eV-TEM: transmission electron microscopy with few-eV electrons

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Appendices

Appendix A Transfer matrices

A sample presents an electric potential to incoming **L**ow-**E**nergy **E**lectrons (LEE). We approximate this as a localized one-dimensional potential:

$$V(x) = \begin{cases} 0 & \text{if } x < \frac{-L}{2} \\ V(x) & \text{if } \frac{-L}{2} \leq x \leq \frac{L}{2} \\ 0 & \text{if } x > \frac{L}{2} \end{cases} \tag{A.1}$$

$V(x)$ is only non-zero in a finite region. To determine the LEE reflectivity or transmissivity of the potential generated by a sample, we have to consider the one-dimensional Schrödinger equation:

$$-\frac{\hbar^2}{2m}\frac{\partial^2 \Psi}{\partial x^2}(x) + [V(x) - E]\,\Psi(x) = 0 \tag{A.2}$$

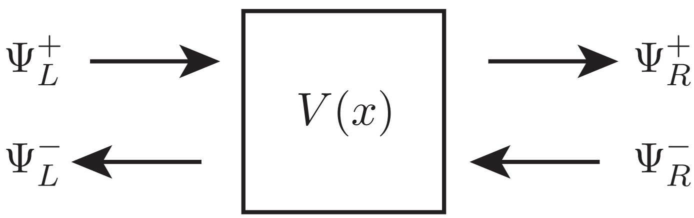


Figure A.1: Representation of the incoming and outgoing plane waves that scatter off $V(x)$.

where $\Psi(x)$ is the wave function and E is the energy of the electron. Since the potential outside the region around the origin is constant everywhere, the wave function is a superposition of plane waves:

$$\begin{aligned} \Psi_L &= \Psi_L^+ + \Psi_L^- \\ \Psi_R &= \Psi_R^+ + \Psi_R^- \end{aligned} \tag{A.3}$$

where the $+$ or $-$ superscript indicates the direction in which the plane wave moves and the R and L subscripts indicate on which side of the potential the wave functions are. This is depicted in figure A.1.

We are interested in the transmission and reflection properties of a potential. We know that an incident electron can be reflected or transmitted from the potential. Hence, when we consider an incident electron from the left (Ψ_L^+), we know that:

$$\begin{aligned} \Psi_L^- &= r\Psi_L^+ \\ \Psi_R^+ &= t\Psi_L^+ \\ \Psi_R^- &= 0 \end{aligned} \tag{A.4}$$

where r and t are the reflection and transmission amplitudes for an incoming electron from the left side. For incident electron form the right (Ψ_R^-) we know:

$$\begin{aligned} \Psi_L^+ &= 0 \\ \Psi_L^- &= t'\Psi_L^+ \\ \Psi_R^+ &= r'\Psi_L^+ \end{aligned} \tag{A.5}$$

where r' and t' are the reflection and transmission amplitudes for an incoming electron from the right side. We can thus write:

$$\begin{pmatrix} \Psi_L^- \\ \Psi_R^+ \end{pmatrix} = \mathbf{S} \begin{pmatrix} \Psi_R^- \\ \Psi_L^+ \end{pmatrix} \tag{A.6}$$

where $\mathbf{S}$ is the scattering matrix. $\mathbf{S}$ is given by:

$$\mathbf{S} = \begin{pmatrix} t' & r \\ & \\ r' & t \end{pmatrix} \tag{A.7}$$

We can also see how the electron wave functions on one side of the potential relate to those on the other side of the potential:

$$\begin{pmatrix} \Psi_R^+ \\ \Psi_R^- \end{pmatrix} = \mathbf{M} \begin{pmatrix} \Psi_L^+ \\ \Psi_L^- \end{pmatrix} \tag{A.8}$$

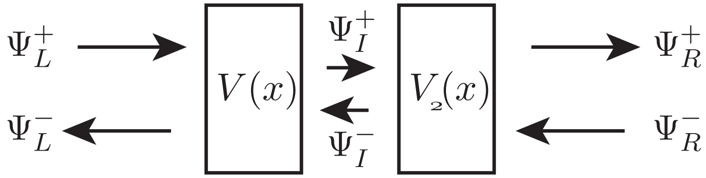


Figure A.2: Representation of the incoming and outgoing plane waves that scatter off $V(x)$ and $V_2(x)$.

Where **M** is the so-called transfer matrix:

$$\mathbf{M} = \begin{pmatrix} t' - \frac{rr'}{t} & \frac{r}{t} \\ -\frac{r'}{t} & \frac{1}{t} \end{pmatrix} \tag{A.9}$$

The reflectivity and transmissivity of the potentials are given by:

$$\begin{aligned} R &= |r|^2 \\ T &= |t|^2 \end{aligned} \tag{A.10}$$

We now know how the wave function on the left side of the potential is related to the one on the right side of the potential. This notation also allows us to use the right side wave function as the left side of another potential, $V_2(x)$, as depicted in figure A.2. We label this intermediate wave function as Ψ_I. The transmission of the combined system is given by:

$$\begin{pmatrix} \Psi_R^+ \\ \Psi_R^- \end{pmatrix} = \mathbf{M}_1\mathbf{M}_2 \begin{pmatrix} \Psi_L^+ \\ \Psi_L^- \end{pmatrix} \tag{A.11}$$

We can define a transfer matrix $\mathbf{M}_{tot}$ of the combined system:

$$\mathbf{M}_1\mathbf{M}_2 = \mathbf{M}_{tot} = \begin{pmatrix} t'_{12} - \frac{r_{12}r'_{12}}{t_{12}} & \frac{r_{12}}{t_{12}} \\ -\frac{r'_{12}}{t_{12}} & \frac{1}{t_{12}} \end{pmatrix} \tag{A.12}$$

Here r_{12} and t_{12} are the reflection and transmission probability amplitudes of the combined system. They are functions of r and t. For example when we take $r_1 = r_2 = r'_1 = r'_2$ and $t_1 = t_2 = t'_1 = t'_2$:

$$t_{12} = \frac{t_1 t_2}{1 - r_1 r_2} \tag{A.13}$$

which takes all different reflections into account. This can be seen by expanding it in a power series:

$$t_{12} = t_1 \left(1 + r_2 r_1 + r_2 r_1 r_2 r_1 + \cdots \right) t_2 \tag{A.14}$$

By multiplying these transfer matrices, the reflectivity and transmissivity of systems with any number of layers can be determined. We call these R_{tot} and T_{tot}.

$$\begin{aligned} R_{tot} &= \left|r_{12}\right|^2 \\ T_{tot} &= \left|t_{12}\right|^2 \end{aligned} \tag{A.15}$$

By taking the phase of the wave functions into account we can determine the effect of quantum interferences on the reflection and transmission. We can do this by introducing a propagation matrix:

$$\mathbf{M}_{prop} = \begin{pmatrix} e^{i\phi} & 0 \\ 0 & e^{-i\phi} \end{pmatrix} \tag{A.16}$$

where ϕ is the phase an electron traveling over a distance d gains, with d the distance between the potentials. ϕ is given by qd were q is the wave vector in between the layers, with d the distance between the layers and $q = \sqrt{\frac{2m}{\hbar^2}\left(E - \phi_w\right)}$, where ϕ_w is the work function of graphene. With this, equation A.12 becomes $\mathbf{M}_{tot} = \mathbf{M}_1 \mathbf{M}_{prop} \mathbf{M}_2$.

The total transmission of a combined system of two potentials is given by:

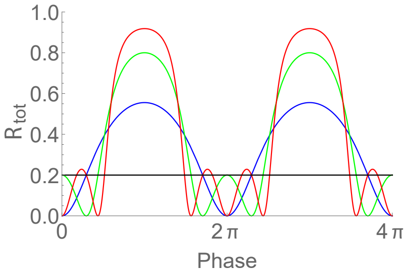
$$T_{tot} = \left| \frac{e^{i\frac{\phi}{2}} t^2}{1 - e^{i\phi} r^2} \right|^2 \tag{A.17}$$

and the reflection by:

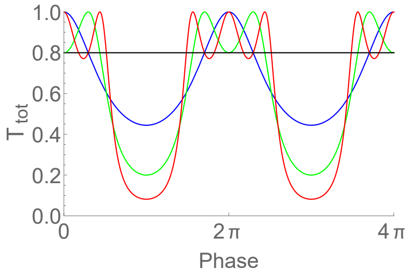
$$R_{tot} = \left| \frac{1 - r e^{i\phi}\left(r^2 + t^2\right)}{1 - r^2 e^{i\phi}} \right|^2 \tag{A.18}$$

This can also be done for three potentials:

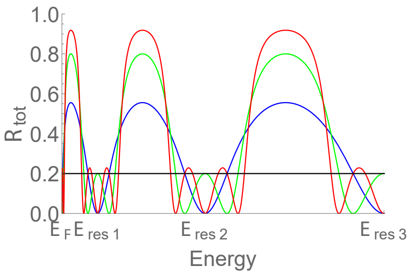
$$T_{tot3} = \left| \frac{t^3}{e^{-i\phi} - 2r^2 + e^{i\phi} r^2 \left(r^2 + t^2\right)} \right|^2 \tag{A.19}$$



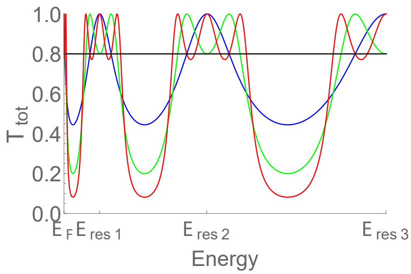
(a)



(b)



(c)



(d)

Figure A.3: Total transmission and reflection (R_{tot} and T_{tot}) of systems consisting of two, three, and four identical potentials (blue, green, and red). Each potential has reflectivity $R = 0.2$ and $T = 0.8$, indicated by the black line. (**a**), (**b**), R_{tot} and T_{tot} as a function of phase. (**c**), (**d**), as a function of energy.

$$R_{tot3} = \left| \frac{e^{i\phi} r \left(1 + e^{i2\phi}\left(r^2+t^2\right)^2 - e^{i\phi}\left(2r^2+t^2\right)\right)}{e^{-i\phi} - 2r^2 + e^{i\phi} r^2 \left(r^2+t^2\right)} \right|^2 \tag{A.20}$$

In figure A.3 the total reflectivity and transmissivity are plotted as a function of phase (in a and b) and as a function of energy (c and d). As a consequence of interference there are high transmission resonances where ϕ is a multiple of 2π. Since the phase and electron energy are related via

$$\phi = d\sqrt{\frac{2m}{\hbar^2}E} \tag{A.21}$$

the energy, $E_{res\,n}$, which the n^{th} resonance occurs is given by:

$$E_{res\,n} = \frac{\hbar^2}{2m}\left(\frac{n2\pi}{d}\right)^2 \tag{A.22}$$

Appendix B Coherence length

An electron beam with a certain energy spread ΔE has an associated spread in momentum $\hbar\Delta k$. The energy and momentum of an electron beam are related via the vacuum electron dispersion:

$$E = \frac{\hbar^2}{2m}k^2 \tag{B.1}$$

The variation in momentum associated with a variation in energy is proportional to the derivative of momentum with respect to energy. ΔE therefore gives Δk via:

$$\frac{\Delta E}{2E_0} = \frac{\Delta k}{k_0} \tag{B.2}$$

The wave function associated with an electron beam with a finite ΔE consists of a sum of many plane waves with different momenta and is thus given by:

$$\psi\left(x,t\right) = \frac{1}{\Delta k}\int_{k_0-\frac{\Delta k}{2}}^{k+\frac{\Delta k}{2}} e^{i(k_0 x+\omega t)}dk = \mathrm{sinc}\left(\frac{\Delta k}{2}x\right)e^{i(k_0 x+\omega t)} \tag{B.3}$$

This gives a coherence length of:

$$x_l = \frac{2\pi}{\Delta k} \tag{B.4}$$

The coherence length of an electron beam with an energy width ΔE, is therefore given by:

$$x_l = \frac{4\pi\hbar}{\Delta E}\sqrt{\frac{E_0}{2m}} \tag{B.5}$$

Appendix C Channel plate calibration for high dynamic range

The detector, described in section 2.1.5, consists of two **M**icro**C**hannel **P**lates (MCP), a phosphor screen and an optical camera with a 12-bit CCD. The signal we want to measure is the current, I_S, that arrives on the MCP where it is amplified. The amplification factor is determined by the MCP voltage, V_{cp}. The amplified current, I_M, is consequently converted into light by the phosphor plate, which is imaged by the light camera which records the image with a CCD. The intensity of the light emitted by the phosphor plate is proportional to I_M. The measured signal is related to the signal, I_s, via:

$$I_M = I_{BG} + I_S e^{gV_{cp}} \tag{C.1}$$

where I_{BG} is the background current that is always present and g is the MCP gain. To determine the value of g we illuminate the MCP with a constant I_S and measure I_M as a function of V_{cp}. This is plotted in figure C.2. The blue crosses are the measured data point and the red line is a fit of those data points with equation C.1. We use the fitting parameters I_B, I_S, and g. We find:

$$\begin{aligned} I_{BG} &= 36.9 \\ I_s &= 2.53 \times 10^{-9} \\ g &= 19.7\,\mathrm{kV}^{-1} \end{aligned} \tag{C.2}$$

We get I_S via:

$$I_S = (I_M - I_{BG})\, e^{-gV_{cp}} \tag{C.3}$$

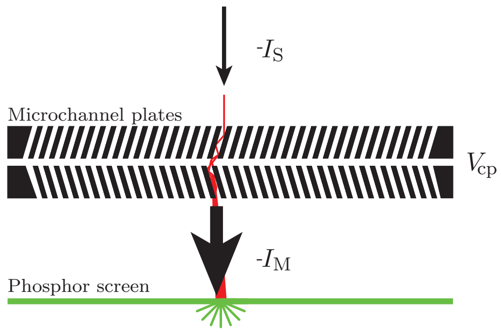


Figure C.1: Microchannel plates and phosphor screen. An incident current I_s (the minus sign is such that the arrow points in the direction the electrons move). This current is amplified to a much higher current I_M with a factor that depends on the MCP voltage V_{cp} (see equation C.1). The current I_M is converted into light with the phosphor screen. Data is recorded by a conventional light camera with a 12-bit CCD, that images the phosphor screen.

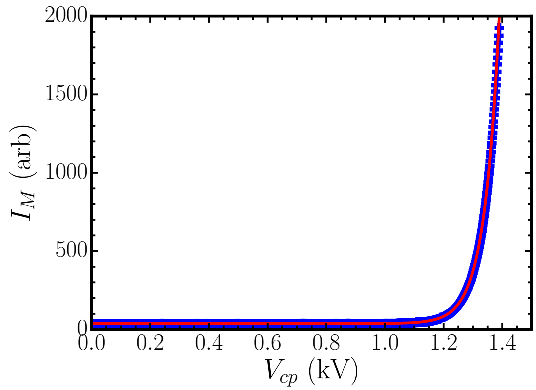


Figure C.2: Response of the MCP as a function of V_{cp}. The blue crosses are measured points and the red line is a fit of the data with equation C.1. The fit parameters are presented in equation C.2.

The conventional way to determine I_S is to just measure I_M with a fixed value for V_{cp}. However, the dynamic range can be dramatically increased when V_{cp} is also controlled. We refer to this measurement method as **H**igh **D**ynamic **R**ange (HDR) measurements. This is especially useful when determining LEE reflection and transmission spectra where I_S can have values in a range that spans several orders of magnitude. With HDR, the amplification can be high when I_s is low and low when I_S is high. The latter is very important, a large I_M can cause permanent damage to the MCP and phosphor screen.

Appendix D Data sets obtained with a different electron source

Reflected and transmitted signals are acquired with two electron sources with different energy widths. The eV-TEM and LEEM electron guns have an energy width of respectively ΔE_T and ΔE_R. The electron source we used for eV-TEM has an energy width of approximately 0.8 eV and the main electron gun used for reflection measurement about 0.25 eV. The reflected data therefore contain higher frequency components than the transmitted data. When two data sets are compared we have to make sure they share the same frequency range. To do this we first note that the measured signal can be written as:

$$\begin{aligned} T\left(E\right) &= \left(f_T * G_{\Delta E_T}\right)\left(E\right) \\ R\left(E\right) &= \left(f_R * G_{\Delta E_R}\right)\left(E\right) \end{aligned} \tag{D.1}$$

Where f_T and f_R are the transmitted and reflected signal if they would have been measured with an infinitely small energy width, $G_{\Delta E}$ is the Gaussian energy distribution with an energy width ΔE, and $(f * g)(t)$ denotes the convolution of f and g, which is defined as:

$$\left(f * g\right)\left(E\right) = \int_{-\infty}^{\infty} f\left(\tau\right) g\left(E-\tau\right) d\tau \tag{D.2}$$

To ensure that the two data sets share the same frequency range the data set with the largest range should be convoluted with a Gaussian such that:

$$R * G_{\Delta E'} = \left(f_R * G_{\Delta E_R}\right) * G_{\Delta E'} = f_R * G_{\sqrt{(\Delta E')^2+(\Delta E_R)^2}} = f_R * G_{\Delta E_T} \tag{D.3}$$

Where we use that $(f * g) * h = f * (g * h)$ and $G_{\Delta E} * G_\epsilon = G_{\sqrt{(\Delta E)^2+\epsilon^2}}$. To compare the two data sets $\Delta E'$ has to be chosen such that:

$$\Delta E' = \sqrt{(\Delta E_T)^2 - (\Delta E_R)^2} \tag{D.4}$$

Appendix E Normalization of reflected and transmitted signals

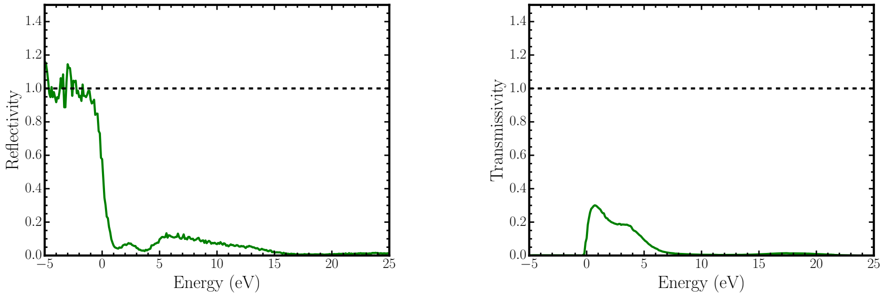
The electron reflectivity or transmissivity can be measured by determining the reflected or transmitted fraction of the incident current, I_0. The normalized signal is given by:

$$\bar{I}_{R,T} = \frac{I_{R,T}}{I_0} \tag{E.1}$$

In a reflection measurement this can easily be done by determining the reflected intensity at a negative incident electron energy. Here, the full incident current is reflected back into the imaging system before any electron reached the sample. The normalized reflected signal is therefore determined by dividing the signal by the average intensity measured at $E < 0$, as shown in figure E.1.

In a transmission experiment, I_0 can be determined by measuring the current transmitted through an open hole in the sample. However, when the electric fields on both sides of the opening are not equal such an aperture acts as an electron lens. This should be taken into account to determine I_0. In figure E.2 ray traces* are presented. Electrons are decelerated as they come closer to the middle electrode, which has a hole in it. Electrons arrive at this electrode with an energy that is indicated below the figures. Due to electric field components perpendicular to the optical axis, electrons in the vicinity of the hole are also transmitted through the hole. This causes an overestimation of I_0 when the lensing effect is not taken into account. At higher electron energies this effect is much smaller. Therefore, the electron current through the hole at higher electron energies is a good measure for the incident electron current in eV-TEM.

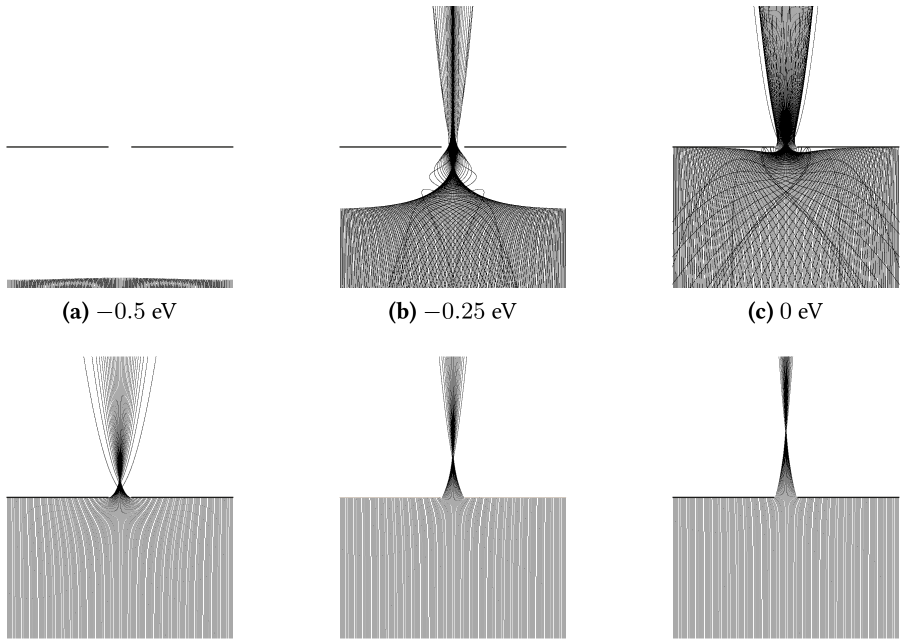
*Calculated with SIMION.



(a) Reflection **(b)** Transmission

Figure E.1: Normalization for reflection and transmission expermiments. **(a)** A typical LEEM reflectivity spectrum. This is measured on triple-layer graphene. The signal is normalized to the intensity measured in mirror mode, i.e. the intensity for $E < 0$. Here electrons are reflected back before they reach the sample. **(b)** An eV-TEM measurement on the same region. Here the incident current cannot be determined from the same region and has to be measured from an uncovered aperture in the sample, nearby the position where the spectrum is obtained.

In figure E.3 we compare an eV-TEM measurement with the electron optical simulation. The intensity is measured in eV-TEM on an uncovered aperture in the sample. Both datasets are normalized to the intensity at 10 eV. The simulations and measurements are in good agreement. This means that we can determine the incident current density in eV-TEM by measuring the transmitted current through a hole with an incident electron energy > 10 eV.



(a) -0.5 eV **(b)** -0.25 eV **(c)** 0 eV

(d) 1 eV **(e)** 5 eV **(f)** 10 eV

Figure E.2: Electron ray traces calculated with SIMION. Electrons arrive from the bottom and are decelerated as they come close to the middle electrode. The electron energy with which they enter the figure is chosen such that they arrive at the middle electrode with the energy that is indicated below the figures. The middle electrode has a hole in it (2.5 μm diameter) through which electrons can travel. 1mm above this figure an electrode (not shown) is placed at $+15$ kV, towards which the electrons are accelerated.

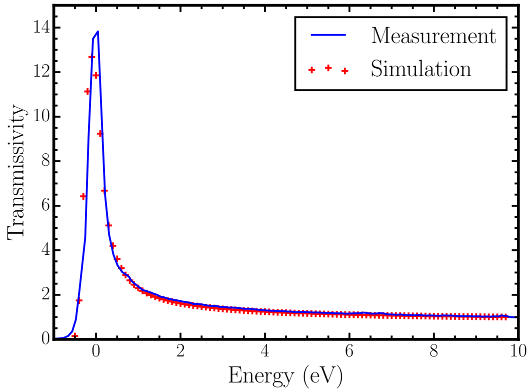


Figure E.3: eV-TEM measurement of the intensity through 2.5 μm hole, compared to the transmission determined from the SIMION ray-traces. The measurement and simulation are in good agreement. At low electron energies, the transmitted current is much higher than I_0. Electrons in the vicinity of the hole are also directed through the hole due to in-plane electric field components. From the simulations we know that at higher energies this effect becomes less important and the transmitted current through the hole approaches I_0. Here the two datasets are normalized to the intensity at 10 eV.