

# Mechanisms of One-Electron Capture by Slow $\text{He}^{2+}$ , $\text{C}^{4+}$ and $\text{O}^{6+}$ Ions in Collisions with $\text{CH}_4$

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Translational energy spectroscopy (TES) has been used to study one-electron capture by  $\text{He}^{2+}$ ,  $\text{C}^{4+}$ , and  $\text{O}^{6+}$  ions in collisions with  $\text{CH}_4$  within the range 200 - 2000 eV amu<sup>-1</sup>. In each case the main collisions mechanisms and product channels have been identified. The measurements reveal significant differences in the way the dissociative and non-dissociative mechanisms contribute to electron capture. However, in all cases, the highly selective nature of the charge transfer process is confirmed in spite of the wide range of energy defects associated with possible product channels.

Keywords: Collisions; Charge transfer

## I. INTRODUCTION

The process of one electron capture in slow exothermic collisions with molecules may be highly selective because of the nature and position of the initial and final potential energy curves describing the molecular systems formed during the collision. Thus, in the general reaction scheme  $X^{q+} + Y_i Z_j \rightarrow X^{(q-1)+}(n, l) + Y_i Z_j^+(\Sigma) \pm \Delta E$  where  $Y_i Z_j^+(\Sigma)$  includes all final bound or dissociative states, only a limited number of product channels leading to excited products  $X^{(q-1)+}(n, l)$  corresponding to energy defects  $\Delta E$  may be involved. Translational energy spectroscopy (TES) can provide a detailed insight into the mechanisms involved in the electron capture process. The main selective capture channels leading to either ground or excited products can be identified (subject to the limitations of the available energy resolution) while the relative importance of the possible dissociative, non-dissociative or autoionizing double capture (ADC) contributions to the total one-electron capture cross section can be determined.

In this work we have studied one-electron  $\text{He}^{2+}$ ,  $\text{C}^{4+}$  and  $\text{O}^{6+}$  ions in collisions with  $\text{CH}_4$  within the range 200 - 2000 eV amu<sup>-1</sup>. These processes are relevant to a better understanding of the interaction of the solar wind with cometary and planetary atmospheres while  $\text{CH}_4$  molecules are also an important building block for more complex molecules. Previous experimental studies are sparse and detailed theoretical studies are difficult. Simple predictions based on Landau-Zener or classical over-barrier approaches have been shown [1] to be of limited value when both dissociative and non-dissociative channels are significant.

## II. EXPERIMENTAL APPROACH

A detailed description of the apparatus and experimental approach has been given in our previous publications (see [2] and references therein) and only a brief summary need be given here. A primary ion beam of well defined energy  $T_1$  is passed through the target gas and the kinetic energy  $T_2$  of the forward scattered  $\text{He}^+$  ions formed as products of single col-

lisions is then measured. The difference in kinetic energy  $\Delta T$  is then given by  $\Delta T = T_2 - T_1 = (\Delta E - \Delta K)$  where  $\Delta K$  is a small recoil correction of the target. Provided the ratio  $\Delta E/T_1 \ll 1$  and the scattering is confined to small angles, the measured change in translational energy  $\Delta T \approx \Delta E$ . The relative importance of collision product channels (characterised by particular values of  $\Delta E$ ) may then be assessed from a careful analysis of the energy change spectra subject to the limitations of the available energy resolution. Energy defects corresponding to specific product channels were identified by reference to the energy level tabulations of Bashkin and Stoner [3] and the photoelectron spectroscopy data of Brundle and Robin [4] and Dujardin *et al* [5] for  $\text{CH}_4$ .

## III. RESULTS AND DISCUSSION

Observed energy change spectra for one-electron capture by  $\text{He}^{2+}$ ,  $\text{C}^{4+}$  and  $\text{O}^{6+}$  ions in  $\text{CH}_4$  are shown in Fig. 1. In the case of  $\text{He}^{2+}$  in  $\text{CH}_4$ , the peak centred on an energy change of about 0.5 eV correlates with the non-dissociative product channel [ $\text{He}^+(n = 2) + \text{CH}_4^+ [^1T_2] + (0.05 - 0.85) \text{ eV}$ ]. The spectra also indicate much smaller non-dissociative contributions from the endothermic product channel [ $\text{He}^+(n = 3) + \text{CH}_4^+ [^1T_2] - (6.7 - 7.5) \text{ eV}$ ]. The broad peak in the spectra corresponding to [ $\text{He}^+(n = 1) + (\text{CH}_4^{2+} \Rightarrow \text{fragmentation}) + (3.3 - 21 \text{ eV})$ ] centred on an energy change of about 12 eV comprises a number of possible transfer ionization product channels involving a variety of fragmentation modes which our TES apparatus is unable to resolve. It increases in relative importance with decreasing energy and provides the main contribution to the total captures at 300 eV amu<sup>-1</sup>.

The energy change spectra for  $\text{C}^{4+}$  ions in  $\text{CH}_4$  are dominated by a well defined peak which corresponds to the non-dissociative product channel [ $\text{C}^{3+}(n = 3) + \text{CH}_4^+ [^1T_2] + (10.7 - 14.2) \text{ eV}$ ]. The spectra also reveal small  $\text{C}^{3+}(n = 4)$  contributions arising through dissociative product channels of large endothermicity.

In the case of  $\text{O}^{6+}$  ions in  $\text{CH}_4$ , the spectra contain a well defined peak corresponding to the non-dissociative  $\text{O}^{5+}(4d)$  product channel [ $\text{O}^{5+}(1s^2 4d) + \text{CH}_4^+ [^1T_2] + (17.1 - 17.9) \text{ eV}$ ].

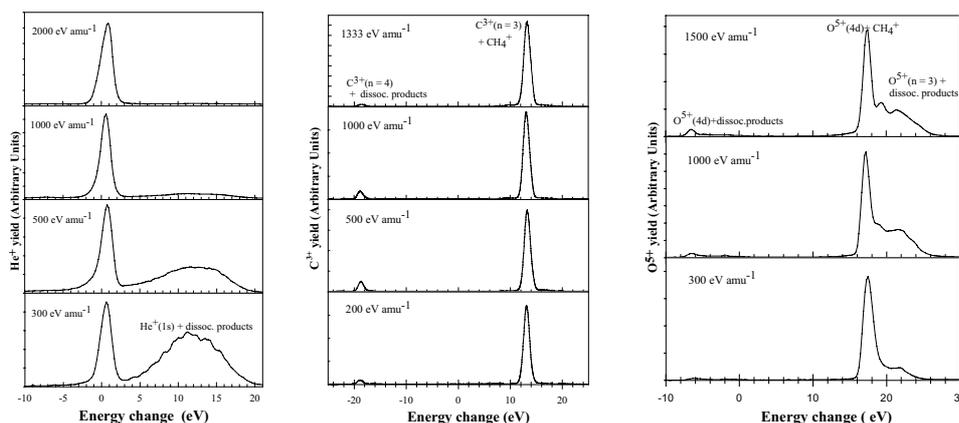


FIG. 1: Energy change spectra for one-electron capture by  $\text{He}^{2+}$ ,  $\text{C}^{4+}$  and  $\text{O}^{6+}$  ions in  $\text{CH}_4$ .

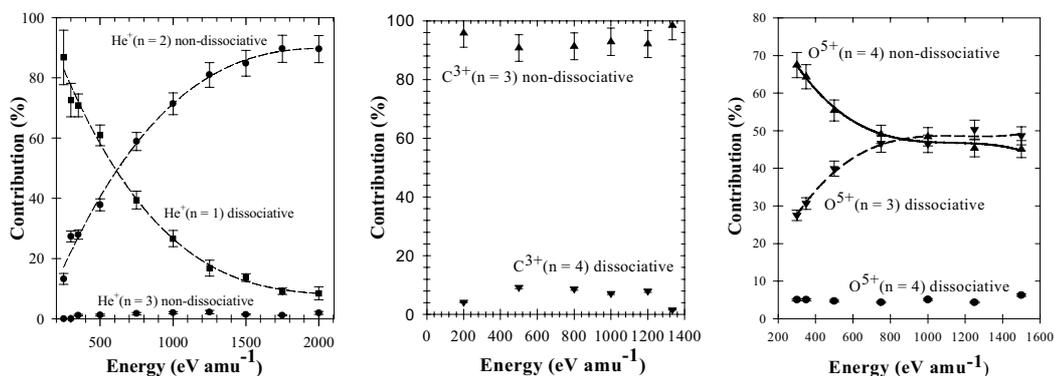


FIG. 2: Relative contributions of the main non-dissociative and dissociative product channels in one-electron capture by  $\text{He}^{2+}$ ,  $\text{C}^{4+}$  and  $\text{O}^{6+}$  ions in  $\text{CH}_4$ .

eV ] which dominates the capture process at low energies. The spectra also show structure corresponding to a series of more highly exothermic  $\text{O}^{5+}(n=3)$  product channels involving the formation of  $\text{CH}_4^{2+}$  which is known to break up into a number of possible fragments [5] which we are unable to resolve. These product channels increase in relative importance with increasing energy. A small endothermic peak in the observed energy change spectra can be ascribed to  $[\text{O}^{5+}(n=4) + (\text{CH}_4^{2+} \Rightarrow \{(\text{CH}^+, \text{H}_2^+, \text{H}); (\text{CH}_2, \text{H}^+, \text{H}^+); (\text{CH}, \text{H}^+, \text{H}_2^+); (\text{C}, \text{H}_2^+, \text{H}_2^+)\} - (4.6 - 8.1) \text{ eV}]$  product channels which, again, can involve many possible fragmentation modes.

The relative contributions to one-electron capture from the main product channels (Fig. 2) show that there are significant differences in the way the dissociative and non-dissociative

mechanisms contribute to electron capture in  $\text{He}^{2+}$ ,  $\text{C}^{4+}$  and  $\text{O}^{6+}$  ions in collisions with  $\text{CH}_4$ . However, in all cases, the highly selective nature of the charge transfer process is confirmed in spite of the wide range of energy defects associated with the possible product channels.

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