Gross and partial ionization cross sections in 6-MeV/amu bare-ion collisions with methane

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Abstract. Gross and partial ionization cross sections of CH₄ molecules were measured in 6-MeV/amu H⁺, He²⁺, C⁶⁺, Ne¹⁰⁺ and Ar¹⁸⁺ ion impact. The gross ionization cross sections in light H⁺ and He²⁺ ion impact were found to be proportional to square of projectile charge q in accord with the first Born approximation. As the projectile charge was increased, the dependence became weaker than q². From mass-spectroscopic measurements, the cross sections for CH₄⁺ and CH₃⁺ ion production were also found to obey the q² dependence in low charge ion impact. In high charge ion impact, their dependence, however, became weak compared with q² scaling. The fragmented ions, such as CH₂⁺, CH⁺ and C⁺, showed charge dependence steeper than CH₄⁺ and CH₃⁺ ions. Multiply charged carbon ions up to C⁵⁺ were also observed.

1. Introduction

Ionization cross sections in energetic (>MeV/amu) heavy ion collisions are important for understanding the ionization mechanisms and fundamentals of astrophysics, cosmic ray physics, plasma physics and so forth. We have studied ionization of rare gases, hydrocarbons, and water in collisions of 6-MeV/amu bare-ions by measurements of absolute cross sections and relative intensities of secondary ions. Ionization processes of CH₄ have been studied under impact of various projectiles; 5-4000 keV protons [1], 1 MeV/amu -F²⁺, -Si⁶⁺, -S⁶⁺, -C⁶⁺ and Ar¹⁸⁺ [2], 1-12 MeV protons [3], up to 3keV electrons [4], 50-6000keV protons and antiprotons [5], and 0.5-3.5 MeV proton and electron [6]. However, a limited number of experiments have been reported in collisions of energetic highly-charged ions. This paper discusses gross and partial ionization cross sections of CH₄ in 6-Mev/amu-H⁺, -He²⁺, -C⁶⁺, -Ne¹⁰⁺ and -Ar¹⁸⁺ ion collisions.

2. Experiment

The experiments were carried out at the Middle-Energy beam course of Heavy Ion Medical Accelerator in Chiba (HIMAC) of the National Institute of Radiological Sciences (NIRS) in Japan. The gross (net) ionization cross sections were measured with a condenser-plate assembly, which was
installed in a collision cell. When projectile ions enter the collision cell, recoil ions are produced. These ions are in turn collected with a parallel-plate electrode by the electric field. The cross sections were calculated from the ratios of total recoil ion current $I_r$ to the projectile ion current $I_p$, $(I_r/I_p)$, measured as a function of the target-gas pressure, which were measured with a capacitance manometer. The systematic uncertainties in the gross ionization cross section were estimated to be 8.7%, and the statistical error was within 1.2% [7].

A mass-spectroscopic technique was applied to separate recoiled ions to get partial ionization cross sections. Secondary ions produced in collisions were extracted with an electrostatic lens, mass/charge analyzed by a double focusing sector magnet and finally detected by a Channeltron detector. Detailed description of the experimental setup and procedure have been reported in previous papers [7, 8].

3. Results and discussion

3.1. Gross ionization cross section

The measured gross ionization cross sections are summarized in table 1 and also shown in figure 1. In figure 1, they were plotted as a function of projectile charge $q$.

| $H^+$ | $He^{2+}$ | $C^{6+}$ | $Ne^{10+}$ | $Ar^{18+}$ |
|------|----------|---------|-----------|-----------|
| 0.403| 1.62     | 12.9    | 30.9      | 83.7      |

Table 1. Gross ionization cross sections for CH$_4$ in bare ion impact (in 10$^{-16}$ cm$^2$)

![Cross section (in 10$^{-16}$ cm$^2$)](image)

Figure 1. Projectile charge $q$ dependence of gross ionization cross sections for CH$_4$. Filled circles show the present results. Solid line represents the $q^2$ scaling.

For light projectiles, $H^+$ and $He^{2+}$, the gross ionization cross sections are well on a line showing $q^2$ dependence, as predicted by the first Born approximation. However the $q$ dependence became weaker as the projectile charge became increased, and the cross sections calculated from $q^2$ dependence results in 1.56 times larger than the experimental value in $Ar^{18+}$ impact, indicating a failure of the first Born approximation for high charge projectiles even in the present collision energy of 6 MeV/amu.
3.2. Partial ionization cross sections

In figure 2, mass/charge spectra for CH$_4$ in He$^{2+}$ and Ar$^{18+}$ impact are shown for comparison. They are characterized by several peaks corresponding to fragmented ions as well as the most prominent peak for parent CH$_4^+$ ions. As can be noted in figure 2, the intensity of CH$_4^+$ and CH$_3^+$ peaks are almost the same in He$^{2+}$ and Ar$^{18+}$ projectiles. Differences in peak intensity appear for fragmented ions, such as CH$_2^+$, CH$^+$ and C$^+$. The intensities of these fragmented ions relative to parent CH$_4^+$ ion increased as the projectile charge increased. In Ar$^{18+}$ impact, multiply charged C$_n^+$ (n=2-4) are clearly seen. Production of C$_5^+$ ions was also confirmed in a separate measurement. Partial ionization cross sections were calculated from the relative intensities of peaks in the mass spectra and the gross ionization cross sections in table 1.

![Figure 2. Mass/charge spectra for CH$_4$ in He$^{2+}$ and Ar$^{18+}$ impact.](image1)

![Figure 3. Partial ionization cross sections for CH$_4$ as a function of projectile charge q.](image2)

Figure 3 shows the obtained partial cross sections as a function of projectile charge $q$. The cross sections for CH$_4^+$ and CH$_3^+$ ions are prominent, and they increase with a slope of around 2 in a log-log
plot. This indicates that they are mainly produced at distant collisions, where the first Born approximation would be applicable. In addition, the intensity ratios of fragmented CH$_4^+$ ions to parent CH$_4^+$ ion were almost independent of $q_i$, indicating that these ions are mainly produced through two-step processes as described by the quasiequilibrium theory (QET) by Rosenstock [9]: An excited state molecular CH$_{4}^{i+}$ ion is first produced in the impact, and this ion then de-excites or fragments statistically with a fragmentation pattern which depend only on the internal energy of the excited CH$_{4}^{i+}$ ion and does not depend on the ionization cross section.

On the other hand, fragmented ions, such as CH$_{4}^{i+}$, CH$^{i+}$ and C$^{i+}$, showed charge dependence steeper than the $q^2$ scaling, indicating deviation from the first Born approximation and that other mechanisms, such as multiple ionization and inner-shell ionization, might contribute to their production. Moreover, the intensity ratios of these ions to parent CH$_4^+$ ions were found to change considerably with projectile charge $q_i$, indicating deviation from the quasiequilibrium theory.

4. Summary
Gross ionization cross sections for CH$_4$ molecule in light H$^+$ and He$^{2+}$ ion impact were found to show $q^2$ dependence, as predicted by the first Born approximation. As the projectile charge was increased, the charge dependence became weaker than $q^2$, indicating a failure of the first Born approximation for high charge projectiles such as C$^{6+}$, Ne$^{10+}$, and Ar$^{18+}$ even in the present collision energy of 6 MeV/amu.

The mass/charge spectra of recoil ions showed that the most prominent product was parent CH$_4^+$ ions in all projectiles studied. Highly fragmented CH$_n^{i+}$ (m=0,1 and 2) ions and multiply-charged C$^{i+}$ (i=up to 5) ions were detected with considerable intensities in high charge Ar$^{18+}$ impact. Deviation from quasiequilibrium theory was observed in production of these fragmented ions.

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