

STUDY OF NANO-STRUCTURED ELECTRON SOURCES USING PHOTOEMISSION ELECTRON MICROSCOPE

A. Kachwala*, M.M. Rizi, S. Karkare, Arizona State University, Tempe, AZ, USA
 D. Kim, E.I. Simakov, Los Alamos National Laboratory, Los Alamos, NM, USA
 D. Filippetto, Lawrence Berkeley National Laboratory, Berkeley, CA, USA
 O. Chubenko, Northern Illinois University, DeKalb, IL, USA
 C.M. Pierce, University of Chicago, Chicago, IL, USA
 J. Maxson, Cornell University, Ithaca, NY, USA

Abstract

Nanostructured electron sources exhibiting simultaneous spatio-temporal confinement to nanometer and femtosecond level along with a low emittance can be used for developing future ordered electron sources to generate unprecedented electron beam brightness and can revolutionize stroboscopic ultrafast electron scattering and steady-state electron microscopy applications. In addition, high current density electron beams generated from nanostructured electron sources can be used for applications that include nanoelectronics and dielectric laser accelerators. In this work, we report our efforts to develop and characterize two kinds of nanostructured electron sources: (i) nitrogen incorporated ultrananocrystalline diamond [(N)UNCD] pyramid tip cathode and (ii) plasmonic Archimedean spiral focusing lens. We demonstrate the ability to fabricate these cathodes and characterize them using a photoemission electron microscope under femtosecond laser illumination thereby demonstrating the ability of these structures to be used for next generation electron sources.

INTRODUCTION

High-density bunched electron beams have numerous applications in various fields such as research in basic sciences, [1] medicine [2] and X-ray generation [3]. Brightness of the electron bunch is a common figure of merit for all these applications. The brightness of the electron bunch is increased by reducing the emittance of the electron bunch [4] at the photocathode. The emittance at the photocathode can be reduced by reducing the emission area from the cathode. The emission area is set by the required bunch charge and the accelerating electric field or by the smallest spot size the laser can be focused to, often limited by the diffraction limit of light. In last few decades, several attempts have been made to reduce the emittance of the electron bunch by reducing the emission area of the photocathode. This involves laser assisted photoemission from nanometer sized tips, photocathodes which are driven by the plasmonic effects at metal-dielectric interface, work function engineered small emission areas to name a few [5, 6]. These nanostructured electron sources exhibiting simultaneous spatio-temporal confinement to nanometer and femtosecond level along with a low emittance can be used for developing future ordered electron sources to generate unprecedented electron

beam brightness and can revolutionize stroboscopic ultrafast electron scattering and steady-state electron microscopy applications. In addition, high current density electron beams generated from nanostructured electron sources can be used for applications that include nanoelectronics and dielectric laser accelerators (DLA) [7,8]. In this work, we report our efforts to develop and characterize two kinds of nanostructured electron sources: (i) nitrogen incorporated ultrananocrystalline diamond [(N)UNCD] pyramid tip cathode and (ii) plasmonic Archimedean spiral focusing lens.

(N)UNCD pyramid tip cathode with a nanometer-sized tip is considered a promising candidate for high peak current applications that include nanoelectronics and dielectric laser accelerators [7–9]. These cathodes have the ability to generate high current density, can withstand high laser intensities and have non-stringent vacuum requirements [5, 10]. However, the non-linear photoemission mechanism of (N)UNCD cathodes is not well understood [5]. In this work we aim to investigate non-linear photoemission from the (N)UNCD pyramid tip cathode using 800 nm, 150 fs laser at peak laser intensities ranging from 10^9 – 10^{10} W/cm² using Photoemission Electron Microscope (PEEM) [11]. This will aid in the better understanding of emission mechanisms from these photocathodes and further their use for high peak current and DLA applications.

It is possible to achieve a nanoscale emission area and femtosecond response time simultaneously using nanopatterned plasmonic cathodes. In such plasmonic cathodes, the surface is patterned at the nanometer scale to excite surface plasmon-polaritons [6]. The coupling of light and surface plasmon polariton results can result in highly concentrated optical fields resulting in multi-photon emission from a small 100 nm scale emission area. In this work, we present the design and fabrication of Archimedean spiral plasmonic structures that can act as plasmonic lenses to produce 100-nm scale emission areas for next-generation ultrafast electron sources. We demonstrate the ability of this structure to achieve ~140 nm electron spot size using FDTD simulation and ability to fabricate these structure using e-beam lithography (EBL).

(N)UNCD PYRAMID TIP PHOTOCATHODE

The diamond deposition process for synthesis of (N)UNCD pyramid tip cathode was performed by Advanced

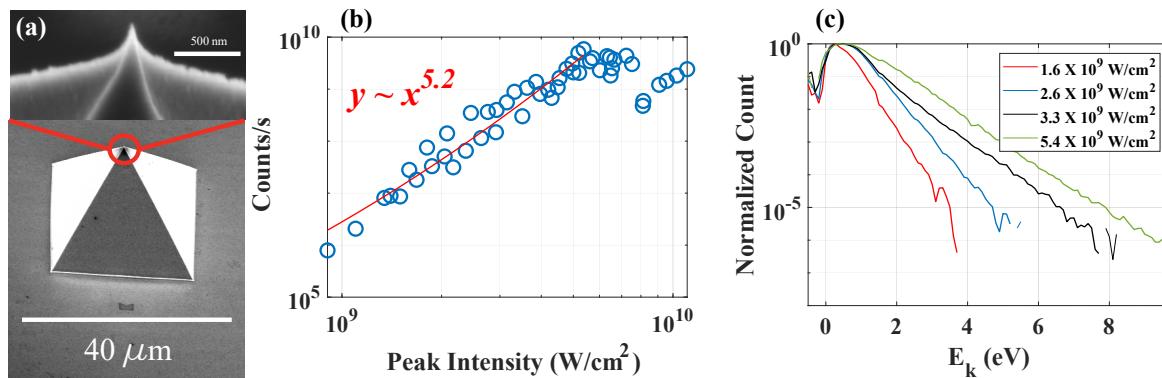


Figure 1: (a) Scanning Electron Microscope (SEM) image of the (N)UNCD pyramid tip photocathode, (b) counts per second as the function of peak laser intensity for (N)UNCD pyramid tip photocathode, (c) photoemission electron kinetic energy spectra for (N)UNCD pyramid tip photocathode.

Diamond Technologies. Details of different steps for the preparation of (N)UNCD are reported elsewhere [12]. Typically, it has 1 μm layer of conductive (N)UNCD film, followed by a layer of 15 μm microcrystalline diamond. Diamond film is brazed to a polished Molybdenum substrate using the brazing material TiCuSil. Scanning electron microscope images of the (N)UNCD pyramid tip photocathode that was tested in this particular experiment is shown in Fig. 1 (a). The photocathode was a diamond square with 3x3 array of pyramids. Each pyramid had a 25 μm base, and the spacing between pyramids was 1000 μm. There was an ultra-sharp tip (with diameter of ~15 nm) on the top of the pyramid as shown in Fig. 1 (a).

The non-linear photo-electron emission from the (N)UNCD pyramid tip cathode was investigated using a 800 nm (1.55 eV), 150 fs laser at peak laser intensities ranging from 10^9 to 10^{10} W/cm² using PEEM. The laser was directed on the sample at an angle of incidence 65° w.r.t the axis of the pyramid. The laser was p-polarized w.r.t the substrate. Fig. 1 (b) shows the counts/second from the (N)UNCD pyramid tip photocathode plotted as a function of peak laser intensity on a double logarithmic scale. Considering the work function (Φ) of (N)UNCD to be ~4.4 eV [13], using 1.55 eV photon energy should result in 3rd order photoemission. However, the exponent of the power dependence suggests a 5th order photoemission process. The 5th order process could be a result of 5th order multiphoton emission in which an electron absorbs 5 photons to get emitted or could be a result of strong-field photoemission in which electrons tunnel out of the surface due to the electric field of light concentrated at the tip [14]. For the case of the 5th order multiphoton emission process the maximum kinetic energy that the electrons can have is $(5 \times 1.55 \text{ eV}) - \Phi \approx 3.5 \text{ eV}$. Fig. 1(c) shows the kinetic energy distribution of the emitted electrons at various laser peak intensities. We see electrons with energies as high as 8 eV at larger powers, suggesting the existence of the strong field emission process.

Beyond peak intensity of $\sim 7 \times 10^9 \text{ W/cm}^2$ the counts/sec vs peak intensity curve flattens (Fig. 1(b)). The emission

spot observed in the PEEM also broadens in size. We attribute the broadening of emission spot and the counts/sec saturation to the Coulomb interaction between electrons emitted from the nano-tip and screening of the electric field by the electron cloud.

PLASMONIC PHOTOCATHODE

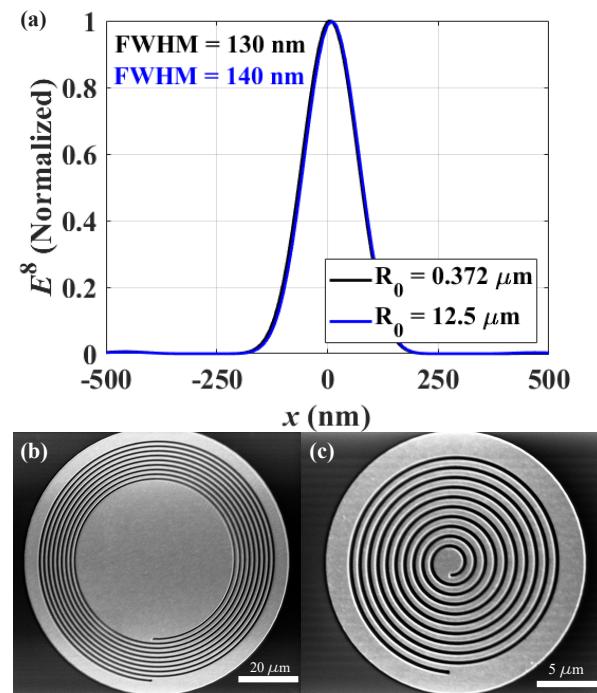


Figure 2: (a) Lumerical FDTD simulation showing E^8 enhancement at the center of the Archimedean spirals. Here R_0 is the start radius. Scanning Electron Microscope (SEM) image of Archimedean gold spiral on the Si substrate (b) with $R_0 = 12.5 \mu\text{m}$, (c) with $R_0 = 0.372 \mu\text{m}$.

As discussed earlier, the emittance of the emitted electron bunch can be reduced by reducing the emission area of the

electron bunch. One way to achieve the same is by using nano-structured Archimedean spiral photocathode. This is similar to the bullseye structure in Ref. [15] which is operated with radially polarized light. However, the Archimedean spiral structures enables the cathode to be operated with the circularly polarized light instead of the more difficult to obtain radially polarized light.

In our previous work, we designed and fabricated the spiral photocathode using a template stripping method with starting radius (R_0) of $0.372\text{ }\mu\text{m}$ [6]. These fabricated spirals were imaged using PEEM after illuminating the sample with $\lambda = 800\text{nm}$, 150 fs laser. However, due to fabrication non-uniformities, several points around the spiral edges were emitting electrons and the small emission area in the center was not observed. To circumvent this issue, we considered two approaches: (i) design spirals with large starting radius such that the fabrication non-uniformities are far away from the desired emission area and (ii) develop a fabrication process that would result in defect free spiral edges. Each of these is discussed in detail in the next two paragraphs.

The plasmonic cathodes are designed using gold for 1.55 eV of photon energy and are known to focus light to few hundred nanometers. Considering the work function of gold to be ~ 5.4 eV, using 1.55 eV of photon energy would result in 4^{th} order photoemission. According to Fowler-Dubridge theory, the current density for n^{th} order photoemission is directly proportional to I^n , where I is the intensity of the laser [16, 17]. In terms of the optical electric field, it is then given by E^{2n} . The numerical FDTD [18] simulation was performed for spirals with two starting radii, $R_0 = 0.372\text{ }\mu\text{m}$ and $R_0 = 12.5\text{ }\mu\text{m}$. Fig. 2(a) shows the E^8 field enhancement at the center of the spirals. As we can see, for both the cases, the electron emission spot size is approximately $\sim 140\text{ nm}$.

Next, we fabricated the spirals using EBL. Double-layer PMMA (185 nm 495 K + 200 nm 950 K) was spin-coated on a Silicon substrate. Then PEEM spiral patterns were written with EBL (JEOL JBX-6000FS). The samples were then developed in MIBK: IPA (1:3) solution for 90 s and rinsed with IPA. After that, the PMMA residue was removed with O_2 plasma (Plasma-Therm 790, O_2 10 sccm, 8 mT, 25W) for ~ 30 seconds. Next, Cr (5nm)/Au (120 nm) was deposited with E-beam evaporation, lifted off in remover PG, and rinsed with IPA. After fabrication, the spirals were imaged using SEM (FEI Nova 200). The SEM images of the fabricated spirals are shown in Fig. 2(b) and (c).

The next step involves demonstrating the electron emission spot size of $\sim 140\text{ nm}$ using PEEM. We also plan to fully characterize the emission from these plasmonic spirals by measuring the emission current density, photoemission electron energy spectra and MTE of the emitted electron beam from these spiral plasmonic photocathodes.

CONCLUSION AND FUTURE WORK

In this paper, we report our efforts to develop and characterize two kinds of nanostructured electron sources: (i) (N)UNCD pyramid tip cathode and (ii) plasmonic

Archimedean spiral photocathode. (N)UNCD pyramid tip cathode showed strong-field laser-induced emission from the nano-tip. The result obtained is consistent with the hypothesis that higher optical field enhancement would be achieved at sharper tips, leading to higher order non-linear photoemission. In other work, we performed the numerical FDTD [18] simulation for plasmonic Archimedean spiral photocathode with two starting radii, $R_0 = 0.372\text{ }\mu\text{m}$ and $R_0 = 12.5\text{ }\mu\text{m}$. For both the cases, the electron emission spot size is on the order of $\sim 140\text{ nm}$. Finally, we fabricated these spirals using EBL. The next steps involves demonstrating the electron emission spot size of $\sim 140\text{ nm}$ using PEEM. We also plan to measure the emission current density, photoemission electron energy spectra and MTE of the emitted electron beam from the Archimedean spiral plasmonic photocathodes for complete characterization.

ACKNOWLEDGEMENT

This work is supported by the NSF Center for Bright Beams under award PHY-1549132, and by the Department of Energy, Office of Science under awards DE-SC0021092, and DE-SC0021213 and the support of the Los Alamos National Laboratory (LANL) Laboratory Directed Research and Development (LDRD) Program. This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Los Alamos National Laboratory, an affirmative action equal opportunity employer, is managed by Triad National Security, LLC for the U.S. Department of Energy's NNSA, under contract 89233218CNA000001. C.M.P. acknowledges US NSF Award PHY-1549132, the Center for Bright Beams and the US DOE SCGSR program. 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.

REFERENCES

- [1] J. C. Nygard and R. F. Post, "Recent advances in high power microwave electron accelerators for physics research", *Nucl. Instrum. Methods*, vol. 11, pp. 126–135, Jan. 1961. doi:10.1016/0029-554X(61)90015-5
- [2] M. Oppelt *et al.*, "Comparison study of in vivo dose response to laser-driven versus conventional electron beam", *Radiat. Environ. Biophys.*, vol. 54, no. 2, pp. 155–166, Jan. 2015. doi:10.1007/s00411-014-0582-1
- [3] B. W. J. McNeil and N. R. Thompson, "X-ray free-electron lasers", *Nat. Photonics*, vol. 4, no. 12, pp. 814–821, Nov. 2010. doi:10.1038/nphoton.2010.239
- [4] I. V. Bazarov, B. M. Dunham, and C. K. Sinclair, "Maximum Achievable Beam Brightness from Photoinjectors", *Phys. Rev. Lett.*, vol. 102, no. 10, Mar. 11, 2009. doi:10.1103/physrevlett.102.104801
- [5] E. I. Simakov *et al.*, "Observations of the Femtosecond Laser-Induced Emission From the Diamond Field Emitter Tips", in *Proc. IPAC'19*, Melbourne, Australia, May 2019, pp. 2130–2133. doi:10.18429/JACoW-IPAC2019-TUPLTS089

[6] C. M. Pierce *et al.*, “Towards High Brightness from Plasmon-Enhanced Photoemitters”, in *Proc. NAPAC'22*, Albuquerque, New Mexico, USA, Aug. 2022, paper TUYD4, pp. 285.

[7] K. P. Wootton, J. McNeur, and K. J. Leedle, “Dielectric Laser Accelerators: Designs, Experiments, and Applications”, *Rev. Accel. Sci. Technol.*, vol. 09, pp. 105–126, Jan. 2016. doi:10.1142/s179362681630005x

[8] E. I. Simakov, H. L. Andrews, M. J. Herman, K. M. Hubbard, and E. Weis, “Diamond field emitter array cathodes and possibilities of employing additive manufacturing for dielectric laser accelerating structures”, *AIP Conference Proceedings*, 2017. doi:10.1063/1.4975877

[9] C.-K. Huang *et al.*, “Modeling of diamond field emitter arrays for a compact source of high brightness electron beams”, *J. Appl. Phys.*, vol. 125, no. 16, p. 164501, Apr. 28, 2019. doi:10.1063/1.5086292

[10] W. Yuan *et al.*, “Highly conductive nitrogen-doped ultrananocrystalline diamond films with enhanced field emission properties: triethylamine as a new nitrogen source”, *J. Mater. Chem. C*, vol. 4, no. 21, pp. 4778–4785, 2016. doi:10.1039/c6tc00087h

[11] FOCUS-IS-IEF-PEEM, <https://www.focus-gmbh.com/peem-nanoesca/peem/>

[12] D. Kim, H. L. Andrews, B. K. Choi, and E. I. Simakov, “Fabrication of Micron-Scale Diamond Field Emitter Arrays for Dielectric Laser Accelerators”, 2018 IEEE Advanced Accelerator Concepts Workshop (AAC), Aug. 2018. doi:10.1109/aac.2018.8659407

[13] A. Kachwala, O. Chubenko, D. Kim, E.I. Simakov, and S. Karkare. “Quantum efficiency, photoemission energy spectra, and mean transverse energy of ultrananocrystalline diamond photocathode”, *J. Appl. Phys.*, vol. 132, no. 22, p. 224901, 2022. doi:10.1063/5.0130114

[14] R. Bormann, M. Gulde, A. Weismann, S.V. Yalunin, and C. Ropers. “Tip-enhanced strong-field photoemission”, *Phys. Rev. Lett.*, vol. 105 no. 14, p. 147601, 2010. doi:10.1103/PhysRevLett.105.147601

[15] D.B. Durham, *et al.*, “Plasmonic lenses for tunable ultrafast electron emitters at the nanoscale”, *Phys. Rev. Applied*, vol. 12 no. 5, p. 054057. 2019. doi:10.1103/PhysRevApplied.12.054057

[16] R.H. Fowler, “The analysis of photoelectric sensitivity curves for clean metals at various temperatures”, *Phys. Rev.*, vol. 38, no. 1, p. 45, 1931. doi:10.1103/PhysRev.38.45

[17] L.A. DuBridge, “A further experimental test of Fowler's theory of photoelectric emission”, *Phys. Rev.*, vol. 39, no. 1, p. 108, 1932. doi:10.1103/PhysRev.39.108

[18] Ansys-Lumerical-FDTD, <https://www.ansys.com>