Guiding of Slow Highly Charged Ions through Nanocapillaries - Dynamic Aspect -

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Abstract. We have studied the properties of guiding of Ne⁷⁺ ions through nanocapillaries in insulating PET polymers for incident energies from 3.5 to 7 keV with a two-dimensional position sensitive detector. During the intensity evolution of transmitted ions, the deflection angle of the transmitted Ne⁷⁺ ions shows a few oscillations before approaching an equilibrium value. The experimental results are interpreted in terms of a scenario where the deflection angles of transmitted ions are governed by charge patches formed on the inner wall of the capillary. In addition, the memory effect of charge patches retained from previous irradiation are studied.

1. Introduction

Interactions between slow highly-charged ions (HCIs) and solid surfaces have extensively been studied experimentally and theoretically [1-3]. HCI-metal surface interactions, especially charge transfer processes from the metallic surface, can be well explained by the so-called classical over-the-barrier model [4]. With metallic microcapillary target, experimental evidence of hollow ions in vacuum and the related charge transfer processes have been shown [5-7]. On the other hand, interactions between ions and insulator surfaces have complex features, which depend on the insulator surface properties.

Recently, Stolterfoht et al. [8] showed that mesoscopic properties of the insulator surface can be used to guide slow ions along the capillary axis of an insulator nanocapillary. Qualitatively, this phenomenon can be explained as follows [8, 9, 10]: (1) ions collide with inner wall of a nanocapillary and a positive charge patch is formed near the entrance region, (2) the following ions are deflected by the charge patch into the capillary axis direction, (3) a considerable amount of ions are transmitted through the nanocapillary without any close collisions with the inner wall.

Until now, various kinds of insulator capillaries (multicapillaries [8, 11-14] and single-glass capillaries [15-17]) have been used to demonstrate the ion guiding effect. Especially for nanocapillaries in a PET foil, ion guiding properties in equilibrium conditions, i.e., after the saturation of the transmitted ion intensity, were reported and the relationship between the guiding ability and the width of the transmission profile was revealed [18-20]. In such
Four jaw slits: 1-2 mm

 Ion beam

400 mm

2 mm φ aperture

500 mm

100 mm

170 mm

PET foil with capillaries

PSD

Figure 1. Experimental setup. Ne$^7^+$ ions are collimated by 4 jaw-slits and a 2 mm aperture before the target, which can be rotated by the angles $\psi$ and $\chi$ as indicated. Transmitted ions are measured by a 2-D position sensitive detector (PSD). An electrostatic deflector can be set just after the target to analyze the charge state of the transmitted ions.

experiments, using a one-dimensional detector, it is rather difficult to measure the two-dimensional angular profiles of transmitted ions changing with time.

In this work, we utilize a two-dimensional (2-D) position sensitive detector (PSD) to measure the transmitted ions. Position and time information of the ions hitting the 2-D PSD was recorded by a list mode. This method [17, 21-25] allows us to measure not only the transmitted intensity but also the spatial change of the transmitted ions in steps of a few seconds [22]. We have observed that (1) the peak shape and the deflection direction of the transmitted ions varies during the intensity evolution, and (2) the deflection direction is affected by the charge patches produced by the previous irradiation, which is referred to as "memory effect" [25].

2. Experiment

We performed the experiments at RIKEN with Ne$^7^+$ ions from a 14.5 GHz electron cyclotron resonance (ECR) ion source [26]. The beam energies were between 3.5 and 7 keV, and the beam intensity ranged from 5 to 200 pA at the target position. The beam divergence was smaller than 0.5°, and the size of the beam at the target was 1-1.5 mm in diameter. The vacuum in the chamber was better than 1×10$^{-5}$ Pa. The experimental setup is shown in Fig. 1. A polyethylene terephthalate (PET) foil was attached to the target holder, which can be rotated by the angles $\psi$ and $\chi$ as shown in Fig. 1. Angles of $\psi$ and $\chi$ were measured from the beam direction. The PET foil included capillaries with a length of 12 μm and a diameter of 200 nm. The capillary density was 4×10$^6$ holes/cm², which was two orders of magnitude lower than that of the target used in previous experiments [8, 11]. To avoid charging up of the PET foil surface, Au was evaporated under 30° on the front and back sides of the PET foil. The Au layer reaches about 150 nm into the entrance and exit regions of the capillaries [8].

A two-dimensional (2-D) position sensitive detector (PSD), which consists of micro-channel plates and a wedge and strip anode is located 170 mm away from the target position. An electrostatic deflector can be set just after the target position, to analyze the charge state of the transmitted ions. The electric field of the deflector is parallel to the $\psi$ direction. We have measured the transmitted ions by the 2-D PSD as a function of time with a constant ion current. The information about the ion hitting the 2-D PSD including its position and time, and also the incident ion current were recorded by a PC program using a list mode. This allows us to measure the change of the profile and position of transmitted ions as a function of charge $Q_d$ deposited on the PET surface. The deposited charge $Q_d$ is a proper parameter to discuss the

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intensity evolution of the guided ions [20].

In this paper, we mainly present the dynamic behavior of the guiding effect for two different initial conditions [25].

(1) "Fresh" surface: to study the charging up processes on the inner wall, we performed experiments with a fresh surface condition. In this case the capillary inner wall was expected to hold initially no charge. To realize such condition, we waited about one week after the previous experiment. The characteristic discharge time of the charge patch in the capillary is of the order of 1-3 hours [8, 23, 25]. One week is expected to be enough to erase the previous memory on charge at the inner wall of the PET capillary.

(2) "Charged" surface: to study the charging up processes in conjunction with the remaining charge patches produced by pre-charging before the experiment.

3. Results and Discussion

Figure 2 shows a typical example of the intensity evolution of the transmitted ions as a function of $Q_d$. Moreover, a couple of 2-D images of the ions are depicted to demonstrate the profile and position of the transmitted ions during the intensity evolution. Here, the charge states of the transmitted ions are analyzed. Expected positions of Ne and Ne$^{7+}$ are shown by the arrows in Fig. 2. When the irradiation starts, only a weak neutral Ne peak is observed. As $Q_d$ increases, the Ne$^{7+}$ peak quickly increases. In the equilibrium condition, i.e., after the intensity saturation, a small number of ions with charge states between 0 and 6 are also observed although more than 90 % of the transmitted ions are Ne$^{7+}$. When we discuss the deflection angle of transmitted ions, we use the peak position of Ne$^{7+}$.

![Figure 2](image-url)

**Figure 2.** Typical evolution of the integrated intensity and the 2-D images of the transmitted ions as a function of charge $Q_d$ deposited on the PET surface. 2-D images for different deposited charge $Q_d$ are shown: A for $Q_d$=0-5 nC, B for $Q_d$=20-25 nC, and C for $Q_d$=80-85 nC. Here, an electrostatic deflector was set just after the target. Expected positions for different charge states of the ions are shown by arrows. In these 2-D images the number of 11ch corresponds to 1°.

During the evolution of the intensity, the 2-D images of transmitted ions vary as seen in Fig. 2. Note that the labels A, B, and C for the upper graphs correspond to those given in the lower graph. As is seen in Fig. 2, the deflection angle of Ne$^{7+}$ varies with $Q_d$. These changes show that the charge patches of the capillary inner wall evolve. In Fig. 2, the peak position of Ne$^{7+}$ moves along the Y-axis, which corresponds to the $\chi$ direction. The direction of this shift is determined by a "memory effect" of the pre-charged capillary inner wall [25]. The "memory effect" will be discussed in 3.2 Charged Surface.
During the intensity evolution, the shape of the transmitted ion profile changes. At equilibrium, the shape of the transmitted ion profile is almost circular and the angular width is about \(1^\circ\) [21, 25]. The angular widths of the transmitted ions depend on the beam energy [21, 25] as reported previously [11, 21]: the higher the beam energy, the smaller the angular width.

### 3.1. Fresh Surface

First we discuss results obtained with a fresh surface. Figure 3 shows the results for two different beam energies of 3.5 and 7 keV with the beam intensity of 50 pA [25]. Furthermore Fig. 4 shows the results for two different beam currents of 5 pA and 67 pA with the beam energy of 3.5 keV.

The PET capillary was tilted by \(\psi=0^\circ\) and \(\chi=2.8^\circ\) with respect to the incident beam direction. The observed intensities and deflection angles of the transmitted ions are plotted as functions of \(Q_d\). The deflection angle is deduced from the mean peak position on the 2-D PSD, which has the error (±0.2°). The deflection angle perpendicular to the tilt angle was constant (\(\psi=0^\circ\)) during the intensity evolution. Thus, only the deflection in the tilt direction is plotted.

The deflection angle moved toward the tilt direction in accordance with the increase of the transmitted intensity. The ion guiding processes can be explained as the deflection of ions by charge patches produced on the capillary inner wall by preceding ion-surface collisions [8, 9]. Thus, the increase of the deflection angle during the intensity evolution should be accompanied by the growth of charge patch in the capillaries. Moreover, the decrease in the deflection angle after the maximum in Figs. 3(a) and 4(a) may be attributed to the growth of an additional secondary charge patch in the capillaries. The oscillatory behavior of the transmitted ions may qualitatively be explained as follows;

(1-1) ions are deposited on the inner wall of the capillary entrance region so that it is charged up positively,

(1-2) the following ions are deflected by the entrance charge patch toward the capillary axis as indicated by the broken line in Fig. 5(a),

**Figure 3.** Incident beam energy dependence. Deflection angles (\(\chi\) direction) and intensity evolutions of Ne\(^{7+}\) ions transmitted through a fresh sample. The tilt angles were \(\psi=0^\circ\) and \(\chi=2.8^\circ\). The ion energies are 3.5 keV (a, c) and 7 keV (b, d). The ion current is 50 pA.

**Figure 4.** Incident beam current dependence. Deflection angles (\(\chi\) direction) and intensity evolutions of Ne\(^{7+}\) ions transmitted through a fresh sample. The tilt angles were \(\psi=0^\circ\) and \(\chi=2.8^\circ\). The ion currents are 67 pA (a, c) and 5 pA (b, d). The ion energy is 3.5 keV.
Growth of additional charge patch

Increasing

Decreasing

Primary charge patch

(a)

(b)

Growth of additional charge patch

Figure 5. Scenario of the oscillatory behavior of the deflection angle due to the growth of charge patches.

(1-3) with the growth of the charge patch the number of the deflected ions increases and also the deflection angle of them increases as indicated by the solid line in Fig. 5(a),

(1-4) with the increase of the deflection angles the ions collide with the inner wall of the capillary and create an additional charge patch as shown in Fig. 5(b),

(1-5) with the growth of the primary and secondary charge patches, the ions are shifted back to smaller deflection angles as indicated by the movement from the broken line to the solid line in Fig. 5(b).

For the lower incident energies of 3.5 keV the deflection angles of ions affected by the primary charge patch are large enough to produce the additional charge patch in the capillary as shown in Fig. 5. Previous studies [19] suggest that the equilibrium accumulated charge $Q_\infty$ at the charge patch does not depend on the beam energy and the effective potential $U$ produced by $Q_\infty$ has a weak energy dependence of $(E_p/q)^{-0.3}$, where $E_p$ is the energy and $q$ the charge state of the ions. With increasing beam energy, the effective potential $U$ becomes weaker [17] and the deflection angle of ions by the primary charge patch becomes smaller. Hence, the additional charge patch may not be produced. Thus, for 7 keV, the oscillatory behavior is not observed.

Our previous studies [22] demonstrated that the fraction of transmitted ions is nearly independent on the incident beam current from 5-67 pA at RIKEN and for 100-1000 pA at HMI, which indicates that a sudden increase in the discharge current depleting the entrance charge patch, though the fraction of the transmitted ions decreases as the incident beam current decreases. For the lower incident beam current of 5 pA (shown in Figs. 4(b, d)), the charge patch size (or the effective potential $U$) of the primary charge patch may be smaller than those for 50 pA (shown in Figs. 3(a, c)) and 67 pA (shown in Figs. 4(a, c)). Then the intensity and the deflection angle of deflected ions by the primary charge patch at the entrance region for 5 pA may be not enough to make an additional charge patch, which deflect back the beam as shown in Fig. 5(b). This effect may cause the obscure oscillation in Fig. 4(b).

Very recently Skog et al. [24] and Stolterfoht et al. [27] reported on the oscillatory behavior of the transmitted 7 keV Ne$^{7+}$ ions through nanocapillaries in SiO$_2$ and 3 keV Ne$^{7+}$ ions through nanocapillaries in PET, respectively. Our observation is in good agreement with their results. Skog et al. [24] have explained the phenomenon with model calculations where charge patches are sequentially formed in the nanocapillaries. They needed three charge patches to explain the deflection angle of the transmitted ions in consistency with the aspect ratio (250:1) of the capillaries. Stolterfoht et al. [27] have explained the phenomenon with the dynamically formed multiple patches in the nanocapillaries for their conditions. In our case, the existence of two charge patches may explain the deflection angle of the transmitted ions considering the aspect ratio (60:1) of our capillaries [25].
Figure 6. Evidence for memory effects. 2-D images of the transmitted ions at the different charge $Q_d$ deposited on PET surface with the tilt angles $\psi=3^\circ$ and $\chi=-2^\circ$. The incident beam is 4.9 keV Ne$^{7+}$ with the beam current of 50 pA. In order to see the deflection angle of the transmitted ions, we don’t use the electrostatic deflector after the PET capillaries. Although the transmitted ions contain all charge states of Ne ions, more than 90% of transmitted ions are Ne$^{7+}$ ions. Values of the deposited charge $Q_d$ are shown in the each figure. Each image is plotted in linear scale and normalized the peak value. Therefore these images only show the position information. (a) Pre-charged tilt angles $\psi=0^\circ$ and $\chi=0^\circ$. The transmitted beam position for the pre-charged tilt angles is indicated by the red star, whose observed angles are $\psi=0^\circ$ and $\chi=0^\circ$. (b) Pre-charged tilt angles $\psi=3^\circ$ and $\chi=0^\circ$. The transmitted beam position for the pre-charged tilt angles is indicated by the blue star, whose observed angles are $\psi=2.8^\circ$ and $\chi=0^\circ$ and agree with the tilt angles within the experimental uncertainty($\pm0.2^\circ$). At first, a weak peak appears at a few nC ((a)-1 and (b)-1), then moves to the direction, indicated by arrows (red solid or blue broken), depending on the memory of the charge patch on the inner wall of capillaries, and then approaches the equilibrium position((a)-6 and (b)-6) after an oscillation. The equilibrium positions agree with the tilt angles within the experimental uncertainty. Initial positions of the weak peaks are indicated by intersections.

3.2. Charged Surface

In the previous subsection, we showed evidence for the change of the charge patch from the variation of the deflection angle. Here, we present the effect of charge patches produced by pre-charging on the evolution of the deflection angle. Although an old charge patch is known to affect the growth of new charge patches, the information about the memory effect is limited so far.
Figure 7. Relationship between the direction to which the peak moves and ”memory” of the surface. The PET foil is located at O. OP1 is the present tilt direction. OP0 and OP0’ are the different pre-charged directions. When the ions enter the capillaries with ”memory”, the deflection angle of the transmitted ions moves as indicated by the red solid arrow or the blue broken arrow depending on the pre-charged direction. The movement may show an oscillatory behavior. Finally, the ions approach the equilibrium direction around OP1.

In Fig. 6, the 2-D images of the transmitted ions at the different $Q_d$ for the same tilt angles ($\psi=3^\circ$ and $\chi=-2^\circ$) with different memory. The capillary inner walls were pre-charged at different tilt angles, i.e., $\psi=0^\circ$, $\chi=0^\circ$ for Figs. 6(a), and $\psi=3^\circ$, $\chi=0^\circ$ for Figs. 6(b).

After the pre-charging at $\psi=0^\circ$ and $\chi=0^\circ$ ($\psi=3^\circ$ and $\chi=0^\circ$) for a few hours’ irradiation confirming the saturation of the transmitted ion intensity, we changed the tilt angles to $\psi=3^\circ$ and $\chi=-2^\circ$, and measured the transmitted ions with the constant beam current of 50 pA. The equilibrium positions for the pre-charged tilt angles are indicated by the red star ($\psi=0^\circ$ and $\chi=0^\circ$) in Figs. 6(a)-1 and the blue star ($\psi=3^\circ$ and $\chi=0^\circ$) in Fig. 6(b)-1. A weak peak appeared as shown in Fig. 6(a)-1 (Fig. 6(b)-1), which moved as indicated by the red arrow (blue broken arrow) and finally approached the equilibrium position shown in Fig. 6(a)-6 (Fig. 6(b)-6). In the Figs. 6(a) and 6(b), the initial weak peak positions of the transmitted ions for the present tilt angles ($\psi=3^\circ$ and $\chi=-2^\circ$) are indicated by intersections.

We have to mention that for the higher beam energy of 7 keV (not shown in this paper), the oscillatory behavior became not so clear as in the case of the fresh surface, which can be attributed to a smaller deflection by the charge patch in the entrance region.

When ions enter the capillaries with a pre-charged inner wall, the ions are deflected by the charge patch in the opposite direction and collide with the inner wall. With the growth of the new charge patch and the decrease of the size of the old charge patch due to the discharge processes, the ions may be finally guided along the capillary axis passing through the capillaries. We expect that the old charge patch may also be reduced by secondary electrons produced by the creation of the new one. Hence, in this case, the old charge patch becomes smaller as the new charge patch grows. In the equilibrium conditions only the new charge patch persists guiding the ions in a self-organized manner along the capillary axis.

The direction to which the deflection angle of transmitted ions moves just after the start of the ion transmission, is mainly determined by the old charge patch produced by the pre-charging, as shown in Figs. 6(a) and 6(b). The relationship between the ion deflection angle and the tilt angle is shown in Fig. 7. For our experimental conditions, P0: $\psi=0^\circ$ and $\chi=0^\circ$, P0’: $\psi=3^\circ$ and $\chi=0^\circ$, and P1: $\psi=3^\circ$ and $\chi=-2^\circ$. At the beginning of the ion guiding, a weak peak appears in between the OP0(OP0’) and OP1 directions, moves to the OP1 direction in the P1-O-P0(P0’) plane, as shown by the red solid(blue broken) arrow, and approaches the OP1 direction after damped oscillations.

4. Summary
In summary, we observed an oscillatory behavior in the deflection angles of ions guided by capillaries during their intensity evolutions. Oscillatory behavior in the ion deflection angle may be understood in terms of a secondary charge patch formed in addition to the main charge.
patch in the capillary entrance region. And also we have demonstrated that the "memory" of the charge patch on the inner wall of capillaries affects the oscillatory behavior.

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