RESONANT TUNNELING AND EXTREME BRIGHTNESS FROM DIAMOND FIELD EMITTERS AND CARBON NANOTUBES

J. D. Jarvis, N. Ghosh, B. L. Ivanov, J. L. Kohler, J. L. Davidson*, and C. A. Brau

Vanderbilt University, Nashville, TN, U.S.A.

Abstract

We report recent advances in the development of electron sources of extreme brightness approaching the quantum degenerate limit. These cathodes comprise either a diamond field emitter or carbon nanotube and an individual adsorbed atom or molecule. Both emitters are covalent carbon structures and thus have the benefits of high activation energy for atomic migration, chemical inertness, and high thermal conductivity. The single adsorbate produces surface states which result in dramatic resonant enhancement of the field emission current at the allowed energies of those states. The result is a beam with a narrow energy spread that is spatially localized to roughly the size of a single atom. Thus far, we have observed short lived (~1 sec) beams from residual gases of ~6 microamps corresponding to a normalized transverse brightness of ~3e18 A/m^2-str. Whereas conventional field emitters have a quantum degeneracy of <1e-4, we estimate the degeneracy of our observed beams to be ~0.1. The use of metal adsorbates should stabilize the effect, allow higher current operation, and provide a long lived source whose brightness approaches the quantum limit.

BACKGROUND

The development of low-emittance, high-brightness electron sources is of significant importance to fields such as electron microscopy [1,2], point-projection microscopy [3], Gabor holography [4,5], and beam-driven radiation sources[6,7]. Emittance, the transverse phase space occupied by the beam, is fundamentally limited by the Heisenberg uncertainty principle, so that

$$\varepsilon = \Delta x \Delta p_x \geq \hbar / 2$$

where $\hbar$ is Planck's constant h divided by 2π. Brightness, the density in phase space, is fundamentally limited by the Pauli exclusion principle to a single spin pair per h^3 [8]. Accordingly, the degeneracy is defined by

$$\delta = \frac{h^3 I}{2 q_e \Delta x \Delta p_x \Delta y \Delta p_y \Delta E} \leq 1$$

where q_e is the electronic charge, h is Planck's constant, and I is the electron beam current. For most electron sources quantum effects are too small to be of any practical consequence. However, the generation of electron beams with quantum-limited emittance and high degeneracy is possible with certain types of field-emission cathodes.

In this paper we discuss two types of field emitters, carbon nanotubes and sharpened diamond pyramids, that can be used to form quantum-limited beams. Multi-wall carbon nanotubes (MWCNTs) have been extensively studied as sources of high-brightness electron beams [9,10]. Recently, chemical-vapor deposited (CVD) diamond field emitter arrays (DFEAs) have demonstrated promise as high-brightness electron sources [11]. The composition and fabrication of these devices are detailed elsewhere [12,13]. Both CNTs and CVD diamond emitters are covalent carbon structures and thus have the benefits of high activation energy for atomic migration, chemical inertness, and high thermal conductivity. These properties are very important for achieving high-intensity, stable emission.

If an atom or molecule is adsorbed on the surface of the emitter it modifies the potential at the emitter-vacuum interface, and can produce surface states through which resonant tunneling can occur. This process is shown schematically in Figure 1. Resonant tunneling can increase the locally emitted current by orders of magnitude while maintaining a narrow energy spectrum. The current through a single adsorbate can extend well into the microampere regime, depending on the adsorbates' binding energy. With this current level, the narrow spectrum (~0.5 eV FWHM) and spatial localization of the enhancement (~1 Å) results in beam brightness near the quantum limit.

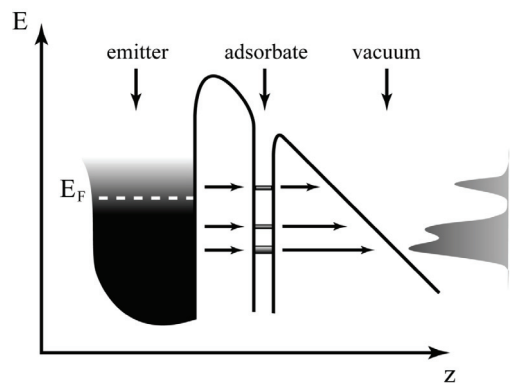

Figure 1: Energy diagram of the emitter-vacuum interface in the presence of an adsorbate and the resulting field emission energy distribution.

EXPERIMENTS AND RESULTS

The experiments using DFEAs were performed in a UHV test stand shown schematically in Figure 2, with a base pressure of less than 10^{-10} Torr. The cathode holder was mounted on a gimballed kinematic mount that allowed the adjustment of the anode-cathode spacing and parallelism during operation. The full details of our spectral measurements on DFEAs are given in our previous report [14]. The important results for the present discussion are shown in Figure 3. In these data an individual adsorbate has increased the current, in a stepwise fashion, by more than an order of magnitude while the spectral width of the emitted beam has remained the same. This means the longitudinal phase space density has increased dramatically. As noted previously the spatial localization of the emission also results in a significant increase in the transverse phase space density. Put together, the brightness has increased by many orders of magnitude. Thus far, these diamond emitters have been shown to support stable emission currents up to 15 μA DC, which is expected to be more than sufficient for production of a degenerate beam.

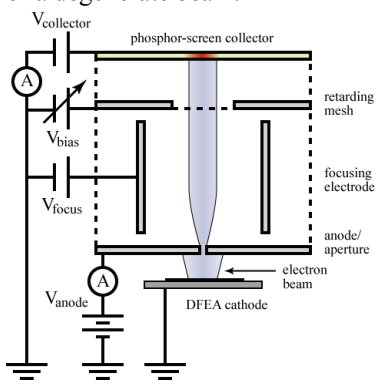

Figure 2: A schematic representation of the energy analyzer/ DFEA system. The 50-μm aperture admits current from a single emitter, which is then collimated and subsequently analyzed with a retarding mesh.

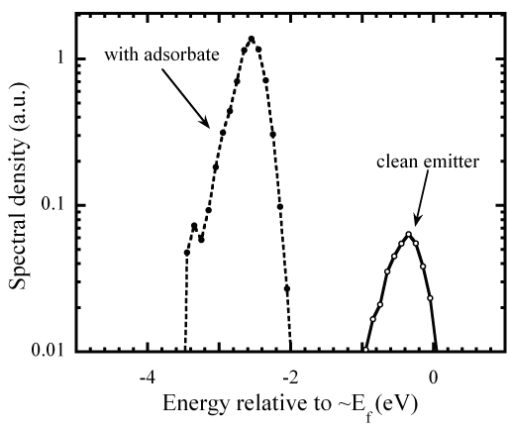

Figure 3: In some cases, single adsorbate events increased the emission current by more than an order of magnitude without affecting the spectral width or shape.

Measurements of the emission from individual MWCNTs are performed in a conventional UHV field-emission microscope (FEM), shown schematically in Figure 4. This apparatus allows for simultaneous acquisition of FEM images and electron energy spectra. A system of electrostatic deflectors enables spectral analysis of different parts of the electron beam spot. A joystick controlled LabVIEW interface allows the user to quickly position the beamlet from an adsorbate over the probe hole of the analyzer. The cathode is typically an individual closed-cap MWCNT (~10 nm diameter, ~1.7-μm length) mounted on a tungsten needle (~35-nm tip radius) by E. C. Heeres at Leiden University [15]. The pressure during high-current operation (>1 μA) is typically less than 10^{-9} Torr.

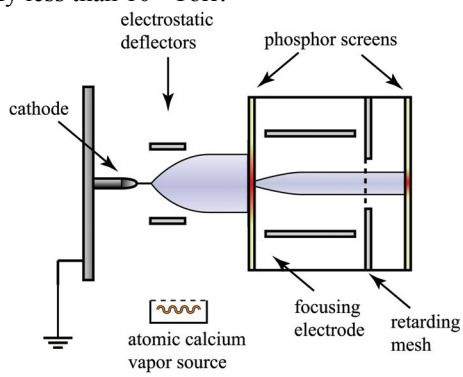

Figure 4: FEM system includes electrostatic deflectors and an electron energy analyzer with a phosphor coated front plate. Various atomic vapor sources can be integrated. Presently, a $CaCO_3$ based Ca source is used.

Our previous measurements in a simplified FEM demonstrate the high currents that are possible through individual adsorbates on MWCNTs [14]. Figure 5 shows a typical high-current adsorbate event taking place on a clean surface of a MWCNT [14]. Prior to taking these data the emitter surface was cleaned by high current operation (>5 μA) for tens of minutes. The cleaning procedure was terminated once pentagonal rings were visible in the emission pattern. The appearance of an adsorbate on the emitter surface is clearly seen in the emission image and in the total current. This adsorbate was likely a residual gas molecule, polarized and attracted by the large field gradient near the emitter.

The full width at half maximum (FWHM) of the spot is roughly 4 mm and the transit time of the electrons to the phosphor is 1.5 ns, so the rms transverse momentum uncertainty is $\Delta p_x \sim 10^{-24}$ kg-m/s. If we assume that the rms emission spot size from a single adsorbate is $\Delta x \sim 10^{-10}$ m, then we see that the transverse emittance is $\Delta x \Delta p_x \sim 10^{-34}$ J-s. This is close to the Heisenberg uncertainty limit and suggests a beam with excellent transverse coherence. Although resonant tunneling through a single adsorbate can produce a much narrower spectrum, if we assume an energy spread ΔE ~ 1 eV for this 6 μA beamlet, we find that the corresponding

degeneracy would be approximately δ = 0.1. The corresponding normalized transverse brightness of ~3e18 A/m^2-str. While the brightness of this beam is impressive, its lifetime is less than impressive. At these current levels, even the most tightly bound residual gases (typically 100's of meV) have short lifetimes due to joule heating. Metal atoms can have binding energies of several eV on the surface of a CNT [16]. The lifetime of an adsorbate on the emitter surface is exponentially sensitive to the binding energy. For example, at 1000 K, the ratio of the lifetimes for adsorbates having binding energies of 2 eV and 0.2 eV is approximately 10^9.

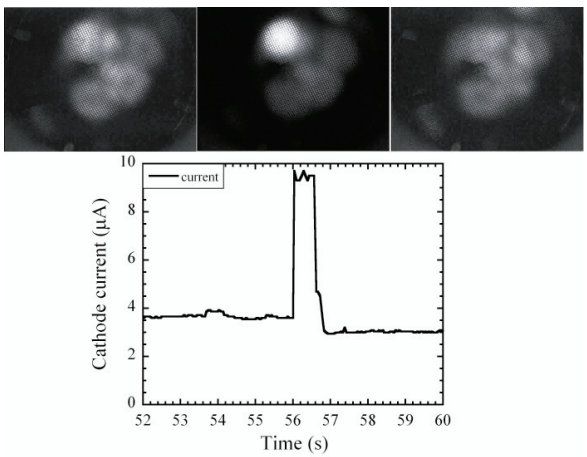

Figure 5: 6 μA beam coming from an individual adsorbate (likely a residual gas molecule); estimated degeneracy (~0.1).

The most straightforward way to deposit metal atoms on the MWCNT surface would be sublimation of a heated metal filament. However this method would likely result in the deposition of large clusters of atoms. This would tend to broaden the emitted energy spectrum and increase the effective source size, thereby reducing the brightness substantially. For single atom deposition we require an atomic metal vapor sources. For simplicity we are beginning with calcium, which is readily available in a convenient carbonate form ($CaCO_3$). The $CaCO_3$ can be dusted onto a filament, such as molybdenum, with a variety of methods. When the filament is heated to ~1250 K the carbonate decomposes leaving calcium oxide on the molybdenum surface. Upon further heating to ~1450 K, the molybdenum is oxidized and atomic calcium is liberated from the surface[17]. The general procedure for single atom deposition will be as follows:

- using the cathode heater filament, elevate the cathode temperature to ~1000 C to clean emitter surface and prevent residual gas adsorption. This temperature should not be sufficient to prevent metal atom adsorption.
- activate metal vapor source while monitoring FEM image and total current, in the manner of Fig. 5.
- upon detection of an adsorption event disengage vapor source
- monitor FEM image while increasing cathode temperature to encourage adsorbate migration to region of highest field.
- reduce temperature to increase adsorbate lifetime while still preventing residual gas adsorption.

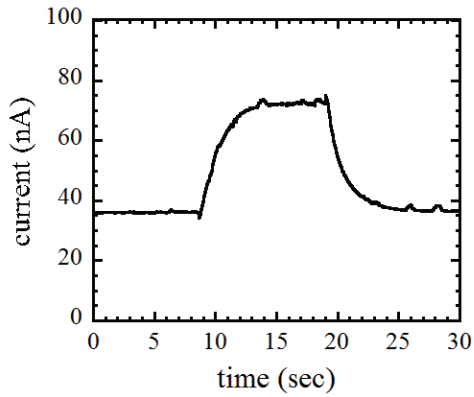

Figure 6: Current vs. time for cathode temp. cycle up to ~1000 C. The smooth increase and stable emission suggests a clean emitter surface.

Presently, we are testing these procedures. Figure 6 shows the emitted current from a clean single MWCNT during a single thermal cycle up to ~1000 C and back. The increase in current is due to increased tunneling at higher energies as a thermal tail develops in the electron population. This is seen more directly in the spectral measurements shown in Figure 7.

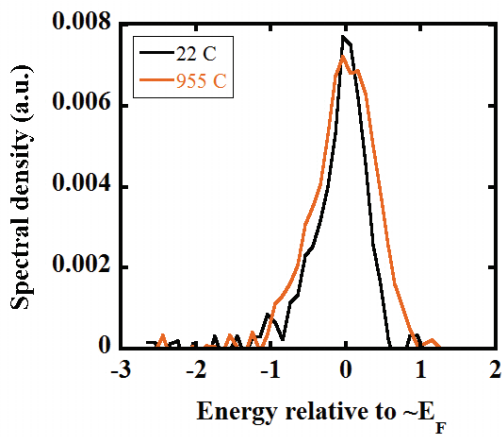

Figure 7: Measured energy spectrum for a clean CNT at room temperature and at ~955 C.

CONCLUSIONS

In conclusion, we have considered the effects of adsorbates on the surface of carbon nanotubes and diamond field emitters. We have demonstrated that adsorbates can be used to produce electron beams of extreme brightness, reaching nearly to the quantum degenerate limit imposed by the antisymmetry of the fermionic wavefunction. We are actively investigating

the use of single atom metal adsorbates as a way of producing a long lifetime quantum degenerate electron beam source. The development of such a source would be a groundbreaking achievement.

REFERENCES

[1] P. W. Hawkes and E. Kasper, *Principles of Electron Optics* (Academic Press, London, 1994)

[2] J. C. H. Spence, *High-resolution electron microscopy* (Oxford, New York, 2009)

[3] J. C. H. Spence, W. Qian, and A. J. Melmed, Ultramicroscopy 52 (1993),473

[4] D. Gabor, Proc. Roy. Soc. London A 197 (1949), 454

[5] J. C. H. Spence, X. Zhang, and W. Qian, *Proceedings of the International Workshop on Electron Holography* (Elsevier, Amsterdam, 1995)

[6] M. J. Rhee, Phys. Fluids **B**, **4**, 1674 (1992)

[7] M. Reiser, *Theory and Design of Charged Particle Beams* (Wiley, New York, 1994)

[8] J. C. H. Spence and M. R. Howells, Ultramicroscopy, **93**, 213 (2002)

[9] N. de Jonge, Y. Lamy, K. Schoots, and T. H. Oosterkamp, Nature **420**, 393 (2002)

[10] N. de Jonge, M. Allioux, J. T. Oostveen, K. B. K. Teo, and W. I. Milne, Phys. Rev. Lett. **94,** 186807 (2005)

[11] J. D. Jarvis, H. L. Andrews, C. A. Brau, B.-K. Choi, J. L. Davidson, W.-P. Kang, and Y.-M. Wong, J. Vac. Sci. Technol. B **27**, 5, pp. 2264-2269 (2009)

[12] W.-P. Kang, J. L. Davidson, M. Howell, B. Bhuva, D. L. Kinser, Q. Li, and J.-F. Xu, Vacuum Microelectronics Conference, 1995. IVMC, pp. 287-291, 30 July – 3 August 1995

[13] W.-P. Kang, J. L. Davidson, M. Howell, B. Bhuva, D. L. Kinser, D. V. Kerns, Q. Li, and J.-F. Xu, J. Vac. Sci. Tech. B, **14**, 2068 (1996)

[14] J. D. Jarvis, H. L. Andrews, C. A. Brau, B. K. Choi, J. Davidson, W.-P. Kang, C. L. Stewart, and Y.-M. Wong, *Proceedings of the International Free-Electron Laser Conference*, TUPC59, (2010), Liverpool, UK.

[15] E. C. Heeres, A. J. Katan, M. H. van Es, A. F. Beker, M. Hesselberth, D. J. van der Zalm, and T. H. Oosterkamp, Rev. Sci. Instrum. 81, 023704 (2010)

[16] E. Durgun, S. Dag, S. Ciraci, and O Gulseren, J. Phys. Chem. B **2004**, 108, 575-582

[17] E. W. Plummer and R. D. Young, Phys. Rev. B **1**, 2088-2109 (1970)