ISBN: 978-3-95450-232-5 ISSN: 2673-7000 doi:10.18429/JACoW-NAPAC2022-TUYD4

TOWARDS HIGH BRIGHTNESS FROM PLASMON-ENHANCED PHOTOEMITTERS

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Abstract

Plasmonic cathodes, whose nanoscale features may locally enhance optical energy from the driving laser trapped at the vacuum interface, have emerged as a promising technology for improving the brightness of metal cathodes. A six orders of magnitude improvement [1] in the non-linear yield of metals has been experimentally demonstrated through this type of nanopatterning. Further, nanoscale lens structures may focus light below its free-space wavelength offering multiphoton photoemission from a region near 10 times smaller [2] than that achievable in typical photoinjectors. In this proceeding, we report on our efforts to characterize the brightness of two plasmonic cathode concepts: a spiral lens and a nanogroove array. We demonstrate an ability to engineer and fabricate nanoscale patterned cathodes by comparing their optical properties with those computed with a finite difference time domain (FDTD) code. The emittance and nonlinear yield of the cathodes are measured under ultrafast laser irradiation. Finally, prospects of this technology for the control and acceleration of charged particle beams are discussed.

SURFACE PLASMON POLARITONS

Surface plasmon polaritons (SPPs) are the evanescent electromagnetic waves that may exist at metal-dielectric interfaces. For a single planar interface, the wave's dispersion relation may be solved exactly starting from Maxwell's equations [3] with the result that,

$$k_x = \frac{\omega}{c}\left(\frac{\varepsilon_1(\omega)\varepsilon_2(\omega)}{\varepsilon_1(\omega)+\varepsilon_2(\omega)}\right)^{1/2}, \qquad (1)$$

where $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ are the relative permittivity on either side of the interface. For a free-electron metal, $\varepsilon(\omega) = 1-\omega_p^2/\omega^2$, where ω_p is the plasma frequency. If we consider a cathode application and take the dielectric to be the vacuum ($\varepsilon(\omega) = 1$) then the dispersion lies to the right of the light line with an asymptote at $\omega_{sp} = \omega_p/\sqrt{2}$ (Fig. 1).

The SPP dispersion relation has two consequences for photocathode physics. Firstly, plasmons may be confined to smaller spatial dimensions and achieve higher optical intensity than free-space light. This is due to the fact that SPPs may have a shorter wavelength than light in a vacuum for optical photon energies. Second, additional momentum may be required to couple laser light in SPPs which in cathodes is commonly supplied with a grating structure.

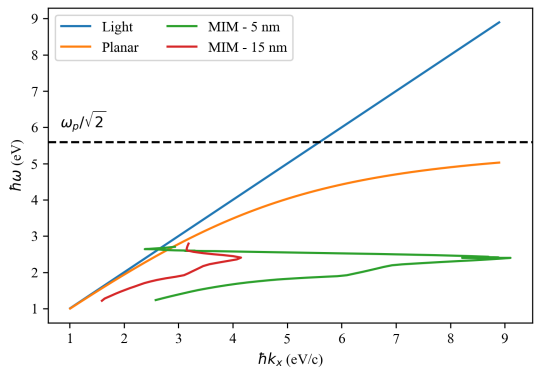

Figure 1: Dispersion relationship of an SPP at a gold-vacuum interface (Planar) for the free electron model (optical parameters from Ref. [4]). Dispersion relationship for two gap widths in a metal-insulator-metal system shown from Ref. [5] (MIM - 5 nm and MIM - 15 nm). Light line shown superimposed (Light).

ENGINEERED NANOGROOVE PHOTOCATHODES

Consider an otherwise flat gold photocathode patterned with a grating made of nanoscale trenches. Typical width and depth of the grooves are ~10 nm and ~50 nm. Typical grating pitch is ~ 700 nm. The two vertical walls of the grooves form a metal-insulator-metal system that supports coupled plasmons at the two interfaces. For small insulator width, the plasmon wavevector can become much longer than for free-space light at IR energies [6] (Fig. 1). Capping the insulator off at the bottom and leaving the cavity open at the top forces to the system to adopt Fabry-Perot like resonant modes [7]. These modes (an example of which is shown in Fig. 2) locally enhance the optical intensity at the groove edges. Prior research has shown that this may increase the nonlinear yield of the system by a factor of 10^6 over that of flat gold [1].

In our work, we fabricated nanogroove photocathodes in a variety of dimensions and studied the effect this had on their optical and photoelectric properties. Four pitches (670 - 700 nm) and groove widths (14 - 17 nm) were chosen with the depth fixed at ~ 50 nm. We measured reflectivity

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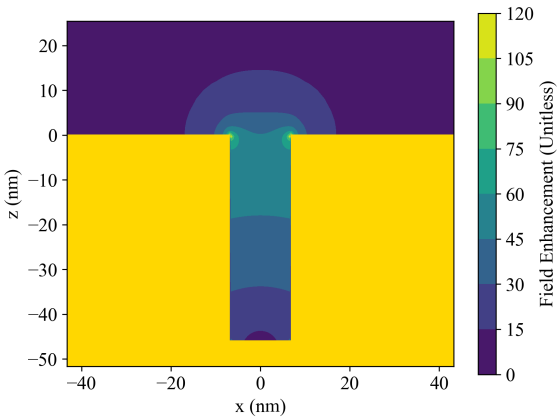

Figure 2: Cross sectional view of a single groove in nanogroove array cathode with up being the direction down the beamline. Field enhancement is shown for illumination with ~800 nm light calculated using an FDTD code [8].

spectra of the cathodes and fit them to FDTD calculations with width (which is ill-determined from the fabrication process) as the free parameter. The absorption peaks' wavelengths and widths correlated well with the calculations (example in Fig. 3) demonstrating an ability to engineer nanogroove arrays (after calibrating out the width).

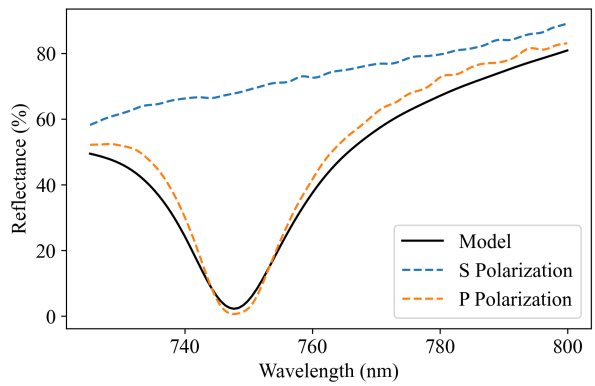

Figure 3: An example measurement of the of the reflectivity for polarization pointing along (S) and against the grooves (P). Calculated reflectivity from an FDTD code [8] is compared with the measurement.

Nonlinear yield was estimated for each sample by measuring photocurrent as optical intensity from the Ti:Saph laser driving the cathode was varied. We found that the cathodes supported a nonlinear yield enhancement of around 10^6 as in Ref. [1]. This yield does correlate with the amount of optical power absorbed by the cathode which is known from the laser's spectrum and the cathode's reflectance data. However, the correlation is imperfect and geometric factors / optical enhancement of the cathode may need to be accounted for to accurately model yield.

Finally, we performed what we believe is the first emittance measurement on this system. The cathode was loaded into a low voltage (20 kV) photoemission electron gun and the emitted beam passed through a solenoid to a final scintillator screen. Using a linear model of the system, the initial phase space moments may be fit using the method described in Ref. [9]. Final analysis is ongoing, but our initial results suggest strong asymetry of the emittance in the directions along and against the grooves (Fig. 4). We believe this asymmetry can be explained by the roughness effect of electrons emitted from the sharp groove edges. Further details on this work can be found in our manuscript awaiting publication.

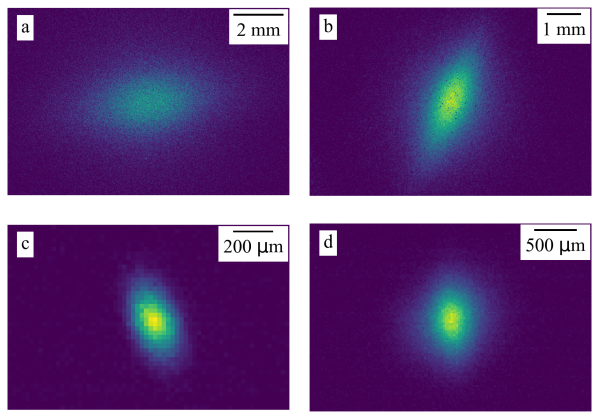

Figure 4: Images of the beam in solenoid scan measurements of the nanogroove cathodes showing strong asymmetry in emittance. Images a-d are for solenoid currents 0 A - 9.5 A.

CHARACTERIZING THE SOURCE SIZE OF NANOLENS EMITTERS

Flat emitters in conventional photoinjectors are limited by diffraction to sizes near ~10 µm depending on the optics used to form the laser spot on the surface. Nanometric tips may achieve smaller sizes, but at the cost of high initial momentum spread of the emitted beam [10]. One suggested concept to achieve small source sizes from a flat emitter is to manufacture a nanoscale diffractive lens on the surface of a metal photocathode [2]. Although the plasmon wavelength at infrared energies (here, 1.5 eV) isn't much shorter than that of free-space light (Fig. 1), the lens may form a focus of only a few hundred nanometers due to what is effectively a high numerical aperture. Then, nonlinearity in the photoemission process can be exploited to further shrink the source size to, what estimates say, is near 150 nm [2].

We have fabricated and studied a nanolens cathode similar to the one in Ref. [2]. Instead of a bullseye pattern, grooves were formed in the shape of a spiral (Fig. 5). This enables the cathode to operate with circularly polarized light instead of the more difficult to produce radially polarized light. Cathodes were placed in the previously mentioned low voltage electron gun and excited with a Ti:Saph oscillator (300 mW

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average power, 80 MHz rep. rate, 15 fs FWHM). Current would appear and then decrease with a characteristic time of 10 s which we attribute to point emitters on the sample burning off.

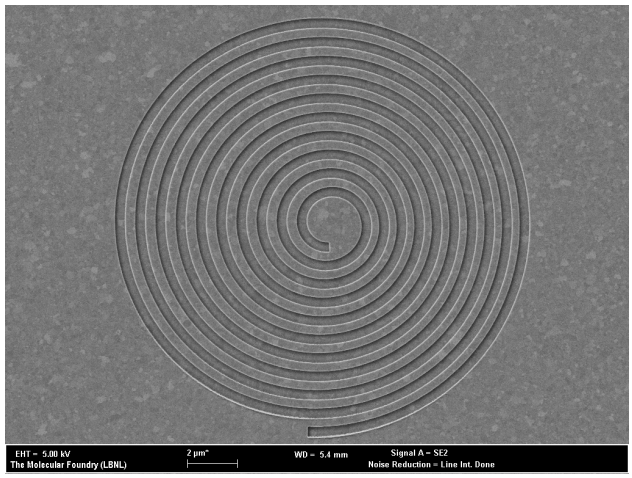

Figure 5: Scanning electron micrograph of one of the spiral nanolens cathodes.

After waiting long enough, we did measure sustained current (1 pA) from the cathode for circularly polarized light. The current vanished with the handedness of the circular polarization was swapped. This is a possible indication of lensing effects as the cathode only operates for one polarization of light. However, the current was too small to be measured at the scintillator screen.

The nanolens cathodes were moved to a photoemission electron microscope where the spatial distribution of the emitted electrons could be imaged. After illuminating the sample with an ultrafast laser, points around the spiral edges could be observed emitting electrons. The points did burn up and clean themselves over time as observed in our earlier measurements, but not enough to see small sources sizes. Work is ongoing to determine methods of fabricating nanolens photocathodes that lack point emitters.

ALL-OPTICAL ELECTRON BEAM CONTROL

Jitter in the RF control systems of accelerators can become an important limit to the time resolution of ultrafast beam-based techniques. Using the same laser that drives the photocathode and pumps the sample to drive control elements would nearly eliminate this problem. However, free-space interactions between electrons and photons are weak and limited to higher order processes, a restriction that does not necessarily apply to evanescent fields such as SPPs.

By engineering structures through which electron beams may pass and that also support and manipulate SPPs, all optical control of electron beams could be realized. As a first blush of this concept, a structure consisting of a thin (~1 µm) gold foil with a ~1 µm hole is considered. Surrounding the hole on both sides of the foil is a bullseye structure as in Ref. [2]. When light is used to excite the structure, optical energy is channeled through the hole. An FDTD calculation of the time dependent on axis field for an unoptimized structure is shown in Fig. 6. Sending an electron through this channel realizes an energy gain per peak field magnitude in the structure of 1.2 eV/(MV/m). Future work could improve the accelerating gradients and length of the interaction region in these structures and also enable bunching of electron beams.

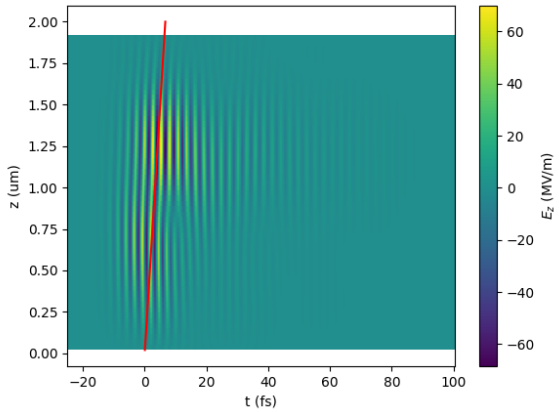

Figure 6: Electric field on axis (IE the path a particle would take) through a hole in a plasmonic structure calculated with FDTD software. The structure is centered at $z = 1$ µm in the simulation volume. In red is the trajectory of an electron traveling through the structure. The electron's initial energy and time of arrival have been optimized for energy gain through the structure.

ACKNOWLEDGEMENTS

Work at the Molecular Foundry was supported by the Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy, under Contract No. DE-AC02-05CH11231. C.M.P. acknowledges US NSF Award PHY-1549132, the Center for Bright Beams and the US DOE SCGSR program. D.B.D. and A.M.M. acknowledge support from NSF Science and Technology Center on Real-Time Functional Imaging (STROBE) under Grant No. DMR-1548924, which included funds for building the DC photoemission test setup used in this work This work made use of the Cornell Center for Materials Research Shared Facilities which are supported through the NSF MRSEC program (DMR-1719875).

REFERENCES

[1] A. Polyakov *et al.*, "Plasmon-Enhanced Photocathode for High Brightness and High Repetition Rate X-Ray Sources," *Physical Review Letters*, vol. 110, no. 7, p. 076 802, 2013. doi:10.1103/PhysRevLett.110.076802

[2] D. B. Durham *et al.*, "Plasmonic lenses for tunable ultrafast electron emitters at the nanoscale," *Physical Review Applied*, vol. 12, no. 5, p. 054 057, 2019.

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doi:10.1103/PhysRevApplied.12.054057

[3] S. A. Maier, *Plasmonics: Fundamentals and Applications*. Springer, 2007. doi:10.1007/0-387-37825-1

[4] P. B. Johnson and R. W. Christy, "Optical Constants of the Noble Metals," *Physical Review B*, vol. 6, no. 12, pp. 4370–4379, 1972. doi:10.1103/PhysRevB.6.4370

[5] A. Polyakov, S. Cabrini, S. Dhuey, B. Harteneck, P. J. Schuck, and H. A. Padmore, "Plasmonic light trapping in nanostructured metal surfaces," *Applied Physics Letters*, vol. 98, no. 20, p. 203 104, 2011. doi:10.1063/1.3592567

[6] J. Le Perchec, P. Quémerais, A. Barbara, and T. López-Ríos, "Why Metallic Surfaces with Grooves a Few Nanometers Deep and Wide May Strongly Absorb Visible Light," *Physical Review Letters*, vol. 100, no. 6, p. 066 408, 2008. doi:10.1103/PhysRevLett.100.066408

[7] S. Zhang, H. Liu, and G. Mu, "Electromagnetic enhancement by a single nano-groove in metallic substrate," *Journal of the Optical Society of America A*, vol. 27, no. 7, p. 1555, 2010. doi:10.1364/JOSAA.27.001555

[8] *Lumerical, Finite difference time domain solutions*, 2011. http://www.lumerical.com

[9] I. V. Bazarov *et al.*, "Thermal emittance and response time measurements of negative electron affinity photocathodes," *Journal of Applied Physics*, vol. 103, no. 5, p. 054 901, 2008. doi:10.1063/1.2838209

[10] J. Vogelsang *et al.*, "Ultrafast Electron Emission from a Sharp Metal Nanotaper Driven by Adiabatic Nanofocusing of Surface Plasmons," *Nano Letters*, vol. 15, no. 7, pp. 4685–4691, 2015. doi:10.1021/acs.nanolett.5b01513