

Carbon Nanotube Field Emitters for Nanoklystrons and other High Frequency Tube Sources

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Abstract—Traditional submillimeter-wave vacuum tube sources (backward wave oscillators), now available only from the former Soviet Union, have been a mainstay for generating modest amounts (mW) of narrow band (phase lockable) tunable CW RF power at frequencies from 50 to 1200 GHz. Recently groups in both Europe and the US have recognized the potential for producing these expensive, hand-assembled sources through monolithic techniques. A major step in this process is the formation of the electron gun – typically a hot cathode operating at high temperature and requiring significant power. We have developed a cold field emission cathode based on multi-walled carbon nanotube bundles. This cathode can be integrated directly onto a silicon circuit (as part of a monolithic process), has demonstrated emission at only a few volts/ μm and current densities (unfocused) of more than $20\text{A}/\text{cm}^2$. This short paper summarizes the design, fabrication and applications for these new field emission devices as components in proposed monolithic terahertz vacuum tube sources.

I. INTRODUCTION

After more than three decades, the acquisition of portable, narrow band, coherent CW sources of tunable RF power in the submillimeter wave frequency bands (300-3000 GHz) remains a major stumbling block for many active (and passive) sensor systems. Although significant progress has been made on solid-state sources derived from power amplifiers and frequency multiplier chains on the low frequency side, and by new quantum cascade lasers at the higher terahertz frequencies, there still exists a significant gap between 500 and 2500 GHz where low cost, modest efficiency, mW level oscillators would provide a tremendous advantage in generating the local oscillator power required by heterodyne downconverters, the source power needed for active imagers and radars or the broadband probe beams required by high resolution spectrometers. Although commercial vacuum tube devices of varying designs (backward wave oscillators and traveling wave tubes) have been available in the millimeter-wave bands, and even up to 1200 GHz for many years, these devices typically require high magnetic fields (kGauss) making them both heavy and bulky; operate at very high voltages (5-10kV); suffer from limited lifetime (due to cathode decay); and generate large amounts of heat (requiring water or forced air cooling). They also require hand assembly with very tight mechanical tolerances driving up the cost and making consistent performance difficult to achieve.

In the past several years groups in the UK and in the US have begun to explore alternative vacuum tube sources based on monolithic construction techniques employing either micromachined or silicon etched cavities and beam guiding structures [1,2]. One way of eliminating the high voltages and strong magnetic fields required by traveling wave tubes and backward wave oscillators altogether is to exploit the short beam tunnel and compact resonator design of the reflex klystron [3]. These tubes were traditionally realized at frequencies as high as 200 GHz in compact (5x5x5cm) metal packages [4]. However the electron stream is still provided by a hot cathode, which necessitates high power dissipation and relies on an extremely high quality vacuum for extended lifetime. Our group has been developing an alternative to the hot cathode using field emission devices based on carbon nanotube (CNT) bundles. These cathodes have already demonstrated low voltage emission (2-5V/ μm) with significant current density ($>20\text{A}/\text{cm}^2$) at modest currents ($>1\text{mA}$). In addition the CNT bundles can be fabricated directly on silicon substrates (with proper catalyst), in a variety of configurations, and with integrated emitter grids. In this form they can serve as the building blocks for a fully integrated, monolithic vacuum tube circuit which we have designated the “nanoklystron” [1].

II. NANOKLYSTRON CIRCUIT

A cross section of the nanoklystron circuit appears in Fig. 1 along with some of the building blocks that have been developed to realize the structure monolithically. The operation is that of a standard reflex klystron analyzed more than 50 years ago [5]. The electrons are injected from the cathode and focused into the short beam tunnel that crosses through a hole in the center of the resonant re-entrant cavity. An integrated field-emission grid and electrostatic focusing elements replace the heater and static magnetic field that form the basis of traditional traveling wave tube oscillators. An accelerating voltage (expected to be between 500 and 1000 V) sends the electrons through the cavity where they are reflected in phase by a repeller (-50 to -250 V) and forced to bunch and resonate within the high Q structure. The power is bled off through a small slot in the cavity wall and coupled out via a single mode waveguide. The entire structure (cavity, beam tube, coupling and waveguide output port) is formed using reactive ion etching (RIE) in two halves on two silicon wa-

fers, which are bonded together and sealed under vacuum to form the completed structure. The cathode (formed from distributed CNT bundles) is grown in place, and the shaped repeller is added in a separate step. To date we have completed various pieces of the complete nanoklystron structure including the re-entrant cavity and stepped waveguide output transformer and the CNT cathode with integrated emitter grid. No RF testing has been performed and we are still developing the repeller circuit, vacuum sealing techniques and integrated RF output window. Much of the focus has been on the field emission cathode which, according to simulations, requires current densities of at least as much as 100 A/cm^2 (over a diameter of 20-50 μm) in order to produce oscillations at frequencies approaching 1 THz.

III. NANOTUBE CATHODES

Traditional high current density hot cathodes can be formed from oxide coated surfaces and series arrays of electrostatic focusing elements. However these cathodes are typically not amenable to monolithic fabrication nor compatible with the modest vacuum environments achievable in silicon RIE processes. Cold cathodes (Spindt devices) employing oxide coated silicon tips, on the other hand, have been monolithically fabricated in very high density arrays [6]. Unfortunately these oxide coated cathodes also require extremely high vacuum and tolerate little or no back sputter. Some years ago we realized that arrays of carbon nanotubes might yield electron densities sufficient to generate oscillations in a small microcavity if enough tips could be made to emit. Moreover carbon nanotubes were known to be more tolerant of electron back-scatter and low vacuum conditions (10^{-5} rather than 10^{-9} Torr). The small diameter of carbon nanotubes (single walled tubes can have a diameter $<1 \text{ nm}$) enables efficient emission at low fields, despite their relatively high work function ($>4.5\text{eV}$). Even much thicker multi-walled tubes emit at a few $\text{V}/\mu\text{m}$.

We began our investigations on highly ordered multi-walled CNT's grown in an alumina honeycomb cell structure [7] (Fig. 2). The ordered pores are produced by anodizing high-purity aluminum substrates. The tip density is about $100 \text{ tips}/\mu\text{m}^2$ with a typical tube diameter of $\sim 40\text{nm}$. Measurements of emitter current were performed in an ultra-high vacuum chamber using a separate grid and adjustable anode. Currents of $\sim 100 \mu\text{A}$ were extracted from an area exceeding $100\mu\text{m}^2$ ($1\text{A}/\text{cm}^2$) indicating that only a very few of the CNT tips were actually emitting. We then proceeded to test a variety of less uniform CNT structures, including single and multi-walled tubes fabricated on various catalysts in ordered and disordered arrangements (Fig. 3). The multi-walled tubes consistently performed better under emission tests. These were then grown on two different substrates (silicon and SiO_2) with two different catalysts, Fe and Ni, and with a large density gradient across the wafer. The samples grown with

Fe catalysts on SiO_2 had the strongest emission, with peak current occurring at very specific densities. On close examination, the region with highest emission was composed of nanotubes that had twisted together to form long rope-like shapes (Fig 4-right). We then catalyzed these rope-like structures over regions of varying diameter (0.2-5 μm) and with varying spacing (2-100 μm) across a wafer (Fig. 4-left). When characterized in the UHV chamber, the highest emission density was achieved from arrays of CNT bundles of 1 μm and 2 μm diameter with 5 μm edge-to-edge spacing (Fig. 5). An emission density in the range of 1.5 to 1.8 A/cm^2 was routinely achieved at fields as low as 4 $\text{V}/\mu\text{m}$. Occasionally we achieved emission density as high as 6.0 A/cm^2 at 20 $\text{V}/\mu\text{m}$. We believe these are the best reported results with this type of diode test and at such low fields [8]. Finally, we integrated emission grids onto the nanotube bundle arrays (Fig. 6) and measured the grid current as a function of applied voltage. Preliminary results yield more than 20 A/cm^2 at fields below 6 $\text{V}/\mu\text{m}$ with currents in the mA range. With added focusing, these bundles are now capable of reaching the current densities required by the THz nanoklystron circuit. Near term plans include incorporation of electrostatic focusing electrodes onto grid bundles, integration of the CNT cathode with the silicon cavity, and fabrication of the shaped repeller. Parallel work is ongoing to produce the nanoklystron in a metallic block that can be tested with a separate hot cathode in the UHV chamber at 100, 300 and 600 GHz.

SUMMARY

Steps towards the realization of a monolithic terahertz vacuum tube source based on a traditional reflex klystron structure have been described. Special emphasis has been given to the development of a field emission cathode formed from bundled arrays of multi-walled carbon nanotubes. By growing the nanotubes on an SiO_2 surface with an Fe catalyst to get a specific tube density, and then spacing both the individual tubes as well as select diameter tube bundles in a regular fashion, record current density has been achieved at low emitter voltages. With the addition of an integrated emitter grid even higher currents have been measured, sufficient, we believe, (with external focusing) to generate the $>100 \text{ A}/\text{cm}^2$ values needed to spark oscillation in a terahertz nanoklystron.

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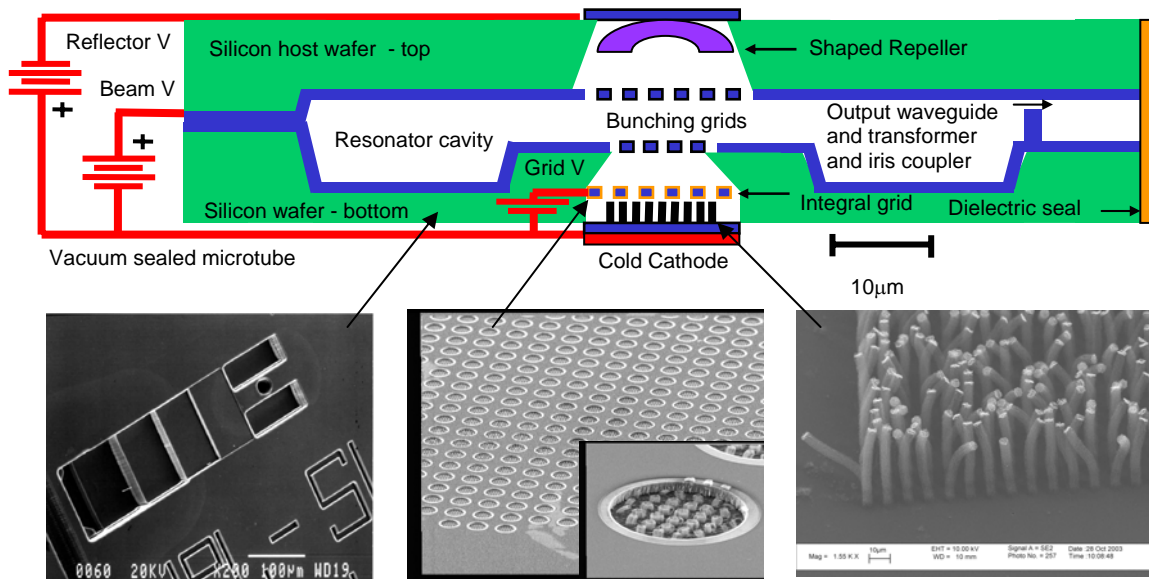


Fig. 1. Schematic cross section of a proposed nanoklystron. The cathode is composed of a carbon nanotube field emitter array with integrated grid. The cavity, beam and output waveguide are etched from two silicon wafers, which are later joined by thermocompression bonding. The repeller and cathode are drop-in parts and vacuum sealing is performed in the last step.

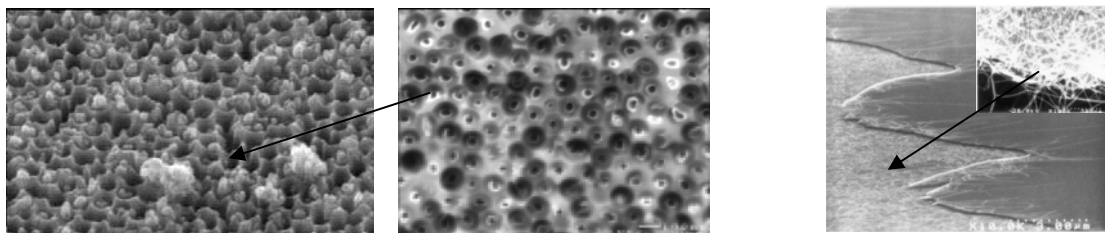


Fig. 2. Carbon nanotubes grown in a porous hexagonal lattice and (right) close up looking straight down on exposed tips.

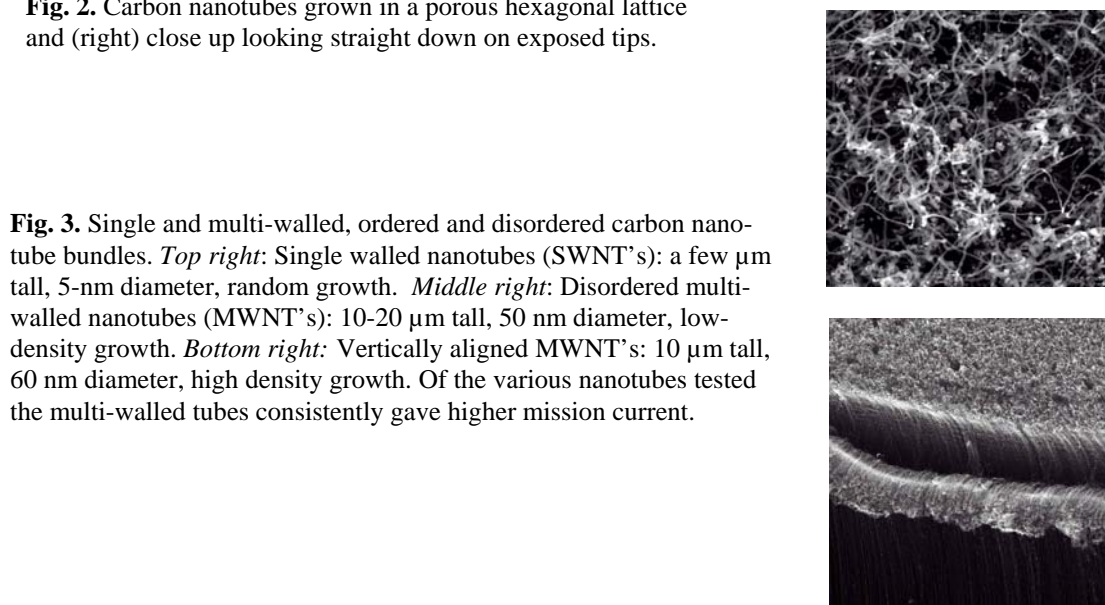


Fig. 3. Single and multi-walled, ordered and disordered carbon nanotube bundles. *Top right:* Single walled nanotubes (SWNT's): a few µm tall, 5-nm diameter, random growth. *Middle right:* Disordered multi-walled nanotubes (MWNT's): 10-20 µm tall, 50 nm diameter, low-density growth. *Bottom right:* Vertically aligned MWNT's: 10 µm tall, 60 nm diameter, high density growth. Of the various nanotubes tested the multi-walled tubes consistently gave higher mission current.

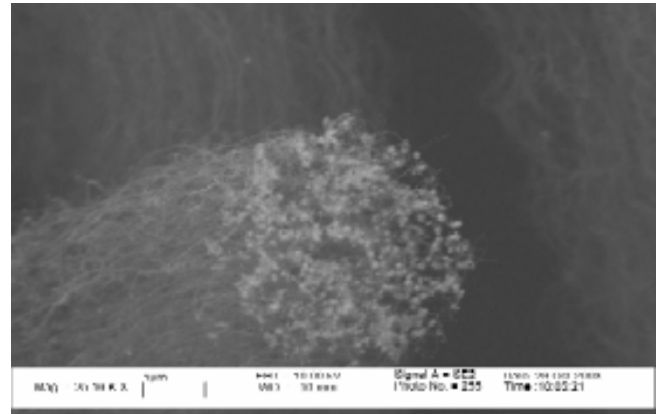
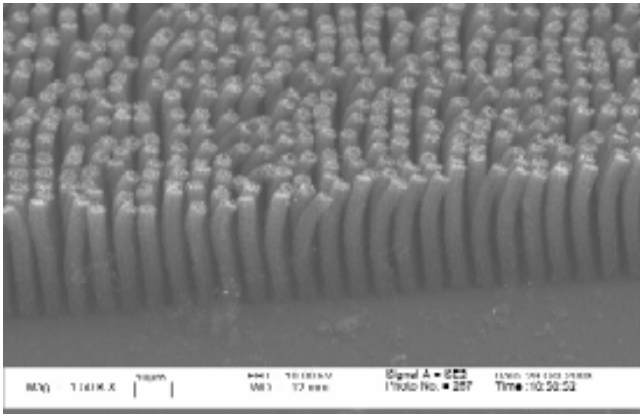


Fig. 4. SEM showing the bundled arrays of multi-walled nanotubes grown on iron catalysts. Each bundle is 5 μm in diameter and spaced 2 μm edge to edge. The MWNT's average 50 μm in height and have a tube diameter of approximately 20nm. *Right:* Close-up of one nanotube bundle in which the individual nanotubes are visible. Note the twisted rope-like structure.

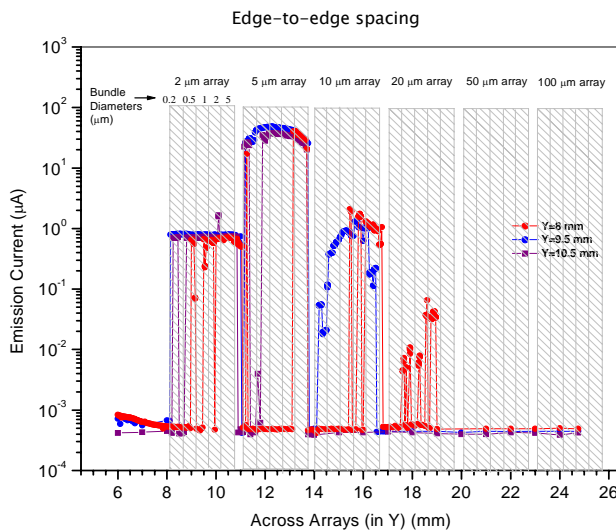


Fig. 5. Emission current as a function of bundle size and spacing as anode is scanned across the CNT wafer.

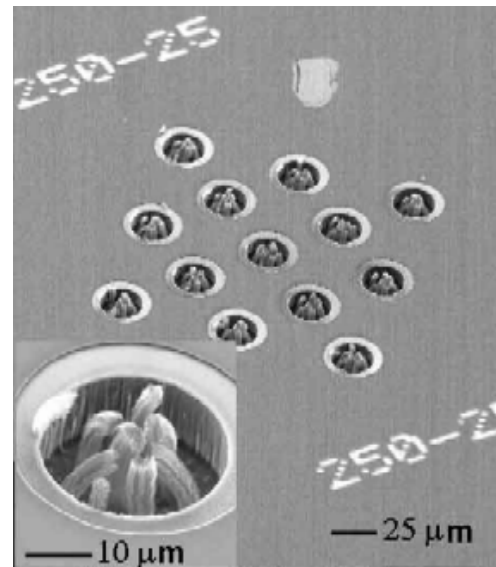


Fig. 6. MWNT arrays with integrated emission grids.

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