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Nonequilibrium Multiphoton photoemission in Metallic Photocathodes near threshold

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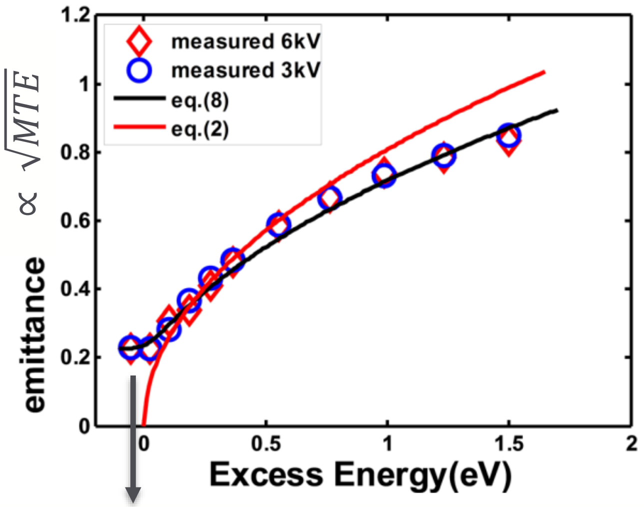
Outline

- History of Photoemission simulation
- Brightness
- Nonequilibrium dynamics - Boltzmann equation
- Result (Static regime)
- Result (Dynamic regime)
- Maximum achievable brightness

Photoemission Simulation (1)

- Dowell and Schmerge Photoemission calculation
  - ➢ Free – electron Fermi gas model (Copper)
  - ➢ Absolute Zero temperature
- Non-zero temperature (Vecchione)
  - ➢ Fermi-Dirac distribution
- Realistic density of states (Feng)
  - ➢ Matches experimental measurements
  - ➢ Static simulation

Feng *et al*.



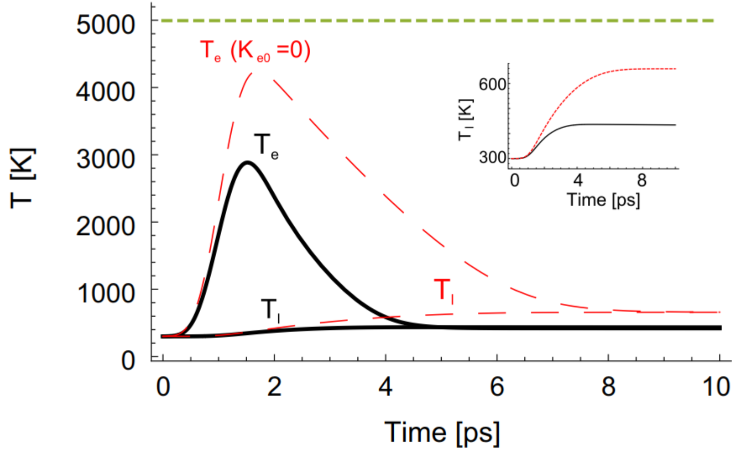
Partition theorem: *MTE ~ kT*

Dowell, Schmerge. *PRST-AB* 12.7 (2009): 074201
Vecchione *et al*. FEL2013 (2013): 424
Feng *et al*. *APL* 107.13 (2015): 134101
Dimitrov *et al*. *J. Appl. Phys*. 122.16 (2017): 165303

Photoemission Simulation (2)

- Two temperature model (Maxson)
  - ➢ Dynamic simulation of photoemission
  - ➢ Single photon absorption
  - ➢ Instant thermalization of electrons
- Boltzmann equation (our work)
  - ➢ Non-equilibrium dynamics (Rethfeld *et al*.)
  - ➢ Multi-photon photoemission
  - ➢ Femtosecond scale

Maxson *et al.*



Maxson *et al. Nucl Instrum Methods Phys Res A* 865 (2017) 99-104
Mueller, Rethfeld. *Phys. Rev. B* 87.3 (2013): 035139
Rethfeld *et al. Phys. Rev. B* 65.21 (2002): 214303

Source Brightness

$$Brightness \propto \frac{I}{\sigma_x \sigma_y \, MTE}$$

- I, photocurrent, depends on application
- $\sigma_{x,y}$, laser spot size, is limited by space-charge beam dynamics
- MTE is determined by material properties inside photocathode

Non-equilibrium dynamics

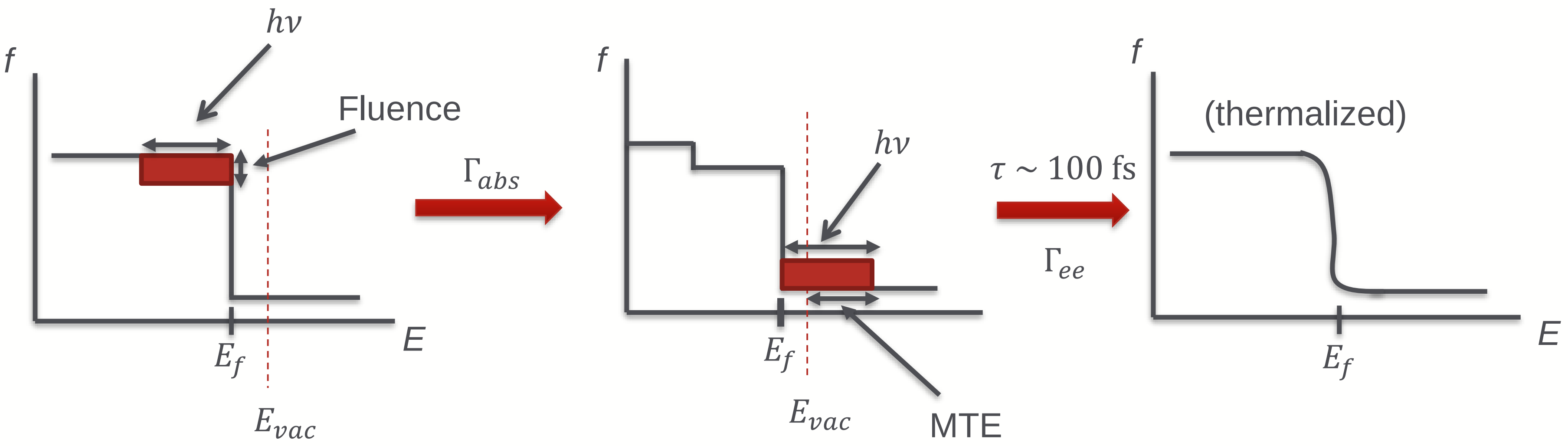
- In a thermal equilibrium, the occupation function *f(E)* follows the Fermi-Dirac distribution

$$f = \frac{1}{1 + e^{(E-E_f)/kT}}$$

- Non-equilibrium *f* can be calculated with Boltzmann equation

$$\frac{\partial f}{\partial t}(E,t) = \left(\frac{\partial f}{\partial t}\right)_{electron-electron} + \left(\frac{\partial f}{\partial t}\right)_{\substack{absorption \\ (electron-photon)}} + \left(\frac{\partial f}{\partial t}\right)_{electron-phonon}$$

Boltzmann Equation diagram (1)



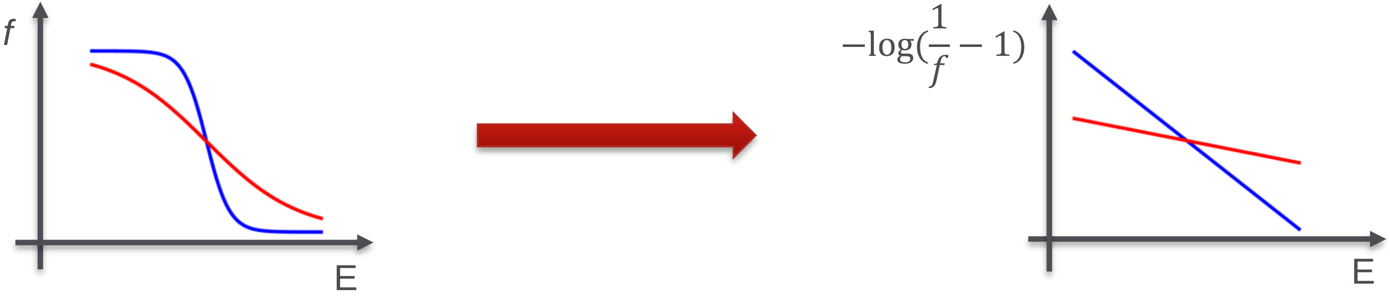
- Photon absorption excites electrons above Fermi level
- Electron energy distribution thermalizes by electron-electron scattering

Non-equilibrium Occupation

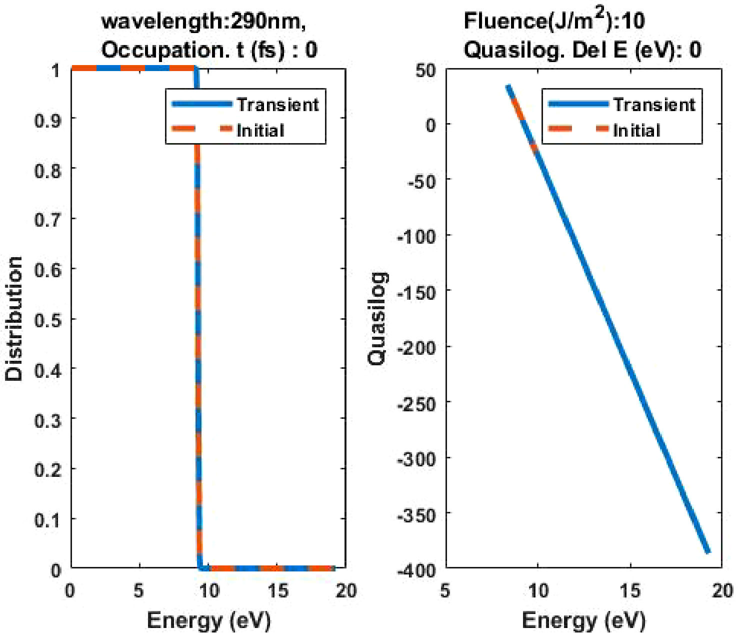
- Non-equilibrium *f* can be visualized intuitively when it’s plotted as

$$-\log\left(\frac{1}{f}-1\right)$$

Linear -> FD distribution, slope ~ 1/kT

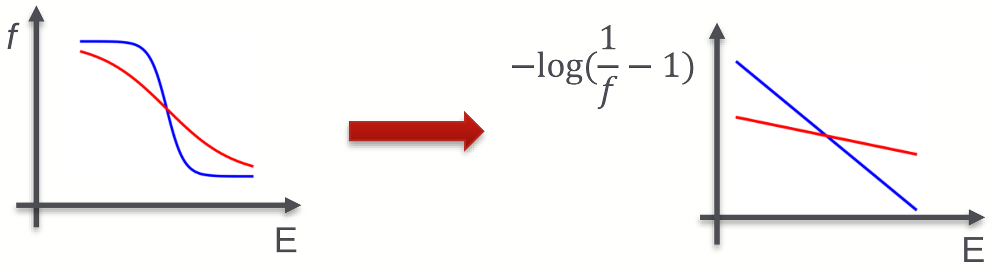


Tracking electron distribution

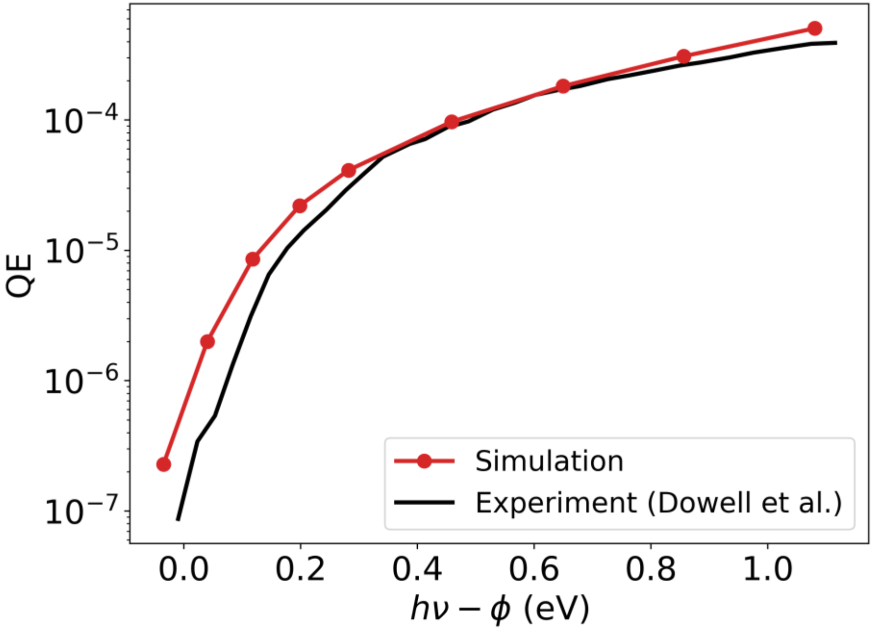

wavelength:290nm, Occupation. t (fs) : 0

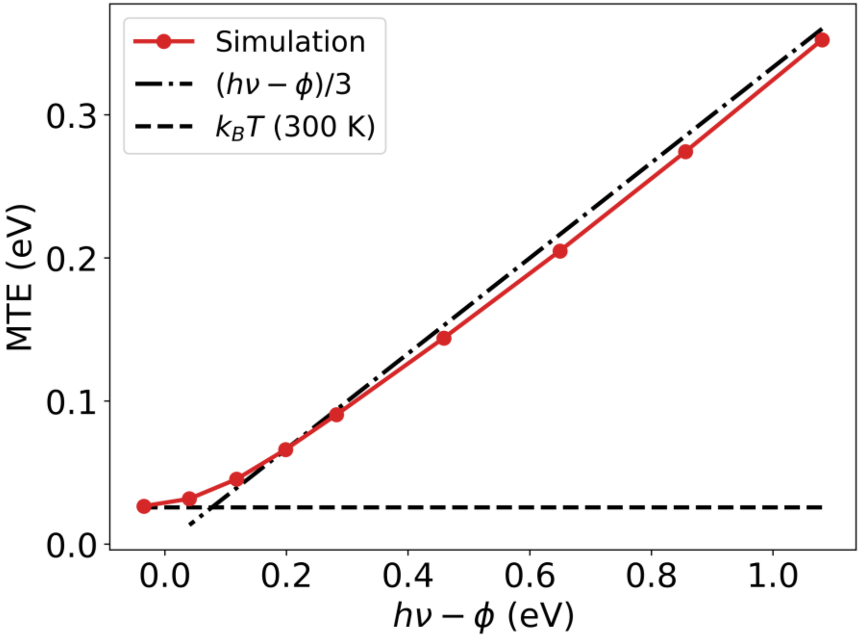
Fluence(J/m^2):10 Quasilog. Del E (eV): 0

- Distribution over time
  - ➢ 50 fs pulse duration
  - ➢ $h\nu$ = 4.3 eV (290 nm)
  - ➢ Absorbed Fluence = 1 mJ/cm^2
- As laser is turned off at 50 fs, Quasi-log ($-\log(\frac{1}{f}-1)$) plot becomes linear, which implies thermal equilibrium



Result (Static / low fluence regime)

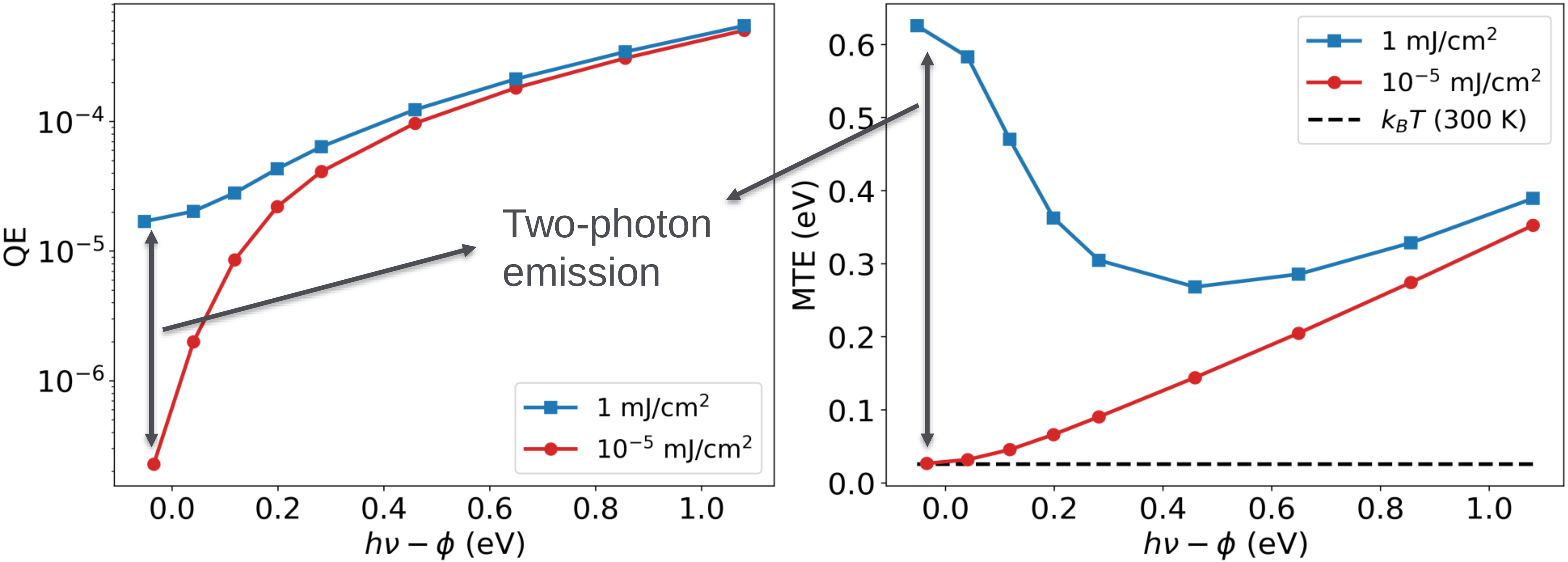




- In low fluence regime ($10^{-5}\ mJ/cm^2$), single photon absorption dominates. Therefore, it agrees well with experiment values and earlier predictions

Dowell *et al. PRST-AB* 9.6 (2006): 063502

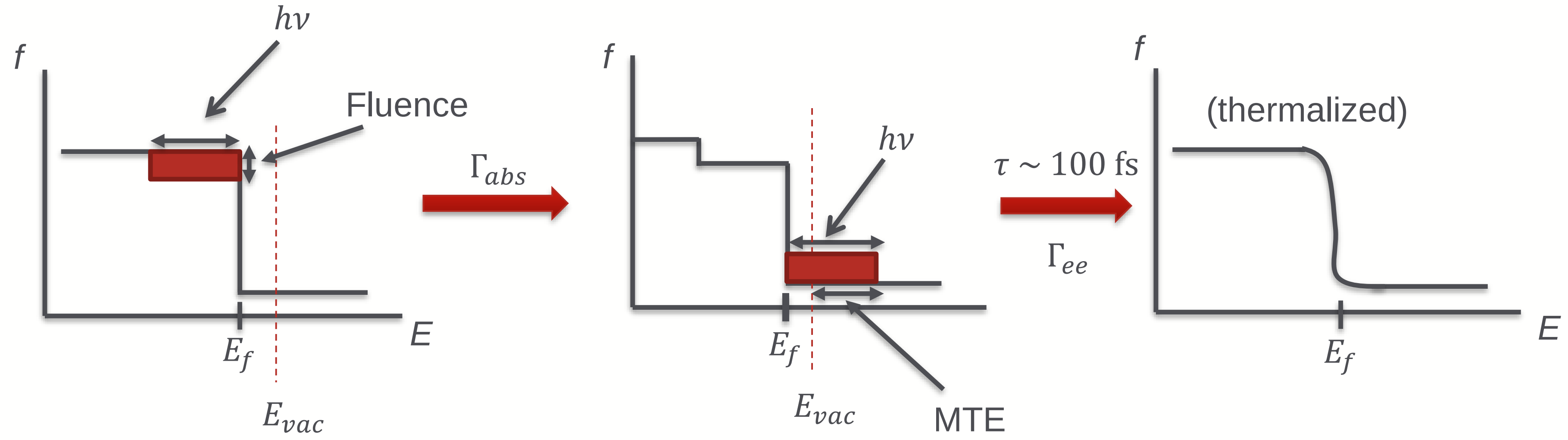
Result (Dynamic / high fluence regime)



Two-photon emission

- No qualitative difference is observed between the low fluence result and high fluence results at high photon energies since single photon absorption dominates at this photon energy
- As the photon energy approaches the threshold, two-photon absorption starts to become significant. Thus, both QE and MTE deviates from the low fluence result

Boltzmann Equation diagram (2)



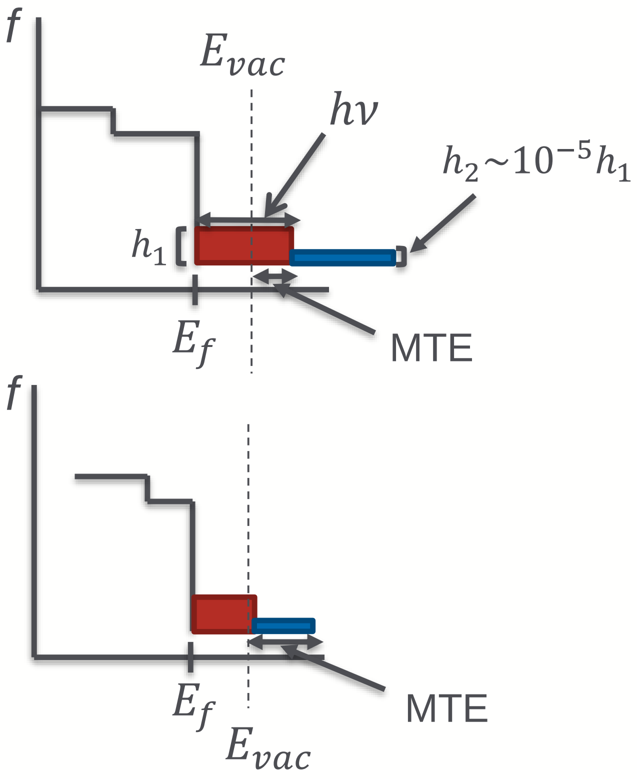
- For a low fluence, the photon energy decides MTE

Non-monotonic behavior of MTE

High photon energy
$(h\nu > \phi)$

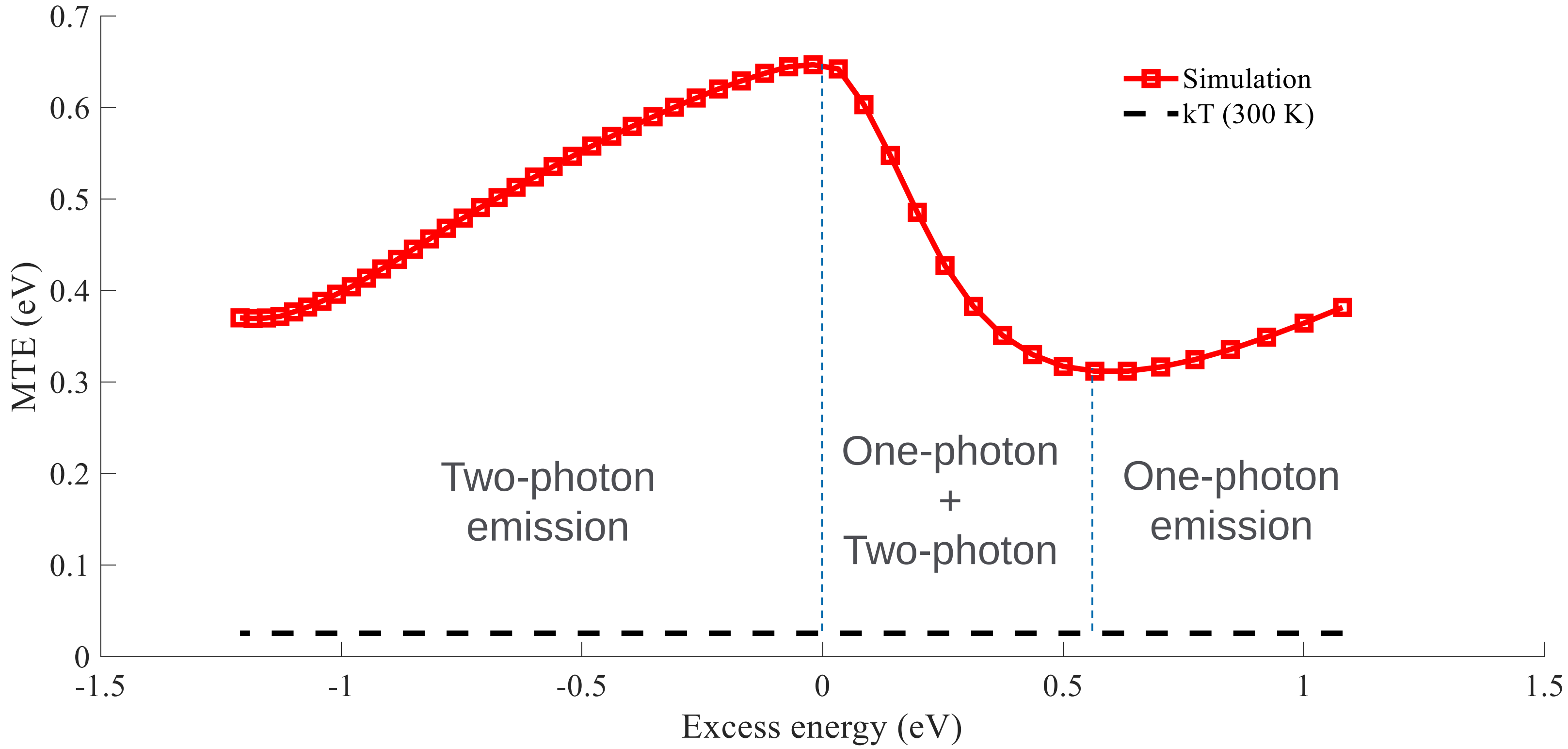
vs

Low photon energy
$(h\nu \sim \phi)$

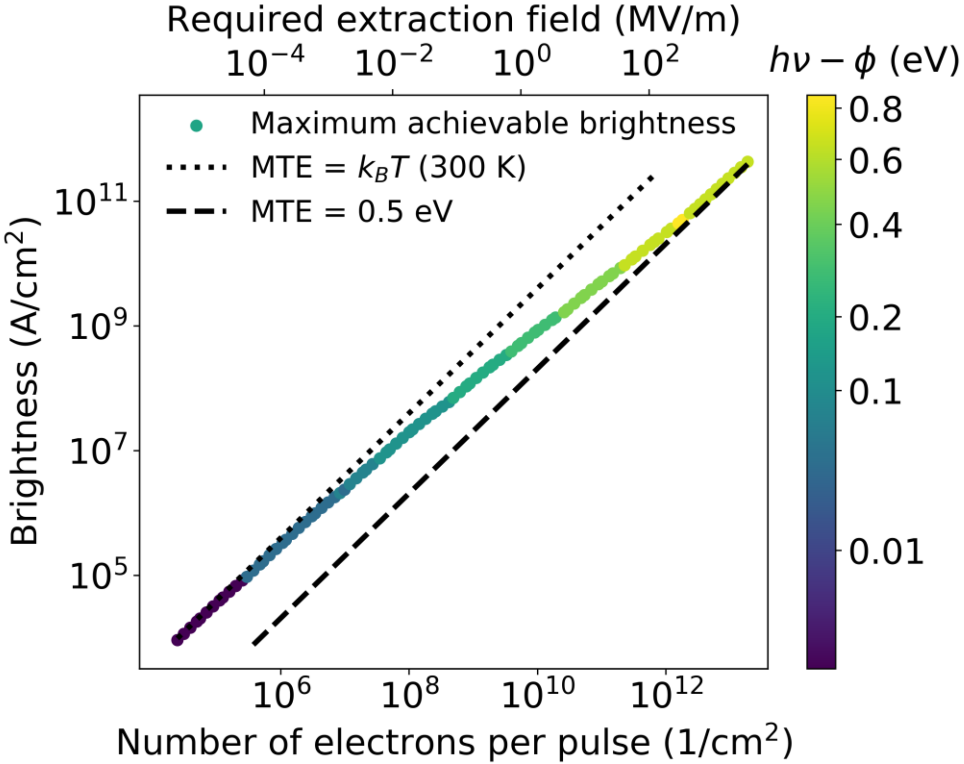


- For a high fluence, the blue box (two-photon absorbed electrons) becomes non-negligible
- Two-photon interaction causes non-monotonic behavior of MTE

MTE below threshold



Brightness



- Optimal photon energy for maximum brightness is no longer near the threshold (color scale)
- For the number of electrons that requires 100 MV/m extraction field, the brightness can decrease by a factor of 12

Conclusion

- Boltzmann equation allows dynamic simulation of photoemission
- Due to two-photon interaction, copper has very high MTE near threshold for high fluence ultrafast laser
- Brightness can be decreased by a order of magnitude from the thermal limit for femtosecond pulses
- Experimental effort is required and other materials (semiconductors) need to be studied

Acknowledgment

- Jared Maxson, Ivan Bazarov, Luca Cultrera, Pietro Musumeci, Siddharth Karkare, Howard Padmore

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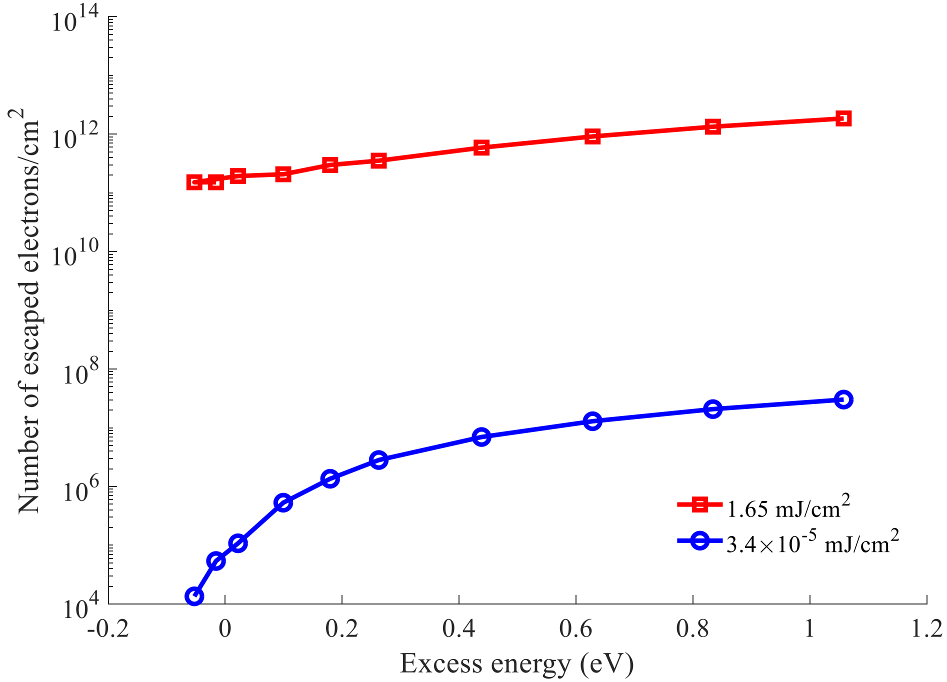
Assumptions

- Thin film approximation
- Diagonal density matrix : no interference
- Limited photon absorption mechanism (Inverse Bremsstrahlung)
- Isotropy : *f(k)* → *f(E)*
- Short laser duration (~50 fs). $\Gamma_{el-ph} \approx 0$
- One band model: $k(E) = \left(3\pi^2 \int D(E')dE'\right)^{1/3}$

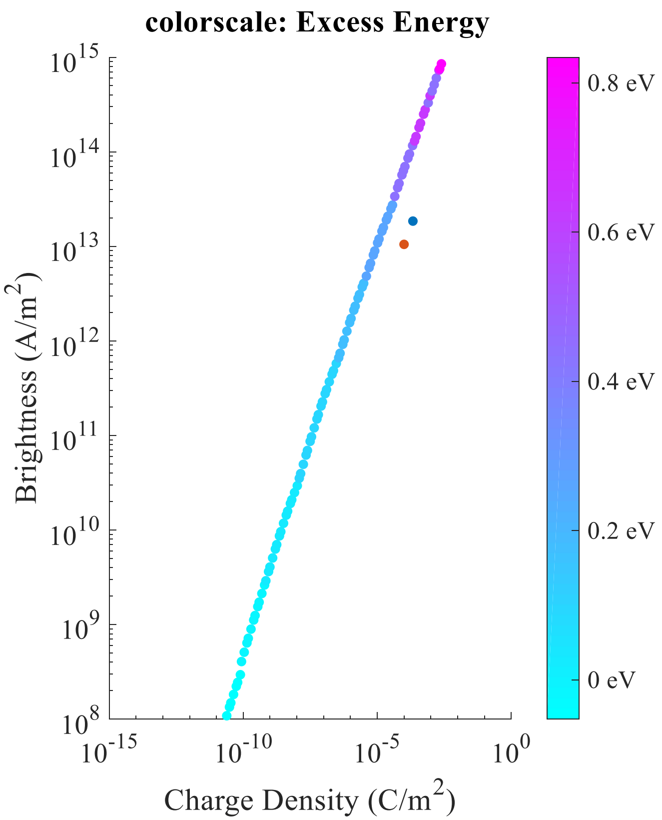
$$\frac{df(E)}{dt} = \Gamma_{ee}(E) + \Gamma_{abs}(I_{laser}, h\nu, E)$$

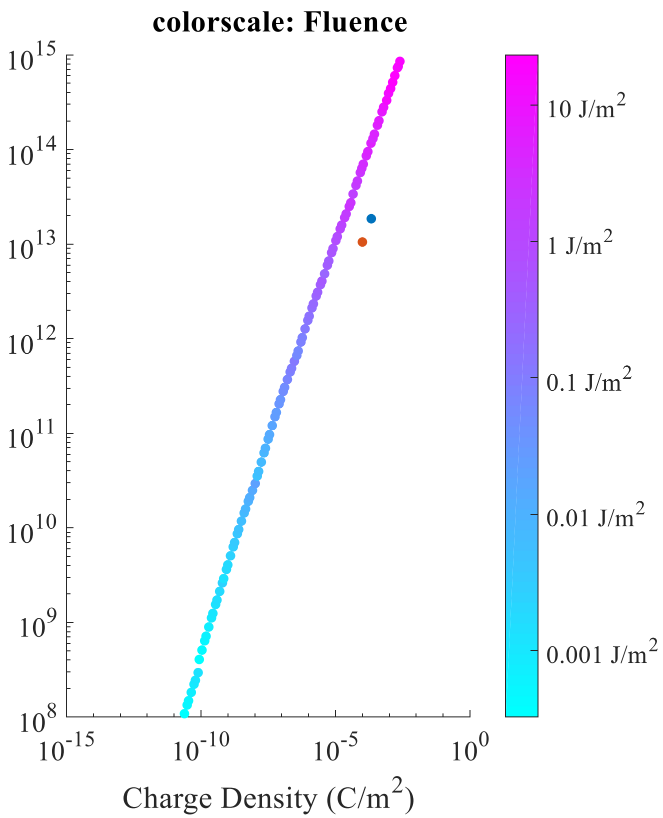
Escaped electrons



- At low photon energy, the number of escaped electrons does not scale with fluence due to multiphoton photoemission

Brightness


colorscale: Excess Energy
Brightness (A/m^2)
10^{15}
10^{14}
10^{13}
10^{12}
10^{11}
10^{10}
10^{9}
10^{8}
10^{-15}
10^{-10}
10^{-5}
10^{0}
Charge Density (C/m^2)
0.8 eV
0.6 eV
0.4 eV
0.2 eV
0 eV


colorscale: Fluence
10^{15}
10^{14}
10^{13}
10^{12}
10^{11}
10^{10}
10^{9}
10^{8}
10^{-15}
10^{-10}
10^{-5}
10^{0}
Charge Density (C/m^2)
10 J/m^2
1 J/m^2
0.1 J/m^2
0.01 J/m^2
0.001 J/m^2

Comparison with experiment (1)

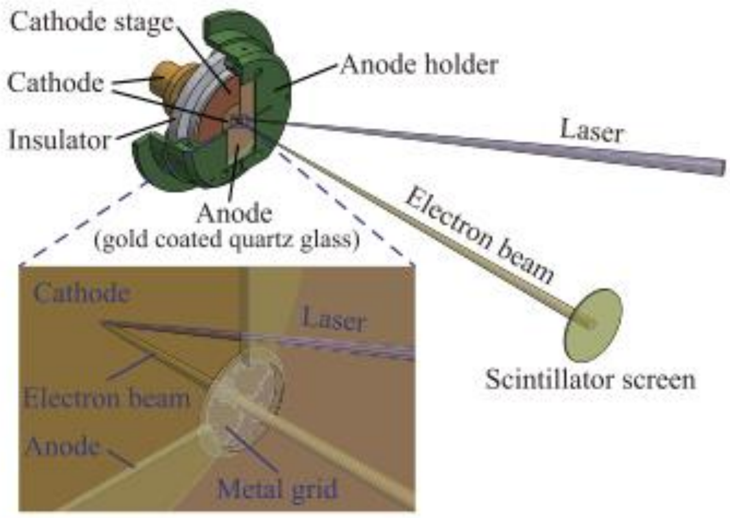
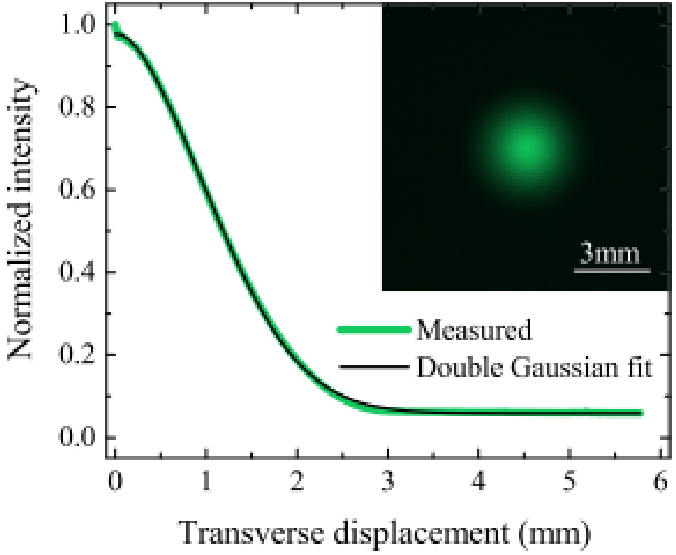
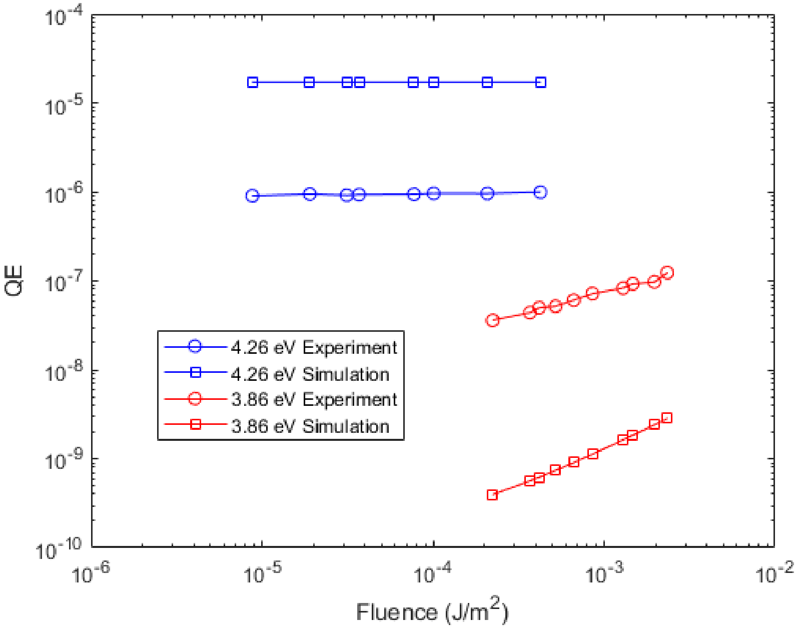


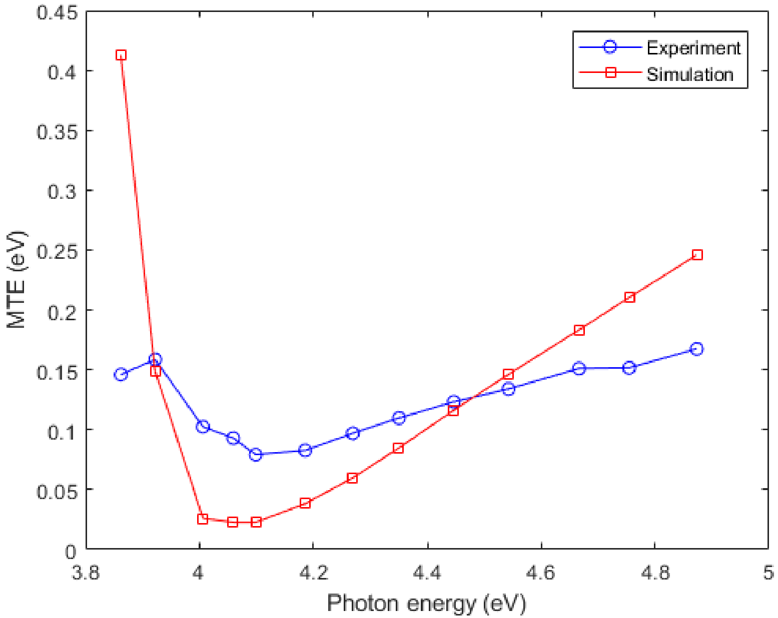
FIG. 1. Mechanical model of measuring apparatus.



$$J = \sigma_1 I + \sigma_2 I^2,$$

Comparison with experiment (2)





Photoemission Calculation

- QE
  - $j(t) = \rho\vec{v} \quad \propto \quad \int_{E_f+\phi}^{\infty} dE \int_{\cos\theta_{\max}}^{1} d(cos\theta)\,[\overset{\rho/e}{\overset{\downarrow}{\boxed{f(E,t)D(E)}}}\ \overset{\vec{v}}{\overset{\downarrow}{\boxed{v(E)cos\theta}}}]$
  - Number of photons $\propto\ < \Delta E >= \int_{E_f+\phi}^{\infty} dE[E(f(E,t) - f_{initial})D(E)]$
- $< p_x^2 > \ \propto \ \int_{E_f+\phi}^{\infty} dE \int_{\cos\theta_{\max}}^{1} d(cos\theta)\ [\boxed{p_x{}^2}\ \underset{j(E,t)}{\underset{\uparrow}{\boxed{f(E,t)\mathrm{D(E)}v(E)cos\theta}}}]$

Non-equilibrium Occupation 3

- We closely follow Mueller and Rethfeld (2013) paper which utilize Boltzmann equation for electron thermalization with high laser illumination.

- Nickel DOS
- Dashed- Final occupation
- Solid – Transient occupation