GROWTH OF LONG, HIGHLY ALIGNED SINGLE-WALLED CARBON NANOTUBES ON CRYSTAL QUARTZ FOR TERAHERTZ SPECTROSCOPY

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Horizontally aligned single-walled nanotubes (HA-SWNTs) with lengths in the range of 500 μm – 1 mm have potential applications in THz devices. We aimed to grow dense, highly aligned arrays on ST and R cut crystal quartz substrates using chemical vapor deposition (CVD). To maximize SWNT length, we tested different catalysts and deposition methods. We hypothesized that Cu would grow longer SWNTs than previously tested catalysts such as iron and cobalt because it uses a tip-growth mechanism. The catalyst was deposited by (1) vacuum deposition or by (2) drop-casting a solution over a physical mask. For CVD we used pure ethanol at partial pressures of 0.1 – 0.2 kPa at 800 C. The longest nanotubes (300 – 500 μm) were grown from Cu on R cut using the solution-based method. The highest density arrays were also grown from Cu and showed a narrower diameter distribution than Co-grown arrays in the 1-2 nm range, suggesting that the Cu sample contained fewer bundled nanotubes. Furthermore, Cu-based SWNTs only grew from very low concentration solutions (0.1 mmol/L $CuCl_2$ and PVP in ethanol). We observed through AFM that the range of catalyst particle sizes was broader for the solution-based method than for vacuum deposition. We suggest that Cu has the most promise for THz applications because it can inherently grow longer SWNTs due to tip-growth. We further conclude that Cu has a more limited range of active particle sizes than other catalysts, such as Co, and that this range can be most easily found by reducing concentration using the solution-based method.

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Introduction

Goal: Optimize procedure to grow horizontally aligned single-walled carbon nanotubes suitable for THz devices.

Method

1. Preparation of crystal quartz substrate
2. Catalyst deposition

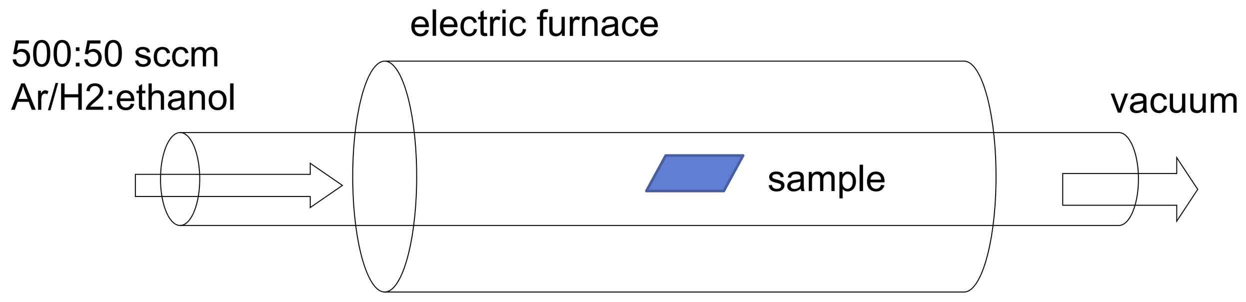
Deposition Method

Dry Method: Photolithography & vacuum deposition • Catalyst nanoparticles from vaporized metal	vs.	Wet Method: Razor scratch over physical mask • Catalyst nanoparticles dissolved in solution

Catalyst Type

Base growth: • Iron (dry method) • Cobalt (wet method)	vs.	Tip growth: • Copper

3. Chemical Vapor Deposition (CVD)
   - Flowed pure ethanol at partial pressure 0.1-0.2 kPa with mixture of 97:3 Ar:H_2 carrier gas for 15 minutes at 800 °C.



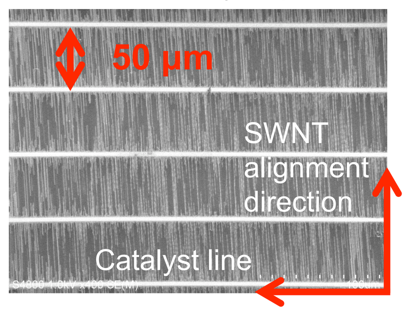
4. Analysis
   - Scanning Electron Microscopy (SEM)
   - Atomic Force Microscopy (AFM)
   - Raman Spectroscopy

Results & Discussion

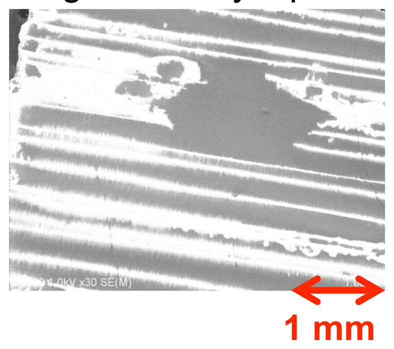
Catalyst Deposition

SEM images of SWNT arrays

Photolithography — Uniform catalyst pattern

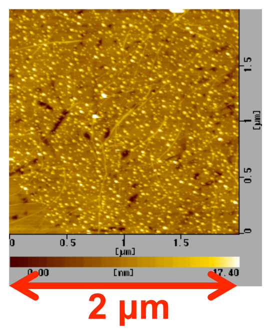


Razor Scratch — Irregular catalyst pattern

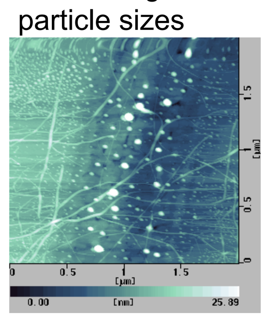


AFM images of catalyst lines

Dry method — Uniform particle sizes



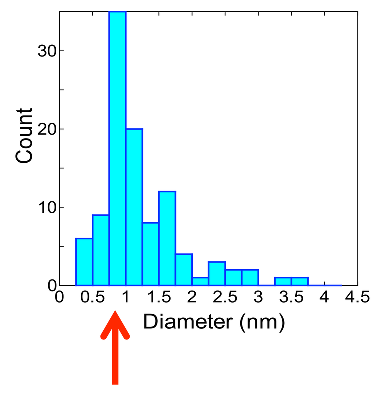
Wet method — Broad range of particle sizes



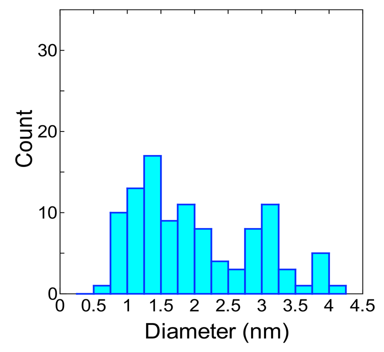
AFM & Raman Analysis

AFM: Diameter Distribution

Copper

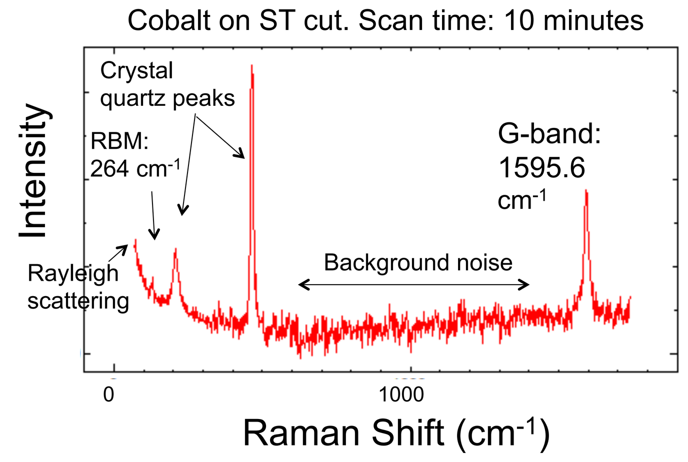


Cobalt



Cu-grown SWNTs show tighter distribution in 1-2 nm range

Raman Spectroscopy: Chirality



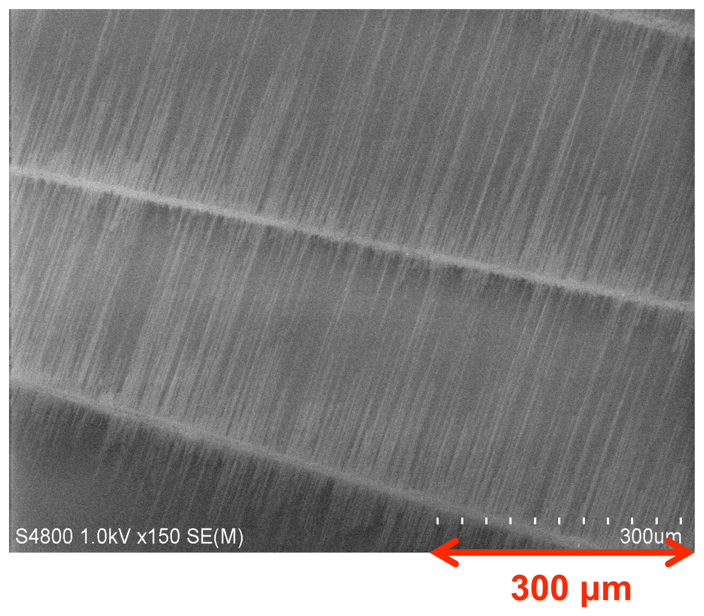
Longest SWNTs: Copper, 300-500 μm

- Results for copper on R cut:
  - Vacuum deposition of 0.2 nm and 1 nm nominal thickness from copper wire X X
  - Solutions (ethanol):

Conc. $CuCl_2$ (mmol/L)	Conc. PVP (mmol/L)	SWNTs?
1	10	X
1	100	X
10	10	X
0.1	1	O

➢ Reducing concentration reduced aggregation, creating smaller catalyst particles

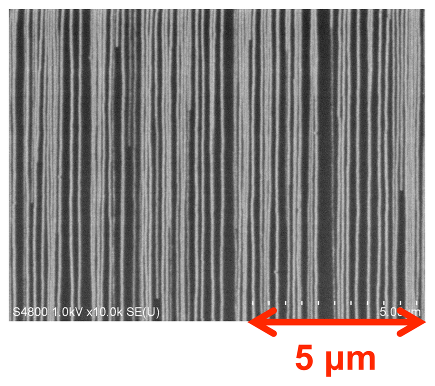
SWNT array grown from 0.1 mmol CuCl2: 1 mmol PVP per liter ethanol solution

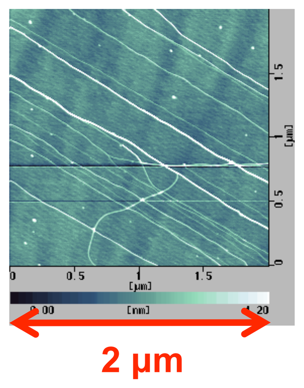


SWNTs show atomic scale alignment for ST cut and etched, annealed R cut

Density: Copper, 7-8 CNT/μm & Cobalt, 5-6 CNT/μm

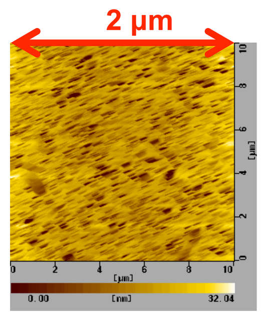
SEM & AFM images of Co-based SWNTs on ST cut



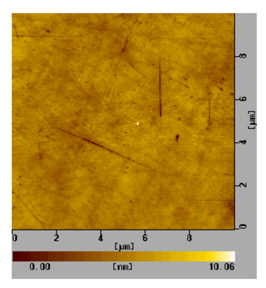


AFM Observations of ST vs. R Cut Surface Roughness

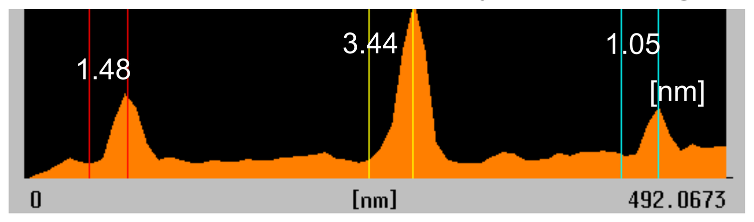
R cut



ST cut



AFM images of ST cut show SWNTs easily distinguishable from smooth quartz surface as opposed to "noisy" R cut image



Conclusion

- Wet method is the best way to test new catalysts because it deposits a broad range of catalyst particle sizes
- Copper is the most promising catalyst for THz applications because of its ability to grow long SWNTs with high density, alignment, and limited bundling
- Copper can only grow SWNTs from a limited range of small catalyst particles
- There are differences between ST and R cut surface roughness, but the effect on SWNT growth is unclear

Acknowledgements

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