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Guiding of low-energy electrons by highly ordered Al_2O_3 nanocapillaries

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We report an experimental study of guided transmission of low-energy (200–350 eV) electrons through highly ordered Al_2O_3 nanocapillaries with large aspect ratio (140 nm diameter and 15 μm length). The nanochannel array was prepared using self-ordering phenomena during a two-step anodization process of a high-purity aluminum foil. The experimental results clearly show the existence of the guiding effect, as found for highly charged ions. The guiding of the electron beam was observed for tilt angles up to 12°. As seen for highly charged ions, the guiding efficiency increases with decreasing electron incident energy. The transmission efficiency appeared to be significantly lower than observed for highly charged ions and, moreover, the intensity of transmitted electrons significantly decreases with decreasing impact energy.

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In 2002, Stolterfoht *et al.* [1] reported an unprecedented experiment of transmission of 3 keV Ne^{7+} ions through nanocapillaries of highly insulating polyethylene teraphthalate (PET). Surprisingly, they found that the majority of Ne^{7+} ions survived the surface scattering in their initial charge state, while the angular distributions of the transmitted highly charged ions (HCIs) indicated propagation of ions along the capillary axis. The authors proposed that a self-organized charge-up process inhibits HCIs from hitting the capillary walls and guides ions along the capillary axis. Thereafter, the guided transmission of charged particles through insulating nanocapillaries has attracted considerable attention. Besides the investigation of hollow-atom formation at large distances from the surface [2], these studies might gain important information about the properties of the inner walls of the capillaries and for possible applications (e.g., manipulation of charged particles on the nanoscale) [3].

To the best of our knowledge, the investigation of the guiding effect has been focused so far on the use of positive ions as projectiles, mainly slow (3–7 keV) HCIs. Apart from the first studies done on PET capillaries [1,4–7], new experimental results have been recently reported for the transmission of HCIs through highly ordered SiO_2 [8] and Al_2O_3 [9] nanocapillaries, as well. The experimental results are partially supported by a classical trajectory simulation that relates the microscopic charge-up to macroscopic material properties [10,11]. Nevertheless, a full understanding of different processes involved in capillary guiding has not been achieved yet. In particular, the characteristics of electron guiding are considered to be unknown at the present.

The investigation of transmission of electrons through insulating nanocapillaries might shed further light on the topic. First, it is interesting to compare the processes of self-organized charge-up for projectiles of the opposite sign. For example, the barrier energy for holes and electrons is different; therefore it might influence the bulk conductivity, and thus the discharging constants for positive (negative) charging [12]. Second, due to the low q/m ratio, low-energy electrons are more affected by the electric field than ions and they are a more sensitive tool for the characterization of nanocapillaries. Electron guiding also represents a new challenge for numerical simulations. Note that the guiding of electrons by insulating nanocapillaries has already been anticipated within the frame of investigation of the electron transparency of micrometer-thick porous alumina membranes with closed pore endings [13]. The authors concluded that the negative charging of insulating pores provides "channeling" of electrons, and increases the "transparency" of the membrane. However, this study is not applicable here, since the electrons are transported along the capillary axis only for normal incidence, and also the experimental geometry is not well defined as the straggling of the electron trajectories after being transmitted through the closed pore endings is unknown.

In the following, we present evidence of guiding of electrons through insulating nanocapillaries. The present results cover incident electron energies (E_0) from 200 to 350 eV and tilt angles (ϕ) 0°–12°. We compare our results to the previous ones obtained for positive ions and discuss the characteristics of the angular distributions of transmitted electrons, guiding ability, and transmission efficiency as a function of E_0. Differences in the guiding properties of electrons and HCIs are mainly attributed to the physical characteristics of the projectile itself (q/E ratio), without going into the complex phenomena of self-organized charging and insulator discharging processes. Note that, even for the lowest used en-

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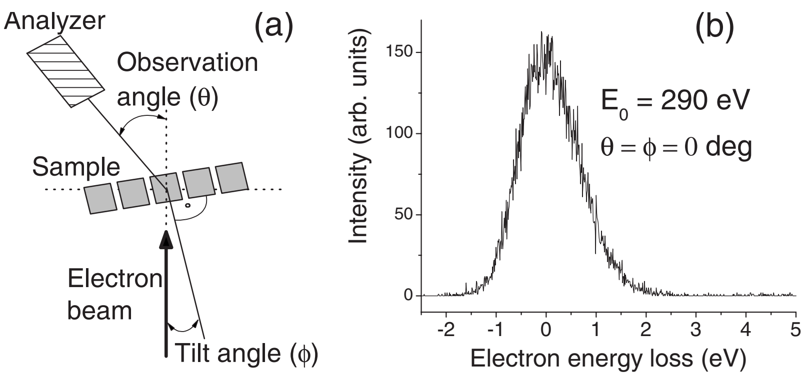

FIG. 1. (a) Definition of the observation (θ) and tilt (ϕ) angles. (b) The energy loss spectrum of transmitted electrons of 290 eV for $\theta=\phi=0°$.

ergy of 200 eV (the limit being imposed by low transmission), the electron velocities are by a factor of 50 larger than for 3 keV Ne HCIs.

A detailed description of fabrication of Al_2O_3 nanocapillaries used in the present experiment has been given recently [9]. Briefly, a highly ordered hexagonally close-packed nanochannel array was prepared using the self-ordering phenomenon during a two-step anodization process of a high-purity (99.999%) 0.5-mm-thick aluminum foil. To prevent a macroscopic charge-up of the target surface, the niobium layers of 20 nm thickness were deposited by dc sputtering on both sides of the final well-ordered honeycomb membrane. The diameter of the used Al_2O_3 capillaries is about 140 nm and the intercapillary distance about 320 nm, while the length is 15 μm. The calculated geometrical transparency is about 8.4%.

The measurements were performed using a modified cross-beam experimental setup, which was described in detail elsewhere [14]. The electron beam is produced by an electron gun, and a sample nanocapillary array was mounted on a target holder, allowing a change of the orientation of the capillary axis with respect to the electron beam direction [see Fig. 1(a)]. The base pressure in the experimental chamber was about 1×10^{-6} mbar. The transmitted electrons were focused into a double-cylindrical-mirror energy analyzer and detected by a single-channel electron multiplier working in single-counting mode. For each experimental point, the intensity was measured at the maximum of the elastic peak obtained in the energy loss mode. A high-pass energy (about 55 eV) of the analyzer was chosen giving a low overall energy resolution of about 1.5 eV [full width at half maximum (FWHM) of the elastic peak], which reduces the dependence of the detection efficiency on the electron energy (see [14]). According to recorded energy loss spectra [e.g., Fig. 1(b)], we conclude that the majority of the transmitted electrons undergo only elastic scattering within the capillaries. During measurements of the angular distributions, the incident beam current was about 14±1 nA. The radius and angular divergence of the incident electron beam were estimated to be 0.5–1 mm and 0.2°–0.5°, respectively (see [14]), thus giving an estimate of 20–80 nA/mm^2 of the primary beam current density. The angular distribution of the primary beam was measured using the analyzer system as a Faraday cup and found to be about 1.8° (FWHM).

The intensities of the transmitted electron beam measured as a function of the observation angle, for tilt angles from −3.5° up to 11.6°, are given in Fig. 2. The data were acquired for E_0 of 200, 250, 290, and 350 eV. The presented distributions were obtained by averaging several measurements and the errors represent the standard statistical error. The centroid of the peak formed by transmitted electrons shifts with changing tilt angle for all electron energies and it matches well with the tilt angle of the capillaries, considering the uncertainty of this angle estimated to be about ±0.5°. It is important to note that the aspect ratio of the capillaries was about 1:110 (diameter:length), so they are geometrically totally closed already at a tilt angle of about 1° (taking also into account the angular spread of the primary beam). Therefore, the presented results give ambiguous evidence of guiding of electrons by insulating nanotubes.

The width of the measured angular distributions does not seem to depend significantly either on ϕ or on E_0. The averaged value of the FWHM is 4.8° for 200 eV (only for zero tilt angle), 4.5°±1.1° for 250 eV, 4.4°±0.2° for 290 eV, and 4.7°±0.7° for 350 eV, where the errors represent standard deviations over all tilt angles. The experimentally obtained widths (Γ_{ex}) should be corrected for the primary beam distribution (Γ_{det}) and the nanocapillary nominal opening (Γ_{nc}). By using the formula $\Gamma_{cp}=\sqrt{\Gamma_{ex}^2-\Gamma_{det}^2-\Gamma_{nc}^2}$ [7], one obtains a corrected average value of Γ_{cp} to be about 4.0°. Although the angular distributions are well fitted by a Gaussian function, as expected (e.g., [4]), we also observed nonsymmetric angular dependencies with a tendency to two-peak distributions (not presented here), which were usually not reproducible during a prolonged observation time. These structures have been predicted by simulations [10] as the result of the formation of a small secondary patch close to the exit surface, which deflects projectiles passing close to it. Of course, the nonsymmetric angular distributions cause a broadening of the Gaussian width, hence giving a larger variation of the FWHM for different tilt angles.

The angular distributions are considerably wider than the primary beam distribution and the value defined by the capillary aspect ratio, as was reported for HCI passing through Al_2O_3 [9] and PET capillaries [1,4–7]. Considering the quite large aspect ratio of the used capillaries, the broad angular distributions suggest a multiple deflection of electrons within the capillary. Still, the broadening of the distribution can also be caused by defocusing the electric field at the capillary exit [8,10,11]. This field could be rather important here, considering the about four orders of magnitude lower m/q ratio of an electron compared to a HCI, as well as the large primary

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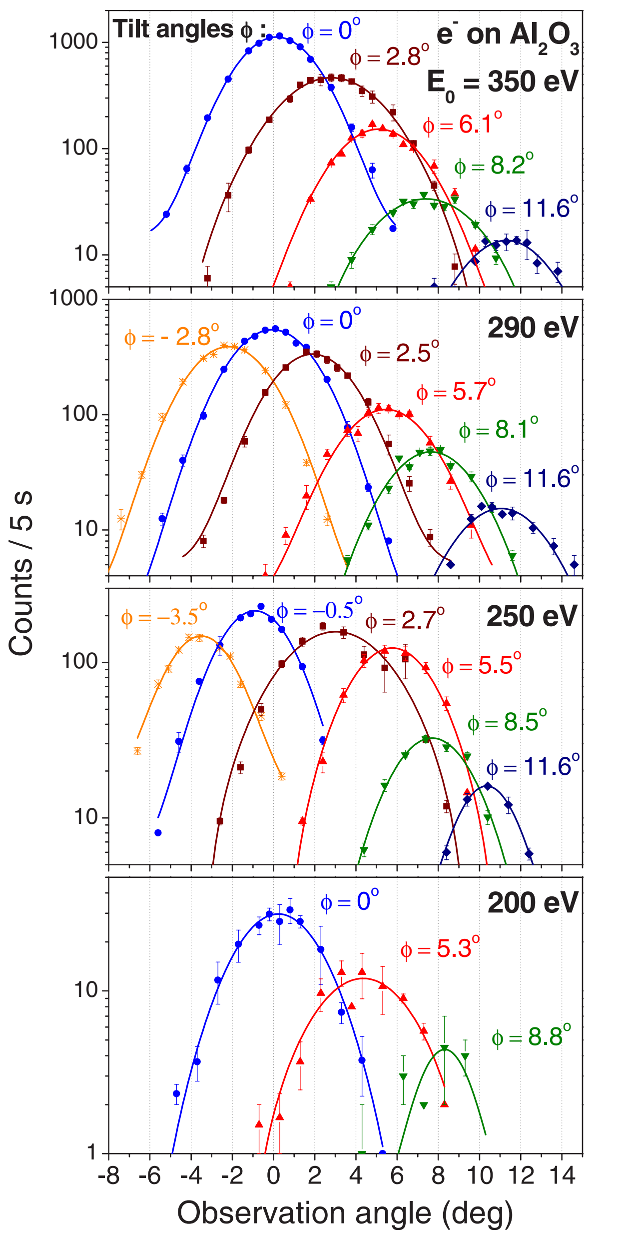

FIG. 2. (Color online) Angular distributions of electrons transmitted through the Al_2O_3 nanocapillaries for different tilt angles (ϕ) and impact electron energies (E_0).

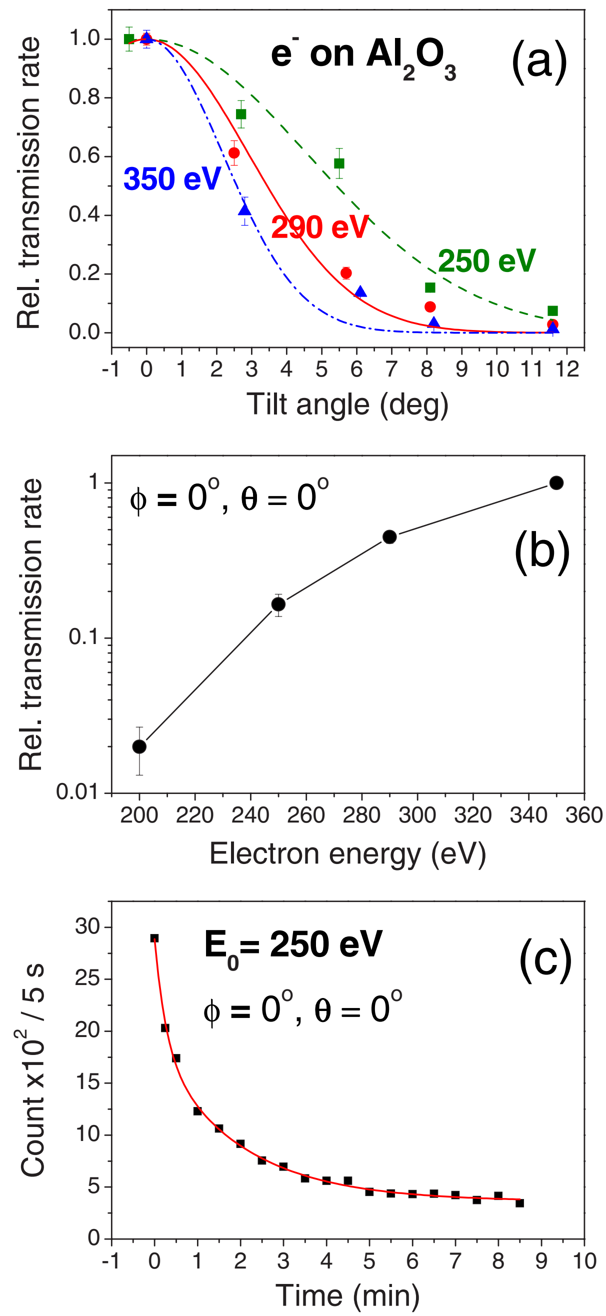

FIG. 3. (Color online) (a) Relative transmission as a function of the tilt angle, for different electron energies (E_0). Experimental points are fitted by the function (1) (see text). (b) Relative transmission as a function of E_0 at zero tilt (ϕ) and observation (θ) angles. (c) Time dependence of the intensity of transmitted electrons at E_0=250 eV and $\phi=\theta=0°$. The data collection starts at about 30 s after the electron beam is turned on, after a discharging time of about 40 min. The experimental points are fitted by a second-order exponential decay function.

current density. On the other hand, the resistivity of nanoporous Al_2O_3 ($10^{11}-10^{14}$ Ω cm) is significantly smaller than for SiO_2 and PET, thus reducing the defocusing effect [8].

The relative dependence of the transmitted electron beam intensity as a function of tilt angle, for different impact energies, is presented in Fig. 3(a). The data are normalized to the intensity measured at 0°. It is evident that by increasing E_0 the slope of the curves gets steeper for increased tilt angle. In order to obtain a quantitative estimation of this effect, the experimental points were fitted to a previously proposed tilt-angle dependence formula [5,9]:

$$f(\phi) = f(0\,°)e^{-\lambda \sin^2 \phi}, \tag{1}$$

where the inverse of the only freely varying parameter λ characterizes the guiding ability of the capillary array. The present results give λ(250 eV)=77±11, λ(290 eV)=192 ±33, λ(350 eV)=344±57. Therefore, the guiding ability of the capillaries increases with decreasing electron energy, as expected. We note that λ=143 was reported for 3 keV Ne^{6+} passing through Al_2O_3 [9]. It is interesting, however, that with decreasing impact electron energy the transmission rate significantly decreases [see Fig. 3(b)]. In our measurements, the guiding was established almost immediately after the beam is turned on, which means that the charging time of the capillaries was much faster than observed in HCI experiments (about 10 min for PET [1]). This is reasonable, however, taking into account the high current density, i.e., while in [1] about 2×10^3 elementary charges enter a single capillary per minute, here about 2×10^5 electrons enter a single capillary per minute. We also note that Doll *et al.* [13] calculated charging time to be less than100 ms for the beam current density of $40\ \mathrm{mA/m^2}$. More interestingly, for discharged capillaries, the measurement of the time dependence of the intensity of transmitted electrons shows a decrease. An example of this effect for 250 eV is given in Fig. 3(c). The experimental points are well fitted by a second-order exponential decay function, with time constants of about $\tau_1=2$ min and $\tau_2=0.3$ min. Finally, the estimated transmission rate (after a sufficient time needed to reach an "equilibrium") at zero tilt and observation angles was about 0.2% for 290 eV electrons, which is about two orders of magnitude lower than the transmission of slow HCIs through SiO_2 and PET.

In our opinion, all these findings could be the consequence of a collective charge-up effect involving simultaneous charging of the entire capillary array near the entrance surface, over the area of the beam spot [11]. For relatively large incoming currents, when charging is faster than discharging, the deflecting electric field increases in time, thus reducing the transmission. Moreover, the large difference in resistivity between Al_2O_3 and the conducting surface will form an electric lens at the capillary entrance (exit), thus affecting strongly the trajectories of incoming electrons. Intuitively, with decreasing impact energy, the electrons are more sensitive to the electric field, which can explain the energy dependence of the transmission [Fig. 3(b)]. By using a simple model of a charged flat surface, one can estimate the influence of a small charged patch on the trajectory of a charged particle passing close to it, for different kinetic energies and charge states. This estimate gives us the ratio of the deflection (y) of the trajectory of an electron with energy E_e to that for an ion with energy E_i and a charge state q_i, which is simply given by $y_e/y_i=M/qi$, where $M=E_i/E_e$. Therefore, as the energy of electrons decreases, they are strongly deflected along randomly formed secondary paths inside the capillaries (as an example, already a $5\times5\ \mathrm{nm}^2$ patch at the capillary entrance, uniformly charged by $5e$, will prevent an electron of 300 eV from reaching the exit in a straight line, for the present capillary aspect ratio $\approx 0.5°$) and influenced by the entrance (exit) lens effects, thus reducing the transmission in comparison with HCIs. Further, it is straightforward to see from the above formula that HCIs are more sensitive to the capillary charging than low charged ions of similar energies, as was recently shown for 3 keV Ne^{7+} and 1.6 keV H^+ [5]. Finally, it is important to note that also the transmission efficiency of HCIs appears to be significantly lower for Al_2O_3 than for SiO_2 and PET [15].

In summary, we have presented evidence of guiding of low-energy electrons (200–350 eV) through highly ordered insulating Al_2O_3 nanocapillaries with a large aspect ratio. The guiding ability depends on the electron energy. For 350 eV, the transmitted intensity decreases up to a factor of 100, when increasing the tilt angle up to about 12°. The angular distributions of transmitted electrons are wider than expected from the capillary aspect ratio, and are weakly dependent on the tilt angle and the electron energy. The transmission of electrons appears to be significantly lower than found for HCI. Also, it decreases with decreasing electron energy. Our results contribute to a better understanding of complex processes involved in guiding of charged particles by insulating nanocapillaries, give solid support to the theoretical investigation of this effect, and suggest the possibility of using the insulating nanocapillaries for manipulation of electron beams on the nanoscale.

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