Nanofabrication and Characterization of Ultrafast Plasmonic Au Nanorod Array Photocathodes

R. G. Hobbs[1], Y. Yang[1], W. P. Putnam[1], P. D. Keathley[1], E. De Leo[1], William S. Graves[1], F. X. Kärtner[1,2], & K. K. Berggren[1]

[1]Massachusetts Institute of Technology, Cambridge, MA 02139
[2]Center for Free-Electron Laser Science, DESY & Dept. of Physics, University of Hamburg, Notkestraße 85, D-22607 Hamburg, Germany
rhobbs@mit.edu

Optically actuated plasmonic nanostructured electron emitter arrays may facilitate the development of ultrafast, ultra-bright photocathodes for next-generation free-electron lasers (FELs) as well as for time-resolved electron microscopy, diffraction, and spectroscopy. [1–4] Nano-optical field enhancement by localized surface plasmons in metallic nanoparticles allows the production of strong localized surface fields on the nanoscale. Such localized surface fields result in electron emission from a minimal area, thereby maximizing electron source brightness. Operation of plasmon-enhanced photocathodes with femtosecond pulsed optical control fields will thus enable the development of tools for x-ray and electron analysis of materials with unparalleled spatial and temporal resolution.

In this work we have fabricated arrays of plasmonic Au nanorods with critical dimensions below 20 nm by electron beam lithography on indium-doped tin oxide (ITO) coated substrates, as shown in Fig. 1. We have excited electron emission from Au nanorods using ultrafast pulses of 800 nm light, with pulse durations as short as 7 fs, and repetition rates as high as 84 MHz. We have studied the emission current as a function of incident laser-pulse energy (Fig. 2 (a)), applied static field, and nanorod array density (Fig. 2 (b)). A transition in the emission mechanism with increasing laser intensity, from a multiphoton absorption process, to strong-field tunneling has been observed, as have space-charge effects with increasing emission current. Additionally we have investigated the timescale of the emission process by interferometric autocorrelation (Fig. 3), and have observed the emission to be prompt *i.e.* on the timescale of the femtosecond optical pulse used to excite emission. Consequently, arrays of plasmonic Au nanorods are promising candidates for applications as high-brightness, ultrafast electron and x-ray sources.

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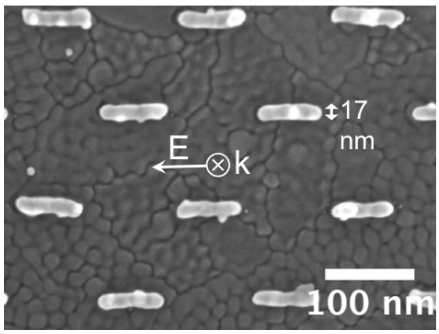
***Figure 1**: SEM image of a Au nanorod array on an 80 nm film of ITO on a sapphire substrate. Orientation of the linear polarization (E) of the light used to excite electron emission is depicted by the arrow as shown, as is the direction of light propagation (k) into the plane of the ITO film.*



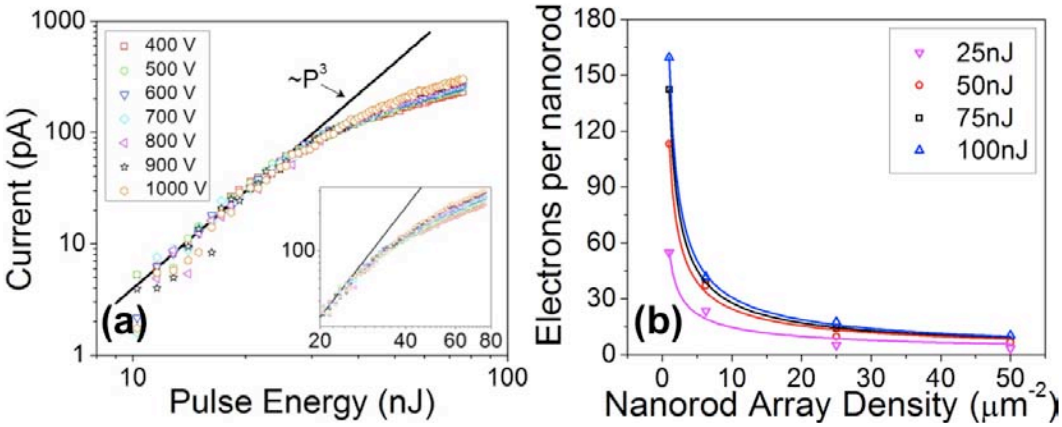


***Figure 2**: (a) Plot of emission current vs pulse energy at various anode bias values, for a 1 μm pitch square array of Au nanorods illuminated by a 90 μm (FWHM) beam of 35 fs pulses of 800 nm light (3 kHz rep. rate). Current scales approximately with the third power of the laser intensity for all bias values up to 27 nJ at which point all data deviates from the power-law scaling irrespective of applied bias, suggesting a transition to strong-field tunneling-like emission. Inset shows a closer view of the point of deviation from the power-law. (b) Plot of electron emission yield per nanorod per pulse vs nanorod array density at four different laser pulse energies (intensities) and a fixed applied static field of ~1 MVm^{-1}. The relationship between electron yield (N), and array density (ϱ), approaches that of $N \propto 1/\varrho$ with increasing laser intensity. Color-coordinated lines represent best fits of the data to a power law ($N \propto \varrho^n$).*

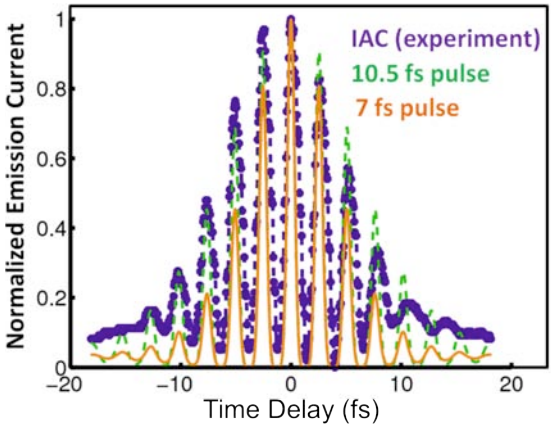


***Figure 3**: Interferometric autocorrelation (IAC) trace of normalized electron emission current vs time-delay for electron emission from a 200 nm × 100 nm rectangular array of Au nanorods (purple dots). IAC trace for optical pulse used to excite emission from Au nanorod array consistent with a 7 fs FWHM (orange line). Dashed line represents the simulated IAC trace for a 10.5 fs pulse. The observed broadening in the IAC trace by the Au nanorods may be due to damping of the localized surface plasmon mode in the nanorods.*