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Guiding of 150 keV O^{6+} ions through nanocapillaries in an uncoated Al_2O_3 membrane: special time dependence of the transmission profile width*

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This paper reports that the transmission of O^{6+} ions with energy of 150keV through capillaries in an uncoated Al_2O_3 membrane was measured, and agreements with previously reported results in general angular distribution of the transmitted ions and the transmission fractions as a function of the tilt angle well fitted to Gaussian-like functions were observed. Due to using an uncoated capillary membrane, our ψ_c is larger than that using a gold-coated one with a smaller value of $\frac{E_p}{q}$, which suggests a larger equilibrium charge Q_∞ in our experiment. The observed special width variation with time and a larger width than that using a smaller $\frac{E_p}{q}$ were qualitatively explained by using mean-field classical transport theory based on a classical-trajectory Monte Carlo simulation.

Keywords: guiding effect, Al_2O_3 nanocapillaries, uncoated membrane, transmission profile width

PACC: 6890

1. Introduction

Studies of nanostructures at surfaces and in solids have received considerable attention during the past few years.[1,2] This work has been motivated by the rapid growth of nanotechnology with the aim of the refinement of interfaces used in microelectronics and related fields. Particular attention has been paid to linear structures of mesoscopic dimensions, such as capillaries. Since it is difficult to study the interior of tiny tubes, as a promising method, an investigation into the transmission of slow ions through nanocapillaries was conducted to probe the physical and chemical properties of the capillary surface. Initially, we used capillaries in conducting metallic materials, whose surface properties are well known, to study the decay process of hollow atoms produced after transmission of highly charged ions（HCIs）through the capillaries.[3]

Recently, Stolterfoht *et al*[4] reported their interesting finding that the majority of HCIs can be transmitted through strongly tilted capillaries in highly insulating polyethylene terephthalate (PET) in their initial charge state, in contrast to the result of conducting capillaries, and that the centroid of the transmission profile shifts relative to the direction of the incident beam to an angle nearly equal to the tilt angle of the capillaries, which was referred to as the guiding effect. Since then Stolterfoht and his co-workers have performed considerable work on ion guiding of capillaries in PET experimentally and theoretically with respect to the profile width and fraction of the transmitted ions. Due to the increasing interest in this subject, several laboratories have started investigations of capillary guiding too, using insulating or semiconducting materials such as PET,[5] SiO_2,[6] SiN,[7] Al_2O_3[8] and personal computer polycarbonate.[9] The projectile energy used by them ranged from 0.05keV/u to 7keV/u. Their results showed that with increasing

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projectile energy, the guiding range for the tilt angle of the capillaries reduced sharply from ±25° down to ±5°, or even smaller. According to Stolterfoht *et al*, the capillary guiding was attributed to the internal electrostatic field produced at the entrance and exit of the capillaries mainly by the charges deposited at the entrance in a self-organizing way. This subject has also been studied theoretically by others, but the guiding effect has not been well understood in detail until now. So far, the transmitted fraction as a function of the capillary tilt angle can be well described mathematically; however, the profile width of the transmitted ions is only regarded approximately to be constant with the capillary tilt angle. In conjunction with our experimental conditions, the aim in the present work is to study the transmission of O^{6+} with a relatively higher energy of 150 keV (in turn a higher value of $\frac{E_p}{q}$) through capillaries in an uncoated Al_2O_3 membrane, with analysis of the profile width variation as an emphasis.

2. Experiment

The experiment was performed at the Institute of Modern Physics, Chinese Academy of Science, Lan Zhou, China. A 14.5 GHz Electron Cyclotron Resonance ion source provided the 150 keV O^{6+} ion beam. The beam, with an average intensity of 0.5 nA, was analysed with a bending magnet, focused by two electrostatic quadrupole doublets, and collimated by a pair of four-jaw slits set 0.85m apart, then entered the experimental chamber. The vacuum in the beam line and in the experimental chamber was below 4×10^{-5} Pa.

A schematic diagram of the experimental setup is shown in Fig.1. In the chamber, a special grounded metallic target holder with five degrees of freedom (two rotational and three translational) was mounted at the end of the manipulator, and the target holder was insulated from the manipulator. The target holder allows an adjustment of elevation angle which helps in the optimization of the target position with respect to the beam direction. 400 mm away from the target, a one-dimensional position sensitive microchanneltron detector (PSMD) with effective length of 90mm and width of 10mm was used to detect the outgoing ions. The charge signals coming from the two ends of the anode of the PSMD were transferred to voltage signals, and then processed with a set of standard electronic elements made by the ORTEC company, including pre-amplifiers, amplifiers, a position sensitive analyser and so on. They turned into the position signals of the ions at the PSMD, and eventually input into a computer to be recorded with multi-parameter acquiring software (MPA-3).

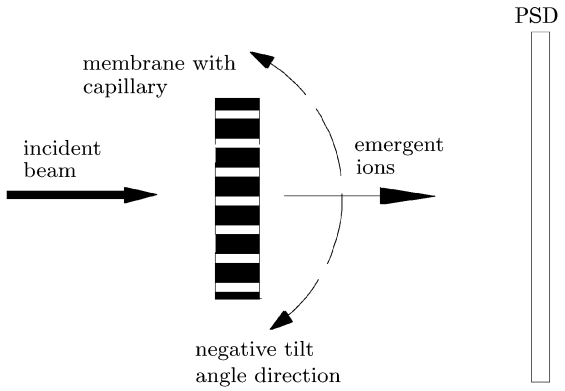


Fig.1. Schematic diagram of the experimental chamber with the target and the one-dimensional position sensitive channeltron detector.

A membrane with a hexagonal pattern of ordered Al_2O_3 nano-capillaries was used. The capillaries have 50 nm diameters and 10 μm lengths, giving an aspect ratio angle of ≈0.3°, and the membrane has a geometrical transparency of approximately 21% corresponding to a capillary density of about 10^8 capillaries per mm^2. We did not have our membrane covered with any conducting material, which differed from nearly all those films used in capillary guiding experiments by others, and to observe the influence of this on the experimental result is also a purpose of the present work. Figure 2 shows the regular distribution of the ordered nano-capillaries over the surface.

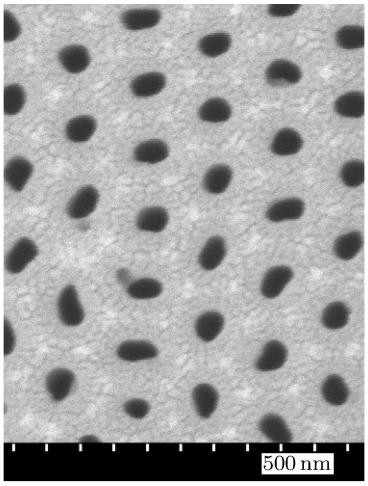


Fig.2. SEM micrograph of the Al_2O_3-capillary membrane.

At first, we performed a fast scanning measurement in steps of 0.5° for tilt angles near and to find 0° tilt angle position by comparing the count rates, where the direction of the capillaries coincides with the direction of the beam and, correspondingly, the amount of the transmitted ions is the largest during the same time stretch relative to that of all the other tilt angles. After finding the 0° position, an optimization of the target position including elevation angle with respect to the beam direction was performed. A −5° tilt orientation of the capillary film relative to the direction of incident beam was the starting point of the formal measurement. From here on, the film was rotated in steps of 1° in a single direction towards the 0° tilt angle position, until across the 0° position the 5° position was reached, an angular distribution spectrum, recorded by a PC, of the transmitted ions being obtained at each tilt angle. Each spectrum measurement cost time of 10 minutes, and the time spacing between two neighbouring tilt angles was also 10 minutes, being used to save the data of the previous tilt angle position, rotate the film to the next tilt angle, and wait for the film to calm down.

3. Results and discussion

The ions transmitted through the Al_2O_3 capillaries were analysed for their charge state distribution by using a pair of deflecting plates, and the result showed that only a small fraction of the ions which passed through the capillaries changed their charge state, and actually this is just the first characteristic featuring the so-called capillary guiding already mentioned in the introduction. Due to this fact, the measurements of the angular distribution of the transmitted ions by others were directed to those in the initial charge state by using a combination of an electrostatic analyser and a channeltron detector rotatable around the target. This fact also justified our use of a position sensitive detector with no charge state separation for the measurements of the angular distributions.

The angular distribution of the transmitted ions as a function of the position at the channeltron plate for different capillary tilt angles ψ is shown in Fig.3. To avoid crowding, the angular distribution spectra for capillary tilt angle −5°, −4° , −3° and 5° are not given.

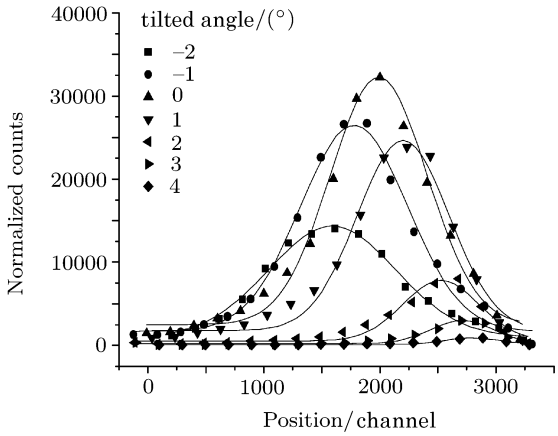


Fig.3. Angular distribution of 150 keV O^{6+} ions transmitted through AL_2O_3 capillaries for capillary tilt angle ranging from –2° to 4°. Experimental data (symbols) and fit of each profile to a Gaussian function (solid lines). In the figure about 240 channels correspond to a position angle of 1°.

First of all, we can see, in accordance with previously reported results, at tilt angles larger than the geometrical opening angle of the capillaries, considerable transmission can still be observed. Also, when the capillary membrane is tilted with respect to the direction of the incident beam, the peak position of the transmitted ions shifts accordingly, to a position angle (similar to the term of observation angle used by others, and defined in Fig.4) nearly equal to the tilt angle，which is another characteristic feature of capillary guiding. The exception to this is the 4° tilt angle whose corresponding peak is very low, even somewhat invisible, and does not shift significantly as those for the other tilt angles did. This shall be discussed later in terms of guiding angle and guiding ability. In the figure, approximately 240 channels correspond to a position angle of 1°. The width of the transmission profile for 0° tilt angle is 3.6° full width at half maximum (FWHM).

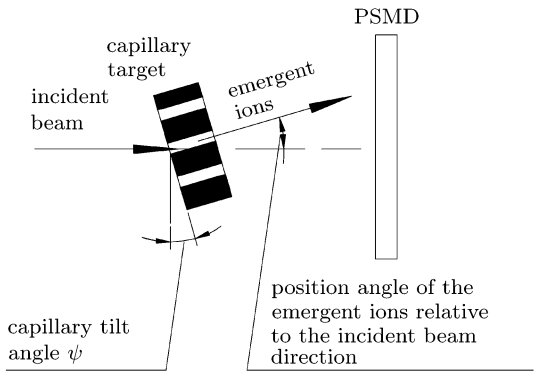


Fig.4. Schematic diagram of the tilt angle of the capillary membrane and the position angle for the transmitted ions.

The integrated angular distribution of each capillary tilt angle is shown in Fig.5. The fraction of transmitted ions was described[10] as Eq.(1),

$$f(\psi) = f(0°)\mathrm{e}^{-\frac{E_\mathrm{p}}{qU}\sin^2\psi}, \tag{1}$$

$$f(\psi) = f(0°)\mathrm{e}^{-\frac{\sin^2\psi}{\sin^2\psi_\mathrm{c}}}, \tag{2}$$

and another expression Eq.(2) was used to fit the integrated ion transmission as a function of the tilt angle, introducing a characteristic tilt angle ψ_c.[11] The guiding angle ψ_c, sometimes also called the critical angle, is a measure for the guiding power specifying the ability of the capillaries to guide ions. After successfully fitting the experimental data to the transmission function Eq.(2), we can get $\psi_\mathrm{c} = 2.13°$. The weakness and stiffness of the peak at 4° tilt angle observed in Fig.3 now can be easily understood, since earlier at the tilt angle of 2.13° the transmitted fraction had already dropped to 1/e of that at 0° tilt angle, and the ability of the capillaries to guide ions at 4° tilt angle would become even poorer, as indicated by the fitting function Eq.(2). In a paper also studying Al_2O_3 capillary guiding,[11] a value of $\psi_\mathrm{c} = 1.1°$ was obtained.

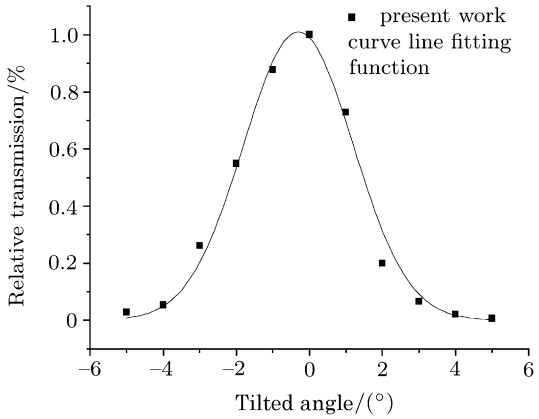


Fig.5. Integrated transmission for tilt angles –5° to 5° in steps of 1° relative to transmission at 0°. Experimental data (squares) and fitting to the transmission function (curved line).

According to Ref.[12],

$$\sin^{-2}\psi_\mathrm{c} = \frac{E_\mathrm{p}}{qU}, \tag{3}$$

$$U = \frac{Q_\infty}{c_\mathrm{e}}\left(\frac{E_\mathrm{p}}{q}\right)^{-0.3}, \tag{4}$$

$$\sin^{-2}\sigma_t = \frac{1}{4\ln 2}\frac{E_\mathrm{p}}{qU_\mathrm{t}}, \tag{5}$$

$$U_\mathrm{t} = \frac{Q_\infty}{c_\mathrm{t}}\left(\frac{E_\mathrm{p}}{q}\right)^{-0.3}, \tag{6}$$

where σ_t represents the profile width of the transmitted ions, E_p and q represent the kinetic energy and charge state of the ions respectively; U_t, U and Q_∞ represent an effective potential responsible for ion defocusing in the exit region of the capillary, an effective potential difference across the capillary diameter in the entrance region responsible for ion loss (or fraction transmitted) in the scattering region, both produced mainly by the charge deposited in the entrance region, and the equilibrium charge deposited primarily in the entrance region, respectively; and c_t and c_e are two constants both similar to a capacity, which determine the potentials U_t and U. Equations (3) and (4) show explicit dependence of ψ_c on $\frac{E_\mathrm{p}}{q}$ and $\frac{Q_\infty}{c_\mathrm{e}}$ so, $\frac{Q_\infty}{c_\mathrm{e}}$ being treated as a constant value,[13] ψ_c would vary with only $\frac{E_\mathrm{p}}{q}$, and the larger the value of $\frac{E_\mathrm{p}}{q}$, the smaller the ψ_c. The value of $\frac{E_\mathrm{p}}{q}$ in our experiment is 25 kV, and bigger than that of 1 kV in Ref.[11]; however, the obtained value ψ_c is bigger than that in Ref.[11] too, as presented above. The only reason for this lies in the fact that our membrane was not gold-coated which, excess deposited charges at the capillary wall not being able to be conducted away via the gold layer, probably caused the greatly enlarged Q_∞, and in turn caused the enlarged ψ_c according to the equations. So, due to using an uncoated membrane, our ψ_c is larger than that in Ref.[11], in spite of our larger $\frac{E_\mathrm{p}}{q}$. Unlike other capillary guiding experiments, in our experiment using uncoated film, the self-organizing charging and discharging of the capillaries seemed to be destroyed. However, the integrated angular distribution in Fig.5 can be well fitted by transmission function Eq.(2) with a larger value of $\psi_\mathrm{c} = 2.13°$, so a larger equilibrium charge Q_∞ seems to exist according to Eqs.(3) and (4).

In Fig.6 the transmission profile width (FWHM) variation with capillary tilt angle is shown. In Ref.[12], the profile width was approximately considered to be independent of the capillary tilt angle, as indicated in Eqs.(5) and (6). However, our result exhibits a monotonously and quasi-linearly decreasing tendency of the width, not with tilt angle, but with time, as seen in Fig.6. It is noted that the sequence of the data points obtaining in Fig.6 is one by one from –5° to 5° by the capillary tilt angle, as mentioned in Section 2. In Refs.[13, 14], a mean-field classical transport theory based on a classical-trajectory Monte Carlo (CTMC) simulation was developed, and the collective

field effects of an entire ensemble of nanocapillaries in the beam spot were taken into account in a mean field F_{mean}. As those capillaries are simultaneously charged, a mesoscopic electric field is generated which is aligned along the capillary axis. Due to the large distance of the charge patches near the entry side from the exit surface (10 μm) the ensemble can be viewed as a charged condenser consisting of two metallic plates (the gold layers) between which there is a dielectric material. The mean field can then be estimated from the total charge Q_∞ deposited near the entrance, the dielectric constant of the insulating material, and the irradiated target area. The mean field leads to additional defocusing at the capillary exit hole. This mean-field theory yielded good agreement with the experimental result in profile width variation. In our case, an uncoated capillary foil was used. However, the foil during the experiment can still be viewed as a charged condenser, with the front surface and the back one being just two parallel plates. Unlike the case using coated foil, the above mentioned mean field in our foil is stronger, since the contribution from the entry surface with higher charge density is included while, in the case using a coated one, only the total charge Q_∞ deposited near the entrance works. This can also explain the observed width of 3.6° at 0° tilt angle in our experiment larger than that of 1.18° in Ref.[12], where a smaller value of $\frac{E_{\mathrm{p}}}{q}$ was used; this, however, contradicts the semi-empirical equations (5) and (6). In our case, the uncoated front surface of the foil was exposed to the incident beam, so it got a very high charge density very soon after the experiment began, which would contribute to the angular spread of the transmitted ions significantly. Initially, when the front surface has a high charge density, while the back surface has nearly a zero one, this is the time when the mean-field is the strongest, and the largest width is produced corresponding to that at –5° tilt angle as shown in Fig.6. Due to the faster surface electric conductivity than bulk conductivity, most of the charges coming from the front surface of the foil and the charge patch deposited near the entry side of the capillaries would diffuse first, along the internal wall of the capillaries, to the back surface, so the charge density of the back surface has a clear dependence on time. Clearly, when the charge density of the back surface becomes larger gradually with time, the mean field produced by the condenser consisting of the front and back surface of the foil would become weaker with time, therefore the experimental fact that the profile width dominated by this mean field has a tendency to decrease with time as seen in Fig.6 is also reasonable.

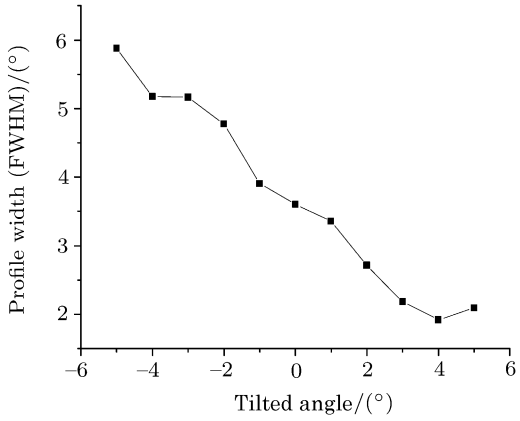


Fig.6. Transmission profile width variation with capillary tilt angle from –5° to 5°. The experimental data are shown as square symbols, and the line is to guide the eye.

It is noted here that properly fitting the transmission fraction as a function of the tilt angle to the transmission function Eq.(2) produced a certain guiding angle of $\psi_{\mathrm{c}} = 2.13°$, so the equilibrium charge Q_∞, deposited near the entry side of the capillaries and responsible for the transmission fraction, may be considered to remain constant approximately during the experiment according to Eqs.(3) and (4). So the contribution from Q_∞ to the angular spread of the transmitted ions is also steady. The increasing charge density of the back surface with time caused the decresing profile width.

Although Eqs.(5) and (6) indicate tilt angle independence of the profile width, both the experimental results[12] and simulation results[14] suggest tilt angle dependence that was not observed in our experiment, which means that the time dependence of the width in our experiment is stronger than the tilt angle dependence.

4. Conclusion

In conclusion, we measured the transmission of O^{6+} ions with a higher energy of 150 keV (and in turn a higher value of $\frac{E_{\mathrm{p}}}{q}$) through capillaries in an uncoated Al_2O_3 membrane, and observed agreements with previously reported results in the general angular distribution of the transmitted ions and the transmission fractions as a function of the tilt angle well

fitted to Gaussian-like functions. Due to using uncoated capillary membrane, our ψ_c is larger than that using a gold-coated one with a smaller value of $\frac{E_p}{q}$, which suggests a larger equilibrium charge Q_∞ in our experiment. We qualitatively explained the observed special width variation with time and a larger width than that using a smaller $\frac{E_p}{q}$ by using a mean-field classical transport theory based on CTMC simulation.

Undoubtedly, the charge density of the back surface of the uncoated foil in our experiment would not increase infinitely: relaxation and successively another circle of the charge density variation corresponding to a periodic variation of the profile width are anticipated. The explanation of the guiding effect has proved to be challenging work. We shall perform capillary guiding experiments using uncoated membranes to further confirm the conclusion drawn above.

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