Inelastic processes associated with electron guiding through insulating PET

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Guiding of relatively fast (500 and 1000 eV) electrons through insulating polymer nanocapillaries of aspect ratio 50 has been studied. The guiding has been attributed to the self-organizing charge-up of the inner walls of the nanocapillaries, which, after a characteristic charging time, deflects the traversing ions causing them to be guided through the sample along the capillary axis. The transmitted electron spectra show significant energy losses that increase with foil tilt angle and incident electron energy, a phenomenon not previously observed for slow highly charged ions or for lower energy electrons. In spite of the substantial energy losses both elastically and inelastically scattered electrons are clearly guided.

Keywords: Electronic excitation; ionization of atoms; surface collisions.

La dirección de electrones relativamente rápidos (500 y 1000 eV) a través de polímero nanocapilares de radio 50 ha sido estudiada. La dirección ha sido atribuida a la propia organización de las paredes interiores del nanocapilares, que, después de un tiempo de carga característico, desvía los iones que atraviesan y las hace ser dirigidas a través de la muestra a lo largo del eje capilar. El espectro electrónico transmitido muestran pérdidas de energía significativas que aumentan con la inclinación del ángulo de la hoja de metal y la energía de electrón de incidente, un fenómeno no observado antes para iones altamente cargados lentos o para electrones de energía inferiores. A pesar de las pérdidas de energía sustanciales los electrones dispersados elástica e inelásticamente pueden ser claramente guiados.

Descriptores: Excitación electrónica; ionización de átomos; colisiones de superficie.

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1. Introduction

The guiding of slow highly charged 3 keV Ne$^{7+}$ ions through insulating PET nanocapillaries of 10 µm length and 100 nm diameter (aspect ratio = 100) was first demonstrated experimentally by Stolterfoht et al. [1], which stimulated many new investigations [2-7,11]. Most of the experimental results have been modeled by Stolterfoht and his group [1-4,11], while Schiessl et al. [3] have performed theoretical simulations to help understand the observations. Recently the guiding of slow electrons with observations similar to positive ions has been reported [6].

A schematic of capillary guiding is shown in Fig. 1, in which the tilt angle $\psi$ of the foil and the observation angle $\theta$ are both measured with respect to the incident beam direction. Both sides of the capillary foil are coated with a thin metallic film to prevent the incident ions from building up charge on the surfaces. Initially, incident ions are deposited on the inner surfaces of the insulating capillaries in a self-organizing manner. Monte-Carlo simulations [3] have shown that most of the incoming ions are deposited in the entrance region of the capillaries and therefore form multiple charge patches in that region. Hence, this entrance region has been called the patch region. Additional charge patches (secondary and tertiary) could also be formed deeper inside the capillaries after the entrance patches are formed but their contribution to ion guiding is less significant [3]. The ion deposition takes place for certain period of time, known as the characteristic charging time with time constant ($\tau_c$) for capillary charging, and produces an electric field inside the capillaries. Under equilibrium conditions this electric field provides an effective “trampoline” for subsequent incoming ions, causing them to be guided along the capillary axis to the capillary exit. In this case the ions are scattered at distances greater than the critical distance

$$R_c = \frac{\sqrt{2q \psi}}{W_\Phi},$$

where $q$ is the charge state of the incident ions and $W_\Phi$ is the work function of the material, inhibiting electron capture by the incident ions from the capillary walls [1-3]. It is found that the angular widths, i.e., full width at half maximum (FWHM) $\Gamma$, of the transmitted ions are nearly constant and independent of the foil tilt angle $\psi$ [1,2-4]. To understand these observations, the capillary is further divided into two regions: scattering and guiding [2-4]. These regions are considered to be independent, i.e., processes occurring in the scattering region do not affect those in the guiding region. In the first region incident ions are scattered by the already deposited ions (entrance patches) from one side of the wall to the other side and retain memory of the incident angle direction. After several scatterings they completely lose their memory of the incident angle and enter the guiding region where they are directed along the axis and exit the capillary with a FWHM independent of $\psi$. As most of the charges are deposited in the entrance region [1-3], it is believed that the
In this paper we report the guiding of 500 and 1000 eV incident electrons through insulating polyethylene terephthalate (PET or Mylar) nanocapillaries of length 10 µm and diameter 200 nm (aspect ratio 50) and associated elastic and inelastic processes. The angular distributions of transmitted electrons show that electrons are guided along the axis of the nanocapillaries in a fashion similar to positive ions [1,2]. The essential aspect of capillary guiding is the ability of the capillaries to transmit charged particles for foils tilted at an angle \( \psi \) with respect to the incident beam direction. This guiding power can be quantified as the characteristic guiding angle \( \psi_c \) [4], defined as the angle at which the transmitted intensity falls to \( e^{-1} \) of its value at \( \psi = 0^\circ \). In order to determine \( \psi_c \), spectra for electron observation angles \( \theta \approx \psi \) (i.e., observation angle \( \approx \) foil tilt angle) were considered as shown in Fig. 3, since these spectra represent the maximum transmitted intensities for each tilt angle. It is seen that the electron energy spectrum for \( \psi = 0^\circ \) has nearly the same profile and width as the spectrum measured for the electron beam obtained from the bare filament (i.e., without the sample) as shown by the dashed curve in Fig. 3a. The small inelastic contribution on the low-energy side of the \( \psi = 0^\circ \) spectrum is due to the angular spread of the incident beam and capillary nonparallelism, which had full width half maximum (FWHM) values of \( \sim 0.25^\circ \) and \( \sim 0.2^\circ \), respectively.

2. Experimental results and discussion

The measurements were performed at Western Michigan University in collaboration with the Hahn-Meitner-Institut (HMI) Berlin, Germany. Figure 2 shows the angular distributions of transmitted electrons for 500 and 1000 eV electrons, demonstrating that electrons are guided along the axis of the nanocapillaries in a fashion similar to positive ion guiding. However, the transmitted electron spectra show considerable energy losses that increase with foil tilt angle (\( \psi \)) and incident electron energy. This energy loss, presumably due to close encounters of electrons with the inner surfaces of nanocapillaries, is phenomenon not observed for the guiding of slow positive ions [1,2,4,5] or lower energy electrons [6]. Nevertheless both elastically and inelastically scattered electrons are clearly guided through the PET capillaries.
Table I. Values of the characteristic guiding angle $\psi_c$ for the elastic and inelastic contributions associated with the electron energy spectra measured in the present work for electron guiding through PET nanocapillaries compared with the corresponding values for positive ions, also for PET foils.

<table>
<thead>
<tr>
<th>$E/q$ (eV)</th>
<th>Elastic</th>
<th>Inelastic</th>
<th>Ions</th>
</tr>
</thead>
<tbody>
<tr>
<td>500</td>
<td>2.0</td>
<td>2.8</td>
<td>4.7$^a$</td>
</tr>
<tr>
<td>1000</td>
<td>1.6</td>
<td>2.1</td>
<td>2.9$^a$</td>
</tr>
</tbody>
</table>

$^a$ From References 4 and 11

Figure 4. (a) Plot of $I_{\text{inelastic}}/I_{\text{elastic}}$ as a function of tilt angle $\psi$ showing that the inelastic contribution dominates the transmitted electron intensity and increases with $\psi$ and the incident electron energy. (b) Exponential fits to the elastic and inelastic contributions to the transmitted intensities for 500 and 1000 eV electrons for determination of the associated characteristic guiding angles $\psi_c$.

Figure 5. Relative transmission intensities vs. tilt angle for incident 500 and 1000 eV electrons. Three contributions – direct, elastic, and inelastic – are required to represent the overall transmitted electron intensity (see text and Ref. 7).

Figure 6. Angular distributions of inelastically and elastically scattered electrons through PET nanocapillaries. Total refers to the integrated transmitted intensity without separating the elastic and inelastic contributions (see Fig. 2).
This result indicates that for \( \psi = 0^\circ \) electrons are transmitted through the foil with negligible interactions with the capillary walls. However, the transmitted electron spectra for \( \psi > 0^\circ \) (Figs. 3b-3f) show significant energy losses, suggesting that a fraction of the incident electrons undergo inelastic scattering with the inner surfaces of the nanocapillaries [7]. This energy loss is quite unexpected in view of earlier results for low-energy positive ions [1,2,4,5] and lower energy (200-350 eV) electrons [6], for which no appreciable energy loss was observed even for rather large foil tilt angles.

The elastic \( I_{\text{elastic}} \) and inelastic \( I_{\text{inelastic}} \) contributions associated with each of the transmitted spectra were then evaluated separately by superimposing the \( \psi = 0^\circ \) spectrum onto the \( \psi > 0^\circ \) spectra and normalizing to these latter spectra as shown in Figs. 3b-3f. The shaded areas are the elastic contributions. The inelastic contribution was then obtained by subtracting the elastic contribution (shaded area) from the corresponding integrated spectrum [7]. A plot of the ratio, \( I_{\text{inelastic}}/I_{\text{elastic}} \), as a function of foil tilt angle \( \psi \) shows that inelastic contributions dominate the transmitted electron energy spectra with the relative contribution increasing with \( \psi \) and incident electron energy as shown in the Fig. 4a.

The characteristic guiding angles \( \psi_c \) for the elastic and inelastic contributions for \( \psi \neq 0^\circ \) were determined for 500 and 1000 eV (see Fig. 4b) and are listed in Table I along with the corresponding values obtained for positive ions [4,9]. It is seen that the values of \( \psi_c \) obtained for electrons in the present work are smaller than the corresponding values for positive ions, indicating that electron guiding falls off faster than for positive ions. Also, the values of \( \psi_c \) are different for the elastic and inelastic contributions, with the elastic contribution decreasing faster. In order to represent the transmitted intensity for all tilt angles a direct contribution (no guiding) corresponding to the large transmitted intensity at \( \psi = 0^\circ \) must be included as shown in the Fig. 5. The direct contribution, which is due to a convolution of the capillary aspect ratio, the beam collimation, and capillary nonparallelism, decreases very rapidly for \( \psi > 0^\circ \) as expected [7].

Using the same superimposing and normalizing technique, the elastic and inelastic contributions associated with each transmitted spectrum for \( \theta \neq \psi \) for each tilt angle \( \psi \) were evaluated separately. The resulting plots of the angular distributions of elastically and inelastically scattered electrons show that the angular centroids for both contributions coincide and are nearly equal to the tilt angle, \( \theta \approx \psi \), demonstrating that both elastically and inelastically scattered electrons are guided as shown in Fig. 6. The large inelastic contribution in the transmitted electron spectra suggests that insufficient (negative) charge is deposited on the inner walls of the capillaries to electrostatically prevent the traversing electrons from interacting strongly with the walls. Consequently, electrons inside the nanocapillaries undergo both elastic and inelastic scattering with the inner surfaces of the capillaries, with the result that some electrons are lost in the capillaries and those that make it through lose energy.

It should be noted that the energy of the incident electron beam (500 and 1000 eV) is much higher than the values of electronic properties (e.g., band gap, electron affinity etc.), the valence and K-shell ionization energies of carbon, oxygen and hydrogen atoms, as well as the covalent bonding energies among the elements of the PET sample [11-14]. As a consequence various inelastic processes are likely to occur due to impact of the high energy electron beam on the PET sample [12-21]. Specifically, the electron beam can excite or ionize the C, H and O atoms leading to possible Auger processes (in C and O) and generation of low energy secondary electrons [13,14,19].

Additionally, inelastic processes can lead to cleavage or rearrangement (e.g., formation of C = C bonds) [13,14,18], stretching (C–H stretching in CH2 groups) [13,14], breaking of the C–C, C–O, C=O, C–H, C–O–C, or any other bonds, as well as affecting even the most stable benzene ring [13,14,21]. As a result various volatile products, e.g., CO2, CO, O2, H2, hydrocarbons (CH4, C2H2, C3H6, etc.), and cations/anions can be produced [13,14,17]. Thermally, the incident electron beam can locally heat up the interaction area, thereby producing significant electron emission from the surface, which in turn, increases the conductivity exponentially, leading to thermal breakdown of the polymer [13,14]. Furthermore, polymers also contain permanent space charges [12,13], causing incoming electrons to become trapped locally and build up a corresponding space charge according to Poisson’s equation \( \nabla \cdot E = \rho/\varepsilon \), where \( \rho \) is the charge density of trapped electrons and \( \varepsilon \) is the permittivity of the polymer [13,16].

Due to these various inelastic processes the PET polymer can undergo both physical and chemical degradation upon irradiation by high energy electron beams [13,14]. It should also be mentioned that the age of the polymer and improper maintenance of the sample can contribute to its degradation [13,14]. Apart from processes leading to degradation, a survey of the literature reveals that the fundamental dynamics of the scattering of electrons with insulators, unlike charged particle interactions with metals [22], has not been extensively studied due to the limitations imposed by charge build-up on the insulator surfaces.

3. Conclusions

We have shown that electrons are guided through PET nanocapillaries in a fashion similar to positive ions. The transmitted electron spectra show significant energy losses which increase with foil tilt angle and incident electron energy. Three contributions – direct, elastic, and inelastic – are required to represent the overall transmitted intensity. Despite significant energy losses, inelastically scattered electrons are guided along with those elastically scattered. Several mechanisms leading to degradation of the polymer by inelastic processes are suggested. As a result inelastic electron guiding may provide a means to understand the fundamental interaction dynamics between electrons and insulators.
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