ACTA PHYSICA DEBRECINA XLVIII, 1 (2014)

DISCHARGING OF INNER CAPILLARY WALLS IN ION TRANSMISSION THROUGH NANOCAPILLARIES

Péter Herczku[1,2], Zoltán Juhász[1], Sándor T.S. Kovács[1,2], Richárd Rácz[1], Sándor Biri[1] and Béla Sulik[1]

[1] Institute for Nuclear Research (Atomki), Debrecen Bem tér 18/c
[2] University of Debrecen, Debrecen Egyetem tér 1

Abstract

We carried out systematic experiments for studying the transmitted intensity of 3-keV Ne^{7+} ions through nanocapillaries. The sample was a polyethylene terephthalate (PET) foil with randomly distributed straight capillaries, perpendicular to the foil surface. The sizes of the capillaries were 200 nm (diameter) and 12 μm (length). The transmission during the charging up and discharging processes was measured at different tilt angles. The main goal of the experiment was to clarify the behavior of the discharging process. The data were fitted by three-component exponential decay functions. Our recent results show that the decay of the guided transmission follows a non-linear behavior.

I. Introduction

Ion guiding through nano- and micrometer sized capillaries with insulator walls attracted increasing interest in the last decade [1-4]. This self-organizing process is governed by the charge deposition on the inner capillary walls. The incident ions collide with the inner surface of the capillary, and they deposit their charge, creating electrostatic fields. After a critical amount of charge is deposited, this field governs the ions towards the capillary exit. The reflection of the ions occurs at relatively far from the wall, therefore the charge state of the subsequently incoming ions does not change. When the sample is tilted relative to the incident beam, a charge patch will be formed at the entrance region of the capillary. During its development this patch may become overcharged, i.e. the ions will be over-deflected by the repulsive field, and a secondary charge patch is formed at the opposite side of the capillary. This leads to oscillations in the mean angle and the transmitted intensity of the ions [5]. These damped oscillations usually end up in a constant equilibrium transmission rate. Sometimes the transmission gets completely blocked by the over-charged entrance patch [7,9,10]. The development of a regular transmission or blocking is determined by the surface characteristics.

Most of the experiments [6] show that the efficiency of transmission only weakly depends on the incoming beam current. When the incoming beam becomes 10 times larger, the transmitted current is usually found almost tenfold as well. For sending the same portion of the beam towards the exit, the electrostatic field inside the capillary should not change significantly. This implies that the voltages inside the capillary also do not change remarkably. Consequently, the accumulated charge in the patches at equilibrium does not or only weakly depends on the incoming current. In addition, because of the similar charge pattern, both the vertical and tangential component of the field is expected to be similar to the low current case.

One of the puzzling fact is that when the formed patch on the wall is supplied by a tenfold increased current its charge - and its voltage - only slightly increases. The explanation is that the depletion current from the patch strongly depends on the incoming beam. Namely, if the incoming beam increases, then the depletion current must increase also nearly in the same way. For realizing this, a small increase of the tangential field, i.e., a small increase of the voltage at the charged patch seems to be sufficient, which can be understood if one consider the non-linear conductivity of insulators.

The relation between the voltage of the patch and the depletion current reminds of the current-voltage characteristic of a forward-biased diode. The conductivity of the insulator capillary wall must very strongly depend on the tangential component of the electrostatic gradient. One of the possible models of this behavior was presented in our previous paper [8]. Nevertheless, verification of this model is not easy.

The connection between the charge of the patch and the depletion current can only be studied by indirect methods due to several reasons: the phenomenon is hidden inside the capillary and occurs in a very small area. It is not possible to carry out measurements with any known probe at such small area. Direct measurements on a spacious flat surface would be possible, but the distribution of electric fields could be very different for such a surface and for the case of a curved surface with 100 nm radius. Thus the information gained from studies at large flat surfaces may not be relevant.

At the capillary samples, we can measure the number and the characteristic properties of the in- and outgoing particles (e,g., kinetic energy, charge state, angular spread) as a function of the tilt angle or the total incoming charge. A deeper understanding can be achieved by energy and charge state analysis. One has to keep in mind, that in the case of ions we may obtain information about the net effect of fields inside the capillary, while the outgoing neutral particles (atoms) carry rather direct information about the location, where they were neutralized at the wall.

The scenario is the most complex when the patch is being charged up by the incoming beam. One has to take into account than, that the beam intensity may fluctuate in time and the beam density distribution sometimes also vary. The dynamics of charging up and depletion has statistical features due to both the random distribution of the capillaries, and the fluctuating number of charge carriers entering a single capillary. The measured transmission is modulated by many statistical and systematic effects.

The charges on the surface also move when the incoming beam is stopped. In this case the time development of the charge distribution on the surface is the only undergoing process until we test the transmission with some short beam pulses. The charge of the patches practically does not increase if we let the beam to the sample only for short periods for testing the transmission. From the time dependence of the transmission one can conclude to the time development of the charge migration. Therefore, the depletion process can be more straightforwardly investigated with the reduction disturbing circumstances. In this work we present the results of such indirect, systematic measurements.

II. Experimental method

The experiments were carried out at the beamline of Electron Cyclotron Resonance Ion Source (ECRIS) of Atomki, Debrecen. The highly parallel $^{22}Ne^{7+}$ beam was collimated to 0.5 mm diameter right before entering to the vacuum chamber. The base pressure was less than 1×10^{-6} mbar. The sample was a PET foil, with straight capillaries of 200 nm in diameter and 12 μm in length (i. e. the thickness of the foil). The capillary density was about 1.2×10^{8} cm^{-2}. Both side of the foil was covered by a thin gold layer (about 20 nm), in order to avoid the macroscopic charge up of the sample. These metallic layers provide connection to ground, to where the deposited charges from the capillaries can migrate.

Two-dimensional profiles of the transmitted particles were collected by a position sensitive detector. In order to separate the different charge states in the transmitted beam an electric field was applied in front of the detector. The charging up and the discharging dynamics were measured in different ways. During charging up, frames were exposed with 9-s durations, which were followed by 1-s breaks. The collections of frames were uninterrupted until the transmission reached nearly an equilibrium state. After, the beam was switched off and the discharging dynamics was measured by sampling the transmission by short beam pulses. In an ideal case, one should use infinitely short pulses in order to avoid recharge of the capillaries. Practically it is not possible, but we applied pulses as short as possible.

The sampling method is described as follows: After the transmission reached equilibrium, a new measurement was started with pre-programmed time sequence of data collection without turning off the beam. The sampling pulses were synchronized with the data collection. During the breaks of the measurement an electrostatic signal was generated by the measuring program in order to control the beam, so the beam could not enter the experimental chamber. During acquisition, however, a zero signal was generated and the beam could enter the set up freely. As a first step a 10 s long acquisition of transmitted particles was taken meanwhile the controlling signal was connected to a chopper which controlled the beam. This was followed by a 1 minute long break. After this, the transmission was sampled by 3 consecutive 2-s long beam pulses with 1-s break between them. The first train of sampling pulses was followed by a 2-minute long break and then a subsequent train of sampling pulses was applied. This procedure of acquisition was repeated with increasing break periods between the trains until a measurable transmission was observed. The reason for three consecutive sample pulse was to check whether the transmitted intensity has an increasing tendency during the sampling procedure, which would indicate recharging effects. We found that the values of intensities were matching within the experimental uncertainties. So we concluded that, during the discharging measurement the capillary walls were not significantly recharged.

III. Results

Some of the measured discharging dynamics of ion transmission can be seen in figure 1. The data were taken at different tilt angels, which are indicated in the figure. It is clearly seen that the maximum of transmitted intensity is decreasing with increasing tilt angle. From the angular dependence, the guiding power of the sample can be determined. The guiding power (Ψ) is identical with the tilt angle, where the transmitted intensity is dropped to its 1/e fraction [1,2]. It is an important characteristic of the sample. The experimental data were taken according to the same timing scheme of the sampling procedure at each angle. This way we could evaluate the time dependence of the "effective guiding power" in the discharging dynamics too, but these results are not presented here. For fully charged up sample in equilibrium we found the guiding power to be $\Psi = 4.5°$.

The time dependence of the decreasing transmission provides indirect but useful information about the charge migration in the first charge patch close to the grounded metal layer, which mainly determines the transmission. If the discharge current would be a linear function of an effective voltage of the entrance patch (if Ohm's law were applicable here), the deflecting field would drop exponentially. Since the capillaries are approximately cylindrical, the drop of the transmission should be *faster* than an exponential drop (except the very beginning of the discharging process), and the transmission should drop to zero within a finite time. Therefore, in a linear (time) - logarithmic (transmission) graph the decrease of the transmission should

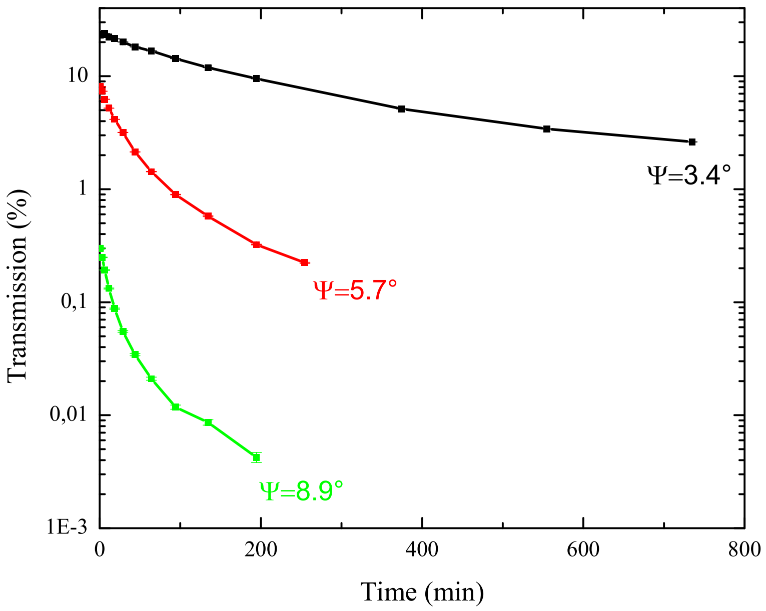

Figure 1: (Color online) The decrease of transmission of the guided $^{22}Ne^{7+}$ ions at $\Psi = 3.4°, 5.4°$ and $8.9°$ tilt angles as a for the PET sample function of time after the incident ion beam is switched off. The starting points are the equilibrium transmission values.

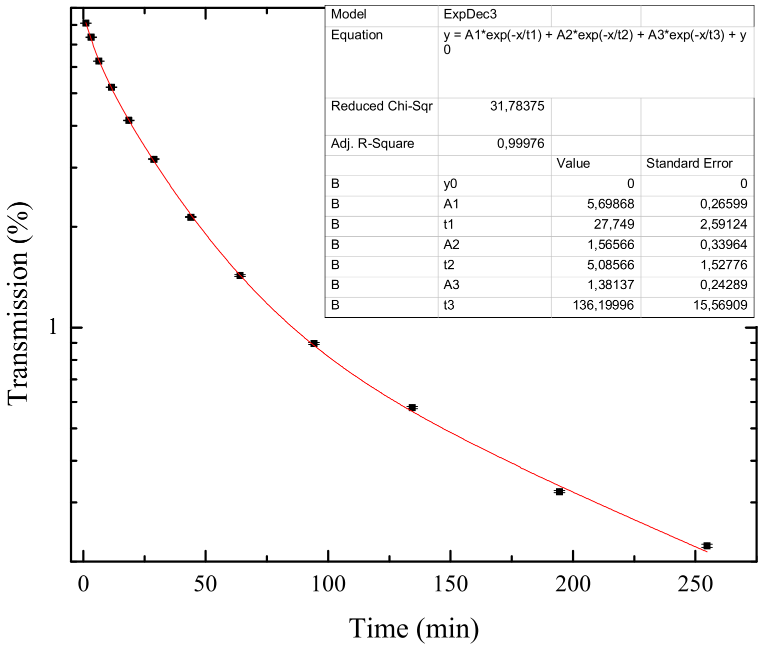

Figure 2: (color online) Decay of the transmission at $\Psi = 5.7^{\circ}$ tilt angle. Dots: experiment; line: fitting by a 3-component exponential model function. The direction of the deviation from a single exponential decay suggests an open-diode type, strongly non-linear dependence of the depletion current on the effective voltage of the first charge patch (see text).

deviate from a straight line what could be expectedfor experimentally decaying transmission. Rather, it should appear more and more below the straight line as the discharging time increases. With other words, for a linear system the logarithm of the transmission should be a convex function of time.

Figure 2 clearly shows that it is not the case. The deviation from an exponential decrease (linear in the lin-log scaled graph) is just the opposite what we expect for a linear discharge of the first patch. At the beginning of the discharge (in the first 60 minutes) the decay of the transmission is fast, but then it slows down significantly. After 4 hours, the transmission is still well measurable. It can be seen, that the decay is monotonically decreasing in the whole studied region, but the rate of the decrease is slowing down with time. Without model considerations, the data was fitted by a tree-component exponential decay function and a good matching was obtained resulting in three significantly different characteristic time constants (5, 28 and 136 min). Further analysis is needed for the identification of the possible processes maybe associated with the three term in the model function.

VI. Conclusion

We measured the time dependence of the intensity of ions transmitted through PET nanocapillaries. Every measurement was started with the charging up of the capillaries, which was followed by sampling the transmission during the discharging of the capillaries. The aim of these experiments was to quantitatively determine the decay of the transmission, and consequently learn about the discharging of the first patch inside the capillary. We found that - after switching off the beam - the guided transmission monotonically decreases with time in such a way that the logarithm of the transmission is a concave function of time everywhere. All the measured data can be well fitted with three-component exponential decay functions. After a few hours, the decay of the transmission becomes extremely slow. This finding is in agreement with the proposed nonlinear model of surface conductivity presented in [3]. The estimated electric field strength in the capillaries is in the non linear regime for conductivity, which gives further support for the model. However, the role of surface and bulk conductivity associated with different characteristic time constants for discharge, which was presented in a linear model [4,6] cannot be ruled out by the present results.

V. Acknowledgement

This work was supported by the Hungarian OTKA Grant no. K83886, as well as by the TÁMOP 4.2.2.A-11/1/KONV-2012-0036 project, co-financed by the EU and the European Social Fund.

References

[1] N. Stolterfoht, et al Phys. Rev. Lett. **88**, 133201 (2002).

[2] N. Stolterfoht, et al Surf. & Coat. Techn. **196**, 389 (2005).

[3] N. Stolterfoht, et al Phys. Rev. A **87**, 012902 (2013).

[4] K. Schiess, et al Phys. Rev. A **72**, 062902 (2013).

[5] N. Stolterfoht, et al Phys. Rev. A **79**, 022901 (2009).

[6] C. Lemell, et al Progr. Surf. Sci. **88**, 237-278 (2013).

[7] P. Herczku, et al Acta Phys. Debr. **XLVI**, 57 (2012).

[8] P. Herczku, et al Acta Phys. Debr. **XLVII**, 67 (2013).

[9] Z. Juhász, et al Nucl. Instr. and Meth. B **279**, 177-181 (2012).

[10] N. Stolterfoht, et al Phys. Rev. A **88**, 032902 (2013).