SIMULATION OF PLASMONIC EFFECTS IN NANOSTRUCTURED COPPER SURFACES FOR FIELD-ASSISTED PHOTOEMISSION

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Abstract

Plasmonics in photoemission have recently gained interest due to their effect on important electron beam parameters such as emittance or quantum efficiency (QE), among others. For example, the presence of nanostructures at the surface of the photoemissive medium can increase the probability of electron emission if their morphological features match that of the plasmonic resonance when laser-irradiated in an electron gun. To determine the optimal size and geometry of these nanostructures and predict the resulting field enhancement, we conducted parametric electromagnetic simulations of nanostructured surfaces using COMSOL®. We investigated several types of geometries commonly found in laser processed copper substrates, optimized their performance under different irradiation wavelengths, and determined the expected maximal enhancement factor attainable.

INTRODUCTION

Photocathode technology plays a key role in modern high energy and charge electron accelerators [1–3]. The electronic properties of the photoemissive medium, coupled with the specifications of the extracting and laser fields, are key components to produce the required electron beam with maximal charge and minimum emittance. The photoelectric performance of photocathodes is characterized by the Quantum Efficiency (QE) and beam emittance [4]; QE is defined by the ratio of electrons emitted per photon hitting the cathode, as in:

\[ QE = \frac{J}{e} \]

where \( J \) is the current density of charges produced by a laser beam of intensity \( I \) and photon energy \( \hbar \omega \). A maximal QE obviously ensures a higher bunch charge for the electron beam produced. In the photoemission process, after electrons are excited by the incident laser pulse, they have to undergo a quantum tunneling through the potential barrier of the metal-vacuum interface to be emitted. Applying an additional electric field \( E \) to the surface results in a lowered work function, \( \phi \) as expected by the Schottky effect (\( \phi_{\text{schottky}} \)). In this sense, the photoemission current density \( J \) expressed in Eq. (1) depends on \( \phi \) and \( E \) as follows [5–7]:

\[ J = -e \oint N(W, k, E, T, \phi) \times D(W, k, E, \phi) dW dk \]

where \( N(W, k, E, T, \phi) \) is the supply function of electrons reaching the surface of the metal with a kinetic energy \( W \) and a transverse momentum \( k \) at a temperature \( T \). \( D(W, k, E, \phi) \) is the probability transmission of one of these electrons tunneling through the potential barrier. As a result, QE will increase as a function of the electric field \( E = E_{\text{RF}} + E_{\text{laser}} \) applied to the surface, a composite of the RF and laser fields. We investigate the possibility to apply this concept in photocathode technology, by using nanostructures at the irradiated surface to induce laser field enhancements via plasmonic effects, with the peculiarity of using photons of energy above the work function. This is what is known as field-assisted photoemission [8–10]. In addition to increasing the effective surface area, designing structures of optimal size and geometry could also excite the metallic plasmon of electrons. This will increase the charge density in a highly localized area, resulting in a laser field enhanced by a factor \( g = |E_{\text{local}}|/|E| \), where \( E_{\text{local}} \) is the local electric field calculated at each point of the metallic surface. In this work, we conduct electromagnetic simulations of deep UV irradiation of metallic nanostructured surfaces using COMSOL®, and determine the expected value of \( g \) for different geometries and parameters of wavelength. We focus our study on copper (Cu) as a photoemissive material, as it is widely used in RF photoinjectors around the globe. We compare the models and performances of periodic ripples in the shape of Laser Induced Periodic Surface Structures (LIPSS) and nanospheres, two types of nanostructures that appear when irradiating metallic surfaces with high repetition rate ultrafast laser beams [11–13].

PLASMONIC SIMULATIONS

The purpose of the simulation is to study the electromagnetic response of a 3D geometric array of nanometric morphologies at a surface, and irradiated by a perpendicular laser beam. We used the simulation software COMSOL® Multiphysics 6.0 to solve Maxwell’s equation in a 3D domain with its embedded Finite Element Method (FEM) solver for differential equations.

The simulation was built on a thick Cu surface placed in vacuum. The copper dielectric parameters were set to \( \varepsilon = 1 \) as for any standard metal, and \( \sigma = 5.998 \times 10^7 \) S/m. Nanostructures of different geometries and periodicities were then built with the Computer-Aided Design (CAD) module and attributed the same Cu parameters. An incident linearly polarized electromagnetic plane wave of wavelength \( \lambda \), defined as \( E = E_0 e^{-ikz} \) with \( E_0 = 1 \) V/m and \( k = 2\pi/\lambda \), was placed above the structures. The FEM meshing refinement was performed with a precision level of \( \lambda/10 \) and the solver was set to a parametric sweep to get a complete overview of the behavior of the plasmonic system. The output enhancement factor \( g \) is the ratio of the surface field over the scattered field.

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Figure 1: (a) 3D visualization of the field enhancement experienced by a LIPSS pattern with a spatial periodicity \( \Lambda = 170 \text{ nm} \) under a \( \lambda = 266 \text{ nm} \) illumination wavelength; (b) maximum field enhancement \( g_{\text{max}} \) of the structured surface, showing a resonance peak shifting for different resonant pairs of \((\Lambda, \lambda)\).

**LIPSS Nanostructures**

Periodic ripples like LIPSS were modelled as semi-cylinders placed periodically on a surface, and separated by smaller semi-cylindrical grooves carved into the surface. The aspect ratio of cylinder to groove was set to 1/5, according to SEM images and profilometric studies of LIPSS [14]. The structures were set perpendicular to the incident wave polarization to maximize the field enhancement factor. For the purpose of this study, only spatial periodicities \( \Lambda \) going from 150 to 300 nm were tested, because they are the typical geometries that have interesting plasmonic properties in the violet to deep UV spectrum, ideal for the purposes of UV laser driven photoinjectors [1]. However, we tested irradiation wavelengths far beyond, covering \( \lambda \) between 200 and 1000 nm to get a complete picture of the phenomena. Figure 1 shows a summary of the results of the model. Note that here we study the surface field data only, that is to say the applied electric polarization of Cu, and not the surrounding field.

The cylindrical arrays display clear plasmonic behavior, for both the field enhancement position and intensity are changing with the excitation wavelength. The orientation of the array was set perpendicular to the polarization of the incident field, as visible in Fig. 1.a because it was found that plasmonic resonance is more intense in that configuration (when put parallel, only the tip of the cylinders are affected and less overall surface area encounters a plasmon). Figure 1.b shows resonance peaks of the structure in the maximum enhancement factor data, shifting towards green resonant wavelength with bigger \( \Lambda \). This is logical, considering that bigger spatial periodicity scales up the characteristic size of the ripples, making the resonant cavities bigger and thus requiring larger wavelengths to resonate. LIPSS produced with DUV light often show periodicity around 170 nm to 200 nm. In this range, resonance is shown to appear at \( \lambda \) between 275 and 320 nm with an enhancement factor \( g_{\text{max}} = 2.58 \). This number, although quite promising, is to put in perspective with the spatial distribution of the field that will eventually decrease the effective experimental enhancement. Moreover, the model isn’t exactly accurate geometrically, for it is known that LIPSS aren’t perfectly periodic but rather quasi-periodic, and have a profile closer to a hyperbola rather than a circle. We may expect the actual numbers of a more accurate model to vary and potentially decrease. The overall plasmonic performance of LIPSS wasn’t quite matching the QE increase results measured experimentally, so we had to look deeper into the structural composition of laser processed Cu surfaces.

**Nanospheres**

Another type of structure observed experimentally in ultrafast Cu laser processing is nanospheres [13]. The size distribution depends on the laser energy during processing, and for DUV we saw that the average size was at a radius of 50 nm, with a spread between 20 and 100 nm.

We then built a far simpler simulation model for studying these nanospheres, which are well known to have unrivalled plasmonic properties due to their high degree of symmetry, making them very good resonators. We placed a Cu sphere of variable nanometric radius \( R \) on a Cu surface with a penetration depth inside the surface set at \( 0.2 \times R \) to match experimental observation. The incident electromagnetic wave, as well as the Cu dielectric parameters, were kept identical. The study was conducted looking at variations of the \((R, \lambda)\) pairs. Here, polarization orientation does not matter as long as it is linear. Figure 2 shows that peak surface enhancement is able to go much higher than what has been obtained with LIPSS. In fact, a 50 nm radius nanosphere excited by a 250 nm incident field is expected to show enhancement with \( g > 3.5 \) according to Fig. 2.c. Once again, plasmonic behavior is clearly displayed in Fig. 2.b, where we see that a nanosphere of bigger size excited by a relatively smaller wavelength will present a plasmonic resonance of mode \((1,1,0)\) \footnote{Spherical cavity modes are characterized by the number of nodes in three directions: the degree, the radial order and the azimuthal order.}. The resonance map in Fig. 2.c also shows that resonant modes are specific to particular couples of val-
ues \((R, \lambda)\). However, we notice an enhancement line that holds for all \(R\) around \(\lambda = 250\,\text{nm}\), which most likely corresponds to the case where \(h\omega = \phi_{\text{Cu}}\), that is to say when photon energy matches the work function level of Cu.

The arrow size and direction shows the intensity and orientation of \(E\) at a given point. We observe that when the field enhancement at the surface of the metal only reaches so high as \(g = 3.5\) for a 50 nm sphere, in the direct neighboring of the surface it goes up to \(g > 6\), inducing a much more intense extraction potential and reducing the barrier even more. The orientation of the field is variable depending on the plasmonic lobes of different modes, emphasizing the role of the phase of the wave \(E\). the plasmonic field will be successively positively and negatively oriented relative to the sphere’s surface, and oscillating through time. We can thus expect the extracting field to be increased only one side at a time, while the other will be decreased, and both will switch at the frequency of the field.

Once again, we can comment that the effect is highly local, even more on such a small nanostructure that only cover a very narrow portion of a typical photocathode surface. The field enhancement is then to be correlated to the size distribution and space density of nanospheres in order to estimate real life performance of such a plasmonic photoemitter.

**CONCLUSION**

Plasmonic effects are a promising approach to improving the electronic properties of photocathodes, as we demonstrate with this first study that important field enhancements can be achieved in the surface of the metal \((g = 3.5)\) and in its vicinity \((g > 6)\). We also demonstrate the good versatility and adaptability of the effect for different wavelengths but also possibly for other materials, as the plasmonic resonant frequency highly depends on the dielectric constants of the medium. Nevertheless, the high locality of the field enhancement effect, together with its sensitivity, demands geometric and electromagnetic parameters to be very finely adjusted to reach peak plasmonic resonance, otherwise the contribution might be very minor, nay counterproductive. In addition, the field distribution problem with the nanostructure density, giving the expected performance of a macroscopic system, is also to be addressed in more details. A more in-depth topological study could be conducted to determine the most optimized nanostructure shapes depending on experimental requirements, as well as their distribution over a large surface to maximize the enhancement.

**REFERENCES**


Figure 2: (a-b) 3D visualization of Cu nanospheres of two different radii \(R = 50\,\text{nm}\) and \(R = 100\,\text{nm}\), irradiated by \(\lambda = 266\,\text{nm}\) light, showing two distinct resonant modes; (c) Resonance space of a Cu nanosphere as a function of the excitation wavelength \(\lambda\) and the radius \(R\).

Figure 3: Representation of the electric field vector component \(E\) in the transverse section of the nanosphere model, in the polarization plane, for two different nanosphere sizes: \(R = 50-100\,\text{nm}\).


