Enhancement of Emission from Metal Cathodes

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Outline

- Some observations about metallic photocathodes for RF guns
  - does the ‘standard model’ of photocathode emission reflect reality?

- Angular dependence of photoemission at very low energy
  - can we improve emission using off-normal incident light?

- Controlling light absorption by creation of plasmons
  - can we control light absorption to create higher QE?

- Guiding light using plasmons and field localization
  - can we guide light in nanostructures to create local intense emission
LCLS Cu Photocathode

- Cu photocathode
- polycrystalline surface
- UV and H ion cleaned
- photon energy ~ 0.5 eV in excess of workfunction
Real cathodes are not random!

- real metals are usually highly textured with well defined grains
- grain structure exists from the macro to nano domains
- each grain has a well defined electronic structure, work function, optical properties….

- \( <111> W = 4.98 \text{ eV}, <100> = 4.5 \text{ eV} \)

- assuming 50 MV/m, 257 nm,
  excess energy \(<111> = 0.1 \text{ eV} \)
  excess energy \(<100>= 0.6 \text{ eV} \)

- 257 nm light leads to factor 36 difference in yield between grains
- emission will be structured at the 0.1 mm scale near the cathode
- emittance should be calculated with a Fowler weighting of emission taking into account the grain fractions.
Real cathodes: a closer look

Many grains themselves are highly textured.

Grain boundaries are sharp at the resolution of 20 nm.

PEEM was able to identify the field enhancement at the diamond turning marks on the substrate (around 20 nm x ~5 micron).

Initial images showed extreme s-p contrast.

S-p contrast vanished after light ion etching (keV).

266 nm, 5 mW CW, 1mm² beam; very rapid laser cleaning.

Annealed polycrystalline cathodes have large grains, together with regions of very small (sub-100 nm) grains. Probably as a result of high temperature annealing and grain growth.

Expected that near cathode emission will be highly filamented.
**Photocathode Quantum Efficiency**

**Step 1**
Absorption of light and excitation of electron
- energy conservation
- conservation of momentum not used

**Step 2**
Probability of reaching surface w/o scattering
- uses calculated or measured mean free path
- assumes single scattering leads to loss

**Step 3**
Emission from surface
- perp. component of energy > work function
- for flat bands leads to Fowler law

- we need a certain excess energy for a defined emittance
  - on an excess energy scale, most metals give same QE
  - Al is factor of 10 lower, as it's a highly reflective FE metal
- at photon energies typical of a photo-injector, no direct optical transitions possible

- transitions possible by inelastic processes + surface states

- is the emittance of each excitation channel the same?
Some observations about metallic photocathodes for RF guns

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Angular dependence of photoemission at very low energy (ARPES)

- can we improve emission using off-normal incident light?

Controlling light absorption by creation of plasmons

- can we control light absorption to create higher QE?

Guiding light using plasmons and field localization

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Characterizing Photocathodes

Techniques

- ultra-low energy angle resolved electron spectroscopy
  - kinetic energies 0 – 1eV
- angle resolved electron yield

- full measurement of momentum distribution and yield as function of:
  - polarization
  - photon energy (2 – 6 eV)
  - photon incidence angle
  - surface preparation
Photocathodes Lab

- 5 axis manipulator; motor scanned theta, LN2 – 1100 K
- Low Energy Electron Diffraction + Auger
- Time of Flight Electron Analyzer
- VIS UV mono
- Tunable 2nd, 3rd, 4th harmonic generation + pulse picker
- Sample transfer
- 80 MHz Ti:S Oscillator
Large off-axis enhancement of photoemission yield (VPE)

- Cu(111) supports a surface state
- 3 step model does not account for change in yield (red)
- model fit simply says that z field is more important than in-plane field

\[ J \propto \left( |E_x|^2 + |E_y|^2 + B|E_z|^2 \right) \]


Original work on this system Pedersoli et al APL 93, 183505, 2008
New work, Greaves et al in preparation
Photoemission from Cu(100)

- No off-axis enhancement of photoemission yield (VPE)
- Cu(100) does not support a surface state
- 3 step model accounts for change in yield (red)

- effect seems to be related to surface states
- check: Mo(100) has a SS and (111) does not, i.e. reverse of Cu
- VPE of 50 reported on ‘polycrystalline’ Mo by JWJ
  - probably not poly by single xtal caused by high temp anneal (as in LCLS Cu cathode)
Emittance: Theoretical Estimate

For Cu(111), $k_{\text{max}} = 0.225 \ \AA^{-1}$, $R = 1 \ \text{mm}$

$$\varepsilon_{x,rms} = 2.2 \times 10^{-7} \ \text{m rad}$$

For Ag(111), $k_{\text{max}} = 0.125 \ \AA^{-1}$, $R = 1 \ \text{mm}$

$$\varepsilon_{x,rms} = 1.2 \times 10^{-7} \ \text{m rad}$$

- surface states limit the max transverse momentum
- ideally want a system of independent 2d planes
  - TaSe$_2$, MoS$_2$, graphite(Li).....
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Surface Plasmons

2-dimensional plasmon oscillation

\[ k_{x,sp} = \frac{2\pi}{\lambda_{x,sp}} \]

In reality complex dielectric function limits max. k. Example Aluminum

\[ k_x = \frac{\omega}{c} \left( \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \right)^{\frac{1}{2}} \]

limit of large \( k_x \) (short \( \lambda_{sp} \)):
\[ \varepsilon_1 = -\varepsilon_2 \]

\[ \omega_{sp}^2 = \frac{\omega_p^2}{1 + \varepsilon_2} \]
Enhancing photoemission: use of surface plasmons

Optical Reflectivity

\[ \lambda_0 = 260 \text{ nm (4.77 eV)} \]
\[ \phi = 4.27 \text{ eV} \]
Al thickness: 19.9 nm

Model For 12.59 nm Al Thickness and 5 nm Oxide Layer

Wavelength = 394 nm
Substrate Transmittance = 0.96

Matching condition for wavelength and wave vector for generation of surface plasmons
Enhancing photoemission: use of surface plasmons

Total electron yield from Al, p-polarization, as a function of angle of incidence for 4 photon energies.

Wavevector matching condition
- ~ 100 normal incidence yield
- this factor is larger than expected based just on modified absorption
- present experiments on optical measurements in Al graded films
- next measurements in UHV on electron yield of single xtal films (eg. Al on MgO)
Enhancing photoemission: use of surface plasmons

- Al (Mg)
  stable and thin (2 nm) surface passivation of oxide can be alloyed with 2% Li (Ba) to reduce the work function to 3 (2.5) eV

- Mo
  higher QE, easy to clean by flash heating also exhibits very high VPE effect (50)

Modification of 3 step model with plasmon fields

QE In SP-coupled System For Photon Energy 0.5 eV Above Workfunction
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Plasmon dispersion and coupling

\[ m \lambda / g = \sin \alpha + \sin \beta \]

g = grating spacing;  \( m \) = diffraction order; \( \lambda \) = wavelength
\( \alpha \) and \( \beta \) = angles of incidence and diffraction
Grating coupled plasmon on an Al surface

- complete absorption in the case of optimized groove depth
- highly angle and wavelength dependent
- 1d grating only
- electric vector must be perpendicular to grooves

Aluminum grating, normal inc.

period \( g = 800, 400, 200 \text{ nm} \)

groove depth \( h = 60, 20, 11 \text{ nm} \)

1:1 land – groove ratio

RCWA calculations
Why Metallic Surfaces with Grooves a Few Nanometers Deep and Wide May Strongly Absorb Visible Light

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It is theoretically shown that nanometric silver lamellar gratings present very strong visible light absorption inside the grooves, leading to electric field enhancement of several orders of magnitude. It is due to the excitation of quasistatic surface plasmon polaritons with particular small penetration depth in the metal. This may explain the abnormal optical absorption observed a long time ago on almost flat Ag films. Surface enhanced Raman scattering in rough metallic films could also be due to the excitation of such quasistatic plasmon polaritons in grain boundaries or notches of the films.

Silver grating, normal inc.

period \( g = 30 \text{ nm} \)

groove depth \( h = 5, 15, 30 \text{ nm} \)

groove width \( w = 5 \text{ nm} \)

- no grating orders possible
- period is too small
- doesn’t need to be periodic
- field enhancement in grooves
- can be made 3d (holes)
Al grooves tuned for UV photoemission

- Groove width and depth tuned to give zero reflectivity at ~ 0.5 eV above work function

- 1d field enhancement in the grooves ~ g/h = 10

- Min. reflectivity < 10^{-4} (< 10^{-7} in the optical regime)
Al grooves tuned for UV photoemission

- ‘groove plasmon’ dip at 4.5 eV is non-dispersive in angle
- dispersive surface plasmon peak at higher energy
Groove plasmon dispersion

- Efficient coupling to groove
- Strong damping in the groove
- Groove depth tunes Fabry Perot $\lambda_{\text{plasmon}}/4$ interference

Plasmon dispersion in groove structure

$$\frac{\varepsilon_1 \sqrt{k^2 - \varepsilon_m k_0^2}}{\varepsilon_m \sqrt{k^2 - \varepsilon_i k_0^2}} = \tanh \left( -\frac{w}{2} \sqrt{k^2 - \varepsilon_i k_0^2} \right)$$

Note scale

Qmax at 5eV ~ 2
- a ‘bad’ cavity
Groove plasmon dispersion

Plasmon dispersion in groove structure
- numerical solutions

\[
\frac{\varepsilon_1 \sqrt{k^2 - \varepsilon_m k_0^2}}{\varepsilon_m \sqrt{k^2 - \varepsilon_i k_0^2}} = \tanh \left( -\frac{w}{2} \sqrt{k^2 - \varepsilon_i k_0^2} \right)
\]

- Efficient coupling to groove
- Strong damping in the groove
- Groove depth tunes Fabry Perot $\lambda/4$ interference

$w = 32, 16, 8, 4, 2, 1$ nm
Field imaging using photoemission

- aberration limited resolution 5 nm at low energy
- can be used for pump probe fsec time resolved measurements
PEEM imaging of Al hole arrays

- strong localization of emission on holes
- strong polarization dependence of the emission
- polycrystallinity of Al sets surface roughness and FIB precision

50 nm holes on 248 nm pitch, Al
(FIB: Hyuck Choo)

p: 266 nm light
PEEM image
7:1 hole to background contrast

s: 266 nm light
PEEM image
1.5:1 hole to background contrast
Plasmon field can be used to accelerate electrons

3 cycle, TiS, 800 nm, 1.5 nJ

Emission by tunneling
Metals are interesting by $K_2SbCs$ is efficient!

- 0.5 – 2% QE at 532 nm
- use SR techniques to pin down structure and chemistry
  - powder diffraction
  - scanning XRF
  - micro EXAFS
- many question to address
  - relationship of structure / chemistry to QE
  - damage mechanisms
  - emittance
  -
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Some observations about metallic photocathodes for RF guns
- cathodes are in reality very complex microstructures

Angular dependence of photoemission at very low energy
- vectorial effects can be strong and potentially useful

Controlling light absorption by creation of plasmons
- absorption can be made complete in any free-electron-like material

Guiding light using plasmons and field localization
- fields can be guided and enhanced; may be useful for 2 photon PE
- can also be used for electron acceleration
  - keV energies can be reached with very short nJ excitation pulses