Discharging dynamics of insulator surfaces irradiated by highly charged ions

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Abstract. Guiding and focusing of keV to MeV ions by insulator micro-capillaries offer exciting perspectives for the production of low divergence micro-sized beams and the spatial control over the irradiated zone. These effects result from the local charging of the capillary inner wall and depend strongly on the charging/discharging dynamics of the insulating material. This dynamics has been studied on various glass insulator surfaces (borosilicate, fused silica and quartz) by grazing incidence highly charged ion beams. We propose simple experimental methods to derive the relevant time constant and study the influence of temperature and different material properties to the charging process.

1. Introduction

Bombardment of insulator surfaces by highly charged ions can be used for the production of nano-structures, opening perspectives for nano-fabrication \cite{1,2}. Besides improving our understanding of the processes leading to material modification, it is equally important to be able to guide the highly charged ions at the desired spot. In this context, recent results on guiding and focusing of highly charged ions by taper-glass micro-capillaries \cite{3,4} appear to be a very promising solution. The guiding, which was previously observed in PET nanocapillaries, was explained by the self-organizing process of the inner wall charge up \cite{5}. Since the ion trajectories are nearly parallel to the inner walls, ions are focused in the tapered glass capillaries due to its geometry. However, a more detailed physical description is needed to better understand the guiding and focusing effects, and choose the best candidate material.

The tubular geometry is not necessarily optimum to access the relevant parameters such as surface and bulk diffusion of charges, secondary electron emission etc., governing space and time distribution of charges in a capillary. This task is best achieved by grazing incidence bombardment of a planar insulator surface by highly charged ions, since the primary mechanisms should be common. Such a geometry also offers a possibility to directly measure the above-mentioned material parameters and better understand the behavior of appropriate micro-capillaries.

We describe two simple methods for measuring discharge dynamics of insulator surfaces at different temperatures charged up by highly charged ions. The influence of the sample history (target
annealing or change of its properties due to the ion bombardment) is also observed [6]. The results give valuable information to guide future experiments.

2. Measurement procedures

The insulator samples were prepared by prolonged heating at temperatures up to 600 K (phase separation, crystallization or a phase transition, in the case of quartz, might occur at higher temperatures) in UHV conditions for several hours. Sputter-cleaning was not employed to avoid preferential sputtering effects. The details of the experimental setup are given elsewhere [6] and its scheme is given in figure 1. The primary ion beam is extracted from an ECR ion source, mass analyzed and collimated before impinging the insulator surface with incident angle $\psi$ between $0.5^\circ$ and $1.5^\circ$. The sample is progressively inserted into the beam so that part of the beam hits the surface while the rest flies over the surface without touching it and directly hits a position sensitive detector (PSD) located downstream. The quantity of ions impinging the surface is proportional to the intensity of scattered (mostly neutral) particles. The projectiles hitting the surface contribute to its charging by formation of electronic holes. Consequently, the surface charges repel the direct beam (thus reducing the rate of ions impinging the surface $\Phi$), and its deflection is monitored on the PSD (figure 1). This way, the ion beam is used for surface charging and monitoring of the charging process at the same time. Diffusion of holes into the bulk also takes place during the charging up, leading to surface discharging. Equilibrium deflection of the direct beam is achieved after some time. It is defined by the equality between the charging rate (proportional to $\Phi$) and the discharging rate. Once this steady state is reached, the beam is turned OFF to allow self-discharging of the surface. The discharging dynamics can be then determined by turning the beam ON for few seconds at regular time intervals to track the time evolution of the average direct beam deflection (here denoted as the beam deflection). The beam deflection decreases in time, which corresponds to surface discharging. This procedure can be used to determine discharging dynamics as a function of temperature.

![Figure 1. Experimental scheme and typical PSD images at the beginning and at the end of the charging process.](image)

If the surface discharging is too fast to be monitored by this method, an alternative approach has to be developed, providing information on temperature dependence of discharging dynamics. As it is shown in the next section, the observed discharging time decay is exponential, so we approximate the discharge rate by $Q/\tau$, where $Q$ represents the overall charge in the subsurface region and $\tau$ is discharge time constant. Hence, the time evolution of $Q$ can be described by the following equation:

\[
\frac{dQ}{dt} = \Phi \cdot (q + \gamma) - \frac{Q}{\tau}
\]

where $q$ is the ion charge and $\gamma$ is the secondary electron yield per single ion. The proposed model is just an approximation since the surface distribution of charges is not considered. Nevertheless, it gives access to the dependence of $\tau$ on the sample temperature $T$, under the assumption that the direct beam deflection is directly proportional to $Q$. The equilibrium beam rate $\Phi_{eq}$, for which a constant deflection (i.e. approximately constant $Q$) is achieved, can be determined as a function of $T$. Then, since $dQ/dt = 0$ in the steady state, the following relation stands: $\tau(T) \sim 1/\Phi_{eq}(T)$. Here we assume that $\gamma$, which strongly depends on surface charge (which reduces electron emission), does not depend significantly on $T$ in the range of interest (typically 300 – 400 K). The explained approach is denoted here as dynamic equilibrium approach.
3. Results and discussion

The proposed methods for measuring the discharge dynamics are illustrated by examples of three different insulators of interest in tapered micro-capillary production: fused silica, quartz and borosilicate glass. The following ions were used in the experiments: H\(^{+}\), Ar\(^{+}\), Ar\(^{4+}\), Kr\(^{9+}\), Xe\(^{12+}\), He\(^{+}\), and He\(^{2+}\). The primary ion energies were in the range of 1-2.5 keV per charge.

The time dependences of the Ar\(^{4+}\) beam deflection from fused silica for two different temperatures are presented in figure 2. The origin of time corresponds to the beginning of the surface charging. Clearly, a temperature difference of 100 K produces a change of the discharge time constant by two orders of magnitude. The possible reason is the increased hole mobility – the residence time in the hole trap is decreased due to thermal activation. Besides, two slopes are visible in the case of higher temperature. As observed in [6], the time constants significantly depend on the sample history: they can be increased by as much as one order of magnitude if the measurement is done on a previously irradiated part of the surface. This fact, together with existence of two slopes in some cases, hinders the determination of \(\tau(T)\). Decrease of \(\tau\) with the incident angle in the investigated range (0.5°-1.5°) is also observed, which possibly implies influence of the charge flux. On the other hand, influence of the ion charge state is not observed [6].

![Figure 2](image1.png)

**Figure 2.** Time dependence of the Ar\(^{4+}\) beam deflection ions from fused silica at temperatures: a) \(T = 450\) K and b) \(T = 550\) K.

![Figure 3](image2.png)

**Figure 3.** Time dependence of the beam deflection of H\(^{+}\) ions from quartz surface. The charging experiment has been performed after 20 h annealing of the sample at \(T = 780\) K.

The single crystal quartz sample has the same composition but quite different structure compared to amorphous fused silica. Its behavior in terms of discharging is completely different. In the first experiments, it was not possible to charge up as–received quartz. However, after a long annealing (20 h at 780 K, which is still below the phase transition temperature at 870 K) it was charged up with H\(^{+}\) beam at room temperature. The discharge was very slow as can be seen from figure 2, which shows the discharge dynamics. A possible explanation of the observed behavior can be related to the fact that the hole traps in fused silica are defects [7], which should have low concentration at the quartz surface. Hence, while the majority of holes quickly migrates into the bulk, the small fraction is slowed down by filling the traps (this is coherent with the different activation energies for hole mobility – 0.16 eV for quartz and 0.6 eV for fused silica). Then, after annealing, the surface concentration of defects
could be significantly increased by their segregation from bulk [6], so the discharging is significantly slowed down.

In contrast to fused silica and quartz, the former measurement procedure for the discharge dynamics could not be applied to the borosilicate sample (\(\text{SiO}_2(80.9 \%)\), \(\text{B}_2\text{O}_3(12.7 \%)\), \(\text{Al}_2\text{O}_3(2.3 \%)\), \(\text{Na}_2\text{O}/\text{K}_2\text{O}(4.1 \%)\)) since the discharging was too fast. However, the electrical properties of alkali doped borosilicates are mainly governed by the alkali ion mobility [8]. The dynamic equilibrium method, described in section 2, was applied to determine the temperature dependence of the discharge time constant. Assuming that \(\tau = \tau_0 \exp(E_a/kT)\), where \(E_a\) is activation energy for mobility of charges and \(k\) is Boltzmann constant, \(E_a\) can be determined from the slope of \(\log(1/\Phi_{eq})\) vs. \(1/T\) (figure 4). In this experiment 22.5 keV Kr\(^{9+}\) ion beam was used. The obtained value of 0.48 ±0.04 eV is close to the activation energy for migration of Na\(^+\) ions bound to non-bridging oxygen atoms in borosilicate glass (0.55 eV) [9]. In other words, it seems that surface discharging is not related to migration of holes, but of sodium ions [6].

4. Conclusion

Two simple experimental methods are proposed to measure the discharging dynamics of insulator surfaces previously charged by grazing incidence impact of highly charged ions. They can be used to determine characteristic time constants and the activation energy of the discharging process. To our knowledge, these magnitudes, providing macroscopic picture of the discharging process, has not been measured till now. On the other hand, measurement of surface charging properties on the nano scale is known for some time [10]. The presented experimental methods propose a link between the nano-scale approach, which provides fundamental information about the charging/discharging processes, and a specific technological problem such as ion guiding through insulator capillaries. Therefore, direct comparison of these two approaches would be valuable for better tailoring of taper-glass micro-capillaries and their future applications for highly charged ion guiding and focusing.

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