SURFACE PLASMON RESONANCE ENHANCED MULTIPHOTON EMISSION FROM METALLIC CATHODE*

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Abstract

We investigate the use of surface plasmonic nanostructures in increasing the efficiency of copper photocathodes for high-brightness electron sources. A nanohole structure was fabricated, using focused ion beam milling, such that it exhibited a strong plasmonic response near 800 nm. The reflectivity of the nanostructured surface was significantly lower compared to that of a flat surface, due to plasmonic resonance effects. A previous experiment [1] demonstrated an increase in charge yield by over a factor of 100, using a slightly off-resonance structure. With this new near-resonant structure, the reflectivity is expected to be even lower and the charge yield enhancement factor even larger.

INTRODUCTION

Recently, progress has been made in using metal photocathodes to generate high brightness electron beams, as an alternative to using semiconductor cathodes [2]. Because of their prompt response, low vacuum requirements, robustness, and fabrication simplicity, metals are typically the preferred choice in high peak brightness photoguns [1]. However, metal cathodes are not without disadvantages, a significant one being that they are often highly reflective at optical and infrared wavelengths and in general offer lower QE than semiconductors [3].

We, at the UCLA Pegasus laboratory, have demonstrated the generation of electron beams through multiphoton emission using a copper photocathode with an 800 nm Ti:Sa incident laser [2]. At 800 nm, copper is ~95% reflective, and thus, most of the photons from the incident laser are wasted and do not participate in the multiphoton emission process [3]. To increase the efficiency of the cathode, an obvious solution would be to increase the absorptivity of copper at 800 nm. One way we propose to do this is to pattern a cathode surface with nanoscale features that take advantage of plasmonic effects.

THEORY

Recent advances in nano-plasmonics have enabled fine control of optical properties of metal surfaces by patterning them with sub-wavelength or nanoscale features [4]. Nanoscale features of the correct geometry can lead to surface plasmon resonance, or a resonant interaction between surface plasmons and incident photons [5]. This produces two major effects [4]: 1) significant reductions in metal reflectivity due to photons

being absorbed by surface plasmons, and 2) large enhancements of local optical field intensity due to photon energy being stored in the surface plasmons. These nano-plasmonic phenomena could be applied to optimize the optical response of a cathode and obtain high efficiencies in photoemission charge yield.

Previously, we reported the first experimental results of the generation of relativistic electron beams from a nanostructured photocathode in a high gradient RF gun [2]. The aforementioned nanohole array structure exhibited a resonant response at ~810 nm. At 800 nm, the reflectivity of the nanopattern was ~60%, still producing a >100-fold enhancement in the multiphoton charge yield. With a nanopattern that is resonant closer to 800 nm, the charge yield can be expected to be greater.

SETUP

The geometry of the nanopatterns consist of rectangular arrays of nanoholes fabricated with focused ion beam (FIB) milling, as shown in Fig. 1. The patterns were milled onto a single crystal Cu(100) substrate to avoid pattern variation due to the random grain sizes and orientations associated with polycrystalline substrates.
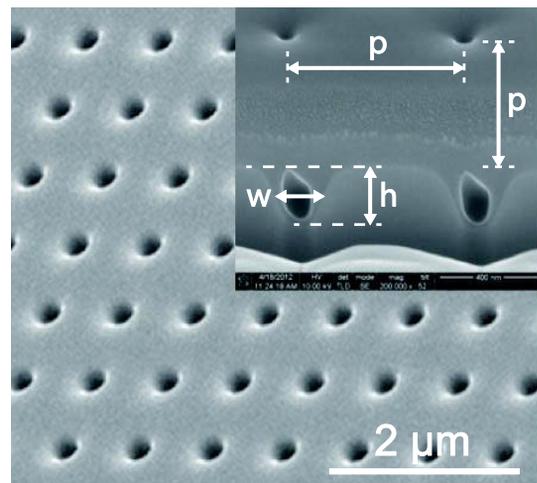
Figure 1 [1]: Scanning electron microscopy images of a nanohole array. Inset: Zoomed in profile view of nanoholes.

When using the nanohole array geometry, the resonant wavelength is much more sensitive to the spacing of the holes than to their depth, width, and even their shape [1]. Each nanohole has an approximately Gaussian profile, mainly due to the current density distribution of the milling ion beam. Finite Difference Time Domain (FDTD) simulations were performed to compare the

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difference in resonant response between Gaussian-shaped holes and ideal rectangular holes, using the same depth, width, and spacing. We found that the resonant wavelength for these two profiles differed only by about 4 nm, as shown in Fig. 2. This finding is important in that it allows us to fine tune the resonant wavelength primarily by changing the hole spacing p, which is the most controllable and reproducible parameter in a FIB device.
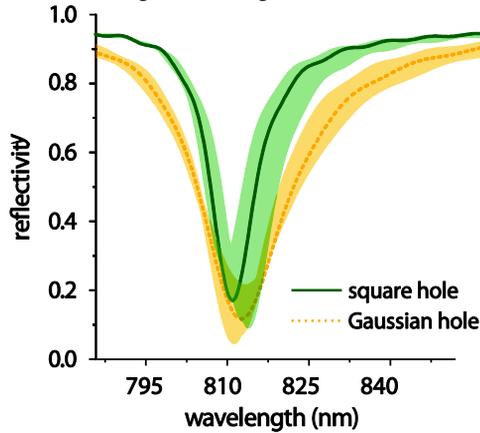
Figure 2 [1]: Simulated reflectivities at normal incidence of two types of nanohole pattern: square holes and Gaussian holes.

In this study, we fabricated nanopatterns with spacing p of 765 nm, shown in Fig. 3. The dimensions of the structure (spacing p, depth h, and FWHM width w of the holes) were optimized through finite difference time domain (FDTD) simulations. The values of h, w, and p are ~300 nm, ~200 nm, and ~765 nm respectively. The middle right pattern is the same as the top and middle left patterns, but rotated 90 degrees. This is to check the possibility of asymmetry in the nanoholes, i.e. the holes could be slightly elliptical rather than perfectly circular. Fig. 4 shows the reflectivity curves for the un-rotated and rotated patterns. When imaging the nanopattern at normal incidence using a 2 nm FWHM laser at the resonant wavelength, the incident light is strongly absorbed (Fig. 3).

Figure 3: Nanopatterns with 200 nm FWHM width and 300 nm depth. Top left: 765 nm spacing. Middle left: 765 nm spacing. Middle right: 765 nm spacing, pattern rotated 90 degrees with respect to first two patterns. Bottom right: practice pattern. Pattern square sizes: ~ 100 μm by 100 μm.

When imaging the nanopattern at normal incidence using a 2 nm FWHM laser at the resonant wavelength, the incident light is strongly absorbed, as shown in Fig. 3. The measured resonant wavelength of this nanopattern is 795 nm with a bandwidth of 34 nm, and corresponds to a reflectivity of 12%. At 800 nm, the nanopattern exhibited a reflectivity of 23%.
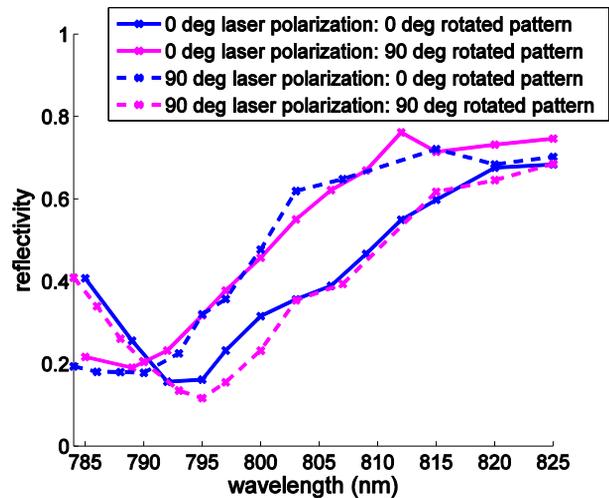
Figure 4: Reflectivity curves for most recent nano-patterns. Solid curves: 0 degree laser polarization. Dashed curves: 90 degree laser polarization. Blue curves: un-rotated pattern. Magenta curves: rotated pattern, 90 degrees with respect to un-rotated pattern.

ANALYSIS

The reflectivity curves in Fig. 4 indicate polarization dependence for the nanostructures. For a pattern, the un-rotated pattern for example, the resonant wavelength shifts when the laser polarization is changed by 90 degrees (blue curves). The rotated pattern exhibits the same behaviour, but in reverse (magenta curves). This suggests that the holes are indeed slightly elliptical in shape. The small shift (< 4 nm) in the plasmonic resonance peak can be explained due to some asymmetry in the FIB machine's focus in the x and y directions in agreement with the simulation prediction.

It is interesting to compare the potential increase in charge yield from the 795 nm resonance pattern to the previous 810 nm resonance pattern. In the previous experiment [1], IR laser pulses (150 fs FWHM) were focused to 120 μm rms and scanned around the nanopattern at near normal incidence (<1°) with a piezo-controlled mirror. The position of the laser spot on the cathode was monitored using a virtual cathode screen, and the generated beam charge was measured by a calibrated high efficiency beam profile camera. The maximum

signal, obtained when the laser spot fully covered the nanopattern, was $Y_{exp} > 1.2 \times 10^2$ times greater than when the laser was hitting only a flat surface [1].

This large increase cannot by fully explained by just the change in IR reflectivity and absorption. The generalized Fowler-Dubridge theory [6] predicts an increase of $Y_{FD}=(1-R_p)^3/(1-R_f)^3=27$ times, where $R_p=64\%$ and $R_f=88\%$ are the measured reflectivities of the previous nanopattern and the flat cathode surface, respectively. The additional enhancement in charge emission can be attributed to the optical intensity enhancement due to surface plasmon resonance (Fig 5.).
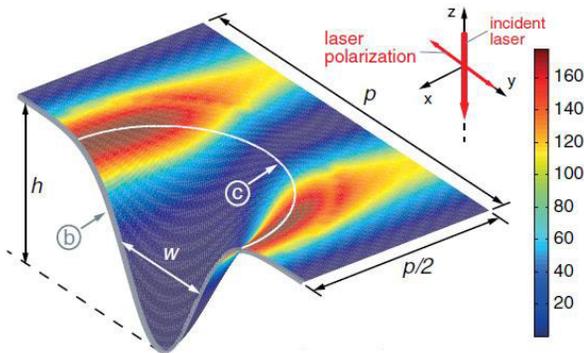
Figure 5 [1]: Intensity distribution on the nanohole surface.

It is estimated that a nanopatterned surface produces ~5 times more charge than a flat surface under equal absorbed intensity [1]. As mentioned, the more recent nanopattern exhibited a reflectivity of $R_p=23\%$. From a reflectivity standpoint alone, this corresponds to a charge enhancement factor of $Y_{FD}=(1-R_p)^3/(1-R_f)^3\sim250$ times. Taking into account the enhancement due to surface plasmon resonance, this suggests that the total enhancement factor could be at least 1000.

It is important to consider possible limitations to this large enhancement factor. It has been demonstrated that structural changes to a nanopatterned cathode surface occur when the absorbed fluence approaches 10 mJ/cm^2 [1]. The result is consistent with the 50 mJ/cm^2 limit reported for a flat copper surface [7] taking into account the ~5 times enhancement due to surface plasmonic effects. This sets an upper limit to incident laser power for nanopatterned surfaces that is lower than the limit for a flat surface, i.e. using an arbitrarily high laser power does not yield an arbitrarily high charge yield. In regards to an upper limit to charge yield, at present, it is unclear whether the localized emission of photoelectrons from a nanostructured cathode sets a different limit on the highest extractable charge density than a flat cathode case, but that should be an interesting topic for future studies.

CONCLUSION

In summary, this work demonstrated the potential to use plasmonic nanostructures to change the optical properties of a copper photocathode. In doing so, we were able to mitigate the inherent ~90% reflectivity of a flat copper surface. We showed a significant increase in charge yield from using a previous nanostructure that was 64% reflective at 800 nm, and thus, we can expect any even larger increase from the current nanostructure, which is 23% reflective. Our next steps include generating an electron beam with this nanostructured cathode and comparing its characteristics (charge yield, intrinsic emittance, and bunch length) to that of a beam from an ordinary flat cathode. We envision this work to be a promising step in a new direction of photocathode research.

ACKNOWLEDGMENT

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