Ultrafast electron emission from metallic nanotip arrays induced by near infrared femtosecond laser pulses

S. Tsujino,^{1,a)} P. Beaud,¹ E. Kirk,¹ T. Vogel,¹ H. Sehr,¹ J. Gobrecht,¹ and A. Wrulich²

¹Department of Synchrotron Radiation and Nanotechnology, Paul Scherrer Institut, CH-5232 Villigen-PSI, Switzerland

²PSI XFEL, Paul Scherrer Institut, CH-5232 Villigen-PSI, Switzerland

(Received 2 April 2008; accepted 19 April 2008; published online 12 May 2008)

The authors explore the impact of femtosecond light pulses on the field-emission properties of single-gate molybdenum field-emitter arrays with nanometer scale tip apex. Despite the small fraction of the emission area, we observed a single-photon photoelectric current from the emitter tips on top of the dc field-emission current under the irradiation of 50 fs laser pulses at a wavelength of 800 nm with an external quantum efficiency up to $\sim 2 \times 10^{-7}$ and the emitter tip quantum efficiency of $\sim 10^{-2}$. The result indicates that metallic field-emitter arrays are promising for applications that require high-brightness short electron beams. © 2008 American Institute of Physics. [DOI: 10.1063/1.2924290]

Laser-induced photoemission in metallic needle cathodes with nanometer-scale sharpness has attracted attention to generate bright, low-emittance, and short electron bunches with durations down to the femtosecond range, ideal for applications such as the time-resolved electron microscopy,¹ compact free-electron lasers,² or scanning probe microscopy.³ Small apex curvatures facilitate the application of an intense dc electric field in the order of 1–10 GV/m at the emitter apex with practical voltages to lower the work function substantially by the Schottky effect. Combined with the dc field, electron bunches in the femtosecond time scale can be generated by irradiating the needle cathodes by near infrared femtosecond light pulses.^{4–7} However, the small emission area of the needle cathodes restricts its applicability to certain high current applications such as the rf power amplifiers and the short-wavelength free-electron lasers.

The field-emitter arrays (FEAs) realized by Spindt⁸ can emit large current by applying low gate bias voltages of ~ 100 V or below. Several authors have pointed out the potential advantage of FEAs for the microwave power amplifier applications but the high current operation at frequencies much higher than ~ 10 GHz has remained to be a challenging task.^{9–11} The high speed switching and photoresponse of silicon FEAs have been explored in detail^{12–15} and the carrier-transport-limited switching down to ~ 100 ps was recently reported.¹⁵ Compared with semiconductor FEAs, metallic FEAs are more robust and potentially more advantageous for the generation of even faster electron bunches with high emission current. However, only a limited number of authors have studied the effect of the laser irradiation on metallic FEAs so far.¹⁶

In this letter, we investigate the field-emission properties of molybdenum FEAs with single-gate electrodes under the irradiation of intense femtosecond light pulses in near infrared. Despite the small fraction of the electron emission area, we observe a single-photon photoelectric current from the emitter tips with a high quantum efficiency, superposed on the dc field-emission current.

The FEA devices (Fig. 1) used for the experiment are fabricated in combination of the molding technique and the self-aligned gate process. The field-emitter and the gate electrode material consist of molybdenum deposited by sputtering at room temperature. The pyramidal-shaped field-emitter structures are aligned in a square or a hexagonal array with 5–10 μm period. The substrate of the emitter structures consists of a 500 μm thick electroplated nickel plate. The distance from the emitter tip to the edge of the gate aperture hole is ~ 1.2 μm . The apex size of the emitters (inset of Fig. 1) is ~ 20 nm in diameter.¹⁷

In the experiment, we measured several FEA devices having 39×39 tip square arrays aligned with 5 μm period (total array area of 200 μm^2). An FEA was mounted on a copper holder and the gate electrode is wire bonded to an insulated electrode. Then the device is loaded in the vacuum chamber which is subsequently evacuated down to the base pressure below 5×10^{-9} mbar. A positive bias voltage is applied to the gate electrode in dc while the substrate and the emitter arrays were held at ground potential. The emitted electrons are collected by a cylindrical shape anode with a diameter of 15 mm, placed in front of FEAs with a separa-

FIG. 1. Scanning electron microscope images of the Mo field-emitters aligned with a 5 μm period. The gate electrode is a 0.5 μm thick Mo film separated from the emitter substrate by a 1.2 μm thick SiO_2 film. Individual emitters are ~ 1.3 μm tall. The inset is the apex of an emitter imaged from the top at higher magnification, showing that the apex diameter is equal to ~ 20 nm.

^{a)}Electronic mail: soichiro.tsujino@psi.ch.

FIG. 2. (Color online) (a) dc field-emission current I_{dc} (filled squares) and the photoelectric signals vs gate bias voltages when the FEA is irradiated at 25 GW/cm² (filled circles), and at 83 GW/cm² (empty circles). (b) The FN plot of I_{dc} (squares) and of the photoelectric signals at these two laser intensities. The linear slope of the relation between $\ln(I_{dc}/V_g^2)$ and $1/V_g$ suggests that the emitter apex field is given by βV_g with β equal to $(5-10) \times 10^7$ m⁻¹.

tion of ~ 20 nm. The anode is dc biased at 1 kV while monitoring the dc current. The ac anode current was detected by a load resistor of 99.2 Ω , amplified, and monitored by a digital oscilloscope. Most of the data were collected by using a 60 dB gain voltage amplifier with 10 MHz bandwidth. An amplifier with 2 GHz bandwidth was used optionally. When the gate bias voltage V_g is increased above ~ 50 V, the device starts to emit a dc field-emission current I_{dc} (Fig. 2) which reaches 0.5–1 mA when increasing V_g to 140–145 V. The relation between I_{dc} and V_g is described well by the empirical Fowler–Nordheim (FN) equation. This is indicated by the linear FN plot in Fig. 2(b), i.e., the relation between $\ln(I_{dc}/V_g^2)$ and $1/V_g$ is approximately linear.¹⁸ The background pressure increased up to $(1-4) \times 10^{-8}$ mbar when operating the FEA at high emission current due to the electron bombardment of the anode.

The near infrared pulses are generated by a Ti:sapphire chirped pulse amplifier system delivering ultrashort laser pulses of 50 fs duration centered at a wavelength of 800 nm at a repetition rate of 2 kHz. The linearly polarized laser beam is focused onto the sample to a spot size of $320 \times 690 \mu\text{m}^2$ full width at half maximum with an incidence angle of $63^\circ \pm 2^\circ$ from the surface normal. Approximately 14% of the pulse energy is coupled to the active 200 μm square part of the devices. The pulse energy was kept below 13 μJ corresponding to the maximum intensity of 94 GW/cm². This is approximately a factor of 10 below the laser ablation threshold of our devices as determined by a separate experiment.

Figure 3(a) shows the photocurrent pulses at various V_g for the excitation light intensity I_{NIR} of 25 GW/cm² and with the optical electric field within the incident plane (p polarization). When increasing V_g from 0 to 70 V, the photocurrent decreases by a factor of 34. When raising V_g further, the photocurrent exponentially grows and reaches, at 140 V, a value larger than the zero-bias signal. For V_g above 70 V, a slow component of the photocurrents in the order of a few microamperes (about 0.1% of I_{dc}) is also observed. The slow signal decays over $\sim 5 \mu\text{s}$ (not shown).

FIG. 3. (Color online) (a) Photoelectric current pulse from the Mo FEA device observed when the gate bias voltages V_g was varied from 0 to 145 V, when the device was irradiated by the p -polarized 50 fs laser pulses at a peak intensity I_{NIR} of 25 GW/cm² (from top to down, V_g was varied by 10 V step between 0 and 70 V and by 5 V between 75 and 145 V; the photocurrent was measured using an amplifier with 10 MHz frequency bandwidth). (b) Photoelectric current signal vs laser intensity with V_g set to 0 V (filled triangle), 130 V (circles), and 145 V (squares). For the latter, the filled signs represent the fast and the empty symbols the slow signal component. The broken curve shows the I_{NIR}^3 dependence of the zero bias signal at low excitation intensity, evidencing the three photon photoemission process which becomes saturated at high excitation. The chain lines are the linear fit of the fast signals for V_g equal to 130 V (double chain) and 145 V (single chain) indicating that these are generated by single-photon photoemission.

The zero-bias photocurrent is assigned to the three-photon photoemission (3PPE) from the gate electrode that occupies 94% of the device area. This can be concluded from the pulse energy dependence of the signal (see below). The application of a positive V_g creates a potential barrier for the electrons at the surface of the gate electrode, and thus inhibits the 3PPE process. On the other hand, the fast component of the photocurrent observed for a bias above 70 V is attributed to the field-emission from the nanotip array enhanced by the incident laser field. The concomitant increase of I_{dc} with the increase of V_g is consistent with this interpretation (Fig. 2). Accordingly, the slow signal is ascribed to the increase of the lattice temperature of the emitters by the light absorption, in the order of 10° , and the resultant transient enhancement of I_{dc} .¹⁸ When the laser polarization is perpendicular to the incident plane (s polarization), the signal at $V_g=130$ V decreases to 65% of the one obtained with p -polarized light. This polarization dependence also supports the above interpretation that we indeed observe the ultrafast electron emission from the emitter tips induced by the laser field.⁴⁻⁷

Figure 3(b) shows the dependence of the peak photocurrent on I_{NIR} for V_g equal to 0, 130, and 145 V. The cubic dependence of the zero-bias signal at low laser intensity confirms the 3PPE process. In fact, the three-photon energy of 4.64 eV compares well to the work function of molybdenum of ~ 4.6 eV.¹⁹ At large I_{NIR} , the increase of the 3PPE signal becomes saturated, which is similar to previous reports.²⁰ In contrast, the I_{NIR} dependence at two finite bias voltages varies as $I_{NIR}^{(1+\epsilon)}$ with ϵ equal to 0.01 ± 0.04 and 0.08 ± 0.09 for V_g equal to 130 and 145 V, respectively. Hence, within the experimental uncertainty, the photocurrent at these conditions is generated by single-photon photoemission. The large dc electric field F_{dc} at the emitter apex and resultant lowering of the work function are the keys to observe the near infrared

single-photon photoemission from the field-emitter structures. Due to the Schottky effect, the work function of the molybdenum is lower than the single photon energy when F_{dc} is larger than 6 GV/m. In fact, by evaluating the relation between F_{dc} and V_g from the slope of the FN plot,¹⁸ we found that, when V_g was equal to 145 V, F_{dc} was equal to 7–14 GV/m.²¹

The actual pulse width of the fast current component is likely to be in the order of the light pulse width and is limited here by the bandwidth of the measurement electronics.²² Therefore, the peak current amplitude is proportional to the emission charge but the quantification of the value requires a separate calibration. This is done by measuring the photocurrent using a dc ammeter at zero V_g when I_{dc} was zero and comparing it to the pulsed peak current amplitude. From this calibration, we found the maximum single-photon quantum efficiency η_{ext} of 2.1×10^{-7} . This was observed at the largest V_g and corresponds to the maximum charge of the fast component of the electronic bunch of 0.27 pC. Assuming that the laser-induced electron bunch has the same pulse width as the excitation pulse and considering the spatial chirp of maximum 600 fs across the 200 μm device diameter due to the oblique angle of incidence,²³ the measured charge corresponds to a peak current of 0.5 A. The bunch length and the peak current can be improved by appropriate compensation.^{23,24}

Finally, we note that η_{ext} can be written as $(1-R)\eta_{tip}(r/p)^2$ where $R=0.56$ is the reflectance of the molybdenum at 800 nm, η_{tip} is the quantum efficiency at the emitter apex, and $(r/p)^2$ is the fraction of the emitter tip area given by the emitter diameter r equal to ~ 20 nm and the emitter period p equal to 5 μm . From these values, we found that η_{tip} is equal to $\sim 3 \times 10^{-2}$. A quantum efficiency for near infrared radiation of the similar order of magnitude was also observed in low work function materials such as the dispenser photocathode.²⁵ From the linearity of the signal, it is likely that the laser-induced emission charge from the FEAs can be significantly increased at the larger light pulse energy when the pulse width is proportionally increased with the pulse energy. The upscaling of the emission charge, the characterization of the emittance, and the elucidation of the electron emission mechanism are the subjects of further study.

In summary, we demonstrated the single-photon photoelectric current from molybdenum FEAs induced by near infrared laser pulses with 50 fs pulse duration. Despite the small fraction of the emission area from the field-emitter tips, up to ~ 0.3 pC of the emission charge from emitter tips

was observed with the external quantum efficiency of $\sim 2 \times 10^{-7}$. Our result indicates that metallic FEAs are promising for applications that require high-brightness ultrashort electron beams.

- ¹A. H. Zewail, *Annu. Rev. Phys. Chem.* **57**, 65 (2006).
- ²C. A. Brau, *Nucl. Instrum. Methods Phys. Res. A* **407**, 1 (1998).
- ³St. Grafström, *J. Appl. Phys.* **91**, 1717 (2002).
- ⁴P. Hommelhoff, Y. Sortais, A. Aghjani-Talesh, and M. A. Kasevich, *Phys. Rev. Lett.* **96**, 077401 (2006).
- ⁵P. Hommelhoff, C. Kealhofer, and M. A. Kasevich, *Phys. Rev. Lett.* **97**, 247402 (2006).
- ⁶C. Ropers, D. R. Solli, C. P. Schulz, C. Lienau, and T. Elsaesser, *Phys. Rev. Lett.* **98**, 043907 (2007).
- ⁷B. Barwick, C. Corder, J. Strohaber, N. Chandler-Smith, C. Uiterwaal, and H. Batellan, *New J. Phys.* **9**, 142 (2007).
- ⁸C. A. Spindt, *J. Appl. Phys.* **39**, 3504 (1968).
- ⁹P. M. Lally, Y. Goren, and E. A. Nettesheim, *IEEE Trans. Electron Devices* **36**, 2738 (1989).
- ¹⁰J. P. Calame, H. F. Gray, and J. L. Shaw, *J. Appl. Phys.* **73**, 1485 (1993).
- ¹¹D. R. Whaley, B. M. Gannon, W. O. Heinen, K. E. Kreischer, C. E. Holland, and C. A. Spindt, *IEEE Trans. Plasma Sci.* **30**, 998 (2002).
- ¹²H. Ishizuka, Y. Kawamura, K. Yokoo, H. Mimura, H. Shimawaki, and A. Hosono, *Nucl. Instrum. Methods Phys. Res. A* **483**, 305 (2002).
- ¹³H. Mimura, T. Ukeda, H. Shimawaki, and K. Yokoo, *J. Vac. Sci. Technol. B* **B22**, 1218 (2004).
- ¹⁴K. X. Liu, C.-J. Chiang, and J. P. Heritage, *J. Appl. Phys.* **99**, 034502 (2006).
- ¹⁵C.-J. Chiang, K. X. Liu, and J. P. Heritage, *Appl. Phys. Lett.* **90**, 083506 (2007).
- ¹⁶O. Yavas, N. Suzuki, M. Takai, A. Hosono, and S. Kawabuchi, *Appl. Phys. Lett.* **72**, 2797 (1998).
- ¹⁷E. Kirk, S. Tsujino, T. Vogel, H. Sehr, and J. Gobrecht, A. Wrulich (unpublished).
- ¹⁸C. A. Spindt, I. Brodie, L. Humphrey, and E. R. Westerberg, *J. Appl. Phys.* **47**, 5248 (1976).
- ¹⁹*CRC Handbook of Chemistry and Physics*, 79th ed., edited by D. R. Lide (CRC Press LLC, Boca Raton, 1998), pp. 12–124.
- ²⁰J. G. Fujimoto, J. M. Liu, E. P. Ippen, and N. Bloembergen, *Phys. Rev. Lett.* **53**, 1837 (1984).
- ²¹The estimation error of F_{dc} arises from the uncertainty of the slope of the FN-plot. It varied in time and on the V_g -scan directions as well. However, after an extensive conditioning procedure, the slope became stable within a factor of two. Similarly to Ref. 18, the emitter apex diameter estimated from the fitting of the FN-plot was smaller than the geometrical area obtained from SEM by a factor of 5–10.
- ²²From an additional experiment with 2 GHz-amplifier, we found that the fast current pulse was shorter than 0.8 ns, which was still limited by the bandwidth of the amplifier.
- ²³P. Baum and A. H. Zewail, *Proc. Natl. Acad. Sci. U.S.A.* **103**, 16105 (2006).
- ²⁴Zs. Bor and B. Racz, *Opt. Commun.* **54**, 165 (1985).
- ²⁵N. A. Moody, K. Jensen, D. W. Feldman, P. G. O'Shea, and E. J. Montgomery, *Appl. Phys. Lett.* **90**, 114108 (2007).