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# Image charge force of keV ions through insulating nanocapillaries

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# Image charge force of keV ions through insulating nanocapillaries.

Previous title: What is wrong with the image charge force of keV ions in insulating nano-capillaries ?

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(Dated: January 20, 2023)

In nano-capillaries of large aspect ratio, the attractive image charge force is strong enough to affect the trajectory of ions passing through capillaries and consequently to diminish the fraction of transmitted beam ions. We calculated the theoretically transmitted fraction, using an expression of the image charge force valid in the case of a static ion and an infinite cylindrical dielectric interface. When comparing the theoretically transmitted fraction to available experimental data for nano-capillaries with an inner diameter of less than 200 nm, we found a surprisingly large disagreement, i.e., the theoretically transmitted fractions were easily an order of magnitude lower than the experimental ones. Noting that the image charge force depends on the velocity of the ion via the frequency dependent relative permittivity of the insulator, we investigated whether the disagreement could be lifted using a velocity depend image charge force. We give the exact expressions of the dynamical image charge force for a plane and cylindrical dielectric interface as a function of the ion velocity. We then re-evaluated the theoretically transmitted fractions in the case of SiO<sub>2</sub> and PET dielectric interfaces. Our findings are discussed in the light of the available experimental data.

## I. INTRODUCTION

Guiding of low-energy ions by insulating capillaries was first reported by Stolterfoht *et al.* in 2002 for nano-capillaries [1] and later by Ikeda *et al.* [2] for macroscopic glass capillaries. Their experimental results showed that even though the capillaries were tilted with respect to the beam axis such as no geometrical transmission was allowed, the beam was steered after an initial charge-up phase through the insulating capillary by self organized charge patches. As a result, a part of the beam could be transmitted, with the ions keeping their initial charge state, indicating that the ions never touched the inner wall. Those pioneering results triggered numerous experimental and theoretical studies allowing a better understanding of the guiding process of keV ions through insulating capillaries [3, 4]. For an overview, the reader may consider [5] and [6]. By combining available experimental data with theoretical predictions, insulating capillaries become a formidable tool to explore the dynamics of excess charges in insulators in the presence or absence of charge injection. Indeed, the time-evolution of the rate, energy and angular profile of the transmitted ions may give indirectly information about the relaxation process of excess charge in insulating capillaries and even about the charge injection process by ion-surface collisions.

Over time, models for the charge dynamics in insulating capillaries were contentiously refined in order to improve the description of the charge injection and charge relaxation in insulating capillaries [7–12]. In particular, the electric field inside the capillary was calculated precisely by considering appropriate boundary conditions at the inner and outer surface of the capillary [9]. In recent models for glass macro-capillaries, the charge injection at the inner surface term accounts even for secondary electrons emitted by ion-surface collisions and subsequently

re-absorbed by the surface [11]. In nano-capillaries, the bulk and surface currents were even considered to depend non-linearly on the electric field as the electric field in nano-capillaries exceeds easily the keV/mm [8]. Eventually, the image charge force acting on the beam ions inside nano-capillaries was added to the dynamics to explain the observed beam-shaping of transmitted ions through muscovite mica nano-capillaries with rhombic and rectangular section [13, 14]. Indeed, when an ion approaches a dielectric interface separating two different dielectric media, the induced surface polarization charges attract the ion toward the interface. In nano-capillaries, the attractive image charge force was shown to be strong enough to sensibly affect the trajectory of passing ions and consequently the fraction and angular distribution of transmitted beam ions.

In this work, the author proposes to have a closer look at the modeling of the image charge force acting on beam ions through cylindrical nano-capillaries. In particular, we want to investigate whether experimentally observed transmission rates through insulating straight nano-capillaries are well reproduced using the expression of the image force inside a cylindrical dielectric. Several experimental studies reported the transmission rates of keV ions through insulating nano-capillaries [4, 13, 15, 16]. Here, we are mostly interested in those cases where a uniform keV ion beam was injected at zero tilt angle and for which the measured transmitted fraction, defined as the transmitted current divided by the injected current, was given. These experimental conditions constitute an ideal setup for which the transmitted fraction can be calculated analytically as the ion trajectories are dominated by the image charge force. Indeed, as will be discussed further in the text, for a uniform beam, aligned with the capillary axis, the charge deposited in the capillary is axisymmetric, yielding an inner radial

electric field that is negligible compared to the image charge force. As a result, ion beams through zero-tilted nano-capillaries are a formidable tool to investigate the image force of ions moving along a cylindrical dielectric interface.

The paper is organized as follow. In section II, we propose and discuss a closed and accurate formula that fits the exact image force of a charge sitting inside a cylindrical dielectric interface. We then calculate the theoretical transmitted fraction in the case where the injected beam is aligned with the capillary axis. Next, we highlight the discrepancy between the theoretical and the experimental transmitted fractions found in the literature. That finding made us question the validity of the expression of the image charge force used here. In section III we propose to improve the expression of the image charge force by explicitly taking into account the velocity of the ion and the frequency dependence of the relative permittivity of the capillary. We then calculate the image charge force as a function of the ion velocity and discussed its influence in the case of an arbitrary dielectric response function. In section IV we re-evaluated the theoretical transmitted fractions using this time the velocity depend image charge force, mainly for those cases for which experimental data is available in the literature. We discuss then in how far the velocity dependent image charge force lifts the initially mentioned discrepancy.

## II. THEORETICAL AND EXPERIMENTAL TRANSMITTED FRACTIONS

The ion trajectories through a capillary is affected by two fields, the electric field generated by the accumulated charges in the capillary wall and by the image force. First, we show that in the case where a keV ion beam is injected with zero tilt angle into a straight insulating nano-capillary, the transmitted beam fraction is mainly controlled by the image force, while the deposited charge has no notable effect on the transmission.

Indeed, for a beam aligned with the capillary axis, the deposited charge distribution has axial symmetry, generating an axisymmetric electric potential  $V(\rho, z)$ . Such a potential yields an inner electric field that competes with the image force only if the accumulated charge density in the bulk exceeds a certain amount. But because the insulator cannot withstand an electric field larger than the dielectric strength field, typically 50 kV/mm [17], without undergoing electrical breakdown and becoming electrically conducting, the accumulated charge in the capillary wall is limited. We performed simulations with our numerical code InCa4D [10] where we followed the trajectories of 7 keV ions through a SiO<sub>2</sub> nano-capillary. Expression (6), which will be discussed later in the text, was used for the image force. We monitored the ion transmission for several hours ( see black curve in figure 1) and found that the transmission is stable in time and neither increased or reduced by the deposited

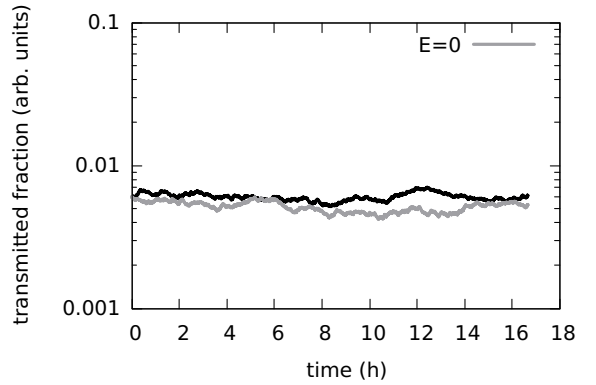


FIG. 1: Simulated transmitted beam fractions (black curve) through an insulating nano-capillary. The gray curve, for which the electric field due to deposited charges was removed, has been added for comparison.

self-organized charge. Details of the calculations are presented in Appendix VI A. Simulations confirm that the effect of the deposited charge on the transmission is negligible and that it is not possible to charge an insulating nano-capillary to the necessary amount to generate an inner electric field that competes with the image force. We can thus safely assume that the transmitted fraction is exclusively controlled by the image force.

### A. Image charge force at a cylindrical dielectric interface

Let us assume a translationally invariant cylindrical dielectric interface of radius  $R$ , separating the inner vacuum from the outer insulating medium of dielectric relative permittivity  $\epsilon_r > 1$ . A charge  $q$  is located inside the cylinder at position  $\rho < R$ , where  $\rho$  is the distance from the symmetry axis Oz. The modeling of the image force acting on the charge  $q$  is described in details in [18, 19] and [20]. Using cylindrical coordinates  $(\rho, \theta, z)$  and introducing the dimensionless radial component

$$\varrho = \rho/R \quad ,$$

the resulting force is given as a sum over angular moments  $m$  and dimensionless wave-numbers  $k$ ,

$$\vec{F}_{\text{im}}(\varrho) = \frac{q^2}{16\pi\epsilon_0 R^2} \sum_{m=0}^{\infty} \int_0^{\infty} dk A_m(k, \epsilon_r) \frac{\partial I_m^2(k\varrho)}{\partial \varrho} \vec{u}_\rho \quad , \quad (1)$$

where  $\vec{u}_\rho$  is the radial unit vector,  $I_m()$  the modified Bessel function of order  $m$  and  $A_m()$  a dimensionless function, accounting for the dielectric permittivity  $\epsilon_r$  of the capillary. The expression of  $A_m()$  is given by Eq. 14. Because of the sum over  $m$  and  $k$ , computing the image charge force  $F_{\text{im}}$  may be quite time consuming in simulations and a computationally faster expression approaching  $F_{\text{im}}$  would be welcomed. In the case of a

perfectly conducting cylindrical interface, Tórkési *et al.* approached expression (1) by the closed expression of the image force of a charge inside a conducting sphere [18, 19, 21]. Extending that idea to a dielectric interface, we propose to approach  $\vec{F}_{\text{im}}$  in a first step by the leading (zero order) term of the image force of a charge inside a dielectric sphere, see appendix VIC,

$$\vec{F}_{\text{im}}^{\circ}(\varrho) = \frac{q^2}{4\pi\epsilon_0 R^2} \frac{\epsilon_r - 1}{\epsilon_r + 1} \frac{\varrho}{(1 - \varrho^2)^2} \vec{u}_{\rho} \quad (2)$$

In order to check how well  $F_{\text{im}}^{\circ}$  approaches  $F_{\text{im}}$ , we compute the ratio  $F_{\text{im}}/F_{\text{im}}^{\circ}$  as a function of  $\varrho$  and for various values of  $\epsilon_r$ . Interestingly, the ratio  $F_{\text{im}}/F_{\text{im}}^{\circ}$  is independent of  $R$  as both  $F_{\text{im}}(\varrho)$  and  $F_{\text{im}}^{\circ}(\varrho)$  scale as  $R^{-2}$ . The ratio can thus be evaluated for arbitrary  $R$ . Here,  $F_{\text{im}}(\varrho)$  was evaluated using  $R = 1$  and the sum over  $m$  was limited to the first 200 angular moments, which was found to be sufficient for an accurate evaluation of (1) up to a radial position  $\varrho \leq 0.98$ . Results are shown in Fig. 2 for a selection of  $\epsilon_r$ . We see that the ratio get globally closer to unit with increasing  $\epsilon_r$ . And for a perfectly conducting cylindrical interface,  $|\epsilon_r| \rightarrow \infty$ , the ratio lies between 1.0027 and 1. Predictably, the deviation is systematically larger for a charge located close to the symmetry axis ( $\varrho \ll 1$ ), but even for  $\epsilon_r = 2$ , the relative deviation in that case is only about 11%. Importantly, close to the interface,  $\varrho \rightarrow 1$ ,  $F_{\text{im}}^{\circ}$  tends asymptotically to  $F_{\text{im}}$ . Indeed for  $\rho \simeq R$ , the cylindrical and spherical interfaces can both be assimilated by a plane interface, and their expressions yield the same limit namely that of the image force (12) of a plane surface. We deduce thus that the exact expression  $F_{\text{im}}$ , is surprisingly well approached by  $F_{\text{im}}^{\circ}$ , considering that the curvatures of cylindrical and spherical interfaces are different. In addition,  $F_{\text{im}}^{\circ}$  is less precise where the force is still relatively weak.

Obviously, an even better fit for (1) can be found. Looking at the data in Fig. 2, one notices that all 5 ratios deviate from unit in a scaled manner. We found that those ratios are well fitted by a function of the form

$$h_{\epsilon_r}(\varrho) = 1 + \alpha(1 - \varrho^2) + \beta(1 - \varrho^2)^{3/2} \quad (3)$$

where  $\alpha$  and  $\beta$  are parameters depending on  $\epsilon_r$ . Checking for a large selection of  $\epsilon_r \geq 2$ , we found that the relation between  $\alpha$ ,  $\beta$  and  $\epsilon_r$  can be written in the form,

$$\alpha = 0.48/(\epsilon_r + 1) \quad (4)$$

$$\beta = 0.00278 - 0.22/(\epsilon_r + 2) \quad (5)$$

We superimposed to the data in Fig. 2 the function  $h_{\epsilon_r}(\varrho)$  with corresponding  $\epsilon_r$ . The excellent agreement, (a relative error of less than  $5 \times 10^{-4}$ ) indicates that the closed formula

$$\vec{F}_{\text{im}}^{\text{cyl}}(\varrho) = \vec{F}_{\text{im}}^{\circ}(\varrho) \times h_{\epsilon_r}(\varrho) \quad (6)$$

with  $h_{\epsilon_r}$  defined by (3),(4) and (5) is an excellent approximation to the exact image force (1) for a cylindrical

dielectric interface. In simulations for which the computational cost for the evaluation of the image force is critical, we can safely suggest the closed formula (6) instead of the exact but CPU consuming expression (1).

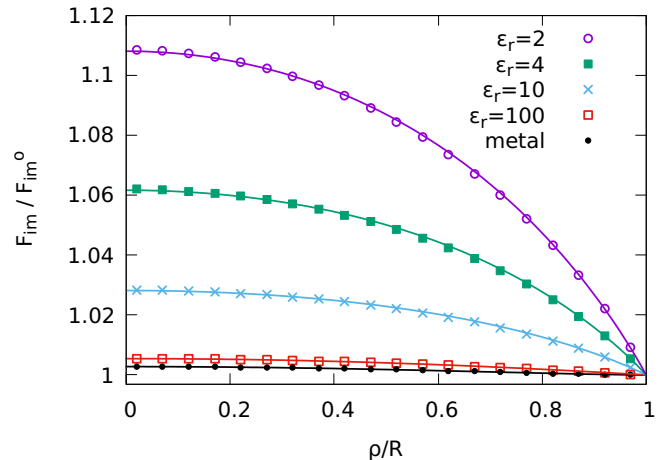


FIG. 2: Ratio  $F_{\text{im}} / F_{\text{im}}^{\circ}$  as a function of  $\rho/R$  for various  $\epsilon_r$ . The label "metal" stands for a perfectly conducting interface. The full curves are the function  $h_{\epsilon_r}(\rho/R)$  for  $\epsilon_r = 2, 4, 10, 100$  and  $|\epsilon_r| \rightarrow \infty$ .

## B. Ion trajectories and the theoretical transmitted fraction

We will now define the theoretically transmitted fraction  $f_{\text{th}}$  of an injected ion beam at zero tilt angle, in the ideal case where the capillary is a straight tube and the uniform, mono-kinetic ion beam has zero divergence. We assume that a charge  $q$  of mass  $m$  and energy  $E_k$  enters the cylindrical capillary with an initial radial distance  $\rho_0 < R$  and with an initial velocity  $\vec{v}_0 = \sqrt{2E_k/m} \vec{u}_z$ , so that at  $z = 0$  one has  $d\rho/dz = 0$ .

The theoretically transmitted fraction  $f_{\text{th}}$  is evaluated using  $F_{\text{im}}^{\circ}$  because the latter allows to establish a simple but useful relation between  $f_{\text{th}}$  and the properties of the capillary and the ion beam. We further check that using the more precise formula  $F_{\text{im}}^{\text{cyl}}$  introduces only a negligible correction to  $f_{\text{th}}$ , giving confidence in the present approach.

The trajectory  $\rho(z)$  of the ion can then be readily computed by solving the differential equation

$$\frac{d^2\rho}{dz^2} = \frac{1}{2E_k} F_{\text{im}}^{\circ}(\rho(z)/R) \quad (7)$$

with the initial values  $\rho(0) = \rho_0$  and  $\left. \frac{d\rho}{dz} \right|_{z=0} = 0$ . Using the dimensionless quantities  $\varrho = \rho/R$  and  $\zeta = z/H$ , equation (7) can be re-written in dimensionless form

$$\frac{d^2\varrho}{d\zeta^2} = \Gamma \frac{\varrho(\zeta)}{(1 - \varrho(\zeta)^2)^2} \quad (8)$$

with  $\varrho_0 = \rho_0/R$  and  $\left.\frac{d\varrho}{d\zeta}\right|_{\zeta=0} = 0$  and where the dimensionless factor

$$\Gamma = \frac{q^2}{4\pi\epsilon_0} \frac{\epsilon_r - 1}{\epsilon_r + 1} \frac{1}{2E_k} \frac{H^2}{R^3} \quad (9)$$

accounts for the length  $H$ , inner radius  $R$  and dielectric constant  $\epsilon_r$  of the capillary as well as the kinetic energy  $E_k$  of the charge. Remarkably, the factor  $\Gamma$  tells us that the dimensionless image charge force scales with the square of the capillary length  $H$  and but with the inverse cube of the capillary radius  $R$ . This explains why, for nano- and macro-capillaries of same aspect ratio  $H/R$ , the force can be neglected in macro-capillaries ( $R \sim 1$  mm) but not in nano-capillaries ( $R \sim 100$  nm). Also note that the force is inversely proportional to the kinetic energy of the ions.

Equation (8) is solved numerically, using a standard 4<sup>th</sup> order Runge-Kutta method. For a given  $\Gamma$  and for a given initial value  $\varrho_0$  the trajectory  $\varrho(\zeta)$  is uniquely defined. If the trajectory is intercepted by the capillary wall, we reduce the initial value of  $\varrho_0$ , otherwise,  $\varrho_0$  is increased. Using the bisection method, a good estimate for the largest value of  $\varrho_0^{\max}$ , for which the ion is still transmitted is found. Assuming that the injected current density  $j_0$  is uniform, one has  $I_{\text{in}} = j_0\pi R^2$  and  $I_{\text{out}} = j_0\pi(\varrho_0 R)^2$ . The ratio

$$\frac{I_{\text{out}}}{I_{\text{in}}} = (\varrho_0^{\max}(\Gamma))^2 = f_{\text{th}}(\Gamma) \quad (10)$$

defines the theoretical transmitted fraction  $f_{\text{th}}$  of the ion beam in the ideal case where the nano-capillary is a straight cylinder and the ion beam has zero divergence. The fraction  $f_{\text{th}}$  depends only on the factor  $\Gamma$  and has to be understood as an upper bound value, obtainable only in ideal conditions, i.e. the injected beam is spatially uniform, mono-kinetic, parallel and the capillary is perfectly cylindrical. In figure 3, we show 3 trajectories for 3 different values of  $r_0$  in the case of  $\Gamma = 1$ . We see that only the ions that are injected at a distance less than  $\varrho_0^{\max} = 0.435$  are transmitted, so that for  $\Gamma = 1$  the transmitted fraction  $f_{\text{th}}$  equals 18.6%. The theoretical transmitted fraction was calculated for a large range of  $\Gamma$  and is shown in Fig. 4 by the full black curve. In the case of macro-capillaries,  $\Gamma$  is typically well below  $10^{-4}$ , yielding  $f_{\text{th}}$  close to 100 %. As expected, this indicates that the effect of the image charge force on the ion trajectory is negligible in macro-capillaries. However, for keV ions through nano-capillaries,  $\Gamma$  is or the order of unit, and the ions transmission is expected to be strongly reduced by the image charge force. For  $\Gamma$  as large as 10, the transmitted fraction falls even below 1%. We add to Fig. 4 the values of  $f_{\text{th}}$  evaluated with  $F_{\text{im}}^{\text{cyl}}$  (red circles) instead of  $F_{\text{im}}^{\circ}$ . We used  $F_{\text{im}}^{\text{cyl}}$  with  $\epsilon_r = 2$ , which corresponds to the case where the correction  $h_{\epsilon_r}$  is the largest. We note that the black curve tends to slightly over estimate  $f_{\text{th}}$  for larger  $\Gamma$  but in general the difference with red circles is rather negligible, indicating that  $F_{\text{im}}^{\circ}$  is already a good approximation for (1).

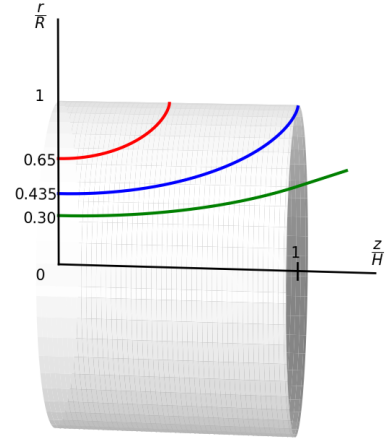


FIG. 3: Trajectories for 3 different initial values of  $\varrho_0$  in the case of  $\Gamma = 1$ . Only for  $\varrho_0^{\max} < 0.435$  are the ions transmitted, so that for  $\Gamma = 1$  the transmitted fraction  $f_{\text{th}}$  equals  $\varrho^2 = 18.6\%$

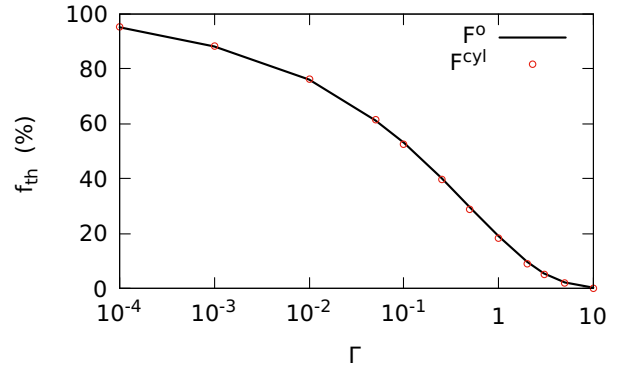


FIG. 4: Theoretical transmitted fraction  $f_{\text{th}}$  (in percentage) as a function of the dimensionless parameter  $\Gamma$  defined by Eq. 9 in the case of a uniform, divergence-less ion beam aligned with the capillary axis. Full black line has been computed using  $F_{\text{im}}^{\circ}$  while the red circles were obtained with  $F_{\text{im}}^{\text{cyl}} = F_{\text{im}}^{\circ} \times h_{\epsilon_r}$  and  $\epsilon_r = 2$ .

### C. Comparison with experimental data

We can now compare the theoretically transmitted fraction to the one found in former published works and discuss the observed discrepancies. This allows us to highlight a disagreement between the theoretical and experimentally observed transmitted fraction and of ions through nano-capillaries. Note that for the following discussion the more precise formula  $F_{\text{im}}^{\text{cyl}}(\varrho)$  was used to evaluate the theoretically transmitted fraction instead of  $F_{\text{im}}^{\circ}(\varrho)$ .

In experimental works on nano-capillaries, authors usually give the transmitted ion rate  $I_{\text{out}}$  through a large number of capillaries rather than the transmitted fraction  $I_{\text{out}}/I_{\text{in}}$ . Indeed, the injected ion rate  $I_{\text{in}}$  has to be deduced from the current density and size of the beam,



as well as transparency of the foil containing the capillaries, parameters which are difficult to obtain with sufficient precision. We nevertheless found three experimental studies which give the transmitted fraction at zero tilt angle.

Surprisingly, when comparing the theoretical transmitted fractions  $f_{th}$  to experimental data found in [4, 15, 16], we noticed a significant disagreement. In a recent work by Stolterfoht *et al.* [16], a transmitted fraction of 50% was observed for an 3-keV  $Ne^{7+}$  ion beam through polyethylene terephthalate (PET) nano-capillaries. The latter have a length of  $H = 12 \mu m$  and a diameter of  $2R = 230$  nm. To avoid charge-up of the insulating film by the ion beam, a gold layer of about 20 nm has been deposited on both sides. This may result in a gold collar reducing the inlet and outlet diameter of the nano-capillaries, see Fig. 5. The inlet and outlet diameters, as measured by scanning electron microscopy (SEM) after gold deposition, may thus underestimate the diameter of the capillary up to 40 nm. While we found no clear experimental evidence that shows that a collar of tens of nm thickness is present, we will nevertheless evaluate  $f_{th}$  for both cases, with and without a 20 nm circular gold collar at the inlet and outlet of the capillaries.

We specify that for the gold deposition process, the capillaries were tilted by 30 deg with respect to the gold vapor and then turned by 180 deg. Accordingly, the collar may not be perfectly circular but rather some kind of ellipse. Nevertheless, for the present purpose we assume that the gold coverage has circular geometry.

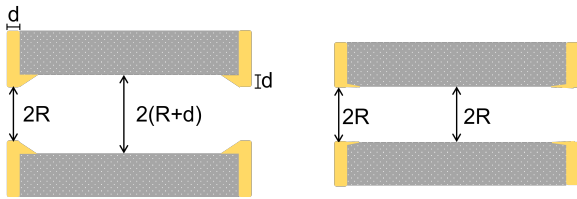


FIG. 5: Schematic representation of the presence (left) or absence (right) of a gold collar of thickness  $d$  at the inlet and outlet of a capillary after a gold deposition occurred on the front and back side of the insulating sheet containing the capillaries.

Using for PET a dielectric constant of 3.0 [?], one has for the parameter  $\Gamma$  a value of 0.55. The theoretical transmitted fraction obtained for  $\Gamma = 0.55$  is 27%, which is lower than the experimental one by almost a factor 2. If we account for a 20 nm collar,  $f_{th}$  increases to 31%. In order to recover the experimental transmitted fraction of 50%, the parameter  $\Gamma$  needs to be reduced by a factor 4. This can be achieved by reducing  $\epsilon_r$  to 1.3, which yields however a non-physically low value for  $\epsilon_r$ . Alternatively, it can be achieved by increasing the diameter  $2R$  of the capillaries by a factor 1.5, that is  $2R \rightarrow 350$  nm. But that would mean that the diameters measured by scanning electron microscopy (SEM) underestimates the actual inner diameter of the capillaries by about 120

nm, which seems unlikely.

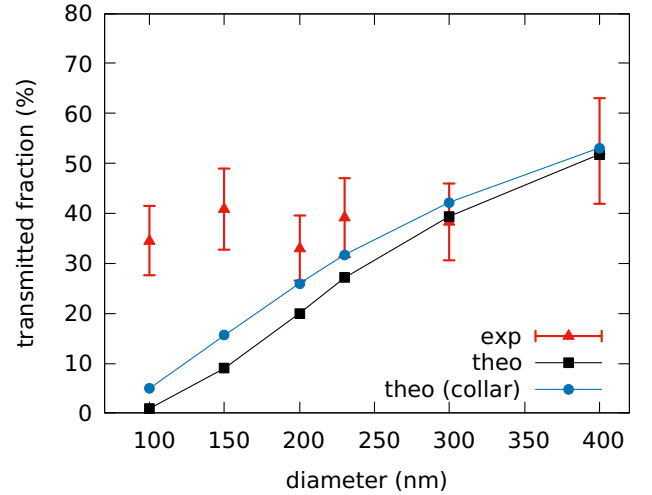


FIG. 6: Comparison between experimentally transmitted fractions (red triangles) extracted from [15, 16] and our theoretically transmitted fractions as a function of the capillary diameter  $2R_i$ . Experimental data are normalized such that fractions coincide for capillaries with 400 nm diameter. Filled blue circles give the theoretically transmitted fractions in the case where the inlet and outlet diameters include a 20 nm collar, filled black squares are evaluated without collar.

In 2010, Stolterfoht *et al.* measured the transmission rates of PET nano-capillaries of various diameter  $2R_i$ , ranging from 100 to 400 nm [15]. They did not give the transmitted fraction for each diameter but rather the relative transmitted ion yield. Obviously, this analysis advocates that, for those measurements, the current density and spot size of the injected ion beam was kept constant. The injected ion yield  $I_{in}$  can thus be considered proportional to the inlet section  $\pi R_i^2$  of the capillaries.

$$I_{in} \propto R_i^2 \quad .$$

Consequently, we find that the experimentally transmitted fraction  $I_{out}/I_{in}$  is proportional to the transmitted ion yield divided by the inlet section,

$$\frac{I_{out}}{I_{in}} \propto \frac{I_{out}}{R_i^2} \quad .$$

In figure 6, we compare  $\propto I_{out}/R_i^2$  to the theoretically transmitted fractions for capillary diameters used in [15]. The proportionality factor was chosen such that  $I_{out}/I_{in}$  coincides with  $f_{th}$  for capillaries with the largest diameter, namely  $2R_i = 400$  nm. We also add the fractions for  $2R_i = 230$  nm, discussed previously. We see that the experimentally transmitted fractions (red triangles) diminishes only slightly with decreasing  $2R_i$ . This is surprising because equation (9) tells us that  $\Gamma$  scales as  $R^{-3}$  and thus a strong dependence of the transmitted fraction on the capillary radius is expected, as shown in figure 4.

Our model predicts that  $f_{\text{th}}$  decays much faster with decreasing capillary diameter. In particular for  $2R_i = 100$  nm, the discrepancy is huge. The theoretical transmitted fraction, without a collar, is 30 times smaller than the observed one. If we consider a 20 nm gold collar at the inlet and outlet of the capillaries (blue filled circles),  $f_{\text{th}}$  increases slightly, especially for smaller diameters. But even then,  $f_{\text{th}}$  is 6 times smaller than the experimental one for  $2R_i = 100$  nm.

In their work of 2006 on highly ordered  $\text{SiO}_2$  nano-capillaries, Sahana *et al.* [4] found that 20% of the  $\text{Ne}^{7+}$  ions, entering the capillaries at zero tilt angle, are being transmitted. They used a 7 keV ion beam and the nano-capillaries had a length of  $H = 25$   $\mu\text{m}$  and a diameter about  $2R \simeq 100$  nm, yielding a large aspect ratio of about  $H/(2R) \simeq 250$ . A 30 nm gold layer was evaporated on both sides of the the film. Taking for the dielectric constant of amorphous  $\text{SiO}_2$  a value of 3.9 [23], we get using (9) for the factor  $\Gamma$  a value of 14.9. For such a large  $\Gamma$ , our simulations find that only those ions which are injected within an initial distance  $\rho \leq 2$  nm from the symmetry axis are transmitted. The theoretical transmitted fraction  $f_{\text{th}}$ , calculated with  $\Gamma = 14.9$  is 0.2%, is lower than the experimental one by a factor 100. If we consider a 30 nm gold collar,  $f_{\text{th}}$  increases to 1%, which is still a factor 20 from the experimental one. In order to recover the experimental transmitted fraction of 20%, the scaling factor  $\Gamma$  should be reduced here by a factor 14. Either the transmitted fraction was overestimated by the authors, or the measurement of the capillary diameter was underestimated or the image force as given by (6) and thus (1) is not appropriate.

The observed transmitted fraction in those three studies [4, 15, 16] are systematically significantly larger than those found theoretically in the ideal case. The experimental data do not corroborate the theoretical scaling of the transmitted fraction with the capillary radius. For keV ions, the intensity of the image force in cylindrical capillaries with a diameter smaller the 200 nm seems thus to be much lower than what is predicted theoretically. One possibility is that the image charge force expressed by  $F_{\text{im}}^{\text{cyl}}(\varrho)$  (6) is not valid for keV ions through nano-capillaries. And as (6) fits the exact expression  $F_{\text{im}}(\varrho)$  extremely well, the same holds for  $F_{\text{im}}(\varrho)$  (1). We remind that expressions (1) was obtained in the electrostatic limit with a static relative permittivity  $\varepsilon_r$ .

#### *Case of rhombic and rectangular capillaries*

Apart from cylindrical nano-capillaries, experimental studies were also done on muscovite mica nano-capillaries with rhombic (long axis 248 nm and short axis 142 nm) and rectangular section ( $215 \times 450$  nm<sup>2</sup>) [13, 14]. The observed beam-shaping of the transmitted ion was shown to be due to the image charge force, indicating clearly that the image charge force influences dominantly the ion trajectories. In their simulations, the image charge

force is a superposition of the image charge force for a plane dielectric surface for each of the 4 plane surfaces. Interestingly, their simulations succeeded to reproduce the observed transmitted fraction of about 1% for 7 keV  $\text{Ne}^{7+}$  through the rhombic shaped muscovite mica nano-capillaries. However, in the case of rectangular capillaries, the simulated transmission rate and experimental values differ considerably (the experimental values are lower), but was attributed to size variations inside the channels.

We conclude this section by noting that significant disagreements were found between measured and theoretical transmitted fractions in the case of cylindrical nano-capillaries made in PET and  $\text{SiO}_2$ . Better agreement was noticed in the case of rhombic shaped mica nano-capillaries, but was not further confirmed in nano-capillaries with rectangular sections. Where disagreement was found, the experimental transmitted fraction was always significantly larger than the simulated one.

### III. VELOCITY DEPENDENT IMAGE CHARGE FORCE

The disagreement highlighted in the previous section made us question the validity of expression (1) of the image charge force in the case where keV ions pass through a nanocapillary of large aspect ratio. There are indeed two effects that we ignored in the evaluation of the image charge force  $F_{\text{im}}(\rho)$ . If the inner surface of the capillary is hit by ions, the accumulated charge may modify the dielectric response of the insulator. For example, in pre-charged PET samples, the trapped charge was found to partially inhibit the dipolar polarization, reducing by about 5% the dielectric permittivity with respect to untreated PET [24]. However, except for [24], which limited their study below the 1MHz domain, no other reference was found. So while the accumulated charge seem indeed to modify the dielectric response, the effect of excess charge on the dielectric response function is hardly documented in the literature and otherwise difficult to estimate by simple considerations. We decide thus for the present study to ignore the effect of the charge on the dielectric response.

The other effect is that the ions are moving and the velocity of the ions influences the induced polarization at the interface. The dynamical effects in the image charge force were already investigated by [26] in the case of a metal plane interface. They found that dynamical corrections become non-negligible when the velocity  $v$  of the moving charge approaches the Fermi velocity of electrons in metals. The reason is that the image charge follows the mirror trajectory but is retarded by a distance  $vt_r$  due to the finite response time,  $t_r$  of the electron gas. Similar findings are presented by Tórkési *et al.* [18] where they studied the dynamical correction of the image force in the case of a cylindrical conducting interface. We will in this section investigate the image charge force for a



moving ion approaching a dielectric plane and cylindrical interface and evaluate in how far dynamical corrections modify the static image force in the case of keV ions.

### A. Frequency dependent relative permittivity

The relative permittivity of bulk describes the ability to polarize a material subjected to an electrical field. In dielectrics, this polarization originates from a number of sources: electron cloud displaced relative to nucleus, relative displacement of atoms, alignment of dipoles in electric field and accumulation of charge carriers at the interface. These sources have different response times so that the relative permittivity  $\varepsilon_r \equiv \varepsilon_r(\omega)$  depends on the angular frequency  $\omega$  of the external electric field. We assume that the dependence of  $\varepsilon_r$  on the wave vector  $\vec{k}$  can be neglected if the spatial extension of the induced dipole moments is small compared to the wavelength  $2\pi/k$ , which is the case for frequencies of the electromagnetic field well below optical frequencies [25]. The function  $\varepsilon_r = \varepsilon'_r + i\varepsilon''_r$  is a complex quantity and the frequency-dependence of the real and imaginary part of  $\varepsilon_r$  is illustrated in Fig. 7.

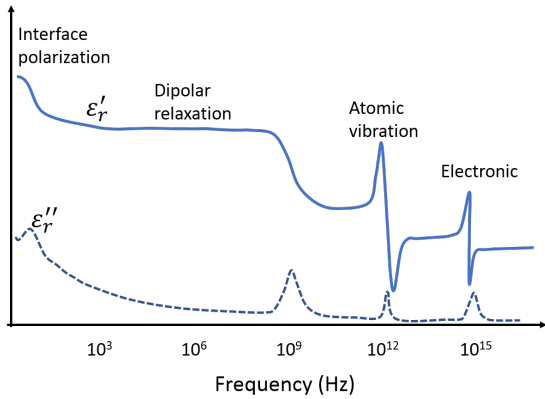


FIG. 7: Arbitrary dielectric permittivity spectrum over a wide range of frequencies. The real  $\varepsilon'_r$  and imaginary  $\varepsilon''_r$  parts of permittivity are shown, and various processes are labeled: Interface polarization, dipolar relaxation, atomic and electronic resonances at higher frequencies.

For a keV ion moving along the interface, the electric field generated at a point of the interface may quickly evolve in time. If the response time of the dielectric function is too low, the patch of induced surface charges may trail the ion, which results in a reduction of the image charge force. Only polarization sources that have a sufficiently fast response time will contribute significantly to the image charge force. For a dielectric interface, the image force is thus expected to depend on the velocity of the ion [18, 26]. Considering a keV ion having typically a velocity of about  $10^5$  m/s and located 10 nm from the interface, the electric field generated by the ion at a point of the interface has a characteristic frequency of  $v/x_p = 10$  THz, which is well in the infrared domain, where the di-

electric response of insulators may vary significantly. In figure 8 we give the real part of the dielectric response of  $\text{SiO}_2$  and PET insulators in the THz domain. Data were extracted from [27–29] and show that the atomic dipole resonances responsible for a large part of the value of  $\varepsilon_e$  are found above 10 THz.

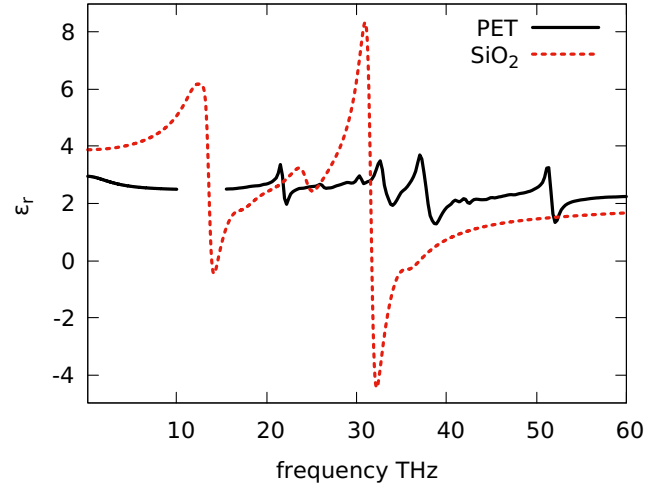


FIG. 8: Real part of the dielectric response function of  $\text{SiO}_2$  (red dashed line) and PET (black full line) extracted from [27] and [22, 28, 29] respectively.

### B. Velocity dependent image charge force

We will now evaluate the influence of the velocity of a moving charge on the image force. In a first step, we investigate the image force in the case of an infinite plane interface, which yields for the velocity dependent image force a handy expression, facilitating the discussion. Then, we check its effect on in the case of a cylindrical interface.

#### 1. Case of an infinite plane interface

Let us consider a homogeneous dielectric which fills the region  $z < 0$ . The region  $z > 0$  is empty. A projectile of charge  $q$  is situated at  $z = z_p > 0$  and moves along the Oz coordinate with the velocity  $v > 0$ . The expression of image charge force acting on a moving ion has been detailed in appendix VIB and is given by

$$F_z(z_p, v) = \frac{q^2}{2\pi\varepsilon_0 v^2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} |\omega| \frac{\varepsilon_r(\omega) - 1}{\varepsilon_r(\omega) + 1} K_1 \left( \frac{2z_p |\omega|}{v} \right) \quad (11)$$

The image charge force depends on the velocity  $v$  of the ion and on the frequency response of the dielectric function  $\varepsilon_r(\omega)$ . For angular frequencies  $\omega \gg v/(2z_p)$ , the modified Bessel function  $K_1(2\omega z_p/v)$  decreases as

$\exp(-2\omega z_p/v)$ , so that the ratio  $v/(2z_p)$  can be regarded as a cut-off angular frequency and only angular frequencies  $\omega$  smaller than  $v/(2z_p)$  will contribute significantly to the image charge force. For example, 7 keV  $\text{Ne}^{7+}$  ions (as used by Sahana *et al.* [4]) positioned  $z_p = 10$  nm from the interface yield a cut-off angular frequency of about  $10^{13}$  radians Hz. The latter is close to the atomic polarization resonances, where the dielectric function varies quickly. Thus, for keV ions, the dielectric response function cannot be assumed constant and the image charge force is expected to depend significantly on the ion velocity.

For much lower ion velocities, the cut-off frequency  $v/(2z_p)$  may fall below 1 Mhz. In this low frequency range, the relative permittivity of insulators is usually constant, interfacial polarization apart. Taking  $\epsilon_r(\omega) = \epsilon_r$  and integrating over  $\omega$  yields the well-known image charge force for a plane vacuum-dielectric interface [? ].

$$F_z(z_p, v \rightarrow 0) = \frac{q^2}{4\pi\epsilon_0} \frac{\epsilon_r - 1}{\epsilon_r + 1} \frac{1}{(2z_p)^2} \quad (12)$$

Equation (12) can thus be seen as the adiabatic limit of Eq. (11), i.e. for vanishing small ion velocities  $v$ .

The dependence of the image charge force on the ion velocity is shown in figure 9. For better readability, we normalize the latter with respect to the adiabatic ( $v \rightarrow 0$ ) image charge force (Eq. 12), which allows to highlight the influence of the ion velocity on the force. The normalized force is shown for three different distances  $z_p$  from the dielectric plane interface, namely  $z_p = 5, 10$  and  $20$  nm. Here, we used the dielectric response function  $\epsilon_r(\omega)$  of  $\text{SiO}_2$  as shown in Fig. 8, which has a large response in the THz domain, due to the ionic and atomic vibrations of the glassy network. For a distance  $x_p = 10$  nm

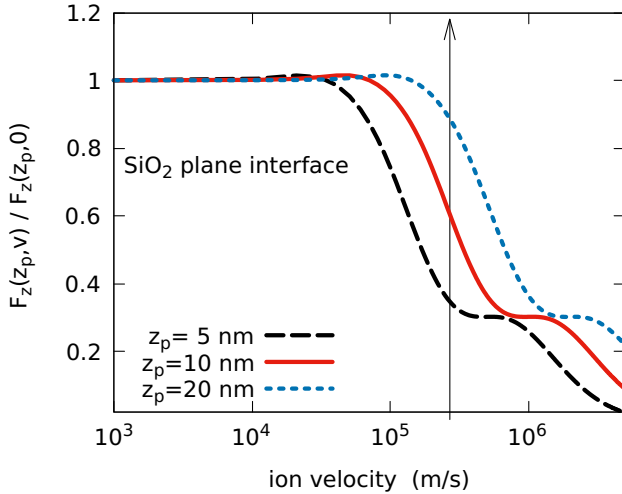


FIG. 9: Normalized image charge force  $F_z(z_p, v)/F_z(z_p, 0)$  as a function of the ion velocity  $v$ , for 3 different distances  $z_p$  from the  $\text{SiO}_2$  interface,  $z_p = 5, 10$  and  $20$  nm. The arrow indicates the velocity of 7 keV  $\text{Ne}^{7+}$  ions as used by Sahana *et al.* [4].

from the interface, the normalized image charge force is

reduced by 40% for ion velocities larger than  $10^5$  m/s. The reason is that for  $v > 10^5$  m/s, atomic and ionic modes that usually lie in the THz range do not respond quickly enough to the time-varying electric field. Only bound electrons respond quickly enough and contribute to the surface polarization. For even larger velocities  $v > 10^7$ , bound electrons are not responding fast enough and the image force tends to zero. With this example we illustrate that the image charge force acting on a charge moving along a cylindrical interface is significantly reduced for velocities above a critical value, which depends on the frequency response of the dielectric function and distance  $z_p$  from the dielectric interface.

## 2. Case of an infinite cylindrical interface

In the next step, we consider an infinite cylindrical interface of radius  $R$  having its symmetry axis along  $Oz$ . The interface separates the inner vacuum from the outer insulating medium of dielectric permittivity  $\epsilon_r(\omega)$ . The latter is assumed complex valued and depending on the frequency. Let a charge  $q$  move in the  $xOz$  plane with the velocity  $v\vec{u}_z$ , so that its time-dependent position is given by the vector  $(\rho, 0, vt)$ , with  $\rho < R$ . The expression of the dynamical image force is similar to the static one (Eq. 1) but takes now explicitly into account the dielectric response and depends on the velocity  $v$ ,

$$F_{\text{im}}(\rho, v) = \frac{q^2}{16\pi\epsilon_0} \sum_{m=0}^{\infty} \int_0^{\infty} dk \frac{\partial I_m^2(k\rho)}{\partial \rho} \Big|_{\rho} A_m(kR, \epsilon_r(kv)) \quad (13)$$

The function  $I_m()$  and  $K_m()$  are the modified Bessel functions and  $A_m()$  is a dimensionless function depending on the dielectric permittivity  $\epsilon_r(\omega)$ , evaluated at the pulsation  $\omega = kv$ ,

$$A_m(kR, \epsilon_r(kv)) = \frac{\epsilon_r(kv) - 1}{\left( \epsilon_r(kv) \frac{I_m(kR)}{K_m(kR)} - \frac{I'_m(kR)}{K'_m(kR)} \right)} \quad (14)$$

Here  $I'_m$  and  $K'_m$  are the partial derivatives of the Bessel functions with respect to  $\rho$ . Note that the imaginary part of  $A_m$  cancels, so that  $A_m$  is real valued. In the case of the adiabatic limit,  $v \rightarrow 0$ , the functions  $A_m()$  are evaluated using for the relative permittivity its static limit,  $\epsilon_r(kv \rightarrow 0) = \epsilon_r$ , yielding expression (1).

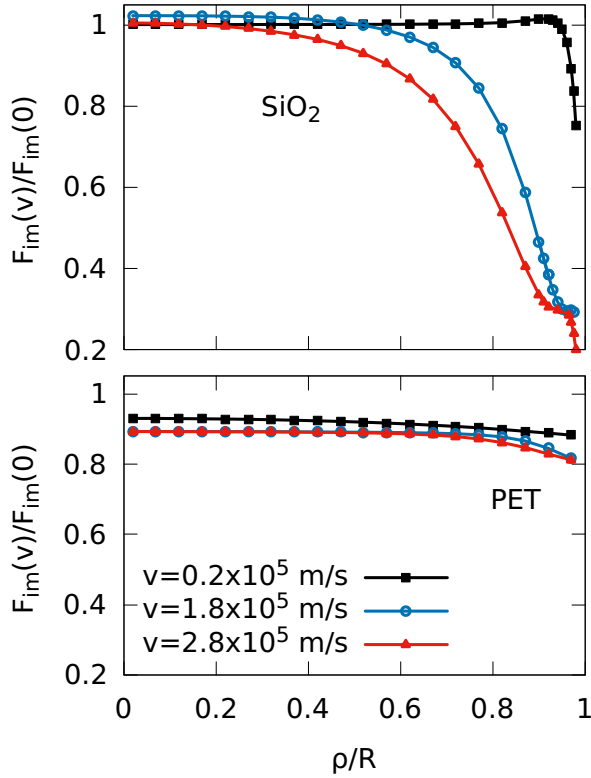


FIG. 10: Normalized image charge force,  $F_{\text{im}}(\rho, v)/F_{\text{im}}^0(\rho)$  of a moving charge inside a cylindrical dielectric interface of radius  $R = 50$  nm as a function of the dimensionless radial distance  $\rho/R$ , for three different velocities, namely  $v = 2 \times 10^4$ ,  $v = 1.8 \times 10^5$ , and  $v = 2.8 \times 10^5$  m/s. Upper panel, dielectric response function of  $\text{SiO}_2$  as given in Fig. 8 was used. Bottom panel, dielectric response function of PET as given in Fig. 8 was used.

#### IV. INFLUENCE OF THE ION VELOCITY ON THE THEORETICAL TRANSMITTED FRACTION

##### A. Image force for keV ions in $\text{SiO}_2$ and PET nano-capillaries

We may now check in how far the velocities of keV ions reduce the image charge force for those cases highlighted in section II C, namely for 7 keV  $\text{Ne}^{7+}$  ions through  $\text{SiO}_2$  nanocapillaries and 3 keV  $\text{Ne}^{7+}$  ions through PET nanocapillaries. The dielectric response functions of  $\text{SiO}_2$  nano-layers and of a thin samples of PET are pictured in Fig. 8. We compute for each case the image force as a function of  $\rho$  and for three different velocities, namely  $0.2 \times 10^5$  m/s,  $1.8 \times 10^5$  m/s and  $2.8 \times 10^5$  m/s. The two latter correspond to  $\text{Ne}^{7+}$  at 3 keV, as used by Stolterfoht et al. [15, 16] and  $\text{Ne}^{7+}$  at 7 keV ions, as used by Sahana *et al.* [4], respectively. The first smaller velocity, is added for comparison. Expression (13) is evaluated numerically using MATHEMATICA [30] for the case of a

cylindrical interface of radius  $R = 50$  nm. The sum over  $m$  was limited to the 100 first angular multipoles. The results are shown in Fig. 10, where the dynamical image force  $F_{\text{im}}(v)$  is normalized with respect to the static one,  $F_{\text{im}}(v = 0)$ . In the case of a  $\text{SiO}_2$  interface and 7 keV  $\text{Ne}^{7+}$ , (red triangles, upper panel,  $v \simeq 2.8 \times 10^5$  m/s), the dynamical image force is reduced by up to a factor 3 compared to the adiabatic image force when approaching the interface. Indeed, for such a velocity, the response time of the first Lorentz oscillator with an eigenfrequency about 12 THz is too low to contribute efficiently to image force. This results in a significant decrease of the image force when the ion approaches the interface,  $\rho/R \geq 0.6$ , which for  $R = 50$  nm, gives a distance of about  $< 23$  nm from the interface. On contrary, for a 10 times smaller ion velocity (black squares), all the dipole oscillators follow the ion in phase and the dynamical corrections on the image force are negligible.

For a PET interface, the ratio  $F_{\text{im}}(v)/F_{\text{im}}(0)$  for a moving charge of  $1.8 \times 10^5$  is about 0.88 and mainly independent on the radial coordinate  $\rho$ . This is due to the fact that the dynamical corrections to the image force come from the finite response time of dipole alignment in PET, as indicated by the small decrease of  $\epsilon_r$  from 3 to 2.6 in the sub 3 THz domain, as shown in Fig 8 and reported by [28]. Also, the first resonance frequency of atomic dipoles in PET lies well above 20 THz and also has a smaller strength than the first resonance in  $\text{SiO}_2$ . As a result, the dynamical image force for charges moving with a velocity up to  $2.8 \times 10^5$  is only slightly affected when approaching the PET interface. We deduce that for keV ions approaching a PET interface, the dynamical corrections are small and the dielectric permittivity of PET is thus well approached by a dielectric constant  $\epsilon(f = 3\text{THz}) = 2.6$ .

##### B. New theoretical transmitted fraction

We may now check to what extent the dynamical corrections to the static image force lift the discrepancy found between the experimentally observed and theoretically evaluated transmitted fraction, as described in section II C. Again we solve equation 7, but this time using the image force as given in Fig. 10. We look for the largest initial value of  $\rho_0^{\text{max}}$  for which 7 keV  $\text{Ne}^{7+}$  ions are transmitted through  $\text{SiO}_2$  nanocapillaries, as those used by Sahana *et al.*. We get this time a theoretical transmitted fraction of 0.3%, only slightly larger than the 0.2% obtained previously using the static image force, but still well below the observed 20% transmission.

Similarly, for 3 keV  $\text{Ne}^{7+}$  ions through PET nanocapillaries, as those used by Stolterfoht et al. [15, 16], dynamical corrections increase only negligibly the theoretical transmitted fraction. With respect to the one obtained with a static image force,  $f_{\text{theo}}$  increases only slightly, from 27% to 28%, which is still almost a factor 2 lower than the observed 50% transmission. We get thus the

unfortunate result that dynamical effects do not alter significantly the theoretical transmitted fraction with respect to the static one. Even if the velocity of the keV ion is taken into account in the evaluation of the image force inside a dielectric cylinder, we are not able to explain the experimentally observed transmission fractions of ions through large aspect nano-capillaries, especially for inner diameters of about 100 nm. It seems thus that the discrepancy hides a phenomenon we did not yet tackle.

## V. CONCLUSION

We evaluated the theoretically transmitted ion beam fraction through straight insulating nano-capillaries by calculating the fraction of ion trajectories that were not intercepted by the capillary wall. We checked via simulations that the only non-negligible force acting on the ions was the image charge force due to the induced polarization at the inner dielectric interface of a straight cylindrical capillary. Instead of using the exact expression of the image force, we proposed a computationally faster formula that approaches very well the image force in the adiabatic limit. The transmitted fraction was evaluated in the ideal case of a mono-kinetic divergence free beam injected at zero tilt angle. When comparing the theoretically transmitted fraction to the three experimental data available in the literature, we found large discrepancies in the transmitted fractions. We took into account in the theory a gold collar of about 20 nm that reduces the inlet/outlet diameters of the capillaries. We found that the collar did increase the theoretically transmitted fractions by a notable amount, but did not succeed to lift the disagreement.

As the relative permittivity of a dielectric material depends on the frequency of the applied electric field, the surface polarization induced by a charge moving along the interface depends on its velocity. We evaluated thus the image charge for a charge moving along a dielectric plane and along a dielectric cylindrical interface and in particular for  $\text{SiO}_2$  and PET interfaces. We found that for keV ions having typically a velocity about  $10^5$  m/s, the image charge force is controlled by the near infrared domain (THz) of the dielectric response function. For a  $\text{SiO}_2$  interface, the velocity of the ions affects notably the image force when the ion is close to the interface. For PET interface, the dynamical effects of the image force are negligible in the chosen keV range.

We then checked in how far the theoretically transmitted fractions are modified when using the velocity dependent image charge force instead of the static (adiabatic) one. The calculations were performed for 7 keV and 3 keV  $\text{Ne}^{7+}$  ions. We found that the newly calculated transmitted fractions are only slightly larger than those found previously in the case of adiabatic limit and still disagree with the experimental results. We conclude thus that a velocity dependent image charge force is not able

to lift the highlighted disagreement. And because the electric field inside the capillary due to the accumulated charge in the walls can be neglected for large aspect ratio capillaries, there is still no convincing explanation for why the theoretical transmitted fraction was found well below the observed one, especially for capillaries with inner diameters below 200 nm. Of course there is always this uncertainty about the real diameter of the capillary in experimental data because of the unknown thickness of the gold collar at the inlet and outlet of the capillaries. But how much it affects the transmitted fraction needs additional investigation.

With this work, we hope to motivate new experimental work on the ion transmission through insulating nano-capillaries of different sections. We think that new data will guide authors to model more accurately the image force of keV ions through capillaries, shedding new light on the phenomenon.

## VI. APPENDIX

### A. Electric field of the deposited charges vs image force on ion trajectories

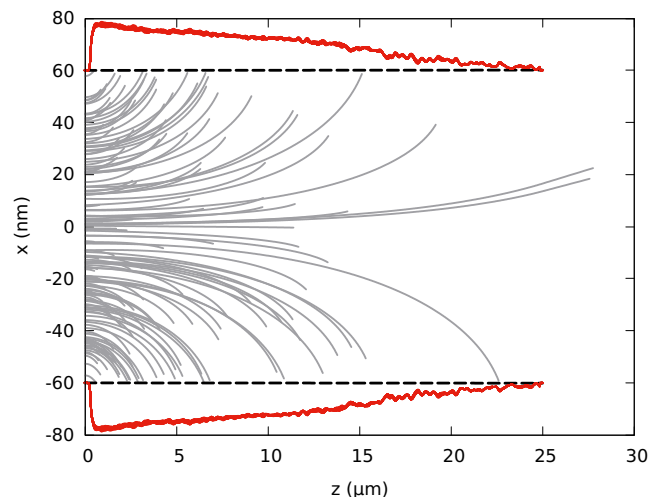


FIG. 11: Simulated ion trajectories (gray curves) through a 25  $\mu\text{m}$  long nanocapillary of inner radius of 60 nm, as indicated by the black dashed line. The red curves indicate the accumulated charge in arbitrary units at the inner interface.

We simulated the trajectories of 7 keV  $\text{Ne}^{7+}$  ions through an insulating  $\text{SiO}_2$  nano-capillary embedded in semi-conducting silicon. The inner radius of the capillary tube is 60 nm, the outer radius is 150 nm and the length is 25  $\mu\text{m}$ . The forces acting on the ions are the image force expressed by Eq. 6 and the Coulomb force due to accumulated charges at the inner capillary surface. The beam axis is aligned with the capillary axis and the injected beam intensity is 10 ions per second, thus about  $10^{-17}$  A. The outer surface is grounded and

accounts for the grounded semi-conducting Si bulk. This system model aims at modeling closely the experiment described in [4]. The bulk conductivity  $\kappa$  of SiO<sub>2</sub> is field dependent and given by the non-linear Poole-Frenkel relation [31, 33]

$$\kappa(E) = \kappa_0 \exp\left(\sqrt{E/E_c}\right) \quad (15)$$

The field independent bulk conductivity  $\kappa_0$  is set to 10<sup>-15</sup> S/m [32] and the critical field  $E_c$  is set to 10<sup>7</sup> V/m, which is the typical range where the conductivity SiO<sub>2</sub> becomes non-linear [17, 33]. We show in figure 11 the computed ion trajectories. The accumulated charge in the capillary wall is shown by the red curves. After about 3 hours, the deposited charge per unit time is balanced by the leakage current and the accumulated charge has reached its asymptotic distribution. The electric field inside the capillary wall reaches at that moment 30 MV/m, putting the conductivity well within the non-linear regime. The transmitted beam fraction as a function of time is shown in Fig. 1, where it is compared to the case where the electric field due to the deposited charges is removed, so as to put into evidence the zero effect of the deposited charge on the transmission.

### B. Image charge at plane interface

Let us consider a homogeneous dielectric of relative permittivity  $\varepsilon_r$  which fills the region  $x < 0$ . The region  $x > 0$  is empty. A projectile of charge  $q$  is situated at  $x_p > 0$ ,  $y_p = 0$  and moves along the Oz coordinate with the velocity  $v > 0$ ,  $z_p = vt$ . The x-component of the electric field at the interface  $x = 0$  generated by the moving charge at position  $\vec{r}_p(t) = (x_p, 0, vt)$  reads

$$E_x^q(x_p, y, z, t) = \frac{q}{4\pi\epsilon_0} \frac{-x_p}{(x_p^2 + y^2 + (z - vt)^2)^{3/2}} \quad (16)$$

Because of the discontinuous jump of the relative permittivity  $\varepsilon_r$  at the interface  $x = 0$ , the electric field  $E_x^q$  induces at the interface surface charge density  $\sigma$ . The electric field generated by the surface charges  $\sigma$  at the dielectric side  $x = 0^-$  is  $-\frac{\sigma}{2\epsilon_0}$  but  $+\frac{\sigma}{2\epsilon_0}$  at  $x = 0^+$ , so that the total electric field at each side reads

$$E_x^- = E_x^q - \frac{\sigma}{2\epsilon_0} \quad , \quad x = 0^- \quad (17)$$

while in the empty region side,  $x = 0^+$ , it reads

$$E_x^+ = E_x^q + \frac{\sigma}{2\epsilon_0} \quad , \quad x = 0^+ \quad (18)$$

In the absence of free surface charges, the x-component of the displacement field  $D_x$  is continuous across the interface. The relation between  $\vec{D} = \varepsilon\vec{E}$  is non-local in time and position and is more comfortably used in the frequency and wave-number domain. The latter relation

is more convenient when expressed in the Fourier space, where it simplifies to

$$\vec{D}(\vec{k}, \omega) = \tilde{\varepsilon}(\vec{k}, \omega) \vec{E}(\vec{q}, \omega) \quad , \quad (19)$$

Let  $\tilde{E}_x^q$  be the Fourier transform in the frequency domain of the electric field  $E_x^q$ ,

$$\begin{aligned} \tilde{E}_x^q(\omega, x_p, y, z, v) &= \frac{-q}{4\pi\epsilon_0} \int_{-\infty}^{\infty} \frac{x_p e^{i\omega t} dt}{(x_p^2 + y^2 + (z - vt)^2)^{3/2}} \\ &= \frac{-q e^{i\omega z/v}}{2\pi\epsilon_0} \frac{|\omega|}{v^2} \underbrace{\frac{K_1\left(\frac{|\omega|\sqrt{x_p^2 + y^2}}{v}\right)}{\sqrt{x_p^2 + y^2}}}_{G(\omega, x_p, y, v)} \end{aligned} \quad (20)$$

where  $K_1()$  is the modified Bessel function of order 1.

In the following we assume that the dielectric function is local in space so that  $\varepsilon_r(\omega, \vec{k}) \equiv \varepsilon_r(\omega)$ . This assumption is valid for  $|k| \ll \pi/a$ , where  $a \simeq 0.5$  nm is the typical length of the molecules responsible for the polarization. Using (19) one has at both sides of the interface  $x = 0$ ,

$$\tilde{D}_x^-(\omega, y, z, v) = \varepsilon_r(\omega) \left( \frac{\tilde{\sigma}(\omega, y, z, v)}{2} + \varepsilon_0 \tilde{E}_x^q \right) \quad (21)$$

$$\tilde{D}_x^+(\omega, y, z, v) = \left( -\frac{\tilde{\sigma}(\omega, y, z, v)}{2} + \varepsilon_0 \tilde{E}_x^q \right) \quad (22)$$

Using the interface condition

$$\tilde{D}_x^-(\omega, x, y, v) = \tilde{D}_x^+(\omega, x, y, v)$$

one obtains the expression for  $\sigma(\omega, y, z, v)$

$$\tilde{\sigma}(\omega, y, z, v) = \frac{-q e^{i\omega z/v}}{\pi} \frac{\varepsilon_r(\omega) - 1}{\varepsilon_r(\omega) + 1} \frac{|\omega|}{v^2} G(\omega, x_p, y, v) \quad (23)$$

The image charge force acting on the projectile is eventually,

$$F_x(x_p) = \frac{q}{4\pi\epsilon_0} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \int dy \int \frac{e^{-i\omega t} x_p \tilde{\sigma}(\omega, y, z, v) dz}{(x_p^2 + y^2 + (z - vt)^2)^{3/2}} \quad (24)$$

Integration over  $z$  gives

$$F_x(x_p) = \frac{-q^2}{2\pi^2\epsilon_0 v^3} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} |\omega|^2 \frac{\varepsilon_r(\omega) - 1}{\varepsilon_r(\omega) + 1} \int_{-\infty}^{\infty} dy G(\omega, x_p, y, v)^2 \quad (25)$$

Noting that

$$\int_{-\infty}^{\infty} dy G(\omega, x_p, y, v)^2 = \pi \frac{v}{|\omega|} K_1\left(\frac{2x_p|\omega|}{v}\right) \quad (26)$$

one has

$$F_x(x_p) = \frac{-q^2}{2\pi\epsilon_0 v^2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} |\omega| \frac{\varepsilon_r(\omega) - 1}{\varepsilon_r(\omega) + 1} K_1\left(\frac{2x_p|\omega|}{v}\right) \quad (27)$$



### C. Image force for a spherical dielectric interface

Assume a spherical cavity of radius  $R = 1$  in a dielectric bulk of dielectric constant  $\varepsilon_r > 1$ . A charge  $q$  is located inside the cavity at a distance  $x < 1$  from the center of the sphere. The image potential at  $x$  is given by [34]

$$V_{\text{im}}^{\text{sph}}(x) = \frac{-q}{4\pi\varepsilon_0} \frac{\varepsilon_r - 1}{\varepsilon_r + 1} \sum_{n=0}^{\infty} \left( \frac{(n+1)x^{2n}}{n+1-\delta} \right) \quad , \quad (28)$$

where  $\delta = \frac{1}{\varepsilon_r + 1} < \frac{1}{2}$ . In order to simplify the notation, we define

$$q' = \frac{-q}{4\pi\varepsilon_0} \frac{\varepsilon_r - 1}{\varepsilon_r + 1} \quad .$$

Noting that  $\left(1 - \frac{\delta}{(n+1)}\right)^{-1} = 1 + \frac{\delta}{(n+1)} + \frac{\delta^2}{(n+1)^2} + \dots$ , we may expand the image potential in powers of  $\delta$ ,

$$\begin{aligned} V_{\text{im}}^{\text{sph}}(x) &= q' \sum_{n=0}^{\infty} \left( \frac{x^{2n}}{1 - \frac{\delta}{n+1}} \right) \\ &= q' \sum_{n=0}^{\infty} \left( x^{2n} + \delta \frac{x^{2n}}{n+1} + \delta^2 \frac{x^{2n}}{(n+1)^2} + \dots \right) \end{aligned} \quad (29)$$

The terms on the right hand side become smaller with increasing power of  $\delta$ , so that the zero order term is the leading term of the image potential. Noting the closed form

$$\sum_{n=0}^{\infty} x^{2n} = \frac{1}{1-x^2} \quad (30)$$

we get

$$V_{\text{im}}^{\text{sph}}(x) = q' \left( \frac{1}{1-x^2} + O(\delta) \right) \quad (31)$$

The zero order term in the  $\delta$  expansion of the image force is eventually,

$$F_{\text{im}}^{\circ} = \frac{q}{4\pi\varepsilon_0} \frac{\varepsilon_r - 1}{\varepsilon_r + 1} \frac{x}{(1-x^2)^2} \quad (32)$$

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