

MULTI-PHOTON PHOTOEMISSION AND ULTRAFAST ELECTRON HEATING IN Cu PHOTOCATHODES AT THRESHOLD

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Abstract

Operating photocathodes near the photoemission threshold holds the promise of yielding small intrinsic emittance, at the cost of significantly reduced quantum efficiency. In modern femtosecond photoemission electron sources, this requires a very high intensity (10s of GW/cm²) to extract a useful quantity of electrons. At this intensity, the electron occupation function is far from equilibrium and evolves rapidly on sub-ps timescales. Thus, ultrafast laser heating and multiphoton photoemission effects may play a significant role in emission, thereby increasing the minimum achievable emittance. In this work, we use a Boltzmann equation approach to calculate the non-equilibrium occupation function evolution in time for a copper photocathode, yielding a prediction of quantum efficiency and mean transverse energy as a function of input intensity.

INTRODUCTION

Ultrafast electron pulses are used in a wide array of research fields that involve the resolution of time-resolved structural dynamics, ranging from free electron lasers [1] to ultrafast electron diffraction [2]. In both of these cases, beam brightness is a critical figure of merit, and is fundamentally limited by the mean transverse energy (MTE) at the photocathode. In the photocathode community, it has long been thought that operating a photocathode at the threshold photon energy is optimal to minimize the MTE because MTE originates from excess energy of electrons after being emitted from the cathode.

Dowell and Schmerge utilized Spicer's three-step photoemission model to obtain expressions for the key parameters of a photocathode, the Quantum Efficiency (QE) and MTE, under following assumptions: free-electron Fermi gas model, single photon absorption, flat density of states, and the absolute zero temperature [3]. This work was further extended to account non-zero fixed electronic temperature (T_e) and realistic density of states [4]. The model predicts asymptotic behavior of MTE when the excess energy ($h\nu - \phi$) is varied: near the threshold ($h\nu \approx \phi$), MTE approaches the thermal energy ($MTE \approx k_b T_e$) while for a photon energy well above the threshold ($h\nu \gg \phi$), MTE linearly increases ($MTE \approx (h\nu - \phi)/3$). This calculation predicts the minimum MTE at the threshold photon energy under the assumption of single photon absorption and constant electronic temperature.

Metallic photocathodes are attractive as an ultrafast electron source due to their prompt response time (< 50 fs) and resistance to poor vacuum conditions, but suffer from low QE ($\sim 10^{-5}$) at the threshold photon energy. Due to the low QE, operating metal photocathodes requires a high laser fluence (10s of mJ/cm²), which for femtosecond pulses results in a very high laser intensity (10s of GW/cm²) [5]. During laser illumination, the photon energy is initially absorbed by only electrons, and the occupation function deviates from the thermal equilibrium Fermi-Dirac distribution. For metallic photocathodes, the electron-electron scattering thermalizes the occupation function over 100 fs time scale, resulting in a few thousand Kelvin of electronic temperature while the lattice temperature stays relatively constant at the initial temperature. The electron-phonon scatterings thermalize electron-lattice temperature over 10 ps time scale. Previously, a two temperature model was used to calculate electron-phonon equilibration dynamics and predicted the photocathode performance under high fluence irradiation with the assumption of single photon absorption and instant thermalization of electron occupation function [5].

In this proceeding, we extend a Boltzmann equation approach originally developed by Rethfeld *et al.* [6–8] to calculate photoemission parameters (MTE, QE). This approach has the capability to account for multi-photon absorption and femtosecond scale non-thermal equilibrium dynamics, and can be utilized to predict the performance of a copper photocathode irradiated by high fluence ultrafast laser.

BOLTZMANN EQUATION

Before the laser irradiation, the initial electron occupation function follows the Fermi-Dirac distribution in thermal equilibrium. For tracking non-equilibrium occupation function over time, we apply the Boltzmann equation with homogeneous thin film assumption, allowing us to neglect spatial variation of the occupation:

$$\frac{df(\vec{k}, t)}{dt} = \left. \frac{\partial f(\vec{k}, t)}{\partial t} \right|_{el-el} + \left. \frac{\partial f(\vec{k}, t)}{\partial t} \right|_{absorb} + \left. \frac{\partial f(\vec{k}, t)}{\partial t} \right|_{el-phon} \quad (1)$$

In this work, we also neglect the electron-phonon scattering term, as all simulations were done in sub-100 fs regime. The Electron-electron scattering term and photon absorption term are described in detail in the following subsections, and are wholly derived from the model proposed in Refs. [6–8].

Electron-electron Scattering

Electron-electron collisions are modeled via Coulomb interaction between two electrons subject to both energy

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conservation ($E_i = E_f$) and momentum conservation ($\vec{\Delta k} = \vec{k}_2 - \vec{k}_1 = \vec{k}_3 - \vec{k}$) [6, 7]. The Fourier transform of the screened Coulomb potential $V_{ee}(r)$ gives the matrix element in an isotropic system:

$$|M_{ee}(\Delta k, \kappa)|^2 = \left(\frac{e^2}{\epsilon_0 \Omega} \frac{1}{|\Delta k|^2 + \kappa^2} \right)^2 \quad (2)$$

where $\kappa^2 = \frac{e^2 m_e}{\pi^2 \hbar^2 \epsilon_0} \int_0^\infty f(k) dk$ is the screening parameter, and Ω is the unit cell volume. The Pauli exclusion principle is expressed with occupation functions, $f_i = f(k_i)$, as [6, 7]:

$$\mathcal{F} = f_1 f_3 (1 - f)(1 - f_2) - f f_2 (1 - f_1)(1 - f_3). \quad (3)$$

Thus, the electron-electron scattering term is written as:

$$\left. \frac{\partial f(\vec{k})}{\partial t} \right|_{el-el} = \frac{2\pi}{\hbar} \sum_{\vec{k}_1} \sum_{\vec{k}_3} |M_{ee}|^2 \mathcal{F} \delta(E_i - E_f). \quad (4)$$

In Ref. [7], this expression is further simplified under isotropic one band system assumption, $|k(E)|^3 = 3\pi^2 \int_0^E d\epsilon D(\epsilon)$, an assumption we also make here.

Photon Absorption

As in the model of [6–8], photon absorption is modeled by the inverse bremsstrahlung process where electrons absorb photons mediated by ion/nuclei collisions to conserve energy and momentum ($\Delta \vec{k} + \vec{k} = \vec{k}_f$). The Coulomb interaction between the electron and ions has the identical mathematical expression with the electron-electron scattering case while the interaction Hamiltonian between the electron and light vector potential, $\vec{A} \cdot \vec{p}$, introduces an additional Bessel function term, $J_\ell^2(e\vec{E}_L \cdot \Delta \vec{k} / m_e \omega_L^2)$, multiplied to the Coulomb interaction matrix element for ℓ -photon absorption. Therefore,

$$\left. \frac{\partial f(\vec{k})}{\partial t} \right|_{absorb} = \frac{2\pi}{\hbar} \sum_{\Delta \vec{k}} \sum_{\ell} |M_{ee}|^2 J_\ell^2 \mathcal{F} \delta(E_i - E_f) \quad (5)$$

where \mathcal{F} is again the Pauli exclusion principle term [6, 7]:

$$\mathcal{F} = f(k_f)(1 - f(k)) - f(k)(1 - f(k_f)). \quad (6)$$

Similar to Eq. 4, the one-band isotropic system further simplifies the expression [7] and is used throughout.

PHOTOCATHODE CALCULATION

In this section, we derive expressions for QE and MTE based on numerically calculated time dependent occupation function $f(E, t)$ from the previous section.

Quantum Efficiency

First, we describe current density that will be used in calculations for both QE and MTE. The current density flux of electrons with energy E in the direction θ away from the normal direction can be expressed as:

$$j(E, t, \theta) = e f(E, t) D(E) v(k(E)) \cos \theta \quad (7)$$

with $v(k(E)) = \hbar k / m_e$. Then, the total number of escaped electrons for unit area is:

$$N_e = \int_0^t dt \int_{E_f + \phi}^\infty dE \int_{\cos \theta_E}^1 d(\cos \theta) \frac{j(E, t, \theta)}{e} \Big| \int_{-1}^1 d(\cos \theta) \quad (8)$$

where $\cos \theta_E = \sqrt{(E_f + \phi) / E}$.

Though the electric field of the laser pulse is used as a simulation input, the inverse bremsstrahlung term (which in our model ignores interband absorption) is known not to produce the correct skin depth of the material [8]. Thus, rather than relying on the input fluence, we calculate the number of absorbed photons post-facto calculating the change of internal energy for unit volume, ΔE :

$$N_{ph} = \frac{\Delta E d_s}{\hbar \omega_L} = \frac{d_s}{\hbar \omega_L} \int_0^\infty dE D(E) (f(E, t) - f(E, 0)) E \quad (9)$$

where 13 nm was used for the skin depth of copper, d_s [5]. We use this change of internal energy as the actual absorbed fluence in subsequent calculations. Then, the QE can be expressed as:

$$QE = N_e(1 - R) / N_{ph} \quad (10)$$

with R as the reflectivity of copper.

Mean Transverse Energy

MTE is the averaged transverse energy ($(p \sin \theta)^2 / 2m_e$) of emitted electrons. For the emitted free electrons, we use $p^2 / 2m_e = E$. Then, MTE is expressed as:

$$MTE = \frac{\int_0^t dt \int_{E_f + \phi}^\infty dE \int_{\cos \theta_E}^1 d(\cos \theta) E \sin^2 \theta j(E, t, \theta)}{\int_0^t dt \int_{E_f + \phi}^\infty dE \int_{\cos \theta_E}^1 d(\cos \theta) j(E, t, \theta)} \quad (11)$$

RESULT

Figure 1 demonstrates the simulation result of 50 fs laser irradiation on copper with absorbed fluence of $(3.4 \pm 0.1) \times 10^{-5}$ mJ/cm². The spectral response agrees well with the experiment values measured from a hydrogen ion beam cleaned copper cathode [9]. Calculated MTE demonstrates an asymptotic behavior as it was predicted from earlier works as well. The small discrepancy on the slope of MTE over excess energy is also observed in Ref. [4] when a realistic density of states was used. In this fluence regime, single photon absorption dominates so that it agrees well with the conventional models.

In Fig. 2, 50 fs irradiation was simulated with absorbed fluence of 1.65 ± 0.05 mJ/cm². For both QE and MTE, it is noteworthy that there is no qualitative difference between high fluence (1.65 ± 0.05 mJ/cm²) results and low fluence ($(3.4 \pm 0.1) \times 10^{-5}$ mJ/cm²) results for photon energies well above the threshold. This is because at high photon energies, the single photon absorption dominates as it is in the low fluence regime. However, as the photon energy approaches the threshold, the number of electrons that can escape just from a single photon absorption decreases rapidly while

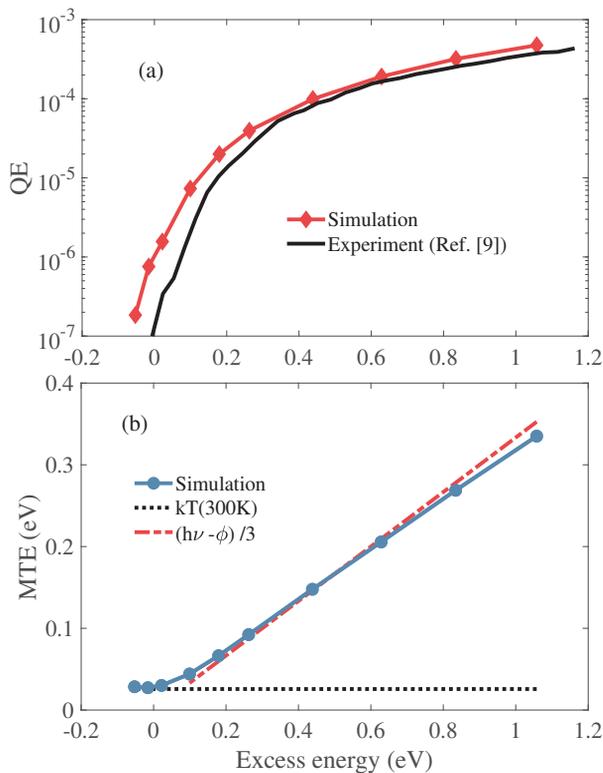


Figure 1: (a) QE calculated by Eq. 10 with absorbed fluence of $(3.4 \pm 0.1) \times 10^{-5} \text{ mJ/cm}^2$. The simulated values agree with the experiment values [9]. (b) MTE calculated by Eq. 11 with absorbed fluence of $(3.4 \pm 0.1) \times 10^{-5} \text{ mJ/cm}^2$.

electrons that absorb more than one photon can still escape. Since allowed directions of escape velocity is significantly suppressed near the threshold energy ($E_f + \phi$) of electrons, the two-photon interaction starts to become non-negligible well above the threshold photon energy. Therefore, the non-monotonic behavior of MTE is caused by increasing portion of electrons that absorb more than one photon as the the photon energy approaches the threshold. Once the photon energy falls below the threshold, the qualitative behavior of MTE is completely dominated by only two-photon absorption. Thus, MTE starts decreasing as the photon energy decreases as if it were when only single photon absorption was considered in earlier works.

CONCLUSION

In this proceeding, we report on Boltzmann equation approach for photocathode calculation to account multi-photon interaction and femtosecond scale non-equilibrium thermal dynamics. For a low fluence irradiation, the simulation result agrees well with experiment and prediction from earlier works. However, in case of a high fluence irradiation, multi-photon absorption becomes significant so that MTE no longer monotonically decreases as the photon energy approaches the threshold. These results demonstrate the importance of both multiphoton effects and thermalized electron

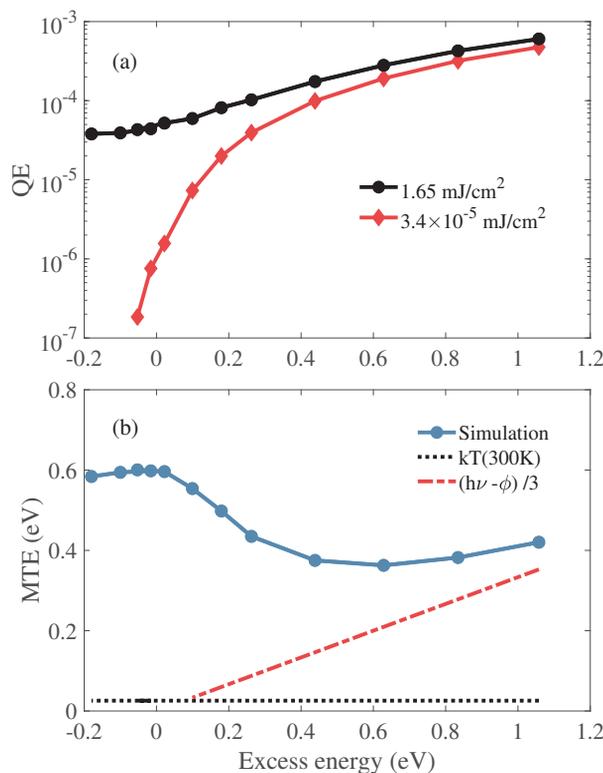


Figure 2: (a) QE calculated by Eq. 10 under high fluence irradiation ($1.65 \pm 0.05 \text{ mJ/cm}^2$) and low fluence irradiation ($(3.4 \pm 0.1) \times 10^{-5} \text{ mJ/cm}^2$). As the photon energy approaches the threshold, the QE deviates significantly due to two-photon absorption. (b) MTE calculated by Eq. 11 with absorbed laser fluence of $1.65 \pm 0.05 \text{ mJ/cm}^2$. Two-photon absorption results in non-monotonic behavior of MTE.

heating in photoemission beam brightness when operating near the photoemission threshold.

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