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Interaction of H_2^+ molecular beam with thin layer graphene foils*

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The interaction of MeV H_2^+ molecular ions with thin layer graphene and graphite foils was studied by using a high-resolution electrostatic analyzer. A large number of fragment protons were observed at zero degree (along the beam direction) when the H_2^+ beam was passing through the monolayer graphene foil, which indicates that the electron of the H_2^+ molecular ions can be stripped easily even by the monolayer graphene foil. More trailing than leading protons were found in the energy spectrum, which means significant wake effect was observed in the monolayer graphene foil. The ratio of the numbers of trailing protons over leading protons first increased with the thickness for the much thinner graphene foils, and then decreased with the thickness for the much thicker graphite foils, which indicates that the bending effect of the wake field on the trailing proton varied with the foil thickness.

Keywords: H_2^+ molecular ions, graphene, electron stripping, wake effect

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1. Introduction

The study of the interaction of molecular ions with solids has been an active topic for the past several decades, which provides a basic technique and gives possibilities to precisely study the properties of materials and incident particles. This interesting field was pioneered by Poizat and Remillieux who used swift H_2^+ molecular ions to collide with carbon targets.[1] When the molecular ions are passing through the targets, electrons can be stripped off quickly and the left positively charged ions are isolated by a repulsive force and experience a so-called Coulomb explosion process.[2] And also, the ions can continuously experience a retarding force from the target molecules and lose energy in the material. When the kinetic energy of the incident ions is several MeV/u, the dominant energy loss process will be excitation or ionization of the target molecules.[3]

Unlike individual ions, the fragmented ions of molecular ions/cluster are proximal and can experience pronounced correlate effects during interaction with material, which is called as the vicinage effect in the cluster research field. When two or more ions, whose internuclear distance are very short, are moving together inside a material, they would interact with the same target molecule simultaneously and cause an interference effect, then the phenomenon is very different from the case of isolated ions.[4–6] For example, Brandt *et al.* measured the energy loss of H_2^+ and H_3^+ molecular ions in a carbon foil, and found that the energy loss of the molecular ions was larger than that of two/three isolated protons and confirmed the existence of vicinage effects for the first time.[4] The vicinage effect is conventionally expressed by the stopping ratio $R = S/\sum_i S_i$, where S is the stopping power of the molecular ions/cluster and S_i is the stopping power of fragment i as an independent ion. Subsequently, extensive measurements have been performed with H_2^+. According to a review work by Arista,[7] the stopping ratios for H_2^+ in carbon had been measured to within 0.8 and 1.4, which varied with the beam energy and target thickness.

Besides the vicinage effect, the wake effect is studied widely when swift molecular ions interact with solids.[8–12] The molecules of the target can be polarized by the incident ions and form one induced field which is called the wake field. Normally the wake field is very weak and only exists within a very short period, which is not easy to detect. But for the interaction of molecular ions with a target where several vicinage ions are moving together, the wake field induced by the leading ion would have strong effect on the trailing ions, which renders the wake field detectable.[13,14] For dicluster ions, the wake effect induced by the leading particle could create a deep potential well in the target, which might capture the trailing particle and bent its direction.[13] The effect has been confirmed experimentally by Susuki *et al.*[9] by using 0.2–0.5 MeV/u H_2^+ molecular ions colliding with graphite foils, where a detector

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with acceptance angle of 0.04° was used to measure the ratio between the numbers of aligned proton pairs and isolated protons. And a much slower decreasing trend of the ratio versus foil thickness was found, which agreed with the theoretical calculation well only when the wake effect was considered.[10]

The physics of the interaction between the H_2^+ molecular ions and thin foils has been studied for a long period, and normally much thicker foils with several tens of atomic layers were used.[8,15–19] It is believed that the electron is stripped off quickly after H_2^+ hit the foil, which is supposed to happen within several atomic layers, or shorter than a femtosecond.[2,4,20–22] And there are some theoretical models which assume that the electrons of molecular ions are stripped off once they contact the first atomic layer of the thin foil.[5,9,23–26] In Ref. [27], the Rutherford backscattering energy spectrum of carbon fragments which were generated by a 1.847 MeV C_2^+ cluster beam interacting with a silicon crystal target was used to measure the time when the Coulomb explosion just happened. It was reported that the electron stripping time was around 0.1 fs for T_{100} spatial period, 0.12 fs for T_{211} spatial period, and 0.2 fs for T_{111} spatial period. However, it is still not easy to directly clarify where the electron is stripped off. Since the position of electron stripping determines the charge state of the molecular ion cluster in its subsequent motion and the main physical processes including the stopping force of the medium and the energy loss per unit length, the process of electron stripping is more important. In our work, very thin free carbon foils whose thickness ranged from monolayer (graphene) to several dozen atomic layers were used to study the collision with MeV H_2^+ molecular ions. Here, the experimental results are reported to answer the questions of how many atomic layers are needed to strip off the electron of H_2^+, and how is the wake effect varying for really thin targets.

2. Experimental setup

The experiment was performed by using an electrostatic accelerator, and the schematic of the device is shown in Fig. 1.[28] A high-frequency ion source and a 2.5-MV electrostatic accelerator were used to generate the H_2^+ molecular ion beam with the energy of a few MeV/u. A 90° vertical bend magnet and a 20° horizontal bend magnet were used to deflect the beam and screen out the required H_2^+ molecular ions from others. A two-dimensional (2D) XY electrostatic deflector, an adjustable XY slit, and a fixed aperture were used to guide the H_2^+ beam to the target, to control the beam size, and to further remove the protons and H particles in the beam. The error of the original beam parallelism was less than $\pm 4.5 \times 10^{-5}$. And the vacuum of the system was better than 5×10^{-4} Pa during operation.

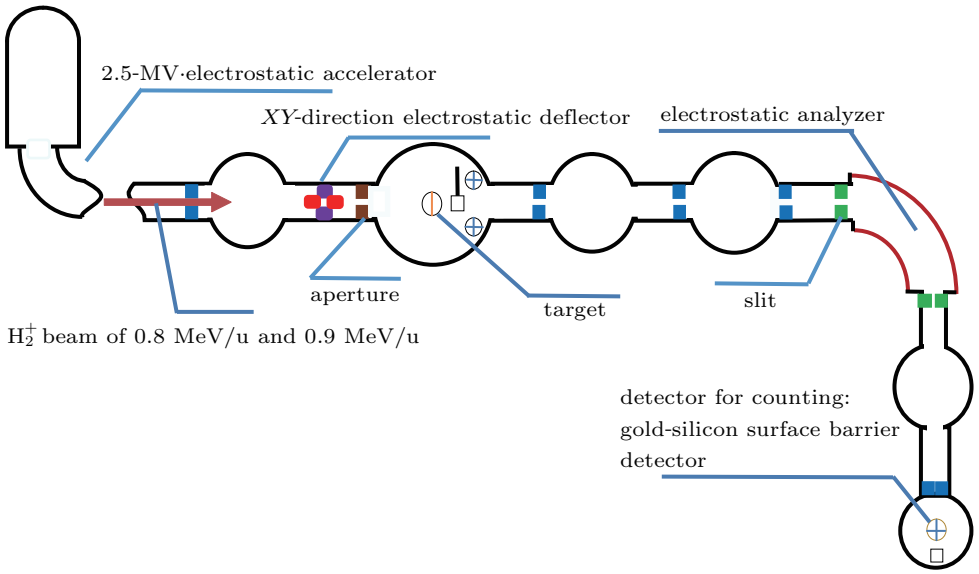


Fig. 1. Schematic of the experiment setup.

In the target chamber, a movable target ladder with 9 slots was used to mount the foils. The graphene type foils with thicknesses of 1, 2, 3–5, and 6–8 layers, and graphite type foils with thicknesses of 7–10 nm and 10–15 nm were used. For convenience, the thickness of mono layer graphene was supposed to be 0.34 nm, and the thicknesses of the graphene type foils were simply expressed as 0.34 nm, 0.68 nm, 1.36 ± 0.34 nm, and 2.38 ± 0.34 nm, respectively. While the thicknesses of the graphite foils were expressed as 8.5 ± 1.5 nm and 12.5 ± 2.5 nm. The graphene sheets were provided by TED PELLA Co., Ltd,[29] which were supported by 2000-mesh copper TEM grids with circular holes of 6.5 μm and pitches of 12.5 μm. The 2000-mesh TEM grid was supported by a 1 mm×2 mm slotted grid to give the required stiffness. The total usable area of the free-standing graphene sheets was approximately 30% due to the pitches, unavoidable folds, and

wrinkles. The graphite type foils were supported by 300-mesh copper TEM grids with circular holes of 58 μm and pitches of 25 μm, which were supplied by Beijing Zhongjingkeyi Technology Co., Ltd.[30]

The H_2^+ molecular ions were adjusted to collide with the foil perpendicularly, and might pass through the graphene/graphite foil or fracture into two protons (secondary beam), and only particles along the beam direction (0°) were tested. The thickness of the TEM grid was 25 μm, which was thick enough to stop all H_2^+ molecular ions and had no influence to the experiment.

An energy measurement system consisting of a 90° electrostatic analyzer (EA, with radius 800 mm) and a gold-silicon surface barrier detector (ORTEC, CA-014-025-1000) was used to test the energy of the fragment protons and transmitted H_2^+, which was placed at 3 m downstream the target. The resolution of the energy measurement system was better than 0.04% for 1.8 MeV H_2^+ ions. An adjustable slit was mounted at the entrance of the EA and only particles with the divergence angle of less than 0.75×10^{-4} rad could be accepted. The energy calibration coefficient was 0.045 keV/V for the EA.[28]

3. Experimental results

Since the internuclear axes of the H_2^+ molecular ions were randomly oriented, the direction of the Coulomb explosion would be random. And most protons would obtain one extra transversal velocity after the Coulomb explosion and would deviate from the beam direction. Only these proton pairs, which were generated from the H_2^+ molecular ions whose internuclear axes were aligned parallel to the beam direction, could keep the original direction and be detected by the EA. The energy of these protons was measured by the EA, and the energy spectra are shown in Figs. 2 and 3. Figures 2(a) and 2(c) are the energy spectra of the fragment protons from 0.8-MeV/u H_2^+ in monolayer graphene and in 10–15 nm graphite, respectively. To ensure that the TEM grid had no influence to the experiment, the energy spectra of the fragment protons from 0.8-MeV/u H_2^+ passing through one empty 2000-mesh copper TEM grid and one empty 300-mesh copper TEM grid are shown in Figs. 2(b) and 2(d), respectively. Figures 3(a) and 3(c) are the energy spectra of the fragment protons from 0.9-MeV/u H_2^+ in monolayer graphene and in 10–15 nm graphite, respectively. And the energy spectra of the fragment protons from 0.9-MeV/u H_2^+ passing through one empty 2000-mesh copper TEM grid and one empty 300-mesh copper TEM grid are shown in Figs. 3(b) and 3(d), respectively. It was found that the peak height of monolayer graphene was 3500 for 0.8-MeV/u H_2^+ and 35000 for 0.9-MeV/u H_2^+, and the peak height of the 10–15 nm graphite was 12000 for 0.8-MeV/u H_2^+ and 45000 for 0.9-MeV/u H_2^+, while the peak height of the empty TEM grid for all energy H_2^+ was only a few dozen. The proton counts in the empty TEM grid might be caused by the

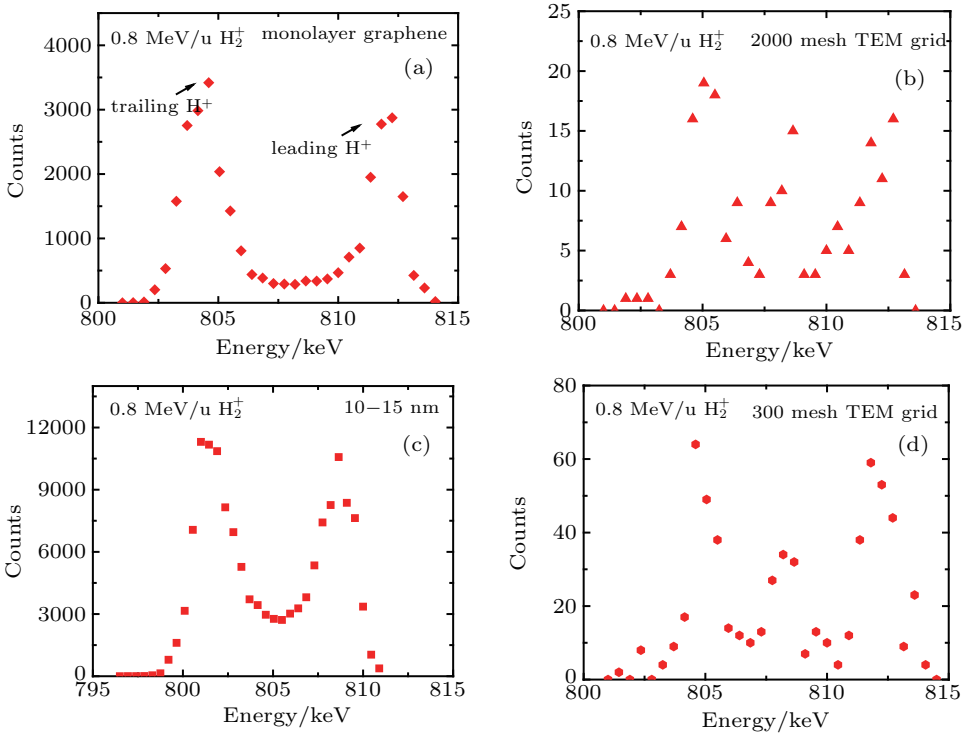


Fig. 2. The energy spectra of fragment protons at 0° generated by 0.8 MeV/u H_2^+ (a) in monolayer graphene foil, (b) in empty 2000 mesh TEM grid, (c) in 10–15 nm graphite foil, and (d) in empty 300 mesh TEM grid. The *X*-axis represents the energy of fragment protons, and *Y*-axis represents the counts of the protons at given energy.

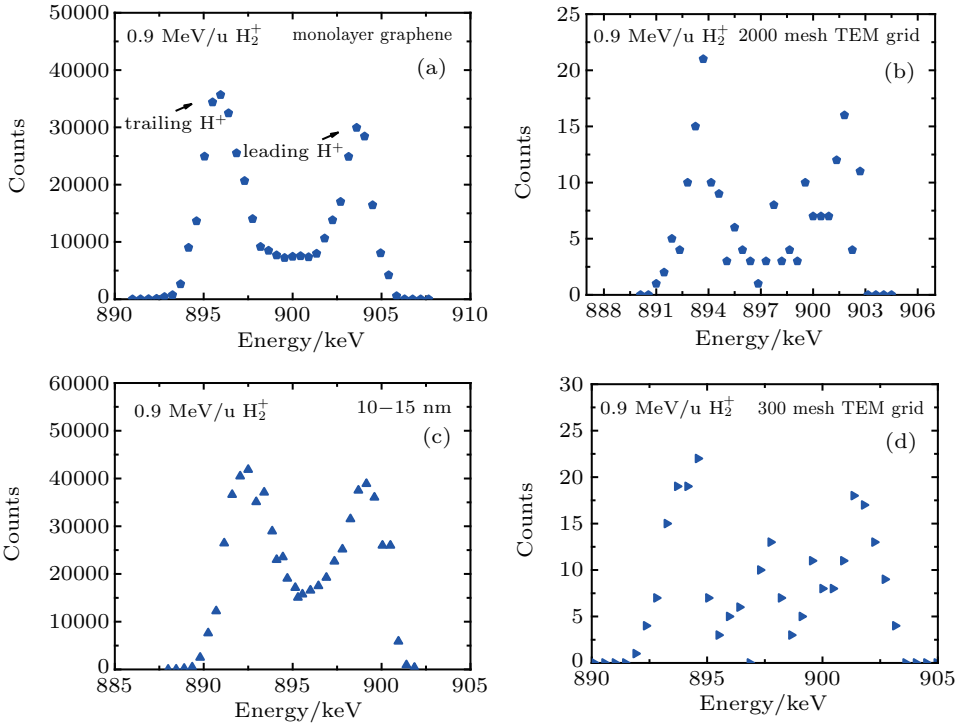


Fig. 3. The energy spectra of fragment protons at 0° generated by 0.9 MeV/u H_2^+ (a) in monolayer graphene foil, (b) in empty 2000 mesh TEM grid, (c) in 10–15 nm graphite foil, and (d) in empty 300 mesh TEM grid. The X-axis represents the energy of fragment protons, and Y-axis represents the counts of the protons at given energy.

collision of H_2^+ ions with residual gases in the vacuum pipeline, because the similar spectrum structure was found when no target was used. Obviously, the influence of the copper TEM grid was negligible, and two peaks in Figs. 2(a), 2(c), 3(a), and 3(c) were caused by the foils (graphene and graphite). Then we could infer that one single layer graphene was able to strip off the electron of H_2^+. It was also found in Figs. 2(a) and 3(a) that there were more trailing than leading protons, which means that the wake effect was available in monolayer graphene. The wake field generated by the leading proton was able to capture the trailing proton nearby and bent it to the beam direction, and some trailing protons which were originally not at 0° could be bent to the beam direction and be detected by the EA.

The energy spectra of the fragment protons from other graphene type foils (0.34 nm, 0.68 nm, 1.36 ± 0.34 nm, and 2.38 ± 0.34 nm) and more thick graphite type foils (8.5 ± 1.5 nm and 12.5 ± 2.5 nm) were tested, and the similar two peak structure were found. The peak height of the trailing/leading proton for all types of foils are shown in Fig. 4(a) for 0.8 MeV/u H_2^+ and Fig. 4(b) for 0.9 MeV/u H_2^+. It was found that the peak height varied within the range of 2500 to 12000 for 0.8 MeV/u H_2^+ and within the range of 20000 to 45000 for 0.9 MeV/u H_2^+. And the peak height for the monolayer graphene foil was not significant smaller than that for the thicker foils, which might indicate that the vast majority electrons of H_2^+ were stripped off by the first layer of the foil. Then the answer to the question was that one single layer is enough to strip off the electron of H_2^+ molecular ions.

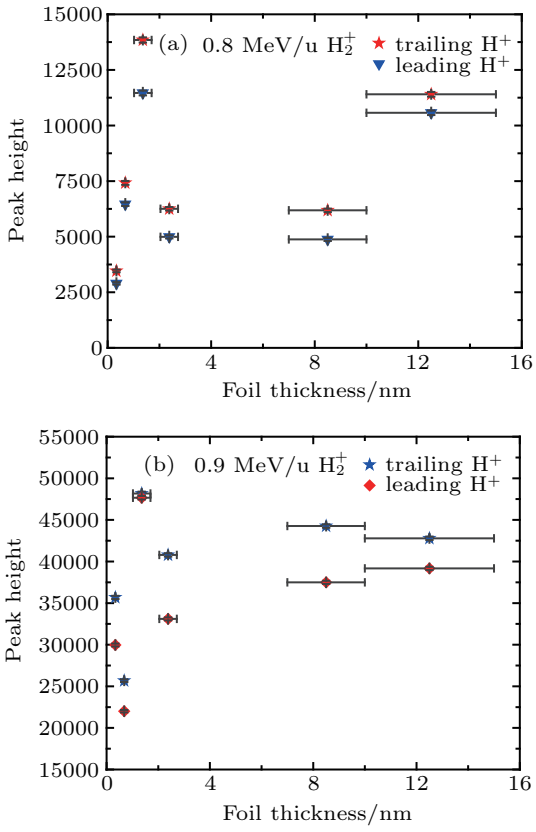


Fig. 4. The peak height of the trailing/leading proton at different foil thickness for (a) 0.8 MeV/u H_2^+ and (b) 0.9 MeV/u H_2^+.

The ratio $R = N_{\text{trailing}}/N_{\text{leading}}$ was used to represent the intensity of the wake effect, where N_{trailing} and N_{leading} are the

numbers of the trailing and leading protons in the proton energy spectrum. The thickness dependences of R at the incident energies of 0.8 MeV/u and 0.9 MeV/u are shown in Fig. 5. The ratio R was larger than 1.0 for all cases, which means more trailing than leading protons were found for all cases, and significant wake effects were observed for all foils. The data was very strange for the 0.9 MeV/u H_2^+ in 3–5 layers graphene, we cannot explain the abnormal data, we guessed that the beam condition may be abnormal for this point. If the abnormal point was discarded, it was found that the ratio R first increased with the thickness for the much thinner graphene foils, and then decreased with the thickness for more thick graphite foils. This indicated that the bending effect of the wake field on the trailing proton increased first and then decreased with the foil thickness.

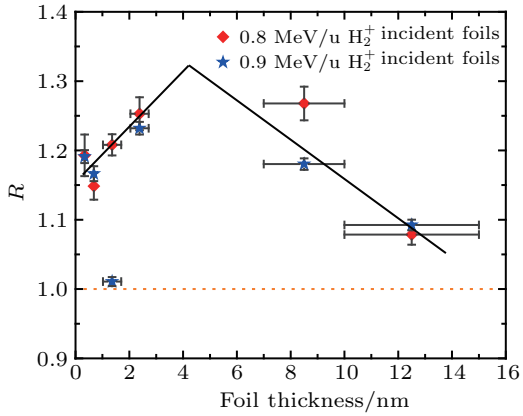


Fig. 5. The ratio R varied with the target thickness. The red solid point and blue solid point represent the experiment values for 0.8 MeV/u and 0.9 MeV/u H_2^+ incident foils, and the black solid line is only used to guide the eye. The ratio R first increased then decreased with the target thickness, except for the case of 0.9 MeV/u H_2^+ in 3–5 layers graphene foil which was abnormal.

In thin foils, the wake field was mainly affected by the inter-nuclear distance between the leading and trailing protons, as well as the thickness of the foil. Generally speaking, the wake field might be stronger for thicker foils because the lasting time of the wake field would be longer, and the ratio R would be larger. When the foil thickness was much thicker, the distance between the leading and trailing protons would be larger, and the wake field might decrease or even reverse polarity, which would make the ratio R much samller, just as shown in Fig. 5.

4. Conclusion

The interaction of MeV H_2^+ molecular ions with very thin graphene and graphite foils were studied. It was found that the vast majority of the electrons of the H_2^+ molecular ions could be stripped off in monolayer graphene foils. And it was very possible that the electrons of the incident H_2^+ were stripped off at the first atomic layer of the target when the H_2^+ molecular ions interacted with the thick foils. The existence of significant wake effect in monolayer graphene was confirmed experimentally, and the bending effect of the wake field on the trailing proton increased first and then decreased with the increase of the foil thickness. These experimental phenomena answer the question of how many atomic layers are needed to strip off the electron of H_2^+, and how the wake effect varies for really thin targets. Especially, it was found that the electron of H_2^+ was stripped off in the first layer, which was valuable to understand the main physical processes, including the stopping force of the medium and the energy loss per unit length after molecular ions enter the medium.

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