Modeling quantum yield, emittance, and surface roughness effects from metallic photocathodes

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Motivation

- Developments in materials design and synthesis have resulted in photocathodes that can have a high quantum yield (QY), operate at visible wavelengths, and are robust enough to operate in high electric field gradient photoguns for application to free electron lasers, in dynamic electron microscopy and diffraction.

- However, synthesis often results in roughness, both chemical and physical, ranging from the nano to the microscale. The effect of roughness in a high gradient accelerator is to produce a small transverse accelerating gradient, which therefore results in emittance growth.

- Although analytical formulations of the effects of roughness have been developed, detailed theoretical modeling and simulations that are verified against experimental data are lacking.

- We aim to develop realistic, verified and validated, electron emission modeling and 3D simulations from photocathodes with controlled surface roughness to enable an efficient way to explore parameter regimes of relevant experiments.
Momentatron experiments allow investigation of emission properties and surface roughness effects.

- Recent advances in material science methods have been demonstrated (H. A. Padmore. Measurement of the transverse momentum of electrons from a photocathode as a function of photon energy, in P3 2014) to control the growth of photoemissive materials (e.g. Sb) on a substrate to create different types of rough layers with a variable thickness of the order of 10 nm.

- Momentatron experiments have been developed (J. Feng et al., Rev. Sc. Instr., 86, 015103-1/5, 2015) to measure transverse electron momentum and emittance.

- It was demonstrated (J. Feng et al., Appl. Phys. Lett., 107, 134101-1/4 2015) recently how data from momentatron experiments can be used to investigate the thermal limit of intrinsic emittance of metal photocathodes.
Overview of our modeling approach

The overall modeling capabilities implemented in the Vorpal Particle-in-Cell (PIC) code framework to simulate electron emission from photocathodes with controlled rough surfaces consist of

- electron excitation in a photoemissive material in response to absorption of photons with a given wavelength
- charge dynamics due to drift and various types of scattering processes
- representation of rough interfaces
- calculation of electron emission probabilities that takes into account image charge and local field enhancement effects (as a function of position on rough surfaces).
- particle reflection/emission updates and efficient 3D electrostatic (ES) solver for a simulation domain that has sub-domains with different dielectric properties separated by rough interfaces.
We have developed simulations with different types of surface roughness.

Figure 1: The surfaces are represented with stair step or cut-cell grid boundaries.
The simulations effectively implement the 3 step model for electron emission from metallic photocathodes.

Figure 2: A simplified diagram indicates the three main processes to model, the importance of electron-electron scattering and temperature effects.

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Electron energy sampling takes into account the density of states of photocathode materials.

Figure 3: Electron energies on occupied states are sampled from a probability distribution with density function $p(\mathcal{E}) \sim g(\mathcal{E})f(\mathcal{E})$, where $g(\mathcal{E})$ is the density of states (DoS), and $f(\mathcal{E}) = 1/(e^{(\mathcal{E} - \mu)/k_B T} + 1)$ is the Fermi function. The DoS used is from published band structure calculations: Sb (left) is from Bullet (1975) and Au (right) is from Christensen & Seraphin (1971).
We implemented two models for electron-electron scattering for Monte Carlo particle simulations.

- For transport in Sb, we used the unified model proposed by Ziaja et al. J. Appl. Phys., 99 033514 (2006) giving mean free path (in Å):
  \[ \lambda_{ee}(\mathcal{E}) = \frac{\sqrt{\mathcal{E}}}{a(\mathcal{E} - \mathcal{E}_{th})^b} + \frac{\mathcal{E} - \mathcal{E}_0 \exp\left(-B/A\right)}{A \ln(\mathcal{E}/\mathcal{E}_0) + B}, \]
  with \( \mathcal{E}_{th} \) a threshold energy for the scattering (\( \mathcal{E}_{th} = 0 \) for metals and \( \mathcal{E}_{th} = E_G \) for semiconductors), \( \mathcal{E}_0 = 1 \text{ eV} \), and \( a, b, A, \) and \( B \) are fitting constants.

- For transport in Au, we implemented a model proposed by K. Jensen et al. J. Appl. Phys., 102 074902 (2007) with a temperature-dependent scattering rate (with only one adjustable parameter, \( K_s \)):
  \[ 1/\Gamma_{ee}[\mathcal{E}(k)] = \frac{8\hbar K_s^2}{\alpha_{fs}^2 \pi m c^2} \left( \frac{\mathcal{E}(k)}{k_B T} \right)^2 \left( \left( 1 + \left( \frac{\mathcal{E}(k) - \mu}{\pi k_B T} \right)^2 \right)^{\gamma} \left( \frac{2k}{q_0} \right) \right)^{-1}. \]
Electron-electron (el-el) scattering is the dominant energy relaxation mechanism in metallic photocathodes.

Figure 4: The el-el scattering rates for Sb and Au used in the simulations are shown in the left plot. The electron-acoustic phonon rates for Au are about an order of magnitude smaller than the el-el rates for the range of photon energies of interest to electron emission.
Electron emission probabilities are calculated using a transfer matrix approach.

The surface potential energy is

\[ V(x) = \mu + \phi - Fx - \frac{Q}{x}, \]

where \( \phi \) is the work function, \( Q = Q_0 \left( K_s - 1 \right) / \left( K_s + 1 \right) \), with \( K_s \) the static dielectric constant of the emitter, and \( Q_0 = q^2 / \left( 16\pi\varepsilon_0 \right) \) with \( q \) the fundamental charge and \( \varepsilon_0 \) the permittivity of vacuum. The Schottky effect is taken into account by using the electrostatic potential in the simulations to evaluate the electric field \( F \) along local normal directions. This is included in the calculation of emission probabilities at positions where electrons attempt to cross the photocathode surface.
We compare simulation results on electron emission from flat and 3-ridge rough emission surfaces of Sb and Au.

- For simulations with uniform work function on the surface, we used $\phi_{\text{Sb}} = 4.5 \text{ eV}$ and $\phi_{\text{Au}} = 4.9 \text{ eV}$ for Sb and Au, respectively.
- A constant potential difference is maintained across the $x$ length of the simulation domain leading to an applied field magnitude in the vacuum region of the order of 1 MV/m (it varies on the rough emission surface).
- We use periodic boundary conditions in the transverse directions.
- Typical parameters for both 3-ridge and flat emission surface simulations: $0.4268 \times 1.182 \times 0.394 \text{ (in } \mu\text{m})$ domain size, with $88 \times 264 \times 16$ number of cells; time step: $\Delta t = 2.5 \times 10^{-16} \text{ s}$.
- Details of the models for photo-excitation, transport, and emission, together with material parameters used, are given in: 
Electron dynamics inside and out of an antimony photocathode at selected simulation times.

Figure 5: Photo-excited electrons (red spheres) have effectively diffusive dynamics in the photocathode. Vacuum electrons (green spheres) move ballistically.
The implemented models for Sb are validated against experiments on quantum yield.

Figure 6: Simulations with the higher electron-electron scattering rate show agreement with experimental data (J. Feng, D. Voronov, and H. A. Padmore, unpublished) on quantum yield from antimony with a flat emission surface.
Simulations with controlled surface roughness show increased quantum yield. 

Figure 7: The increased QY is likely due to a geometric effect: electrons excited on the sides of the ridges are effectively closer to the emission surface than electrons excited in Sb with a flat vacuum interface.
Modeling intrinsic emittance: 3-step model with constant DoS, \( T = 0 \) K

- If there is no correlation between transverse position and momentum distributions of emitted electrons, the intrinsic emittance \( \epsilon_y \) per mm of rms laser spot size \( \sigma_y \) is (Dowell & Schmerge, *PRSTAB*, 12, 073401, 2009):

\[
\frac{\epsilon_y}{\sigma_y} = \sqrt{\left< p_y^2 \right> / m_e c},
\]

where \( p_y \) is a transverse momentum component of an emitted electron.

- A 3-step model with a constant DoS and \( T = 0 \) K leads to

\[
\left< p_y^2 \right> / m_e = (\hbar \omega - \phi) / 3,
\]

and predicts zero intrinsic emittance for \( \hbar \omega = \phi \).
When temperature effects and the DoS of the photocathode material are taken into account, the intrinsic emittance can be obtained by numerically calculating the mean transverse energy (J. Feng et al., Appl. Phys. Lett., 107, 134101-1/4 2015) from:

$$\frac{\langle p_y^2 \rangle}{m_e} = \frac{\int_{\mu+\phi-\hbar\omega}^{\infty} g(E)f(E)(E + \hbar\omega)h(E, \phi, \hbar\omega) dE}{\int_{\mu+\phi-\hbar\omega}^{\infty} g(E)f(E) \left(1 - \sqrt{\frac{\mu+\phi}{E+\hbar\omega}}\right) dE}, \tag{2}$$

where $h(E, \phi, \hbar\omega) = \frac{2}{3} - \sqrt{\frac{\mu+\phi}{E+\hbar\omega}} + \frac{1}{3} \left(\frac{\mu+\phi}{E+\hbar\omega}\right)^{\frac{3}{2}}$. 
Intrinsic emittance from simulations is in agreement with experimental data on emission from flat Sb surfaces.

Figure 8: The antimony DoS and temperature effects had to be included in the implemented model to obtain agreement with experimental data (J. Feng et al., Appl. Phys. Lett., 107, 134101-1/4 2015). Simulations with the higher electron-electron scattering rate in Sb are in agreement with both the QY and intrinsic emittance data.
Surface roughness effects on intrinsic emittance from antimony.

Figure 9: Intrinsic emittance increases due to emission from the sides of the ridges and to presence of transverse electric field components near the rough emission surface.
3 step model for quantum yield

We have started to compare simulation results on QY from Au with the 3 step model for metallic photocathodes (Dowell et al., PRSTAB, 9, 063502, 2006; Dowell & Schmerge, PRSTAB, 12, 073401, 2009):

\[
QY(\omega) = \frac{\int_{\mu+\phi-\hbar\omega}^{\infty} dE \, p(E, \omega) \int_{\cos \theta_{\text{max}}}^{1} d(\cos \theta) \, F_{e-e}(E, \omega, \theta)}{2 \int_{\mu-\hbar\omega}^{\infty} dE \, p(E, \omega)},
\]

(3)

with probability density to excite an electron from \( E \) to \( E + \hbar \omega \):

\[
p(E, \omega) \propto g(E + \hbar \omega) (1 - f(E + \hbar \omega)) g(E) f(E),
\]

cosine of maximum incident angle allowing conservation of transverse momentum: \( \cos \theta_{\text{max}} = \sqrt{(\mu + \phi)/(E + \hbar \omega)} \) and probability that an excited electron reaches the emission surface without el-el scattering:

\[
F_{e-e}(E, \omega, \theta) = \frac{\lambda_{ee}(E + \hbar \omega) \cos \theta}{\lambda_{opt}(\omega) + \lambda_{ee}(E + \hbar \omega) \cos \theta}.
\]
3 step model for quantum yield: constant DoS, $T = 0 \text{ K}$

- Analytical formula for the QY can be obtained under the additional approximations that $\cos \theta \approx 1$ when $\theta_{\text{max}}$ is small and the electron-electron mean free path $\lambda_{ee}(E + \hbar \omega)$ is slow varying over the range of excited electron energies considered in some experiments:

$$QY_0(\omega) = \frac{F_{ee}(\omega)}{2\hbar \omega} \left(1 - \sqrt{\frac{\mu + \phi}{\mu + \hbar \omega}}\right)^2 (\mu + \hbar \omega), \quad (4)$$

where $F_{ee}(\omega) = 1/ \left(1 + \lambda_{opt}(\omega)/\lambda_{ee}(\omega)\right)$,

$$\lambda_{ee}(\omega) = \frac{1}{\hbar \omega - \phi} \int_{\mu + \phi}^{\mu + \hbar \omega} \lambda_{ee}(E) \, dE,$$

and $\hbar \omega > \phi$.

- Under these approximations, $QY_0(\omega) \equiv 0$ for $\hbar \omega \leq \phi$. 
Quantum yield from Au is strongly affected by its DoS in the 3-step model.

Figure 10: Simulations using the Au DoS from relativistic band calculations leads to markedly lower QY compared to the 3-step model with constant DoS and \( T = 0 \) K. Including the DoS and temperature effects in the 3-step model improves the agreement. However, a model using only direct optical transitions for photon absorption might be sufficient to explain QY from gold (compare Christensen & Seraphin, PRB, 4 3321 (1971) to Krolikowski & Spicer, PRB, 1 478 (1970)).
The DoS of Au indicates that a significant number of photoexcited electrons will not contribute to emission.

Figure 11: In the 3 step model, only electron energies in the narrow region near the Fermi level contribute to emission. Small changes in the DoS in this region will lead to large changes in quantum yield.
Physical and chemical surface roughness effects on quantum yield from Au

Figure 12: Surface roughness and variable work function effects could lead to a crossover in the QY relative to emission from a flat surface.
Summary

- Simulation results on quantum yield and intrinsic emittance are in agreement with experimental data on emission from Sb photocathodes with flat emission surfaces.
- Agreement with experimental data was obtained only after including the Sb DoS and temperature effects in the modeling.
- Results on QY from gold also show strong dependence on its DoS.
- The relative MTE growth due to controlled surface roughness is largest near emission threshold and at the emission surface (around 50% in the Sb simulations).
- Simulation results on QY vs wavelength from Sb and Au are higher from the controlled rough surfaces vs flat ones when using uniform work functions and light absorption. This is due to photo-exited electrons effectively closer to the rough emission surface.
- Future work will focus on using additional data from band structure calculations, modeling of nonuniform light absorption on rough surfaces, and electron heating.
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