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Electron transmission through suspended graphene membranes measured with a low-voltage gated Si field emitter array

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Abstract. We experimentally demonstrate the transmission of electrons through different number (1, 2, and 5) of suspended graphene layers at electron energies between 20 eV and 250 eV. Electrons with initial energies lower than 40 eV are generated using silicon field emitter arrays with 1 µm pitch, and accelerated towards the graphene layers supported by a silicon nitride grid biased at voltages from –20 V to 200 V. We measured significant increase in current collected at the anode with the presence of graphene, which is attributed to the possible generation of secondary electrons by primary electrons impinging on the graphene membrane. Highest output current was recorded with monolayer graphene at approximately 90 eV, with up to 1.7 times the incident current. The transparency of graphene to low-energy electrons and its impermeability to gas molecules could enable low-voltage field emission electron sources, which often require ultra-high vacuum (UHV), to operate in a relatively poor vacuum environment.

Keywords: graphene, electron transmission, low energy, field emission

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1. Introduction

Graphene is an atomically thin layer of hexagonally arranged carbon atoms, which has exceptional electrical and mechanical properties [1, 2]. It has been demonstrated that a thin membrane made of one or multiple graphene can be impervious to larger molecules, but transparent to electrons [3, 4]. This is due to the low interaction cross-section of electrons with the graphene, which is largely empty, thereby enabling electrons with sufficient kinetic energy to pass through [5]. This is a particularly interesting property that could enable the encapsulation of devices such as field emission electron sources, to separate an ultra-high vacuum (UHV) region where field emission occurs, from a poor vacuum region where electrons are transported [6]. For mechanical strength, multiple layers of graphene are necessary in order to withstand the pressure difference between the electron emission and transport regions [7, 8]. UHV is essential for reliable field emission performance due to the barrier height sensitivity to adsorption/desorption of gas molecules, and variations in this barrier height cause exponential variations in current [9]. In addition, a non-pristine vacuum can also lead to the generation of energetic ions that erode the emitter tips, rendering the tips blunt and deteriorating electrical performance [10]. Operation in poor vacuum using a graphene-encapsulated field emitter device at voltages less than 50 V was demonstrated in our previous work, which showed promise for poor vacuum electron transport [11]. However, the current-voltage characteristics were noisy especially at lower electron kinetic energies. Assessing the electron transmission through graphene membranes to such low energy electrons is therefore essential. Several studies such as Li et al [12] and Jeon et al [13] were conducted using higher acceleration voltages (> 0.5 keV), where graphene layers have shown excellent transparency. Measuring the electron transparencies at lower energies is difficult due to the strong electrostatic field needed to extract electrons by field emission. Planar graphene devices have been demonstrated for low-energy electron field
emission measurements from graphene; however these devices do not provide an independent measure of electron transmission through graphene membranes with different number of layers [14]. In this work, we instead use atomically sharp Si emitters with self-aligned gates to extract electrons at voltages of less than 20 V as a separate electron source. Such devices have been reported previously in [10, 15], having tip radius, $r \sim 3-8$ nm, self-aligned poly-Si gate apertures of approximately 350 nm in diameter, and integrated high aspect ratio nanowires current limiters; arrays ($10^8$ emitters/cm$^2$) of these emitters have low turn-on voltage, $V_{ON}$, of 8.5 V, low operating voltage (40 V), high current density (150 A·cm$^{-2}$) and long lifetime (> 300 hours) [15]. Using this low-voltage gated Si field emission source, we characterize the low-energy electron transmission through suspended membranes with different number of graphene layers.

2. Methods

2.1. Graphene membranes fabrication and characterization

Mono-layer graphene was grown on 25 µm thick Cu foils (MTI Corp, Richmond, CA, USA), which was pre-treated with 1:6 HCl:H$_2$O. The growth was carried out at 965 °C at 10 mbar pressure in an Aixtron Black Magic Pro (Aixtron SE, Herzogenrath, Germany) cold wall chemical vapor deposition (CVD) chamber. The foil was first annealed for 15 mins at 10 mbar in H$_2$ before a flow rate of 10/60 sccm CH$_4$/H$_2$ diluted in 500 sccm of Ar was used to grow graphene for 10 mins. Initial cooling to 400 °C was carried out with 500 sccm of H$_2$, before final cooling in Ar to less than 100 °C when the foil was unloaded. PMMA A8 was spin-coated at 4000 rpm and baked at 95 °C for 5 mins, after which, the backside of the foil was etched with O$_2$ plasma in a barrel asher. Graphene transfer was then performed by etching the Cu foil in 1 M FeCl$_3$ and cleaning steps in diluted HCl (1:3) and de-ionized water. Multiple layers were prepared by transferring the floating graphene/PMMA on a subsequent
foil and re-etching. The substrates were holey (hexagonally arranged, 2 µm diameter and 4 µm pitch) 200 nm thick silicon nitride with 0.5 mm by 0.5 mm square area transmission electron microscope (TEM) grids (TedPella Inc. Redding, CA, USA). Au/Ti (50/5 nm) was deposited on both sides of the grids by electron beam evaporation prior to graphene transfer. This step was carried out to avoid the charging of the grid as observed in our previous work [16]. The PMMA/Gr stack was “fished” using the grid substrate, which was then allowed to dry at an angle in a convection oven at 60 °C for at least 4 hrs. The substrate was then baked on a hot-plate at 120 °C for 3 hrs. After the transfer was complete, the PMMA stamp was removed by gently immersing in acetone and soaked overnight. IPA (isopropyl alcohol) was then used to remove any acetone residues by soaking for 30 mins, before drying the sample by placing the substrate vertically in ambient for at least 2 hrs [7, 17].

Mono-layer (1L), two-layer (2L) and five-layer (5L) graphene were characterized by scanning electron microscopy (SEM, Model: Supra™ 40, Carl-Zeiss AG, Oberkochen, Germany) at an acceleration voltage of 2 kV. The number of holes was measured from the SEM images using Fiji [18] and by determining the area of the holes, we categorized the integrity of the graphene membranes using the percentage area torn/uncovered. Atomic force microscopy (AFM, Model: D3100, Veeco, Plainview, New York, USA) scans of the membranes were performed in tapping mode. Raman spectroscopy (InVia Reflex Confocal Microscope, Renishaw Inc., West Dundee, IL, USA) at an excitation wavelength of 532 nm was used to extract the intensity ratio of the 2D peak to the G peak. The substrates were subsequently mounted on a separate probe with vacuum-compatible conductive Ag epoxy for electrical characterization.
2.2. Device fabrication and characterization

Si FEAs (square array of 10,000 tips) with 1 µm pitch, and integrated Si nanowire current limiters, as reported by Guerrera and Akinwande [15], were used as the electron source. The fabrication process is detailed in previous reports [11, 19]. Current-voltage ($I-V$) characteristics of fabricated FEAs in UHV ($<2 \times 10^{-9}$ Torr) were first measured as a three terminal device, using three source-measurement units (SMUs), to obtain a baseline of the electrical performance. With the grid, the measurement was carried out as a four terminal device. The vacuum was maintained by an ion pump and the pressure was recorded using a Bayard-Alpert ion gauge. The four SMUs (Keithley Instruments Model 2657A) were connected to the device by means of miniature high voltage (MHV) electrical feedthroughs and tungsten micro-manipulator probes. A tungsten ball of (~0.5 mm in diameter) was used as the anode, positioned at approximately 3 mm from the surface of the chip and biased at a constant voltage of 1.1 kV for all the electrical characterization. All voltages were in reference to the gate voltage, $V_G$, which was biased at 0 V. The gate-emitter ($V_{GE}$) voltage was varied from 0 to 40 V whereas the grid voltage ($V_{Grid}$) was maintained at a fixed bias. The transfer characteristics ($I_A$ vs $V_{GE}$) were performed in the sequence where $V_{Grid}$ was 0, 10 V, 20 V, 50 V, 200 V and −20 V respectively. All the grids were connected to a probe to ensure they are at the same heights, and the grid under consideration was aligned to the array by means of a microscope. To obtain the baseline data without the grids, the probe holding the graphene grids (see Supporting Information) was first moved out of the way to measure the device with an anode as a three-terminal device so that all data was obtained in-situ without breaking the vacuum. To analyze the data, we used the Murphy-Good equation that corrected a significant error in the Fowler-Nordheim (FN) theory to extract slope with absolute value, $b_{FN}$, and intercept $\ln(a_{FN})$ by fitting the FN plot using (1) [20]:

$$\ln\left(\frac{I_A}{V_{GE}^2}\right) = \ln(a_{FN}) - b_{FN}\left(\frac{1}{V_{GE}}\right)$$
The field factor, $\beta$, was calculated from $b_{\text{FN}}$ using (2):

$$\beta = \frac{s_{\text{SN}} \cdot B \cdot \phi^{3/2}}{b_{\text{FN}}}$$

(2)

where $s_{\text{SN}}$ is the slope correction factor of about 0.95 at the fields of around 2 V·nm$^{-1}$, and $B$ is a constant in the FN formulation with a value of 6.83 eV$^{-3/2}$·V·nm$^{-1}$ [21], and $\phi$ can be approximated by the Si electron affinity, $\chi_{\text{Si}}$, of 4.05 eV. Transmission characteristics ($I_A$ vs $V_{\text{Grid}}$) were recorded by fixing $V_{\text{GE}}$, and sweeping $V_{\text{Grid}}$ from $-20$ V to 200 V in 5 V steps.

2.3. Modeling

Finite element modeling was performed using COMSOL Multiphysics® 5.0 using a 3D structure with the electrostatic and the charged particle modules. Graphene was modeled as a 2 nm layer because this was the smallest thickness that could be defined in the software. The layer was used as a pass-through wall, and the terminal was biased at $V_{\text{Grid}}$. The electron source was modeled as a conical surface (with bottom diameter of 1 µm and top diameter of 100 nm, and height of 100 nm) located at the center of the aperture but 25 µm below the grid. The angular spread of the beam was set to $0^\circ$ as the goal was to investigate the trajectories of secondary electrons generated by a narrow focus electron beam. A third terminal located at 25 µm above the grid and biased at 250 V was used as the anode. The bias voltages and distances used were smaller than in the experiment in order to reduce the number of elements, and hence computation time while providing a relatively accurate account of the electron trajectories. The value of $V_A$ was selected to ensure there will always be an electric field from the anode to the grid causing electrons to accelerate towards the anode if a maximum $V_{\text{Grid}}$ of 200 V is used. A time step of 0.1 ps was used and the trajectory maps (1 µm above grid) were obtained at a time point of 0.1 ns for different initial electron energy and different $V_{\text{Grid}}$ values. Secondary emission was modeled as an isotropic hemisphere with the generation of 2 particles for each interaction with the kinetic energy scaled for each particle generated. This
number was chosen as the secondary yield of graphene on Cu measured at energies lower than 300 eV was found to be approximately 1.5 as reported by Cao et al. \cite{22}. At lower energies, this yield tends to rise first before it decreases, and therefore a value of 2 secondary particles could be reasonably assumed \cite{23}.

3. Results

3.1. Graphene membrane characterization

Figure 1 illustrates the characterization of the graphene membranes using SEM, AFM and Raman spectroscopy. The membranes were inspected using SEM images as shown in figure 1(a), from which the percentage of partially covered membranes and completely broken membranes was determined as shown in figure 1(b). From AFM scans shown in figure 1(c) and (d), we find that the five-layer graphene membranes had a larger deformation (\(\sim 75 \text{ nm}\)) within the aperture. Raman spectra shown in figure 1(e) confirm the presence of the graphene of different number of layers. Since the spectra were taken in the graphene membrane region, no peak at 521 cm\(^{-1}\) is observed, which would have demonstrated the presence of Si peaks due to the Si\(_3\)N\(_4\) membrane or on the Si wafer. The quality of the transfer, as assessed by Raman spectroscopy, is shown in figure 1(e) confirming the reduction (from 3.4 to 0.49) of the intensity ratio of the 2\(D\) to the \(G\) peak as the number of layers increased from 1 to 5. Median and interquartile range of the \(I_{2D}/I_G\) ratios are shown in the box and whisker plot in figure 1(f) and the position of the peaks in the spectra measured is depicted in figure 1(g). In both figure 1(f) and (g), the variations for the mono-layer graphene were much larger than the two-layer and the five-layer graphene. The position of the peaks also red-shifted by \(\sim 10 \text{ cm}^{-1}\) (figure 1(g)) from 1L to 2L but blue-shifted by \(\sim 7 \text{ cm}^{-1}\) from 2L to a 5L membrane.
Figure 1. Membranes with monolayer and multilayer characterization. (a) SEM of different number of graphene layers on nitride grid; (b) Bar plot of partially torn and fully torn membranes analyzed for over 600 membranes; (c) AFM scan of membranes; (d) Profile scan of membrane with different number of layers; (e) Raman spectra of different number of graphene layers on the membranes; (f) $I_{2D}/I_G$ ratio box and whisker plot for 20 membranes; and (g) Mean and standard deviation locations of the $I_G$ and $I_{2D}$ peaks.

3.2. Field emitter array transfer characteristics

Figure 2 illustrates the characterization of the Si FEA. The device, schematically shown in figure 2(a), had a nanowire diameter of $\sim 100$ nm and gate thickness of 200 nm. Field emission current measured at the anode, $I_A$, from the 100 by 100 Si FEA with $V_{GE}$ is shown in figure 2(e). The Si FEAs turned-on at $V_{GE} \approx 15$ V and anode currents, $I_A \approx 72$ $\mu$A were measured at $V_{GE} = 40$ V as shown in figure 2(e). We extracted a $b_{FN}$ value of 320 V and intercept $\ln(a_{FN})$ of $-9$ from FN plot shown in figure 2(f). The $b_{FN}$ value corresponded to $\beta = 0.164$ nm$^{-1}$ at the emitter tip, and $r$ of approximately 3 nm, assuming the work function is the electron affinity of Si. With turn-on fields of approximately 2 V·nm$^{-1}$, this value of $\beta$ corresponded to a $V_{ON}$ of 12 V, which was reasonably close to the experimentally measured
value of 15 V.

Figure 2. Field emission source based on Si nanotip arrays with self-aligned gate apertures. (a) Schematic of the electron source; (b) Cross-sectional scanning electron microscope (SEM) image of tip with nanowire and oxide stripped for clarity; (c) Tilted view of an array of emitters with self-aligned apertures; (d) Optical micrograph of device with 10,000 tips with contact pad extension; (e) Field emission characteristics of device showing turn-on voltage at 15 V, and anode currents higher than 50 µA at 40 V; and (f) Fowler-Nordheim plot to extract slope, $b_{FN}$ and intercept, $\ln(a_{FN})$, of the anode current. (b) and (c) are reprinted, with permission from [24] ©[2020] IEEE.

3.3. Transfer characteristics with graphene

To measure the electron transmission through a graphene-mounted grid, it was positioned, with the graphene layer facing the electron source, at a height of $\sim$1 mm above the device, and $\sim$3 mm below the anode as illustrated schematically in figure 3(a). The geometrical transparency was calculated using figure 3(b), where the unit cell was a hexagon inscribed in a circle with diameter, $d_h$, of 4 µm containing a circular aperture with diameter, $d_c$, of 2 µm. The area of the hexagon is $3\sqrt{3}a^2/2$, where $a$ is $0.5d_h$ and using the area of the circle $\pi d_c^2/4$, we find that the geometrical transparency is 30.2%. Electrical characteristics at two different grid voltages, $V_{Grid}$, of 0 V and 50 V are shown in figure 3(c) and figure 3(d), respectively. All combinations —grid but without graphene (0L), 1L, 2L, and 5L —were compared to the
case without the grid (and without graphene) from figure 2(e). In these measurements, since \( V_G \) was invariably at 0 V, \( V_{\text{Grid}} \) was the acceleration voltage from the gate to the grid, and accordingly, the energy of the electrons emerging from the grid would be essentially equal to \( q(V_{\text{Grid}} + V_{\text{GE}}) \), where \( q \) is the elementary charge \((1.6 \times 10^{-19} \text{ C})\). The anode current ratio, \( \gamma \), comparing the currents with a grid structure to the case without the grid, was calculated using (3):

\[
\gamma = \frac{I_A, \text{ with graphene and grid}}{I_A, \text{ without grid and without graphene}}
\]

The inclusion of the torn graphene probability has not been accounted for in (3); however, since it is only in the range of 5-10% for fully torn-graphene, it would not affect the \( \gamma \) significantly.

Figure 3. Electrical characteristics observed with graphene layers on nitride membrane between the anode and the FEA: (a) Schematic of the measurement set-up showing the different voltages and distances; (b) Schematic of hexagonal arrangement of apertures; (c) Anode currents measured with different graphene layers on the grids with \( V_{\text{Grid}} = 0 \); and (d) Anode currents measured with different number of graphene layers with a grid bias of 50 V. (c) and (d) are reprinted, with permission from [25] ©[2020] IEEE.

Using a conductive membrane was essential, as we found in our previous work [16] that
without the presence of the conductive layer of Au/Ti, a maximum $\gamma$ of only 0.19 using a 5L graphene membrane at $V_{GE} \approx 42$ V was measured. This was lower than the case of 0L membrane (grid only), where $\gamma \sim 0.45$ at $V_{Grid} = 0$ V was achieved [16]. We concluded that by using an insulating grid, charge accumulation on the graphene was significant and the resulting repulsion decreased the effective transmission of electrons through the grid apertures. In addition, there would be poor contact between the graphene and the probe if the graphene layer was not continuous, or between the grain-domain boundaries. Hence all the membranes had the metal layers for the measurements in this work.

We compared the maximum $I_A$ and FN slope from the $I$-$V$ characteristics of different number of layers as shown in Table 1.

At $V_{Grid} = 0$ V, $I_A$ was smaller than 72 $\mu$A when using a grid compared to without it, hence $\gamma < 1$. Still, for a graphene mounted grid, $I_A$ was larger than the case with a grid without graphene ($\gamma > 0.3$). For a mono-layer graphene grid, $I_A$ was considerably larger than without the grid ($\gamma \sim 1.7$) at a $V_{Grid}$ of 50 V. With 0 V on the grid, the $b_{FN}$ was similar for the case without the grid, in the range of 300-320 V. In contrast, $b_{FN}$ increased to $>320$ V in the presence of a grid compared to without it when $V_{Grid} = 50$ V.

**Table 1.** Extracted maximum $I_A$ and FN slopes for different grid types biased at 0 V and 50 V, compared with the case without the grid.

| Grid type | $I_A$ [$\mu$A] @ $V_{Grid} = 0$ V | $V_{Grid} = 40$ V | $V_{Grid} = 50$ V | $b_{FN}$ [V] @ $V_{Grid} = 0$ V | $V_{Grid} = 50$ V |
|-----------|---------------------------------|------------------|------------------|----------------------------|------------------|
| None      | 71.6                            | —                | 325              | —                          | —                |
| 0L        | 24.6                            | 10.7             | 299              | 362                        |                  |
| 1L        | 35.4                            | 119.4            | 303              | 337                        |                  |
| 2L        | 38.1                            | 76.3             | 294              | 343                        |                  |
| 5L        | 51.1                            | 85.4             | 313              | 318                        |                  |
3.4. Transmission characteristics

The transmitted electrons through the grid contribute to $I_A$, whereas the intercepted electrons are collected by the grid, measured by the grid current, $I_{\text{Grid}}$. The $I_A - V_{\text{Grid}}$ transfer characteristics for different number of graphene layers are shown in figure 4(a). For comparison, the $I_A - V_{\text{Grid}}$ characteristics are plotted in figure 4(b). Therefore, the transmission ratio, $T$, across the grid can be estimated by (4):

$$T = \frac{I_A}{I_A + I_{\text{Grid}}}$$  \hspace{1cm} (4)

It is shown in figure 4(a) that $\gamma$, in the presence of the graphene covered grid compared to the case where there was no graphene, increases with $V_{\text{Grid}}$ in the range 0-100 V before decreasing after 100 V. Without the graphene, a $\gamma$ of $\sim 0.3$ is measured, consistent with the grid transparency. However, with graphene, $\gamma$ increased as high as 1.2 at $\sim 80$ V on the grid. It was observed (figure 4(b)) that at higher $V_{\text{Grid}}$, more electrons were intercepted and consequently, a lower transmission ratio (figure 4(c)) was achieved at the anode. As the number of layers increased, the general trend was a lower $T$ as shown in figure 4(c).

In order to understand the measurements we performed simulation on the equipotential lines at the aperture region with and without graphene as shown in figure 4(d). The presence of graphene flattens the lines, and using a negative $V_{\text{Grid}}$ caused the equipotential lines to converge. Simulation of the electrons passing through the graphene is shown in figure 4(e) incorporating secondary emission physics. Without the secondary emission, the number of electrons emerging from the grid was smaller than the number of incident electrons (1000). Electron scattering with graphene layers led to secondary electron emission and higher number of electrons emerging from the grid. At higher $V_{\text{Grid}}$, the electrons diverge from the central region (figure 4(e)). In addition, the initial electron energy emerging from the emitter, $E_{\text{init}}$, also caused a decrease in the number of secondary electrons generated when $E_{\text{init}}$ increased from 20 to 40 eV (figure 4(e)). By extracting the total number of electrons at
different values of $V_{\text{Grid}}$, we found that the retarding potential of $-20$ V (figure 4(f)) caused the highest yield. The trend for each of the energy analyzed in figure 4(f) was similar to observations from experiment, where increasing $V_{\text{Grid}}$ initially caused $I_{\text{A}}$ to rise and then to decrease with further increase in $V_{\text{Grid}}$. At negative $V_{\text{Grid}}$ values, the electrons are slowed down in the layer causing longer interactions with graphene and much higher multiplication factors (figure 4(f)). Simulations with a layer than has a depth in the cavity were also performed (see Supporting Information), which demonstrated that as the depth increases, due to the sagging of the graphene layer, there could be some focusing of the electrons generated at the interface. However, the trend in electron multiplication was similar to a flat layer except more

Figure 4. (a) Anode current, $I_{\text{A}}$ measured with grid voltage $V_{\text{Grid}}$ with $V_{\text{GE}} = 40$ V; (b) Grid current, $I_{\text{Grid}}$ measured with grid voltage, $V_{\text{Grid}}$, for all the layers; (c) Transmission ratio of the different grids at $V_{\text{GE}} = 40$ V; (d) Equipotential lines at the grid interface with and without a graphene membrane biased at two different voltage polarities (+20 V and $-20$ V); (e) Maps (square of 5 µm) of electrons collected at 1 µm above the grid biased at different grid voltages, $V_{\text{Grid}}$, with and without secondary emission across the graphene membrane demonstrating large number of scattered electrons, and reduced secondary emission with higher initial energy, $E_{\text{init}}$ and; (f) Anode current ratio collected at 1 µm above grid for different $E_{\text{init}}$ and $V_{\text{Grid}}$. Source was 1000 electrons.
pronounced due to the increased interaction between the electrons and the cavity.

4. Discussion

We have demonstrated that the multiple layers of graphene tend to sag in the aperture, perhaps due to the decreased Young’s modulus of multi-layer graphene, or the increased van der waal’s forces between the layer and the aperture wall. This could also explain the larger proportion of torn membranes in the five-layer graphene, as compared to the one and two layer graphene. The large variation in the $I_{2D}/I_G$ ratio for the monolayer graphene could be due to the nucleation sites especially on Cu foil, which is not perfectly smooth. These sites result in some locations having more than 1L during the nucleation process. When several layers are transferred, this variation averages out, thereby reducing the distribution of the $I_{2D}/I_G$ ratio for multiple layers. The inconsistent trend in the peak positions can be explained by the stacking of the graphene layers, which is random due to the multiple transfers involved, causing a change in the peak position for the five layers.

In electrical characterization, the larger $I_A$, when using graphene layer grids as compared to when there was no grid, resulted in $\gamma > 1$. This could be explained by either wrinkles or grain boundaries in graphene producing electrons by inelastic scattering. The non-ideal coverage – both broken apertures and non-flat surface – could also lead to electron scattering by stray field lines that eventually impact the trajectories of the electrons emitted from the device. Hence, transmissive secondary electrons could be generated [26]. The energy of the primary electrons determines the yield of secondary electrons. In this work, the primary electrons at $V_{GE} = 40$ V impinging the surface at $V_{Grid} < 50$ V would have energies of 90 eV. The kinetic impact of these electrons in the graphene layer could generate secondary electrons internally, and the interaction time increases with increasing number of layers. As graphene is atomically thick, the secondary electrons and the lower energy primary electron could be
transported to graphene surface facing the anode, and then escape from the solid-vacuum interface [23]. The free electrons would then be accelerated towards the anode due to the potential difference between the grid and the anode.

However, this was not observed in the case of the grid without any graphene, which demonstrated a value of $\gamma$ of less than 0.5 as expected from its geometrical transparency. Although the geometrical transparency and the ratio of 0L to None should be approximately equal, here the $\gamma$ was 0.45 whereas the calculated geometrical transparency was 0.30. This can be accounted for by the broadening of the apertures during lithography and etching processes, increasing the diameter from 2 µm to approximately 2.5 µm, which would increase the calculated geometrical transparency to 0.47. Therefore, the larger anode current ratios would be mainly due to the graphene. This is demonstrated by simulations comparing the case with and without secondary electrons, where the primary electrons are slowed down in the thicker layer, enabling secondary emission of electrons that emerge on the side facing the anode. In 1L and 2L graphene cases, the electrons generated are able to escape as there are fewer collisions as compared to the five-layer graphene. In addition, the larger deformation of the five-layer potentially generates further electrons due to collisions with Au on the grid walls. Multiple layers also allow more time for collisions and, hence increasing the probability of secondary electron generation. When the incident electrons are not perpendicular to the graphene layer, this could lead to shallower penetration and could enable multiple collisions and even larger interception as observed with the 5L graphene. At higher $V_{\text{Grid}}$, the interception of both primary and secondary electrons would increase leading to the higher $I_{\text{Grid}}$ and lower $T$ as shown in figure 4(b) and figure 4(c). Comparatively, at lower $V_{\text{Grid}}$, the first peak [figure 4(b)] could be due to the generation of electrons in the graphene layer, which are then collected by the grid probe at $V_{\text{Grid}} < 80$ V and by the anode when 80 V $< V_{\text{Grid}} < 110$ V.
Since the grid and graphene were far (\(\sim 1 \text{ mm}\)) from the emitter, they would not cause any significant field enhancement on the device to account for the large anode currents measured. The polarity of \(V_{\text{Grid}}\) could also influence the force experienced by the electrons in the absence of the graphene layer, causing electrons with initial energy lower than 20 eV when emerging from the gate to be stopped. Although graphene has been reported to have low secondary yield, and often used to suppress secondary emission from surfaces, we found that the yields at low energies was significant [27, 28]. From the simulations, we have demonstrated that secondary electrons may be the main reason for this increase, it could also be caused by electron-hole pair generation leading to further enhancement in multiple layers [29]. The mechanism of electron transparent graphene layers at low energy would therefore need to incorporate the effect of secondary electron generation and perhaps electron-hole pair generation, with the electrons escaping into vacuum.

Despite our inference that the multiplication of electrons in the graphene membrane could be due to secondary emission, further work should be performed to study the causes, such as residues in the graphene, which could dope the layer. In the future, using a modified structure and measuring the energy spectra of the electrons emerging from the aperture could also give indication of the origin of the electron multiplication, which is presently difficult to perform due to the short distances between the anode and the grid. Nonetheless, the ability of selective transparency of graphene to electrons relative to gas molecules opens up new avenues for an interface which separates an ultra-high vacuum region to a poor vacuum region. At low energies, secondary electrons or electron multiplication could be used in applications for high electron flux, such as in low-resolution x-ray imaging, or electron impact ionization.
5. Conclusions

In conclusion, we characterized the electron transmission through different number of suspended graphene layers on a nitride grid using a low-voltage field emission source. Interestingly, in the presence of the graphene layers, at certain voltages larger currents are measured at the anode, compared to the case without the grid or the graphene. This suggests that low-energy electrons impinging on the graphene layer generate inelastically scattered electrons that are transmitted to the anode. This was verified by modeling the electron trajectories and incorporating secondary emission physics in a graphene membrane. This work demonstrates that although graphene is electron transparent, the secondary emission should be taken into consideration at lower kinetic energies.

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References

[1] Morozov S V, Novoselov K S, Katsnelson M I, Schedin F, Elias D C, Jaszczak J A and Geim A K 2008 Physical Review Letters 100 URL https://dx.doi.org/10.1103/physrevlett.100.016602
[2] Lee C, Wei X, Kysar J W and Hone J 2008 Science 321 385–388 URL https://dx.doi.org/10.1126/science.1157996
[3] Weatherup R S 2018 Topics in Catalysis 61 2085–2102 URL https://dx.doi.org/10.1007/s11244-018-1075-2
[4] Sun P Z, Yang Q, Kuang W J, Stebunov Y V, Xiong W Q, Yu J, Nair R R, Katsnelson M I, Yuan
S J, Grigorieva I V, Lozada-Hidalgo M, Wang F C and Geim A K 2020 Nature 579 229–232 URL https://dx.doi.org/10.1038/s41586-020-2070-x

[5] Meyer J C, Girit C O, Crommie M F and Zettl A 2008 Nature 454 319–322 URL https://dx.doi.org/10.1038/nature07094

[6] Stoll J D and Kolmakov A 2012 Nanotechnology 23 505704 URL https://dx.doi.org/10.1088/0957-4484/23/50/505704

[7] Wang L, Williams C M, Boutiller M S H, Kidambi P R and Karnik R 2017 Nano Letters 17 3081–3088 URL https://dx.doi.org/10.1021/acs.nanolett.7b00442

[8] Bunch J S, Verbridge S S, Alden J S, van der Zande A M, Parpia J M, Craighead H G and McEuen P L 2008 Nano Letters 8 2458–2462 URL https://dx.doi.org/10.1021/nl801457b

[9] Gomer R 1994 Surface Science 299-300 129–152 URL https://dx.doi.org/10.1016/0039-6028(94)90651-3

[10] Karaulac N, Guerrera S A, Akinwande A I, Cole M T and Milne W I 2017 Field emission from silicon tips embedded in a dielectric matrix 2017 30th International Vacuum Nanoelectronics Conference (IVNC) (IEEE) URL https://dx.doi.org/10.1109/ivnc.2017.8051582

[11] Rughoobur G, Zhao J, Jain L, Zubair A, Palacios T, Kong J and Akinwande A I 2020 Enabling atmospheric operation of nanoscale vacuum channel transistors 2020 Device Research Conference (DRC) (IEEE) URL https://dx.doi.org/10.1109/drc46940.2019.9046454

[12] Li C, Cole M T, Lei W, Qu K, Ying K, Zhang Y, Robertson A R, Warner J H, Ding S, Zhang X, Wang B and Milne W I 2013 Advanced Functional Materials 24 1218–1227 URL https://dx.doi.org/10.1002/adfm.201300322

[13] Jeon H, Choi Y C, Park S, Kang J T, Go E, Lee J W, Kim J W, Jeong J W and Song Y H 2017 Carbon 119 371–377 URL https://dx.doi.org/10.1016/j.carbon.2017.04.046

[14] Murakami K, Igar T, Mitsuishi K, Nagao M, Sasaki M and Yamada Y 2019 ACS Applied Materials & Interfaces 12 4061–4067 URL https://dx.doi.org/10.1021/acsami.9b17468

[15] Guerrera S A and Akinwande A I 2016 Nanotechnology 27 295302 URL https://dx.doi.org/10.1088/0957-4484/27/29/295302

[16] Rughoobur G, Jain L and Akinwande A I 2019 Towards vacuum-less operation of nanoscale vacuum channel transistors 2019 Device Research Conference (DRC) (IEEE) URL https://dx.doi.org/10.1109/drc46940.2019.9046454

[17] Wagner S, Weisenstein C, Smith A D, Östling M, Kataria S and Lemme M C 2016 Microelectronic Engineering 159 108–113 URL https://dx.doi.org/10.1016/j.mee.2016.02.065
[18] Schindelin J, Arganda-Carreras I, Frise E, Kaynig V, Longair M, Pietzsch T, Preibisch S, Rueden C, Saalfeld S, Schmid B, Tinevez J Y, White D J, Hartenstein V, Eliceiri K, Tomancak P and Cardona A 2012 *Nature Methods* 9 676–682 URL https://dx.doi.org/10.1038/nmeth.2019

[19] Rughoobur G, Karaulac N, Jain L, Omotunde O O and Akinwande A I 2020 *Nanotechnology* 31 335203 URL https://dx.doi.org/10.1088/1361-6528/ab8edf

[20] Fowler R H and Nordheim L 1928 *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences* 119 173–181 URL https://dx.doi.org/10.1098/rspa.1928.0091

[21] Forbes R G and Deane J H B 2007 *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences* 463 2907–2927 URL https://dx.doi.org/10.1098/rspa.2007.0030

[22] Cao M, Zhang X S, Liu W H, Wang H G and Li Y D 2017 *Diamond and Related Materials* 73 199–203 URL https://dx.doi.org/10.1016/j.diamond.2016.09.019

[23] Shih A, Yater J, Hor C and Abrams R 1997 *Applied Surface Science* 111 251–258 URL https://dx.doi.org/10.1016/s0169-4332(96)00729-5

[24] Rughoobur G, Sahagun A, Ilori O O and Akinwande A I 2020 *IEEE Transactions on Electron Devices* 67 3378–3384 URL https://dx.doi.org/10.1109/ted.2020.3001082

[25] Rughoobur G, Jain L and Akinwande A I 2020 Low energy electron transmission through suspended graphene layers 2020 33rd International Vacuum Nanoelectronics Conference (IVNC) (IEEE) URL https://doi.org/10.1109/IVNC49440.2020.9203389

[26] Hallam T, Cole M T, Milne W I and Duesberg G S 2013 *Small* 10 95–99 URL https://dx.doi.org/10.1002/smll.201300552

[27] Luo J, Tian P, Pan C T, Robertson A W, Warner J H, Hill E W and Briggs G A D 2011 *ACS Nano* 5 1047–1055 URL https://dx.doi.org/10.1021/nn102579f

[28] Nguyen H K A, Mankowski J, Dickens J C, Neuber A A and Joshi R P 2018 *AIP Advances* 8 015325 URL https://dx.doi.org/10.1063/1.5019360

[29] Phan V N and Fehske H 2012 *New Journal of Physics* 14 075007 URL https://dx.doi.org/10.1088/1367-2630/14/7/075007