Simulation of Charged Particle Guiding Through Insulating Nanocapillaries

K Schiessl¹, C Lemell¹, K Töksi² and J Burgdörfer¹

¹Institute for Theoretical Physics, Vienna University of Technology, Wiedner Hauptstraße 8 – 10, A-1040 Vienna, Austria, EU
²Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), H-4001 Debrecen, Hungary, EU
E-mail: klaus@concord.itp.tuwien.ac.at

Abstract. We report on our model for transport of charged particles through insulating nanocapillaries. The dynamics of transmitted ion beams prior to equilibrium is investigated. Extension of the transport theory to electron projectiles reveals that different mechanisms are responsible for their deflection in capillaries. Charge-up is no longer a prerequisite to establish guided transmission.

1. Introduction
The field of guided transmission of charged particles through insulating nanocapillaries has recently received fast-growing interest. Experiments with highly charged ions (HCI) have been pioneered by Stolterfoht et al. who observed large angle deflection by polyethylene terephthalate (PET, “Mylar”) nanocapillaries [1]. The key property of what is called “guiding effect” is the transmission of HCIs in their initial charge state for angles of incidence $\theta$ larger than the geometric opening angle of the capillary. The following scenario of HCI guiding has emerged [2]: during the charge-up phase, HCIs hit the capillary wall and deposit their charge. These charges are transported along the surface and into the bulk of the target material. Projectiles entering the capillary subsequently are deflected due to the Coulomb field of the deposited charges and may eventually be transmitted. Guiding (stable transmission conditions) sets in as soon as a dynamical equilibrium between projectiles hitting the capillary wall (charging) and transport into the bulk or to the capillary exits (discharging) has been established. Based on this scenario numerical simulations could reach reasonable agreement with experiments.

Since this first observation a variety of systems with different geometry and/or different insulating materials have been investigated, such as SiO$_2$ [3, 4], Al$_2$O$_3$ [5, 6], and glass [7]. The qualitative picture observed appears to be similar in all cases. Moreover, macroscopically large, millimeter-sized, tapered glass capillaries have paved the way to the formation of HCI microbeams [8, 9] with a size of $\gtrsim 1$ $\mu$m. Such beams have successfully been used in irradiation experiments of biological material [10].

The most recent development in this field is the observation of guided transmission of electron beams through insulating Al$_2$O$_3$ nanocapillaries [11]. The observation of a strong inelastic component of transmitted projectiles [12, 13], however, points to a fundamentally different mechanism for guided transport. We have recently developed a classical transport
theory (CTT) describing electron transmission through nanocapillaries which allows inclusion of quantum diffraction effects and is able to describe key findings of the experiment [14]. The most important mechanisms for establishing guided transmission of electrons are identified to be elastic and inelastic scattering at the internal surface as well as in the bulk. Contrary to the case of HCI guiding, charge-up is no longer *conditio sine qua non* for electron guiding. One consequence of our model is the prediction of guided transmission of slow electrons also through metallic capillaries.

In this contribution we first review our classical transport theory and then provide recent applications for both electrons and HCIs as projectiles. After describing the method (Sec. 2) we focus on the dynamics of HCI guiding in Sec. 3. Key aspects of guided electron transmission are discussed and first simulation data conclude our contribution (Sec. 4).

2. Classical Transport Theory of Charged Particle Guiding

A theoretical description and *ab-initio* simulation of guiding of charged projectiles poses a considerable challenge in view of the widely disparate times scales simultaneously present in this problem [2], spanning more than fifteen orders of magnitude: while the microscopic charge-up and charge transport occurs on an atomic (i.e. sub-fs) time scale, the time between two projectiles entering the same capillary is already of the order of a fraction of a second for typical experimental parameters [1]. The macroscopic discharge of the internal surfaces of the insulating materials may take some hours or even days. Such multi-scale problems represent a major challenge for any *ab-initio* simulation. We have therefore developed a mean-field classical transport theory (CTT) [15] based on a microscopic classical-trajectory Monte Carlo simulation for the projectiles $Z^q$ with charge $q$. The forces governing the trajectory are self-consistently coupled to the charge-up of and charge transport at the internal capillary walls [2]. The model adopted relates the microscopic projectile-surface interaction and projectile transport to known macroscopic properties of the materials involved such as conductivity, dielectric constant, material composition, and electronic structure. Despite of the complexity of the processes involved, we aim at a description of the projectile guiding without resorting to freely adjustable parameters or additional ad-hoc assumptions [16]. Figure 1 shows a schematic view of the capillary geometry considered.

2.1. Wall interaction at impact

We study the interaction between charged projectiles, either electrons or HCIs, and the internal surface of the capillary. Upon impact on the internal capillary wall HCIs undergo a sequence of charge transfer process eventually leading to complete neutralization [17]. The secondary electron emission coefficient is low for the HCI parameters (velocity, charge state) considered in this work. Moreover, as long as secondary electrons do not leave the nanocapillary, the aggregate
charge deposited by the HCI impact is equal to its initial charge state \( q \). In the simulation it is assumed that \( q \) elementary charges are Gaussian distributed with a width of distribution \( r_d \approx 1000 \) a.u.. The electrostatic potential in the capillary is updated accordingly after each impact.

For electrons, the interaction with a solid surface is clearly different. In particular, at grazing incidence electrons may scatter quasi-elastically at the attractive surface potential without penetration of the material. This genuinely quantal reflection takes place with a probability that is largest for low incident energies and \( \theta_{in} \rightarrow 0 \). When penetrating into the bulk electrons undergo a cascade of scattering processes which may result in either absorption and deposition of their charge, or in re-emission of the incident projectile. Each of these two processes can be accompanied by secondary electron emission (SEE) leaving then a positive charge \(+|e|\) behind. Depending on the SEE coefficient \( \delta \) the charge-up may even be of a polarity opposite to the (negatively charged) projectile.

Electron transport within the target material is modelled using a classical transport simulation described in more detail in Ref. [18]. In brief, electrons entering the target material with initial kinetic energy \( E_{in} \) are subject to elastic scattering at constituent atomic potentials and to inelastic scattering at the joint electron gas of the compound material. At the top layer of the surface coherent reflection at one-dimensional planar-averaged surface potential [14] is accounted for. The potential is derived from density functional theory (DFT) calculations of the target material. For DFT calculations the program package “ABINIT” [19] is employed. The reflection probability can be approximately described as \( P_s(\theta_{in}) \approx \exp(-\theta_{in}/\theta_c) \) with \( \theta_c \) depending on the surface material’s properties [14]. For PET and \( E_{in} = 500 \) eV, e.g. \( \theta_c \approx 0.95^\circ \).

Elastic scattering cross sections have been calculated with the ELSEPA package [20]. For metal targets muffin-tin potentials are used, bare atomic potentials for insulator constituents. \( Al_2O_3 \) is described as an amorphous compound of 40% aluminum and 60% oxygen atoms, PET (sum formula \( C_{10}H_8O_4 \)) as a compound of about about 45.5% carbon and 18.2% oxygen. The fraction of hydrogen atoms can be safely neglected due to the small elastic cross section of hydrogen as compared to the other components. From the energy-dependent total cross sections and the fractional densities of constituent atoms the elastic mean free path can be derived. For elastic scattering the atom type is randomly chosen according to the fractional densities and the scattering angle is determined from the energy-dependent differential cross sections for this atom.

The doubly-differential inelastic scattering mean free path is derived from the momentum and energy dependent dielectric constant of the bulk material \( \varepsilon_b(q,\omega) \) [21]. The energy loss in an inelastic collision is transferred to the most weakly bound electrons at the upper edge of the electronic band and a secondary electron is released. The total number of electrons included in our simulation is thus non-conserved. Note that energy loss and SEE represent the major difference of electron-surface interaction as compared to HCI-surface interaction.

### 2.2. Charge transport

Once charge carriers have been deposited on the internal capillary walls, the finite conductivity will lead to charge transport at the surface as well as into the bulk. We note that charge transport is treated on equal footing for both HCI and electron transmission.

The materials employed are extraordinarily good insulators with a bulk conductivity of the order of \( \sigma_b \approx 10^{-16} \) \( \Omega^{-1}m^{-1} \) in the case of PET. Charge transport along the surface, however, may be much faster. In the case of PET the ratio of surface and bulk conductivity is of the order of 100 [22]. Because of the presumable importance of surface conductivity we treat the latter explicitly by performing a two-dimensional random walk for the charge carriers that is governed by a diffusion constant \( D_s \). The diffusion constant is related to the conductivity via \( \sigma_{b,s} = \frac{n e}{kT} D_{b,s} \) using the experimentally determined value for the charge carrier density in Mylar.
$n \cong 10^{18} \text{ m}^{-3}$ [23]. Hence, $D_b$ should be of the order of $10^{-17} \text{ m}^2 \text{s}^{-1}$ and $D_s \approx 10^{-15} \text{ m}^2 \text{s}^{-1}$. We use, in fact, $D_b = 2 \cdot 10^{-17} \text{ m}^2 \text{s}^{-1}$ and $D_s = 100 \cdot D_b$ unless otherwise stated. With a probability of $D_b/D_s$ the charges are allowed to enter the bulk material. Subsequently, charge transport is governed by bulk conductivity which is approximated by an exponential decay of the charges with time constant $\tau_b \approx (5a)^2/D_b$.

Within the framework of linear response, the bare charges $q$ are screened by the dielectric medium [16]. Denoting the bare Coulomb interaction between a projectile and the charge deposited by $V_C$, the effective screened interaction, denoted by $V_{SCR}$, depends on the location of the deposited charge $q$. In the limiting case that the charge has already diffused into the bulk, bulk screening would apply, i.e. $V_{SCR} = V_C/\varepsilon_r$. We use the static limit $\varepsilon = \varepsilon(q \to 0, \omega \to 0)$ of the dielectric function since the characteristic time for build-up of screening (time interval $\Delta t$ between two subsequent projectiles entering the capillary $\approx 10^{-11} \text{ sec}$) is very large compared to the characteristic time for optical excitations. The dielectric screening of surface charges is determined by the induced polarization, i.e. the “image charge” having the weight $\chi_s = (\varepsilon_r - 1)/(\varepsilon_r + 1)$. The effective residual charge and hence the effective potential is given by $V_{SCR} = V_C(1 - \chi_s) = 2V_C/(\varepsilon_r + 1)$. We employ the effective potential for surface screening which we have recently shown to compare well to experimental data of HCI guiding [16].

3. Dynamics in Guiding of Highly Charged Ions
We consider capillaries manufactured from PET ($\varepsilon_r = 3.3$) with a length of $L = 10 \mu\text{m}$ and a diameter $2a = 200 \text{ nm}$. So far, this is the only target that has been explored systematically, therefore data is available for many incident energies and charge states [25]. Over a large range of parameters, our theory agrees well with the experiment [16]. In previous work we have focused in particular on transmission rates [2], the angular width of the transmitted beam [24], and on the effect of different levels of dielectric screening [16].

In the present work we focus on the investigation of the dynamics of HCI guiding prior to equilibrium which has recently attracted much interest. Already in early experiments direct observation of charging effects, i.e. increase of the transmission rate before equilibrium has been achieved [1]. It was found that the transmission can be approximately described by $T(\theta_{in}, t) \approx T(\theta_{in}, 0) \times \left[1 - \exp(-t/\tau_c)\right]$, where $\tau_s$ and $\tau_c$ are the saturation time at which guiding sets in and the charging time constant, respectively. $T(\theta_{in}, 0)$ represents the equilibrium value for the transmission rate at an angle of incidence of $\theta_{in}$. Such a behavior is, indeed, well reproduced by the CTT simulation for the exemplary case of 3 keV Ne$^{7+}$ ions incident under $\theta_{in} = 3^\circ$ onto a capillary with $2a = 200 \text{ nm}$ (Fig. 2 b). The smooth charging characteristics points to a continuous charging process that is finally balanced by beam loss. Expressed in terms of the aggregate charge, $\int_0^t j_{in} dt'$, where $j_{in}$ is the incident current per capillary, $\tau_c$ is connected to a value of $\tau_c \approx 4000 \text{ ions}$. For a beam diameter of about 1 mm this value corresponds to roughly 114 nC of charge deposited on the overall capillary target. Recent experimental work [26] has found a value of about 76 nC for parameters very similar to the ones used here ($\theta_{in} = 2.8^\circ$ and $E_{in} = 3.5 \text{ keV}$), while other measurements (Ref. [27]) find $\tau_c \approx 100 \text{ nC}$ for parameters identical to the present calculations.

In equilibrium, a single principal charge patch is formed at the impact region near the entrance of the capillary (in the first 10-15% of the capillary) deflecting subsequent projectiles (see Fig. 3). Additionally, some charge is accumulated near the exit at later stages (> 200 nC). This secondary patch is, in fact, wide spread. Its center is situated on the side of the capillary that is opposite to the principal charge patch.

Indeed, early simulations [2] of HCI guiding have already shown the transient formation of secondary charge patches in addition to the main charge patch always present. The former are weak compared to the principal patch but they further deflect the trajectories well inside the capillary and lead to a transmitted beam not collinear with the capillary axis. Analogously to
Figure 2. Time dependence of guiding for a PET capillary with a diameter of 200 nm. Ne$^{7+}$ ions are incident at an angle of $\theta_{\text{in}} = 3^\circ$ with $E = 3$ keV. a) Motion of the (one-dimensional) center-of-mass position of the angular distribution of transmitted projectiles. b) Transmitted fraction as a function of aggregate incident charge (time). The blue dashed line represents an exponential fit to the transmission curve, $\tau_c \approx 4000$.

Figure 3. Charge density along the capillary axis for different charge depositions, i.e. different moments in time, as indicated in the figure. A single charge patch near the entrance is dominating, at later times some charge also accumulates near the exit.

the principal patch the secondary patch(es) build-up in time, too, and the transmitted beam is not stationary (Fig. 2 a ) ). In equilibrium, however, the beam is centered at the capillary axis. Note that the additional charge patch does not necessarily dissolve. On the contrary, in the long run a weak secondary patch near the exit may remain and support the guiding of projectiles along the axis.

Earlier theoretical work [2] has suggested the existence of two classes of “guided” trajectories - the first class being deflected only once at the primary charge patch as well as those that are also deflected by secondary patches near the exit. Transient behavior of the transmitted beam prior to equilibrium has attracted considerable interest only recently. Several groups have been able to observe the angular motion of the transmitted beam, e.g. in the case of PET [27, 26] as well as for 7 keV Ne$^{7+}$ ions guided through SiO$_2$ capillaries [4]. Our present results compare well to the recent experiments of Refs. [27, 26].

4. Guided Transmission of Electrons
We have recently extended our CTT to include the interaction of electrons with insulator surfaces (see Sec. 2.1 and Ref. [14]). Inelastic scattering during electron-surface interaction is key to understand the energy distribution of electrons guided through PET nanocapillaries (diameter of $2a = 200$ nm and $L = 10 \ \mu$m [12]). Our simulation yields guided transmission even if charge-up is neglected, i.e. guiding should be observable instantaneously after beam switch-on.
Figure 4. Normalized energy spectrum (lower panel) as well as distribution of $(\theta_{\text{out}}, E)$ (upper panel) for 500 eV electrons incident under $\theta_{\text{in}} = 3^\circ$ with respect to a PET capillary’s axis. The energy and angular spread of the incident beam is 20 eV and 1° FWHM. Solid blue triangles show experimental data [12], solid lines result from our electron transport simulation without charge-up.

Figure 4 shows key aspects of the mechanism proposed. The upper panel presents the two-dimensional distribution of exit angles and final energies $(\theta_{\text{out}}, E)$ of transmitted projectiles for $E_{\text{in}} = 500$ eV and $\theta_{\text{in}} = 3^\circ$. Irrespective of the final energy, projectiles are “guided”, i.e. they emerge around $\theta_{\text{out}} \approx 0^\circ$. However, the term “guiding” may be misleading in this context as guiding implies elastic deflection of projectiles well above the capillary wall. As a consequence, truly guided projectiles would suffer negligible energy loss. In the case of electrons elastically transmitted projectiles constitute only a minor fraction of the total transmitted beam. Here, the elastic transmission results from both scattering at the planar-averaged surface potential as well as from small-angle atomic scattering. An energy spectrum of transmitted projectiles (integration over $\theta_{\text{out}}$) is shown in the lower panel of Fig. 4 in direct comparison with experimental data [12]. Good agreement is found.

The influence of charge-up depends on the material properties. The SEE coefficient $\delta$ for grazing incidence becomes particularly important. For $\delta < 1$ the surface will be charged negatively and may support transmission due to repulsive charge-up analogously to the HCI case. However, at the incidence energies considered, $\delta \gtrsim 1$ for many materials. The limiting case of $\delta \approx 1$ would lead to a (globally) uncharged surface. “Guided” transmission (Fig. 4) is then only possible due to multiple scattering. For $\delta > 1$, on the other hand, charge-up does take place but with a polarity opposite to that of the projectile. Transmission is thus suppressed rather than enhanced and is expected to decrease as a function of time until a dynamical equilibrium may be reached. Evidence for such a behavior was found for electrons incident onto Al$_2$O$_3$ capillaries with zero degree tilt with respect to the axis [11, 13].

Figure 5. Charge density at the internal wall of the capillary (parameters as in Fig. 4, incident current density of 0.2 nA/mm$^2$). While the total deposited charge is slightly negative, in the impact region near the arclength $b/(2a\pi) = 0.5$ a local maximum of the charge density is observable.
Whether charge-up is enhancing, suppressing, or insignificant for transmission depends on the subtle interplay between secondary electron emission and charge transport. For the particular case of PET nanocapillaries and a tilt of $\theta_n = 3^\circ$, charge-up turns out to be unimportant. Although the total charge is negative, the modulus of the charge density features a local minimum exactly in the region of projectile impact (arclength $b/2\pi a = 0.5$). This is in clear contrast to the case of HCI where the charge density is maximum in this region and thus supports guided transmission [14]. Accordingly, we do not find strong indication of charging in the time-dependence of the total transmission.

5. Conclusions

We have simulated HCI transmission through insulating nanocapillaries using a classical transport theory developed recently. This approach is based on order-of-magnitude estimates of material properties and does not invoke any ad-hoc assumptions nor any freely adjustable parameters. Surface screening is employed for the dielectric response to the charging which has been shown to compare best to recent experimental data [16].

The dynamics of the charging process and the angular characteristics of the transmitted beam has attracted much interest recently and is now revisited in addition to our previously given time-integral analysis of the phenomena observed. We explicitly follow the movement of the transmitted beam prior to equilibrium and find up to two charge patches contributing to guiding for capillary presently investigated.

Guided transmission of electrons is treated by an extension of our approach involving electron transport inside the bulk solid upon scattering at the surface. Key experimental findings such as a strong inelastic component of the transmitted beam can thus be understood. Depending on the subtle interplay between secondary electron emission and charge transport, charge-up may be weak or even hindering transmission due to its positive polarity. In this case guided transmission is still possible by scattering at the internal capillary walls, an effect that might be present also in metallic capillaries.

Acknowledgments

This work was supported by the Austrian Fonds zur Förderung der wissenschaftlichen Forschung under grants No. FWF-SFB016 “ADLIS”, No. 17449, No. 17359, and the TeT Grant No. AT-7/2007. One of us (K.T.) was partially supported by the grant “Bolyai” from the Hungarian Academy of Sciences and the Hungarian National Office for Research and Technology. K.S. also acknowledges support by the IMPRS-APS program of the MPQ (Germany). We are grateful to Aleksandar Milosavljevic for valuable discussions.

References

[1] Stolterfoht N, Bremer J H, Hoffmann V, Hellhammer R, Fink D, Petrov A and Sulik B 2002 Phys. Rev. Lett. 88 133201
[2] Schiessl K, Palfinger W, Tókési K, Nowotny H, Lemell C and Burgdörfer J 2005 Phys. Rev. A 72 062902
[3] Sahana M B, Skog P, Vikor G, Kumar R T R and Schuch R 2006 Phys. Rev. A 73 040901
[4] Skog P, Zhang H and Schuch R 2008 Phys. Rev. Lett. 101 223202
[5] Mátéfi-Tempfli S, Mátéfi-Tempfli M, Piraux L, Juhász Z, Biri S, Pekete E, Iván I, Gáll F, Sulik B, Vikor G, Pálinkás J and Stolterfoht N 2006 Nanotechnology 17 3915–3919
[6] Skog P, Soroka I, Johansson A and Schuch R 2007 Nucl. Instr. and Meth. B 258 145–149
[7] Bereczky R, Kowarik G, Aumayr F and Tókési K 2009 Nucl. Instr. and Meth. B 267 317 – 320
[8] Ikeda T, Kanai Y, Kojima T M, Iwai Y, Kambara T, Yamazaki Y, Hoshino M, Nebiki T and Narusawa T 2006 Appl. Phys. Lett. 89 163502
[9] Ikeda T, Kanai Y, Kojima T M, Iwai Y, Kanazawa Y, Hoshino M, Kobayashi T, Pokhil G P and Yamazaki Y 2007 J. Phys. Conf. Ser. 88 012031 (9pp)
[10] Iwai Y, Ikeda T, Kojima T M, Yamazaki Y, Maeshima K, Imamoto N, Kobayashi T, Nebiki T, Narusawa T and Pokhil G P 2008 Appl. Phys. Lett. 92 023509
[11] Milosavljević A R, Vikor G, Pešić Z D, Kolarž P, Šević D, Marinković B P, Mářeti-Tempfli S, Mářeti-Tempfli M and Piraux L 2007 Phys. Rev. A 75 030901
[12] Das S, Dassanayake B S, Winkworth W, Baran J L, Stolterfoht N and Tanis J A 2007 Phys. Rev. A 76 042716
[13] Milosavljevic A R, Jureta J, Vikor G, Pesic Z D, Sevic D, Matefi-Tempfli M, Matefi-Tempfli S and Marinkovic B P 2009 Europhys. Lett. 86 23001
[14] Schiessl K, Tökési K, Solleder B, Lemell C and Burgdörfer J 2009 Phys. Rev. Lett. 102 163201
[15] Burgdörfer J and Gibbons J 1990 Phys. Rev. A 42 1206–1221
[16] Schiessl K, Lemell C, Tokesi K and Burgdörfer J 2009 Journal of Physics: Conference Series 163 012081
[17] Burgdörfer J, Lerner P and Meyer F W 1991 Phys. Rev. A 44 5674–5685
[18] Solleder B, Lemell C, Tökési K, Hatcher N and Burgdörfer J 2007 Phys. Rev. B 76 075115
[19] Gonze X and et al 2002 Comput. Mater. Sci. 25 478–492
[20] Salvat F, Jablonski A and Powell C J 2005 Comput. Phys. Commun. 165 157–190
[21] Reinhold C O and Burgdörfer J 1997 Phys. Rev. A 55 450–465
[22] dupontteijinfilmscom 2008 Mylar product information
[23] A C Lilly J, Lowitz D A and Schug J C 1968 Journal of Applied Physics 39 4360–4364
[24] Schiessl K, Palfinger W, Tökei K, Nowotny H, Lemell C and Burgdörfer J 2007 Nucl. Instr. and Meth. B 258 150–154
[25] Stolterfoht N, Hellhammer R, Bundesmann J and Fink D 2008 Phys. Rev. A 77 032905
[26] Kanai Y, Hoshino M, Kambara T, Ikeda T, Hellhammer R, Stolterfoht N and Yamazaki Y 2009 Phys. Rev. A 79 021271
[27] Stolterfoht N, Hellhammer R, Fink D, Sulik B, Juhász Z, Bodewits E, Dang H M and Hoekstra R 2009 Phys. Rev. A 79 022901