CH_4 fragmentation from single and double ionization by proton and electron impact

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Synopsis The fragmentation of methane by impact of electrons and protons was studied experimentally to reveal the mechanisms behind fragmentation by projectiles with opposite charges. Coincidence measurements separate the single from double ionization, in the case of protons, and the DETOF (delayed extraction time-of-flight) technique was used, for electrons, to obtain kinematic signatures of single vacancy (e.g. one-electron–two-vacancy satellite states vs. vertical transitions) production. The substantial differences in the fragmentation cross sections observed between the two projectiles, when several hydrogen bonds are broken, are here attributed essentially to single ionization and interpreted as due to the interference term between the mechanisms of shakeup and the excitation ionization by double impact that appears in the perturbative expansion of the ionization cross section.

In this work we discuss the results of a comprehensive study of ionization followed by fragmentation of the methane molecule by impact of electrons and protons, in the velocity regime of 1 u.a. < v < 12 a.u [1]. The experiments were performed using two different techniques. For electron impact the single ionization cross sections and the energy distribution of the *supra thermal* fragments were measured using the "DETOF" technique [2]. For protons, a time-of-flight spectrometer was used together with the multiple-coincidence technique, which allows us to obtain single and double ionization channels of the molecule [3].

For higher degrees of fragmentation (e.g. production of CH^+ and C^+) a remarkable difference between the electron and proton cross sections are observed for v < 8 a.u. Exclusive coincidence ionization measurements for proton impact showed that these differences are only due to the single ionization, and can be associated to interference between two competing dissociation paths after single ionization (i.e. shake-up and two-step ionization excitation).

For electron impact, an experimental signature of the vertical and satellite contributions to fragmentation was obtained via the distribution of kinetic energy of the ionic fragments produced by electrons with energies between 22 and 800 eV. This finding constitutes a step forward to understand the influence of shake-up and vertical transition paths to fragmentation. The weight of these contributions to the single ionization cross sections were also estimated using a simple model based on the first Born approximation to fit the C^+, CH^+ and CH_2^+ ratios to the CH_4^+ .

For double ionization induced by proton impact, an analysis of our results, together with the data available in the literature [5,6], shows that there are two distinct velocities regions where the interaction is dominated by: a sequential ionization of the molecule, known as TS2 (two-step ionization), which occurs for velocities below 8 a.u., and another, for velocities above 8 a.u., which is dominated by a decay of the post-collisional Auger 1a1 orbital carbon K shell.

References

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