

CHARACTERIZATION OF POLARIZATION-DEPENDENT EMITTANCE FROM AN ARRAY OF AU NANORODS USING VELOCITY-MAP-IMAGING SPECTROMETER*

Hong Ye^{†1,2,3}, Arya Fallahi¹, Sebastian Trippel^{1,3}, Giulio Maria Rossi¹,
Oliver D. Mücke^{1,2}, Jochen Küpper^{1,2,3}, and Franz X. Kärtner^{1,2,3}

¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY,
22607 Hamburg, Germany

²Department of Physics, Universität Hamburg, 22761 Hamburg, Germany

³The Hamburg Center for Ultrafast Imaging, Universität Hamburg,
22761 Hamburg, Germany

Abstract

Electron beams of high quality, e. g., low emittance, are of crucial importance for cutting-edge scientific instruments, such as x-ray free-electron lasers (XFELs) and ultrafast electron diffraction (UED) setups. A velocity-map-imaging (VMI) spectrometer was implemented to characterize the intrinsic root-mean-square (rms) normalized emittance from photocathodes. The spectrometer operated in both, spatial map imaging (SMI) and VMI modes. Therefore, spatial- and velocity-coordinates were recorded independently. The technique allows for fast complete emittance measurements within minutes. A 75-μm pitch array of Au nanorods of dimension 100 × 30 nm, was studied under strong-field emission regime by 100-fs 1-kHz 1.3-μm laser pulses. A patterned electron bunch was observed, each beamlet emitted from a single nanorod within the array. A polarization-dependence photoemission study was performed showing a smaller rms normalized divergence of 0.8 mrad with the laser polarization normal to the sample surface, compared to 1.15 mrad for the parallel case.

INTRODUCTION

Electron beams with ultra-low emittance are paramount for advanced applications, such as x-ray free-electron lasers (XFELs) [1], ultrafast electron diffraction (UED) experiments [2] and inverse Compton scattering (ICS) systems, to increase source brightness. Normalized transverse emittance (NTE) of an electron beam, according to the definition $\epsilon_{n,x} = \frac{\sqrt{\langle x^2 \rangle \langle x'^2 \rangle}}{mc}$, strongly depends on both the electron beam size $\langle x^2 \rangle$ and divergence $\langle x'^2 \rangle$. Photoemission from nanostructures is of particular interest due to its high potential to pursue ultra-low emittance electron beams [3]. The

nanostructured emitter geometrically limits the emission area and induces the local field enhancement of the laser field to approach the strong-field regime at intensities well below the damage threshold.

A velocity-map-imaging (VMI) spectrometer with both spatial- and velocity-mapping capabilities, which measure $\langle x^2 \rangle$ and $\langle x'^2 \rangle$, respectively, allows the direct characterization of the NTE of various photocathodes within minutes [4]. Utilizing this technique, in this paper, we characterized the NTE of the electron bunches photoemitted from a field-emitter array of Au nanorods spaced by 75 μm. The ultrafast laser pulses obliquely illuminated the field-emitter array. An array of electron bunches, consistent with the structure of the field-emitter array, is detected under the spatial-map-imaging (SMI) mode. In the velocity map imaging mode, we observe the variation in velocity distribution of photoemitted electrons upon changing the polarization of the incident laser beam. Assuming that the emission area is restricted to the physical dimensions of the nanorod, an ultra-low, sub-nm, NTE is deduced for strong-field emission from an individual nanorod.

EXPERIMENTAL LAYOUT

Figure 1(a) shows the configuration of the VMI spectrometer. The spectrometer employs three electrodes, named repeller (R), extractor (E) and ground (G), to perform the electrostatic imaging. During the experiments, the spatial- or velocity-map imaging mode is adjusted using the corresponding extractor voltages (E: -5580 V for SMI mode and E: -4880 V for VMI mode, respectively) while the repeller and ground voltages (R: -6000 V and G: 0 V) are kept fixed. The whole configuration is shielded using μ-metal to minimize the influence of the stray electromagnetic fields on the photoemitted electrons. To be able to measure the nascent electron spatial/velocity coordinates and minimize the scattering effects on the flight to the detector, the experiments are performed in ultra-high vacuum (UHV) of 10⁻¹⁰ mbar. The detector system consists of a position-sensitive micro channel plate (MCP) combined with a phosphor screen fluorescing when impacted by electrons followed by a CMOS camera for recording of the fluorescent images. In order to

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[†] hong.ye@desy.de

improve the signal-to-noise ratio, the MCP was gated by a high-voltage switch with a duration of 150 ns.

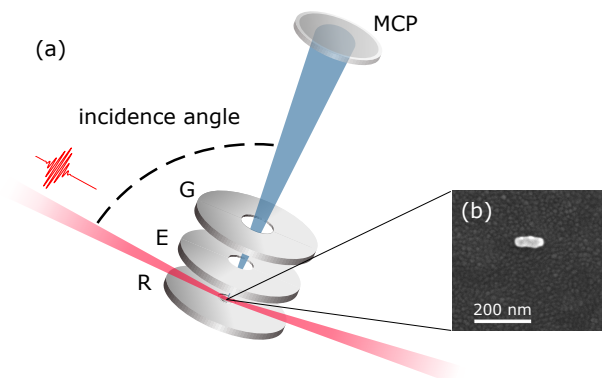


Figure 1: (a) VMI spectrometer consisting of three electrodes: repeller (R), extractor (E) and ground (G); (b) SEM image of the 75-μm pitch field-emitter array of Au nanorods.

The field-emitter array was mounted flat on the sample holder which is centered and contacted with the repeller plate to stay at the same potential, i. e., -6000 V, and to preserve the field configuration for the electrostatic imaging. A scanning electron microscope (SEM) image of the 75-μm pitch field-emitter array of Au nanorods resting on a 30-nm thick indium tin oxide (ITO) layer is shown in Fig. 1(b). The dimension of the nanorod itself is 100 nm in length and 30 nm in width. During the experiments, the field-emitter array was excited with a femtosecond laser pulse with 100 fs pulse duration, at a center wavelength of 1.3 μm and a repetition rate of 1 kHz. The femtosecond pulses were tightly focused onto the sample surface with a glancing incidence angle of ~84°, resulting in a projected intensity spot size of approximately 300 × 30 μm² on the sample.

EXPERIMENTAL RESULTS

Operating the spectrometer in the SMI mode, a spatially patterned electron bunch was observed (see Fig. 2(a)). The entire emission area on the sample is in accordance with the laser focus spot size, which covers five field emitters. Clearly, the active emission area is limited to the nanostructured field emitters. The spatial-mapping image indicates that each electron bunch was photoemitted from a single nanorod within the field-emitter array and therefore spaced by 75 μm. During the experiments, we varied the polarization of the incident laser to investigate the velocity distribution of photoemitted electrons. The direction of the laser polarization is indicated in Fig. 2(b). The optical wave propagates along the long side of the nanorod. The s-polarization is defined when the laser electric field direction is parallel to the nanorod top surface, so to the detector plane, while the p-polarization denotes that the field at the interaction point is normal to it. Moreover, we shifted the laser focus position on the field-emitter array to study the sensitivity of the photoemission from the nanostructured emitters. Figures 2(c)-(f) display the electron velocity-mapping images

of two different positions, which are ~150 μm apart, under both p-polarization (marked “VMI-p”) and s-polarization (marked “VMI-s”). All of the velocity-mapping images were integrated over 6×10^4 laser shots. We observe dramatical changes in the quantum efficiency, strongly related to the local nano-emitters. Fortunately, the rms normalized divergence for the two positions are characterized and basically the same, which are 0.8 mrad and 1.15 mrad for p- and s-polarizations, respectively. From the spatial image, it is evident that the electrons are emitted from the nanorod area which is geometrically limited to $100 \times 30 \text{ nm}^2$. Therefore, the normalized transverse emittance of a beamlet from a single nanorod under p-polarization indicates that $\varepsilon_x = 0.08 \text{ nm-rad}$ and $\varepsilon_y = 0.024 \text{ nm-rad}$ is experimentally achievable.

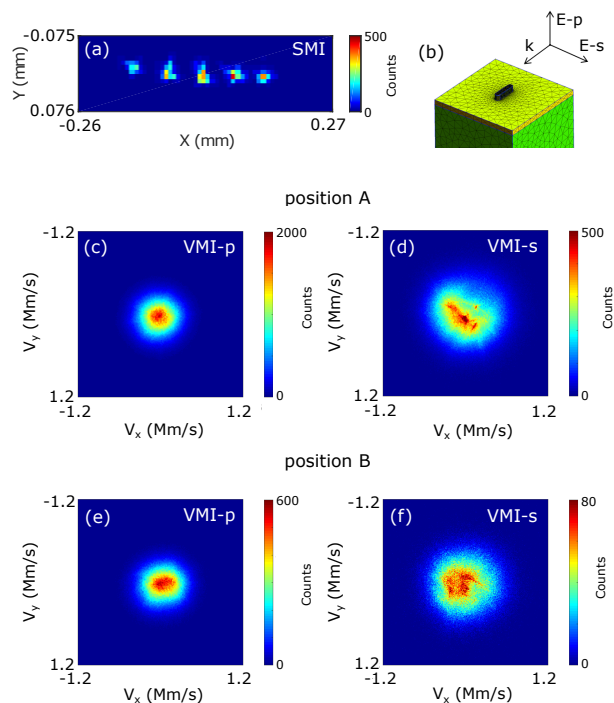


Figure 2: (a) SMI image of a 75-μm pitch field-emitter array of Au nanorods; (b) sketch of laser polarization with respect to the experiment, where E-p and E-s denote the electric field directions under p- and s-polarization, respectively; VMI images of position A under (c) p-polarization and (d) s-polarization; VMI images of position B under (e) p-polarization and (f) s-polarization.

With the laser peak intensity of $2.4 \times 10^9 \text{ W/cm}^2$ (referring to the projected focus spot size) used during the mapping and a field enhancement factor of ~5, the Keldysh parameter is 2.38, implying the emission approaches the tunneling regime. Therefore, the electrons are steered by the laser field upon photoemission. In addition, the maximum number of electrons generated per pulse during the experiments is in the order of one. Therefore, the space charge effects can be excluded. For the s-polarized laser field, the electric field results in a gain of the transverse momentum of photoemit-

ted electrons, giving rise to an increased velocity spread in the transverse direction, which leads to broader transverse velocity distributions. We believe that the VMI images taken under illumination with s-polarized light contain also the information of the nanorod morphology, which needs further investigation. The acceleration in the transverse direction amplifies the effect of photoelectrons emitted from the edges of the nanorods on the velocity distributions. Therefore, the VMI images slightly differ from each other. In contrast, for a linearly p-polarized laser field, the VMI images of two positions exhibit great similarity. As there is a field component perpendicular to the substrate surface, which accelerates electrons towards the detector and the transverse component of the field is strongly reduced between the input and the output laser fields, therefore, electrons photoemitted only gain an additional longitudinal momentum from the laser field. With the transverse momentum kept as initialized, the divergence angle of photoelectrons is decreased, and therefore leads to a smaller normalized emittance.

CONCLUSION

We have reported the spatial- and velocity-map imaging characterization of normalized emittance of a 75- μm pitch field-emitter array of Au nanorods and derived from that the normalized transverse emittance for the different cases. The experimental results show that the field emission under p-polarization from a nano-emitter is promising for improving the emittance. Future interest would be on the further understanding of photoemission process and advanced control

of the liberated electrons by femtosecond laser pulses [5, 6], which benefits not only FEL facilities but also attosecond science and ultrafast electron holography.

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REFERENCES

- [1] B. W. J. McNeil, and N. R. Thompson, X-ray free-electron lasers, *Nature Photonics* **4**, 814 (2010).
- [2] G. Mourou, and S. Williamson, Picosecond electron diffraction, *Applied Physics Letters* **41**, 44 (1982).
- [3] S. Tsujino, P. Das Kanungo, M. Monshipouri, C. Lee, and D. R. J. Miller, Measurement of transverse emittance and coherence of double-gate field emitter array cathodes, *Nature Communications* **7**, 13976 (2016).
- [4] H. Ye, S. Trippel, M. Di Fraia, A. Fallahi, O. D. Mücke, F. X. Kärtner, and J. Küpper, Velocity-map imaging for emittance characterization of multiphoton-emitted electrons from a gold surface, *Physical Review Applied* **9**, 044018 (2018).
- [5] M. Krüger, M. Schenk, and P. Hommelhoff, Attosecond control of electrons emitted from a nanoscale metal tip, *Nature* **475**, 78 (2011).
- [6] W. P. Putnam, R. G. Hobbs, P. D. Keathley, K. K. Berggren, and F. X. Kärtner, Optical-field-controlled photoemission from plasmonic nanoparticles, *Nature Physics* **13**, 335 (2016).