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# NEAR THRESHOLD PHOTOEMISSION FROM GRAPHENE COATED Cu SINGLE CRYSTALS

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*Abstract*

The brightness of electron beams emitted from photocathodes plays a key role in the performance of accelerator applications like X-ray free electron lasers (XFELs) and ultrafast electron diffraction (UED) experiments. In order to achieve a maximum beam brightness, the electrons need to be emitted from photocathodes with the smallest possible mean transverse energy (MTE). Recent studies have looked at the effect that a graphene coating has on the quantum efficiency (QE) of the cathode [1]. However, there have not yet been any investigations into the effect that a graphene coating has on the MTE. Here we report on MTE and QE measurements of a graphene coated Cu(110) single crystal cathode at room temperature and cryogenic temperatures. At room temperature, a minimum MTE of 25 meV was measured at a 4.20 eV photon energy (295 nm). This MTE remained stable at 25 meV over several days. At 77 K, a minimum MTE of 9 meV was measured at a 4.28 eV photon energy (290 nm). We perform density functional theory (DFT) calculations to look at the effects of a graphene coating on a Cu(111) surface state. These calculations show that the graphene coating reduces the radius of the surface state, allowing for emission from a lower transverse energy state in comparison to bare Cu(111).

## INTRODUCTION

The performance of accelerator applications like x-ray free electron lasers (XFELs) and ultrafast electron diffraction (UED) and microscopy (UEM) experiments is dependant upon the brightness of the electron beam emitted from photoinjectors. For XFELs a brighter electron beam will lead to an increase of x-ray lasing and pulse energies [2], as well as the development of smaller and more accessible university scale facilities [3]. For single shot UED, an increase in brightness will open new doors by enabling the study of larger crystal lattices like proteins [4]. And for stroboscopic UED/UEM experiments, a brighter beam will reduce the signal to noise ratio and the data acquisition time [5]. The key figure of merit in determining the brightness of the electron beam generated from a photoinjector is the mean transverse energy (MTE) of the photocathode [6]. Hence, understanding and reducing the MTE is necessary to achieving brighter electron beams from future, and existing, photoinjectors.

For metallic cathodes, the MTE is approximately equal to MTE = $E_{excess}/3$, where the excess energy ($E_{excess}$) is defined as the difference between the photon energy ($\hbar\omega$) and the work function ($\phi$) of the photocathode [7]. For excess energies that are low and negative, the electron emission occurs from tail of Fermi distribution and the minimum MTE is $k_bT$ where $k_b$ is the Boltzmann constant and T is the temperature of the electrons in the lattice [8]. At low laser fluences the lattice and electrons are in equilibrium and thus their temperatures are equivalent. At room temperature, the minimum MTE is 25 meV, a result that has been experimentally verified with Sb photocathodes [9]. By reducing the temperature of the cathode to cryogenic temperatures, MTEs below 25 meV have been demonstrated experimentally [10]. However, as the cathode reaches cryogenic temperatures it becomes challenging to reach this minimum MTE. This is because near threshold the transmission probability is very small for low energy electrons [10, 11]. In addition, the surface nonuniformities of physical roughness and work function variation can lead to an increase in MTE [9, 12]. In order to minimize the effects that these nonuniformities have on the MTE, it is necessary to use atomically ordered single crystalline cathodes rather than polycrystalline cathodes [13].

For single crystals, the transverse momentum is conserved during photoemission. Therefore to minimize the MTE, it is crucial that cathodes are chosen that allow only electron emission from electronic states with a low transverse momentum [14]. By utilizing the low transverse energy states of a Cu(100) single crystal, a 5 meV MTE has been demonstrated when operating near the photoemission threshold and at 30 K [10].

A recent investigation looked into the effect that a graphene coating has on the QE of a Cu(110) single crystal [1]. They found that the graphene coating does not negatively impact the QE, and instead increases the stability by preserving the QE even up to atmospheric conditions. While this result is promising, further studies are needed to investigate the impact that a graphene coating has on the MTE.

In this paper we measure the MTE of a graphene coated Cu(110) single at room temperature and cryogenic temperatures. Our measurements show that we reach the thermal limit of 25 meV at room temperature, and that the MTE remains stable over several days. When the cathode is cooled down to 100 K and 77 K, we measured an MTE of 13 meV and 9 meV respectively. We show that these low, sub-25 meV, MTEs can be attained with a single anneal at 345°C for 3 hours rather than the intense in-situ surface prepara-

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tion that was required for the 5 meV result [10]. Lastly by using density functional theory (DFT) we have found that a graphene coating on a Cu(111) single crystal can lower the Fermi level and therefore restrict emission to electronic states with a lower transverse momentum near the photoemission threshold.

## EXPERIMENTAL SETUP

For this work a commercially purchased, mirror-polished, single crystal Cu(110) sample was sent to the Global Innovation Center at Kyushu University in Japan where it was coated with a monolayer of graphene. The graphene was grown using chemical vapor deposition with the procedure outlined here [15]. A high temperature anneal at the beginning of the growth process ensured that the graphene was grown on an atomically pristine Cu(110) single crystal. The sample was then transported to Arizona State University over several weeks in atmospheric conditions before being loaded into a UHV chamber with a base pressure of $5 \times 10^{-10}$ Torr and annealed at 345°C for 3 hours. It was then allowed to cool for 2 hours before being transported into the analysis chamber which has a base pressure of $1 \times 10^{-10}$ Torr. The analysis chamber contains low energy electron diffraction and Auger electron spectroscopy experiments as well as an electron energy analyzer. The Auger spectrum and LEED pattern indicated that the sample was an atomically pristine, and contaminant free, surface.

The electron energy analyzer consists of the sample and a time-of-flight based delayline detector arranged in a parallel plate configuration and separated by 4 cm $\pm$ 0.1 cm. A sub-picosecond laser was focused down to a sub-100 $\mu$m spot on the sample, and the emitted electrons were accelerated towards the detector by a voltage of 72 V. The delayline detector measures the transverse positions of the emitted electrons as well as the time of flight and the transverse momenta and energies are calculated trivially. Further details of this energy analyzer can be found here [16].

For this work a 130 fs pulse width, 500 kHz repetition rate laser was used along with a tunable wavelength optical parametric amplifier. The sample was cryogenically cooled and data was collected at 300 K, 100 K, and 77 K for wavelengths between 260 nm and 295 nm.

## RESULTS

Figure 1 shows the MTE plotted as a function of the time elapsed since the sample was annealed. We see that the MTE remained stable at the thermal limit for over 4 days (97.5 hours). This time is an order of magnitude longer than bare Cu(110). The sample was cooled down to cryogenic temperatures between 117 hours and 143 hours and the MTE was measured. After the cryocooled measurements concluded, the sample was allowed to reach room temperature and an MTE of 29.72 $\pm$ 10% meV was measured at just under 7 days (164.5 hours) since the sample was annealed. This indicates that the graphene coating significantly increases the stability of the cathode.

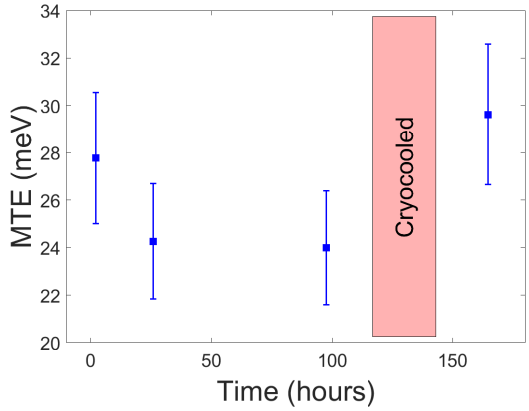


Figure 1: MTE measurements at room temperature as a function of the time elapsed since the sample was annealed. The error in the measurement was estimated to be 10%.

Figure 2 shows the MTE at wavelengths above, and near, threshold for 300 K, 100 K and 77 K. Between 260 nm and 280 the measured MTEs from all three temperatures are roughly the same. This is because the excess energy is large enough where it roughly obeys the $E_{excess}$=MTE/3 relationship established by Dowell and Schmerge. As we approach the photoemission threshold, the emission occurs more from the tail of the Fermi distribution and as a result the MTE decreases with temperature. The minimum MTE measured for 300 K was the thermal limit of 25 meV and occurred at 295 nm. For the cryocooled temperatures, it was not possible to measure linear photoemission at 295 nm. Instead for 100 K and 77 K, the minimum MTE occurred at 290 nm and was measured at 13 meV and 9 meV respectively.

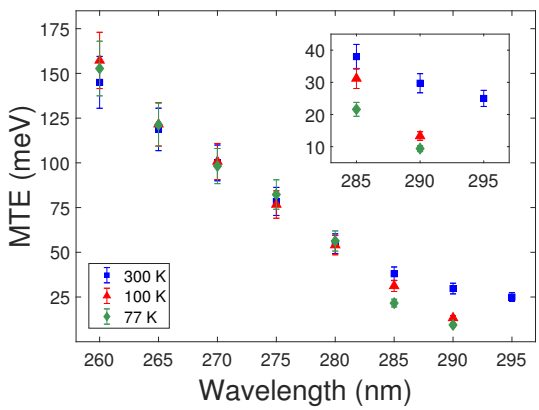


Figure 2: MTE measurements for near threshold photoemission from a graphene coated Cu(110) surface at 300 K, 100 K, and 77 K. The MTE for the cooled measurements reached a minimum of 13 meV and 9 meV for 100 K and 77 K respectively. For 100 K and 77 K it was not possible to measure linear data for 295 nm. The error in the measurement was estimated to be 10%.

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Figure 3 shows the QE between 260 nm and 295 nm for the three temperatures. The current was measured by treating every count measured by the delayline detector as an electron. However, this method does not account for every electron photoemitted by the cathode and instead puts a lower limit on the current. Hence, the actual QE is some unknown factor larger than the QE shown in Fig. 2. The QE for all three temperatures follows a similar trend as it approaches threshold, and it decreases with temperature as expected. At 290 nm we see that the QE drops by an order of magnitude. In spite of this drop, this QE is still sufficient for stroboscopic UED/UEM experiments.

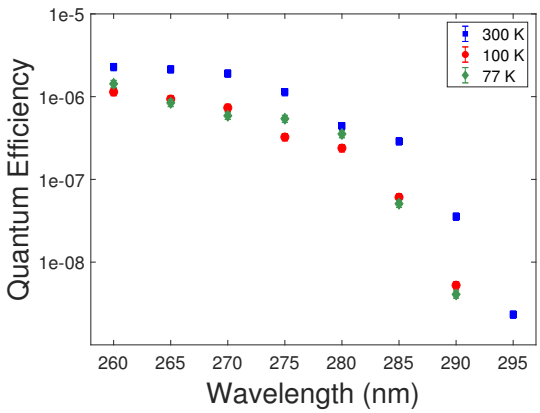


Figure 3: Measured QE values for 300 K, 100 K, and 77 K. The current measured represents only a fraction of the actual current emitted, and therefore the measured QE is some unknown fraction lower than the actual QE. For 100 K and 77 K it was not possible to measure linear data for 295 nm. The error in the measurement was estimated to be 10%.

An additional advantage that the graphene coating provides is a significant reduction in the surface preparation that is required to achieve the atomically ordered surface that is necessary to reach the thermal limit. Single crystals typically require repeated ion sputtering and high (600°C) annealing cycles in order to achieve a well ordered, atomically pristine surface. Current photoinjectors do not have those capabilities, and adding them requires significant and costly modifications. By capping the cathode with graphene, the well ordered single crystal is achieved during the high temperature anneal in the growth process, and the samples can be shipped in atmosphere to anywhere in the world. Then a single anneal at 345°C is all that is needed in-situ to have the cathode ready for operation in the photoinjector. These results represent a significant step forward into the viability of these graphene coated single crystals as realistic candidates for photoinjectors.

### *Density Functional Theory*

DFT calculations were performed on other copper single crystals to see the effects that a graphene layer had on the band structure. Figure 4 shows that for Cu(111) the surface state decreases in size due to a charge transfer between the copper and graphene. Therefore the electrons will emit from a smaller transverse energy state which will lead to a reduction in MTE near the photoemission threshold.

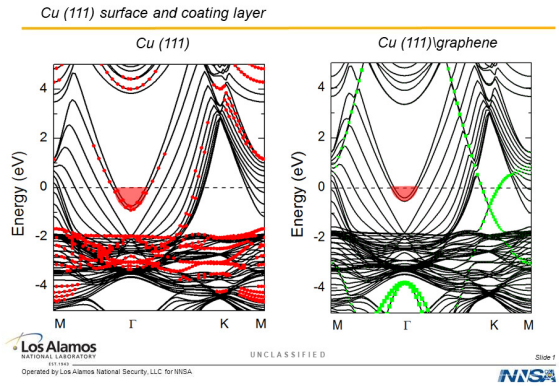


Figure 4: Comparison of the band structure for bare and graphene coated Cu(111). The presence of a graphene layer on Cu(111) leads to a reduction in the radius of the surface state at the center of the Brillouin zone. In comparison to the bare Cu(111), this allows electrons to emit from a smaller transverse energy state and therefore leads to a reduction in the MTE.

## CONCLUSION

In this work, we present MTE measurements for a graphene coated Cu(110) single crystal at room and liquid nitrogen temperatures. We show that a 25 meV MTE can be attained at room temperature with minimal surface preparation, and that it remains stable for several weeks. In addition, a sub-25 meV MTE can be attained when cooling the cathode down to cryogenic temperatures. The simplified surface preparation and low MTE makes graphene coated Cu single crystals a realistic candidate for low charge density accelerator applications. Lastly we present DFT calculations that show a graphene coating on a Cu(111) single crystal lowers the Fermi level and thus limits emission to lower transverse energy states.

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