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Peculiarities of proton transmission through tapered glass capillaries

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Summary. — A study of the 150–320 keV proton beam transmission through tapered glass (borosilicate) capillaries with different diameters of the input and output of the capillary was performed. The focusing effect was observed. The areal density of the transmitted beam is enhanced by approximately 20 times. It was shown that changing a taper angle from 0.5 deg to 1.7 deg evidences increase of the transmission coefficient by more than 300 times keeping the initial energy spectrum of ions. The ion transmission through self-ordered nanoporous alumina membranes prepared by anodic oxidation of high-purity aluminium was studied for different energies of ions.

PACS 34.35.+a – Interactions of atoms and molecules with surfaces.

1. – Introduction

Slightly tapered glass capillaries with micro or submicro outlet diameters attract much attention as ion beam focusing lens [1-4]. The smallest beam spot size obtained so far was $0.3\,\mu$m [1]. Previous studies have demonstrated the focusing effect with respect to many kinds of ions and energies [1-4]. The focusing factor of a glass capillary, defined as the ratio of areal current density at the inlet and outlet, depends on the ion species, the incident energies and the shape of glass capillary. This factor of 8 keV Ar^{8+} is about 10 and for 2 MeV He^{+} beam amounts to about 1000 [1-4].

The purpose of the present paper is to study the dependence of proton transmission through capillaries on capillary taper angles. An ion energy region was chosen to satisfy the experimental conditions of local implantation, local elemental and structure analysis, micro- and nanolithography, X-ray radiography, and applications in biology and medicine [3,5]. For example, this technique enables in-air PIXE measurements of various samples that are not compatible with the vacuum environment [2] (wet solids, liquids and gases). Slightly tapered glass capillary optics is applied to work as a differential

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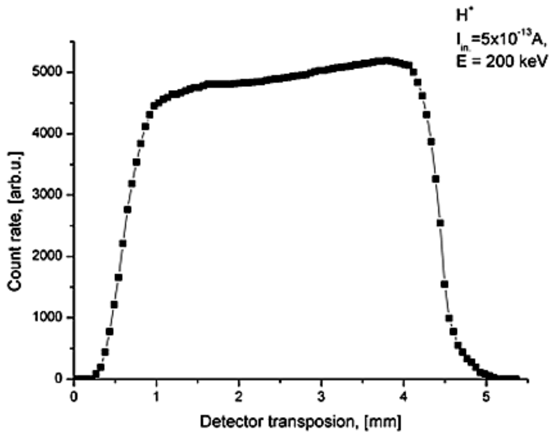

Fig. 1. – Shape and angular distribution of a 200 keV proton beam transmitted through the capillary.

pumping orifice as well as a focusing lens [2]. Recently, the processes of interaction of accelerated protons with the surface of cylindrical insulating capillaries as well as time and angular distributions of ions transmitted through such capillaries have been studied in our paper [6].

2. – Experimental technique

A schematic of the experimental setup designed for studying the transmission of accelerated ions through capillaries has been reported in our paper [6] (fig. 1 in this paper.) This setup, which enters the implantation complex formed on the basis of 1 MeV electrostatic ion accelerator, consists of four units: 1) a system of ion beam formation, 2) a scattering chamber, 3) a measuring chamber, and 4) a system for detecting scattered ions. The parameters of a setup are as follows: the error in determining angles in measurements of angular distributions is not larger than $3.3 \cdot 10^{-3}$ deg and the error in the capillary orientation with respect to the beam axis is not larger than $2.5 \cdot 10^{-2}$ deg.

The used proton beam energy of 150–320 keV was determined by a magnetic analyzer with an error of 0.1%.

We measured the angular and the time distributions of protons transmitted through glass capillaries with the inlet diameters of 3.5 and 0.5 mm and the outlet diameter of 0.1 mm and the length of 150 mm in the range of taper angles of 0.5 to 2.2 deg. Input ion currents from 10^{-13} to 10^{-9} A were used in these experiments. Angular distributions and energy spectra of transmitted protons were measured directly using mobile silicon surface barrier detector positioned at a distance of 90 cm from a capillary holder. The total measured energy resolution of the recording system does not exceed 16 keV.

3. – Experimental results and discussion

The angular distribution of protons transmitted through the tapered capillary with an inlet diameter larger than an ion beam diameter and an outlet diameter of 100 μm is shown in fig. 1. It should be mentioned that a practically uniform distribution of ion

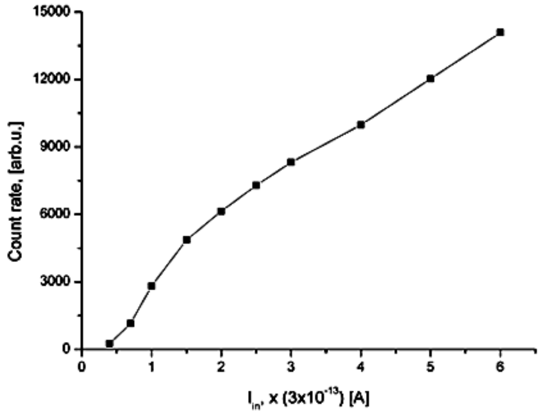

Fig. 2. – Count rate of particles transmitted through the capillary *vs.* proton current at the input of the capillary.

beam density with sharp edges is registered in this case. Moreover, a spot size amounts to 3.8 mm that corresponds to a beam divergence of 0.13 deg. An initial beam divergence was only 0.015 deg. An integral of an area under the curve in fig. 1 shows that the fraction of transmitted protons is equal to 80%, *i.e.* the number of transmitted ions relative to those entering into the capillary. Therefore, taking into account that the outlet diameter of the capillary is 100 μm and the initial beam diameter amounts to 500 μm, the focusing factor reaches up to 20. An effort to measure angular distributions of transmitted protons at high currents at the input of the capillary was failed as the measuring system functioning in a regime of individual particle registration was overloaded. Scanning of transmitted ion beams, at input beam currents of the order of 10^{-9} A, showed that angular distributions of beams transmitted through the capillary are independent of the incident current. In case of this capillary oriented along the beam axis, the count rate *versus* the beam current at the input of the capillary is shown in fig. 2. The results presented in fig. 2 demonstrate a strong nonlinear behavior of the current at the output of the capillary *versus* intensity of the input beam up to an input current of $5 \cdot 10^{-13}$ A. It is well accepted that such protons are guided electrostatically due to the charging up of the inner wall of capillaries made of insulating material [7].

Figure 3 shows the time dependences for swift protons transmitted through cylindrical and tapered capillaries with the diameter of the inlet hole less than the diameter of the ion beam. In spite of the practically equal currents at the input of these capillaries, time evolutions of beam currents at the output of the capillaries are strongly different in shapes and frequencies of the beam intensity oscillation. Current pulse frequencies of ions transmitted through the tapered capillary exceed essentially those for the cylindrical capillary. On the contrary, shorter pulse durations are typical for the tapered capillary. It should be also noted that a pedestal at the level of 1300 pulse/s is observed in fig. 3b. In the case of the tapered capillary such an interpulse transparency is not revealed.

The above-mentioned experimental results confirm our recent assumption [8] on the dominant role of charging up a face part of the capillary in the transformation of continuous ion beams into oscillating ones. This effect is not observed if an input hole of capillaries exceeds the beam diameter. A conducting protection of the face part of the

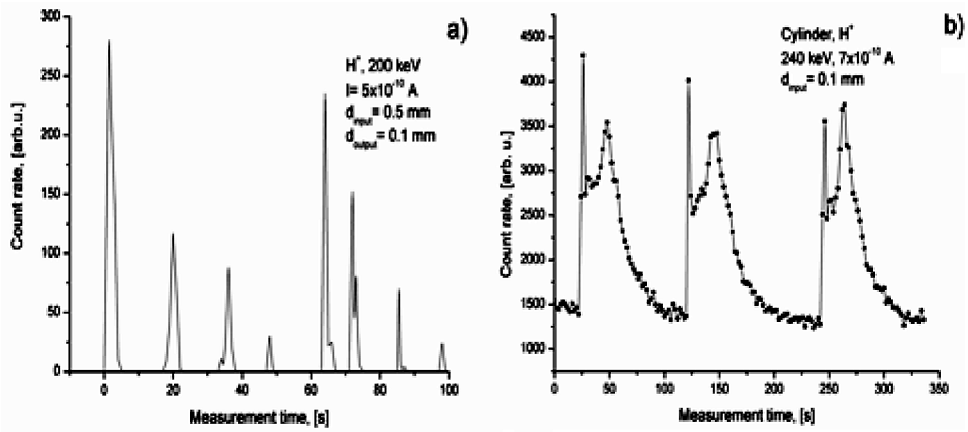

Fig. 3. – Time distributions of protons transmitted through a tapered capillary (a) and cylindrical capillary (b).

tapered capillary (fig. 3a) causes a total suppression of this effect (fig. 4). The angular distribution of transmitted particles in fig. 4 looks like the distribution presented in fig. 1. Nevertheless, the transparency of this capillary with a taper angle of 0.5 deg is a few hundred times less than this parameter for the capillary with a taper angle of 1.7 deg. An analogous strong decrease of the transmission coefficient was observed in the capillary with a taper angle of 2.2 deg (fig. 5). At zero entrance angle (fig. 5) three peaks are observed in the central part of the distribution. Such a behavior has been revealed for an angular distribution of 240 keV protons transmitted through a cylindrical capillary with a diameter of 0.5 mm and length of 178 mm (fig. 3 in ref. [6]).

The energy spectra of the initial and transmitted protons are shown in fig. 6. The most of ions transmit the capillary without energy loss, however, the low energy tail certainly exists (fig. 6, curve 2). It means that those transmitted ions moving with higher

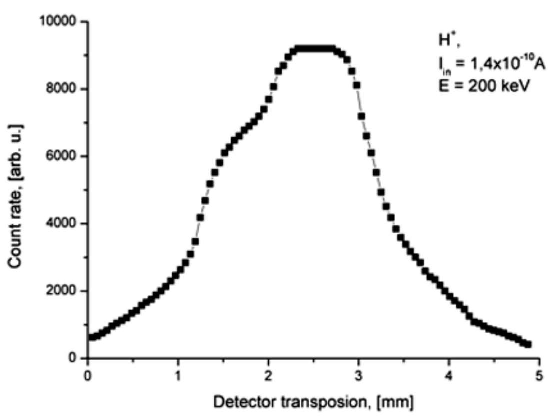

Fig. 4. – Shape and angular distribution of 200 keV protons transmitted through a capillary with a taper angle of 0.5 deg.

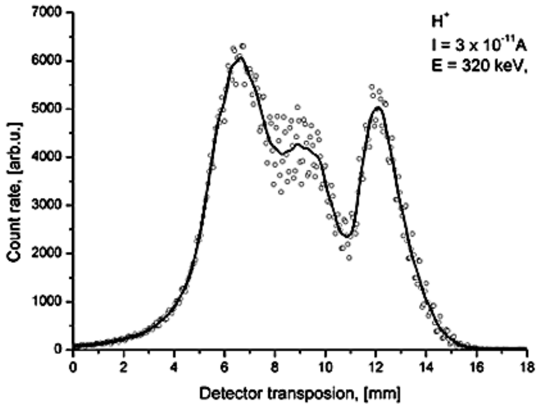

Fig. 5. – Angular distribution of 320 keV protons transmitted through a capillary with a taper angle of 2.2 deg.

transverse energies and suffering the small angle scattering lose their energy interacting with an inner surface of the capillary. It should be noted that these particles cause only a very modest widening of the initial energy distribution (less than 5 to 6%).

Generally speaking, the key to the understanding of these experimental observations is the notion of the two distinctly different time constants for formed charge transport along the surface and within the bulk of capillaries. The interplay between these time constants is responsible for providing a dynamic equilibrium keeping up a Coulomb field large enough to deflect charged particles and small enough not to initiate a Coulomb blockade at the entrance of the capillary.

Proton transmission through nanoporous alumina was also studied. The samples were prepared by two-step anodization of 0.6 mm thick high-purity (99.999%) aluminium foils following the procedure of Masuda and Fukuda [9]. The remaining aluminium layer was

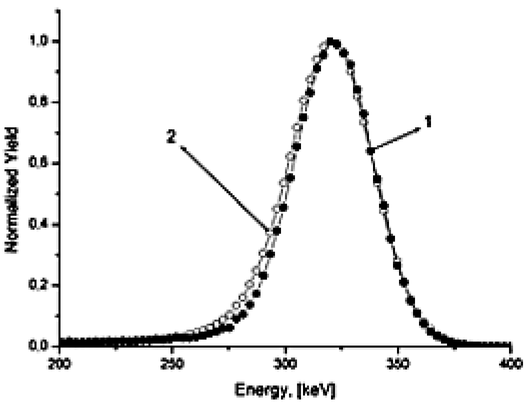

Fig. 6. – Energy spectrum of transmitted protons with an initial energy of 320 keV with and without capillary. The taper angle is 1.7 deg. Curve 1 is for the initial beam and curve 2 is for the transmitted beam.

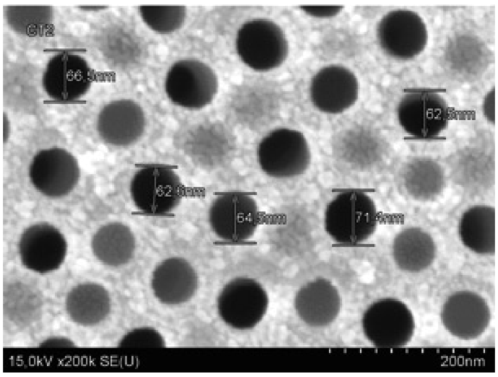

Fig. 7. – The SEM picture of the nanoporous alumina sample from a surface.

removed by saturated $HgCl_2$ solution. Pore ends were opened in a 5% H_3PO_4 solution. The samples were finally cleaned in deionised water. Alumina samples with thickness of $42\,\mu$m were used in these experiments (fig. 7). The diameter of pores was in the range of 63 to 72 nm and the density of pores amounts to $1.2 \cdot 10^{10}\,\text{cm}^{-2}$. The relative surface area covered by pores was about 45 to 50%.

Figure 8 shows a cleaved cross-section of this sample imaged by SEM at different magnifications. As can be seen, the pores extend through the entire depth. The dominant part of capillaries displays a regular and parallel behaviour. However, due to unavoidable irregularities in the nanochannel growing process of anodized Al_2O_3 capillary systems, deviations from the ideal, parallel-arranged structure of the individual capillary, distortions as well intersections are revealed in the examined samples.

Orienting the capillaries parallel to the proton beam was a complicated process in general. Before each set of measurements, the optimum positions were searched by seeking the minimal deviation and maximal transmission of the proton beam in order to find the correct zero point of the title angle scale. Figure 9 shows the angular distributions of protons with the energies of 150 and 320 keV transmitted through untitled capillaries ($\varphi = 0°$). It can be seen that each of both the angular distributions consists of a sharp

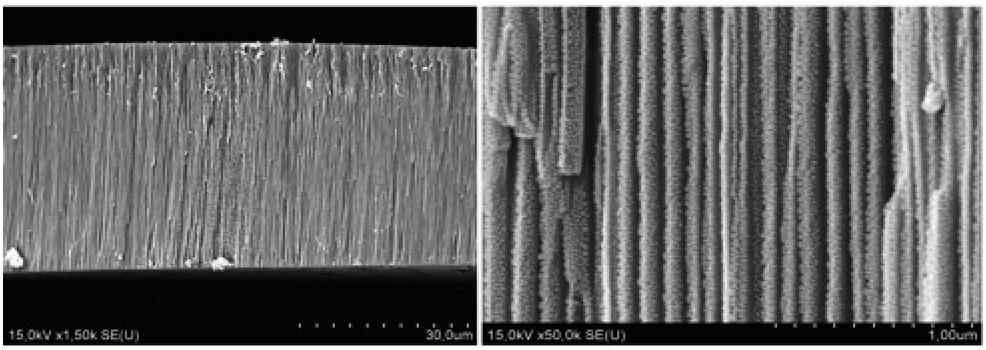

Fig. 8. – The SEM image of a side view of the nanoporous alumina sample at different magnifications.

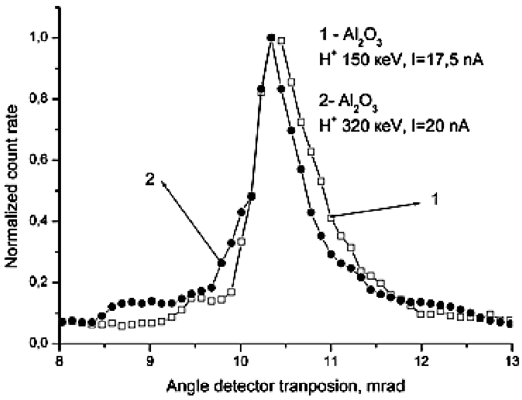

Fig. 9. – Angular distribution of H^+ ions transmitted through capillaries in the Al_2O_3 membrane without metallic coverage at 0° tilt angle: 1–150 keV, $I = 17.5$ nA; 2–320 keV, $I = 20$ nA.

peak and two broad low-intensity peaks disposed symmetrically around the central peak. It should be mentioned that for protons with an energy of 150 keV these low-intensity side peaks are displayed more clearly than for the proton beam with an energy of 320 keV. Moreover, the width of the central transmission peak for low energy protons (fig. 9 (curve 1)) is approximately up to 20% exceeding the angular distribution width for protons with an energy of 320 keV. It should be also noted that the angular width of the ion beam behind the Al_2O_3 membrane is practically equal to the angular width of the initial beam. The tapered glass capillary makes it considerably broader (by a factor of 4 and more). In our opinion, the side maxima in fig. 9 are caused by the above-mentioned non-parallelism of capillaries and structure imperfections (see fig. 8). The ion beam transmission through the Al_2O_3 membrane, defined by integrating the angular distributions, has amounted to about $2 \cdot 10^{-4}$. In order to improve transparency of such a capillary system the inlet and outlet surfaces of a membrane should be covered with a thin metal layer (10 to 20 nm in thickness). It enables us to suppress influences of sample surface charging and increase the transparency coefficient up to a few percents [8]. Non-parallelism of nanocapillaries and other capillary imperfections may also strongly influence the guiding parameters and, therefore, the Al_2O_3 membrane transparency.

The proton beam intensities presented in fig. 9 were measured behind the sample with a thickness of 42 μm that considerably (more than one order of magnitude) exceeds ranges of protons in this material. This is an evidence of the anomalous motion of ions like the hyperchanneling of charged particles along the low-index directions of crystal lattices.

4. – Conclusion

– We have confirmed that a few hundred keV proton beams are successfully focused by the tapered capillary optics.

– The areal density of the transmitted beam is enhanced by approximately 20 times.

– Charging up a face part of the capillary causes the transformation of continuous ion beams to oscillating ones.

– The most of protons (94–95%) in the energy range of 150 to 320 keV transmit the capillary without energy loss.

– Changing a taper angle from 0.5 deg to 1.7 deg evidences increase of the transmission coefficient by more than 300 times keeping the initial energy spectrum of ions.

– Compared with the conventional micro-beam facilities, the usage of tapered capillaries is certainly simple and low-cost, thus providing an interesting technique of submicron RBS or PIXE elemental analyses. Moreover, if the ion species are extended to heavier elements, the present method provides highly local versatile maskless ion implantation technique.

– Angular width of the ion beam behind the Al_2O_3 membrane is practically equal to the angular width of the initial beam. It means that nanochannel alumina is a promising template for pattern transfer by ion lithography.

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