

NEAR-THRESHOLD NONLINEAR PHOTOEMISSION FROM Cu(100)

C. J. Knill*, S. S. Karkare, Arizona State University, Tempe, USA
H. A. Padmore, Lawrence Berkeley National Laboratory, Berkeley, USA

Abstract

Photocathodes that have a low mean transverse energy (MTE) are crucial to the development of compact X-ray Free Electron Lasers (XFEL) and ultrafast electron diffraction (UED) experiments. For FELs, low MTE cathodes result in a lower requirement for electron energy when lasing at a defined energy, and for a defined electron energy result in lasing at higher energy. For UED experiments, low MTE cathodes give a longer coherence length, allowing measurements on larger unit cell materials. A record low MTE of 5 meV has been recently demonstrated from a Cu (100) surface when measured near the photoemission threshold and cooled down to 30 K with liquid Helium [1]. For UED and XFEL applications that require a high charge density, the low quantum efficiency (QE) of Cu (100) near threshold necessitates the use of a high laser fluence to achieve the desired charge density [2]. At high laser fluences the MTE is limited by nonlinear effects, and therefore it is necessary to investigate near photoemission threshold at these high laser fluences. In this paper we report on nonlinear, near-threshold photoemission from a Cu (100) cathode, and its affect on the MTE.

INTRODUCTION

Electron beam brightness from photocathodes is central to the performance of accelerator applications like XFEL and UED experiments. The beam brightness is proportional to the square of the intrinsic emittance of the photocathode, which is described by:

$$\epsilon_n = \sigma_x \sqrt{\frac{\text{MTE}}{mc^2}} \quad (1)$$

where MTE is the mean transverse energy of the photoemitted electrons, σ_x is the rms laser spot size on the cathode, m is the rest mass of an electron, and c is the speed of light in vacuum [3]. From Eq. (1), the true figure of merit in determining the emittance is the MTE of the photocathode. Thus finding photocathodes with the lowest possible MTE is essential to the advancement of these applications.

In recent years, several investigations have shown that the MTE is dependant on the difference between the photon energy and the work function (excess energy) [3], the lattice temperature [1], the nonlinear effects of multiphoton emission and electron heating [2, 4], and the band structure of the photocathode [5]. By minimizing contributions from the above factors, it has been demonstrated that an MTE of 5 meV can be achieved from a Cu (100) sample when measured near the photoemission threshold and cryocooled down to 30 K with liquid Helium [1].

In general, reducing the excess energy reduces the MTE. Therefore it is crucial to investigate the performance of the cathode using wavelengths that are very close to the photoemission threshold [3]. In order to achieve the smallest MTE, it is necessary that emission does not occur from electronic states with a large transverse momentum [5]. Hence selecting single crystal photocathodes, rather than polycrystalline photocathodes, with an appropriate band structure is essential to minimizing MTE. While the use of single crystals and near threshold photoemission have been investigated experimentally, the influence of nonlinear effects on MTE has only briefly been investigated experimentally on polycrystalline Cu [6], and not at the wavelengths and fluences typically used in photoinjectors. From theory we know that these nonlinear effects can alter the MTE [2, 4] significantly and it is critical to study them further.

When measuring the MTE under typical wavelength and fluence conditions used in photoinjectors, it is difficult to distinguish between nonlinear effects and space charge effects. Hence, there have not been any previous investigations into purely nonlinear effects at wavelengths (~265 nm) and fluences (0.1 mJ/cm² to 3 mJ/cm²) that are typically used in photoinjectors. Due to the affect that nonlinear photoemission can have on MTE, it is important to explore their contributions to MTE in this wavelength and fluence regime under experimental conditions that are free of space charge effects.

In this paper we present measurements of nonlinear photoemission near threshold on Cu (100), and investigate its affect on the MTE. In addition, we seek to investigate a previously unexplored transition between linear and nonlinear photoemission with increasing fluence for a single wavelength below threshold. Using measured data for below threshold nonlinear photoemission, and above threshold linear photoemission, we extrapolate to obtain a lower limit on the MTE at various fluences for 265 nm. With this extrapolation we obtain experimental information on the affects of nonlinear photoemission on the MTE at wavelengths and fluences typically used in photoinjectors.

EXPERIMENTAL SETUP

For this work a commercially bought, mirror-polished, single crystal Cu (100) sample was used. The sample was annealed for 2 hours in a UHV chamber with a pressure in the low 10⁻¹⁰ torr without ion bombardment. Hence, the sample can be considered bulk Cu (100) with a layer of oxide. This is expected to alter the work function, but as in this case there is no surface state emission, it is not expected to change the bulk Cu photoemission. The sample was then transferred under UHV into a time-of-flight based electron

* cknill2@asu.edu

energy analyzer that measures the 3-D electron energy and momentum distribution.

The electron energy analyzer consists of a sample and a delay-line-based detector arranged in a parallel plate configuration and separated by $4 \text{ cm} \pm 0.1 \text{ cm}$. A femtosecond pulsed laser with a pulse length of $150 \text{ fs} \pm 50 \text{ fs}$ is focused down to $40 \mu\text{m}$ onto the sample. A flip mirror is placed before the chamber that directs the laser beam towards a beam profiler in order to measure the spot size. The beam profiler and the sample are located an equal distance from the flip mirror. Neutral density (ND) filters are used to select a low enough laser intensity that results in at most one electron emitted per pulse. This is essential as the detector can resolve at most one electron per pulse, and it also ensures that space charge effects do not exist. The emitted electrons are accelerated towards the detector by an accelerating voltage ranging from 4 V to 72 V. The detector measures the x and y position of the electrons on the detector as well as their time-of-flight, and from that the transverse and longitudinal energies can be calculated in a straight forward manner. Further details on the energy analyzer can be found elsewhere [7]. A tunable wavelength optical parametric amplifier (LightConversion Orpheus) operating at a repetition rate of 500 kHz was used for this experiment.

RESULTS

In order to investigate both above, and below, near threshold photoemission, data was collected for photon wavelengths ranging from 260 nm to 290 nm in steps of 5 nm. The data was collected at room temperature for 20 minutes at each wavelength and power. By applying a linear fit to the square root of QE for two photon energies (4.59 eV and 4.68 eV) in the linear photoemission regime, the work function of the sample was found to be 4.56 eV. This is in good agreement with the work function of 4.59 eV from literature [8] which indicates that the surface is possibly quite clean. From Fig. 1 we see that the minimum value of MTE occurs at approximately the same photon energy as the determined work function.

In order to thoroughly investigate how changing the laser power results in a transition from linear to nonlinear photoemission, data was collected at 280 nm at several fluences. This wavelength was chosen because it is right below threshold, and a wide range of ND filters can be used to get a wide range of fluences that emit at most one electron emitted per pulse. At lower laser fluences emission is dominated by linear photoemission from the tail of the fermi distribution. At higher fluences the fraction of two-photon emission increases significantly and the photoemission process starts becoming nonlinear.

In Fig. 2a we have plotted the number of electrons per second against the fluence on a log-log scale. We see that the slope of counts vs fluence increases on the log-log plot indicating an increasing fraction of nonlinear emission and quantum efficiency that increases with laser fluence. Similarly, in Fig. 2b we see that the MTE increases as the emission

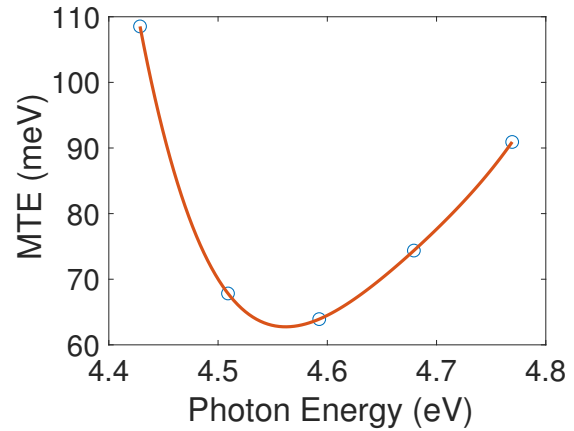


Figure 1: MTE measured for several wavelengths around the photoemission threshold for a laser fluence of 10^{-7} mJ/cm^2 . From the fitted curve we see the minimum value of the MTE occurs at approximately the same energy as the measured work function. MTE increases below threshold due to effects of nonlinear photoemission.

transitions from linear to nonlinear. The increase in MTE is significantly more sensitive to nonlinear effects than the increase in the quantum efficiency.

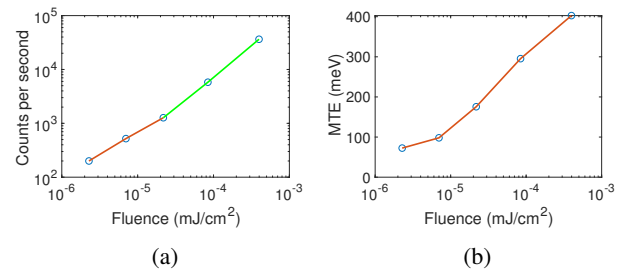


Figure 2: (a) The electron counts per seconds as a function of fluence for 280 nm. The shift from the red slope to the green slope at $2.18 \times 10^{-5} \text{ mJ/cm}^2$ is due to the transition between linear and nonlinear emission. (b) MTE as a function of fluence for 280 nm. We see that the MTE increases as the emission transitions from linear to nonlinear.

Extrapolating to 265 nm

While the above data contains a lot of interesting information, it isn't involving a wavelength or fluence that is particularly useful to photoinjectors. For that we would want to explore how the nonlinearities affect the MTE for 265 nm in the 0.1 mJ/cm^2 to 3 mJ/cm^2 range. And so we have used the data we collected for 265 nm at 10^{-7} mJ/cm^2 as a linear contribution, the data for 290 nm at 10^{-4} mJ/cm^2 as a nonlinear contribution, and extrapolated it to get the MTE as a function of the fluence F using Eq. (2).

$$\text{MTE} = \frac{1}{N} \left(\frac{N_l \cdot \text{MTE}_l}{F_l/F} + \frac{N_{nl} \cdot \text{MTE}_{nl}}{(F_{nl}/F)^2} \right) \quad (2)$$

where $N = \frac{N_l}{F_l/F} + \frac{N_{nl}}{(F_{nl}/F)^2}$ and N_l , MTE_l , and F_l correspond to the electron counts per second, the MTE, and fluence, respectively, for the linear contribution. While those with “nl” subscript correspond to the nonlinear contribution. The specific data that was used in this extrapolation is shown in Table 1.

Table 1: Experimental Data Used in Extrapolation

(nm)	Counts/sec	MTE (meV)	Fluence (mJ/cm ²)
265	46500 (N_l)	74 (MTE_l)	3.82×10^{-7} (F_l)
280	200 (N_l)	72.52 (MTE_l)	2.28×10^{-6} (F_l)
290	52300 (N_{nl})	533 (MTE_{nl})	5.63×10^{-4} (F_{nl})

This method essentially assumes that the contribution to MTE due to nonlinear photoemission does not change significantly from the wavelengths of 290 nm to 265 nm, and that the MTE due to nonlinear photoemission remains constant over a wide range of fluences above 5.63×10^{-4} mJ/cm². Due to these assumptions, this method gives only a lower limit to the MTE, and also results in an MTE estimate that becomes invariant with fluence for very large fluences as can be seen in Fig. 3.

Since we have several data points for 280 nm at different laser powers, this method was checked for accuracy with the measured data and the results are presented in Fig. 4. For this accuracy test, the 280 nm, 14.3 nW data is used as a linear contribution. Although this wavelength is below the photoemission threshold, from Fig. 2 we see that at this low power we are still observing linear photoemission from the Fermi tail of the distribution. From Fig. 4 we see that this extrapolation provides a lower limit on the MTE that becomes less accurate with increasing fluence as expected.

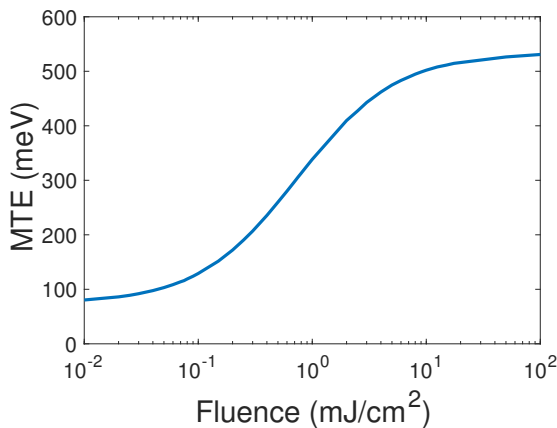


Figure 3: MTE calculated from Eq. (2) for a wide range of laser fluences for 265 nm. The MTE increases by a factor of 3 to 4 in the fluence regime (10^{-1} mJ/cm² to 10^2 mJ/cm²) typically used in photoinjectors. This dramatic increase can be attributed to nonlinear effects.

Despite its accuracy limitations, this method does provide useful information for which fluences these nonlinear effects begin to affect the MTE. Hence we apply it to 265 nm with the data shown in Fig. 3. We show it over a wide range of fluences to see at which point the MTE goes from being dominated by purely linear contributions, to the point where the nonlinear effects begin to effect MTE, and finally to the region where nonlinear contributions dominate. When focusing on fluences that are typically used in photoinjectors, the MTE increases by a factor of 3 to 4 between 0.1 mJ/cm² and 2 mJ/cm² as a result of the nonlinear photoemission effects becoming more significant.

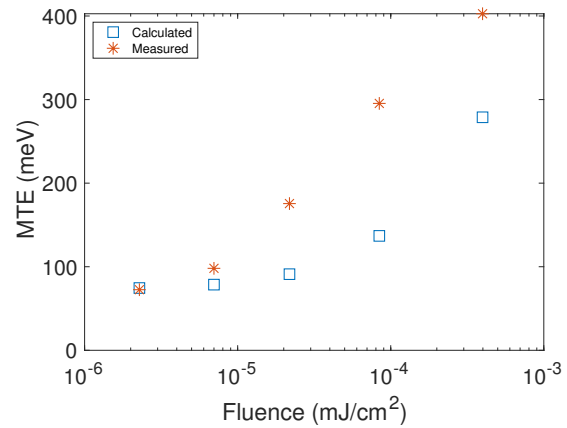


Figure 4: Comparison of the measured MTE for 280 nm (red stars) and the MTE calculated (blue squares) using Eq. (2) to determine the accuracy of this extrapolation. We see that it provides a lower limit and becomes less accurate with increasing fluence.

CONCLUSION

In this work, we have measured nonlinear photoemission of Cu (100) near threshold, and explored its affect on the MTE. We have shown that the MTE increases significantly due to nonlinear effects near threshold. Lastly, we have used the collected linear and nonlinear photoemission data, and extrapolated it to provide a lower limit for MTE at 265 nm with laser fluences typically used in photoinjectors. It was calculated that the MTE increases by a factor of 3 to 4 in the fluence range of 0.1 mJ/cm² and 3 mJ/cm² as a result of nonlinear photoemission. Experiments in the near future will look at the same effects, with pristine atomically clean Cu(100). Future studies will also be performed to use laser pulses of ~10 ps in length which are more often used in photoinjectors.

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