CHARACTERIZATION OF FEMTOSECOND-LASER-INDUCED ELECTRON EMISSION FROM DIAMOND NANO-TIPS*

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Abstract

Nanocrystalline diamond is a promising material for electron emission applications, as it combines robustness of diamond and ability to easily conform to a pre-defined shape, even at nano-scale. However, its electron emission properties are yet to be fully understood. Recently, we started to investigate femtosecond-laser-induced strongfield photoemission from nanocrystalline diamond field emitters with very sharp (~10 nm apex) tips. Initial results show that the mechanism of electron emission at $\sim 10^{10}$ W/cm² light intensities in the near UV to near IR range is more complex than in metals. We present our latest experimental results obtained at Stanford University, while LANL's strong-field photoemission test stand is being commissioned. We show that strong-field photoemission occurs not only at the nano-tip's apex, but also on flat diamond surfaces (e.g., pyramid sides), that is why extra care needs to be taken to differentiate between emission spots on the chip. Qualitatively, we discuss the models that explain the observed dependences of electron emission on the optical power, polarization of the light, etc.

INTRODUCTION

This work supports development of a dielectric laser accelerator (DLA) [1], which will benefit from a very compact laser-triggered electron source. Diamond nano-tips are attractive candidates for that role [2], and a manufacturing technique for such samples has been fully adopted and refined by LANL. Experimental characterization of the strong-field photoemission from diamond nano-tips was conducted jointly with Stanford University DLA laboratory, within the framework of the Accelerator on a Chip International Collaboration (ACHIP) [3].

DIAMOND SAMPLES

Diamond nano-tip samples were fabricated as diamond field emitter arrays (DFEA) [4, 5]. Currently, manufacturing technology cannot produce equally sharp tips, even within one chip. Prior to testing, sharpest tips with the apex curvature radius of ~10 nm are identified by SEM, because those supposedly produce high emission currents as both pDC and optical field enhancement factor is expected to be larger for sharper tips. Different array configurations were manufactured, including linear arrays on a narrow (about 300 µm wide) substrate (Fig. 1).



Figure 1: SEM image of a linear nano-tip array (Cu-03), top view. Pyramids have $25 \ \mu m$ base side length.

EXPERIMENTAL SETUP

The layout and photo of LANL's strong-field photoemission test stand is shown in Fig. 2. The layout is based on Stanford University's test stand with some modifications intended to make the system more flexible and modular. Experimental procedure outlined below is universal with respect to different test stands, the actual measurements' results reported here were obtained at Stanford University.

Laser parameters need to be carefully considered in the design of the beam line so that the intensity of the light at the focal spot can achieve $\sim 10^{10}$ W/cm², where strong-field photoemission is typically observed [6].

Characteristics of individual diamond nano-tips are obtained by focusing the laser light onto one tip at a time, which is established by observing the scattered laser light with a sample microscope camera and fine-tuning the laser focal spot position to achieve maximum electron emission current. Typically, for a setup that employs a small vacuum chamber and a focusing lens just outside of a vacuum viewport, one can focus a 5 mm diameter laser beam down to several tens of microns, which is commensurate with a DFEA pyramid's size (Fig. 1). Larger DFEA chip sizes (e.g., 5x5 mm²), necessitate the use of oblique angle of incidence of 5-10 degrees with respect to the substrate plane in order to avoid incident laser beam being partially occluded by the substrate, while narrow substrates (Fig. 1) allow laser beam alignment parallel to the substrate.

DC field emission without laser illumination is used to align a nano-tip with the anode axis, subsequent strong-

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field photoemission measurements are performed with accelerating gradients below the field emission threshold.

Images formed on the fluorescent screen after the microchannel plate (MCP) intensifier are captured by the camera, calibration of image intensity vs electron current is performed with the help of intermittent current measurements through the Faraday cup at different MCP gains.



Figure 2: Scheme (top) and photo (bottom) of LANL's strong-field photoemission test stand.

LANL's strong-field photoemission test stand (Fig. 2) that is currently being commissioned employs a 1 MHz repetition rate laser operating at 1035 nm [7] (Fig. 3), which would increase the average emission current under any given conditions by a factor of 10. It is a significant improvement considering technical difficulties in measuring currents down to sub-femtoAmpere levels reported here, with all datasets obtained at 100 kHz laser repetition rate.



Figure 3: LANL's newly commissioned femtosecond laser.

EMISSION FROM FLAT DIAMOND SURFACES

Although the main purpose of this study is to contribute to understanding of the strong-field photoemission from diamond nano-tips, recently at Stanford University DLA laboratory we observed strong-field photoemission from flat nanocrystalline diamond, particularly from pyramid sides. In general, when a laser spot is swept across a DFEA sample, emission pattern changes greatly in response to instantaneous position of the focal spot. For example, when the laser is focused on flat surface between the pyramids, relatively weak electron emission is typically observed, consistent with high reflectivity of the substrate at a near grazing (5-10 degrees) angle of incidence. When a focused spot has a significant overlap with a pyramid, much stronger emission is observed, having a distinct pattern consisting of up to four approximately center-symmetrical lobes, sometimes accompanied by a center beamlet, presumably associated with emission from a nano-tip. We confirmed rotation of the pattern along with the rotation of sample; positions of the lobes correspond to the sides of the pyramid, not its edges.

Fine fringed structure is often observed on the lobes, with the spacing between the fringes approximately proportional to the laser wavelength (Fig. 4). Coincidentally, prolonged exposure of the pyramids to the high intensity laser beam would cause a specific damage on the pyramids' side surface, featuring several horizontal stripes [8]. We attribute fringed emission structure of the lobes and horizontal damaged stripes to formation of a standing wave in the region of interference between the incident laser beam and the one reflected from the substrate.



Figure 4: Beam patterns registered by MCP screen that show optical interference effects on emitted electron beam profile for different wavelengths.

POLARIZATION DEPENDENCE

While exact alignment of the optical electric field with a nano-tip's axis cannot be achieved with oblique angle of incidence, qualitatively the observed dependence of a nano-tip's emission on the polarization of the incident laser light is in agreement with previous reports for metal nano-tips [6], featuring sharp peaks and extended flatter valleys and 180 degrees periodicity [8].

However, as compared with the dependence for a metal nano-tip, the dependence appears distorted. We attribute the origin of the distortion to a fine structure of the electron beam profile: the beams are somewhat hollow and asymNorth American Particle Acc. Conf. ISBN: 978-3-95450-223-3

metrical, and separate beamlets can be identified in the pattern with somewhat different polarization dependences, their superposition giving rise to "distorted" polarization dependence for the total current. In turn, we attribute the fine structure of beam profiles to irregularities of each individual tip's geometry and/or composition, which will require further investigation.

POWER DEPENDENCE

Extension of Keldysh theory of strong-field photoionization [9] onto photoemission from metals fairly well describes power dependences of electron emission from tungsten nano-tips [10]. Low-power ends of such dependences typically exhibit well-defined slopes in accordance with the order of the multi-photon excitation, i.e., the ratio of the work function and the photon energy.

Figure 5 shows power dependences of the electron yield measured on a diamond nano-tip for several wavelengths. Unlike metal nano-tips, no systematic dependence of the low-end slope on the photon energy is observed, with all slopes falling in the range between 8- and 12-photon orders (dashed lines in Fig. 5). We are aware of one report on diamond-coated tungsten nano-tips [11] that interprets deviation from existing strong-field photoemission models through the notion of different "channels" of photoemission for different photon energies, but that approach could not explain our data. So far, interpretation of the power dependences reported here remains inconclusive.





In order to understand the origin of the roll-off observed at the higher power end of the dependences in Fig. 5, we studied the evolution of the beam profile with the increasing power (Fig. 6). While the current is rapidly increasing è with the optical power, in the beginning the spot size remav mains approximately constant, and a fine structure of the work beam mentioned above is clearly visible (Fig. 6, left insert). The spot size starts to grow rapidly above ~10 electhis trons/shot, and the beam profile becomes very smooth and from Gaussian-like (Fig. 6, right insert). Therefore, we attribute roll-off region of the dependence to Coulomb interaction Conten between electrons emitted from the nano-tip and screening of the electric field by their cloud, analogous to Child-Langmuir law for flat cathodes.



Figure 6: Power dependences of electron yield and beam size for Mo45 1-3 nano-tip at 1024 nm laser wavelength. Inserts: beam profiles at low and high laser intensities.

CONCLUSIONS

This paper represents an update on LANL project "New Science and Technology for a Tabletop Accelerator". Strong-field photoemission test stand is being commissioned at LANL and will become operational in September. We reported the results of the measurements of strong-field photoemission from diamond nano-tips, fabricated as DFEA chips, conducted at Stanford University. The range of the laser wavelengths in this study was between 512 nm and 2 µm, as compared with nanocystalline diamond photoemission threshold of about 230 nm. Sharpest nano-tips usually produce strongest electron emission, so pre-selection of the tips based on their SEM images is found to be useful. Laser light polarization dependences of electron emission are in qualitative agreement with previous reports on metal nano-tips. Optical power dependences of electron emission can be interpreted by Coulomb interaction between electrons at high currents, while interpretation of the lower end by multi-photon electron excitation does not provide satisfactory agreement with the data and thus will require further studies. Additionally, strong-field photoemission was observed from flat diamond surfaces, with the features indicative of optical interference effects between incident and reflected laser beams in the vicinity of the substrate. We plan to use the new LANL test stand to determine whether the power law exponent is constant at very low light intensity levels or the power dependence follows a different law, which should facilitate understanding and modeling of strong-filed photoemission from diamond nano-tips.

RFERENCES

- E. A. Peralta *et al.*, "Demonstration of electron acceleration in a laser-driven dielectric microstructure", *Nature*, vol. 503, p. 91-94, 2013.
- [2] Evgenya I. Simakov *et al.*, "Diamond field emitter array cathodes and possibilities of employing additive manufacturing for dielectric laser accelerating structures", *AIP Conference Proceedings*, vol. 1812, p.060010, 2017.
- [3] Accelerator on a Chip International Collaboration, https://achip.stanford.edu.
- [4] J. D. Jarvis *et al.*, "Uniformity conditions of diamond field emitter arrays", *Vac. Sci. Technol. B.*, vol. 27, no. 5, p.2264, 2009.
- [5] D. Kim *et al.*, "Fabrication of micron-scale diamond field emitter arrays for dielectric laser accelerators", in *Proc.* AAC'18, Breckenridge, Colorado, US, Aug. 2018, pp. 1-3.
- [6] Peter Hommelhoff *et al.*, "Field emission Tip as a Nanometer Source of free electron femtosecond pulses," *Phys. Rev. Lett.* vol. 96, no 7, p. 076601, 2006.
- [7] Monaco 1035 Industrial Femtosecond Laser, https://www.coherent.com/assets/pdf/COHR_Monaco1035_DS_0318_5.pdf
- [8] E. I. Simakov et al., "Observations of the Femtosecond Laser-Induced Emission From the Diamond Field Emitter Tips", in Proc. 10th Int. Particle Accelerator Conf. (IPAC'19), Melbourne, Australia, May 2019, pp. 2130-2133.
 - doi:10.18429/JACoW-IPAC2019-TUPTS089
- [9] L.V. Keldysh, "Ionization in the field of a strong electromagnetic wave", Sov. Phys. JETP, vol. 20, no. 5, p.1307 1965.
- [10] C. Ropers *et al.*, "Localized multiphoton emission of femtosecond electron pulses from metal nanotips", *Phys. Rev. Lett.*,vol. 98, no. 4 p.043907, 2007.
- [11] A. Tafel et al., "Femtosecond laser-induced electron emission from nanodiamon-coated tungsten needle tips", 2019 https://arxiv.org/pdf/1903.05560.pdf