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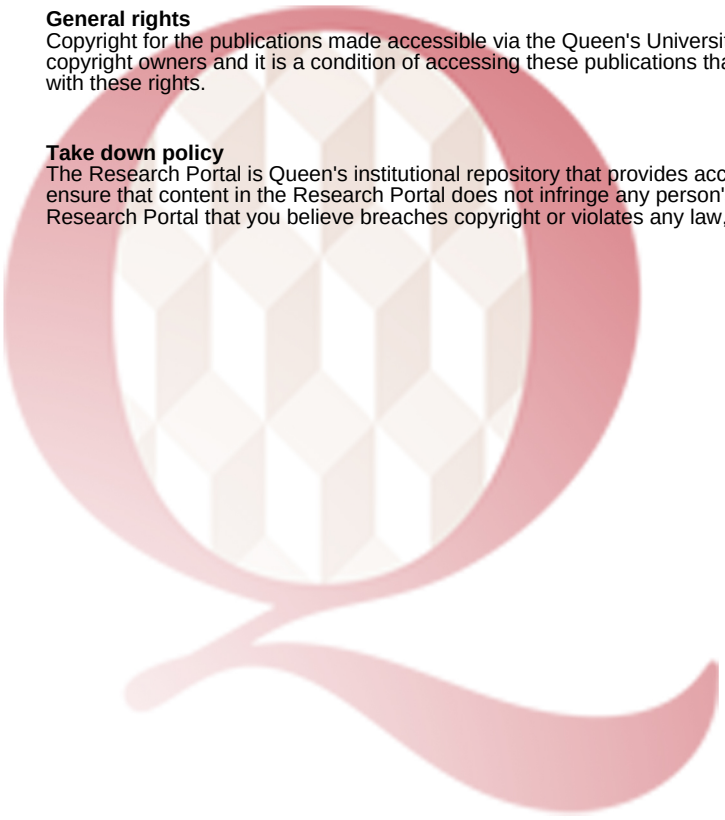

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Energetic beams of negative and neutral hydrogen from intense laser plasma interaction

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We present observations of intense beams of energetic negative hydrogen ions and fast neutral hydrogen atoms in intense (5×10^{19} W/cm^2) laser plasma interaction experiments, which were quantified in numerical calculations. Generation of negative ions and neutral atoms is ascribed to the processes of electron capture and loss by a laser accelerated positive ion in the collisions with a cloud of droplets. A comparison with a numerical model of charge exchange processes provides information on the cross section of the electron capture in the high energy domain. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4850456]

The method of production of fast neutrals and negative ions is rather general and can be applied for production of energetic negative ions and neutral atoms of other species and for measurements of the cross sections of the charge exchange processes.

High power lasers can be used to accelerate ions to energies of tens of MeV.[1] The acceleration of positive ions through a number of laser-driven processes is a fairly well understood phenomenon. There have also been, in the past, a limited number of observations of negative ions accelerated from laser plasma interactions.[2,3] In those experiments, the mechanism of negative ion acceleration could not be determined clearly but the origins of negative and positive ions were considered to be accelerated independently, through separate processes. In contrast, recent experiments have shown that the formation of negative ions is strongly correlated to the accelerated positive ions. A mechanism, where the negative ions and neutral atoms of MeV energies are formed during the collision of positive ions with the atoms at rest in a medium, was proposed in Ref. 4. Up to 10^9 negative ions per srad in 5% energy bandwidth are produced, and the estimated brightness of the negative ion source exceeds 10^8 A · cm^{-2} sr^{-1}. The electron capture and loss processes have higher cross sections than for inelastic collisions when the fast ion velocity is comparable with the orbital velocity of an electron in the atom at rest. The angular distribution of the negative ions resulting from this process is very narrow, ~1° in Ref. 5. The ion that captures or loses an electron propagates essentially in the same direction and with the same energy as before the collision. This scenario explains fairly well the experimental findings in Ref. 4, provides a consistent explanation of earlier results[2,3] and also predicts the existence of fast neutral atom beams, supported by indirect experimental evidence. Accelerated neutral atoms of argon with energies of a few keV per nucleon from laser interaction with argon clusters have been reported in Ref. 6, where the observations were ascribed to the same charge exchange phenomena.

These findings can be attractive for a number of applications.[4,7,8] Compared to standard negative ion sources, which are large scale, complicated devices,[9,10] a laser driven source can be very compact, and with an average power limited only by the laser performance,[6] properties which could be advantageous for applications in industry and fusion research.

The double-charge-exchange method was the one employed in early 1960s to produce negative ions.[11,12] The beam of positive ions (usually H^+ or D^+), accelerated to several keV, was converted into negative ions after passing through alkali metals vapours. However, the efficiency of such sources was very low: only a small fraction of positive ions collected two electrons and became negatively charged.

In this paper, we present the direct observation of beams of energetic negative hydrogen ions and fast neutral hydrogen atoms, generated during intense laser plasma interactions. The use of laser accelerated ions and of a spray of water or ethanol droplets results in an increase of the number of measured particles by 6 orders of magnitude as compared to earlier observations of negative hydrogen,[3] where, in order to obtain a detectable signal, one had to accumulate the ions from more than 10^4 laser shots. The choice of the molecules in the droplets and optimized focusing conditions allowed us to increase more than 100 times the number of negative ions measured in each laser shot. A comparison with a theoretical model allowed us to identify the relevant processes and to evaluate their cross sections, some of which are not known. The scenario of formation and acceleration of negative and neutral hydrogen beams proposed in Ref. 4 is confirmed and

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explained with a simple theoretical model that allows an estimation of electron capture cross section.

The experimental setup is similar to the one presented in Ref. 4. A fully characterized spray of water (H_2O) with droplet diameter of 150 nm[13] and ethanol (C_2H_5OH) with 180 nm[14] droplets was produced with a solenoid valve having an average molecular densities of $n_0 = 2 \times 10^{18}$ and $\sim 10^{19}\,\mathrm{cm}^{-3}$, respectively. A 40 fs, 1 J Ti:sapphire laser pulse was focused to a diameter $d = 4\,\mu$m at an intensity of $\sim 5 \times 10^{19}\,\mathrm{W/cm}^2$ inside of a spray of thickness $2l = 2$ mm. The absolute spectra of positive and negative ions were measured with a Thomson spectrometer in a single laser shot.[13,14] A similar set-up had been employed in the work reported in Ref. 15, where protons with the energies up to a few MeV were measured.

The spectra of H^+ and H^- along with the C^+ and C^- accelerated from an ethanol spray in the laser propagation direction are shown in Fig. 1. Along the parabolic spectral trace, ions with higher energy lay close to the so called "zero point" where undeflected emissions (i.e., x-rays and neutrals) hit the detector. In the attempt to achieve the highest possible yield of H^-, we optimized the interaction temporally and spatially: (i) by adjusting the delay between the spray and the laser pulse, and (ii) by adjusting the laser focus position inside the spray in the plane at fixed distance from the nozzle exit. The highest yield of H^- was achieved when the laser irradiated a high density region of the spray; this requires both temporal and spatial scans in order to identify the most favourable conditions (see Figs. 3 and 4 in Ref. 13). The H^- yield does not depend on the distance of the focal spot from the nozzle, because in this case the spray's areal density remains approximately constant: $n_0 l = \mathrm{const}$ (see Figs. 2 and 3 in Ref. 13) and therefore the charge exchange probability: $P = \sigma n_0 l$, where σ is the cross section of interaction, is not changed.

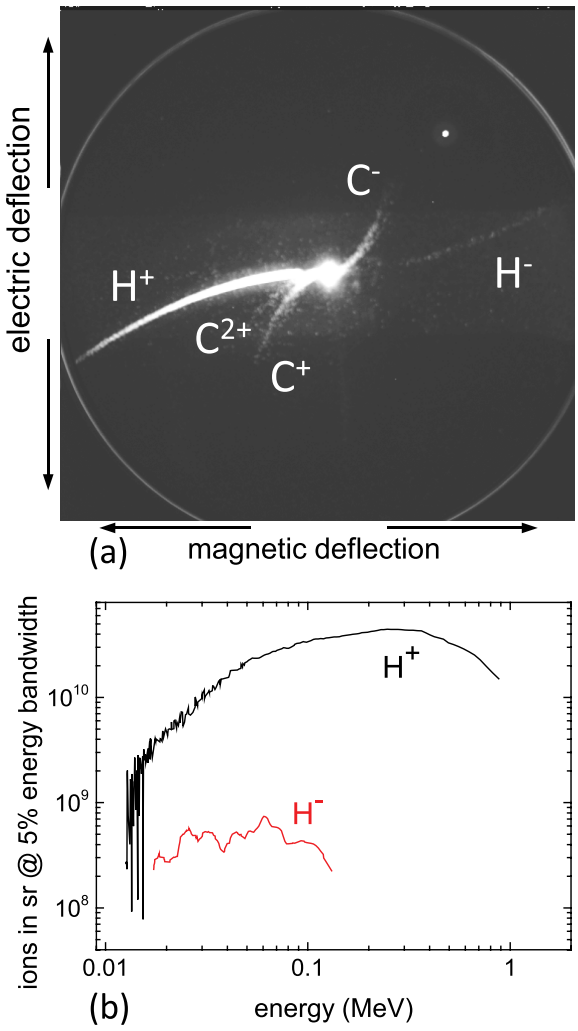

FIG. 1. Thomson parabola measurements of ion production from the ethanol spray in the direction of laser propagation: (a) raw image of H^+ and H^-, and carbon ion tracks; (b) corresponding spectra of H^+ and H^- (black and red lines, respectively). The tracks of carbon ions in (a) are not clearly shown and the "zero-point" is rather extended due to a reduced resolution of the spectrometer as the main goal was to quantify the H^- ions.

In the ethanol spray, the maximum energy of H^+ was $\sim$0.9 MeV, while for H^- it was $\sim$0.14 MeV. In the water spray, the maximum energy of H^- was similar to the case of the ethanol spray but with a 4 times smaller number of particles. This is broadly consistent with the average density of the ethanol spray being $\sim$5 times higher than the water spray.[14]

These observations justify the choice of two liquids, water, and ethanol, differing in their molecular densities by a factor of five, but having a similar ratio of hydrogen to "other constituent atoms" as 2:1. These atoms have similar charge-exchange[7,14] and elastic collision cross sections for fast protons — the stopping power of 100 keV protons in both liquids is $\sim 100\,\mathrm{keV \cdot cm^2/mg}$.[16] Hence, the main difference between the two media was the fact that the mean free path length of protons in ethanol spray is five times smaller as compared to water, which allowed us to verify the role of charge-exchange processes.

The fast neutrals have been measured with the plastic CR-39 nuclear track detector (chemical formula $C_{12}H_{18}O_7$).[17] An energetic particle impinging in the plastic leaves a damage track along its propagation path, which can be made visible by etching. To retrieve the incident particle number, the CR-39 plate was etched in 6 N NaOH solution and the resulting pits were counted using an optical microscope.

To make sure that the interaction conditions were favorable for negative ion generation, a symmetric setup configuration was used similar to Ref. 18, taking the advantage of symmetric ion emission from the spray. Two lateral, symmetrically located Thomson spectrometers were employed; one used a MCP to record "on line" ion spectra, while a CR-39 plate was used on the other spectrometer.

The image of the "zero point" or undeflected emission detected on a CR-39 plate through a 1 mm pinhole is shown in Fig. 2(a). The zoomed picture in panel (b) shows that the "zero-point" consists of pits with two distinct sizes: $\sim$4 and $\sim 2\,\mu$m. They correspond to the pits measured in oxygen and hydrogen spectral traces on the CR-39. The ratio of hydrogen to oxygen neutral atoms was counted to be equal to 2.

The energy of neutrals was measured by the time of flight (TOF) technique using the same "symmetric" setup configuration:[18] In one arm, an electron multiplier tube (EMT) collected the fast neutrals in a 3×10^{-5} sr solid angle, (188 ± 0.5) cm far from the ion source, while the reference spectra were measured with a spectrometer on the opposite side arm. A transverse electric field, 100 cm in front of the EMT, deflected the charged particles away from the detector. The resolution of the TOF setup was limited by the space available in the target area.

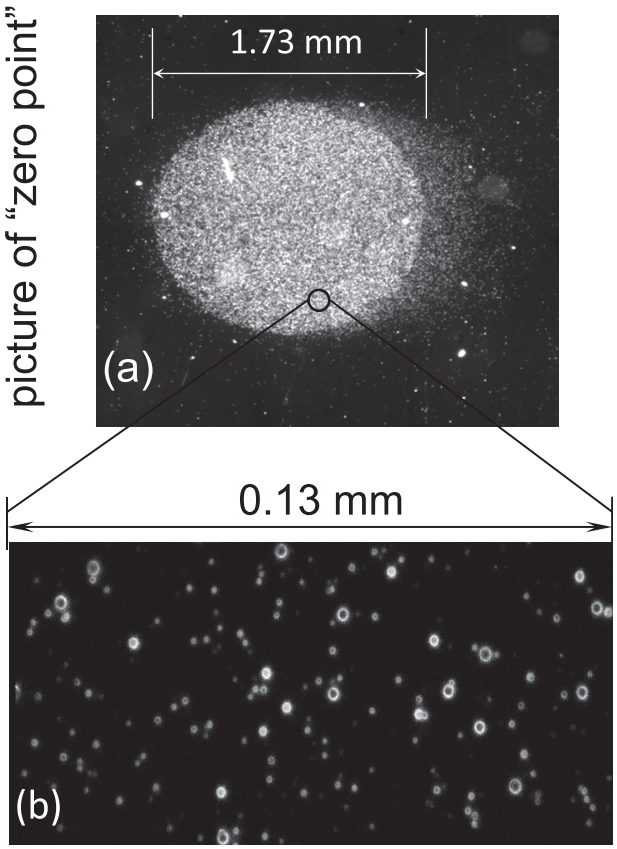

FIG. 2. Neutral atom detection on CR-39: (a) Picture of "zero point" detected on CR-39. (b) Zoomed view of "zero-point" consisting of hydrogen and oxygen atoms, distinguishable by their size.

According to the scenario discussed above, where the fast negative ions and neutrals are formed from the energetic positive ions, we correlated the energy of neutrals and ions on a one-to-one basis. For the recorded spectrum of H^- in the energy range from 140 to 20 keV, the TOF signal of neutral hydrogen was expected to vary from 0.36 to 0.96 μs. The EMT show a clear TOF signal between 0.5 and 1.6 μs (cf. Fig. 3), which corresponds to neutral hydrogen with 7–80 keV energies.

In order to achieve a more quantitative understanding of the processes leading to the formation of the neutral and negative beams, the processes of charge exchange undergone by H^+ ions propagating across the spray were described in the

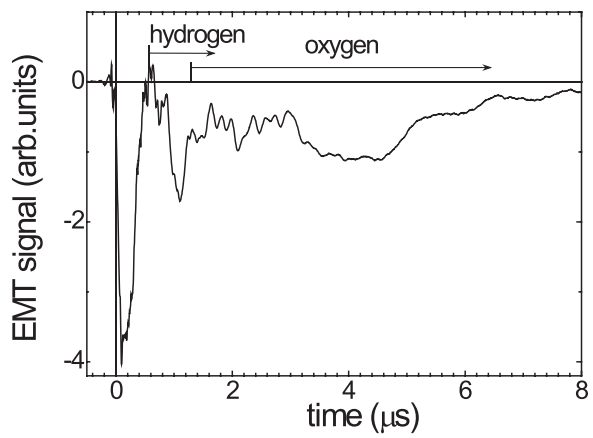

FIG. 3. TOF signal of energetic neutral particles: hydrogen and oxygen detected by the EMT.

approximation of one electron capture and loss: $A^{(q)} \rightarrow A^{(q\pm1)}$ with the cross-section $\sigma_{q,q\pm1}$, where q is the charge of the fast ion before collision: $q = 1, 0, -1$. The populations of the ion species with densities $N_q(\varepsilon,z)$ were modeled by the transport equations

$$\frac{1}{n_0}\frac{dN_q}{dz} = N_{q+1}\sigma_{q+1,q} + N_{q-1}\sigma_{q-1,q} - N_q\sigma_{q,q-1} - N_q\sigma_{q,q+1}, \tag{1}$$

where ε is the particle energy, z is the distance from the source, and n_0 is the average density of the spray. Processes of double charge exchange, elastic scattering, and energy loss were neglected as the corresponding cross sections are much smaller. Since in the charge exchange collisions the angular diagram is very narrow,[5] the particle energy ε is a parameter in Eq. (1). It is assumed that a point-like source at $z = 0$ (as $d \ll l$) emits protons with a given energy distribution: $N_1(\varepsilon,0)$, and $N_0(\varepsilon,0) = N_{-1}(\varepsilon,0) = 0$. There is no charge exchange at $z > l$.

The cross sections of electron capture and loss are known for projectiles with energies up to a few tens of keV/amu[16,19] and, essentially, for hydrogen targets. There is much less data available for heavier atoms, e.g., carbon or oxygen. Here, we have focused on water spray as the behaviour of the water medium is better known, although some cross sections are missing also for this case. The cross sections used in our model for all the processes are obtained in an independent atom approximation. In other words, the charge exchange cross sections of a projectile proton with a water molecule are obtained by adding the corresponding cross sections for the hydrogen and oxygen atoms with the weighting ratio 2:1. In the cases, where the value for the oxygen cross section could not be found, the value for hydrogen was used for oxygen as well. The corresponding cross sections are shown in Fig. 4(a) for the water molecules. The same cross sections were used for the analysis of the results with the ethanol spray, but the atomic density was taken to be 5 times larger than for water.

The spectrum analysis is limited to the interval from 20 to 140 keV where the H^- ions have been measured (Fig. 1). Figure 4(b) shows the interpolated measured distributions of H^- and H^+ together with the calculated distribution of neutral atoms. The proton spectrum at the source is also shown for further comparison.

The solution to Eq. (1) with these cross sections for the projectile H^+ in the energy range from 10 to 100 keV shows that a thickness of 1 mm is sufficient for establishing equilibrium charge states distribution in the ethanol spray. As the saturation level of N_{-1} is much lower than the one for N_0 in first approximation, one can restrict the analysis to H^+ and neutral hydrogen.

Moreover, the cross section for production of negative ions is at least one order of magnitude smaller than the others. Thus, neglecting as a first step the negative ions, the stationary solution of Eq. (1) at $q = 1$ can be presented in the form

$$N_0 = \frac{\sigma_{1,0}}{\sigma_{0,1}} N_1. \tag{2}$$

This relation allows us to find N_0 from the measured spectrum of protons and the known cross section shown in

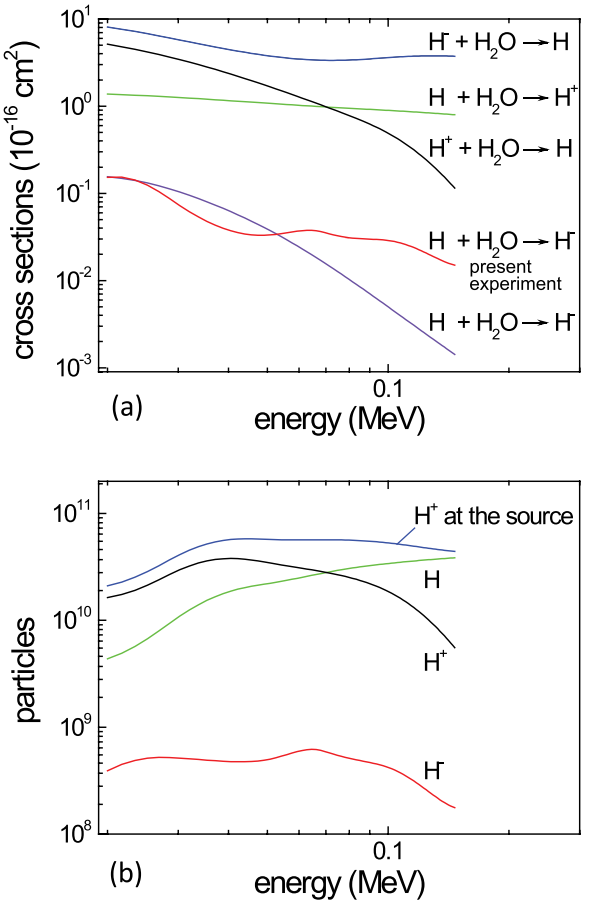

FIG. 4. (a) Electron capture and loss cross sections per atom obtained from the literature[6,19] (blue, black, green, and violet lines) and the calculated cross section of the negative ion production ($\sigma_{0,-1}$, red line) which reproduces the experimental spectrum of H^-. (b) Interpolated measured distributions of H^- (red line) and H^+ (black line) together with calculated distributions of neutral atoms (green line). The reconstructed spectrum of protons at the source $H^+(0) = (N_0 + N_1)$ (blue line).

Fig. 4(b) with the green line. As the total number of particles $(N_0 + N_1)$ is conserved, it gives us the spectrum of particles at the source. This is shown as a blue line in Fig. 4(b).

Knowing the proton spectrum at the source, we consider now the negative ion production by solving the full system (1). The spectrum of negative ions obtained with the known cross sections deviates from the measured one for high energies. The most probable reason is that the charge exchange cross sections on oxygen atoms are not known in this energy range. So, we consider another approach. By using the measured spectrum of negative ions, we reconstructed the cross section for electron capture, which is shown in Fig. 4(a) as a red line.

The cross section for negative ion production agrees within a factor of 2 with the data available in the literature for energies lower than 50 keV, while there is a clear difference for higher energies. At the moment, it is difficult to provide a definite explanation for this. It might be that the unknown electron capture cross section of $H + O$ or $H + C$ is $3 - 4$ times higher than that in the $H + H$ collision. This hypothesis can be checked with detailed numerical calculations.

Another possible hypothesis[6] relates the enhanced attachment cross section to the excited states of neutral atoms. Indeed, x-ray emission and fast electrons from the plasma created in the laser focus may produce excited atoms. However, it is not evident that this would apply to the collisions at high energies where the interaction time is much smaller than the period of excited electron rotation in the atom. Further calculations are needed to evaluate the possible contribution of this effect.

It is also necessary to measure the spectrum of neutral atoms and to increase the precision of the negative ion measurements. The measured flux of H^- ions is relatively low. This can cause an error in the measurement of ion beam intensity (20% accuracy) and in the estimate of ion cut-off energy. (In the present experiment the cut-off energies are similar for both sprays in contrast to[14] where higher ion energies (~MeV range) have been observed from ethanol spray). Further studies are required to elucidate the negative hydrogen acceleration scenario, where special attention has to be paid to neutral particle characterization, to provide a precise estimate of the effective length of collisions, and to the control of density fluctuations in the sprays.

In summary, the experiments have shown that the interaction of high-intensity and high-contrast laser pulses with liquid sprays is an efficient source of fast negative ions and neutral hydrogen atoms. The electron capture and loss processes taking place when energetic positive ion interact with cold spray atoms can explain most of the observed features. Model calculations reproduce the experimental results relating to the formation of negative hydrogen at energies below 50 keV. Further investigations are necessary to elucidate the physics of the charge transfer phenomena at higher energies.

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