Wake effects in the evolution of fast molecular ions through thin foils

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Abstract

In order to improve the geometrical characterization of molecular ions using the Coulomb Explosion Imaging method, we discuss the need to include wake effects in a full description of the interactions felt by the swift dissociated atomic ions moving through thin foils. Comparison of simulations and experimental results for the structure of diatomic (HD^+) and triatomic (CH_2^+) molecular ions is significantly improved when including the polarization effects induced in the target by the passage of swift ions.
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Different techniques are used to determine experimentally the structure of molecular ions, most of them based in obtaining spectroscopical data. Complementary results are obtained by using the foil-induced Coulomb Explosion Imaging technique [1], which allows the direct measurement of the spatial structure of small molecular ions.

In the Coulomb Explosion Imaging technique single molecular ions (whose structure is to be determined) are accelerated to high energy and then directed to a thin target foil, where they are stripped of their binding electrons in less than $\sim 10^{-16}$ s [2], a much shorter time than the typical ones for molecular vibrations ($\sim 10^{-14}$ s) or rotations ($\sim 10^{-12}$ s). After this rapid dissociation, the

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remaining ionic fragments evolve feeling, among other interactions but mainly, their mutual Coulomb repulsive interaction, which takes place both inside and, mostly, outside the foil. After getting their asymptotic velocities (when the initial potential energy of each molecular ion has been transformed to kinetic energy of its constituents) the dissociated fragments reach detectors specially designed to register and store the relative coordinates and thus the relative velocities of the fragments pertaining to each molecular ion.

On the other hand, the Test Storage Ring at the Max-Planck-Institut für Kernphysik, in Heidelberg, is an experimental setup where molecular ions are stored at a relatively high velocity during a variable time, after which they are extracted and analyzed thus following the spatial configuration at different stages of vibrational relaxation.

During the latter years the Coulomb Explosion Imaging method in combination with the Test Storage Ring has been used to characterize the geometrical structure of a broad variety of di- and tri-atomic molecular ions [3,4]. Using the measured relative velocities of the ionic fragments and assuming that the laws driving the molecular dissociation are known, it is possible to obtain information about the geometrical structure prior to dissociation. For this purpose, the experimental data are compared to the results of a Monte Carlo simulation algorithm, which models the fragmentation process taking into account relevant properties of the experimental setup including the target foil [5].

Besides the dominant Coulomb explosion, there are other effects influencing the motion of the dissociated fragments through the target, namely multiple scattering, charge exchange processes, and the electronic excitations of the target induced by the passage of a swift charged projectile. The latter effects (referred to sometimes as polarization effects) were never fully incorporated in the standard data analysis procedure used to reconstruct the original molecular structure, therefore, early studies [6] were designed to suppress them by restricting the analysis to molecular fragments impinging the target foil with a suitable orientation, a procedure that strongly reduces the statis-

tics and is not applicable for non-planar molecular ions. Another possibility to minimize the polarization effects was to use very thin foils to induce the molecular fragmentation, as could be deduced from the short time ($\sim 10^{-15}$ s) spent by the swift fragments moving inside the target, as compared with the long time in which Coulomb explosion takes place ($\sim 10^{-6}$ s), however this procedure is not convenient either as we will see later on.

In order to quantify the consequences of taking or not into account the polarization effects in the analysis of Coulomb Explosion Imaging experiments, we have studied, both experimentally and theoretically, the spatial evolution of two simple molecular ions (HD^+, linear and CH_2^+, planar) incident with velocity $v = 4.38$ a.u. on a 1.5 µg/cm^2-thick diamond like carbon foil [12,11].

The passage of a swift projectile through the target induces a polarization potential whose results are a self-retarding force (acting on the projectile that creates the electronic perturbation) and a wake force (acting on its neighbour fragments). These forces have been calculated using the dielectric formalism framework [7,8], and their effects on the energy loss and evolution of simple [9] or more complex [10] molecular ions have been evaluated.

To illustrate qualitatively how the intensity and direction of the wake forces between two interacting atomic ions depend on their nature and relative positions, we show in Fig. 1 the wake forces between two protons (left panel) and between a C^{4+} ion and a proton (right panel) moving with $v = 4.38$ a.u. through a diamond like carbon target; the C^{4+} ion has been chosen because it is the dominant charge state arriving to the detector after dissociation of CH_2^+ [11]. As expected the intensity of the wake forces increases with the charge state of the interacting atomic ions and decreases with the distance between them. It is important to notice that in both cases the wake force tends to align the atomic ions along the beam direction, changing their relative positions. This suggests that in order to obtain the geometrical structure of molecular projectiles using the Coulomb Explosion Imaging method, the wake forces between the atomic ions that constitute the molec-
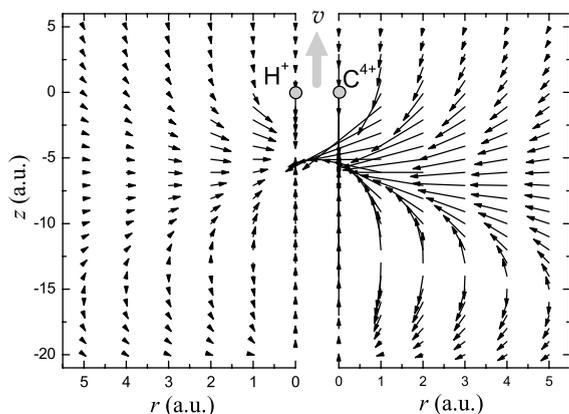
Fig. 1. Wake forces created on a proton by H^+ (left) and C^{4+} (right) moving with velocity $v = 4.38$ a.u. through a diamond like carbon target; z and r are the directions parallel and perpendicular, respectively, to the projectile motion.
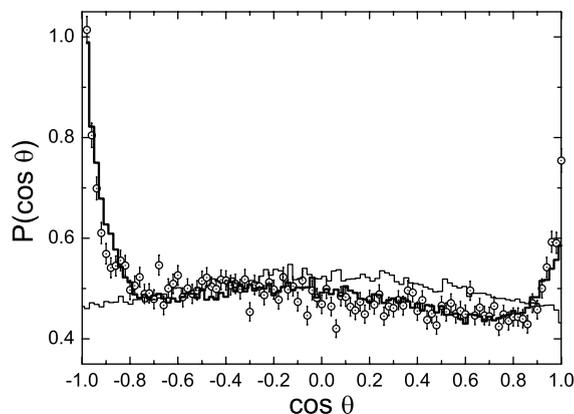

Fig. 2. Probability distribution of the angle θ between the axis joining the fragments dissociated from HD^+ (at their arrival to the detector) and the beam velocity, after impinging with velocity $v = 4.38$ a.u. on a 1.5 μg/cm²-thick diamond like carbon foil. Experimental data [12] are represented by symbols; simulations are represented by histograms: thin line, without polarization effects, and thick line, including polarization effects.

ular projectile should be properly described and considered in the simulations.

We assume that the molecular projectiles extracted from the Test Storage Ring are randomly oriented, so the aligning effect of the wake forces can be observed by analyzing the distributions of the molecular orientation with respect to the beam velocity axis, which will be denoted by z in what follows. In Fig. 2 we show the orientation probability distributions $P(\cos\theta)$ of HD^+ molecules after traversing a diamond like carbon target foil having a thickness of 1.5 μg/cm². θ is the angle of the internuclear axis with respect to the beam axis. The symbols represent the experiments performed using the Coulomb Explosion Imaging technique at the Test Storage Ring; the thin solid line corresponds to the results when the polarization forces are not included in the corresponding simulation, whereas the thick solid line show the results when the simulation takes into account these polarization forces. The experimental data of the orientation probability distribution $P(\cos\theta)$ for HD^+ show a strong alignment effect, demonstrated by the two peaks observed at $P(\cos\theta) \sim -1$ and ~ 1, which corresponds to the H^+ fragment trailing the D^+ partner and viceversa, respectively. This behaviour only is reproduced by the simulation code when considering the polarization forces [12].

For the case of a planar molecular ion other angles have to be used to determine its orientation. The direction of the plane formed by the molecular ion CH_2^+ with respect to the beam velocity is characterized by the angle Θ; values of $\Theta = 0°$ (or $180°$) and $90°$, respectively, indicate that the molecular plane is perpendicular and parallel, respectively, to the beam velocity. The orientation of the nuclei within the molecular plane is given through the angle Ψ; when $\cos\Theta > 0$, the two hydrogen atoms travel in front ($\Psi = 0°$) or behind ($\Psi = 180°$) of the carbon atom. These angles are depicted in Fig. 3.
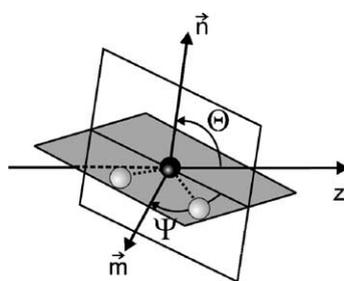

Fig. 3. Angles Θ and Ψ used to characterize the orientation of the CH_2^+ molecular ion fragments reaching the detector. The vector $\vec{n}$ is normal to the molecular plane, and the vector $\vec{m}$ bisects the H–C–H angle.
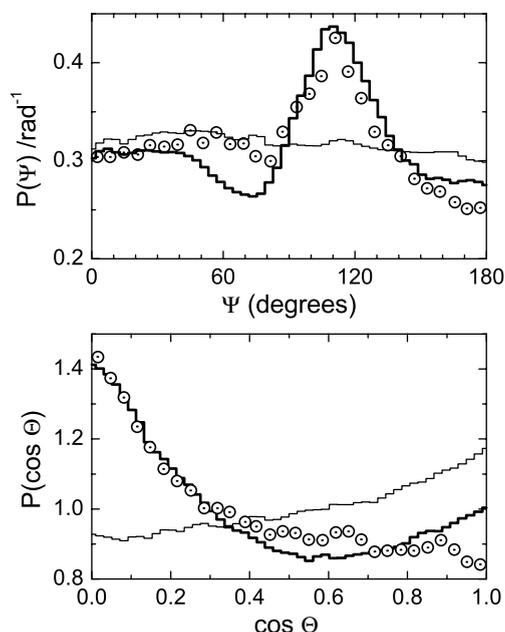
Fig. 4. Probability distributions of the angles Ψ and Θ (defined in Fig. 3 and the text) that characterize the orientation of the dissociated fragments from CH_2^+, at their arrival to the detector after impinging with velocity $v = 4.38$ a.u. on a 1.5 μg/cm^2 diamond like carbon foil. Symbols correspond to experimental data, whereas simulations are represented by histograms: thin line, without polarization effects, and thick line, including polarization effects [11].

Fig. 4 shows the probability distributions $P(\Psi)$ and $P(\cos\Theta)$ that the fragments dissociated from a $v = 4.38$ a.u. CH_2^+ molecular ion after traversing a 1.5 μg/cm^2-thick diamond like carbon foil reach the detector in the relative orientation indicated by these angles. Considering a random orientation of the molecular ions before entering the foil, uniform distributions should be detected if no target effects were operating. But the experimental data (depicted by symbols) are non-uniform, which can be clearly interpreted in terms of polarization effects, as the simulations (histograms) we have done including them (thick solid lines) reproduce fairly well the main experimental features, whereas there is a visible disagreement when these effects are not taken into account.

The results we have discussed show that even for very thin foils wake effects are sizeable, affecting the geometrical evolution of dissociated frag-

ments and should be included in simulations used to analyze Coulomb Explosion Imaging experiments.

In conclusion, the geometrical structure of small molecules can be deduced using the Coulomb Explosion Imaging technique, where fast molecules pass through a thin foil, being stripped of their binding electrons almost instantaneously; following that the charged fragments repel each other, reaching macroscopic distances at detectors installed several meters downstream. From the recorded asymptotic velocities of these fragments, the initial geometry of the molecular ions can be deduced, but in order to obtain a reliable molecular reconstruction, all the relevant interactions, besides the dominant Coulomb repulsion, that take place during the motion of the dissociated fragment should be considered, even the so called minor effects. While most of these effects (such as multiple scattering and charge changing effects) are well known and have been included in the standard data analysis procedure, the dynamic polarization of the target material induced by the passage of the charged fragments (giving rise to the self-retarding and wake forces) has now been added to the simulation code [11]. A good agreement was reached, facilitating now detailed studies also of non-planar molecules, were the effects of target polarization cannot be avoided by selecting suitable orientations of the dissociating molecules.

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