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Five picocoulomb electron bunch generation by ultrafast laser-induced field emission from metallic nano-tip arrays

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Laser-induced field emission from metallic field emitter array cathodes excited by femtosecond near infrared laser pulses is explored. When 50 fs laser pulses irradiated a 1.2×10^5-tip emitter array under a DC field emission bias, electron bunches with bunch charge up to 5.2 pC were observed. The variation of the bunch charge at different laser intensities and polarizations indicated that electrons were produced from the field emitters by a photofield emission process. The result demonstrates the feasibility of metallic field emitter array cathodes for high-charge short-pulse electron source applications. © *2011 American Institute of Physics*. [doi:10.1063/1.3631634]

Laser-induced field emission from sharp metallic tips[1–3] combines the high-brightness of the field-emission electron beam with femtosecond response times of electrons at metallic surfaces. These unique features can open up the exciting possibility to use field emitters for ultrafast electron microscopy.[4] Applications of field emitters for compact free electron lasers[5,6] and THz vacuum electronic sources have also been proposed. However, the electron bunch charge from a single etched-wire needle emitter is limited to $\sim10^3$ electrons per ultrafast laser pulse[3] due to the small emission area. To produce fast electron bunches with 10^7–10^9 electrons per laser pulse, which is required for x-ray free electron laser applications,[6] one needs to use high emission area cathodes, such as micro-fabricated field emitter array (FEA) cathodes[7] consisting of many ultrasharp metal tips on a single chip.

Electron bunches of femtosecond duration can be generated from metallic emitters owing to the small laser penetration depth and their short energy relaxation time.[8] The generation of laser-induced electron pulses from metallic FEAs was demonstrated only recently using Mo FEAs.[9] However, due to limited number of emitters in the tested array, extracted charge was not sufficient for accelerator applications.

In this letter, we explore high charge generation capability of the laser-induced field emission process using large area metallic FEA cathodes. By combining femtosecond near infrared laser irradiation with a strong DC bias, we demonstrated generation of electron bunches with up to 10^7 electrons per laser pulse. Under the strong DC bias electrons are emitted from the nanometer scale tip apexes via a single-photon photofield emission process.

We used single-gate all-metal FEAs, inset of Fig. 1, with 1.5-μm-base and 5-μm-pitch emitters having the tip apex radius of curvature of 10 nm fabricated by molding and a self-aligned gate process.[10] Both the emitter tips and the gate electrode were made of Mo. In Fig. 1, we show DC field emission characteristics of three FEAs without laser irradiation having nominally the same tip apex radius of curvature, with different number of emitters (100, 1600, and 1.2×10^5 tips from bottom to top). The characteristics shown were taken after a repeated DC-scan conditioning operation for several days.[7,10] We observed that for a given bias voltage V_{ge} between the gate and the emitter, DC field emission current was approximately proportional to the number of emitters. In the laser-induced field emission experiment described below, we used the FEA with 1.2×10^5 emitter tips arranged in a 2.2 mm-diameter circle. The maximum anode current I_a for this FEA was 6 mA for V_{ge} of 91 V. The field emission current intercepted by the gate-electrode was below 10^{-5} A (not shown). The I_a-V_{ge} characteristic of the 1.2×10^5-tip FEA was fitted well with the Fowler-Nordheim equation,[7] $AV_{ge}{}^2\exp(-B/V_{ge})$, with $A=(2\pm0.1)\times10^{-2}$ A V^{-2} and $B=917\pm6$ V as shown by the curve in Fig. 1. The DC electric field F_{DC} at the tip apexes is approximately given by βV_{ge} with the field enhancement factor β written as $b\varphi^{3/2}/B$ using B obtained from the Fowler-Nordheim fitting of I-V characteristics, the work function φ, and a constant b equal to 6.83089 eV$^{-3/2}$ V nm^{-1}. Assuming

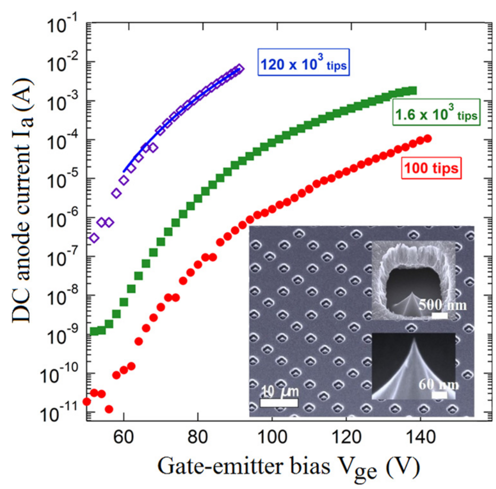

FIG. 1. (Color online) Current-voltage characteristics of three single-gate Mo FEAs with different number of tips. For the 1.2×10^5 tip array data, a fitting by a Fowler-Nordheim equation (curve) is also shown. The insets are SEM micrographs of single emitter cell and a high-magnification image of single emitter tip.

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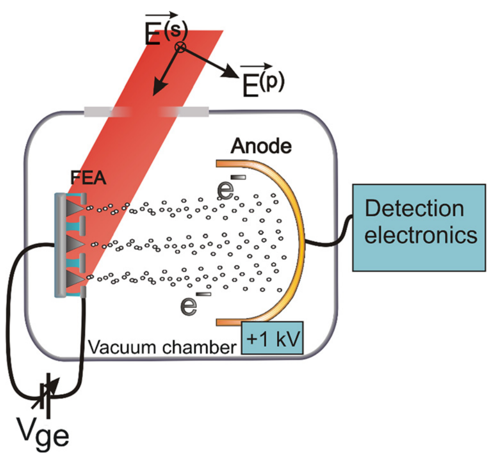

FIG. 2. (Color online) Schematic drawing of setup of the laser-induced field-emission experiment with metallic FEAs excited by femtosecond near infrared laser pulses.

φ of 4.6 ± 0.1 eV for Mo, we found that β was $(7.1–7.6) \times 10^7$ m^{-1} and F_{DC} was 6.1-6.6 GV/m for V_{ge} of 91 V.

Figure 2 illustrates the experimental setup. FEAs were installed in a vacuum chamber with a base pressure of 1×10^{-8} mbar. We irradiated FEAs with 50 fs laser pulses at a wavelength of 800 nm generated from a 2 kHz repetition rate Ti:Sapphire laser system. Linearly polarized laser pulses irradiated the FEA chip through a glass window with 60° incident angle from the chip surface normal. In the case of *p*-polarization, the optical electric field E_l is parallel to the incident plane, as shown in Fig. 2, having a component along the emitter tip. We focused the laser beam to an elliptical spot such that the irradiated footprint on the FEA was round with a diameter of ~2.5 mm in full-width at half-maximum. We irradiated the FEAs with laser intensities of at most 86 GW/cm^2 at the FEA surface to avoid laser-ablation. Electrons emitted from FEAs were collected by a Faraday cup, separated by 10 mm from the FEA. The Faraday cup was DC biased at 1–2 kV via a bias-T with 1 GHz bandwidth, allowing for simultaneous measurements of the DC emission current and the charge Q of the laser-induced electron pulse.

In Fig. 3(a), we show the relation between Q and V_{ge} for two laser polarizations. The V_{ge}-dependence of Q was different for V_{ge} below and above ~70 V: (i) for $V_{ge} < 70$ V, Q decreased with the increase of V_{ge} and (ii) for $V_{ge} > 70$ V, Q increased steeply with the increase of V_{ge}. The dependences of Q on laser intensity and laser polarization were also different in the two regions as shown in Fig. 4(a) for V_{ge} of 0 and 91 V. When V_{ge} was 0 V, Q exhibited a marginal polarization dependence and increased superlinearly, saturating at intensities higher than 40 GW cm^{-2}. In contrast, when V_{ge} was 91 V, Q increased linearly with the laser intensity and strongly depended on the laser polarization. For *p*-polarized laser pulses Q was 5.6 ± 0.1 times larger than for *s*-polarized laser pulses. These observations indicate that Q was generated by different emission processes depending on the applied V_{ge} values.[9] We ascribe the signal for V_{ge} of 0 V to three-photon photoemission from the planar part of the gate electrode occupying ~90% of the device area, whereas the signal for $V_{ge} > 70$ V (region (ii) Fig. 3(a)) is ascribed to photofield emission[11,12] from the sharp emitter apexes. At the gate surface facing the anode, the increase of V_{ge} can suppress three-photon photoemission (see

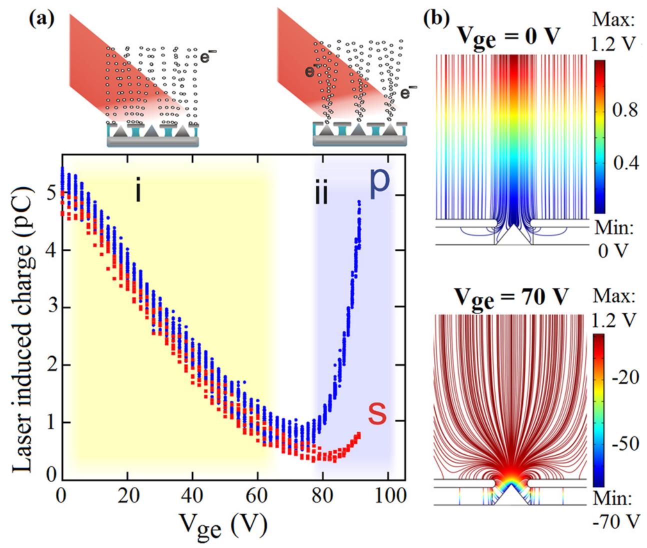

FIG. 3. (Color online) (a) The relation between the laser-induced charge Q and DC gate-emitter potential V_{ge} at a laser intensity of 72 GW/cm^2 for *p*- and *s*- laser polarizations. For V_{ge} below (i) and above 70 V (ii), the $Q-V_{ge}$ characteristics were qualitatively different (see text). (b) DC electric field lines for $V_{ge} = 0$ V and $V_{ge} = 70$ V illustrating suppression of electron emission from gate electrode in region (i).

region (i) Fig. 3(a)) since the fringe field near the gate aperture edge enhances the potential barrier height with the increase of V_{ge}, as the electrostatic simulations shown in Fig. 3(b) indicate. This is consistent with the linear reduction of Q with the increase of V_{ge}. At the emitter tip apexes, rising V_{ge} reduces the potential barrier and exponentially enhances the tunneling probability of electrons excited by near infrared laser pulses. The linear laser intensity dependence of Q indicates that emission of the electrons excited by one-photon energy was most efficient because of the reduced height and thickness of the tunneling barrier at the emitter apex under strong F_{DC}.

The factor 5.6 ± 0.1 higher Q observed for *p*-polarized laser pulses in region (ii) is likely an indication of enhancement of the optical electric field $E_l^{(p)}$ for *p*-polarized laser pulses at the emitter tip apex Ref. 13 and concomitant

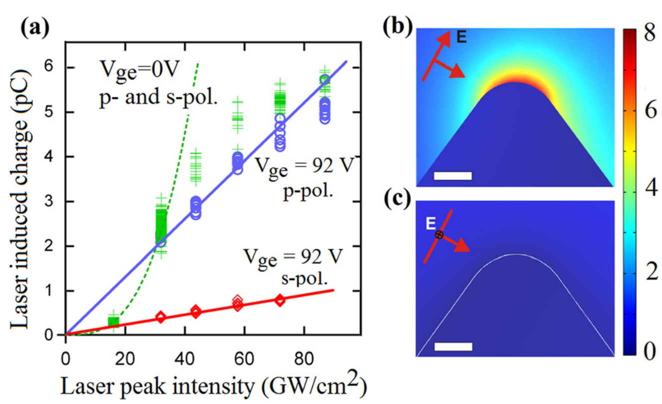

FIG. 4. (Color online) (a) Laser intensity dependence of the laser-induced charge Q at V_{ge} of 0 V (crosses) and 91 V (open circles) for *p*-polarized and for *s*-polarized laser pulses (diamonds), respectively. The lines show the single-photon photoemission dependence of signals at $V_{ge} = 91$ V obtained by linear fitting. The dashed curve show a three-photon photoemission dependence of Q at $V_{ge} = 0$ V fitted with the low laser intensity characteristic. (b) and (c) show the calculated optical electric field distribution near the Mo field emitter tip apex with the radius of curvature of 10 nm for *p*- and *s*-polarized beams with 800 nm wavelength irradiating with 60° incident angle. *Scale bars* represent 10 nm.

enhancement of the photon absorption. Taking into account the linear laser-intensity dependence of Q, we estimated the ratio $E_l^{(p)}/E_l^{(s)}$ at tip apexes from the square-root of the ratio (5.6 ± 0.1) of Q generated by p-polarized laser pulses to Q generated by s-polarized laser pulses (Fig. 4(a)) and found a value equal to 2.4. This value is similar to the recently reported enhancement factor of ∼3 for Mo FEAs[14].

To confirm the $E_l^{(p)}$ enhancement at sharp tip apexes, we simulated the three-dimensional laser field distribution near a sharp Mo emitter apex by using a finite element Maxwell equation solver (COMSOL multiphysics). Figures 4(b) and 4(c) show the calculated optical electric field distribution over a cross section through the center of the emitter tip for p- and s-polarized laser beams, respectively. Norm of the electric field, normalized by the incident wave field value is displayed in these figures. We assumed an axisymmetric Mo emitter with apex radius of curvature of 10 nm and irradiated the emitter by a plane wave of 800 nm wavelength with 60° incident angle. Additional two-dimensional simulation indicated that the gate electrode had only a marginal effect on the optical field distribution near the tip apex.

As shown in Fig. 4(b), the simulation showed that the p-polarized electric field was locally enhanced at the emitter tip. This strongly supports the conclusion that the observed laser-induced charge in region (ii) was generated from the nanometer-scale tip apexes by photofield emission. In the simulation, the maximum enhancement factor of $E_l^{(p)}$ was 7.4 at a position 1 nm shifted from the emitter apex to downstream. The averaged $E_l^{(p)}$ enhancement factor around the emitter apex was 5.8, which is compatible with the experiment. Here we averaged the field within 25° around the tip apex, since the current density is estimated to be one half of the maximum for our emitters at this angle. For s-polarization, there was no enhancement. For more precise estimate of the field-enhancement, one should take into account the influence of the gate electrode[15] and femtosecond dynamics of excited electrons.[16] These are beyond the scope of this letter.

As an alternative laser-induced field emission process at intense E_l, optical field emission has been discussed in the literature[1,3] as a way to produce electron bunches with a few femtosecond duration. Similarly to DC field emission, optical field emission is an electron field emission process induced by electric field oscillating with optical frequency. As such, the bunch charge is expected to exhibit very strong laser polarization dependence and nonlinear laser intensity dependence.[1,3] These were not observed in our experiment, which we tentatively ascribe to insufficient laser intensity; in our experiment, the estimated optical electric field perpendicular to the emitter surface was below ∼4.6 GV/m taking into account the field enhancement factor of 5.8 from the simulation described above. This is an order of magnitude lower than values required to observe optical field emission as reported by Bormann *et al.*[3]

Finally, we note that the maximum quantum efficiency η_{tip} at the emitter tip apex was $\sim 4 \times 10^{-3}$ in our experiment, estimated by taking into account the ratio $(S_{total}/S_{tips}) \sim 1 \times 10^{-5}$ of the total device area S_{total} to the total tip area S_{tips}. Our η_{tip} value was an order of magnitude higher than estimated maximum quantum efficiency of $\sim 6 \times 10^{-4}$ in strong optical field emission experiment by Bormann *et al.*,[3] indicating that photofield emission regime with large F_{DC} at the emitter tip apex (∼6.6 GV/m in our experiment) is advantageous to obtain large electron bunch charge.

In summary, we demonstrated laser-induced electron emission with up to $\sim 10^7$ electrons per pulse from metallic FEAs with 1.2×10^5 tips under strong DC-electric field bias condition. The observed 5.2 pC bunch charge generation also indicates the feasibility of using FEA cathodes for new applications such as vacuum electronic amplifiers and oscillators into submillimeter/THz range as well as for accelerator applications such as the SwissFEL x-ray free electron laser.[6] Further increase of the beam brightness by increasing the emitter tip density combined with a FEA structure with a collimation gate electrode[17–19] is the subject of intense research.

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