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SURFACE CONDUCTIVITY AND CHARGE MIGRATION ON THE INNER CAPILLARY WALL

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Abstract

We performed experiments to study the role played by the surface conductivity in the development of guiding of slow highly charged ions through nanocapillaries. At present, the related open questions in the literature are: What is the effect of charge migration on the transmission of ions? What happens after the charging up process when we stop the charge deposition? In what extent are the processes governed by linear and non-linear terms? Our recent results supports that the processes follow a dominantly non-linear law.

I. Introduction

In the last decade the phenomena of ion guiding through nano- and micrometer sized capillaries attracted increasing interest [1-6]. These capillaries are located in highly insulating materials. Such materials can be polyethylene terephthalate (PET), polycarbonate (PC), Al_2O_3 or SiO_2. The ion guiding is a self-organizing process, which is governed by the charge deposition on the inner capillary wall. The incident ions collide with the inner surface of the capillary where they deposit their charge in a way to build up an electrostatic repulsive field (see Fig. 1). After a critical amount of charge

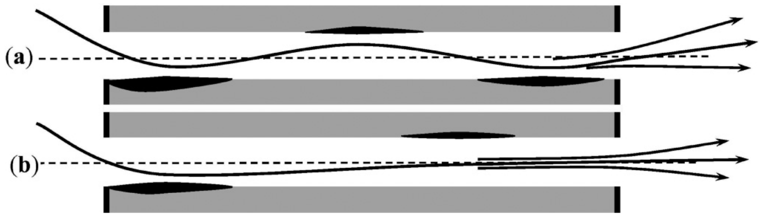

Figure 1: The development of the guiding through a capillary of insulating material. In panel (a) a typical path of the ion in the dynamic stage of the development is shown. In panel (b) the equilibrium transmission is demonstrated. The black areas indicate the charged patches. During the development only the first patch remained significant, most of the other, temporary patches lose their importance.

is deposited, this field will be able to deflect the ions towards to capillary exit. This deflection occurs at relatively far from the surface therefore the charge state of the later incoming ions will not change. If the sample is tilted relative to the incident beam, a charge patch will be formed in 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 will be formed on the opposite side of the capillary. This leads to oscillations in the mean angle and the transmitted intensity of the ions. These damping oscillations usually end up in a constant equilibrium transmission rate.

II. Theory

Recently, N. Stolterfoht published simulations and analytic calculations to study the role played by the surface conductivity in the development of the guiding mechanism [7]. In the simulations he considered, that the positively charged incident ions, which collide with the surface of the inner wall of the capillary, deposit all of their charge in one step. In fact, they are neutralized due to multiple electron capture. This way they make "holes" in the surface. These holes have effective positive charges. At the insulator surface, or inside the insulator, the holes can move only on a jump by jump basis. The deposited charge migrates by this hopping mechanism.

The hole-motion velocity is determined by the electric field, **E**, which is formed by the deposition itself. The electric field can be determined from the electrostatic potential, which is due to the deposited charges. It can be written as follows:

$$V(\mathbf{r}) = \sum_{i}^{n} \frac{q_{sc}}{(|\mathbf{r} - \mathbf{r_i}|^{\mathbf{2}} + \mathbf{r_c^2})^{\mathbf{1/2}}} \tag{1}$$

Here $\mathbf{r}_i$ is the position vector of the ith deposited charge on the surface, and q$_{sc}$ is the (uniform) deposited charge. The r_c term was introduced to avoid the singularty of Coulomb potential. For the moving ions, within this field the Newtonian equation of motion is solved in the next form for the ions trajectories:

$$m\frac{d^2\mathbf{r}}{dt^2} = q\mathbf{E} \tag{2}$$

Here the $\mathbf{r}$=(x,y,z) is the position vector (expressed in descartes-coordinates) of the ion, m is the mass of the ion, $\mathbf{E}$(x,y,z) is the electrostatic field, the gradient of $V(\mathbf{r})$ of Eq. (2). When as a result of (2) the calculated trajectory has an end in the inner capillary wall, then the corresponding ion will deposit its charge. According to Eq. (2), this deposition will change the electric field, so $V(\mathbf{r})$ and $\mathbf{E}$ should be recalculated for determining the trajectory of the next ion. Since the charge migration has significant importance in the formation of charged patches we have to take this into account, when calculating the electric field. From this point of view the drift velocity of holes is important. To the drift velocity one can obtain the formula:

$$v_s = \mu\ \mathbf{E} exp\left(\sqrt{\frac{E}{E_c}}\right) \tag{3}$$

Here μ is the hole mobility, E_c is a characteristic field governing the exponential increase of the surface current. This implies two regions for the velocity, where $E \ll E_c$ the velocity follows a linear field dependence and for $E \geq E_c$ it is determined by a nonlinear term. A similar expression can be derived for the surface conductivity, which determines the motion of holes. It follows similar nonlinear expression which calculated by Frenkel [8].

$$\sigma_s = \sigma \ exp\left(\sqrt{\frac{E}{E_c}}\right) \tag{4}$$

Here σ is the surface conductivity for $E \rightarrow 0$. E_c is a characteristic field governing the exponential increase of the surface current. Experiments qualitatively show the signatures of this nonlinear behavior. The fact that the transmission is only weakly depends on the current density, strongly supports a nonlinear model. Nevertheless, the charging up dynamics depends on many other parameters too, and the values of σ and E_c cannot be extracted from the measured data easily. Without an input current, the equilibrium charge arrangement in the capillary slowly decays, and this process is governed only by the conduction mechanisms at the surface. We note that bulk conduction effects are negligible compared to surface processes. In the present work, we measure the time dependent decrease of the transmission, after switching off the ion current. Of course, from time to time we need to let the current on the sample for a short period, to measure the actual value of the transmission.

III. Experimental method

The experiments were performed at the beamline of Electron Cyclotron Resonance Ion Source (ECRIS) of Atomki. The extracted 3 keV kinetic energy Ne beam was collimated to 0.5 mm before entering into the vacuum chamber. The background pressure was about $1 \times 10^{-}$ mbar during the experiments. The experimental setup in the case of the time dependence measurement of the transmitted intensity can be found in ref [9]. Here only a brief description is presented: We collect 2-dimensional transmission profiles [9] with a position sensitive detector. In front of the detector we apply an electrostatic deflection field, which separates the transmitted ions and neutrals. In a charging down experiment, we are interested in the time dependence of the transmission rate due to the spontaneous discharge of the inner capillary wall without ion beam. The problem is that from time to time we need to measure the transmission by a current pulse, which charges up the capillary wall again. In an ideal charging down experiment, one should use infinitely short current pulses. In a real experiment, the length of the current pulses is of course finite. For keeping it short, we synchronized the switching on and off of the beam and the data acquisition time. The beam was on the sample only during the measurement of the transmission. The algorithm of the measurement is as follows: After the charging up process (equilibrium transmission) we turn the beam off for 1 minute, and then we sample the transmitted ion yield by 3 consecutive pulses of ion current, 1 second duration each, with 1 second breaks between them. This way, we measure the transmission due to the remaining charges on the capillary wall. In addition, the three consecutive pulses provide information about the distortion of the discharge effect due to charging up by the sampling current pulses. The next group of sampling pulses is applied after 2 minutes break, and then the breaks between the test pulse trains increase in time. Our sample is a PET foil which consist capillaries wit diameter of 170 nm and 12 μm length. The aspect ratio is about 1:70. The density of the capillaries is about 10 cm^{-2}. To avoid the macroscopic charging up effect both side of the sample were covered with a thin gold layer (about 20 nm).

IV. Results and discussion

In the present work we measured the transmitted intensity of Ne ions during both the charging up and charging down periods. A typical transmitted intensity as a function of time can be seen in figure 2. The deposited charge, measured on the surface of the sample is increasing nearly linearly with time. Therefore to avoid the beam fluctuations and any disturbing effects we plotted the transmitted intensity as a function of deposited charge. At the initial stage of the curve the oscillations, belonging to the transient formation of the charged patches can nicely be seen. When the first patch is overcharged, the transmission will decrease because a large part of the ions will impinge to the opposite side of the capillary. When this secondary patch has been charged up, the transmission increases again. We note that not only one capillary is included in this process, and the statistical scatter of them somewhat damps the oscillations.

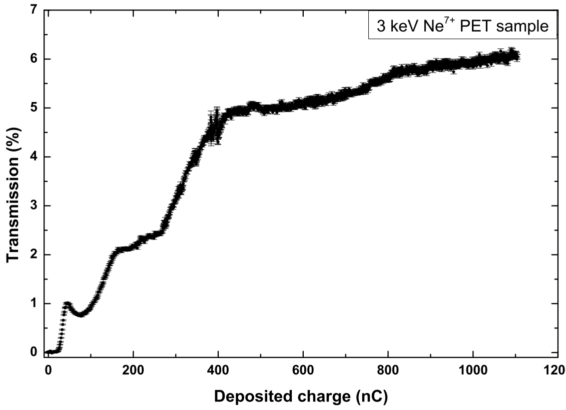

Figure 2: The time development of the guided transmission of 3 keV Ne ions at Ψ=3° tilt angle as a function of deposited charge in the case of a PET sample. According to the ion guiding scenario, when a new charged patch is formed, we see an oscillation on the leading edge of the transmission profile. Later these secondary patches lose their importance and an equilibrium transmission is formed.

After the dynamic development period, the transmission achieves equilibrium. In this regime, the incoming current, the speed of charge deposition, the charge migration from the patch to the metal layer and the transmitted current are in equilibrium. In this regime, without significant fluctuation of the incoming beam, the transmission becomes stable.

The damping oscillation of the mean angle of the transmission is shown in Fig. 3. The dynamics is similar to that presented and analyzed in Ref [10]. It is nicely seen that a charge patch formation corresponds to a half period of angular oscillation and a full period of intensity oscillation. For studying the charging down process, we applied three short pulses (with 1 sec. duration) in every sampling point to test the transmission of ions through the capillaries. We found that the measured transmission data ”triplets”, belonging to the three test shots, are matching within 15 percent to each other. Moreover, they did not show any monotonically increasing pattern. We concluded that the charge arrangement was not significantly modified by the test current pulses, so they practically did not change the

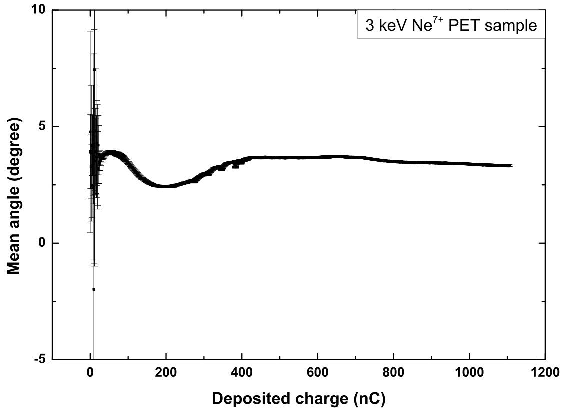

Figure 3: The oscillation of the mean angle of 3 keV Ne ions around Ψ=3° tilt angle in the case of PET sample as a function of deposited charge. According to the ion guiding scenario, the oscillation follows the formation of the charge patches. Note that the frequency of the angular oscillation is around the half of that for the intensity. In equilibrium, the mean angle of the transmission is equal to the tilt angle.

field inside the capillary. The average transmission data as a function of the decay time are shown in Fig. 4.

The plot of an exponential function in a semi-logarithmic scale is a straight line. One gets such a straight line, e.g., if a capacitor is discharged via a linear resistor, and its voltage is displayed. If the conductance of the discharging resistor depends on the voltage, the resistor is nonlinear, and the time dependence will deviate from the pure exponential decay. In Fig. 4, it is clearly seen that the transmission decays fast when the guiding field is closed to its equilibrium value. This corresponds to a maximum charge in an equivalent capacitor, and a maximum value of an equivalent voltage. According to Eq. (4), the effective conductance of the surface is also large here. With the developing discharge process, the effective voltage decreases. It follows from Eq. (4), that the nonlinear surface conductance also decreases. Accordingly, the decrease of the effective potential, the corresponding guiding field and, in turn, the guided transmission slows down as the discharging process develops. This effect is clearly seen in Fig. 4. At

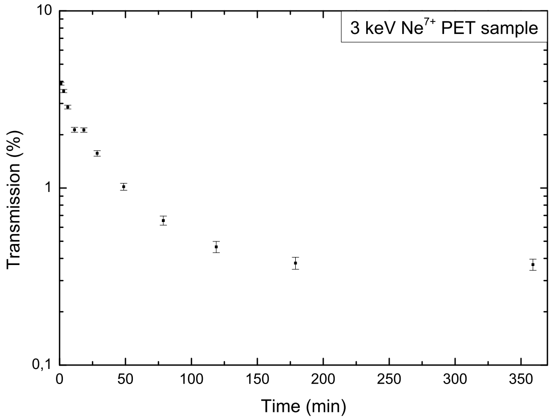

Figure 4: The decrease of the guided transmission of 3 keV Ne ions at Ψ=3° tilt angle in the case of PET sample as a function of time without a continuous incoming ion current. The starting point was the equilibrium state of the system. The method of the measurement is detailed in the text.

the beginning of the discharging process (in the first 125 minutes), the decay of the transmission is fast, while after 5 hours, a finite transmission is still measurable, and its decay gets extremely slow. It is seen that the change of the speed of the discharge is monotonically decreasing in the whole studied time region. This behavior is in agreement with the nonlinear conductance picture of Eq. (4). Of course, in reality, the "capacitors" and "resistors" inside a capillary is not just an RC term, they form a distributed "network", but the linearity of it does not depend on its complexity. Moreover, the measured transmission is not identical with an effective voltage. Nevertheless, the strong deviation of the decay curve of Fig (4) from an exponential pattern supports that Eq. (4) is a correct model of the nonlinear surface conductivity at the capillary surface.

V. Conclusion

We measured two dimensional transmission profiles for studying the role played by the surface conductivity in the formation of charged patches on the inner capillary wall. We found that the velocity of the decay of the guided transmission monotonically decreases with time after switching off the input current. This behavior is in agreement with the nonlinear conductance model of Eq. (4), where the characteristic field along the surface governs the speed of the charge migration.

Acknowledgement

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