Plasmons and Field Enhancement

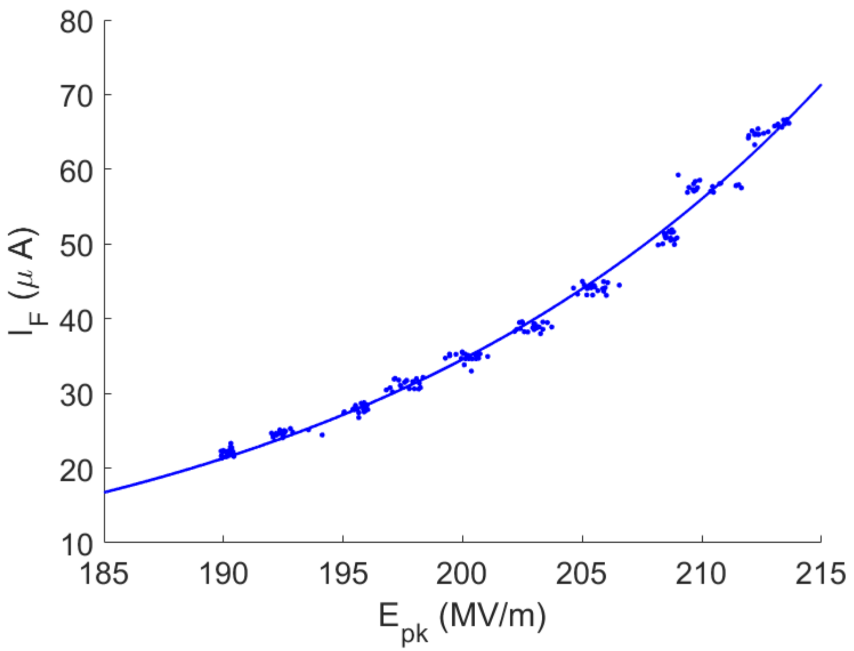
Jan Paszkiewicz, Walter Wuensch

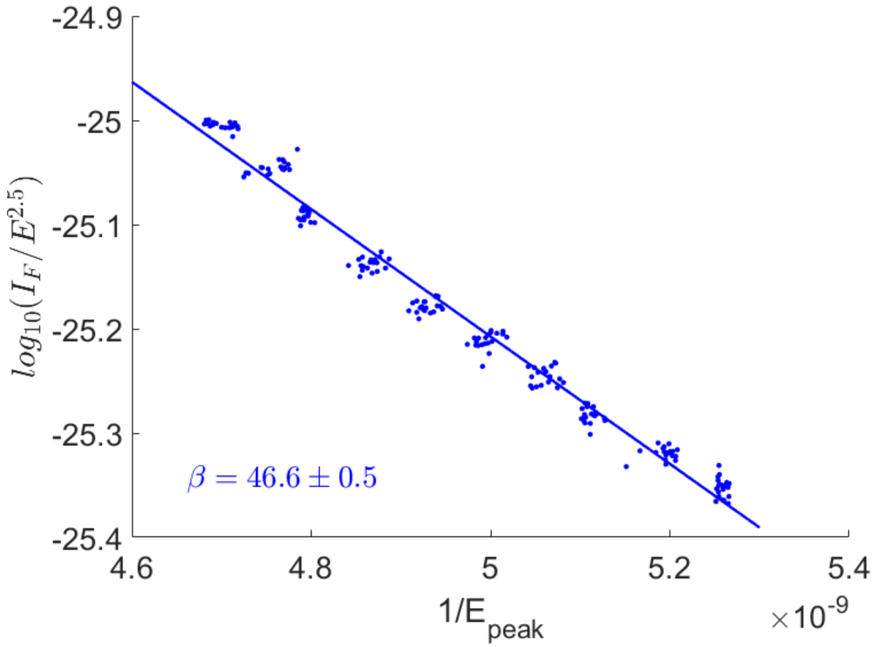
11/11/2020

Mini MeVAarc 2020

Motivation

- Many (all?) experiments with field emission involve fitting a field-enhancement factor β to experimental data to achieve consistency with the Fowler-Nordheim equation.
- Typical range for β factors between 20 and 300 based on the slope of Fowler-Nordheim plots.

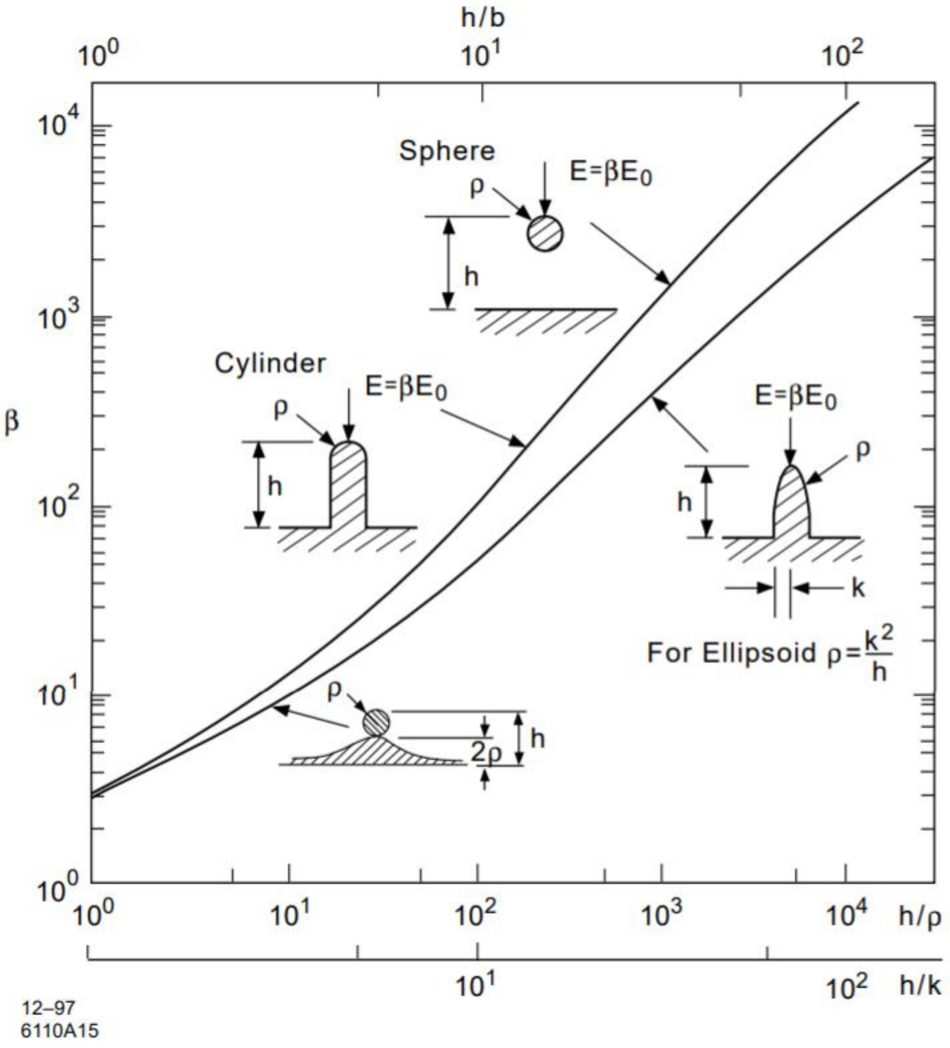




Motivation

- Field enhancement due to surface protrusions is normally used to explain this.
  - This would require extremely narrow structures, which have not been seen on Cu surfaces.
- The work function can also be lowered slightly due to impurities, crystal structure, etc…
  - But also not enough to explain the high β values that we see.

Field enhancement factor vs. aspect ratio of surface protrusions:

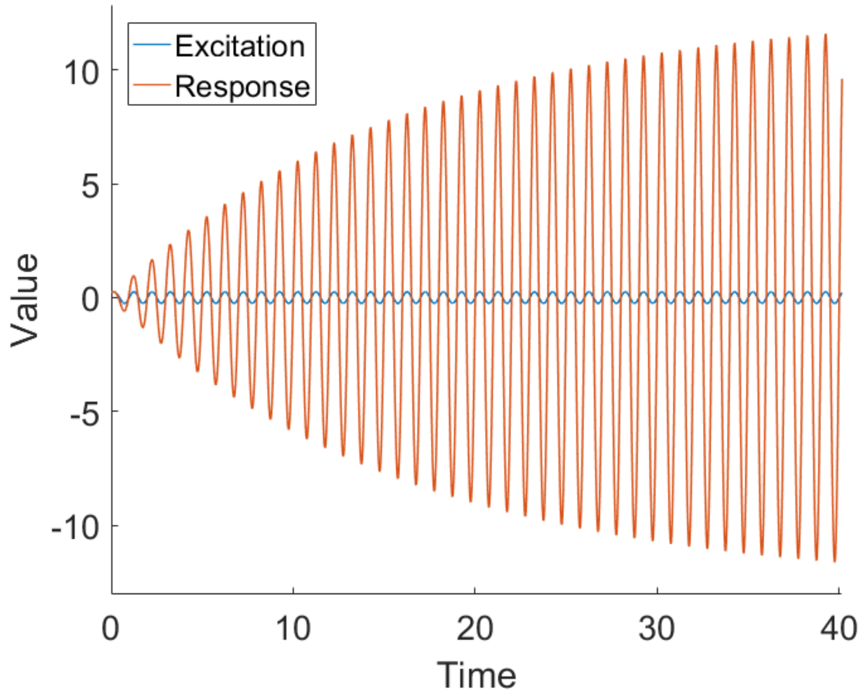


J. W. Wang & G. A. Leow, SLAC-PUB-7684

Motivation

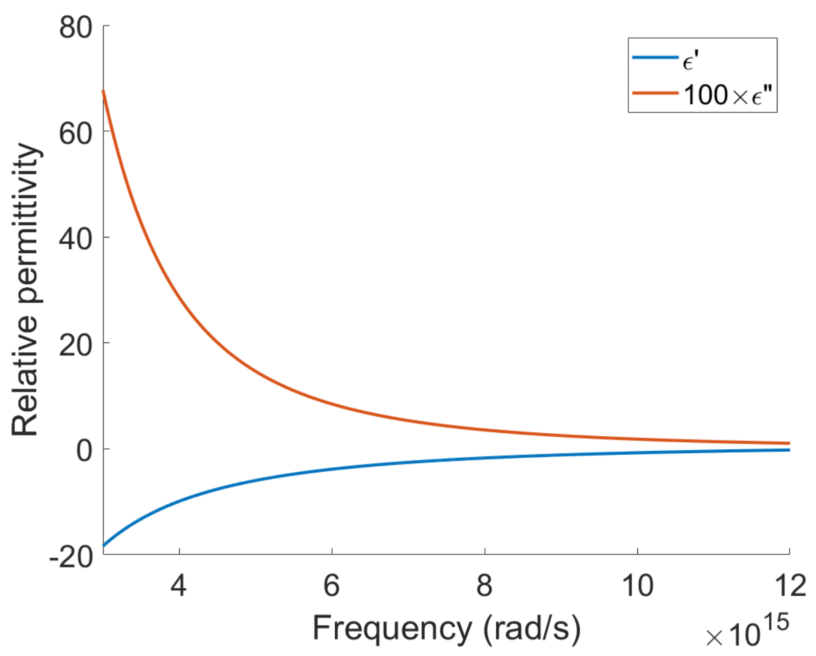
- One avenue to be explored are dynamic effects which result in resonances.
- Resonators are well-known for storing energy and amplifying the amplitude of a driving signal.
    - An initial spontaneous emission of an electron could cause the system to self-excite and produce very high surface fields.
- Surface plasmon-polariton waves are a candidate mechanism for setting up such resonances.

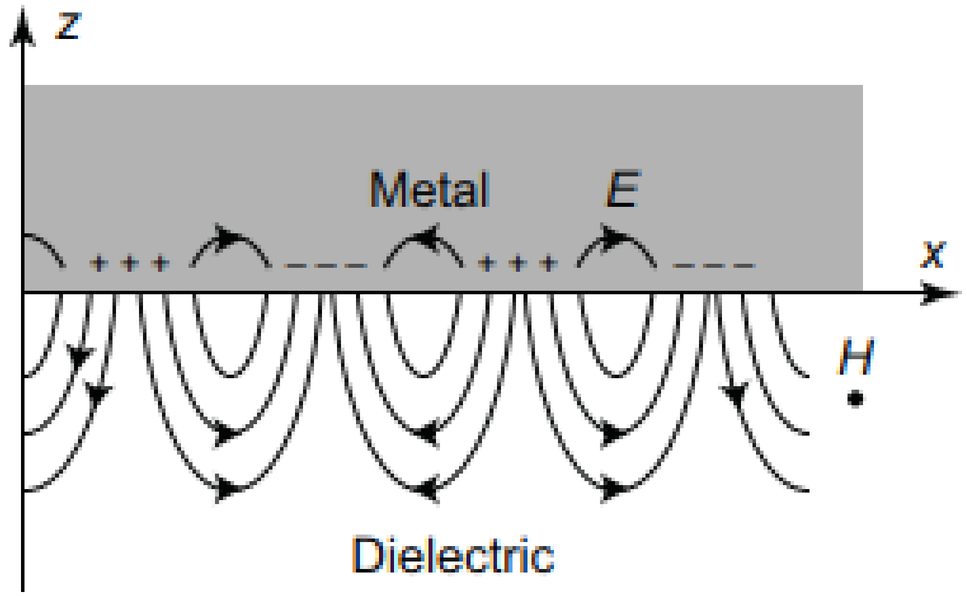
Response of a generic resonator to harmonic excitation



Surface Plasmon Polaritons

- Arise from the negative permittivity of metals at certain frequencies.
- In the Drude model, this is explained as the result of the inertia of the mobile electrons in the metal.
  - Oscillating electric field is accompanied by variations in electron density in the metal.

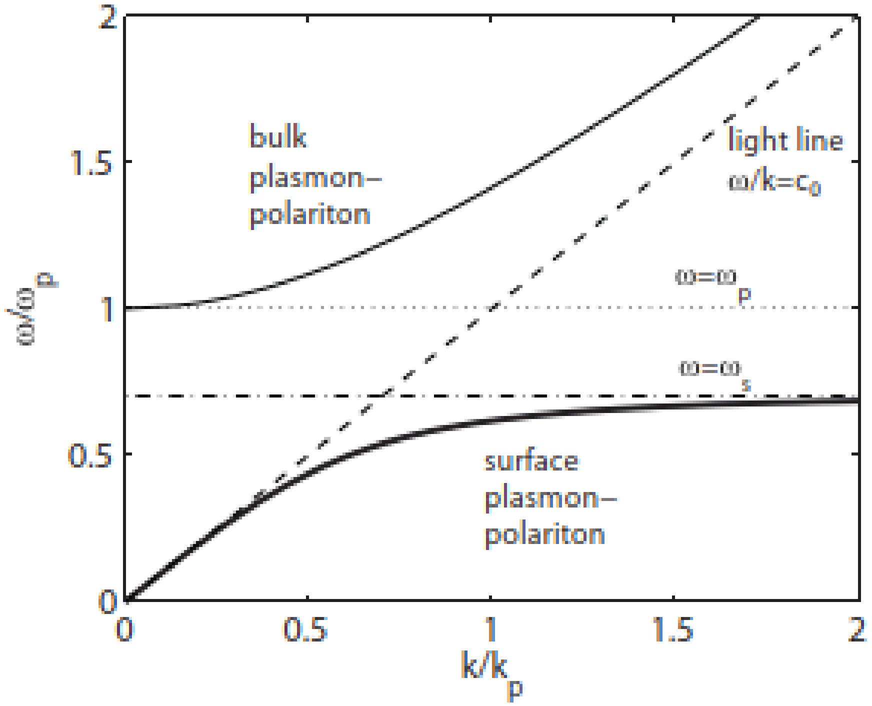




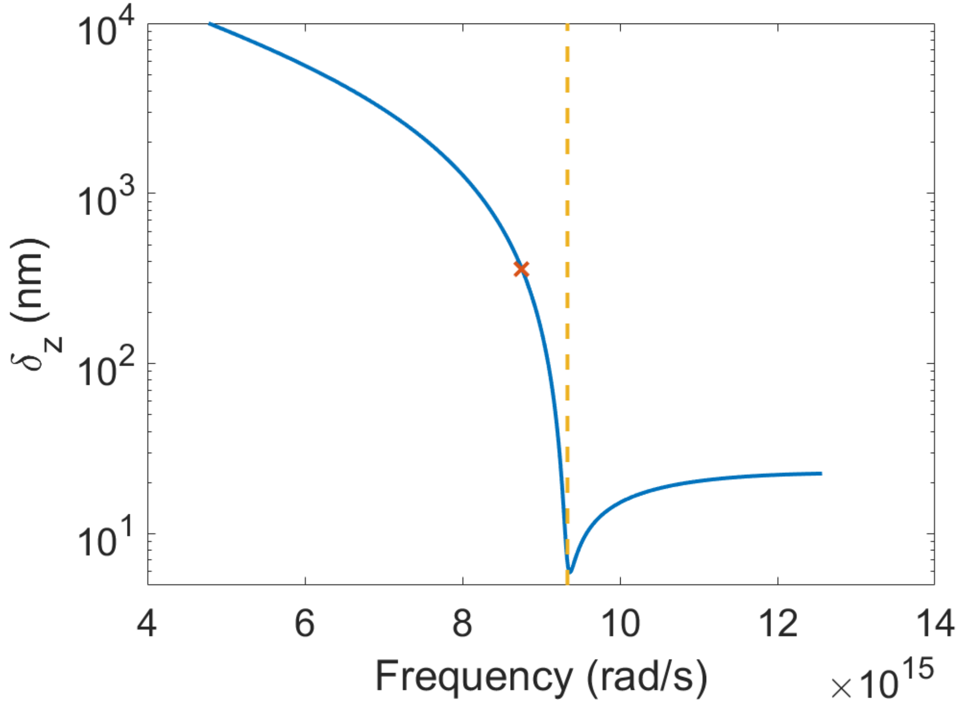
Surface Plasmon Polaritons

- Plasmons are very localised:
  - Surface mode, confined to the interface, extending max. << 1μm into the dielectric.
  - Wavelength much shorter than light for a given frequency.
  - Nano-scale resonators can be made, but short wavelengths do not propagate even without any special structure to confine them.

Dispersion curve

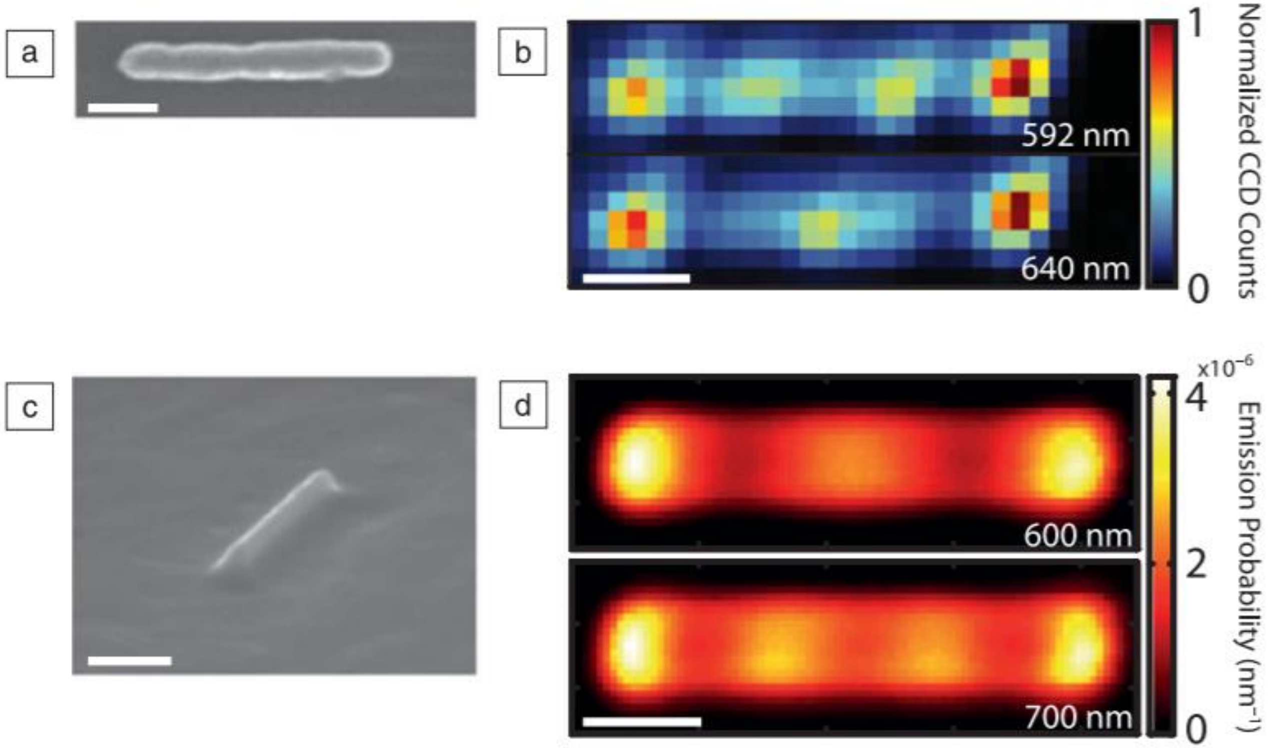


Decay length normal to interface



Surface Plasmon Polaritons

- Plasmons are very localised:
  - Surface mode, confined to the interface, extending max. << 1µm into the dielectric.
  - Wavelength much shorter than light for a given frequency.
  - Nano-scale resonators can be made, but short wavelengths do not propagate even without any special structure to confine them.



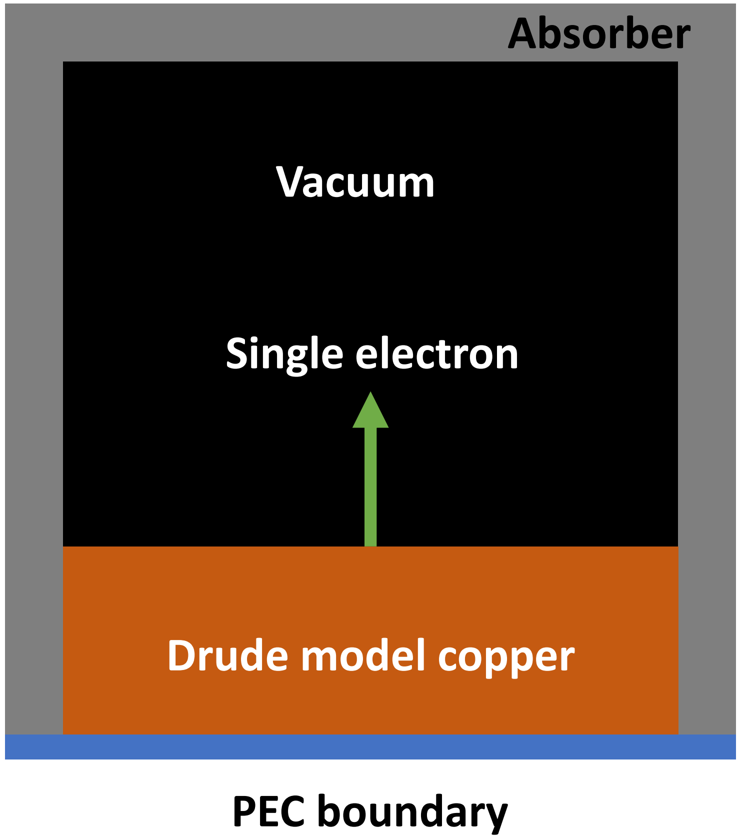
T. Coenen , E.J.R. Vesseur , A. Polman , ACS Nano 6 , 1742 (2012).

Investigation with PIC Simulations

- Gain insight on how plasmons may be excited by field emission, and what kind of fields we can expect.
- 2D PIC solver developed, allowing mutual interaction between electrons and fields.
- Copper represented by Drude model plasma:
    - $\boldsymbol{D}(t) = \varepsilon_0 \left(\boldsymbol{E}(t) + \frac{\omega_p^2}{f_c} \left(1 - e^{-f_c t} \right) * \boldsymbol{E}(t) \right)$
    - Material can be polarised and remembers past values of $\boldsymbol{E}(t)$.
    - Collision frequency f_c defines decay time.
- Variety of scenarios of electron impact and emission studied.

Simulation Setup

- 2D Finite-Difference Time Domain (FDTD) solution of Maxwell's equations
  - TM polarisation with field components Ex, Ez and By (B-field normal to screen).
  - Charge of particles spread over 20 nm in y direction.
  - 2 nm spatial resolution, 0.0025 fs time resolution
  - Sketch of simulation domain shown to the right.
  - 400 nm x 400 nm domain
  - 24 nm thick resistive absorber on three boundaries, PEC on bottom boundary.

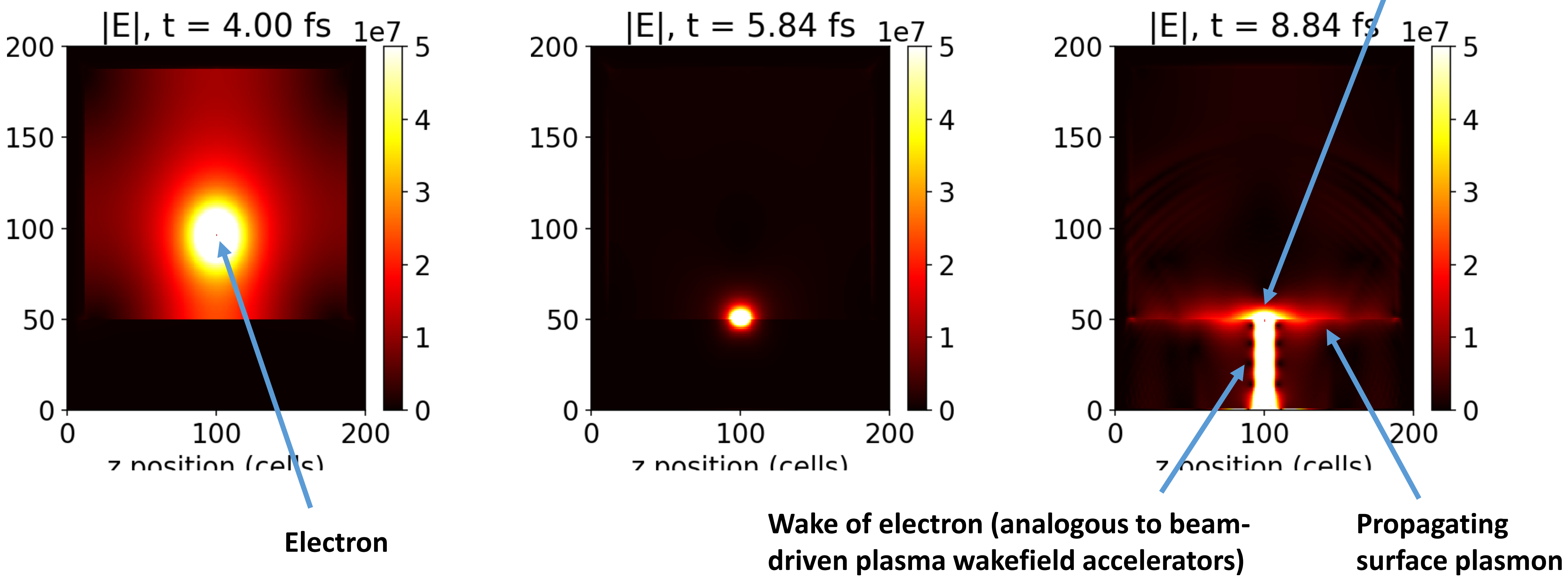


Impact Simulations

- It is unclear exactly how to best represent quantum tunnelling in a purely classical simulation.
- Thus, simulations of electrons impacting the surface were performed first.
- A single electron of velocity between 5e6 m/s and 5e7 m/s was injected from the top of the domain. Corresponds to kinetic energies between 71 eV and 7.1 keV.

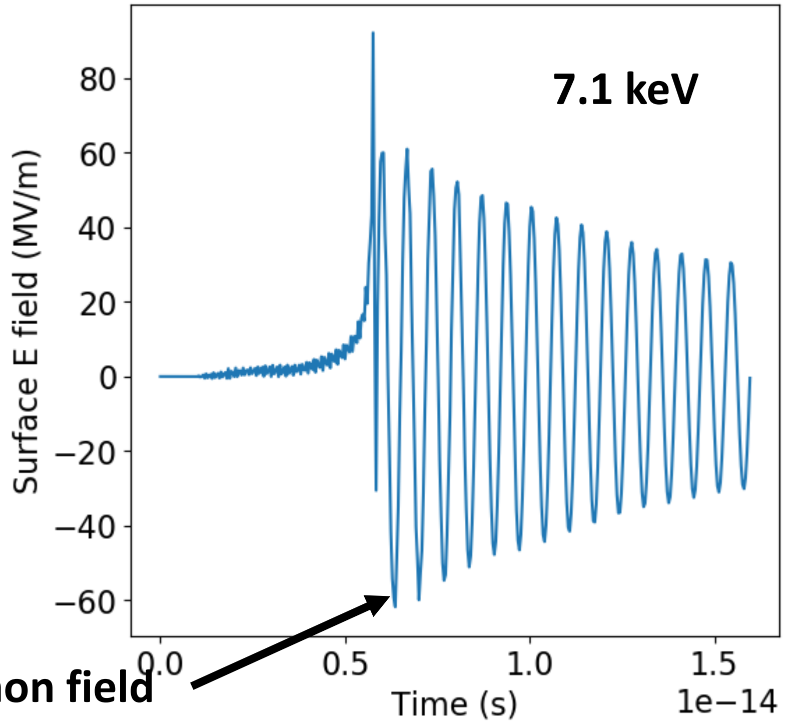
Impact Results – Example at 5e7 m/s (7.1 keV)

- Plots show E-field magnitude in V/m
- Impact excites localised plasmon oscillations. Low frequency components have nonzero group velocity and propagate out. High frequency components remain at the impact location.

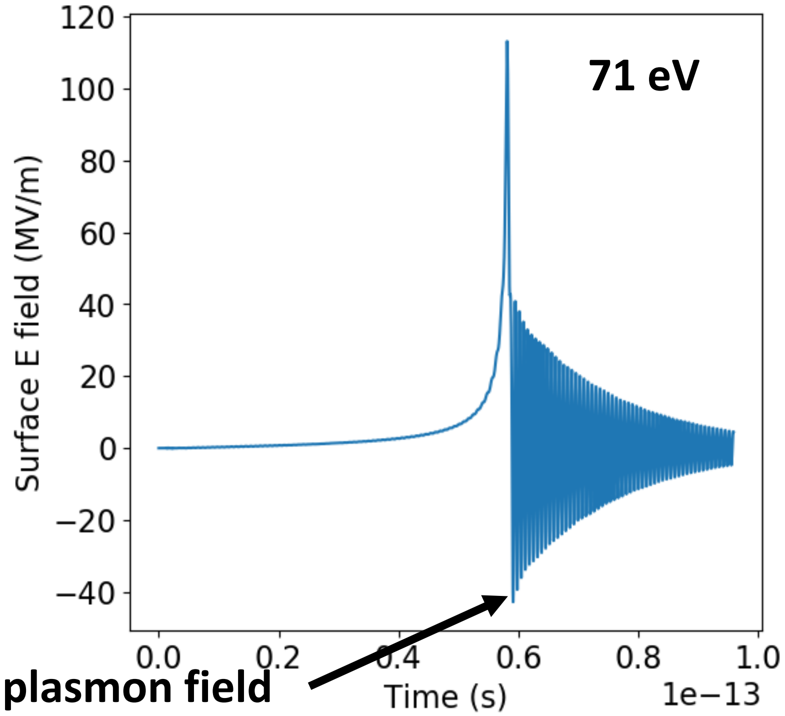


Impact Results – Intense Surface Fields

- Impact excites very large oscillating fields on the copper surface (peak field close to electron)
- Oscillation decays over some tens of femtoseconds due to the f_c term.
- Peak plasmon amplitude does not seem to vary much with electron energy. (Negative fields = electron emission)
- Note: domain thickness was 20 nm – peak electric fields are likely at least an order of magnitude higher in 3D.



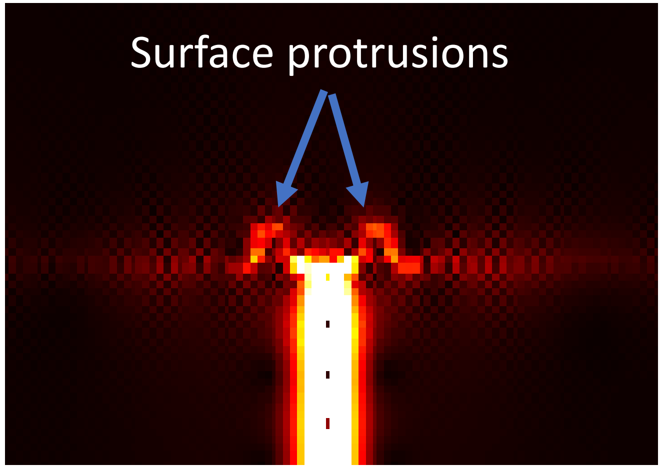
Peak plasmon field

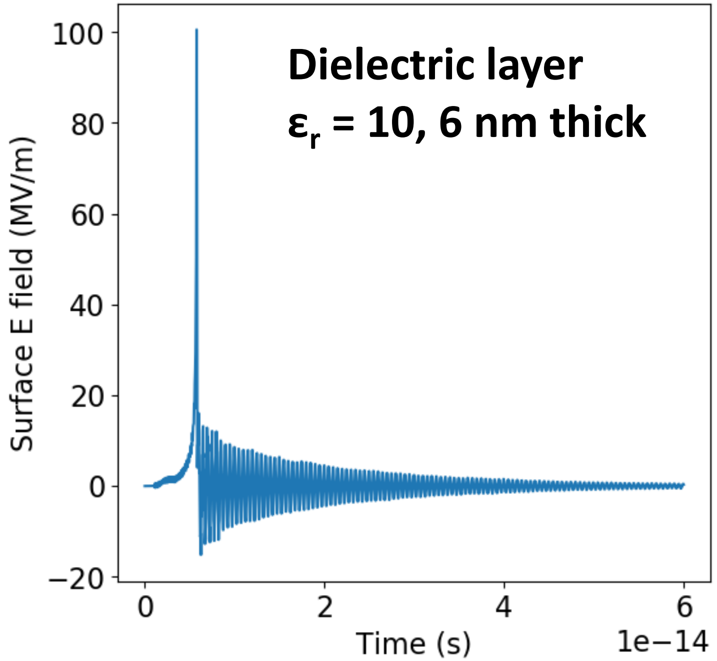


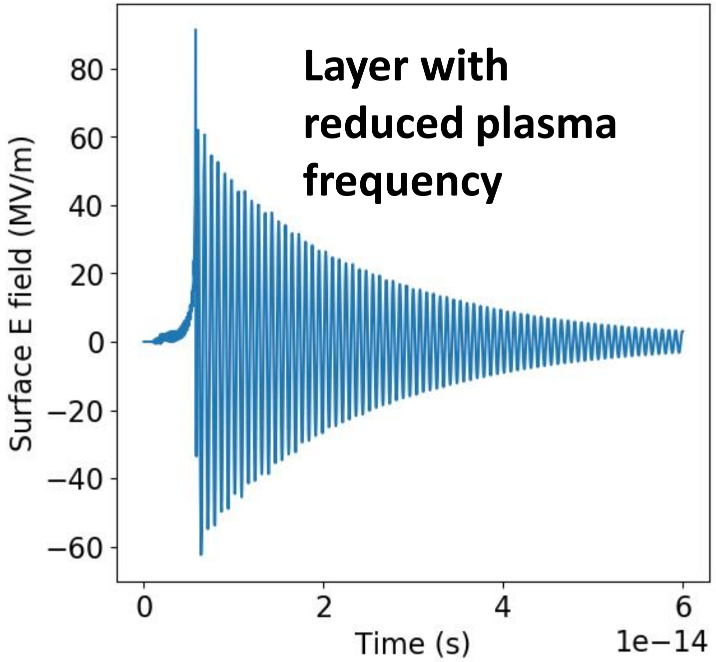
Peak plasmon field

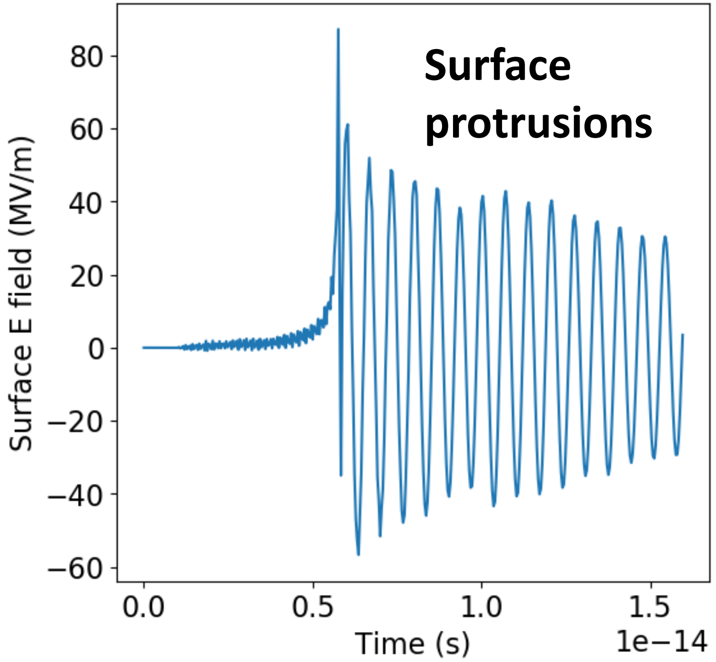
Effect of Surface Modifications

- Modifications to a flat copper surface were tried to see if the peak plasmon field could be increased, with 7.1 keV energy in each case.
- None of these seemed to produce a greater surface field than a flat surface.
  - Most of the plasmon's energy is in the low-vg components – so no resonant structure is necessary to confine the plasmon.







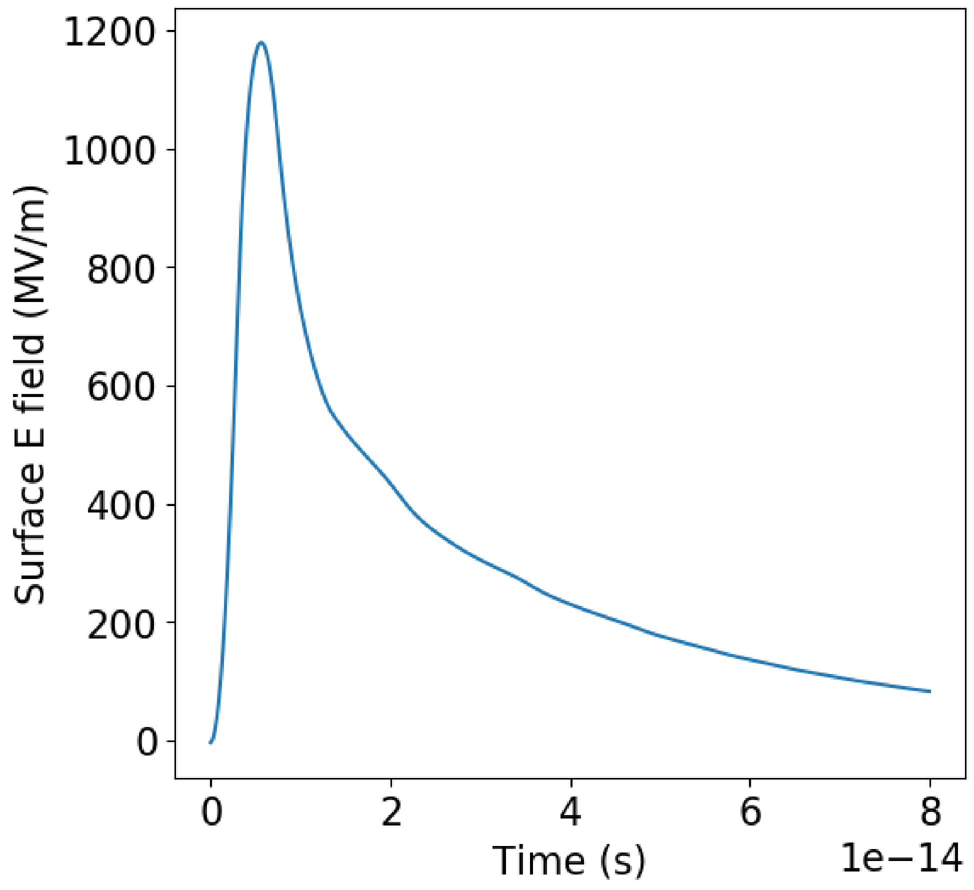


Thermionic Emission

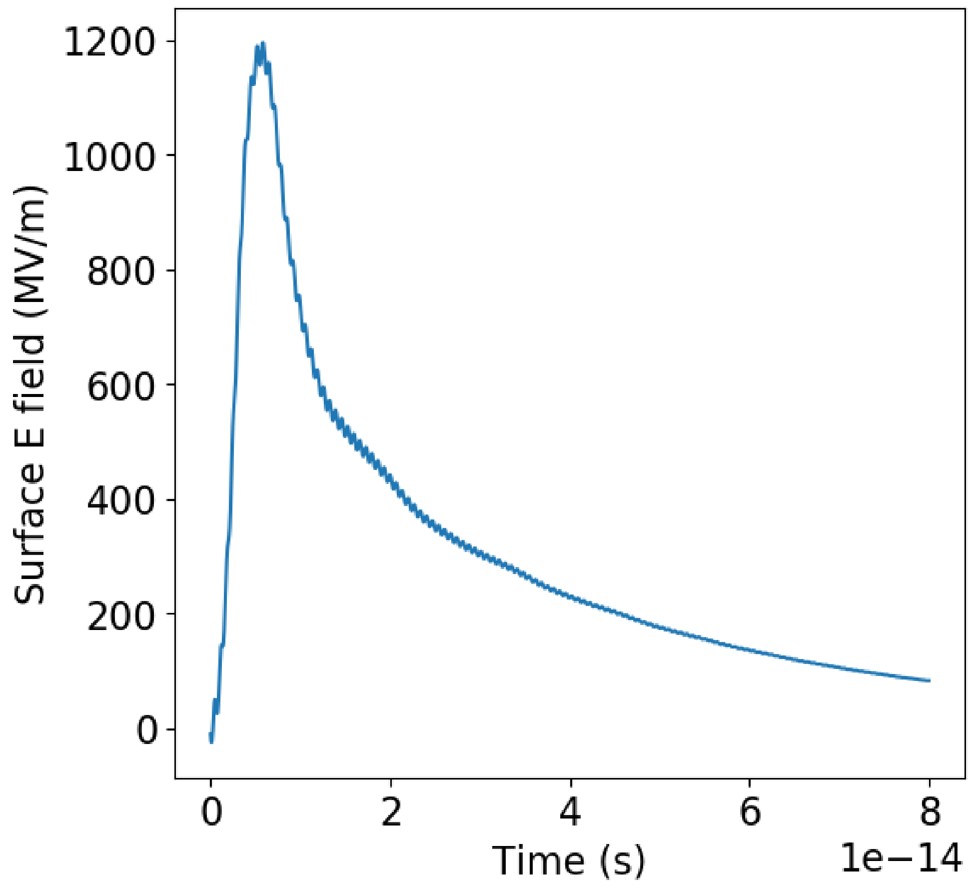
- Tracked electric field at the emission site on the copper surface over time.
- Different initial energies were tried. In each case, the electron was also accelerated by a 100 MV/m field in vacuum.
- Domain depth was 2 nm this time, showing about 10x higher peak fields than the impact simulations.
- For realistically low energies, the plasmon decayed before the electron was far enough away from the surface for further emission to occur.
- But, this may again be an issue with the 2D nature of the simulation, since the electric field due to a charge depends on $E \propto 1/r$ instead of $E \propto 1/r^2$

Thermionic Emission

1.2 eV initial energy, copper represented by PEC:

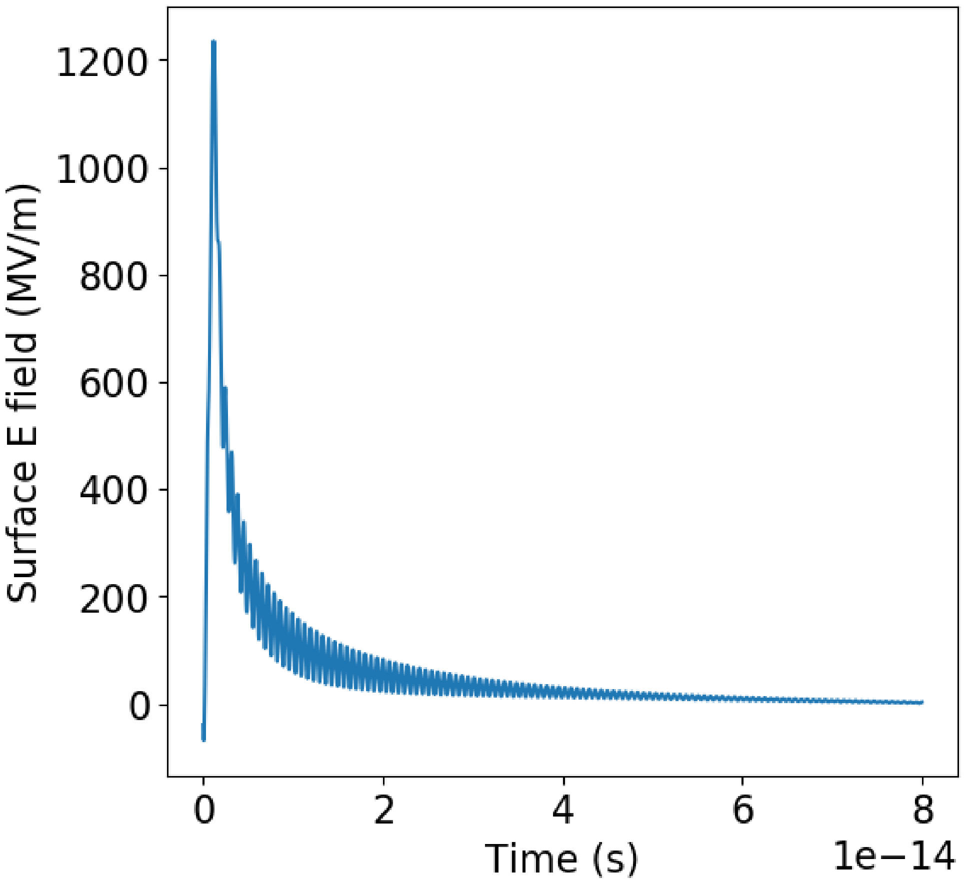


1.2 eV initial energy, copper represented by Drude model:

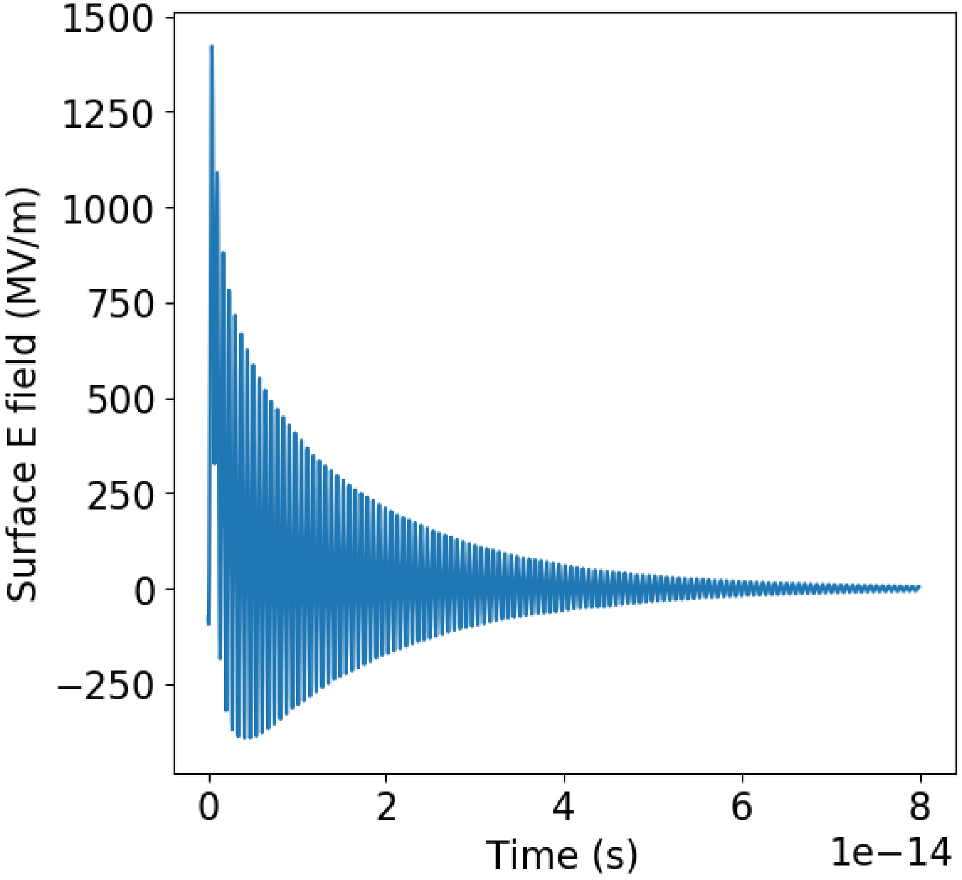


Thermionic Emission

20 eV initial energy, copper represented by Drude model:



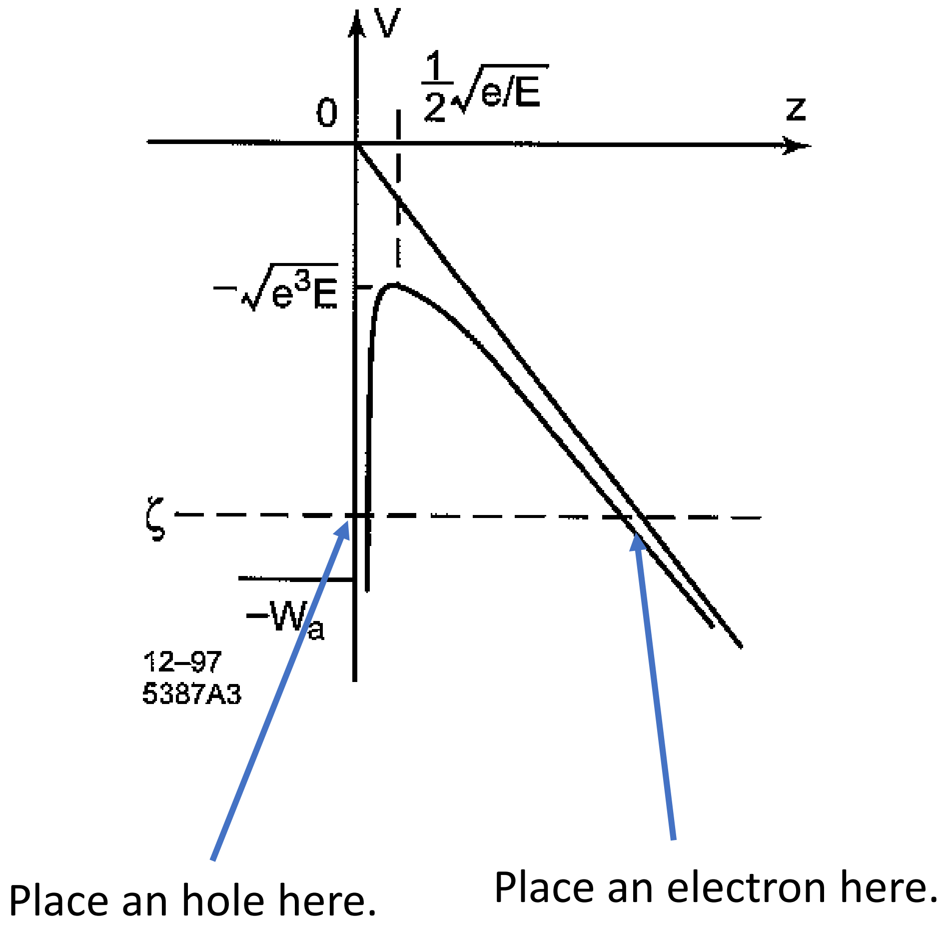
100 eV initial energy, copper represented by Drude model:



‘Charge Teleportation’

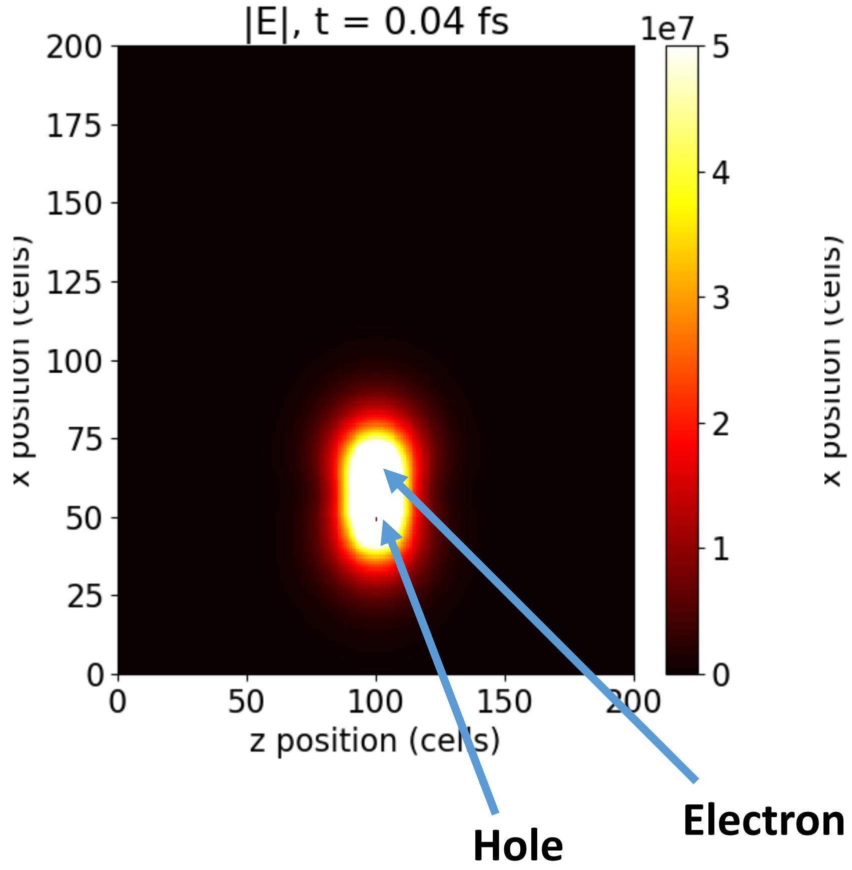
- See if spontaneous field emission at low fields could excite plasmons.
- Represent tunnelling by an instantaneous displacement of a charge across the potential barrier.
- 100 MV/m DC field: electron placed 30 nm above the surface
- Since Drude-type materials behave like vacuum at timescales much shorter than a plasmon period, the initial E-field was a dipole in vacuum.

Simulation setup

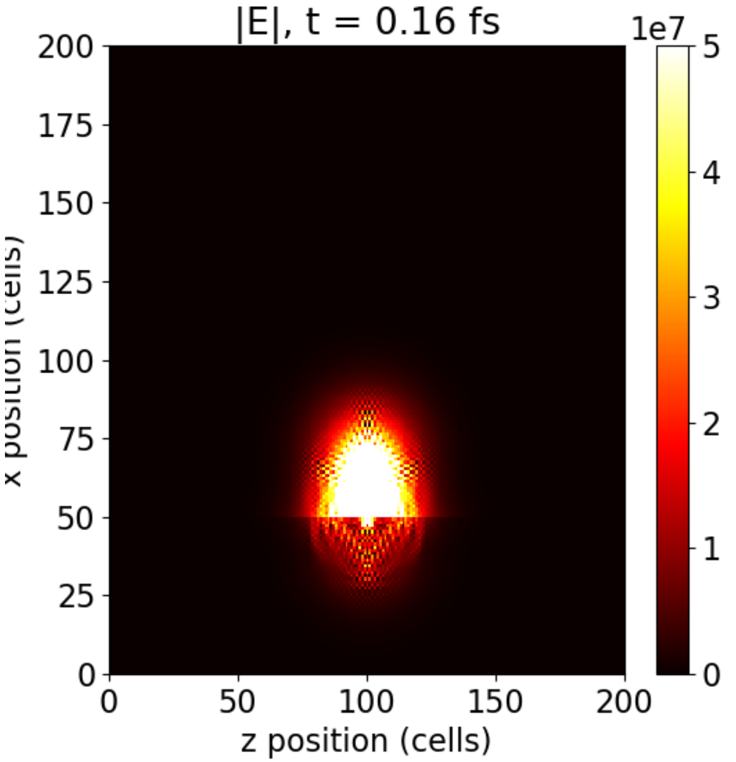


Charge Teleportation Results

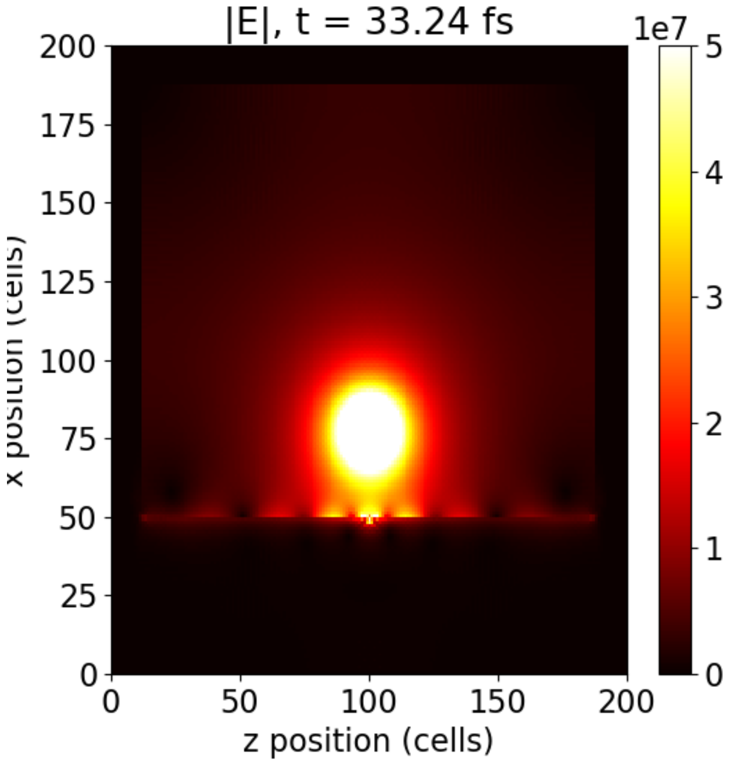
Initial electric dipole field:



The field inside the copper collapses in about 0.1 fs:

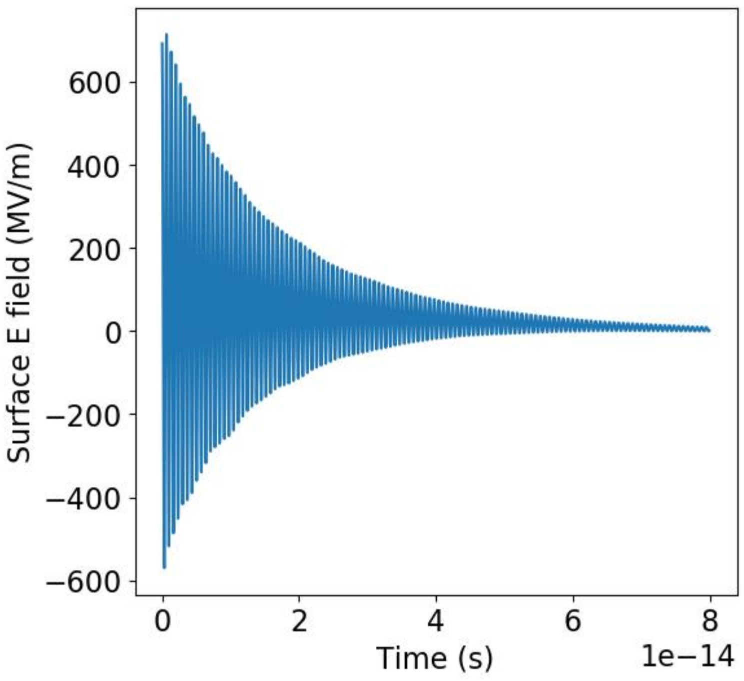


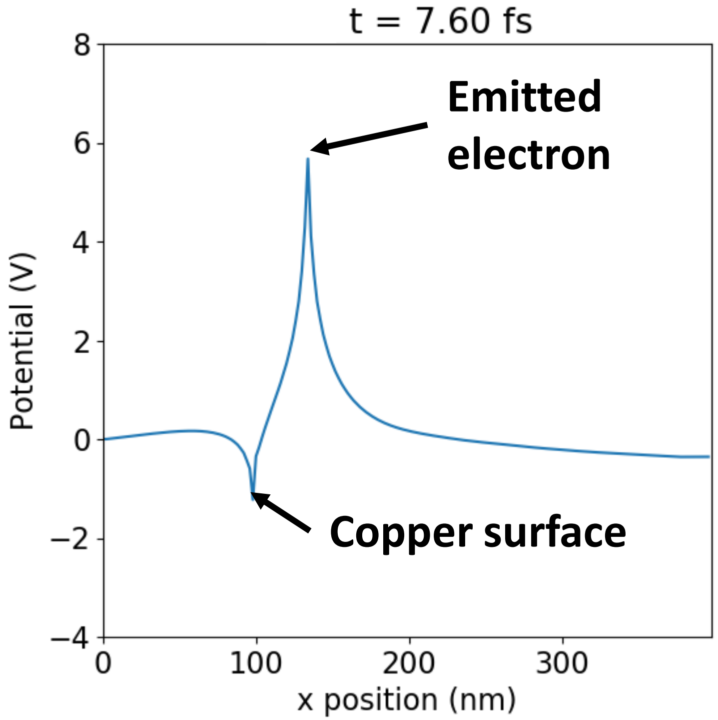
Plasmon oscillations decay while the electron is slowly accelerated away from the surface:



Charge Teleportation Results

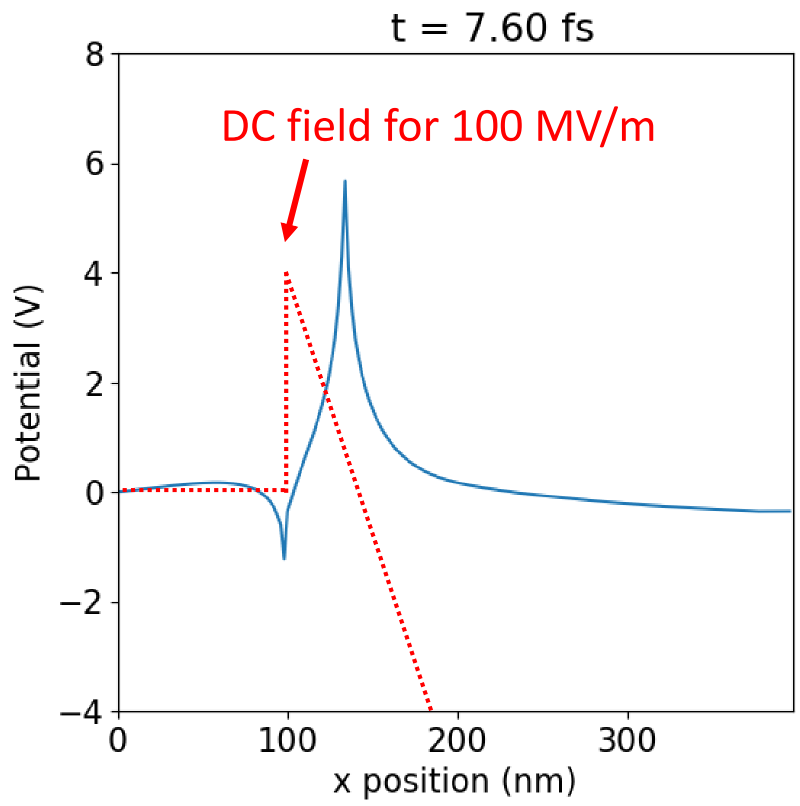
- The large discontinuity in charge excites plasmons effectively.
- The peak electric field on the emission site is quite high, but the previously emitted electron may still present another potential barrier.
- The potential plot on the right was obtained by integrating the electric field at an instant in time along a line normal to the surface crossing the emission site.
    - Does not take into account the work function, DC electric field, or transit time effects.





Further Thoughts on Potentials

- The plasmon field extends into the metal:
  - Can it help electrons gain kinetic energy before reaching the potential barrier?
- Need a local depletion of electrons at the emission site.
- As can be seen from the potential plot, the lowering of the potential barrier can be very significant.
- With a 100 MV/m DC field, the electron needs > 100 fs to move significantly away, much longer than the plasmon decay time of 10 fs.



Conclusions

- Under the right conditions, a single low-energy electron can excite plasmon oscillations with very large peak fields.
- The potential of a plasmon may be a more useful quantity to look at than the surface electric field. (Electrons can gain kinetic energy before reaching the barrier if the electric field extends into the metal)
- Plasmons decay relatively quickly, which poses two problems:
  - A relatively large current is needed to sustain a continuous plasmon oscillation at an emission site (on the order of $f_c q_e \approx 17\ \mu A$ per emitter, possibly up to an order of magnitude less depending on losses assumed)
  - The emitted electron that excited the oscillation can only move a short distance in this time, potentially preventing any emission from that site.
- Quantum phenomena such as interband transitions may have a very large effect on these results.

Thank You!

Drude Model

- All plasmon calculation in this study used a Drude model fit to experimental measurements of copper (but the frequency range over which these fits are valid is unclear).
  - The values used are:

$$\omega_p = 1.32 \times 10^{16}\ rad\ s^{-1}\ (8.68\ eV)$$
$$f_c = 1.05 \times 10^{14}\ s^{-1}\ (0.434\ eV)$$

  - Different sources generally agree on the value of ω_p, but not f_c.
  - J. Phys. Chem. 1987, 91, 634-643 gives $f_c = 0.0955\ eV$, and also claimed that Au, Ag, and Cu nanoparticles exhibit Raman enhancements (related to the Q of the plasmon resonance) much higher than theoretical predictions.
- Quantum phenomena (interband transitions) are also likely to have a very significant impact on the dispersion curve, resonant frequency, and losses, as shown in the plots on the right.
  - In the bottom plot, 'free electron loss' refers to the Drude model as used in these calculations. The Drude model predicts a plasmon resonant frequency on a Cu/vacuum interface of 6.13 eV, well into the region where interband losses are significant.

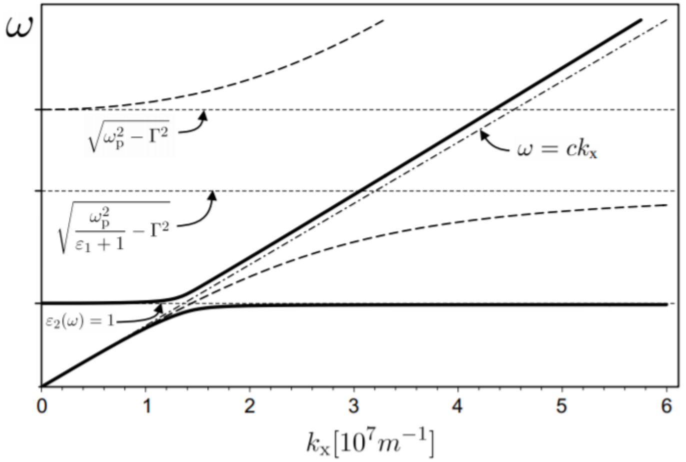


Figure 12.5: Dispersion relation of surface-plasmon polaritons at a gold/air interface. The solid line is the dispersion relation that results from a dielectric function accounting for a single interband transition. The dashed line results from using a Drude type dielectric function. The dash-dotted straight line is the light line $\omega = c \cdot k_x$ in air.

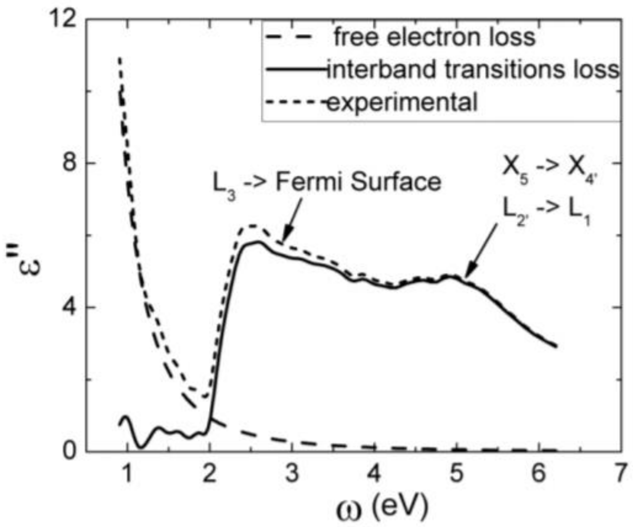


Figure 1 The losses in Cu shown as the sum of interband losses and free electron losses [41,62]. Annotations identify the interband transitions responsible for peaks in ε''.

Top figure: https://www.photonics.ethz.ch/fileadmin/user_upload/Courses/NanoOptics/plasmonss.pdf
Bottom figure: https://arxiv.org/pdf/0911.2737.pdf