DEVELOPMENT OF DIAMOND FIELD-EMITTER ARRAYS FOR FREE-ELECTRON LASERS

Jonathan D. Jarvis, Heather L. Andrews, Charles A. Brau, Department of Physics and Astronomy, Vanderbilt University, Nashville, TN 37235, U.S.A.
Bo Kyoung Choi, Jimmy Davidson, Weng Kang, Supil Raina, Yong Mui Wong, Department of Electrical and Computer Engineering, Vanderbilt University, Nashville, TN 37235, U.S.A.

Abstract

We report recent advances in the development of diamond field-emitter arrays (DFEA) as a promising electron source for free-electron lasers. Both sparse and close-packed arrays have been produced using an inverse-mold transfer process. High-pitch arrays have been used in the development of conditioning techniques that drive the emitters toward uniformity in a self-limiting fashion. Properties of these cathodes including I-V response, emitted energy spread, transverse emittance, temporal stability, and operational lifetime are being examined in two DC test stands. Highly uniform, stable emission current of 15 μA/tip (DC) has been achieved. The resulting high-input-power density destroyed the phosphor anode locally; therefore, higher currents could not be attempted. In an RF gun, pulsed picosecond operation will allow much higher peak currents, and back bombardment from sublimated anode material will not be present. The maximum DC-current densities observed scale to approximately 300 A/cm^2 for fine-pitch arrays, demonstrating great promise for use in free-electron lasers.

INTRODUCTION

Field-emitter arrays have long been considered as potential cathodes for electron-beam-driven radiation sources over a wide range of the electromagnetic spectrum. These devices range from backward-wave oscillators in the THz and GHz, to free-electron lasers in the x-ray and infrared. One of the greatest challenges in the development of FEAs has been reliable scalability and the ability to condition them for uniform emission. The development of early microfabrication technologies in the 1960s led to the invention of the well known Spindt FEA [1]. While these molybdenum FEAs have demonstrated the capability for large current densities, they have only been scalable to areas of 1 mm^2 with tip numbers of less than 50,000. Recent experiments have demonstrated the potential of diamond field-emitter arrays as cathodes for free-electron lasers [2, 3, 4]. Diamond field emitters have several advantages over metallic devices. Since diamond is a covalent solid, temporal stability from clean diamond emitters is superior to that of metals. The high thermal conductivity of diamond mitigates self Joule heating which can lead to the explosive vaporization that plagues metallic field emitters. Additionally, diamond emitters are chemically inert and perform well in poor vacuum conditions. Unlike existing photocathode technology, DFEAs can be stored and transported in air and have proved to be extremely rugged, operating reliably at high per-tip current levels (15 μA/tip DC) in poor vacuum (~10^{-6} Torr). Furthermore, FEAs eliminate the need for a laser window and there is insignificant heat generation at the cathode. These devices should readily integrate with normal and superconducting RF systems by a variety of emission gating techniques. In this report we present recent measurements of DC per-tip-current capability, transverse emittance, and emitted energy spread. We also present uniformity conditioning techniques, including one which yields highly uniform emission in a self-limiting fashion.

FABRICATION

Diamond field-emitter arrays are produced using an inverse-mold-transfer process [5]. Oxidized Si wafers are patterned in preparation for an anisotropic KOH etch that produces pyramidal molds with an opening angle of 70°. These molds are sharpened by oxidation in preparation for diamond deposition. The oxide grows preferentially on the walls of the mold, avoiding the corners (Figure 1).

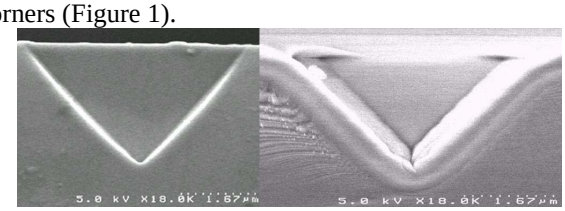

Figure 1. Si mold before (left) and after (right) oxide sharpening.

The result is a sharp recess in the tip of the mold where the faces of the pyramid converge. By using mask holes that are slightly rectangular we can produce two sharp tips rather than a single. Over etching of the mold prior to oxidation allows production of quad-tip emitters. After sharpening, the mold is pretreated by ultrasonication in a diamond slurry, a step which provides nucleation sites for diamond growth. Diamond is then grown in the mold by microwave-plasma chemical-vapor deposition (MPCVD). A variety of growth recipes are used to achieve a desired combination of sp^2, sp^3, dopant concentration, and nitrogen content. A thin, conformal, nanodiamond layer is deposited first, while microdiamond is used to back fill the bulk of the structure. The diamond is then sputtered with a Ni/Ti coating that serves as a buffer/adhesion layer during substrate brazing. TiCuSil braze is used to attach the cathode-mold structure to a polished Mo substrate. After brazing, the protective Si mold is removed with a

KOH etch, and the sharpening oxide is removed with a buffered oxide etch (BOE). Following standard cleaning procedures, the cathode is ready for testing. A completed diamond field-emitter array is seen in Figure 2.

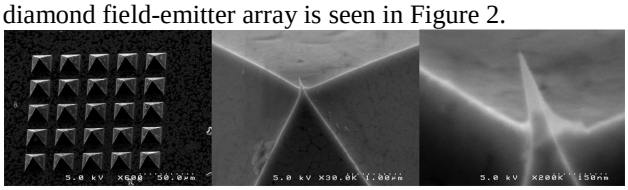

Figure 2. Completed diamond FEA with tip detail.

UNIFORMITY CONDITIONING

In this work we consider three main conditioning schemes for enhancing uniformity of diamond FEAs. The first is a thermal annealing of the cathode while it is emitting, called vacuum thermal electric conditioning (VTEC). The second is the selective exposure of the cathode to various gaseous backgrounds. During gas exposure, we have tested the effects of low and high field and emission operation, as well as the application of heat. The final conditioning technique we have considered is prolonged operation of the cathode at moderate to high current levels. All testing thus far has been performed in a DC mode. Pulsed conditioning techniques will be examined in the coming months.

A schematic of the experimental configuration for these conditioning procedures is shown in Figure 3.

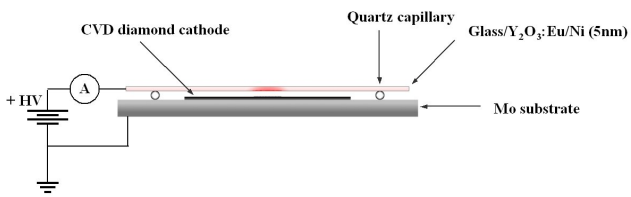

Figure 3. Uniformity apparatus schematic.

An ungated diamond field-emitter array (Figure 2) is placed in a close-diode configuration with a phosphor anode. The anode-cathode gap is set using precision quartz capillaries allowing the application of known electric fields up to ~30 V/μm. The conditioning test stand has a base pressure of ~10^{-8} Torr and allows a variety of gaseous environments (~10^{-3}-10^{-8} Torr) to be achieved using a fine leak valve. The cathode can be heated as high as 350 C by a tungsten filament under its base.

VTEC

Experimental observations show emission dominated by weakly bound adsorbates for low and moderate current operation of unconditioned cathodes (Figure 4). VTEC appears to drive off these weakly attached adsorbates while activating tightly bound species, allowing them to migrate to the region of highest field. These adsorbates enhance the emission by a dipole lowering of the local-surface-energy barrier, resonant tunneling, or other effects. Following VTEC treatments of ~200-300 C, emission uniformity is greatly improved, turn-on field is decreased, and current fluctuation due to adsorbate diffusion is dramatically reduced.

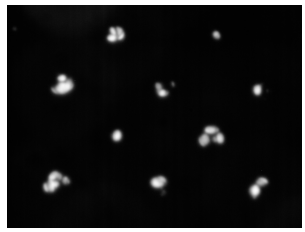

Figure 4. Beamlets from individual adsorbates on diamond nanotips (centroid spacing ~300 μm).

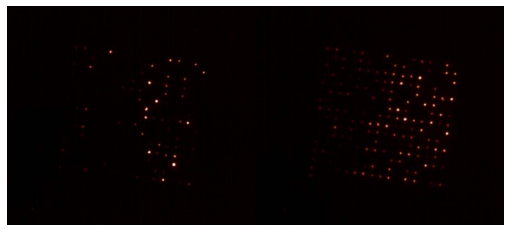

Figure 5. (left) before VTEC ~30% of tips active, (right) after VTEC ~60% of tips active.

Selective gas exposure

Thus far, exposure to a background gas at 10^{-5}-10^{-3} Torr, while emitting at any level, has resulted in an increased turn-on field for every tip. Lower pressures have produced no significant effect. Following high-pressure-gas exposure, VTEC has improved emission somewhat, however the damage is mostly irreversible. An apparatus is under development for heating the cathode to high temperatures (~800 C) in the presence of atomic hydrogen. This hydrogen termination and the negative electron affinity surface it produces may enhance the emission properties substantially.

Moderate/High current operation

Operation at high currents has resulted in the highest degree of uniformity of any conditioning method investigated. The mechanism of conditioning is believed to be thermal-assisted field evaporation of the diamond nanotips to a uniform effective radius. At high emission current, it is expected that the temperature of the emitting surface rises significantly, many hundreds of degrees C. Combined with the high electric field at the emitter sites, negatively charged carbon clusters can be evaporated into the vacuum. Because this process is extremely sensitive to electric field and current, the best emitters are selectively targeted leading to uniform tip geometry across the entire array. The beam from a 5x5 array undergoing conditioning is shown in Figure 6. The field and current were progressively increased over approximately two hours of operation (left to right). The final current in the right-most frame was ~15 μA/tip. Figure 7 presents SEM micrographs of four tips in this array before and after high-current conditioning. The reduction in tip sharpness is clearly visible. To rule out tip etching due to back bombardment as a conditioning

mechanism, we have performed an SEM study of a large number of tips during a conditioning progression [3]. Our observations suggest that thermal-assisted field evaporation is indeed the primary conditioning mechanism.

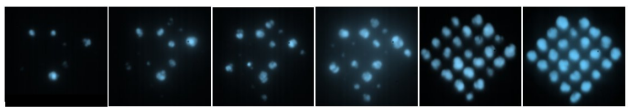

Figure 6. High-current conditioning of a 5x5 DFEA, 200 μm pitch.

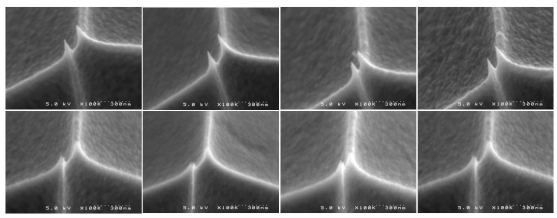

Figure 7. Emitters before (top) and after (bottom) high-current conditioning.

In addition to uniformity conditioning by evaporation, we observe field-induced-self-healing effects, which serve to correct certain fabrication or processing defects. These include the straightening of bent tips and the separation of double tips that are stuck together [3].

TRANSVERSE EMITTANCE

The transverse emittance is of critical importance for beam transport and beam-wave interaction in the lasing process. Specifically, the transverse emittance of the electron beam must be small compared to the laser wavelength to ensure acceptable volume overlap during the interaction. To measure the transverse emittance from ungated diamond field-emitter arrays we utilize a pepperpot technique in a low-energy-DC test stand (Figure 8). The anode-cathode gap is set using quartz capillaries, 330 μm in diameter. When the thickness of the diamond film is included, the gap is ~300 μm. In this arrangement, fields up to ~17 V/μm can be applied. The pepperpot is fabricated from a single SOI wafer and has 30 μm square holes with a 200 μm pitch. In the current experiment, the beamlet that emerges from the pepperpot comprises emission from fewer than a dozen tips. As a result, the beam has nonzero correlations that slightly preserve the aperture's square shape in the final image.

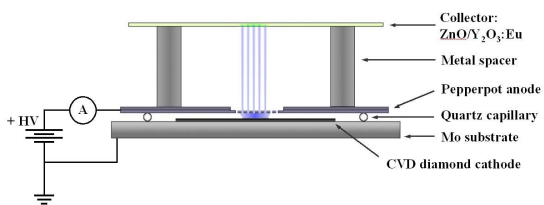

Figure 8. Emittance apparatus schematic.

Figure 9 shows one of the collected beamlets with the approximate location of a linescan that is used to estimate the cathode's rms angular divergence.

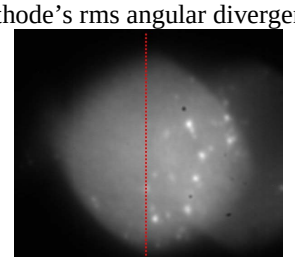

Figure 9. Collected beamlet from pepperpot after 5 mm drift length.

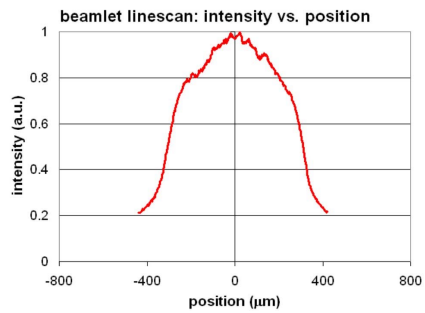

Figure 10. Linescan of observed beamlet in Fig 9.

The rms radius of the beamlet is estimated at approximately 250 μm, which corresponds to an rms angular divergence of ~50 mrad. The pepperpot aperture acts as a diverging lens and increases the measured divergence at the collector. The focal length is given by $f = 4L$, where L is the anode-cathode spacing. The focal length of 1.2 mm reduces the measured rms angular divergence to ~38 mrad. A single tip simulated in POISSON and GPT produces an estimated rms angular spread of 40 mrad, in good agreement with experiment [5]. For a beam energy of 2 kV $(\beta\gamma \approx 0.09)$ and a cathode diameter of 1mm, the normalized x-emittance is

$$\varepsilon_{N_x} = \beta\gamma\sigma_x\sigma_{x'} \approx 1.4 \text{ mm-mrad}. \qquad (1)$$

In the next set of emittance measurements our results will be refined by using arrays with more than ten times the emitter density of the current array.

ENERGY SPREAD

To measure the emitted energy spread we have developed a high-resolution retardation energy analyzer based on previous work at UMER [6]. The measured energy spread in a standard retardation analyzer is artificially high due to trajectories having nonzero transverse momentum. By including a cylindrical focusing electrode, the energy resolution can be improved by several orders of magnitude. A schematic of the analyzer accompanied by a cathode is shown in Figure 11.

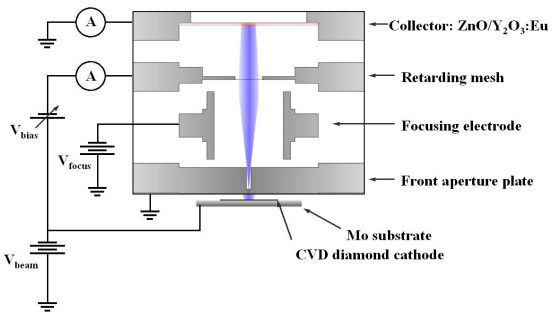

Figure 11. Energy analyzer schematic (with cathode).

In order to predict performance of the analyzer we have modeled the system in SIMION 7.0, an ion/electron optics workbench. We find that the analyzer's resolution, when the focusing voltage is optimized (97% of the beam energy), is better than the kinetic energy error in the simulation (~10 ppm).

The analyzer is incorporated into a UHV-compatible DC test stand capable of providing fields up to $\sim 50\mathrm{V}/\mu\mathrm{m}$. The cathode holder is fixed to a kinematic-style mount that allows real-time adjustment of anode-cathode planarity and spacing during high-voltage operation. Connecting the retarding mesh and cathode to the same power supply allows the collection of energy spectra relative to the cathode's Fermi level. A fiberoptically-coupled bias supply with millivolt stability has been developed for controlling the energy scans programmatically. The data acquisition system is capable of energy scans lasting a fraction of a second, enabling the observation of energy spectra from transient surface adsorbates.

The analyzer has been successfully tested using a 3x24 diamond field-emitter array. The array was conditioned for uniform emission prior to testing. Energy spectra were averaged over hundreds of scans and were taken as a function of focusing voltage to find the optimum setting. The results in Figure 12 reproduce the value suggested by simulations, $V_{focus} = 0.97 V_{beam}$.

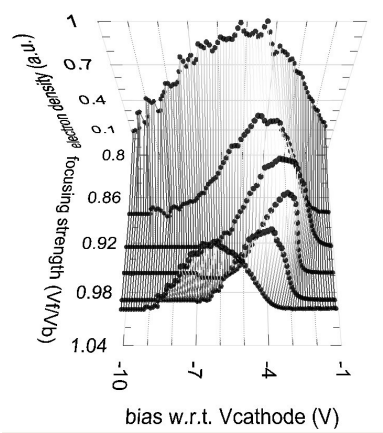

Figure 12. Electron energy spectra as a function of focusing voltage. Optimum focusing is found to be 97% of the cathode voltage.

The minimum measured energy spread has a FWHM of ~1.3 eV, which is large compared to that of metallic field emitters (~0.3 eV). This disparity may be due to adsorbate modification of the energy spectrum, or it may be a property of the diamond cathode. There may be no reason to expect that a complicated carbon structure with a mixture of sp^2 and sp^3 bonded carbon should behave as a metallic emitter in this respect.

In the next round of experiments spectra will be taken at UHV as opposed to the high pressure of the current experiments, ~10^{-7} Torr. Self Joule heating of the nanotips during high-current operation will remove surface adsorbates and enable measurement of energy spectra from clean diamond emitters

CONCLUSIONS

We have presented preliminary results in the exploration of various properties of diamond FEAs. Ungated diamond FEAs have demonstrated per-tip-current capability which scales to 100-300 A/cm^2 DC for fine pitch arrays. We expect that RF-pulsed operation in a higher vacuum environment will unlock new levels of performance for these devices. The measured transverse emittance for ungated arrays is encouragingly low and suggests sufficient beam quality for successful use in FELs. The emittance will be further reduced by self-aligned collimating electrodes built in to gated devices. We measure energy spreads from these emitters of ~1.3 eV, suggesting interesting emission physics that departs from Fowler-Nordheim-emission theory. These measurements will be refined in coming months as the full potential of diamond FEAs continues to be explored.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge Mic Howell for assistance with the diamond CVD systems, and Travis Wade for phosphor procurement. This work was supported by the Office of Naval Research under grants N000014-06-0572 and N000014-07-1-1037.

REFERENCES

[1] C. A. Spindt, J. Appl. Phys. **39,** 3504 (1968)
[2] J. D. Jarvis et al, *Toward Energy Spread Measurements of Diamond Field-Emitter Arrays*, these proceedings
[3] J. D. Jarvis et al, *Uniformity Conditioning of Diamond Field-Emitter Arrays*, these proceedings
[4] J. D. Jarvis et al, *Transverse Emittance of Diamond Field-Emitter Arrays*, these proceedings
[5] W. P. Kang, J. L. Davidson, M. Howell, B. Bhuva, D. L. Kinser, D. V. Kerns, Q. Li, J. F. Xu, J. Vac. Sci. Technol. B, **14**, 2068 (1996)
[6] Y. Cui, Y. Zou, A. Valfells, M. Reiser, M. Walter, I. Haber, R. A. Kishek, S. Bernal, and P. G. O'Shea, Rev. Sci. Instrum. **75**, 8 (2004)