

Collision mechanisms in one-electron capture by He^{2+} ions in collisions with hydrocarbons

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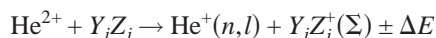
Translational energy spectroscopy has been used to study one-electron capture by 200–2000 eV amu^{-1} He^{2+} ions in CH_4 , C_2H_4 , and C_2H_6 . The main $\text{He}^+(n,l)$ excited product channels have been identified and their relative importance assessed. Although significantly different patterns of behavior are observed, all three cases exhibit the highly selective nature of the electron capture process in spite of the large number of possible nondissociative and dissociative product channels associated with a wide spread in energy defects ΔE .

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I. INTRODUCTION

The process of one-electron capture by He^{2+} ions in slow collisions with simple 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 reaction



where $Y_i Z_j^+(\Sigma)$ includes all final bound or dissociative states, only a limited number of product channels leading to excited products $\text{He}^+(n,l)$ corresponding to energy defects ΔE may be involved. In addition to identifying and assessing the relative importance of the main product channels, the extent to which dissociative and nondissociative collision mechanisms contribute to the selective electron capture process is also of considerable interest in view of the many possible fragmentation modes involving a wide spread in values of ΔE .

In previous work in this laboratory, we have used the well-established technique of translational energy spectroscopy (TES) to provide identification and a quantitative assessment of the main product channels and the collision mechanisms involved in many different processes. Such information is particularly important in cases where reliable theoretical models of the collision have not yet been established. For example, our earlier TES studies of He^{2+} - H_2 collisions in this laboratory [1] provided direct evidence of the great importance of highly selective dissociative excitation mechanisms in the one-electron capture process at low energies. Our subsequent TES studies [2] of one-electron capture by 200–1000 eV amu^{-1} He^{2+} ions in CO have also confirmed the dominant role of processes involving dissociative electron capture at low energies.

In the present work, we have used TES to study one-electron capture by He^{2+} ions in CH_4 , C_2H_4 , and C_2H_6 in the range 200–2000 eV amu^{-1} . There are no detailed theoretical predictions for electron capture collisions with these hydrocarbons, and in our previous work (see, for example, [3]), we have shown that simple predictions based on reaction windows using a Landau-Zener approach are of very limited value when both dissociative and nondissociative channels are important. It is of interest to determine to what extent the general pattern of behavior in these cases is similar to that found in our previous studies of collisions of He^{2+} with H_2 and CO. These processes are also relevant to a better under-

standing of collisions of He^{2+} ions (as an important component of the solar wind) with cometary and planetary atmospheres (see, for example, [4]). In magnetically confined fusion plasmas, these measurements are also relevant to improved modeling of the transport of He^{2+} in edge plasmas containing hydrocarbon impurities arising from the use of carbon composite facing materials [5,6].

II. EXPERIMENTAL APPROACH

A detailed explanation of the apparatus and experimental approach has been given in our previous publications (see [7] and references therein) so that only a brief summary need be given here. In the TES approach, the He^{2+} 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 He^+ ions formed as products of single collisions is then measured. The difference in kinetic energy ΔT is then given by

$$\Delta T = T_2 - T_1 = \Delta E - \Delta K$$

where ΔK 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 (characterized by particular values of ΔE) may then be assessed from a careful analysis of the energy change spectra subject to the limitations of the available energy resolution.

A beam of He^{2+} ions, produced by an all-permanent-magnet 10 GHz ECR ion source, was extracted into an accelerator beamline held at a potential -4 kV. This beam was then momentum analyzed using a 90° double-focusing magnet and passed through two hemispherical electrostatic analyzers. The energy of the emergent beam was then adjusted to that required by passage through a cylindrical electrostatic lens system before entering a voltage-labeled target gas cell. The target gas of interest flowed into the cell at constant rate low enough to ensure single-collision conditions. The forward-scattered He^+ product ions emerging from the cell (within an acceptance angle of $\pm 3^\circ$) were then energy analyzed by a third hemispherical energy analyzer and recorded by a computer-controlled position-sensitive detector. By scanning the retarding voltage, a translational energy spectrum of the product ions could be obtained while maintaining an energy resolution of about 1 eV. An analysis of the posi-

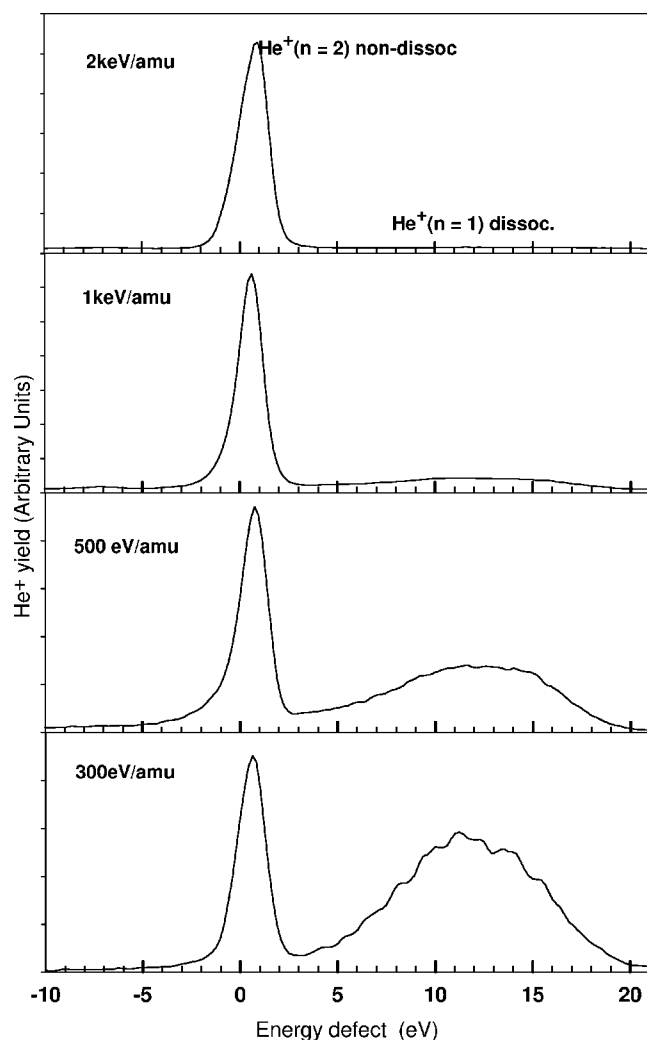


FIG. 1. Energy change spectra for one-electron capture by 300–2000 eV amu⁻¹ He²⁺ ions in CH₄.

tions and magnitudes of the peaks in the observed spectra (using PEAK-FIT software) allowed identification and determination of the relative importance of each product channel. In principle, cross sections for each observed product channel could then be derived by normalizing the sum of the relative yields to total one-electron capture cross sections if available. Energy defects corresponding to specific product channels were identified by reference to photoelectron spectroscopy data and we have assumed that Franck-Condon transitions are valid throughout. For CH₄ we used the data of Brundle and Robin [8], Dujardin *et al.* [9], and Rabalais *et al.* [10], for C₂H₄ the data of Pollard *et al.* [11], and for C₂H₆ the data of Mackie *et al.* [12].

III. RESULTS AND DISCUSSION

Figure 1 shows our observed energy change spectra for one-electron capture by 300–2000 eV amu⁻¹ He²⁺ ions in CH₄. At 2000 eV amu⁻¹, the spectrum is dominated by the peak centered on an energy change of about 0.5 eV which (by reference to photoelectron spectroscopy data) appears to correspond mainly to the nondissociative product channel

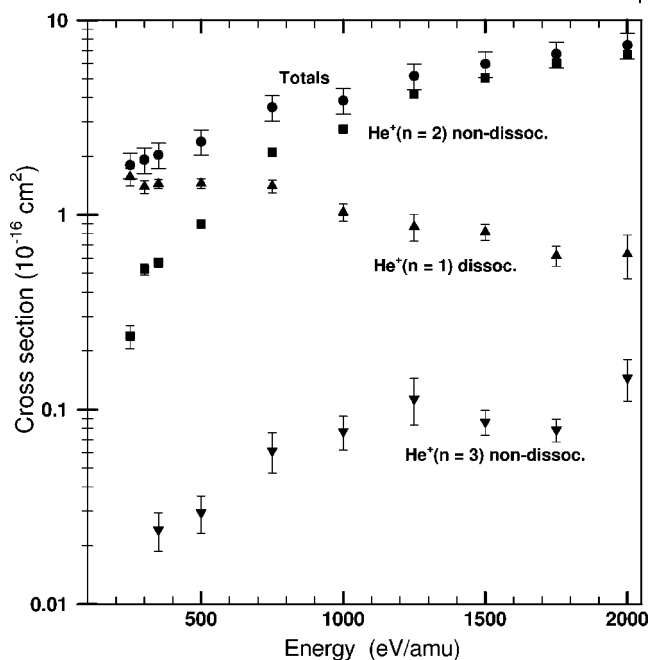
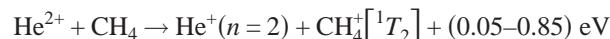
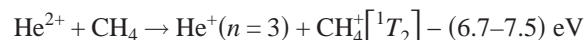


FIG. 2. Measured cross sections for main product states of He⁺ formed in one-electron capture by 300–2000 eV amu⁻¹ He²⁺ ions in CH₄ together with total one-electron capture cross sections.

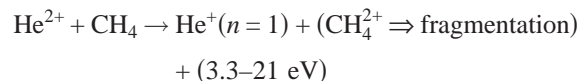


leading to He⁺(*n*=2) formation. The spectra also indicate the presence of a much smaller nondissociative contribution from the endothermic channel,



leading to He⁺(*n*=3) formation.

The spectra in Fig. 1 also show the presence of a broad peak centered on an energy change of about 12 eV. This peak, while small at 2000 eV amu⁻¹, becomes larger with decreasing energy and at 300 eV amu⁻¹, it provides the main contribution to the total captures. This broad peak comprises a number of possible product channels involving a variety of fragmentation modes,



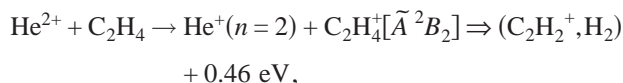
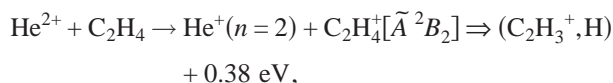
through transfer ionization accompanied by He⁺(*n*=1) formation. The energy resolution available in our TES measurements is insufficient to distinguish between the many possible fragmentation modes. As in our previous studies of one-electron capture by He²⁺ ions, the present spectra provide no evidence of contributions from autoionizing double capture. Overall, in spite of the many different product channels involving a wide range of energy defects Δ*E*, one-electron capture in He²⁺-CH₄ can be seen to be highly selective in the energy range considered with only He⁺ (*n*=1, 2, and 3) formation observable. In Fig. 2, we show cross sections for He⁺ (*n*=1, 2, and 3) formation which have been derived from the measured energy change spectra by normalization to total cross sections for one-electron capture. These

TABLE I. Measured cross sections (in units of 10^{-16} cm 2) for one-electron capture by He^{2+} ions in CH_4 leading to $\text{He}^+(n=1, 2, \text{ and } 3)$ formation compared with total electron capture cross sections.

| Energy (keV amu $^{-1}$) | Totals | $\text{He}^+(n=1)$ | $\text{He}^+(n=2)$ | $\text{He}^+(n=3)$ |
|---------------------------|-----------------|--------------------|--------------------|--------------------|
| 0.25 | 1.80 ± 0.27 | 1.56 ± 0.16 | 0.23 ± 0.03 | |
| 0.30 | 1.91 ± 0.28 | 1.39 ± 0.10 | 0.52 ± 0.03 | |
| 0.35 | 2.03 ± 0.30 | 1.44 ± 0.07 | 0.56 ± 0.03 | 0.02 ± 0.02 |
| 0.50 | 2.37 ± 0.35 | 1.44 ± 0.08 | 0.89 ± 0.04 | 0.03 ± 0.01 |
| 0.75 | 3.56 ± 0.53 | 1.40 ± 0.10 | 2.09 ± 0.10 | 0.06 ± 0.01 |
| 1.00 | 3.86 ± 0.57 | 1.02 ± 0.10 | 2.76 ± 0.13 | 0.07 ± 0.01 |
| 1.25 | 5.16 ± 0.77 | 0.86 ± 0.13 | 4.18 ± 0.21 | 0.11 ± 0.03 |
| 1.50 | 5.95 ± 0.89 | 0.81 ± 0.07 | 5.05 ± 0.25 | 0.08 ± 0.01 |
| 1.75 | 6.71 ± 1.00 | 0.61 ± 0.07 | 6.01 ± 0.30 | 0.07 ± 0.01 |
| 2.00 | 7.43 ± 1.11 | 0.62 ± 0.15 | 6.65 ± 0.33 | 0.14 ± 0.03 |

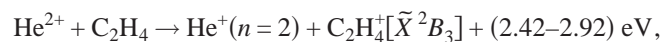
total cross sections were measured using a simple attenuation technique in which we normalized our values to a value measured at 2 keV amu $^{-1}$ by Hoekstra [13]. Strictly, our measured cross sections, which necessarily include contributions from two-electron capture, are an upper limit to the total one-electron capture cross section. All our measured cross sections for one-electron capture by He^{2+} ions in CH_4 are listed in Table I.

Figure 3 shows energy change spectra for one-electron capture by 215–2000 eV amu $^{-1}$ He^{2+} ions in C_2H_4 . At 215 eV amu $^{-1}$ the main peak can be correlated with $\text{He}^+(n=2)$ formation through one of the following possible dissociative channels



which our available energy resolution cannot separately distinguish. This peak can be seen to decrease in relative importance with increasing impact energy.

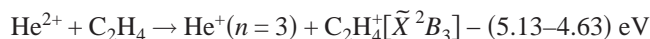
At 2000 eV amu $^{-1}$, the peak centered on an energy change of about 2.6 eV corresponding to the nondissociative channel



leading to $\text{He}^+(n=2)$ formation can be seen to provide the main contribution to the total although there is little evidence of this at our lowest impact energy. Small endothermic contributions from the channel



and, at the higher energies,



are also evident.

There are no measurements of total cross sections for one-electron capture by He^{2+} ions in C_2H_4 in the present energy range so in Fig. 4, we show simply the relative cross sections for excited state formation derived from our energy change spectra. While both dissociative and nondissociative capture both lead to $\text{He}^+(n=2)$ formation, the dissociative capture process is clearly dominant at our lowest impact energy. Our measured relative cross sections for one-electron capture by He^{2+} ions in C_2H_4 are listed in Table II.

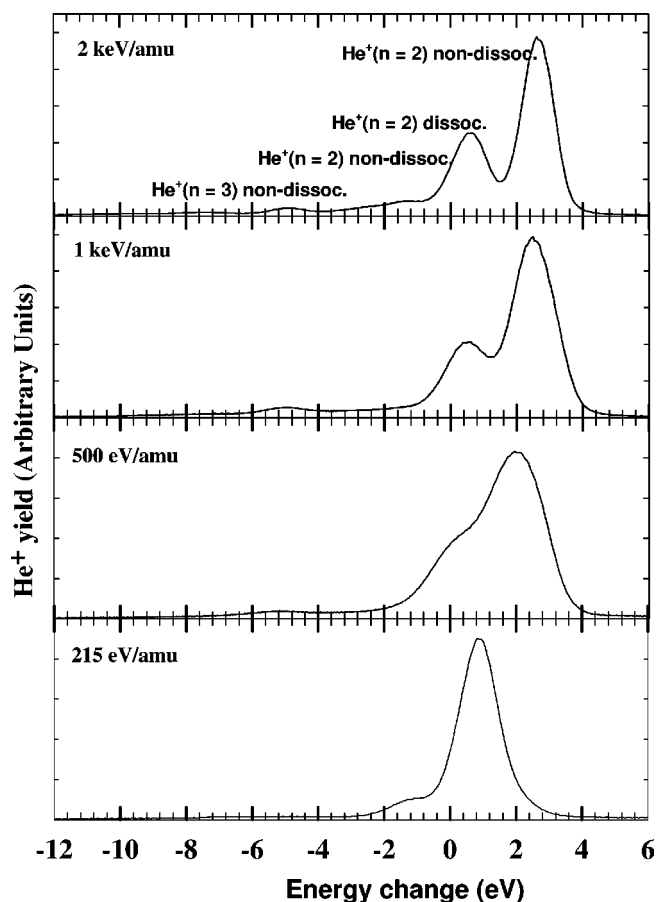


FIG. 3. Energy change spectra for one-electron capture by 215–2000 eV amu $^{-1}$ He^{2+} ions in C_2H_4 .

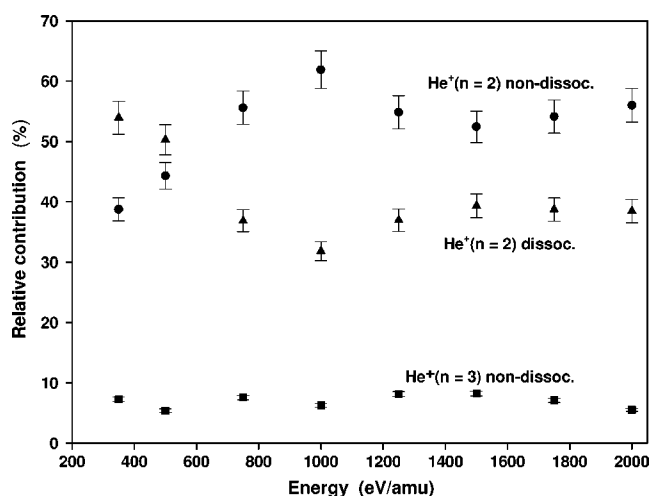
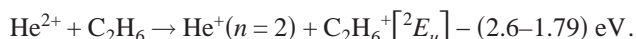
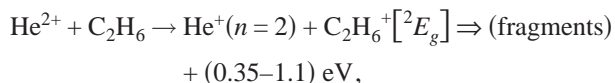
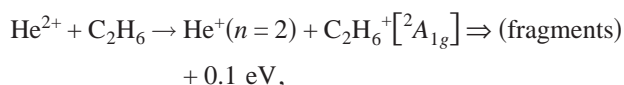


FIG. 4. Measured relative cross sections for main product states of He^+ formed in one-electron capture by 215–2000 eV amu^{-1} He^{2+} ions in C_2H_4 .

Figure 5 shows energy change spectra for 200–2000 eV amu^{-1} He^{2+} ions in C_2H_6 . In this case also, $\text{He}^+(n=2)$ formation is the dominant excited product. At the lowest energy, rather surprisingly, the spectrum is dominated by a single endothermic peak associated with the nondissociative channel



However, at the higher energies the spectra indicate increasing contributions from the channels



in which, according to the photoionization measurements of Mackie *et al.* [12], the main breakup fragment is C_2H_4^+ . At

TABLE II. Measured relative cross sections for one-electron capture by He^{2+} ions in C_2H_4 leading to $\text{He}^+(n=2$ and 3) formation through both dissociative and nondissociative processes expressed as a percentage of the total captures.

| Energy (keV amu^{-1}) | $\text{He}^+(n=2)$ nondissoc. | $\text{He}^+(n=2)$ dissoc. | $\text{He}^+(n=3)$ nondissoc. |
|------------------------------------|----------------------------------|-------------------------------|----------------------------------|
| 0.35 | 38.76 ± 1.93 | 53.94 ± 2.69 | 7.28 ± 0.36 |
| 0.50 | 44.30 ± 2.21 | 50.28 ± 2.51 | 5.40 ± 0.27 |
| 0.75 | 55.58 ± 2.77 | 36.85 ± 1.84 | 7.55 ± 0.37 |
| 1.00 | 61.89 ± 3.09 | 31.82 ± 1.59 | 6.28 ± 0.31 |
| 1.25 | 54.85 ± 2.74 | 36.97 ± 1.84 | 8.17 ± 0.40 |
| 1.50 | 52.43 ± 2.62 | 39.33 ± 1.96 | 8.22 ± 0.41 |
| 1.75 | 54.13 ± 2.70 | 38.74 ± 1.93 | 7.12 ± 0.35 |
| 2.00 | 56.00 ± 2.80 | 38.46 ± 1.92 | 5.52 ± 0.27 |

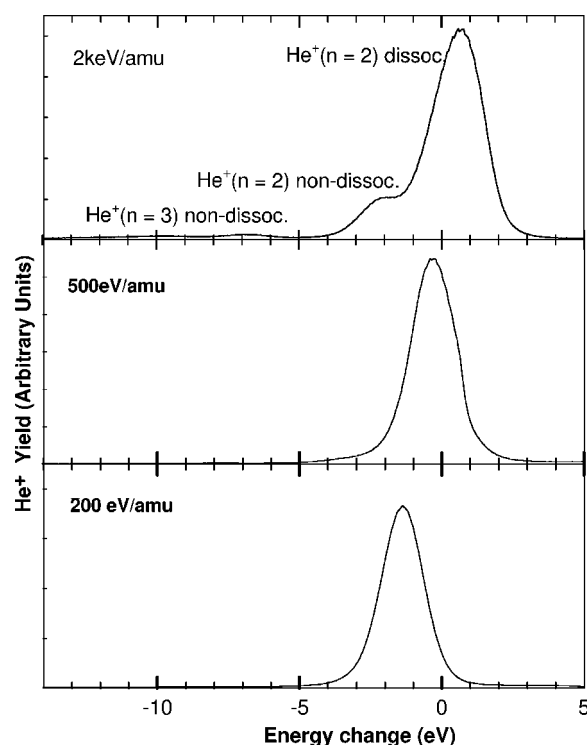
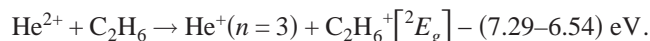


FIG. 5. Energy change spectra for one-electron capture by 200–2000 eV amu^{-1} He^{2+} ions in C_2H_6 .

2000 eV amu^{-1} , there is evidence of a small $\text{He}^+(n=3)$ contribution from the nondissociative endothermic channel



IV. CONCLUSION

Our TES studies of one-electron capture by 200–2000 eV amu^{-1} He^{2+} ions in CH_4 , C_2H_4 , and C_2H_6 have revealed significantly different patterns of behavior. However, in spite of the large number of possible nondissociative and dissociative product channels associated with a wide spread in energy defects ΔE , all three cases confirm the highly selective nature of the electron capture process leading to a very limited number of product states $\text{He}^+(n, l)$. In the case of He^{2+} ions in CH_4 , while nondissociative electron capture into the $\text{He}^+(n=2)$ state is dominant at the highest energies considered, at low energies $\text{He}^+(n=1)$ formation through exothermic channels is dominant. This behavior is similar to our previous observations [2] of one-electron capture by slow He^{2+} ions in CO. The present TES results for He^{2+} ions in C_2H_4 exhibit a very different behavior with $\text{He}^+(n=2)$ as the main product ion arising through a variety of different dissociative and nondissociative channels. Dissociative channels of small exothermicity are found to provide the main charge transfer contribution at the lowest energies. Our TES results for He^{2+} ions in C_2H_6 also identify $\text{He}^+(n=2)$ as the main product ion. This arises from both dissocia-

tive and nondissociative channels but, unlike C_2H_4 , electron capture at low energies takes place predominantly through endothermic capture channels.

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