SUPPORTING INFORMATION

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Sample Preparation and Characterization

VACNT arrays were grown by PECVD from sputtered nickel catalyst dots patterned with the desired spacing and symmetry through electron beam lithography; an overview of the processes is summarised here.

1. Electron beam lithography was used to pattern registration marks to aid beam focussing, then dot arrays from which catalyst dots were formed.

1.1 A 4” diameter Si wafer of 500um thickness with 28um thermal oxide was patterned with registration marks as follows:

• Sonication, 10 mins at room temperature

• Rinse in IPA and blow dry with N2

• Bake on hotplate at 180°C for 2 mins

• Spincoat with UV3, 60rpm for coating then 5000rpm for 1 min to dry • Soft bake on hotplate, 120°C for 1 minute • Load into nanobeam and pattern registration indices followed by registration marks at a dose of 0.3 C.m-2

• Remove and post bake (chemical activation) at 140°C for 1 minute

• Blow cool with N2

• Develop in CD26:DI water 1:1 for 1 min

• Gently swirl in DI water for 1 min

• Gently blow dry
• Sputter 50nm W (100W for 480s in ~3.5mBar Ar)

• Lift off and sonicate

1.2 Array

• Clean wafer as in previous procedure

• Spincoat with resist (PMMA A4 495) as above • Hard bake at 180°C for 30s •
Pattern dot array (dose 6 C.m-2, beam current 5nA)

• Develop in MIBK:IPA for 70s

• Blow dry with N2

2. Catalyst sputtering

• Sputter ~7nm ITO (50W, 45s, 3.5mBar Ar)

• Sputter ~15nm Ni (100W, 105s, 3.5mBar Ar)

• Lift off overnight in acetone

• Dice wafer

3. CNT growth

• Grow CNTs with Black Magic system (750°C, 200sccm NH3, 50sccm C2H2, 10nm/s at ~5mBar)
4. Imaging of CNT arrays

Before performing any optical analysis upon the VACNT arrays it was first necessary to characterise their physical structure, a process best carried out using scanning electron microscopy (SEM). Each array was imaged at normal and tilted (30°) incidence to determine, respectively, the actual spacing and length of VACNTs in the array. The SEM images were then analysed by using a MATLAB script written specifically for the purpose. The script measured the length of the scale bar in the datazone of the SEM image then cropped the image to remove the area below the top of the datazone, leaving only the image of the nanotubes. The image was then analysed with ImageJ, a free software package with image processing and particle analysis capabilities. The analysis detected the nanotubes and fitted ellipses to their shapes, then produced a table of data based on the properties of the ellipses. The data was then further analysed with MATLAB to produce a nearest-neighbour map to determine the average spacing between the CNTs.

Figure S1: (a) SEM image of hexagonal CNT array (Wafer 3 C0) viewed from above. The overlaid red ellipses have been generated by particle analysis and their centroids used for the nearest neighbour analysis. (b) Nearest neighbour distances, showing
discontinuities where defects or edges cause larger than expected distances to nearest neighbour.

5. Sample W3 C0 CNT Length Distribution Histogram

![Figure S2: Histogram of nanotube length distribution](image)

6. Details of the samples used

**Table S1: Summary of CNT length and spacing distribution for samples used in optical bandgap measurements (av. CNT diameter = 80nm).**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Symmetry</th>
<th>Av. Spacing (nm)</th>
<th>STDEV</th>
<th>Av. Length (nm)</th>
<th>STDEV</th>
<th>CNT count</th>
</tr>
</thead>
<tbody>
<tr>
<td>W3-B1</td>
<td>Square</td>
<td>576</td>
<td>28</td>
<td>601</td>
<td>74</td>
<td>24</td>
</tr>
<tr>
<td>W3-B0</td>
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<td>28</td>
<td>561</td>
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<td>43</td>
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<tr>
<td>W3-B0</td>
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<td>33</td>
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<tr>
<td>W3-C4</td>
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<td>49</td>
<td>1014</td>
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