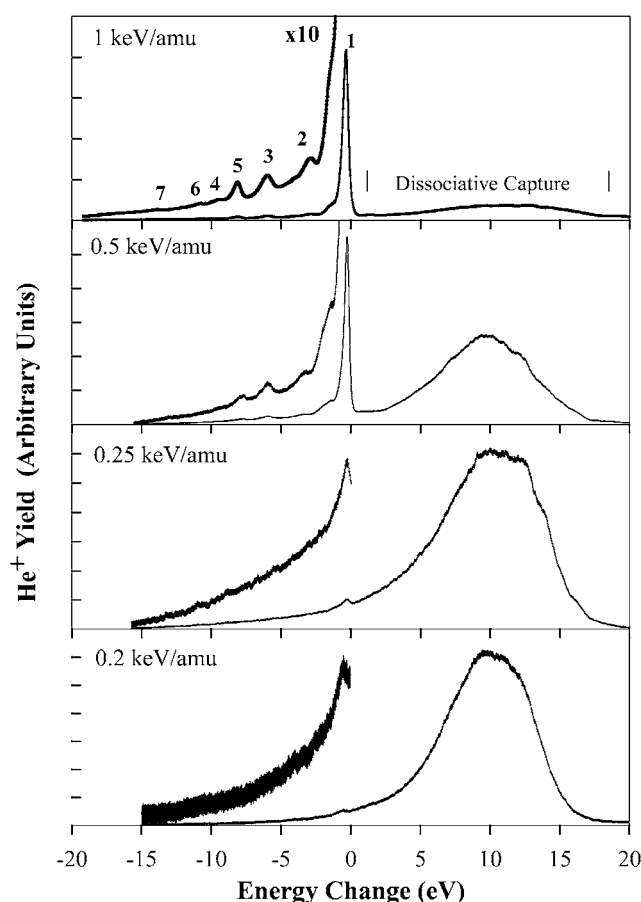


Figure 1. Energy change spectra for one-electron capture by 0.2, 0.25, 0.5 and 1 keV amu^{-1} He^{2+} ions in CO. Peaks 1–7 correspond to non-dissociative electron capture channels (see text), while the broad peaks for $\Delta E > 0$ contain channels involving dissociative electron capture.

was also shown to amount to more than 50% of process (3) at 10.7 keV amu^{-1} . The measurements of Folkerts *et al* (1997) at 4 keV amu^{-1} have confirmed the importance of these and other channels. At 4 keV amu^{-1} they find that fragmentation channels account for about 46% of the total products of the ionization of CO. Detailed information on the energy defects of channels such as (3) and (4) are unavailable. However, if such channels do contribute significantly to (2) at the lower energies considered in this work, they will presumably form part of the single broad unresolved peak in figure 1 ascribed to dissociative electron capture processes.

We have analysed the energy change spectra shown in figure 1 using a peak-fitting procedure (as in our previous work) to obtain the relative contributions to electron capture for each of the channels observed. Cross sections for each of these channels were then obtained by normalizing these yields to the total cross sections (in some cases at interpolated energies) for one-electron capture measured by Ishii *et al* (1999). Our derived cross sections are summarized in table 1 together with the fractional contribution provided by each channel to one-electron capture.

Table 1. Electron capture cross sections (10^{-16} cm^2) for specified product channels derived from the measured energy change spectra. Percentages of the total capture for each channel are also shown. Cross sections σ_{tot} (some at interpolated energies), to which our individual measured values are normalized, were measured by Ishii *et al* (1999). In addition to the specified individual uncertainties, all the present values are subject to an estimated additional uncertainty of $\pm 20\%$ arising from the normalization procedure.

Product channels	Energy (eV amu^{-1})			
	1000	500	250	200
$\text{He}^+(1s) + \text{CO}^+$ (dissociative states)	3.24 ± 0.22 (51.3%)	4.65 ± 0.11 (81.5%)	≥ 4.25 ($\geq 99\%$)	≥ 4.11 ($\geq 99\%$)
$\text{He}^+(n=2) + \text{CO}^+[\text{X } ^2\Sigma^+]$ (peak 1)	2.46 ± 0.22 (38.9%)	0.82 ± 0.10 (14.3%)	≤ 0.04 ($\leq 1\%$)	≤ 0.04 ($\leq 1\%$)
$\text{He}^+(n=2) + \text{CO}^+[\text{A } ^2\Pi_i]$ (peak 2)	0.22 ± 0.02 (3.5%)	0.1 ± 0.01 (1.8%)	—	—
$\text{He}^+(n=2) + \text{CO}^+[\text{B } ^2\Sigma^+]$ (peak 3)	0.17 ± 0.01 (2.7%)	0.07 ± 0.01 (1.2%)	—	—
$\text{He}^+(n=2) + \text{CO}^+[\text{C } ^2\Sigma^+]$ (peak 5)	0.11 ± 0.01 (1.8%)	0.05 ± 0.01 (0.4%)	—	—
$\text{He}^+(n=3) + \text{CO}^+[\text{X } ^2\Sigma^+]$ (peak 4)	—	—	—	—
$\text{He}^+(n=3) + \text{CO}^+[\text{A } ^2\Pi_i]$ (peak 6)	0.11 ± 0.06	0.02 ± 0.01	—	—
$\text{He}^+(n=3) + \text{CO}^+[\text{B } ^2\Sigma^+]$ (peak 7)	(1.8%)	(0.4%)	—	—
$\text{He}^+(n, l) + \text{CO}^+(\Sigma)$ σ_{tot}	6.33	5.70	4.30	4.15

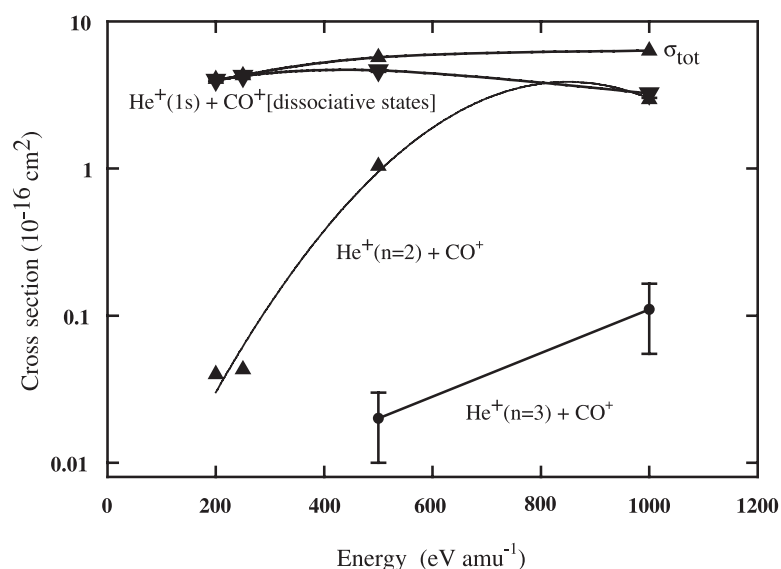


Figure 2. Present cross sections for one-electron capture by He^{2+} ions in CO leading to the product channels $\text{He}^+(n=2) + \text{CO}^+$, $\text{He}^+(n=3) + \text{CO}^+$ or $\text{He}^+(1s) + \text{CO}^+$ [dissociative states]. Total cross sections (σ_{tot}), to which the present data are normalized, are due to Ishii *et al* (1999).

The strong energy dependence of these contributions can clearly be seen. Thus while the main endothermic channel corresponding to peak 1 provides 38.9% of the total charge

transfer at 1 keV amu⁻¹, the corresponding contribution at 0.2 keV amu⁻¹ is only about 1%. In contrast, the contribution from dissociative electron capture increases from 51% at 1 keV to about 99% at 0.2 keV amu⁻¹. The dominant role of dissociative electron capture at the energies considered is further illustrated by the plots in figure 2.

4. Conclusion

The present TES measurements for one-electron capture by 0.2–1 keV amu⁻¹ He²⁺ ions in CO have confirmed the dominant role of processes involving dissociative electron capture. A similar low energy dominance of dissociative electron capture channels was previously observed in our TES studies of He²⁺–H₂ collisions (Hodgkinson *et al* 1995). In this paper, cross sections are shown to be strongly energy dependent with the contribution from exothermic dissociative electron capture channels associated with He⁺(1s) formation accounting for about 99% of the total cross section at the lowest energy considered. For the first time, endothermic excited product channels in non-dissociative one-electron capture have been identified. These lead to either He⁺(*n* = 2) or He⁺(*n* = 3) formation and various states of CO⁺ with some evidence of vibrational excitation. By far the most important of these channels is that leading to He⁺(*n* = 2) + CO⁺[X ²Σ⁺] formation. This accounts for about 40% of the total cross section at 1 keV amu⁻¹, although the contribution falls to about 1% at 0.2 keV amu⁻¹.

Acknowledgments

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