Electron Transmission Characteristics and the Production of Narrow Beams Using Glass Optics

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ELECTRON TRANSMISSION CHARACTERISTICS AND THE PRODUCTION OF NARROW BEAMS USING GLASS OPTICS

by

Buddhika Senarath Dassanayake

A Dissertation
Submitted to the
Faculty of The Graduate College
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Advisor: John A. Tanis, Ph.D.

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TRANSMISSION OF ELECTRONS THROUGH AN INSULATING SINGLE CYLINDRICALLY-SHAPED GLASS CAPILLARY OF MICROSCOPIC DIMENSION HAS BEEN INVESTIGATED. SAMPLES MADE WITH BOROSILICATE GLASS (PYREX 7740) WERE SUBJECTED TO BOMBARDMENT OF 300-1000 eV ELECTRONS. TRANSMITTED ELECTRONS WERE ANALYZED USING A PARALLEL-PLATE SPECTROMETER COUPLED TO A CHANNEL ELECTRON MULTIPLIER.

THE TRANSMITTED ELECTRON INTENSITY WAS FOUND TO DECREASE WITH INCREASING SAMPLE TILT ANGLE RELATIVE TO THE DIRECTION OF THE PRIMARY BEAM. TWO REGIONS OF TRANSMISSION WERE FOUND: DIRECT WHERE THERE IS NO INTERACTION OF THE BEAM WITH THE INNER CAPILLARY WALL, AND INDIRECT WHERE THE BEAM DOES INTERACT WITH THE WALL. THE RATE OF TRANSMISSION FALLOFF IN THE DIRECT REGION WAS INDEPENDENT OF THE PRIMARY BEAM ENERGY, WHEREAS A MAXIMUM IN THE RATE FOR 500 eV WAS OBSERVED FOR THE INDIRECT REGION. RUTHERFORD SCATTERING WAS FOUND TO BE DOMINANT AT LOWER ENERGIES (< 500 eV), WHILE COULOMBIC REPULSION DUE TO CHARGE DEPOSITION TOOK OVER AT HIGHER ENERGIES. WHEN THE SAME EXPERIMENT WAS REPEATED USING AN ANGULAR RESOLUTION SPECTROMETER WITH TEN TIMES BETTER RESOLUTION, THE TRANSMITTED ELECTRON INTENSITIES REVEALED TWO DISTINCT REGIONS WITH DIFFERENT CHARACTERISTICS WITHIN THE INDIRECT REGION INSTEAD OF JUST ONE. THE REGION OF LOWER SAMPLE TILT ANGLES WAS DOMINATED BY TRANSMISSION DUE TO COULOMBIC REPULSION, WHILE FOR LARGER TILT ANGLES INELASTIC SCATTERING OF Incoming ELECTRONS AT THE CAPILLARY WALL DOMINATED THE TRANSMISSION.
Charge deposition inside the capillary was also studied as a function of time and found to be time (charge) dependent, confirming the existence of transmission based on Coulombic repulsion. The transmission intensity showed oscillatory behavior in the indirect region at equilibrium, suggesting a sudden discharge of the capillary followed by slower recovery as the charge build up goes toward equilibrium. Stable transmission equilibrium was never reached due to repeated sudden partial discharge of the inner capillary from time to time. Evidence of initial beam deflection due to charge accumulation at the capillary entrance is in agreement with observations on slow highly charged ions.
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CHAPTER I
INTRODUCTION

Transmission of slow (a few keV) highly charged ions (HCIs) through insulating nanocapillaries has been the focal point of many experiments in recent years due to the need for fundamental understanding of ion insulator interactions [1], as well as future technical applications [2, 3]. The ability of HCIs to traverse through insulating nanocapillary foils with a large fraction of them retaining their initial charge state and negligible energy loss [1] is called "guiding", which is due to charge build up on the inner walls of the capillaries. Investigations of the transmission phenomenon and the variation with time of the transmitted beam position have proved to be significant in learning about the dynamics of the guiding process. These studies for slow HCIs have also revealed that a small finite number of secondary charge patches are sequentially formed inside the capillaries before the transmitted pattern reaches equilibrium [4-6].

Studies with insulating nanocapillary foils suggest that the guiding effect for ions should also exist in single glass capillaries of macroscopic dimensions [7], an effect that was observed in recent measurements [8, 9]. In fact, investigations [10] with glass capillaries have observed multiple oscillations of the transmitted beam on a position sensitive detector, suggesting the dynamics of the charging process are in agreement with a charge patch model.

Unlike slow ion guiding, transmission of faster electrons through nano- and macrocapillary foils has been reported to initiate almost immediately [11, 12]. However, compared to the enormous amount of research for slow ions [1-10], so far
little has been done to investigate electron transmission dynamics, experimentally [11-13] or theoretically [14]. Consequently, many questions regarding the transmission process remain unsolved.

In the following discussion, HCI transmission through nanoscale capillaries and microscopic glass capillaries, as well as electron transmission through nanoscale capillaries, are discussed briefly in order to signify the essentiality and importance of the research that is being done.

**HCI transmission through nanoscale capillaries**

The ability of HCIs to traverse through capillaries with a large fraction of them retaining their initial charge state with negligible energy loss triggered the attention of many groups. This followed the discovery study of the guiding process in 2002 by Stolterfoht et al. [1]. As a consequence of this guiding, both the tilt angle of the sample with respect to the incident beam and the observation angle at the point of maximum transmitted beam intensity through the nanocapillary at a given tilt angle, have been found to vary in a linear fashion.

It has been proposed that the observed ion transmission through the insulating capillaries is governed by charge deposition and the resulting beam reflection. With ions incident on the entrance to the nanocapillary the deposited charge increases until the resulting electrostatic field is strong enough to deflect incoming ions towards the capillary exit. After some time, the deposition close to the entrance remains steady at an amount which is sufficient to maintain the field for ion deflection at the equilibrium state of the transmission [15]. At the exit of the capillary, the transmitted ions are subjected to another electric field due to charge deposition, and the outgoing beam is defocused (broadened), giving rise to an enlargement of the transmitted beam.
When the ions transmit through a capillary, additional weaker secondary charge patches can also be created following the first charge patch inside the capillary. The exact number of charge patches for a given case is believed to be dependent on material properties, capillary geometry, as well as the charge and the kinetic energy of the incident ions. The formation of these secondary charge patches can cause oscillatory ion trajectories, which, in fact, have been experimentally observed [4].

It has also been reported that an increase in incident current does not significantly affect the entrance charge patch, since additional current can flow along the surface of the capillary at the equilibrium state [15]. The amount of charge deposited in the primary charge patch formed close to the entrance has been found to be nearly constant at equilibrium, irrespective of the capillary diameter [16]. Furthermore, surprisingly, the ion guiding ability has also been found to be independent with diameter variations. So, the guiding ability of nanocapillaries (capillary diameters of few hundred nanometers), as well as macrocapillaries, which can have diameters two orders of magnitude larger with respect to nanocapillaries, is believed to have almost the same guiding capabilities [16].

Time evolution studies of ion guiding through nanocapillaries can provide evidence for the existence and progression of secondary patches inside the capillary. Several studies of dynamic charging inside the capillaries have yielded evidence for this scenario. In a recent study of variations in the mean emission angle of the exiting ion beam with respect to deposited charge on the capillary [5], it has been reported that the aspect angle of the capillary plays a role in determining the number of temporary secondary charge patches formed. Consequently, capillaries with larger
diameters produce fewer charge patches. After reaching the equilibrium (or steady state) of transmission, ions were found to be guided along the capillary axis, even when the sample was tilted to higher angles.

The guiding effect of slow HCIs has been observed for insulating capillaries of different densities and diameters, such as polyethylene terephthalate (PET) [1, 5, 15, 16], SiO$_2$ [4, 17], Al$_2$O$_3$ [18], and polycarbonate [5], in the last few years. The overall transmission mechanism for these samples has been found to be similar, irrespective of the capillary material, though the quantitative transmission parameters were found to be somewhat different for each case.

**HCI transmission through single glass capillaries**

Many of the experiments on capillary guiding reported so far have focused on insulating foils with randomly distributed capillaries [1, 4-6]. Thus, the collective effect of all the capillaries has to be taken into account when studying the guiding process, making the problem complex. On the other hand, studying the transmission through a single capillary makes the situation simpler and offers the possibility of producing microbeams, which can be useful in various technical applications.

Studies with insulating nanocapillary foils suggest that the guiding effect for ions should also exist in single glass capillaries of macroscopic dimensions [7], an effect that was observed in recent measurements [8, 9]. In these studies both the tilt angle of the sample and the observation angle of the transmitted beam were found to vary in a linear fashion, providing clear evidence for the guiding process. However, transmission only up to a tilt angle of about 5° has been observed for the case of straight glass capillaries [8], in comparison to about ~ 20° for insulator PET nanocapillaries [1].
Time evolution properties of the HCI transmission through straight glass capillaries have been found to be similar to what has been reported for ion guiding through insulating nanocapillary foils. This results in an exponential increase of transmission before leveling off at the equilibrium state, giving rise to a constant transmission [8]. Furthermore, investigations [10] with tapered glass capillaries in the recent past have reported multiple oscillations of the transmitted beam on a position sensitive detector, suggesting the dynamics of the charging process are in agreement with a charge patch model.

**Electron transmission through capillaries**

Recently, experiments have been conducted with intentions of investigating whether the guiding phenomenon seen for ions through insulating nanocapillaries also exists for incident electrons. This has been done for different types of capillary foils. Transmission of faster (200-350 eV) electrons through an Al₂O₃ nanocapillary foil [11] have revealed observations similar to slow ions [1, 4, 5], from which it was concluded that electrons also guide through alumina nanocapillaries. However, for still faster (500-1000 eV) beams [13], it was found that electrons transmit through insulating PET (or Mylar) nanocapillaries but they suffer significant energy losses, unlike the case of alumina.

The energy losses seen for electrons through PET capillaries indicate that electrons suffer one or more close collisions with the inner capillary walls before being transmitted, giving rise to inelastic transmission of electrons. In contrast, Coulombically deflected electrons from accumulated charges inside the capillary and
elastically scattered electrons from the capillary wall give rise to an elastic transmitted fraction of electrons. Despite considerable energy losses, both the inelastically and the elastically scattered electrons are believed to be guided through the sample [13].

The guiding power of electrons is found to be considerably smaller than that of HCIs, due to low transmission of electrons through the capillaries [13]. It has been suggested that close collisions of electrons with the capillary walls, which can give rise to excitation and ionization of atoms or molecules, are the likely causes for the observed lower transmission of electrons compared to slow positive ions [19].

More recently, in theoretical work on electron guiding through PET nanocapillaries it was suggested that the charge up of the surface only plays a minor role in the guiding process, as opposed to ion guiding where strong electrostatic forces are essential for the guiding to be operative [14]. Claims such as these have to be proved still, however, since a proper time evolution study for electron transmission through either nano- or macro- capillaries have not been conducted so far. Thus, the necessity for experimental work to address such claims is vital within the capillary community.

The prime motive behind this dissertation research is to find a model for electron transmission through single glass capillaries by experimentally investigating the properties of transmitted electrons, and to address some of the unanswered questions regarding the fundamentals of the electron transmission process. Furthermore, transmission dynamics of electrons through straight glass capillaries are
studied for the first time together with the energy dependence, presenting a comprehensive analysis of the much less discussed electron transmission process. The findings of this research work should facilitate the future developments of the capillary field.
CHAPTER II
INTERACTION OF ELECTRONS WITH INSULATOR SURFACES

When a solid is irradiated by a beam of electrons, the bombarded surface emits electrons which can be classified into two categories: secondary electrons (SE) which have energies < 50 eV, and inelastically and elastically scattered electrons which have energies less than or equal to that of the primary incident electron beam [20]. When the solid of interest is an insulator, charging of the sample under irradiation, as well as the generation of a surface potential and electromagnetic fields, causes the electron emission process from the surface to become more difficult to predict. As a result there have been many attempts in the last few decades to comprehend the experimental results and theoretical models to justify the observations for the sake of better understanding of the electron interaction with the insulator surface.

When primary electrons (PE) bombard the sample surface, the average number of SE and back scattered electrons (BE) are defined as the secondary electron yield (emission coefficient) $\delta$ and the backscattered electron yield (electron reflection coefficient) $\eta$, respectively. The sum of $\delta$ and $\eta$ is called the total electron emission coefficient $\sigma$. These values are not only functions of the primary beam and its energy, but also depend on the surface topology and composition of the sample [21]. The knowledge of these processes plays a significant role when studying electron interactions with surfaces and the resulting emission.

Variation of the SE yield $\delta$ with incident primary electron energy $E_{PE}$ has a general shape as shown in Figure 1 for all materials (uncharged surfaces): $\delta$ increases
with increasing $E_{PE}$ until it reaches its maximum at $\delta^m$ at $E^m$ before it decreases for higher energies [20]. At points $E_I$ and $E_{II}$ (crossover energies) of the standard emission curve, $\sigma$ is equal to one, where statistically one electron is emitted by each incident electron. Both $\delta^m$ and the corresponding $E^m$ are dependent on the material properties.

![Standard emission curve](image)

**Figure 1. Standard emission curve.**

There have been many attempts both theoretically and experimentally in the last few decades conducted towards better understanding of the secondary emission process. Almost all of the theories predict the functional dependence of emission but not the magnitude [22].

**Conventional approach for secondary electron emission**

The charging mechanism of insulators under electron irradiation has long been considered to be based on the dependence of $\sigma$ on $E_{PE}$. Seiler [20], Lin [23], Agrawal [24] and many others have proposed numerous theoretical mechanisms and have come up with different methods to interpret the total yield variation in the standard
emission curve in the past few decades using the *conventional approach* (or total yield approach) by simplifying the actual sequence of the process. The conventional approach accounts for emission from insulators based on factors such as surface potential (variation of effective beam energy), penetration ($R$), and escape depths ($X_s$).

It is assumed that the escape probability of SE produced at a distance $x$ from the surface decreases as $e^{-x/\lambda}$, where $\lambda$ is the mean escape depth of the material of interest [20]. In Shih *et al.* [22] the shape of the emission curve has been explained in terms of the penetration depth of electrons and the escape depth. At very low primary beam energies ($E_{PE}$) for which $R << X_s$, the internal SE produced can escape efficiently, but due to the low $E_{PE}$ only a few are generated, giving rise to the low yield at the beginning of the curve shown in Figure 1. As SE production increases with the beam energy, the observed yield rises with increasing $E_{PE}$. At very high energies for which $R >> X_s$, though the SE generation is higher, the number of internal SE that escape decreases offsetting the increase in generation of internal SE. Consequently the observed yield decreases with $E_{PE}$. The maximum of the yield is observed when $R \approx X_s$ [22].

When the primary energy of the electrons $E_{PE}$ lies between $E_I$ and $E_{II}$, the emitted current from the sample is higher than the incident current. Thus, the target charges positively and a positive surface potential $V_s$ appears on the sample. Consequently, incoming primary electrons approach the sample with an effective energy of $E_{eff} = E_{PE} + |e|V_s$, greater than the initial primary electron energy $E_{PE}$. On the other hand, if the primary electron energy lies outside the above interval, the emitted current will be lower than the incident and as a result the sample will charge negatively.

According to the conventional method, if the primary energy of the electrons
If the primary energy of the electron beam $E_{PE} > E_t$, the total electron emission coefficient $\sigma$ will follow the standard emission curve until point $E_{Hu}$, at which an emission equilibrium is reached by statistically emitting one electron for every incident electron. The surface potential attained at equilibrium is then given by the difference between the accelerating potential corresponding to point $E_{Hu}$ and the primary initial accelerating potential [25].

If the primary energy of the electron beam $E_{PE} < E_t$, the effective energy of the incident electrons decreases due to the change in surface potential caused by the evolution of the secondary electron emission yield. As a result, the conventional method suggests the total transmitted yield $\sigma$ moves along the standard curve until the effective energy vanishes [25].

Although the conventional method prevailed for a long time and was used to explain the secondary emission from uncharged surfaces [20, 26, 27], it has been substantially revised in last decade for several reasons. The total yield approach is solely focused on the potential created on the sample to describe the emission evolution, whereas the field effects on the emitted electrons have been completely neglected. Especially because the majority of the secondary electrons emitted from the surface have low kinetic energies, the emission of secondary electrons must have a strong dependence on the extrinsic and intrinsic electrostatic fields of the insulator. Factors affecting the emission yield such as primary beam characteristics, chemistry of the insulator and physical properties of the sample, and trap sites also have not been included in the approach. As a result, several authors such as Melchinger, Hofmann [28] and Cazaux [29] have taken more advanced and comprehensive approaches to solve the problem by paying attention to physics beyond the standard emission curve and have attempted to interpret the charging and field effects theoretically by the **dynamic double layer model**.
Dynamic double layer model (DDM)

The model is based on the assumption that the trapped charges are settled in the insulator in two distinct homogeneous uni-axially cylindrical shaped layers. The layer close to the surface is positively charged whereas the other is relatively thick and negatively charged, lying deep inside the insulator. Since the formation of the double layer is solely caused by the electron–insulator interactions that take place in the bulk, it is important to discuss the fate of the primary electrons once they enter the insulator bulk through the sample surface.

When primary electrons enter the insulator, they can scatter elastically from atoms close to the insulator surface, changing the initial direction, or they can lose energy due to various inelastic processes such as ionization, or excitation of inner shell electrons of material atoms, sometimes creating electrons and holes. Due to the large effective mass of holes, they can be easily trapped compared to electrons. Trapping effects are caused by the presence of defects in the material such as impurities, vacancies, dislocations, or grain boundaries [25]. These trapped electrons and holes also give rise to an electric field which can alter the trajectories of the PE as well as the SE. Due to the influence of the generated internal field, trapped particles can also start to drift [30] as shown in Figure 2. The drifting particles can encounter three different circumstances: (1) if the trap site is empty, the particle will settle in the site, (2) if a site is occupied by a charge of the same sign as that of the drifting particle, the particle continues the drift by following the electric field lines acting on it, (3) if the trap is already occupied by a charge of opposite sign, the two charges recombine, causing a disappearance the charges and freeing the trap site [30].
If an electron with very low energy (about 1 eV) is sufficiently accelerated by the internal fields, it is possible to excite an electron-hole pair. This process is called an "avalanche effect". It is not so significant for the internal field to be strong in order for avalanche to take place. Even if the electron can travel a sufficiently large distance under the influence of a weak internal field, it can gain enough energy for the avalanche process to occur [25].

The significance of the DDM is that it pays attention to all of the above mentioned processes and their correlations when discussing the charging effects of insulators, making it more realistic than the conventional method. According to
Melchinger et al. [28] the charge distribution and currents in the DDM method can be viewed as in the figure below.

![Diagram of charge distribution and electron currents](image)

Figure 3. A schematic drawing of charge distribution and electron currents in the dynamic double layer model proposed by Melchinger and Hofmann [28].

The figure indicates the thin positively charged cylinder $Z_s$ close to the surface with a total charge of $Q_s$, from which SE are assumed to originate. Deep in the material a thick negatively charged cylinder is shown by $Z_m$ with a total charge of $Q_m$. The model suggests that the lateral dimension of the positively charged layer $r_s$ is greater than that of the negatively charged layer $r_m$. The difference between the dimensions can be explained by the effects of backscattered electrons. As mentioned
earlier, the positively charged layer is a result of emitted SE. About 40-80% of these SE originate from excitation processes through BEs rather than PEs. Since BE can reach surface areas far away from the primary electron beam diameter on the sample surface, the positive charge created just below the surface can reach higher lateral distances making $r_s$ considerably bigger than $r_m$ [28].

When the primary beam enters the insulator with a current $I_{PE}$, the current associated with emitted secondary electrons $I_{SE}$ is equal to $I_{PE}\delta$. The emitted SEs leave behind an equivalent positive charge $I_{PE}\delta t$, where $t$ is the irradiating time. If the PEs penetrate further into the bulk without generating SEs, they can be reflected (backscattered) from the material at distances less than the penetration depth. The resulting current $I_{RE}$ can be given as $I_{PE}\eta$. Consequently, a negative charge of $I_{PE}(1-\eta)t$ is left within the bulk. The total charge balance of the material depends on these two factors, whereas it is unaffected by the generation of recombination processes of electrons and holes.

According to the DDM, primary electrons continuously excite electrons into the insulator conduction band allowing the electrons to be mobile. This process (which is also called radiation induced conductivity) takes place between the two oppositely charged layers giving rise to the current $I_R$ [28]. In addition, leakage currents $I_L$ can direct the charge carriers through the surface, as well as the bulk to ground, affecting the total charge of the sample. It is noteworthy to mention that bulk current is believed to be smaller compared to surface current in most of the common cases [31]. Charge carriers can also drift and diffuse from irradiated to un-irradiated regions through the material expanding the charged region [32].

The total balance of the currents should obey the conservation of charge at any instance of irradiation, so,
\[ I_{PE} = I_\sigma + I_Q + I_L \]

where, \( I_\sigma = I_s + I_\eta \) is the overall electron current emitted from the surface, which is the addition of both secondary and backscattered currents, and \( I_L \) and \( I_Q \) are the leakage current and the displacement current due to charge trapping, respectively. It is possible to generally express this as

\[ I_Q = \frac{dQ}{dt} = \frac{(dQ_+ + dQ_-)}{dt}, \]

where \( Q \) is the algebraic sum of both positive \((Q_+)\) and negative \((Q_-)\) charges. In the course of irradiation these charges can be represented as,

\[
\frac{dQ_+}{dt} = I_{PE} \delta \left( \frac{E_{PE}}{E_t} \right) N_t^- \omega_t^- 3\lambda - I_R
\]

\[
\frac{dQ_-}{dt} = I_{PE} (1 - \eta) \left( \frac{E_{PE}}{E_t} \right) N_t^+ \omega_t^+ R_0 - I_R
\]

where \( E_t \) is the generation energy of SEs, \( N_t^\pm \) are bulk densities of capture centers of electrons and holes, respectively, \( \omega_t^\pm \) are effective capture cross sections, and \( R_0 \) is the maximum penetration depth of PEs. The equation for positive current is formulated by assuming that SEs are emitted from a thin surface layer of thickness \( 3\lambda \), where \( \lambda \) is the mean free path of SEs [32].

Charge carriers trapped in the material can be released (or detrapped) and once released they may be trapped again depending on the local conditions. Detrapping can occur under the influence of temperature as well as electric field. Though the thermal detrapping alone is not so efficient, when a considerably strong electric field is present conduction can be significantly increased by processes such as the Pool-Frenkel effect (field assisted thermal ionization), in which the potential barrier of the trap is lowered allowing detrapping to accelerate, hence increasing conductivity. Furthermore, when the trap site is deep beneath the surface of the insulator the relaxation of the accumulated energy of the trap site can cause a material
breakdown in the worst case scenario [25].

**Steady-state emission**

SE emission itself is a time dependent process. When electrons are incident on the insulator surface they continue to be trapped in $Z_m$ (for the case of $+\eta < 1$), where the electric field strength is low. The maximum field strength of the system can be found somewhere between $Z_m$ and $Z_e$, where a maximum of electron-hole pair dissociation can be seen and electrons are pushed towards the surface and holes towards the bulk. Electrical neutrality between the layers is attained when the field is large enough to detrap the charges from shallow traps. As a result a depletion zone is formed and a uniform field is generated between the charge layers. The steady state of the system is attained when any additional charge cannot be trapped. This implies that electron and hole pairs generated by newly arriving PEs are rapidly recombined in the bulk, $\frac{dQ}{dt} = 0$, and $\delta + \eta = 1$ [29].

According to Renoud et al. [25], saturation of the insulator also can be determined by the available trap sites in the bulk. With increasing effective beam energy and penetration depth, the available trap sites for the incoming electrons also increase, so a material with a higher density of traps will take longer to reach saturation compared to a low density material. Saturation of the yield is common for both negative and positive charging of insulators, but the formation and distribution of charges in the sample can be different for these two cases.

**Negative and positive charging**

As can be seen in Figure 1, the insulator can be negatively or positively charged depending on the incident primary energy. In either case, due to the generated charge layers, the resulting electric field is directed towards the center of
the negative distribution in the bulk. A large number of electron and hole pairs can be excited due to the field and the electrons moving toward the surface are accelerated along the path. But when the intensity of the electric field becomes too high, detrapping processes start and charges migrate to less populated areas, consequently decreasing the electric field strength with time.

During negative charging of the insulator, the surface acquires a negative potential. As a result, a considerable slowing down is experienced by the primary electron beam before hitting the sample surface, consequently lowering the effective energy \( E_{\text{eff}} = E_{PE} - |e|V_s \). During the same time, a converse effect takes place on the field above the charged surface. When secondary and reflected electrons appear on the surface, this field acts as an accelerating field on these charges and consequently emits electrons that gain an additional energy equal to \( |e|V_s \). As a result, the electron spectra emitted from the surface can undergo the following transformation: At the beginning when \( t = 0 \), the emitted spectra will cover the range from 0 — \( E_{PE} \), which comprises SEs from 0 — 50 eV and reflected electrons from 0 — \( E_{PE} \). When the surface potential is built up after time \( t \), first the emitted spectra narrows to 0 — \( E_{\text{eff}} \) and once electrons are exposed to the accelerating field the range shifts to \( E_{\text{eff}} - E_{PE} \) [32]. So the end result is the emitted SEs from the surface acquire little additional kinetic energy upon leaving the insulator surface.

In the case of positive charging of the sample, resulting in a positive surface potential, the emitted electrons experience a completely opposite circumstance to the above described situation. Most of the electrons emitted from the surface are low energy SEs (less than about 10 eV). In the case of a positive surface potential, these SEs can be driven back to the surface and sometimes even neutralize the residing positive charges on the surface. Eventually the attracted electrons go on to make a
trapped negatively charged ring around the impact zone of the primary beam [25]. As a result the surface potential of the insulator reduces and consequently the number of SEs driven back to the surface also declines. The process continues until the emission reaches its equilibrium [33].

**Equilibrium and time evolution of secondary emission**

Secondary emission from insulator surfaces due to electron irradiation is a time dependent process, which ultimately leads to a steady state of emission at equilibrium phase. At the equilibrium state of emission, internal currents due to scattering and straggling of primary electrons, excitation of SE and holes and their ballistic flight as electrons and holes, respectively, their attenuation and drift as electrons and holes in self consistent fields followed by recombination or trapping, and/or Pool-Frenkel detrapping from localized traps, adds up to zero. Consequently, sometime after the electron beam is irradiated onto the sample surface (depending on beam intensity) the surface potential comes to a constant value as shown in Figure 4. As a result the total emission yield $\sigma$ reaches unity at the steady (equilibrium) state [34, 35]. Under these conditions, no more charge will be stored in the bulk and the final stationary state is reached.

Since the introduction of the DDL model to interpret the secondary emission from insulator surfaces, it has been accepted as a viable and reliable explanation for the process, and the theory has not been challenged since then. Most of the theoretical and experimental work done so far has been concentrated on normal incidence of the primary beam and grazing incidences have rarely been discussed. It is also noteworthy to mention that most theoretical work has not paid considerable attention to possible defects the primary beam can cause on the insulator sample and its effects
on the emission process.

Figure 4. Overall SE emission and surface potential of a bulk silica target as a function of irradiation time for an incident beam energy of 30 keV and a current density of $10^{-5}$ A/cm$^2$ [34].
CHAPTER III

EXPERIMENTAL PROCEDURE

In this chapter the experimental procedure, the different equipment used, and the data acquisition methodology is described. The experiment was carried out in the tandem Van de Graaff accelerator facility at Western Michigan University (the accelerator was not used). A commercially available filament was used as the source of electrons. Transmitted electrons were analyzed with an electrostatic parallel-plate analyzer placed few centimeters behind the sample. Electron events were counted using a channel electron multiplier (CEM) coupled to the analyzer. The background pressure in the scattering chamber was \( \sim 10^{-6} \) Torr. A \( \mu \)-metal shield was used to minimize the magnetic field effects inside the chamber. A schematic of the experimental setup is shown in Figure 5. A LabView® program was used to communicate with the electronics as well as to control the electronic devices to acquire data. Here the operation of the electron gun and goniometer, the parallel-plate spectrometer, as well as the data acquisition system is discussed in detail.

Electron gun and goniometer

A commercially available tungsten filament (\(~ 20-50 \) W) was used for the source of electrons of desired energy by biasing the filament. The beam which comes through a 1.1 mm exit aperture of the electron gun was collimated by either a set of two apertures of diameters 1.5 and 2.0 mm which were 10 mm apart or just by one aperture of diameter 1.5 mm. The electron beam was allowed to strike the sample and was controlled by focusing and vertical/horizontal steering of the beam.

The sample was mounted in a goniometer with two degrees of rotational freedom, namely, the tilt angle \( \psi \) rotation about a vertical axis (from \(-20^\circ\) to \(+20^\circ\)) and
azimuthal rotation $\phi$ about a horizontal axis (from 0° to 360°) with respect to the incident beam, for precise positioning. The goniometer was controlled using the LabView® software.

Figure 5. Schematic of the experimental chamber and apparatus. $\psi$ and $\theta$ are the sample tilt and observation (spectrometer) angles measured with respect to the incident electron beam direction.

Spectrometer and channel electron multiplier

Two spectrometers having similar energy resolutions but different angular resolution values were employed for the experiment. Both spectrometers were made with 45° plane mirror analyzers which can deflect incoming electrons by 90° towards the channel electron multiplier (CEM) as shown in Figure 6. The spectrometer had
the freedom to rotate about the vertical rotational axis of the goniometer with respect to the incoming beam from $\theta = -30^\circ$ to $+30^\circ$. The spectrometer also had several modes of operation depending on the desired energy resolution and the electron energy range to be investigated [36]. In the present experiment the low resolution and high energy mode of the spectrometer was used.

![Figure 6. Schematic diagram of the spectrometer and CEM. The back plate of the parallel-plate spectrometer was negatively biased to select the desired energy of the transmitted beam through the sample, while the front plate was grounded.](image)

The energy $E$ of the deflected electrons was determined by the voltage applied to the spectrometer plates. The deflected energy of the electrons is related to the voltage $V$ between the plates as [36],
where \( k \), the spectrometer constant, is determined by the geometry of the spectrometer and given by \([36]\),

\[
k = \frac{eV}{k}
\]

Here \( Z \) is the separation between the entrance and exit slits and \( l \) is the plate separation as shown in the Figure 6. The spectrometers used in the experiment had a spectrometer constant of 0.60.

The energy resolution \((R)\) of the spectrometer is given by \([36]\),

\[
R = \frac{\Delta E}{E} = \frac{2S}{Z}
\]

where \( \Delta E \) is the energy spread of the transmitted electrons and \( S \) is the width of entrance and exit slits \([36]\). According to the geometry of the spectrometers employed in the experiment, the energy resolution was found to be 0.03 (3%) for both of them.

By supplying the desired voltage \((-kE)\) to the back plate of the spectrometer (using the LabView\textsuperscript{®} program), only the preferred energy electrons are then deflected by 90° and sent towards the CEM. The first grid at the exit to the spectrometer is given the same voltage as the back plate to avoid low energy electrons from entering the channeltron and the second grid is grounded. An electron energy spectrum can be obtained by stepping the back plate voltage, allowing electrons with different energies to enter the CEM as the back plate voltage varies.

Transmitted electrons passing through the grid just in front of the CEM were then counted using a channel electron multiplier. The CEM used in the experiment was a Burle\textsuperscript{®} model 4821, which had a gain of \( \sim 1.0 \times 10^8 \) \([37]\). The cone of the CEM
was biased to +200 V at the entrance for initial acceleration of the electrons and to +2700 V at the tail respectively for optimal detection efficiency. The CEM pulses were normalized primarily with respect to the current as read on the sample (goniometer), or to the current on the collimator when the sample current was too low to read.

Data acquisition system

A block diagram of the electronics used for the experiment is shown in Figure 7. A BiRa Systems 6700-SCB Computer Automated Measurement and Control (CAMAC) power crate was used to host most of the data acquisition and control the equipment, while a Highland Technology M210 CAMAC Serial Crate Controller was used for data exchange between the PC and the instruments in the CAMAC Crate.

Even though there were three high voltage power supplies and Digital Multi-Meters (DMMs) in Figure 7, only one was employed as just one spectrometer was used at any particular time for all measurements. A Kinetic systems 3388 General Purpose Information Bus (GPIB) allows the three DMMs and the electrometer to be monitored via the same crate controller, which is used to communicate with other devices.

A LeCroy 2415 High Voltage Power Supply was utilized to provide the necessary voltages to the spectrometer plates. The stepper motor which was used to precisely position the entire spectrometer assembly (with respect to the beam direction) was controlled by a Joerger SMC-R Stepping Motor Controller. Verification of exact position of the spectrometer was done with a Joerger CS-5 Optically Isolated Input Register.
As shown in the figure, signals from the CEM were fed into a Fast Amplifier (EGG & ESN FTA 410), which amplified the signal 200 times to a voltage of ~ 2 V. This signal was then input into a constant fraction discriminator (Ortec 463) to eliminate electronic noise below ~ 0.5 V. The NIM logic output from the constant fraction discriminator was then sent to channel 0 of the Scaler (LeCroy 2551), which was read by the PC. The signal received from the CEM was normalized with respect to the current as read on the sample. The sample current was read by a Programmable Electrometer (Keithley 617), which gives a 0-2 V output signal depending on the intensity of the sample current. This voltage output was dropped across a 1 MΩ resistor to convert the voltage into a current. The current was then input to a Digital Current Integrator (Brookhaven 1000), and the digital TTL pulse (~ +5V) obtained from the current integrator was then sent to a Gate and Delay Generator (Ortec 416) to convert the TTL signal into a NIM before sending it to channel 2 of the scaler. The pulses from the CEM (scaler channel 0) were then counted and normalized with respect to the current on the sample (scaler channel 2). The corresponding electron energy spectra were then generated from the PC. Typical spectra can be seen in Figures 12 and 26 of Chapter IV.
Figure 7. Block diagram of the electronics.

RP - Roughing Pump
DP - Diffusion Pump
DMM - Digital Multi Meter

RP: Roughing Pump
DP: Diffusion Pump
DMM: Digital Multi Meter
CHAPTER IV
ENERGY DEPENDENCE

Interaction of energetic beams with solid surfaces depends on several factors as was discussed in the Chapter II. For beams with glancing angles of incidence, this type of interaction can be divided into two different categories in terms of their impact parameter values [38]: (1) charged particles which approach close enough to the target atoms can undergo Rutherford scattering and lose energy, and (2) particles that are reflected by the surface potential, diverting from the initial direction of trajectory and losing little or no energy. Which type of interaction is dominant for a given instance depends on factors such as incident angle, material topology, and most importantly incident energy of the charged particles.

In this chapter the transmission of 300, 400, 500, 600, 800 and 1000 eV electrons through single cylindrically-shaped glass capillaries of macroscopic dimensions is measured and discussed. It is noted that these speeds are about 100 times faster than those used in studies of slow HCI transmission which have been conducted so far [1-6, 8-9, 15-18, 39-40]. The measurements focus on the dependence of the intensity of the transmitted electrons on the capillary tilt angle $\psi$ and the incident electron energy. A significant transmission for electrons has been observed for all incident energies, up to sample tilts angle $\psi$ of 5° to 6°. However, the intensity of the transmitted electrons is found to exhibit different characteristics than has been observed in nanocapillary foils for slow ions [1, 2, 4, 5, 17, 18, 39] or for electrons [13, 19, 41, 42], as well as ions through single glass macrocapillaries [8, 9, 40]. The possibility of Rutherford scattering combined with charge buildup being responsible for the transmission process is investigated.
Sample preparation

The straight glass capillaries used for the experiment were made of Borosilicate glass. Borosilicate (also commercially known as PYREX 7740) is a composition of SiO$_2$ (80.6%), B$_2$O$_3$ (13.0%), Na$_2$O (4.0%), and Al$_2$O$_3$ (2.3%) compounds. Some of the material properties of Borosilicate are given in the table below.

Table 1. Properties of Borosilicate glass.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>2.23 g/cm$^3$</td>
</tr>
<tr>
<td>Coefficient of thermal expansion</td>
<td>$3.3 \times 10^{-6}$ °C for 20°C–300°C</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>1.14 W/m°C at 20°C</td>
</tr>
<tr>
<td>Band gap</td>
<td>2-3 eV</td>
</tr>
<tr>
<td>Electrical resistivity</td>
<td>$\sim 10^{16}$ Ω cm</td>
</tr>
<tr>
<td>Electrical breakdown</td>
<td>200 kV/cm at 300 °C</td>
</tr>
<tr>
<td>Melting/softening point</td>
<td>440–840 °C</td>
</tr>
</tbody>
</table>

The material is known for its low chemical reactivity, except in the presence of hydrofluoric acid, hot phosphoric acid, and hot alkalis. The low thermal expansion and high working temperatures ($\sim 240$ °C) make it a good candidate to study the electron transmission process even at higher beam energies.

The glass capillaries used for the experiment were prepared at the Institute of Nuclear Research of the Hungarian Academy of sciences (ATOMKI), Debrecen, Hungary. The capillaries were made by uniformly heating a straight glass tube about
1 cm diameter and stretching it by applying a constant force at the two ends. Commercially available equipment such as “pipette pullers” can be easily utilized for the production process. Diameter of the final product can be controlled by tuning the temperature and the force applied. The desired length of the capillary can be obtained by cutting the capillary using conventional machining, or with a focused ion beam method (typically 40-50 keV Ga\(^+\) beam) [43]. The capillary is then mounted into an aluminum holder, and the front side of the capillary tube and its holder which face the incident beam as well as the outer surface of the capillary are coated with a layer of graphite to carry away excess charges that fall onto it [44]. A picture of a typical glass capillary is shown in Figure 8.

![Figure 8. Picture of a glass capillary sample [44].](image)

Two glass capillaries with slightly different geometries were used for the energy dependence measurements. The samples are hereafter referred to as sample A and B of diameters \(d = 0.18\) and \(0.23\) mm, lengths \(l = 14.4\) and \(16.8\) mm (aspect
ratios $l/d = 80$ and 73), and half-widths due to aspect ratio $R_{\text{aspect}} = \alpha/2 = (d/2l) \times 57.12^\circ = 0.36^\circ$ and $0.39^\circ$. The geometry of the capillary is shown in Figure 9.

Figure 9. Geometry of the glass capillary.

Electron transmission through glass capillaries

Beam divergence and direct beam component

The beam which comes through the 1.1 mm exit aperture of the electron gun was collimated by a set of two apertures of diameters 1.5 and 2.0 mm, respectively. The beam collimation $I_{\text{coll}}$ can be found from the geometry of the setup as follows. Since it is the 2.0 mm collimator in Figure 10 which limits the beam, we can write,

$$\frac{2.0}{1.1} = \frac{(41 - x)}{x}$$

Then, $x = 14.5$ mm. So, the value for $\alpha$ can be found from,

$$\tan^{-1}\left(\frac{2.0/2}{31+10-14.5}\right) \approx 2.16^\circ$$

This means the collimation of the beam $I_{\text{coll}}$ is $\approx 2.16^\circ$. 
The direct region of transmission occurs when the transmitting electron beam makes no interaction with the capillary inner wall and travels on a straight line path. As shown in Figure 11, direct transmission takes place from $\psi = 0^\circ$ until a particular $\psi^*$ depending on the sample geometry and beam collimation. When the tilt angle is increased from its zero position, the direct transmission gradually decreases and finally drops to zero when overlapping between entrance and exit of the capillary no longer exists.
Figure 11. Direct transmission of the beam. Lower panel indicates decreasing overlapping of capillary entrance and exit with increasing sample tilt angle. $\psi^*$ is the angle at which the direct transmission is completely lost.

In order to find a numerical value for the direct region of transmission a convolution formula [45] was used.

$$I_{\text{direct}}^2 = I_{\text{aspect}}^2 + I_{\text{coll}}^2$$  \hspace{1cm} (5)

By substituting the known values of $I_{\text{coll}}$ and $I_{\text{aspect}}$ it was found that $I_{\text{direct}} \approx 2.2^\circ$.

This suggests that when the tilt angle $\psi$ exceeds this value, transmitting electrons will no longer travel in a straight line and will make at least one interaction with the inner wall of the capillary before exiting.

Transmitted electron spectra and angular distribution

Before collecting the angular dependence data for electron transmission, the “zero position” of the sample with respect to the incoming beam direction was found
by varying the tilt angle $\psi$, azimuthal angle $\phi$, and spectrometer angle (observation) $\theta$ in small steps until maximum transmission of electrons through the capillary was obtained. Then these three angles were redefined as the zero position. ($\psi = \theta = \phi = 0^\circ$). The spectrometer used for this part of the experiment had an energy resolution of $\sim 3\%$ and an angular resolution of $\sim 2.4^\circ$.

During the first phase of the experiment, the transmission of 300, 500, 800, and 1000 eV electrons through sample A and 300 and 500 eV through sample B were investigated. The transmitted intensities were normalized with respect to the incident beam. Angular dependence data were taken after transmission reached the steady state at every tilt angle $\psi$, about 1-2 hours after first putting the beam on the sample. The spectrometer angle $\theta$ was varied in small steps to collect transmitted spectra, keeping $\psi$ constant. The process was repeated for different tilt angles at each energy. The background pressure in the scattering chamber was $\sim 10^{-6}$ Torr during the experiment.

Significant intensities of the electrons transmitted through the capillary were observed up to $\psi \sim 6^\circ$ for 500, 800, and 1000 eV, whereas, intensities were observed only up to $3^\circ$ for 300 eV for both the samples. These angles clearly exceed the angle for electrons to travel in a straight line without touching the inner walls of the capillary, suggesting that the electrons interact with the inner wall of capillary at least once before being transmitted through the sample.

Some of the measured electron energy spectra obtained at tilt angles 0°, 0.5°, 1.0°, 1.5°, 2.0° and 3.0° for $\psi \approx \theta$ at 500 and 1000 eV, where maximum transmitted intensity was observed, are shown in Figure 12. These are the nominal energies, the value of which was set on the high voltage power supply. The actual energies were found to be a little higher than these values when the data were taken. It can be seen
that the overall transmitted intensity decreases strongly as the tilt angle increases. Notably, the spectra show evidence for increasing energy losses when the sample is tilted to larger angles suggesting that a fraction of the electrons undergo inelastic scattering with the inner surface while traversing the sample. This is most prominent when the tilt angle exceeds 2.0°-2.5°, where the direct beam dominance is lost.

The spectra in Figure 12 also indicate the existence of two different regions of transmission due to dominance of inelastically transmitted electrons at higher tilt angles compared to elastically transmitted electrons at lower angles. Some previous studies of electron guiding in PET nanocapillaries for 500-1000 eV electrons also showed the transmitted electrons lose energy when they traverse through the capillaries [13, 19, 41, 42], whereas a study of Al2O3 nanocapillaries for slightly lower energy electrons (200-350 eV) has reported no observation of appreciable energy loss [11]. Furthermore, no considerable energy loss has been observed for slow HCIs through nanocapillary foils [1, 4, 5, 17, 18, 39], or for single straight- and tapered-glass capillary studies [2, 3, 8, 40] so far.

Figure 13 shows the angular distributions of the transmitted electrons for 300, 500, 800, and 1000 eV for sample A. Sample B revealed almost similar distributions of transmission. The angular distributions of transmitted electron intensities were obtained by integrating the entire regions of transmitted spectra, i.e., the ranges 200-330, 400-550, 700-860 and 700-1080 for 300, 500, 800 and 1000 eV, respectively. All the intensities are normalized with respect to the current measured on the sample. The acquired integrated transmitted intensities are plotted as a function of spectrometer angle $\theta$ for different sample tilt angles $\psi$ for all the energies. It can be seen from the Figure 13 that electron transmission depends strongly on the foil tilt angle $\psi$ and the incident electron energy.
Figure 12. Transmitted electron energy spectra for 500 (sample B) and 1000 eV (sample A). All the spectra were taken for $\psi \approx \theta$, where maximum transmitted intensity was observed.
Observation Angle $0$ (deg.)

Angular distributions of the normalized integrated intensities as a function of observation angle $\theta$ for various tilt angles $\psi$ for 300, 500, 800 and 1000 eV for sample A. The solid lines represent symmetrical Gaussian fits to the data.

Figure 13. Angular distributions of the normalized integrated intensities as a function of observation angle $\theta$ for various tilt angles $\psi$ for 300, 500, 800 and 1000 eV for sample A. The solid lines represent symmetrical Gaussian fits to the data.
The transmitted electron intensities show a decrease with the tilt angle $\psi$ agreeing with observations for slow ions [1, 4, 5, 8, 17, 40] and previous electron studies [11, 13, 19, 41, 42]. It is evident from Figs. 12 and 13 that the intensities are found to display two distinct regions with a steep decay at lower tilt angles and slower drop at higher tilt angles. This feature will be discussed later in more detail. It should be noted that no significance should be paid to the angular widths of the symmetrical Gaussian fits of the integrated transmitted spectra, since the widths are greater than the resolution limit (~ 2.4°) of the particular spectrometer used for the data collection process. Consequently, the change of the angular widths of the transmitted electron distributions with tilt angle cannot be addressed in this study.

![Graph showing variation of centroid spectrometer angle $\theta$ with sample tilt angle $\psi$ for 300, 500, 800 and 1000 eV for sample A. Uncertainties of the linear fittings are shown in the bottom left corners for each energy.](image)

Figure 14. Variation of centroid spectrometer angle $\theta$ with sample tilt angle $\psi$ for 300, 500, 800 and 1000 eV for sample A. Uncertainties of the linear fittings are shown in the bottom left corners for each energy.
In addition to the transmitted intensities, the centroid observation angles (spectrometer angle at which the highest integrated transmitted intensity is observed) and the respective tilt angles of Figure 13 are plotted for 300, 500, 800 and 1000 eV for sample A in Figure 14 (Sample B revealed almost similar results). The results show a linear relationship for all the energies giving evidence that electron transmission through glass macro-capillaries is in agreement with what has been observed for slow HCI guiding [1, 8] and electron transmission through nanoscale capillaries [13, 19, 41, 42].

It is also noteworthy to mention that 500 eV showed the best agreement to the linear relationship and 300 eV the least with more than seven times the uncertainty in linear fitting compared to the other energies. This suggests that the electron transmission process at lower energies is somewhat different than the known HCI guiding mechanism resulting from charge patch formation [1].

**Energy loss, centroid energy and FWHM of transmitted spectra**

Unlike slow HCIs, the energy spectra of electrons transmitted through the single glass capillary exhibit significant energy losses as seen in Figure 12. In order to understand this energy loss, the centroid energies (weighted mean values of the transmitted spectra) and full-width-half-maximum (FWHM) values of collected energy spectra were analyzed. The centroid energies were found from \( \frac{\sum E_i I_i}{\sum I_i} \), where \( E_i \) and \( I_i \) are the energy and corresponding intensity at a given point of the spectra, respectively. The FWHM values were found from \( 2\sqrt{2\ln 2} \sigma \), where \( \sigma \) is the standard deviation of the Gaussian distribution. Both the parameters were calculated using Origin 8.0 software.

In order to study the variation of energy loss with capillary tilt angle \( \psi \), the
centroid energy values for all the $\theta = \psi$ spectra (angular centroids) for 300, 500, 800 and 1000 eV electrons were calculated and plotted against the different tilt angles. As shown in Figure 15, the general trend of centroid energies for all incident electron energies shows a decrease with increasing sample tilt angle.

![Graph showing centroid energy vs. sample tilt angle for 300, 500, 800, and 1000 eV electrons.](image)

Figure 15. Centroid energy vs. sample tilt angle $\psi$ at 300, 500, 800, and 1000 eV for sample A. Red lines show first order exponential decay fittings. Respective decay constants (DC) are shown at the bottom of each graph.

In Figure 15 the data were fitted with a first order exponential decay curve, $B + Aexp(-\psi/\alpha)$, where $\alpha$ is the decay constant and $A$ and $B$ are amplitude constants, to understand the progression of the energy loss. The centroid energy values below $\psi \approx 2^\circ$ are nearly constant for all energies. For $\psi > 2^\circ$ a fast decay starts,
followed by a much slower fall-off at higher tilt angles for most energies (500, 800 and 1000 eV). Since an electron beam of divergence ~ 2.2° was used for the measurements, $\psi < 2.5°$ represents the direct transmission region.

The decay constant for 300 eV was found to be as high as 2.1° whereas for 1000 eV it was less than half this value, showing a noticeable decrease with increasing energy. This result indicates higher incident electron energies suffer the larger energy losses with increasing tilt angle, with elastic like characteristics at lower energies compared to more inelastic behavior at high angles. The energy drop is intense around the indirect/direct boundary region (2.0°-2.5°), indicating that incident beam interactions with the inner surface of the capillary play an important role in energy loss process.

The FWHM values for all the $\psi \approx 0$ spectra are also plotted as a function of tilt angle at 500, 800 and 1000 eV for sample A and at 300 eV for sample B as shown in Figure 16. The variation of FWHM values with increasing tilt angle was found to be out of phase with that of the centroid energy values shown in Figure 15. As expected, an abrupt increase in the FWHM value was observed near the boundary between the direct and indirect regions for all incident beam energies. Since an increase in FWHM is attributed to a broadening of the transmitted energy spectra, the sharp increase of FWHM in the boundary region suggests an increased energy loss with increasing sample tilt angle, agreeing with the previous result on the centroid energies. Furthermore, different transmission characteristics are visible in the two regions, with sharp increases in FWHM in the indirect region compared to the direct region, a result that can be attributed to continuous energy loss with increasing tilt angle in the indirect region.
To analyze elastic and inelastic characteristics of the transmitted spectra further, the variation of the ratio of inelastically to elastically transmitted electron intensities with sample tilt angle $\psi$ was analyzed for $\theta \approx \psi$ spectra for all energies for both samples. The inelastically transmitted intensity was obtained by integrating the inelastic portions of the spectra, energies of 200-300, 400-500, 700-800 and 700-990 for 300, 500, 800 and 1000 eV, respectively, whereas energies of 300-330, 500-550, 800-860 and 990-1080 for 300, 500, 800 and 1000 eV, correspondingly, were considered as the elastically transmitted range. The obtained results for
\( \frac{I_{\text{inelastic}}}{I_{\text{elastic}}} \) are illustrated in Figure 17. As the results indicate, all ratios follow the same trend, i.e., an increase in inelasticity with increasing tilt angle. For tilt angles beyond 2.5° (indirect region) the inelastic transmission reaches a constant value of about 2:1 to the elastic contribution for all incident energies. This result also demonstrates the difference in transmission characteristics in the direct and indirect regions, with the indirect region showing more inelastic characteristics compared to the direct region.

![Graph showing variation of inelastic to elastic ratio of transmitted electrons with sample tilt angle for both samples at different electron energies.](image-url)

Figure 17. Variation of inelastic to elastic ratio of transmitted electrons with sample tilt angle for both samples at different electron energies.
In Figure 17, lower energies show a higher inelastic contribution for smaller
tilt angles, suggesting the small percentage of the beam which makes inner wall
interactions can still make a considerable contribution to the total transmitted fraction
even after losing some of its initial energy. A detailed discussion on electron
transmission through the capillary and their surface interactions follows later in this
chapter.

Coulombic repulsion has been established as the driving force behind HCI
guiding through capillaries for which beam interactions with the capillary surface are
inhibited due to charge accumulation, giving rise to elastic transmission of charged
particles. Such guiding effects are believed to exist in electron transmission as well
[11, 13, 19], but it is less effective compared to HCIs due to the inability of electrons
to make strong charge patches. Since only a small amount of charge needs to be
deposited close to the capillary entrance at smaller tilt angles to deflect succeeding
electrons, together with contributions of the direct beam, transmission at lower $\psi$ in
Figure 15 is more elastic (higher centroid energy values). But when the tilt angle is
increased, the direct beam contribution decreases and more charge is needed to
elastically deflect the incoming electrons to a larger angle. Hence, Coulomb repulsion
diminishes, giving way to more inelastic (lower centroid energy) and slowly decaying
scattering processes.

The escape probability of secondary electrons produced at a distance $x$ from
the surface decreases as $e^{-x/\lambda}$, where $\lambda$ is the mean escape depth of the material [20].
When an electron beam with higher primary energy (higher penetration depth)
interacts with an insulator surface, electrons will have longer escape depths to reach
the surface. This explains why the first order exponential fittings to the decay curves
in Figure 15 show decreasing (faster) decay constants for higher energies.
The variation of centroid energies as a function of $\theta$ for different sample tilt angles $\psi$ at 500 eV for sample A is shown in Figure 18 (only $\psi = 0.0^\circ$, 1.0°, 2.5°, 3.5°, 4.5° and 5.0° are plotted for better visualization). At $\psi = 0.0^\circ$ the centroid energy is almost equal to the bare beam energy value (energy of the beam without the sample, shown by the horizontal solid line at 519 eV).

![Figure 18](image)

Figure 18. Centroid energy vs. observation angle $\theta$ for sample A at 500 eV. Solid line at 519 eV indicates the bare beam (without the sample) energy value. Only some tilt angles measured are shown for simplicity.

The results in Figure 18 show that the majority of the transmission through the capillary at $\psi = 0.0^\circ$ is due to the direct beam with no interaction with inner wall, as the centroid energies near the centroid angular value ($\theta \approx \psi$) are almost equal to the bare beam energy (519 eV). The centroid energy appears to depend strongly on the
sample tilt angle and the transmitted electrons tend to lose a certain amount of energy when the tilt angle is increased, agreeing with the previous observation in Figure 15. The average centroid energy values reside on a plateau for almost all tilt angles and decrease only at the edges of the distribution.

Broadening of the transmitted ion beam at the exit of the capillaries in the case of HCIs has been reported and discussed earlier [1, 5]. It is believed that the defocusing (broadening) of the guided ions, giving rise to higher angular FWHM values, is a result of the electric field produced by the ions deposited at the exit of capillaries. The deposition of charges at the capillary exit gives rise to a symmetric potential $U$ as shown in the Figure 19. Due to this potential, emitting charged particles can gain a perpendicular energy $E = qU$. The emission angle of the charged particles ($\alpha$) can be obtained from $\sin \alpha = V_\perp / V_\parallel = (qU/E_p)^{1/2}$, where $V_\perp$ and $V_\parallel$ are the velocities perpendicular and parallel to the capillary axis, while $q$ and $E_p$ are the projectile charge and energy respectively [46].

It is obvious that the exit potential, $U$, of the capillary has a direct impact on the emission angle of the charged particles. Since $U$ depends on the charge deposition on the capillary, material properties, such as electrical resistivity, play a key role in determining the emission angle for a given material and in turn, FWHMs. In recently published results for 3 keV Ne\textsuperscript{7+} transmission through PET nanocapillaries (aspect ratio 50), FWHMs of 3° and 2.8° at $\nu = 0°$ have been reported in two different cases [15, 46], whereas for 4.5 keV Ar\textsuperscript{6+} transmission through a macroscopic glass capillary of aspect ratio 68 (which has about $10^4$ lower electrical resistivity compared to PET) an FWHM of 2.2° has been seen at a similar tilt angle [44]. This lower transmission width is attributed to the inability of the material to hold the charges with decreasing resistivity. Furthermore, in Stolterfoht et.al [15] increasing angular FWHM upon
charge deposition has also been reported, which also indicates that increasing exit potentials yield broader transmission profiles.

Defocusing effects have also been observed in most of the electron transmission experiments conducted using both nanocapillary foils [13, 19], as well as straight glass capillaries [47], suggesting the existence of a charge distribution at the exit. But compared to broader transmission profiles for HCIs observed in similar energy and aspect ratio cases (4.5 keV Ar$^{9+}$ through a macroscopic glass capillary of aspect ratio 68), electron transmission profiles obtained in this experiment (500 eV electrons through a glass capillary of aspect ratio 73) show almost a two times lower FWHM value (1.1°) at $\psi = 0°$. This result is presented later in the chapter. This suggests that the electron distribution at the capillary exit is weaker than that of ions under similar conditions.

The observation of lower centroid energies at the outer edges of the transmitted broadened beam in the present work (Figure 18) would then be a result of weakness of the exit charge distribution leading to interactions of the transmitted electrons with the capillary inner surface at the point of exit. Additionally, since the intensity of the electron beam seen at the capillary exit is much smaller than that at the entrance, charge up time for the exit can be longer and perhaps never reaches its equilibrium during the course of the measurement.

It is emphasized that in the behavior seen in Figure 15, the beam interactions with the inner capillary exit have been minimized (or excluded) by considering electron transmission along the centroid angular position (i.e., along the axis of the capillary) for $\theta = \psi$. So, energy losses seen in Figure 15 have to be caused by the weakness of the entrance charge patch to deflect incoming electrons to larger angles, and are not caused by the exit charge patch.
Figure 19. Exit charge patch formation inside the capillary. Transmitted beam of electrons is broadened (defocused) at the exit point due to the effect of electric field lines. $V_\perp$ and $V_{\parallel}$ are charged particle velocities perpendicular and parallel to the capillary axis, whereas $\alpha$ is the emission angle.

It is evident that the incident electron beam is subjected to energy loss due to its interaction with capillary inner wall, resulting in two regions, direct and indirect, having pronounced differences in transmission characteristics. This result points to the fact that electron transmission is a fundamentally different process than HCI transmission, as no energy loss or inelastic contributions to the transmission have been reported for HCIs thus far. These differences and the energy loss mechanism will be discussed in detail later in the chapter.

**Characteristic guiding angle and electron transmission**

Studies on slow HCI guiding through various kinds of nanocapillary foils have revealed that the fraction of transmitted ions depend on the primary energy $E_p$ of the incident beam as well as the tilt angle $\psi$ in an exponential manner. According to
previous work, the fraction of ions transmitted is given by \( f(\psi) = f(0)e^{\left(-\frac{Ep}{qUs}\sin^2\psi\right)} \),
where \( f(0) \) is the fraction transmitted at \( \psi = 0^\circ \), \( q \) is the initial charge state of the incident ions, and \( U_s \) is the average potential across the capillary diameter due to charge deposition at the entrance [46, 48, 49, 50]. So, for small tilt angles the transmitted fraction can said to be fall off as \( \exp(-\psi^2) \).

In order to find the nature of electron transmission, the logarithm of the transmitted fraction was plotted against both \( \psi \) and \( \psi^2 \) to see which agrees the best. The results for the indirect region of transmission are shown in Figure 20. In the figure the transmitted electron fractions were obtained by the dividing peak heights of the symmetric Gaussian fittings of the angular distributions of Figure 13 by the peak height at \( \psi = 0^\circ \) of the corresponding energy. According to this result, the \( \psi^2 \) fittings show almost double the error compared to \( \psi \). Fittings to other energies also showed the same result. So, the transmitted electron intensity seems to fall off as \( \exp(-\psi) \) rather than \( \exp(-\psi^2) \). This result has already been observed for electrons through a PET foil [13, 19, 42], which varies with the quadratic exponent dependence of slow HCl guiding [46, 48-50]. Although the reason for this cannot be fully explained, this results points out that electron and ion transmission are different processes [13, 19].

It is obvious from Figures 13, 15, 16 and 17 that there are two different regions for the fall off of intensities. To understand these transmission characteristics better, the natural logarithm of the maximum peak height of the Gaussian fit to the angular distribution for each capillary tilt angle at each energy from Figure 13 was plotted against the respective tilt angle for sample A as shown in Figure 20. The transmitted intensities were found to reveal two distinct regions, as expected, which have very different characteristics. For tilt angles \( \psi < 2.5^\circ \) (direct region) the transmission falls off with a steeper slope than in the second region for \( \psi > 2.5^\circ \).
The characteristic guiding angle $\psi_c$, defined as the tilt angle for which the transmitted intensity falls to $1/e$ of its value at $\psi = 0^\circ$, is a measurement of the ability of the material to guide the charged particles along the capillary [50, 51]. For the case of slow HCIs, it can be determined from $I_\psi = I_0 \exp \left( -\frac{\sin^2 \psi}{\sin^2 \psi_c} \right)$, where $I_\psi$ and $I_0$ are the transmitted intensities at a given tilt angle and at $\psi = 0^\circ$ respectively [46, 49]. As for the case of HCI transmission, since the intensity falls off as $\exp(-\psi)$ the characteristic guiding angle for electron transmission can be found from the slope of the $\ln(I_\psi)$ vs. $\psi$ curve with transmitted fraction given by $I_\psi = I_0 \exp \left( -\frac{\psi}{\psi_c} \right)$ [13, 19].

The acquired results for the guiding angles are also given in Figure 21.
Figure 21. Natural logarithmic plots of the maximum peak heights of the Gaussian fits to the angular distributions for sample A from Figure 13. Intensities for 800 and 1000 eV have been shifted by -2 and -4, respectively, along the vertical axis for ease of visualization. Values of $\psi < 2.5^\circ$ represent the direct transmission region of electrons, and values of $\psi > 2.5^\circ$ the indirect transmission region. Values of $\psi_c$ are the characteristic guiding angles. A value could not be calculated for 300 eV because of insufficient data.

In the region of direct transmission of Figure 21 where $\psi < 2.5^\circ$, the characteristic guiding angle was found to be the same for all the energies within the experimental uncertainties, and has a smaller value than in the indirect region where $\psi > 2.5^\circ$. In the indirect region, $\psi_c$ increases (slower fall-off) with increasing energy
with the likely exception of energies below 500 eV. This indicates a clear difference between slow positive ion transmission characteristics [52] and previous results for fast electron emission [13, 19], which showed a decrease in the guiding ability with increasing energy for all incident energies. The large scattering of data points in the direct region is likely due to incomplete charge-up of the inner walls. Increase in tilt angle corresponds to an increase in surface charge density as the beam is deposited in a smaller area [53]. Therefore, the charging process at smaller tilt angles is slower than for larger tilt angles.

Table 2. Comparison of characteristic guiding angles for ions and electrons through a PET foil and through a glass-capillary.

<table>
<thead>
<tr>
<th>E/q (V)</th>
<th>$\psi_e$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>HCIs</td>
</tr>
<tr>
<td>PET</td>
<td></td>
</tr>
<tr>
<td>[52, 54]</td>
<td>5°</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Elastic (PET)</td>
<td></td>
</tr>
<tr>
<td>[19]</td>
<td>2°±0.3°</td>
</tr>
<tr>
<td>Inelastic (PET)</td>
<td></td>
</tr>
<tr>
<td>[19]</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The values for $\psi_e$ for ion and electron transmission through PET nanocapillaries have shown different characteristics from each other. It has been observed that $\psi_e$ for ions are greater than that for both elastic and inelastic regions of the transmitted spectra of electrons through PET as shown in Table 2. The characteristic angle for the case of the glasscapillary is smaller than the above cases for 500 eV, while it is larger for 1000 eV. This suggests that the guiding ability through PET is smaller for electrons than ions and it is even smaller for the case of low energy electrons through glass capillaries. An interesting remark of the above
table is that for both ions and electrons through PET $\psi_c$ decreases with increasing energy, whereas it increases for the case of glass capillaries, suggesting an increase in electron guiding ability with energy, while it decreases for the other two cases.

The differences in electron and slow ion transmission can be attributed to several reasons. Electrons are less efficient at charge deposition on the surface of the capillary compared to ions. In the case of electron transmission, they can scatter off the surface at the point of impact and make the surface more positive due to secondary electron emission. This makes the charge patch weaker, whereas ions have the advantage of causing secondary electron emission that always makes the patch stronger. Therefore, the resulting charge patch due to electrons is reduced making it difficult to create a sufficient Coulombic field to repel the incoming electrons from the wall to guide them along the capillary axis. As a result, more of the incoming electrons penetrate into the surface undergoing inelastic scattering and become lost, or they are transmitted through the capillary again after losing energy, if able to escape from the bulk.

For electron transmission through PET and glass capillaries, one transmission channel in the case of the glass capillary compared to millions in PET are the likely reason for the differences in the $\psi_c$ values. Factors such as non-parallelism of PET capillary foils can result in an exaggerated $\psi_c$ value. On the other hand, differences in material properties such as electrical resistivity, which determine the ability of the capillary to retain charges on the walls, is a defining factor for the guiding ability as well.

So, according to the above calculations, when the problem is analyzed from the guiding ability perspective, it seems that there are two distinct regimes of indirect transmission in terms of the primary energy of the beam. Guiding ability in the
indirect region apparently increases below and above 500 eV with this energy being about the lowest point. To understand this scenario in the indirect region, the possibility that Rutherford scattering is responsible for the observed results was explored.

**Rutherford scattering**

If the inner surface of the capillary is insufficiently charged so that the field produced at the entrance is not strong enough to deflect the incoming beam, the electrons will interact with the sample surface and/or bulk and scatter as a result. In order to investigate if the scattering effects which take place in the indirect region of transmission obey the single collision Rutherford predictions, the experimental data were compared with theoretical predictions. For this, the yield at a given tilt angle and energy can be written in terms of the Rutherford cross section for the observed distributions, which is given by, 
\[
\frac{d\sigma}{d\Omega} = C_n \frac{Z_1 Z_2}{E_p^{2} \sin^{4}\left(\frac{\psi + \alpha}{2}\right)}
\]
Here, \(C_n\) is a constant obtained by normalizing the cross section to unity, \(Z_1\) the charge of an electron and \(Z_2\) the charge of the nucleus of the atom which it scatters from, \(E_p\) the incident electron energy, \(\psi\) the sample tilt angle, and \(\alpha\) a small offset angle introduced to remove the singularity of the cross section at \(\psi = 0^\circ\) [55]. The ratio \(Y(\psi)/Y(\psi=0)\) was calculated for the obtained data for tilt angles \(\psi > 2.5^\circ\) for all the energies, where \(Y(\psi)\) and \(Y(\psi=0)\) are the total electron yields at angles \(\psi\) and \(\psi = 0^\circ\), respectively.

Experimental data were compared with the curve
\[
\frac{Y(\psi)}{Y(0)} = \frac{\sin^{4}\left(\frac{\psi + \alpha}{2}\right)}{\sin^{4}\left(\frac{\psi}{2}\right)}
\]
and the results are plotted in Figure 22. Good agreement between the calculation and experimental data was found for 300 and 500 eV, but for the higher energies, 800 and 1000 eV, there is an evident deviation from the Rutherford calculations. This suggests the dominance of single-collision Rutherford scattering at lower incident energies and
the emergence of a different process at the higher energies.

Figure 22. \( Y(\alpha)/Y(0) \) of the obtained data vs. tilt angle for all energies. \( Y(\alpha) \) is the yield at tilt angle \( \psi \) and \( Y(0) \) is the yield at angle \( \psi = 0^\circ \). Dots indicate the experimental data points and the solid lines are the theoretical predictions of the Rutherford scattering cross section. The 300 eV data are from sample B and the other data from sample A. Results for 500 eV have been multiplied by 2 for better visualization.

According to Rutherford predictions, the yields, as well as the characteristic guiding angles, should decrease with increasing energy. Agreeing with predictions, the yield for 500 eV in the region \( \psi > 2.5^\circ \) (where Rutherford scattering is important) is less than at 300 eV. Also, the total electron yields due to secondary electron emission, i.e., inelastically back scattered and elastically reflected emission from an insulating surface, such as Borosilicate (which is 80% SiO\(_2\)), are a maximum at a primary electron energy of \( \sim 400-450 \) eV [20]. So, scattering effects at 800 and 1000
eV should result in smaller yields than at lower energies. As the primary electron energy increases, the penetration depth of the electrons increases, which would give smaller characteristic guiding angles (faster fall-off) for $\psi > 2.5^\circ$.

However, contrary to the Rutherford predictions the results show an increase in the yield and the guiding angles (slower fall-off) for $\psi > 2.5^\circ$ above 500 eV. Therefore, a second process may be taking over for the transmission at higher energies. This second process is attributed to charge deposition resulting in Coulombic repulsion.

When electrons are incident on the sample surface, they are either repelled electrostatically due to Coulombic forces from already existing charge patches, shown by the dashed lines in Figure 23, or they interact with the surface or bulk. If lower energy electrons interact with the surface, they can scatter (quasi-) elastically from atoms close to the surface and be transmitted toward the exit as shown by the dot-dashed lines in the figure. As a result, transmission is governed by a combination of charge patch deflection and Rutherford scattering for the lower energies. But, if the incident energy is higher (solid lines), electrons penetrate further into the bulk of the capillary and lose energy due to various inelastic processes such as ionization or excitation of inner-shell electrons. If the escape depths of these electrons are sufficiently small, they can be transmitted through the capillary with energy loss, and, if not, the electrons will become lost within the bulk of the material. However, for deeper penetration into the bulk, the probability of secondary electrons to escape the sample is smaller, which causes more charge buildup on the surface. So, both charge deposition and Rutherford scattering co-exist at a given energy, with both Rutherford scattering and Coulomb deflection causing transmission at lower energies and the charge deposition dominating at higher energies.
Figure 23. Collision geometry of the scattering of lower and higher energy electrons. Coulombic scattering of electrons from surface charge buildup is shown by the dashed lines. Lower energy electrons (dot-dashed lines) scatter (quasi-) elastically from atoms close to the capillary surface and can be transmitted towards the exit. Higher energy electrons (solid lines) penetrate deeper into the bulk of the capillary and lose energy due to various inelastic processes, causing loss of the electron within the sample or transmission through the capillary if the escape depth is low enough.

Secondary electron emission

In order to understand secondary emission from the glass capillary sample, studies were conducted at 300, 500, 600, 800 and 1000 eV for both sample A and B using spectra at $\theta \approx \psi$. Both low and high resolution spectrometers were used for this study. The data were acquired by obtaining a full spectrum of the transmission for a few selected sample tilt angles. Spectra obtained at 500 eV for sample B are shown in
Figure 24. Transmitted electron energy spectra for 500 (sample B) using the low resolution spectrometer. All the spectra were taken for $\psi \approx \theta$.

The transmitted electron intensity within the secondary emission region was found by integrating the spectral region below 50 eV (Figure 25 a,b), and the fraction of secondary electrons transmitted was obtained by dividing the intensity by the total
transmitted intensity (Figure 25 c,d). The normalized SE yields decrease with increasing sample tilt angle, while SE fractions increase. It is also evident from the results that the SE yields as well as fractions tend to decrease with increasing energy in general.

At lower primary beam energies, though the internal secondary electrons produced can escape efficiently due to low escape depths, relatively few secondary electrons are generated. As the primary energy increases, the secondary electron emission rises [20]. But at the highest energy of 1000 eV, even though the secondary electron generation is bigger, the nature of the escape process causes a rapid decrease in the number of internal secondary electrons that escape from the sample compared to the increase in generation of internal secondary electrons [22]. This effect can be seen in the results shown in Figure 25, where the fraction of SE produced, as well as SE yields, decreases with increasing energy for both the low and high resolution data. The increase in SE fraction with sample tilt angle is attributed to the increasing interaction of primary electrons with the inner sample surface for these angles.

It is also evident from Figure 25 that for deeper penetration into the bulk (higher energies), the probability of secondary electrons to escape the sample is smaller. This causes the sample to charge more negatively and further inhibit close collisions with the capillary wall. So, the majority of electron transmission through the capillary at higher energies is driven by Coulombic reflection of the primary beam. However, due to the inability of electrons to make strong charge patches, surface interactions of electrons are possible at any given energy [47].
Figure 25. Transmitted secondary electron fraction vs. tilt angle for 300, 500, 600, 800 and 1000 eV incident electrons. Sample A was used for 300 and 1000 eV with the low resolution spectrometer, and sample B was used for the 500 eV case (Figures (a) and (c)). For all of the high resolution spectrometer data (Figures (b) and (d)), sample A was used. The inset to panel (b) shows a magnified image of the normalized yield variations at higher tilt angles.

FWHM analysis using high resolution spectrometer

Transmission of 300, 400, 500, 600, 800, and 1000 eV electrons through sample A was investigated again during the second phase of the experiment to study the angular FWHM with sample tilt angle for different energies using a high resolution spectrometer. The data collection process was similar to what has been described earlier in this chapter and all the angular dependence data were taken after transmission reached the steady state at every tilt angle $\psi$, i.e., more than 2 hours after
shining the beam on to the sample.

The new parallel-plate spectrometer had ~ 10 times better angular resolution compared to the previous low resolution one where it was ~ 2.4°. The energy resolution of the new spectrometer was as same as the previous one and was about 3%. A comparison of angular resolutions of the two spectrometers is given in Figure 26.

Figure 26. Comparison of angular profiles for the low and high resolution spectrometers. Each data point is the integrated intensity of the obtained spectrum at the given spectrometer angle at 500 eV. Data are fit with symmetric Gaussians indicated by the solid lines, giving values for the FWHM of 2.4° and 0.3°, respectively, for the low (black points) and high (blue points) resolution spectrometers.
The experiment was conducted using an electron beam which had a divergence of ~ 0.6°. The direct region of the transmitted electrons was found to be \( \psi < 1° \). FWHMs of transmitted intensities were obtained by fitting symmetric Gaussians functions to the angular distributions of transmitted data, similar to that of Figure 13. Since the angular widths were well beyond the instrumental errors, a FWHM analysis of the angular distributions was possible, unlike with the previous spectrometer.

The transmitted spectra at \( \theta \approx \psi \) (where the maximum intensity of electron transmission is seen) for 500 eV electrons at tilt angles \( \psi = 3°, 4°, 5°, \) and \( 6° \) are shown in Figure 27. Expectedly, the intensity decreased with increasing tilt angle and the transmitted spectra show increased energy losses with tilt angle, similar to that reported for electron transmission through nanocapillary foils [13] and previously measured single straight capillaries [47]. Unlike the previous results, however, the new spectra showed more elastic characteristics in the transmission up to about \( \psi = 4° \) (it was seen only up to \( 1.5° - 2° \) with the previous spectrometer) and more inelasticity for larger angles.

As mentioned previously, the electron transmission through insulating capillaries is basically governed by two processes: (1) Coulombic reflection by already deposited charges, and (2) inelastic/elastic scattering from inner surfaces or the bulk. Even though Coulombic repulsion is the driving force behind HCI guiding [1], for which beam interactions with the capillary surface are inhibited, giving rise to elastic transmission of charged particles, such guiding for electrons is less effective compared to HCIs due to the inability to make strong charge patches [47]. Since only a small amount of charge needs to be deposited close to the capillary entrance at smaller tilt angles to deflect succeeding electrons, transmission at lower \( \psi \) is more elastic, as seen in Figures 27 (a,b). But when the tilt angle becomes larger, more
charge is needed to deflect to a larger angle. Hence, Coulomb repulsion diminishes, giving way to more inelastic and slow decaying scattering processes, as seen in Figures 27 (c,d).

Figure 27. Spectra of transmitted electrons at 500 eV for $\theta = \psi$ using the higher resolution spectrometer. The energy losses suggest electrons undergo inelastic as well as elastic collisions with the capillary inner wall.

Electron interactions with the inner surface are inevitable due to poor electron accumulation in and around the area of impact of the primary beam. When a solid is irradiated, the electrons emitted from the surface can be either true secondary
electrons (SE), which have energies ranging from 0 to 50 eV, or elastically and inelastically backscattered electrons (BE), which have energies that go downwards from that of the primary beam [20]. Since the BEs can reach surface areas far from the point of impact of the primary electron beam [28], the scattered electrons can be emitted even at higher sample tilt angles.

Due to charge deposition on the inner surface, potentials can be created in the entrance and exit region. The nature of potential distribution in the two regions can be different from one another due to the symmetry/asymmetry of the charge distribution. Unlike in the entrance region, the charge distribution and the field in the exit region are more radially symmetric with respect to the capillary axis [56]. Therefore, the deflection of the electrons is also radially symmetric giving rise to the Gaussian like profiles which were seen in Figure 13.

The 10 times better angular resolution of the spectrometer used in the present work allowed more detailed investigation of the angular FWHM variations with sample tilt angle. As stated, the angular FWHMs were obtained by fitting symmetric Gaussians to the angular distributions at different tilt angles and energies. The results are shown in Figure 28. As seen, the width rapidly increases to a maximum at $\psi = 1^\circ$ for all the energies, followed by a fast falloff up to $\sim 3.5^\circ$, before beginning a much slower decay. This gives evidence for the existence of two regions, Indirect–1 and Indirect–2, compared to just one region seen previously in Figure 21. The narrowness in angular spread of transmitted intensity at $\psi = 0^\circ$ for all energies is likely due to the absence of “guided electrons” within the transmitted intensity since the majority of the intensity contribution is from the direct beam component. On the other hand, the higher divergence of the FWHM at the beginning of the Indirect–1 region can be attributed to the onset of guiding and scattering events. Also, the FWHM in this
region, where elastic Coulombic repulsion is more dominant, has a much faster decay in width compared to the more inelastic Indirect–2 region. This feature was not so prominent with the previous low resolution spectrometer data (2.4° angular resolution) because of the inability to distinguish the variation of spectra going from Indirect–1 region towards Indirect–2, which spans only about 2° of the tilt angle range. As a result both Indirect–1 and 2 regions appeared as just as one. These features for FWHM variations for electron transmission through nano- and micro-scale capillaries are different from those obtained with slow HCI [54], where narrower transmission profiles were observed for small tilt angles.

Figure 28. Variation of the angular FWHM with sample tilt angle for different energies, showing two distinct regions within the indirect region.
Average angular FWHM values for $\nu > 3.5^\circ$ seem to approach a common equilibrium value for all the angles as shown in Table 3. Although the exact reason for this is not clear, the decrease in transmitted intensity with tilt angle and lack of charging at the capillary exit may play a role in it.

With the help of the higher resolution spectrometer, the existence of two regions within the indirect transmission region for electrons is evident. Electron transmission in the Indirect–1 region is largely governed by Coulomb repulsion due to charge deposition, whereas in the Indirect–2 region more inelastic transmission occurs due to scattering events from the sample surface and bulk. In the forthcoming chapter on time dependence of electron transmission, distinctive variations of the transmitted intensity with respect to time (charge) will be discussed in detail, as well as the variation of the elastic/inelastic nature of transmitted electrons with respect to transmission dynamics. This should give a better understanding of the electron transmission process.

Table 3. Comparison of FWHMs in the Indirect–1 and Indirect–2 regions at different energies.

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>Avg. FWHM of Region 1</th>
<th>Avg. FWHM of Region 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>0.78°±0.33°</td>
<td>–</td>
</tr>
<tr>
<td>500</td>
<td>1.43°±0.34°</td>
<td>0.90°±0.13°</td>
</tr>
<tr>
<td>600</td>
<td>1.44°±0.35°</td>
<td>0.83°±0.09°</td>
</tr>
<tr>
<td>1000</td>
<td>1.37°±0.38°</td>
<td>0.82°±0.09°</td>
</tr>
</tbody>
</table>
CHAPTER V

TIME (CHARGE) EVOLUTION OF ELECTRON TRANSMISSION

Investigations of the transmission phenomenon and its variation with time (or charge) of the transmitted beam position have proved to be significant in learning about the dynamics of the guiding process [1, 4, 6, 11, 12, 14, 53, 57-60]. Such investigations are essential for understanding the transmission mechanism due to Coulombic deflection, especially since this process, which is one of the two governing processes of the electron transmission through capillaries, is in fact a time dependent phenomenon.

Time evolution studies for slow HCIs using nanocapillary foils have revealed that a small finite number of secondary charge patches are sequentially formed inside the capillaries before the transmitted pattern reaches equilibrium [4, 6, 57]. Recently, Kanai et al. [6] found that the charge patch formation was explained in terms of a five step process: (1) ions are deposited on the inner wall of the capillary entrance region, which charges it up positively forming the primary charge patch, (2) subsequent ions are deflected by the primary charge patch towards the capillary exit as shown by the blue dashed lines in Figure 29 in the upper panel indicated by A, (3) with the growth of the primary charge patch, the number of deflected ions as well as deflection angle increases as indicated by the solid blue line in the upper panel of Figure 29 indicated as B, (4) with the increase of the deflection angle the ions collide with the inner wall of the capillary and create an additional charge patch (secondary charge patch) as shown in the lower panel of Figure 29, and (5) with further growth of the primary and
secondary charge patches upon charge deposition, the ions are shifted to a different
deflection angle as indicated by the movement of the blue solid line to the green solid
line (from B to C) of Figure 29 lower panel. This movement of the ion beam gives
rise to an oscillatory behavior of the transmitted ion beam which has been found to be
a slow gradual increasing process. For the case of PET it has been reported that the
charge up time before reaching the transmission equilibrium is about 10 minutes [1].

![Diagram of primary and secondary charge patches](image)

Figure 29. Change in ion beam direction due to charge patch formation inside a
nanocapillary [6].

Unlike slow ion guiding, transmission of faster electrons through nano- and
macrocapillary foils has been reported to initiate almost immediately [11, 12]. In
previously reported 200-350 eV electron transmission through an Al₂O₃ nanocapillary
foil [11], an exponential decay for $\psi = 0^\circ$ was observed initially, with the intensity
decreasing by about a factor of five before reaching stable transmission after about 5 minutes of beam time using a very intense 20-80 nA/mm² electron beam. Furthermore, Wang et al. [12], in which almost similar beam conditions to Ref. [11] have been utilized, found electron transmission for 1100-1500 eV to be almost immediate and constant over the course of the experiment for the case of a bent (by 15°) SiO₂ tube.

However, compared to the relatively large amount of research done on time evolution of HCIs [1, 4, 6, 57], so far little has been done to investigate the time dependence of electron transmission, experimentally [11, 12, 58] or theoretically [14]. Consequently, many questions regarding the transmission process remain unsolved.

In this chapter, the time evolution for transmission of 500 and 800 eV electrons through the glass capillary at different tilt and observation angles is discussed in detail. The centroid energies (weighted mean values of the energy spectra) and the corresponding full-width-half-maximum (FWHM) energy values associated with the transmitted intensities at different capillary tilt angles $\psi$ and different observation angles $\theta$ have been calculated to explain the charging characteristics of the capillary. Conditions to reach a stable equilibrium are also discussed and transmission characteristics are found to be very different from what has been reported so far for both nano- and macrocapillaries. This is especially due to features such as sharp oscillations in the transmitted intensity, indicating a mechanism of sudden discharge followed by slower recovery.

**Experimental procedure**

The same two glass capillaries which were used in the energy dependence measurements described in the last chapter, with diameters $d = 0.18$ and $l = 14.4$
(sample A) and $d = 0.23$ and $l = 16.8$ mm (sample B), were used for the measurements presented here. The beam was collimated by an aperture of diameter 1.5 mm and allowed to strike the samples. Transmitted electrons were analyzed by the same electrostatic parallel-plate analyzer coupled to the channel electron multiplier, which was located a few centimeters behind the sample. Both high and low angular resolution spectrometers were used at different stages of the experiment. The background pressure in the scattering chamber was kept under $\sim 10^{-6}$ Torr.

Before the collection mode of time evolution measurements, angular dependent data were taken by varying the analyzer angle $\theta$ in small steps to collect the transmitted spectra, while keeping the tilt angle $\psi$ constant. After determining the angular behavior of the transmission, the analyzer (spectrometer) was moved to the desired position $\theta$ and fixed. The beam was then blocked and the sample was allowed to discharge for more than 12 hours before starting the collection mode of the time evolution data. The same procedure was repeated for all tilt angles at both the energies.

The transmission dynamics were studied in two different manners. First, electron transmission was examined by placing the spectrometer at the centroid observation angle ($\theta \approx \psi$) where the transmitted intensity was maximum for a particular $\psi$, in order to understand the transmission dynamics along the geometrical capillary axis. Transmission variations beyond the capillary axial position were also studied by making measurements while keeping the spectrometer away from the centroid angular position of the transmission. Both sets of results will be discussed.

**Transmission dynamics along the capillary axis**

The dependence of transmitted electron beam intensity on integrated charge
for 800 eV at $\psi = 2^\circ$ for sample B and for 500 eV at tilt angles $\psi = 0^\circ$, $1^\circ$, $2^\circ$ and $3^\circ$ for sample with A are discussed in this section. All the measurements were made by keeping the spectrometer at the centroid angular position to study the transmission dynamics along the capillary axis.

The results obtained for the cases of 500 and 800 eV are presented in Figures 30a and b, respectively. Each data point represents the integrated energy spectrum over a period of 40 s for 500 eV (channels 480-530) and 110 s for 800 eV (channels 650-850). The integrated intensities for all tilt angles have been plotted with respect to incident charge per capillary. The $\psi = 2^\circ$ data for 800 eV were taken using a slightly de-focused beam, which had about 6 times lower flux (~ 1.5 pA/mm$^2$) compared to 500 eV. Geometrical calculations of the setup, including beam divergence, indicate that electrons interact with the inner wall of the capillary at least once before being transmitted for $\psi > 1^\circ$, whereas for $\psi < 1^\circ$ electrons can travel in a straight line without touching the inner wall of the capillary. The intensity at $\psi = 1^\circ$ for 500 eV is likely to have contributions from electrons that interact with the wall and those that do not interact with the wall.

For 500 eV at $\psi = 0^\circ$ electron transmission was detected almost immediately after the beam was put on the sample and remained nearly constant over the course of the measurement as shown in Figure 30a. This result is somewhat different from previously reported electron transmission through an Al$_2$O$_3$ nanocapillary foil [11], where an exponential decay was observed initially, with the intensity decreasing by about a factor of five. This is likely due to the beam flux in the work of Milosavljević et al. [11] being three orders of magnitude greater than that used in the present study.

The centroid energies and corresponding full-width-half-maximum (FWHM) values of the obtained energy spectra were calculated for all tilt angles $\psi$ shown in
Figure 31 for 800 eV and Figure 32 for 500 eV. The centroids were found from
\[ \frac{\sum E_i I_i}{\sum I_i} \]
where \( E_i \) and \( I_i \) are the energy and corresponding intensity at a given point of the spectra, respectively, and the FWHM values were obtained from single Gaussian fits to the energy spectra as shown in Figure 33. In both cases Origin 8.0 software was used to do the calculations. The figures also show the values for the bare beam, i.e., without the sample, by the solid lines.

Figure 30. Intensity variation of transmitted spectra with time for 500 and 800 eV. Transmitted intensities are normalized with respect to current on the sample at \( \psi = 1^\circ \) for 500 eV. Intensities for \( \psi = 0^\circ, 2^\circ \) and \( 3^\circ \) in part (a) have been multiplied by 4, 3 and 4, respectively, for better visualization. All data were taken at an observation angle about equal to tilt angle \( (\theta \approx \psi) \), where maximum transmitted intensity was seen.
Discharging data for 800 eV shown in Figure 31a were taken immediately after the charging measurements as a continuation. It is seen that discharging occurs quite rapidly (in about an hour) and that the transmission goes nearly back to the value it had at the beginning of the measurements.

The transmission at $\psi = 0^\circ$ for 500 eV is mainly due to the direct beam, which is due to those electrons that go through the capillary in a straight line. The process is much slower in the indirect region ($\psi = 2^\circ$ and $3^\circ$), where it took $1-2 \times 10^9$ e/cap (electrons per capillary) for substantial electron transmission to begin for both 800 eV (Figure 30b) and for 500 eV (Figure 30a).

The centroid energies and FWHM values for 500 eV at $\psi = 0^\circ$ remain almost constant with integrated charge as shown in Figures 32e and 32i and never reach the bare beam values. According to geometrical calculations, including the beam divergence and finite opening of the sample, about 20% of the incoming beam hits the inner surface at $\psi = 0^\circ$, which can give rise to inner wall charge up of the capillary. The charging rate depends directly on the flux density of the incoming beam [53]. For $\psi = 0^\circ$ the beam flux was held at a lower value and also run only to $1.5 \times 10^9$ e/cap (~ 60 minutes), as seen in Figs. 30a and 32a. At $\psi = 1^\circ$, where some component of the direct beam still passes through the capillary without striking the wall, Figures 32f and 32j show the centroid and FWHM energy values have a similar trend as $\psi = 0^\circ$, falling below and above the bare beam values, respectively.
Figure 31. Variation in transmitted beam intensity (a), centroid energy (b), and FWHM values (c) for 800 eV electrons at $\psi = 2^\circ$ as a function of integrated charge. The blue solid line shows the bare beam (i.e., no sample) centroid at 815.6 eV, and the red solid line the bare beam FWHM at 24.0 eV. The dotted lines indicate sudden discharging of the capillary. Numbers i-iv and letters w-z in (a) are related to Figure 33. Discharging data are also shown for a span of about 150 minutes and are fitted with a first order exponential decay curve. The decay constant ($\tau$) is given at the bottom of the discharging panel.
Figure 32. Variation of intensity, centroid energies, and FWHMs with integrated charge for 500 eV at different tilt angles $\psi$. The centroids have been calculated for all points of each spectrum. For the FWHMs an average of 5 spectra has been considered for better statistics for $\psi = 0^\circ$, $1^\circ$ and $3^\circ$, whereas for $\psi = 2^\circ$ the FWHM is plotted for every point. Blue and red solid lines show the centroid (510.4 eV) and FWHM (18.0 eV) values for the bare beam. Letters $w$-$z$ in panel (c) show the sudden discharging of the transmitted intensity at $\psi = 2^\circ$. 
Figure 33. Gaussian fits to the energy spectra at $\psi = 2^\circ$ for 500 eV at different times (incident charges). Incident charge values are mentioned in the upper left corner. The corresponding locations of the spectra for (c) and (d) in Figure 31(c) occur just before and after $x$.

In the indirect region (at least one capillary wall interaction) for $\psi > 1^\circ$, the centroid energies increase while the widths decrease with integrated charge for the tilt angle $2^\circ$ at 800 eV (Figures 31b,c) and for tilt angles $2^\circ$ and $3^\circ$ for 500 eV (Figures 32g,h,k,l), finally leveling off at about the bare beam values. The centroid energies and FWHMs for 500 and 800 eV at $2^\circ$ took nearly $7.0-7.5 \times 10^9$ e/cap to reach equilibrium. The FWHMs for 800 eV are larger by the 3% constant energy resolution of the spectrometer used. The centroids and widths for 500 eV at $\psi = 3^\circ$ reach the respective bare beam values for three times less integrated charge than those for $\psi = 2^\circ$. 
A progression of the transmitted spectra for 800 eV at $\psi = 2^\circ$ shown in Figure 34 indicates an overall increase in intensity as it approaches equilibrium. Some of the spectra for 500 eV are shown in Figure 33. The increasing dominance of (quasi-) elastically transmitted electrons over time compared to inelastically transmitted electrons is evident from the sequence of the spectra.

The process for electron transmission through an insulating macrocapillary is somewhat different from ions, which are more effective at charge deposition on the surface compared to electrons [58]. For electrons in the indirect region of transmission, it is possible for them to be repelled electrostatically due to Coulombic forces from existing charge patches, or due to Rutherford scattering. At energies lower than ~ 500 eV the transmitted electrons were shown to agree with single collision Rutherford scattering as discussed in the previous chapter, suggesting that they can either scatter (quasi-) elastically or inelastically from atoms close to the surface and still traverse the capillary.

When the beam is incident on the inner surface initially, some electrons start to introduce charge up in a small area around the impact point of the beam, while other electrons penetrate into the bulk of the capillary and scatter elastically or inelastically. If the escape depths of these latter electrons are sufficiently small, they can still be transmitted through the capillary with some energy loss, giving rise to the transmission spectra for smaller times (integrated charges) shown in Figures 34a,b,c. The initial "pure" Rutherford contribution before any charging up can be seen in Figure 34a. These initial spectra will have lower average energies corresponding to lower centroid values and larger FWHMs near the beginning, as seen in Figures 31b,c and 32g,h,k,l. If the escape depths are large, electrons become lost within the bulk of the material, lowering the overall transmitted intensity for these Rutherford electrons.
Secondary electron (energies ≤ 50 eV) production could also play a role in the development of the charge patch, most dominantly at the beginning of charging. These electrons, ejected when a primary electron strikes the surface tending to make it positive, are the likely reason for the initial low transmitted intensity. As primary electrons continue to strike the sample, the secondary electron emission yield decreases and the surface slowly becomes negatively charged, until it reaches the point where it can guide electrons toward the capillary exit.

Figure 34. Measured electron energy spectra for different integrated charges for 800 eV at $\psi = 2^\circ$. The corresponding locations for each spectrum are indicated in Figure 30(a) by the letters $i - iv$ and $w, x$. The elastically transmitted fraction of electrons generally increases with time, compared to the inelastic contribution. Sudden oscillations in the transmitted intensity can be clearly seen from panels (e)-(f) and from (g)-(h).
Deposited negative charges on the capillary surface are spread along the projection area of the beam with time, eventually creating a stronger charge patch close to the entrance which deflects subsequent electrons towards the exit. This reduces beam interactions with the surface and gives rise to elastically transmitted electrons due to Coulombic repulsion as seen in Figures 34d-34h. As a consequence, the average centroid energy of transmitted electrons increases and the FWHM decreases, eventually leveling off near the bare beam values when transmission equilibrium is reached (Figures 31b,c and 32g,h,k,l). In addition, increase in tilt angle corresponds to an increase in surface charge density as the beam is deposited in a smaller area [53]. Therefore, the charging process at \( \psi = 3^\circ \) for 500 eV is faster than at \( \psi = 2^\circ \), which is seen in Figure 32 by the centroids and FWHM energies for the former angle reaching equilibrium for three times less integrated charge than the latter angle. A comparison of experimental values of centroid energies and FWHMs in the equilibrium state is given in Table 4.

In the previous chapter on energy dependence, the prominence of inelastically transmitted intensity with increasing tilt angle was observed. On the other hand, elasticity of the transmitted electrons was observed for 500 eV at \( \psi = 2^\circ \) and \( 3^\circ \) in Figures 32g and h and for 800 eV at \( \psi = 2^\circ \) in Figure 31b. This is consistent with previous angular measurements obtained using the high angular resolution spectrometer, as seen in Figure 27, which suggests the above mentioned results are in fact within the Indirect–1 region (as shown in Figure 28) where elastic Coulombic effects dominate over scattering effects.

From the observations of transmission dynamics along the capillary axis, it is evident that the charge deposition close to the entrance with time gradually creates a field distribution on the inner surface, which elastically deflects further incoming
electrons from the charging area towards the exit of capillary. Transmission comes to "equilibrium" when compensation between charging and discharging processes are reached. It is emphasized, however, that stable equilibrium is never reached due to repeated sudden partial discharge of the charge patch from time to time. Such nonequilibrium has not been previously observed for electron transmission in capillaries. Oscillatory characteristics seen in the transmission process will be discussed in detail later in this chapter.

Table 4. Values of the average centroid energy and corresponding FWHM for the different tilt angles $\psi$ and for the bare beam at 500 eV. Selected time ranges considered for $\psi = 2^\circ$ and $3^\circ$ are within the "equilibrium" region.

<table>
<thead>
<tr>
<th>Tilt angle $\psi$</th>
<th>Range considered (e/cap)</th>
<th>Average centroid energy (eV)</th>
<th>Average FWHM (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0^\circ$</td>
<td>$&gt;4.2 \times 10^8$</td>
<td>$507.6 \pm 0.4$</td>
<td>$20.9 \pm 0.3$</td>
</tr>
<tr>
<td>$1^\circ$</td>
<td>$&gt;1.3 \times 10^9$</td>
<td>$503.6 \pm 1.0$</td>
<td>$19.7 \pm 1.5$</td>
</tr>
<tr>
<td>$2^\circ$</td>
<td>$&gt;7.5 \times 10^9$</td>
<td>$510.6 \pm 0.5$</td>
<td>$18.5 \pm 0.7$</td>
</tr>
<tr>
<td>$3^\circ$</td>
<td>$&gt;3.2 \times 10^9$</td>
<td>$511.2 \pm 0.7$</td>
<td>$17.9 \pm 0.6$</td>
</tr>
<tr>
<td>Bare beam</td>
<td></td>
<td>$510.4 \pm 0.1$</td>
<td>$18.0 \pm 0.3$</td>
</tr>
</tbody>
</table>
Transmission dynamics away from the capillary axis

Charge evolution is studied at observation angles slightly away from the centroid angular position for both 500 and 800 eV. Both cases share similar features compared to the time evolution results discussed previously. All the results were acquired by employing the low resolution spectrometer using the sample B.

The dependence of transmitted electron beam intensity on the incident charge for 500 and 800 eV at \( \psi = 2^\circ \) is presented in Figure 35. Each data point represents the integrated energy spectrum over a period of 105 s for 500 eV and 110 s for 800 eV. Both integrated intensities and centroid energies have been plotted with respect to incident electrons per capillary (e/cap). Different beam collimations were used for the two energies. Geometrical calculations show the direct transmission of electrons (no interaction with the capillary inner wall) to occur for \( \psi < 2.5^\circ \) and \( \psi < 1^\circ \) for 500 and 800 eV, respectively, due to different distances from the electron source to the collimation system that were used. The figures also show the centroid energy values for the bare beam without the sample by the solid lines.

The variation of intensity for both cases has similar characteristics with a sudden burst of intensity near the beginning followed by more stable and constant transmission. Periodic intensity drops are visible in the later region for both energies. The centroid energy for 500 eV decreases after an initial fast rise, and then levels off at \( \sim 4 \) eV below its primary beam value, whereas 800 eV shows a nearly constant value throughout the course of the measurement, which is \( \sim 1 \) eV below its bare beam value on average.
Figure 35. Variation of intensity (black points) and centroid energies (red points) with integrated charge at $\psi = 2^\circ$ for 500 and 800 eV at $\theta = 0^\circ$ and $0.5^\circ$, respectively. The centroids have been calculated for all points of each spectrum from $\frac{\sum E_i I_i}{\sum I_i}$, where $E_i$ and $I_i$ are the energy and corresponding intensity at a given point of the spectrum. Blue solid lines show the centroid (518.4 eV and 815.6 eV) values for the bare beam without the sample. Black arrows point to the sudden intensity drops that occurred periodically.

In the previous charge evolution study, which concentrated on the centroid energies and FWHMs at the centroid of angular positions (where $\theta \approx \psi$), both centroids and FWHMs were found to reach the respective primary beam values after
~7.5×10^9 e/cap at \( \psi = 2^\circ \), which is consistent with the tilt angles in Figure 35. In the present charge evolution results, both sets of measurements were taken a little bit away from the centroid angular position. Interestingly, neither of the energies seems to have reached the respective bare beam values even in the "equilibrium" state. This is likely due to the fact that the measurements were done for an observation angle that is slightly off axis.

Due to continuous charge deposition on the capillary inner wall, Coulombic repulsion of further incoming electrons gradually begins. The deflection of the electrons at initial stages will be small (and nearly along the capillary wall) due to the weakness of the charge patch. The beam deflection slowly increases with the increasing deposition of electrons and aligns with the capillary axis, which lies through the centroid of the angular position. Since the spectrometer was positioned at \( \theta = 0^\circ \) for 500 eV and at \( \theta = 0.5^\circ \) for 800 eV in Figures 35a and b, the burst of intensity between 1 – 3×10^9 e/cap and 0.5 – 1×10^9 e/cap for 500 and 800 eV, respectively, is likely caused by movement of the electron beam due to the charge accumulation process.

As seen in Figure 18, the more divergent parts of the beam, (i.e., \( \theta \) considerably smaller or bigger than \( \psi \)), have lower centroid energies compared to transmission near the centroid angular position for all tilt angles. This is attributed to a weaker exit charge distribution compared to ions leading to interactions of the transmitted electrons with the capillary inner surface at the point of exit. In addition, the reason behind 500 eV (\( \psi = 2^\circ \) and \( \theta = 0^\circ \)) lying ~ 4 eV below and 800 eV (\( \psi = 2^\circ \)
and $\theta = 0.5^\circ$) lying $\sim 1$ eV below their respective primary beam values in Figure 35, whereas results in Figures 31b and 32g showed elastic characteristics by leveling off at the primary beam value for $\theta \approx \psi = 2^\circ$ (centroid angular position), is attributed to the increasing energy losses with increasing divergence of the exiting beam. This is reinforced by the fact that $\theta = 0^\circ$ is more divergent than $\theta = 0.5^\circ$ as measured with respect to capillary axis for which $\theta = 2^\circ$.

**Intensity oscillation of the transmission**

A striking feature of the charging dynamics is the sharp oscillatory behavior for both along and away from the capillary axis, as seen at $\psi = \theta = 2^\circ$ for 500 and 800 eV (Figure 31 and 32) and at $\psi = 2^\circ \neq \theta$ for 500 and 800 eV (Figures 35a and b) respectively. This result is unseen for the direct beam. It is also possible to see some oscillations for both $\psi = 1^\circ$ and $3^\circ$ at 500 eV (Figures 32b,d), but they are not as pronounced as at $2^\circ$. Since some of the direct beam still gets through the capillary at $1^\circ$, the oscillations can be overshadowed by the more intense direct beam component. In the case of $3^\circ$, lower statistics of the transmitted intensity (about 5 times worse than $2^\circ$) likely caused the oscillations not to be as sharp.

Closer attention to recovery after breakdown for the intensity oscillations is shown in Figure 36. The prominent recovery behaviors seen from $w-x$, $x-y$ and $y-z$ for 800 eV (flux density = 1.5 pA/mm$^2$) and 500 eV (flux density = 8.7 pA/mm$^2$) in Figure 31a and Figure 32c, respectively, are plotted, along with a similar result observed for 500 eV at $\psi = 2^\circ$ and $\theta = 0^\circ$ (flux density = 3.5 pA/mm$^2$) from Figure 35. All data presented represent recoveries after the first breakdown in the respective spectra. The data are fitted with first order exponentials. Solid red lines indicate the
fitting curve \( B + A e^{-Q/O_o} \), where \( A, B, \) and \( O_o \) are fitting constants and \( Q \) is the incident charge.

The results for the three curves of each set share similar features with the initial recovery times in Figures 36a,d,g (initial charging) having a higher charge constant \((O_o)\) than the subsequent recovery times as seen in Figures 36b,c, 36e,f, 36h,i, which are smaller and have nearly equal values for all three cases. It is noteworthy to mention that charging constants were almost the same, except for the initial charging, even beyond the oscillations shown in Figure 36. The charge constants after the first recovery for 800 eV are found to have slightly higher values than those for 500 eV, whereas the high flux 500 eV results (Figures 36d,e,f) were found to yield higher \( O_o \) values compared to low flux 500 eV (Figures 36g,h,i) data.

From the bottom two panel rows of Figure 36 for 500 eV, it can be inferred that the flux density of the beam has a direct impact on capillary charging. More than two times higher beam flux in Figures 36d,e,f has increased \( O_o \) by \( \sim 1.5 \) times for the case of charge up in Figures 36e,f compared to 36h,i. Higher charging constants found for 800 eV compared to those for 500 eV indicate the ability to hold more charge at the higher energy. This is attributed to lower secondary electron emission at higher energies.
Figure 36. First order exponential fittings to the recoveries after breakdown in the intensity oscillations for 800 and 500 eV seen at w-x, x-y and y-z in Figure 31(a) and Figure 32(c), respectively. Similar results are plotted for 500 eV at \( \psi = 2^\circ \) and \( \theta = 0^\circ \) for the three recoveries after the first breakdown of the capillary in Figure 35. Red solid curves represent the fittings. Flux density for each case is given in the first panel column. Decay constants obtained from the fittings are given at the bottom of each plot.

The oscillatory characteristics for both the cases (along and away from the capillary axis) were observed after the transmission equilibrium was reached. The transmission equilibrium is a competition of both charging and discharging processes.
occurring at the same time. As charges deposit on the inner surface, they can migrate along the surface changing the distribution, as well as into the bulk. As the deposited charge density increases and equilibrium is reached between charging and discharging, the primary beam interaction with the surface minimizes as it interacts more strongly with the charge patch. The sharp oscillations seen in both the 800 and 500 eV intensity curves in Figures 31 and 32 (at $\psi = 2^\circ$) after equilibrium indicate a sudden (partial) discharge of the capillary followed by slower recovery. This indicates that when deposited charges near the capillary entrance reach a critical value, a sudden discharge occurs along the surface to the conducting coating at the entrance, or less probably to the bulk, leading to a sudden drop in the transmitted intensity. The drops in intensity are not likely due to beam deflections as their sudden nature suggests preclusion of this effect, indicating a charge breakdown instead.

Furthermore, the quasi-elastic peaks of transmitted electron spectra in Figures 34e-f and 34g-h indicate the source of transmission remains the same just before and just after an oscillation. Figures 34f and 34h indicate the remaining charge patches after the partial discharge are still capable of elastically deflecting the incoming electrons. After partial discharge, the charge accumulation slowly recovers and the process continues. Although there are reports of variations in transmitted intensities due to discharging effects for ion transmission with different geometries [59, 60], such discharging effects have so far not been reported for electron transmission through nano- or macro-capillaries.
CHAPTER VI

CONCLUSION

Transmission of electrons of energy 300 – 1000 eV through single straight glass capillaries of microscopic scale was studied in order to gain more insight into already reported electron transmission through various insulating nanocapillary foils. The two-fold experimental study has revealed new information on both energy dependence and time (charge) evolution of the electron transmission process through the capillary optics.

The angular distributions of the transmitted electrons reveal that electron transmission through capillaries is dependent on the tilt angle $\psi$ of the sample, with the transmitted intensity decreasing with increasing $\psi$, similar to what has been observed for slow HCIs. Measurements conducted using a 2.4° angular resolution spectrometer revealed two significant regions of transmission, a direct transmission region, where electrons do not interact with inner capillary wall before being transmitted through the capillary, and an indirect transmission region, where they interact at least once before being traversed.

Existence of an entrance charge patch due to electron deposition at the capillary entrance is evident from the elastic transmission of electrons at lower sample tilt angles. In this region transmission of electrons through the capillary is similar to the case of reported HCI transmission, where the ions are believed to be guided due to the field distribution resulting from charge deposition at the entrance.

The transmitted electron spectra revealed significant energy losses with increasing sample tilt in the indirect region, due to inelastic scattering with the inner surface while traversing the capillary tube, similar to observations made for electrons
through insulating PET nanocapillary foils. Furthermore, higher incident electron energies were seen to suffer the largest energy losses with increasing tilt angle, with more elastic behavior at lower angles (direct region) compared to more inelastic behavior at high angles (indirect region). The decay constants obtained from first order exponential fits showed that the higher energies are subjected to faster energy losses compared to lower energies due to larger penetration depths (and larger escape depths) of electrons at the elevated energies. Moreover, the angular profile of the transmitted beam as a function of observation angle was found to exhibit lower centroid energies at the more divergent parts of the beam compared to transmission near the centroid angular position for all tilt angles. This effect is attributed to electron interactions with the capillary walls at the exit leading to additional inelastic scattering of the electrons in this region.

Broadening of the transmitted beam when exiting the capillary is also seen from the experimental results, and this is believed to be due to formation of a symmetric potential from charge deposition at the capillary exit. Narrow transmission beam FWHMs (almost two times lower) in the electron transmission studies compared to similar work conducted using HCIs suggest a weak exit charge deposition at the exit for the case of electrons.

The transmitted electron intensity was found to fall off as $\exp(-\psi)$ similar to the case of electrons through PET foils, which is contrary to the quadratic exponent dependence of slow HCI guiding. Although the reason for this cannot be fully explained, this points out a difference between electron and ion transmission processes.

The guiding ability of electrons was analyzed by calculating the characteristic guiding angle $\psi_c$ at different electron beam energies. Guiding ability in the indirect
region was found to increase below and above 500 eV with this energy being the 
lowest point, while that of the direct region was found to be the same for all measured 
energies within the experimental uncertainties. To understand this scenario in the 
indirect region, the possibility of Rutherford scattering being responsible for the 
observed results was explored, and good agreement was found between the 
Rutherford predictions and experimental data for lower energies (300 and 500 eV), 
but an evident deviation for the higher energies (800 and 1000 eV) was observed. 

Furthermore, it was understood from the results that, when electrons are 
incident on the sample surface, (1) if the incident energy is lower, electrons can 
scatter (quasi-) elastically from atoms close to the surface and be transmitted toward 
the exit; (2) if the incident energy is higher, electrons penetrate further into the bulk 
of the capillary and lose energy due to various inelastic processes such as ionization 
or excitation of inner-shell electrons; (3) if the escape depths of these electrons are 
sufficiently small, they can be transmitted through the capillary with energy loss, and, 
(4) if not, the electrons become lost within the bulk of the material. 

At higher energies (800 and 1000 eV), due to larger penetration of electrons 
into the bulk, the probability of secondary electrons emitting from the surface is low. 
As a result, electrons can make stronger charge patches on the capillary surface and 
the Coulombic repulsion process becomes more operative at elevated energies. 

When angular dependence measurements were repeated using a 10 times 
better angular resolution spectrometer (0.25°), unlike the previous results, transmitted 
electron intensities revealed three distinct regions with increasing \( \psi \) with different 
characteristics instead of just two. Apart from the direct region, which was already 
observed in previous measurements, two distinct regions in the indirect region were 
recognized. In the Indirect-1 region (smaller tilt angles), the majority of transmission
is due to Coulombic repulsion by charge deposition at the capillary entrance. However, when the tilt angle is larger (Indirect-2), more charge is needed to deflect electrons to a larger angle. Hence, Coulomb repulsion diminishes in importance, giving way to more inelastic and slow decaying scattering processes.

The time (charge) evolution study was twofold, in which the transmission of electrons along and away from the geometrical capillary axis was studied. Integrated charge dependence of electron transmission for incident 500 and 800 eV electrons at different capillary tilt angles (within Indirect-1 region) was studied for transmission along the capillary axis. As the transmitted intensity goes to equilibrium, the centroid energies and corresponding energy values of the FWHM of the transmitted electron distributions are found to vary in phase and out of phase with the transmitted intensity, respectively. Stable equilibrium was not fully reached even for large integrated charge due to sharp oscillations in the transmitted intensity.

Low transmission at the start of each time evolution measurement is mainly attributable to inelastically scattered electrons from interactions with the inner capillary surface. Charge deposition close to the entrance with time gradually creates a field distribution on the inner surface, which elastically deflects further incoming electrons from the charging area towards the exit of capillary. Transmission comes to equilibrium, as evidenced by attaining a centroid energy close to the respective primary beam value, when compensation between charging and discharging processes are reached. A stable equilibrium is never reached due to repeated sudden partial discharge of the charge patch from time to time.

The same behavior was studied away from the geometrical axis of the capillary (at \( \psi \neq \theta \)) for incident energies of 500 and 800 eV. After equilibrium of transmission, electrons had lower average centroid energies than the respective
primary beam values. The oscillatory characteristics seen in the electron transmission intensity with time are in agreement with the experimental observations along the capillary axis. Furthermore, the results give evidence for beam movement across the capillary exit, attributed to the changing electric field distribution caused by charge accumulation close to the capillary entrance at the beginning until equilibrium is reached. The dynamics of the centroid energy values as a function of time (charge) elapsed indicate that the initial inelastic interactions of the transmitted electrons with the inner walls becomes less prominent with increasing charge deposition on the capillary surface. This gives rise to more relatively elastic transmission at the end, agreeing with previous observations for the case along the capillary axis.


44. R. J. Bereczky, private communication.


55. N. Stolterfoht, private communication.


