Homogeneity improvement of field emission beam from metallic nano-tip arrays by noble-gas conditioning

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The authors explored control of field-emission electron beam homogeneity emitted from metallic nano-tips fabricated by molding. Improved beam homogeneity and increased maximum bunch charge were observed after DC operation of cathodes in low-pressure argon gas environment. They also observed average current per tip of 4.5 μA from a 1×10^4 emitter array device after conditioning in neon gas. Evolution of Fowler-Nordheim parameters as a result of noble-gas conditioning was discussed. © 2011 American Institute of Physics. [doi:10.1063/1.3624705]

Field-emitter array (FEA) cathodes with high emission current are promising for applications that require high beam brightness such as free electron lasers^{1,2} and microwave/THz vacuum electronic amplifiers. Using metallic FEAs fabricated by molding,³ sub-nanosecond electrical switching in high acceleration electric field up to 30 MV/m [Refs. 1 and 2] and ultrafast near infrared laser-induced field emission with up to 5 pC electron charge pulse [Ref. 4] were demonstrated recently. However, field emission beam imaging experiments^{1,2} indicated that only a limited number of tips emitted electrons. This is partly because of the distribution of the tip radii of curvature R_{tip} ^{5–8} that demands a procedure to improve it. This is particularly important for high current operation, since, when gate voltage is increased for higher current, current density at sharp emitters that are already switched on at low-voltages, could easily exceed the critical value in the order of 10^7 – 10^8 A cm^{−2} and trigger destructive vacuum arc.⁹ In this letter, aiming at homogenizing the R_{tip} -distribution, we explored the effect of *in-situ* exposure of FEA tips to low pressure noble gas and studied evolution of beam image and current-voltage (*I*-*V*) characteristics.

Three single-gate FEAs, FEA1 (5 μm -pitch, 1.2×10^5 -tips), FEA2 (5 μm -pitch, 3×10^4 -tips), and FEA3 (10 μm -pitch, 1×10^4 -tips) were fabricated by molding and a self-aligned gate process.³ These consisted of pyramidal shape molybdenum field emitters with nominal R_{tip} of ~ 5 nm, ~ 1.2 - μm -height, and 1.5- μm -base-size. Inscribed circle diameters D of the gate aperture holes were ~ 2 μm . To observe the field emission beam image, we integrated FEAs in a high-voltage diode gun of SwissFEL gun test facility.^{1,2} In the diode gun, a negative 200 keV pulse with 250 ns duration was applied to the cathode. Field emission electron pulses from FEAs with ~ 0.5 ns duration were synchronously generated when the cathode voltage was at the largest. The field emission switching was done by applying negative potential $-V_{\text{ge}}$ to the emitter substrate with respect to the gate electrode, which was in contact to the cathode holder of the gun.^{1,2} V_{ge} consisted of a DC bias and a ~ 1 ns pulse. DC bias was kept small for the imaging experiment (with DC

emission current below $\sim 10^{-5}$ A). The field emission beam image was detected by a scintillating screen. Pulse charge was measured by inserting a Faraday-cup. Before the diode gun experiment, we first conditioned FEAs in a preparation chamber. The initial conditioning^{2,10} involved a repeated DC-bias scan or an application of a 50 Hz-AC- V_{ge} at the base pressure of $(1\text{--}5) \times 10^{-9}$ mbar while slowly increasing the maximum V_{ge} over a few days until *I*-*V*s became stable. During the conditioning in the preparation chamber, we collected electrons by an anode biased at a positive voltage V_a of 1 kV, while measuring the anode current I_a , the gate current I_g , and the current I_{em} injected into the emitter substrates. We then transferred FEAs to the diode gun through air, and again DC conditioned those at the base pressure of $(0.5\text{--}1) \times 10^{-8}$ mbar. In the diode gun, we were able to monitor only I_{em} during conditioning with DC V_{ge} with no additional potential between cathode and anode.¹¹

In Figure 1(a), we show an image of a 0.5 ns-long field emission pulse from FEA1 with 0.7 pC charge and 200 keV beam energy. V_{ge} of 78 V (DC 45 V superposed by 33 V pulse) was applied to generate the pulses. Granular spots indicated that beam was emitted from a small number of emitters.² Attempted pulsed-current tip-blunting^{8,12} by application of higher V_{ge} pulses was not successful, leading to a partial loss of emitters (approximately one half) by vacuum arc. We, therefore, tested Ar gas effect on FEAs. For that, we stopped the acceleration voltage and V_{ge} pulse and applied a DC V_{ge} up to 80 V to the chip under Ar gas pressure of 10^{-4} mbar for 2 h. This resulted in a gradual increase of I_{em} to ~ 7 mA at 80 V as shown in Fig. 1(d). The increased conductance near zero V_{ge} showed that I_g increased substantially. Nevertheless, we were able to extract 0.7 pC of bunch charge at V_{ge} of 83 V (DC 50 V and 33 V in pulse) as shown in Fig. 1(b). This was acquired with the same gun and camera setting as in Fig. 1(a) and displayed with the same contrast scale. Although granular emission pattern persisted, emission emerged from some areas (near bottom edges) where it was absent before the Ar conditioning. Also less bright spots were observed. To quantify this beam homogenization effect, we compared normalized and angle-averaged autocorrelation $f_{\text{ac}}(r)$ (r is the spatial scale on the screen) as shown in Fig. 1(c). We found that the width

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FIG. 1. (Color online) Beam images of 0.5 ns-long field-emission electron pulses from FEA1 with 0.7 pC bunch charge, (a) before and (b) after Ar conditioning. The bar in (a) corresponds to 2 mm on the scintillating screen. (c) Autocorrelation of beam images shown in (a) and (b), together with that of a 1.3 pC beam. The curve shows a simulated autocorrelation for a uniform beam with the elliptic beam envelope (broken curves in (a) and (b)). (d) I - V s of FEA1, before and after Ar conditioning. Open marks show I_{cm} (current injected to the emitter substrate) measured with DC V_{ge} . Filled marks indicated actual emission current I_a as evaluated from the bunch charge of the accelerated beam pulses divided by the pulse duration of 0.5 ns.

$r|_{70\%}$ increased 4 times after the Ar conditioning (we defined $r|_{70\%}$ by $f_{ac}(r|_{70\%}) = 70\%$). It was 32% of the f_{ac} -width of an elliptic uniform beam with the envelope indicated by broken curves in Fig. 1. In addition, we were able to increase V_{ge} to 94 V (DC 61 V and 33 V in pulse) to produce electron pulses with 1.3 pC bunch charge, which exhibited the same $r|_{70\%}$ -width as the 0.7 pC bunch charge beam [Fig. 1(c)]. In contrast, we observed no effect when nanosecond pulses drove a FEA for 3 h with 10 Hz repetition rate under the same Ar gas pressure.

Aiming at realizing more homogeneous field emission beam, we next repeated experiment with FEA2 but with careful conditioning so as not to lose any emitter tips by vacuum arc before beam imaging. In particular, we kept DC V_{ge} below ~ 50 V during conditioning in the diode gun, which avoided the increase of I_g . In Fig. 2(a), we show the beam image of FEA2 with 2 pC charge and 200 keV beam energy. The beam homogeneity and the beam current density were higher than FEA1. The autocorrelation $r|_{70\%}$ -width of the beam was 57% of a uniform elliptic beam (broken curve of Fig. 2(a)).

To understand the Ar gas conditioning process on FEA2 quantitatively, we examined evolution of I - V s, a to d in Fig.

FIG. 2. (Color online) (a) Field emission image of FEA2 with 2 pC charge and 200 keV beam energy together with its autocorrelation. The curve shows a simulated autocorrelation of a uniform beam with elliptic beam envelope (broken curve in the inset) (b) DC and pulsed I - V s of FEA2 in the preparation chamber and in the diode gun.

2(b), before the imaging experiment (in the gun, I_a was approximated by I_{em}). The data were fitted well by a function (curves in Fig. 2(b)),^{13,14}

$$I_a = A_{FN}(V_{ge}/B_{FN})^n \exp(-B_{FN}/V_{ge}) \quad (1)$$

with n equal to 2. The fitting parameters A_{FN} and B_{FN} were summarized in Table I. This function is obtained from the Fowler-Nordheim (FN) approximation of I_a

$$I_a = S(ae^{bc^2/W^{1/2}}/W)F^2 \exp(-bW^{3/2}/F) \quad (2)$$

where S is the total emission area given by $\sim N R_{tip}^2$ (N is the average number of current-emitting tips), F is the electric field at the emitter apex, W is the work function, and a , b , and c are parameters equal to 1.541434×10^{-6} A eV V^{-2} , 6.830890 eV $V^{-3/2}$, and 1.199985 eV $V^{-1/2}$ nm $V^{-1/2}$ respectively.^{13,14} From Eqs. (1) and (2), one finds that A_{FN} is proportional to $N R_{tip}^2 W^2 e^{bc^2/W^{1/2}}$. B_{FN} is proportional to $W^{3/2}(R_{tip})^{0.5}$. This is obtained by approximating F with $2\beta V_{ge}/D$, where β is the field enhancement factor at the emitter apex proportional to $\sim 1/(R_{tip})^{0.5}$ for our pyramidal emitters.¹⁵

In Figure 2, the I - V of FEA2 denoted as a was observed after conditioning at the base pressure in the preparation chamber. After subsequent conditioning in 4×10^{-5} mbar-Ar gas for 5 h in total (b in Fig. 2(b)), A_{FN} became 13.3 ± 3.1 times higher and B_{FN} increased by $17\% \pm 2\%$. These values suggested an increase of $N R_{tip}^2$ and an increase of R_{tip} . It is difficult to account for an order of magnitude increase of A_{FN} and fractional increase of B_{FN} by sole variation of W . I_g (b' in Fig. 2(b)) was below 9% in the V_{ge} range,

TABLE I. Summary of the Fowler-Nordheim parameters of I - V s of FEA2 in Fig. 2(b).

Index	A_{FN} (A)	B_{FN} (V)
A	$(0.779 \pm 0.091) \times 10^4$	647.27 ± 5.01
B	$(10.358 \pm 2.104) \times 10^4$	756.98 ± 9.35
C	$(8.741 \pm 1.868) \times 10^4$	677.83 ± 8.35
D	$(8.754 \pm 1.500) \times 10^4$	662.99 ± 6.78

therefore, it does not affect the interpretation. We then transferred the chip from the preparation chamber to the diode gun through ambient air. After reaching 1×10^{-8} mbar, DC V_{ge} conditioning was conducted for a day (c in Fig. 2(b)) and subsequently in 10^{-5} mbar-Ar gas for 30 min (d in Fig. 2(b)) before the imaging experiment. The comparison of b and d shows that, as a result of the transfer of the chip through air, A_{FN} changed by a factor of 0.84 ± 0.29 and B_{FN} decreased by $12\% \pm 2\%$ (we approximated I_a with I_{em} because of the small difference between the pulse current and I_{em}). This suggested a reduction of R_{tip} and a degradation of the beam homogeneity due to, for example, a partial oxidation of emitter tips in air.

It is an interesting question whether other gas has the same effect as argon. We, therefore, tested the effect of Ne gas. Using FEA3 and examining evolution of I - V s in the preparation chamber, we again observed increased FN parameters; Fig. 3(a) shows the variation of pressure, the maximum I_a , the maximum V_{ge} , and A_{FN} and B_{FN} in time under AC 50 Hz V_{ge} . FN parameters obtained from the fitting of I - V s, a - f in Fig. 3(b), were summarized in Table II. After initial conditioning (a in Fig. 3), we repeatedly operated the chip under 10^{-4} mbar-Ne gas environment and at the base pressure. This resulted in the increase of A_{FN} and B_{FN} (the variation from a to b). The periodical increase of I_a during this time resulted from a combination of decreasing I_a and subsequent manual increase of the maximum V_{ge} . A gradual decrease of A_{FN} and B_{FN} followed after returning to the base pressure (the variation from b to c). Still, A_{FN} and B_{FN} of c remained higher than a . After three additional cycles, the resultant I - V (d in Fig. 3) exhibited A_{FN} and B_{FN} , respectively, 16.2 ± 1.0 times and 1.95 ± 0.01 times larger than those of a . We then tested high current operation of FEA3 in this state in the preparation chamber with 92-ns-long pulsed V_{ge} . The data, e and f , in Fig. 3(b) show two scan results with different maximum V_{ge} . The I_a - V_{ge} of e were approximately the same as d but f with maximum V_{ge} of 160 V shifted to higher V_{ge} and A_{FN} and B_{FN} increased as well. This e -to- f variation of I - V appeared to be due to current-induced tip-blunting,^{8,12}

FIG. 3. (Color online) Variation of the maximum anode current I_a , the maximum V_{ge} amplitude, pressure with Ne-gas, and FN parameters A_{FN} and B_{FN} under 50 Hz-AC- V_{ge} for FEA3. (b) Evolution of I_a - V_{ge} characteristics. a to d were measured with 50 Hz-AC- V_{ge} . e and f were obtained from a pulsed measurement with 92 ns-FWHM V_{ge} pulse.

TABLE II. Summary of the Fowler-Nordheim parameters of I - V s of FEA3 in Fig. 3(b).

Index	A_{FN} (A)	B_{FN} (V)
A	$(0.303 \pm 0.012) \times 10^4$	485.6 ± 1.59
B	$(7.413 \pm 0.427) \times 10^4$	911.77 ± 3.52
C	$(1.026 \pm 0.024) \times 10^4$	696.29 ± 1.34
D	$(4.903 \pm 0.238) \times 10^4$	946.96 ± 3.42
E	$(2.109 \pm 0.917) \times 10^4$	890.38 ± 35.7
F	$(5.215 \pm 0.918) \times 10^4$	1048.5 ± 16.4

but no arc was observed until I_a reached the maximum value of 45 mA at 160 V. This current corresponded to $4.5 \mu\text{A}$ per tip and was higher than $2.5 \mu\text{A}$ per tip from a 1600-tip molded single-gate FEA reported previously.^{3,16}

As one underlying mechanism of the emission homogeneity improvement, we consider that bombardment of emitters by noble-gas ions, that were generated by electron impact ionization near current-emitting tips, caused sputtering⁶ or ion-implantation.¹⁷ From the expected R_{tip} -dependencies for A_{FN} ($\sim N R_{tip}^2$) and B_{FN} ($\sim (R_{tip})^{0.5}$) with the FN exponent n of 2, the observed increase of A_{FN} and B_{FN} was compatible with the increase of R_{tip} by sputtering-induced tip-blunting and removal of adsorbate and native oxide. Preferential sputtering of oxygen atoms from partially oxidized molybdenum emitter surface could also contribute to this.¹⁸

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