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New source of MeV negative ion and neutral atom beams

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The scenario of “electron-capture and -loss” was recently proposed for the formation of negative ion and neutral atom beams with MeV kinetic energies. However, it does not explain why the formation of negative ions in a liquid spray is much more efficient than with an isolated atom. The role of atomic excited states in the charge-exchange processes is considered, and it is shown that it cannot account for the observed phenomena. The processes are more complex than the single electron-capture and -loss approach. It is suggested that the shell effects in the electronic structure of the projectile ion and/or target atoms may influence the capture/loss probabilities. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4935234>]

I. INTRODUCTION

The possibility of generating negative and neutral atoms with MeV energies would be of very high interest for a number of applications in science and technology. Negative ions play a major role in several areas of physics and chemistry, in high current tandem accelerators, ion beam microscopy, and lithography. Beams of neutral atoms are essential in fusion experiments where they are used for additional heating of the fusion plasma. The typical procedure to generate neutral atom beams is to produce positive and negative ions in a suitable source, to accelerate them, and then to neutralise them. However, the efficiency of such a process is very low.

Additionally, there is a strong fundamental interest in negative ions: here in the screening of the nucleus, the inter-electronic interactions become relatively more important than the electron–nuclear interactions. In the interplay of these attractive and repulsive interactions, the electron correlation plays an important role.

Recently, in laser-plasma interaction experiments energetic O^- ion emission was observed from a water spray target,¹ and on the basis of the experimental findings the electron capture and loss mechanisms were proposed to explain the observations. The proposed “electron capture and loss” scenario suggested that independently from the location of the positive ion source, the capture or loss of an electron occurs when a fast positive ion passes through the spray. Follow-up experiments in which the ion source was separated from the spray (i.e., the positive ions were first accelerated

from a foil target and then propagated through a cold spray) provided an additional confirmation of the above mentioned “electron capture and loss” scenario.²

A better understanding of electron capture and loss processes, however, requires a more thorough discussion. The conditions of equilibrium-charge-state distributions in Ref. 2, which were established in the beam during its propagation through the spray, are not sufficient to infer the absolute cross-sections for the processes, although certain ratios and trends for the cross-sections may be deduced. For example, a strong enhancement of low charge state densities may indicate a contribution of multiple electron capture or loss processes. The distortions of the distribution in specific energy regions may originate from the shell effects in the electronic structure of the projectile ion and/or target atoms. Additionally, the excitation of the atoms in the spray prior to a charge-changing collision may influence the probabilities of electron capture and loss in subsequent collisions. Unfortunately, the physics of charge exchange processes in clusters at high projectile energies is not known and only very scarce data about the cross sections exist in the literature. This does not allow predicting relative fractions of ions in different states, and new experimental data are needed. Additionally, the cross sections are very complex quantities and it is rather difficult to provide an adequate quantitative description³ of all the processes involved. However, a sufficiently accurate direct experimental measurement of charge distributions may define most of the parameters included in theory. For instance, we evaluated the hypothesis that the excitation of atoms by fast electrons and x-rays emitted from the laser plasma source might affect the charge exchange rate.

In fact, the efficient electron transfer from Rydberg excited clusters to projectile ions within a sheath around the laser focus was suggested recently^{4,5} to explain observations of negative Ar ions in a similar experimental context. Similar

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to the conditions of the work reported in Ref. 1, the cluster medium used in Refs. 4 and 5 is highly collisional, and accelerated ions may undergo a number of collisional ionisation and recombination processes. It is not possible to detect from the experimental data the exact location where the charge state of ion has been changed. In contrast, in the approach we reported in Ref. 2, complexities related to the acceleration processes itself (e.g., temporal, spatial, and characteristics of electrons and ions within the laser plasma source) do not need to be considered and the condition of the target atoms can be controlled precisely. The electron capture and loss mechanisms can thus be studied separately and any influence of excited atoms can be determined.

II. MEASUREMENTS AND DISCUSSION

The experimental arrangement is presented in Fig. 1. The ion beam was accelerated from 5 μm polymer CHO or Ti foil targets by irradiating them with 40 fs, 1 J Ti:sapphire laser pulses at an intensity of about $5 \times 10^{19} \text{ W/cm}^2$. A well characterised water,⁶ or ethanol⁷ spray, positioned at distances of 22–380 mm from the foil target, allowed us to study the ion beam–spray interaction. The Thomson spectrometer enables absolute measurements of both positive and negative ions, and neutral atoms, in a single shot.^{8,9}

Particles with opposite charges are deflected in opposite directions in the parallel magnetic and electric fields of the spectrometer thus creating oppositely directed spectral traces on the detector screen. The position of the particle on the spectral trace depends on its energy: the higher is the ion energy, the less it is deviated from the “zero point,” where undeflected emission (x-rays and neutral atoms) hits the detector. Typically, a magnetic field of about 0.27 T and an electric field of 2–4 kV/cm were applied; the ions were spatially selected by a 200 μm pinhole at the entrance of the spectrometer.

The interaction region of the spray, controlled by a skimmer, has a thickness of up to 2 mm. The spray consists of spherical droplets with a diameter of 150 nm in the case of water (H_2O) or 180 nm in the case of ethanol ($\text{C}_2\text{H}_5\text{OH}$) sprays.

FIG. 1. Experimental setup for studies of electron capture and loss processes of fast positive ions in a liquid spray. Laser accelerated fast positive ions from the foil target propagate through a cold spray. The double ended arrows indicate that the spray position between target foil and spectrometer could be adjusted over a broad range of distances.

The average molecular densities in the spray are 2×10^{18} and $\sim 10^{19} \text{ cm}^{-3}$ for the water and ethanol sprays, respectively.

In order to elucidate to which extent the excitation of atoms in the spray may affect the charge exchange rate, we compared the negative ion and neutral atom yield when the spray was placed at distances of 22 mm and 380 mm from the laser-plasma ion source while maintaining the position of the detector fixed. An increase of the distance between the laser-plasma ion source and the spray by a factor of 17 reduces the intensity of all plasma emissions, which may excite the atoms in the spray (x-rays, fast electrons, or optical emission) by a factor ~ 300 . Consequently, one would expect a substantial reduction of excited atoms in the spray.

The measurements showed that the ion yield is independent of the ion source to spray distance. Typical CCD pictures of ion spectra from the phosphorous screen of the MCP and the corresponding energy distributions are shown in Fig. 2. Similar proton energy distributions and same cut-off energies were measured without spray and after the passage through the spray (panels (c) and (d)). This is to be expected as the liquid spray is transparent for protons with energies down to a few hundred keV (the proton energy loss in the water spray is less than 50 keV/mm). Thus, the proton signal can serve as a reference to verify that in different laser shots similar ion beams were produced, which is fundamental for the comparison of ion spectra obtained with and without the spray. The similar proton energy distributions confirm the identical interaction conditions in these two laser shots, and therefore one can infer that initially identical carbon ion distributions were produced in the two events.

A similar amount of neutral and negative ions was measured for both 22 mm and 380 mm distances between the ion source and the spray. The measured energy distribution of ion charge states also was unchanged for the different spray positions. The fluxes of positive and negative carbon ions are comparable, with a very high brightness higher than

FIG. 2. Thomson-parabola traces of carbon ion and proton spectra (a) accelerated from the Ti foil target and (b) accelerated from Ti foil target and propagated through the water spray. From the foil target, only positive carbon ions with the charge states 1+ up to 4+ and H^+ are visible. The negative carbon ions are measured only after the spray. The bright circle in the centre is formed by neutrals and energetic photons, which are undeflected by the E/B fields in the spectrometer. Corresponding energy distributions of ions are shown in panels (c) and (d).

10^{10} particles per MeV per steradian. We conclude that in our conditions the role of excited atoms in the electron capture or loss processes forming negative ions and neutral atoms is negligible. This conclusion is also valid for the conditions of the experiments described by Rajeev *et al.*^{4,5} as their ensembles of clusters have a similar average molecular density and thickness as our target.

III. SUMMARY

The phenomena of controlling the charge state and distribution of an energetic positive ion beam by using a liquid spray with remarkable stable and reproducible characteristics^{6,7} opens unique possibilities for efficient and compact sources of energetic negative and neutral atom species of a broad variety.

Our experimental results, obtained in a series of campaigns, appear to exclude the role of atomic excited states in the charge exchange processes in clusters, which has been suggested by other authors.^{4,5} We therefore conclude that in our conditions the role of excited atoms in the electron capture or loss processes forming negative ions and neutral atoms is negligible.

It is not yet understood, however, why the interaction of a fast projectile ion with a cluster media is much more efficient than with an isolated atom. Screening of nuclear charge and inter-electron interactions may become more important in a dense system, thus affecting the interplay between attractive and repulsive forces. Although we do not have a clear explanation and theoretical framework for the process, the results suggest that the ion interaction with the medium is more complex than the single electron capture and loss processes, and that the shell effects in the electronic structure of the projectile ion and/or target atoms may influence the probabilities.

Additional studies are required to further elucidate the negative ion and neutral atom formation scenario. Special attention has to be paid to precise measurements of the neutral atom characteristics and to detailed quantum mechanical calculations of the cross sections of electron capture and loss in dense multi-atom configurations. However, experimental results so far indicate a sufficiently high probability of such processes for colliding particle energies up to hundreds of keV/nucleon.

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