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Three Dimensional Imaging of Deformed Nanotubes by Metal Contacts using HAADF-STEM Tomography

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Contact resistance in carbon nanotube devices has been a significant barrier to the widespread applications of carbon nanotubes. It varies by orders of magnitude and scales exponentially with the nanotube diameter [1-2]. To understand why, high-angle annular dark field electron tomography using a scanning transmission electron microscope (STEM) was used to investigate the three-dimensional contact geometry between nanotubes and metal contacts at nanometer resolution.

Nanotubes in ethyl ether were sonicated and dispersed on a copper grid coated with a holey carbon film. Afterward, 11 Å of gold (Au), 15 Å of gold-palladium (Au-Pd), or 25 Å of titanium (Ti) were deposited by E-beam evaporation. During electron tomography, the specimen was tilted over ±70° while acquiring projected images at 2° increments. A high dynamic-range pre-amplifier was implemented to improve detective quantum efficiency from 22 % to over 90 %. This was essential for simultaneously detecting both light and heavy atoms such as carbon and gold. The simultaneous iterative reconstruction technique was used for tomographic reconstructions [3].

Figure 1a shows the reconstructed nanotube with Au clusters. The inset is a view of only the Au clusters whose contact surface to the nanotube is flat. The finding suggests that the nanotube must be deformed to accommodate the flat contact surface. A direct observation of the deformation is shown in Figure 1b, a bright field STEM image of a nanotube deformed by the Au cluster. Like Au clusters, Au-Pd clusters also deform nanotubes. Figure 2a shows the reconstructed nanotube with Au-Pd clusters. A cross-sectioned view shown in Figure 2b reveals the radial deformation of the nanotube.

Since radial deformations can open up band gaps in previously metallic nanotubes [2,4], the deformation signifies a change in the electronic structure of the nanotube. Hence, the Au and Au-Pd contacts can open up a local band gap by deforming the nanotube, creating a tunnel barrier at the contact. Electron energy-loss spectroscopy confirms a change in carbon K-edge of the nanotube deformed by the Au-Pd contacts (Figure 2c), indicating the change in the electronic structure due to the deformation. The induced tunnel barrier by the nanotube deformation can increase the contact resistance between the nanotube and the metal. To avoid the tunnel barrier then, the contact metals need to preserve the nanotube shape. We find that Ti wets to nanotubes almost completely, wrapping around the nanotubes as shown in Figure 3a and 3c. The cross-sectioned views shown in Figure 3b and 3d reveal the curved contact surface preserving the shape of the nanotube.

Previous research on nanotube contact resistance has focused on the Schottky contribution, which is determined by the band misalignment at the contact. However, we note that another significant contribution to the contact resistance is a geometrically induced tunnel barrier due to the nanotube deformation by the contact metals. Our findings suggest that an amorphous metal that completely wet to nanotubes will preserve the nanotube shape and preclude the geometrically induced tunnel barrier which will increase the contact resistance.

References
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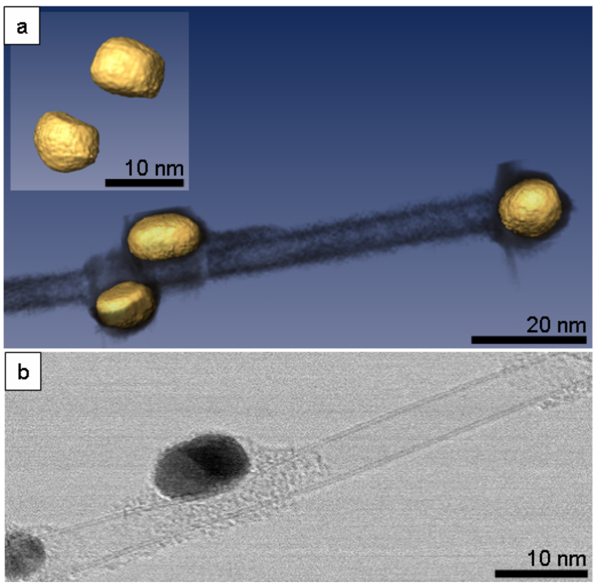

Figure 1. Deformation of carbon nanotube by Au contacts. **a)** Reconstructed nanotube (black) with Au (yellow) clusters. The inset is a view of only the Au clusters whose contact surface is flat. **b)** Bright field STEM image of the nanotube with Au cluster shows the kink at one side of the nanotube where the Au cluster is nucleated on.

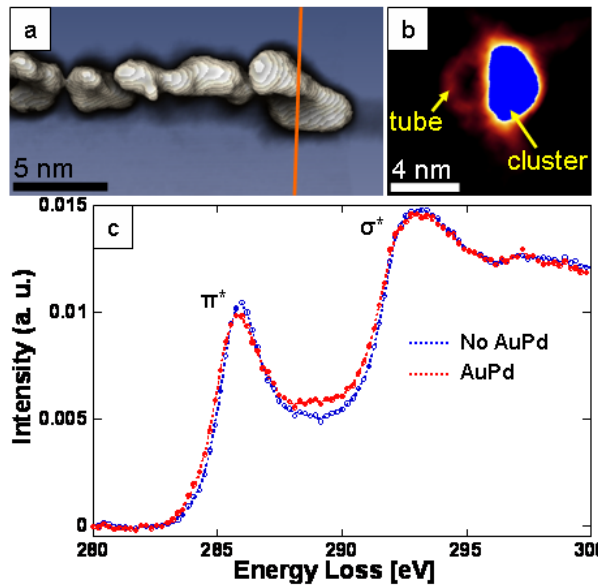

Figure 2. Carbon nanotube with Au-Pd contacts. **a)** Reconstructed nanotube (black) with Au-Pd (white) clusters. **b)** Cross-sectioned view of a Au-Pd cluster in contact with the nanotube. The nanotube is clearly deformed. **c)** EELS carbon-K edge from a Au-Pd cluster and Au-Pd cluster-free region of a nanotube.

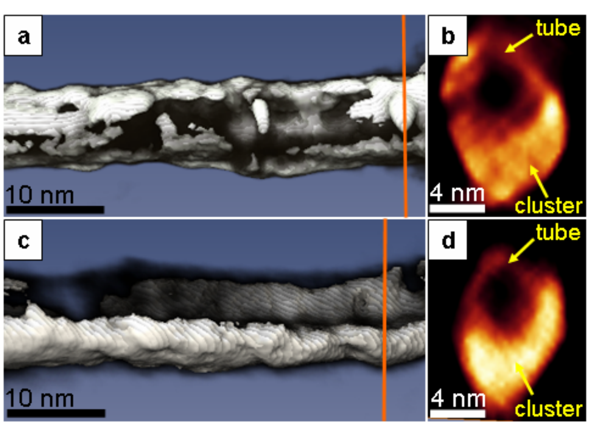

Figure 3. Carbon nanotube with Ti contacts. **a, c)** Reconstructed nanotubes (black) with Ti (white) clusters. **b,d)** Cross-sectioned views from **a)** and **c)** respectively. The contact surface is curved to follow the curvature of the nanotube. Hence, the shape of the nanotube is preserved, eliminating the deformation-induced tunnel barrier.

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