Pressure-Induced Structural Transitions in Multiwall Carbon Nanotubes

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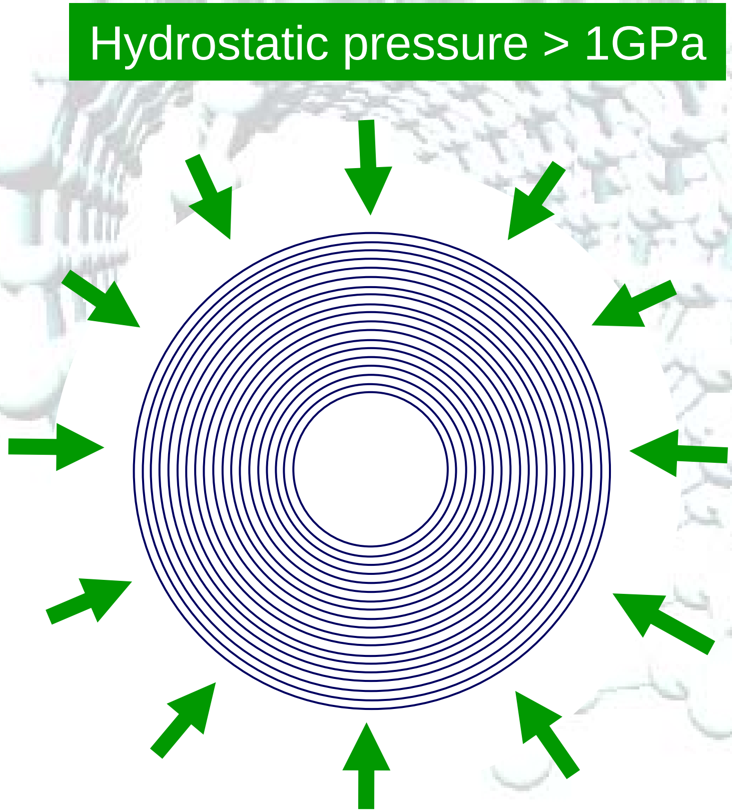
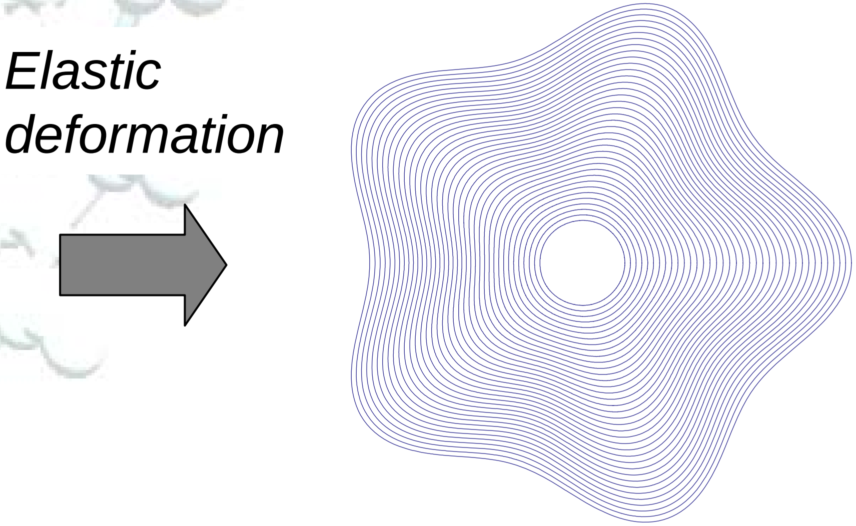
See H.Shima and M.Sato, *Nanotechnology in press*

1-5 September 2008, Trend in NanoTechnology 2008 (TNT2008) in Oviedo, Spain

0. Main Finding

= Pressure-induced **Radial Corrugation** of MWNT





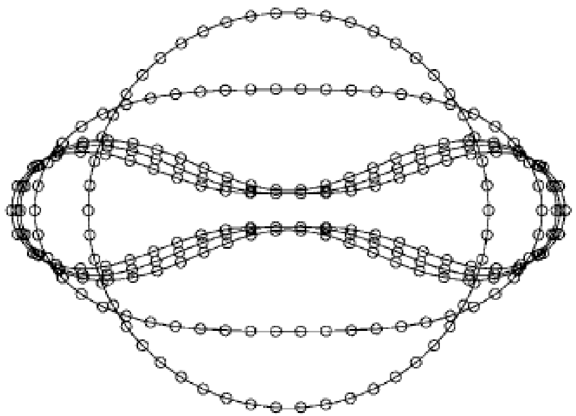
- The cross-sectional shape changes from circular to **radially corrugated** one.

1. Radial collapse of SWNTs

Carbon nanotubes are

- extraordinarily stiff in the axial direction, but
- highly flexible in the radial direction.

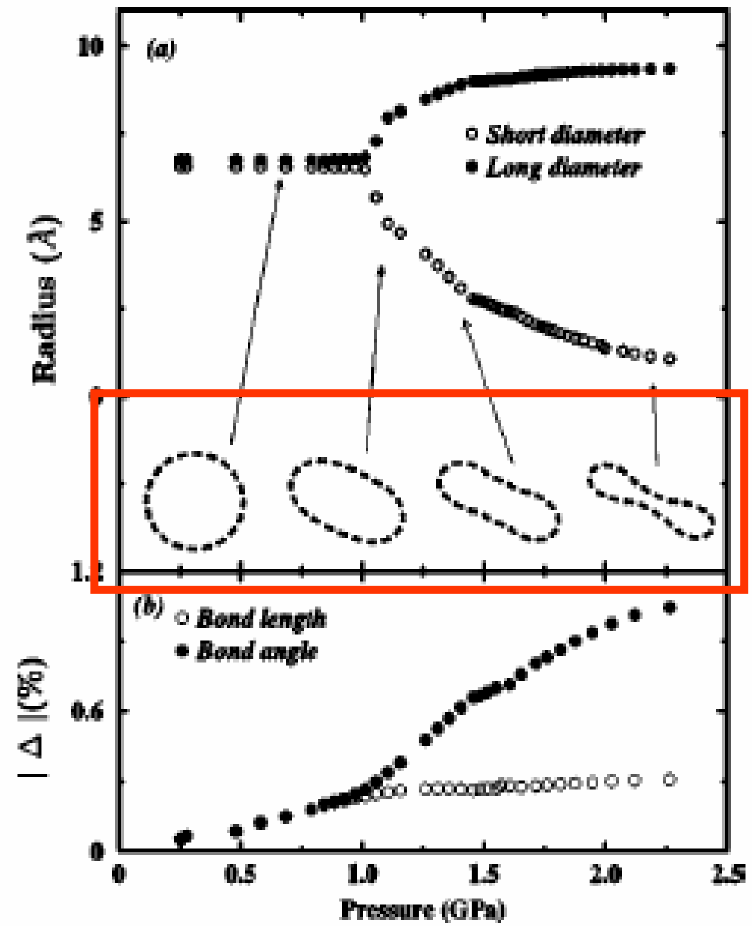
→ **Radial collapse** occurs at 1.0GPa



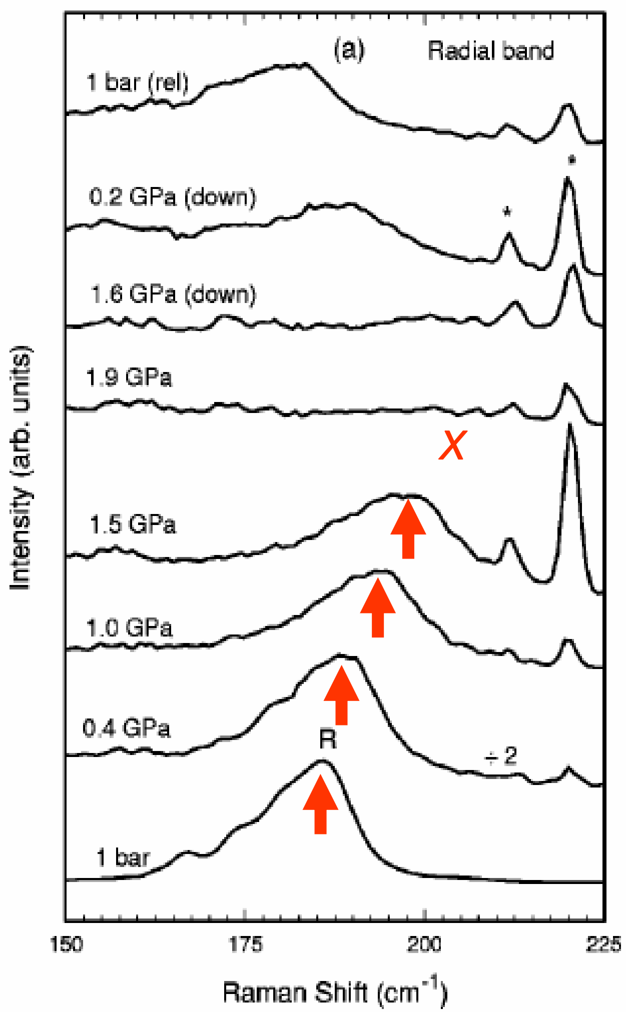
S. Zhang *et al*., PRB 73 (2006) 075423.

Results of MD simulations

Sun *et al*., PRB 70 (2004) 165417



1. Radial collapse of SWNTs



<u>Raman spectroscopy:</u>

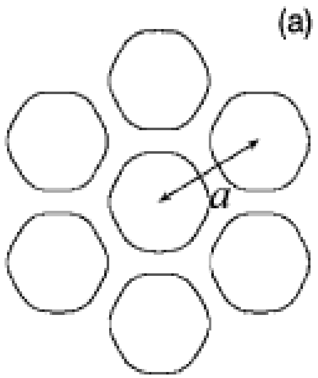
Vanishing a radial breezing mode

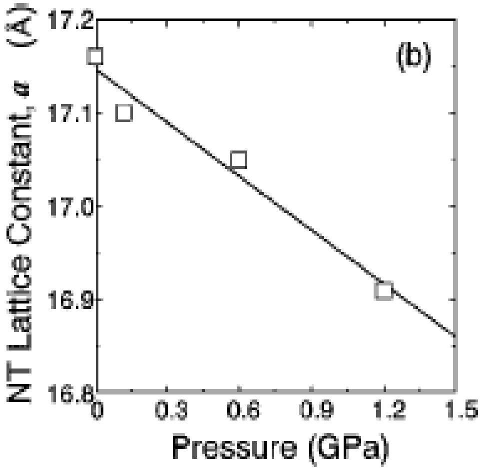
Venkateswaran *et al.*, PRB 59(1999) 10928

<u>X-ray diffraction:</u>

Polygonization of SWNT-bundle

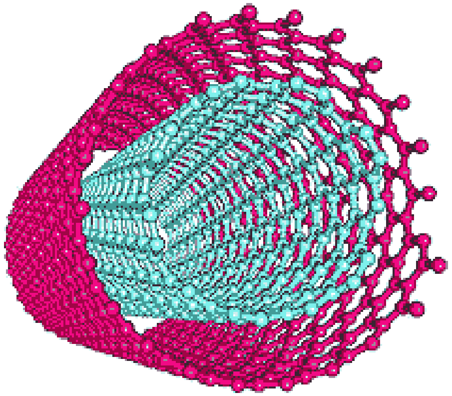
Tang *et al.*, PRL 85 (2000) 1887





2. Motivation

What takes place in Multi-walled nanotubes?



Structural features of multi-walled carbon nanotubes:

- Multiple concentric walls **interact with each other** through the intermolecular forces.
- External pressure leads to a **mechanical instability in outside walls** due to their large tube diameters.
- **Inner walls** are relatively stiff in the radial direction so that they can push back the surrounding outer walls.

Atomic-scale simulations for MWNTs = Very challenging!

⟶ Alternative approach: **Continuum elastic approximation**

3. Model and Method

Continuum elastic-shell model for MWNT

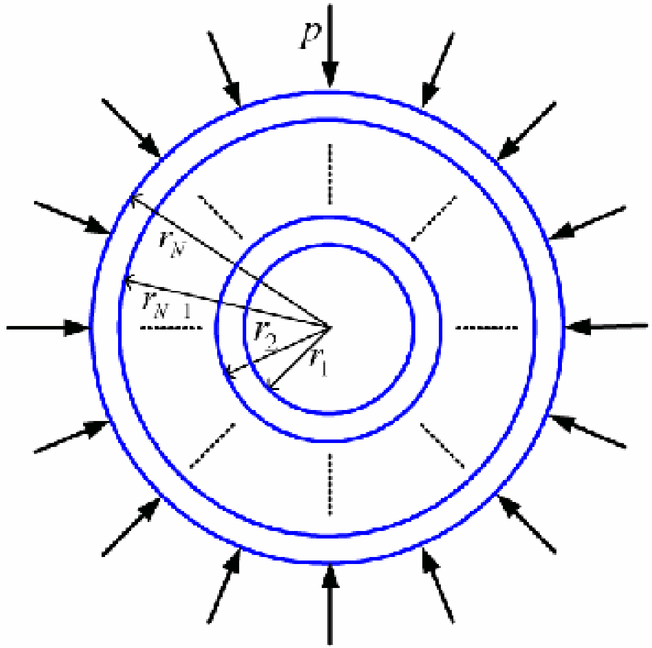
Ru, PRB 62 (2000) 16962

Sudak JAP 94 (2003) 7281

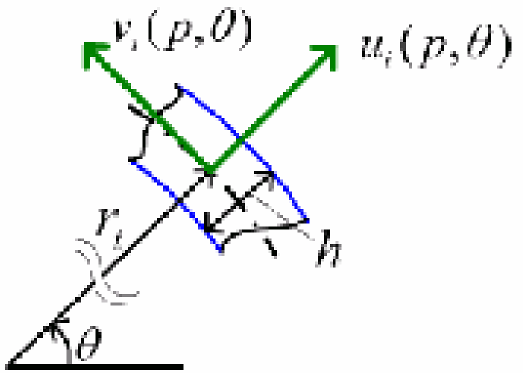
Leung PRB 71 (2005) 165415

He, J. Mech. Phys. Solids 53 (2005) 303

Wang JAP 99 (2006) 114317

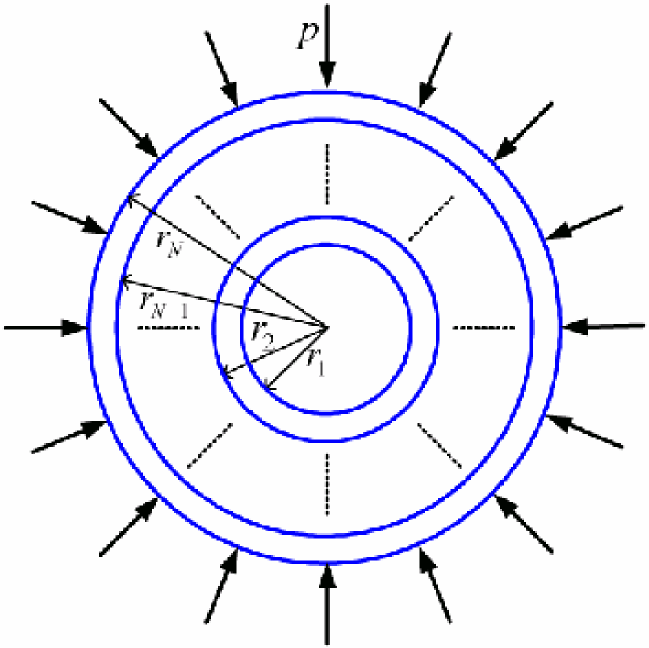


- **Displacements** of a surface element:
  - radial $u(\theta, p)$
  - circumferential $v(\theta, p)$



→ Obtaining the **mechanical energy** of the deformed MWNT, and then its stable cross-sectional shape under high hydrostatic pressure

3. Model and Method



The mechanical energy of MWNT:

$$U\left[p, u_i(p), v_i(p)\right] = U_D + U_I + \Omega$$

→ Apply a **variational method** to U in terms of u_i, v_i in order to determine the **stable cross-section** under high pressure p

Deformation energy: $$U_D = \sum_{i=1}^{N} \int_0^{2\pi} \left\{ \frac{Eh}{2(1-\nu^2) r_i} \left[u_i + v_i' + \frac{(u_i' - v_i)^2}{2 r_i} \right]^2 + \frac{Eh^3}{24(1-\nu^2)} \frac{(u_i'' - v_i')^2}{r_i^3} \right\} d\theta$$

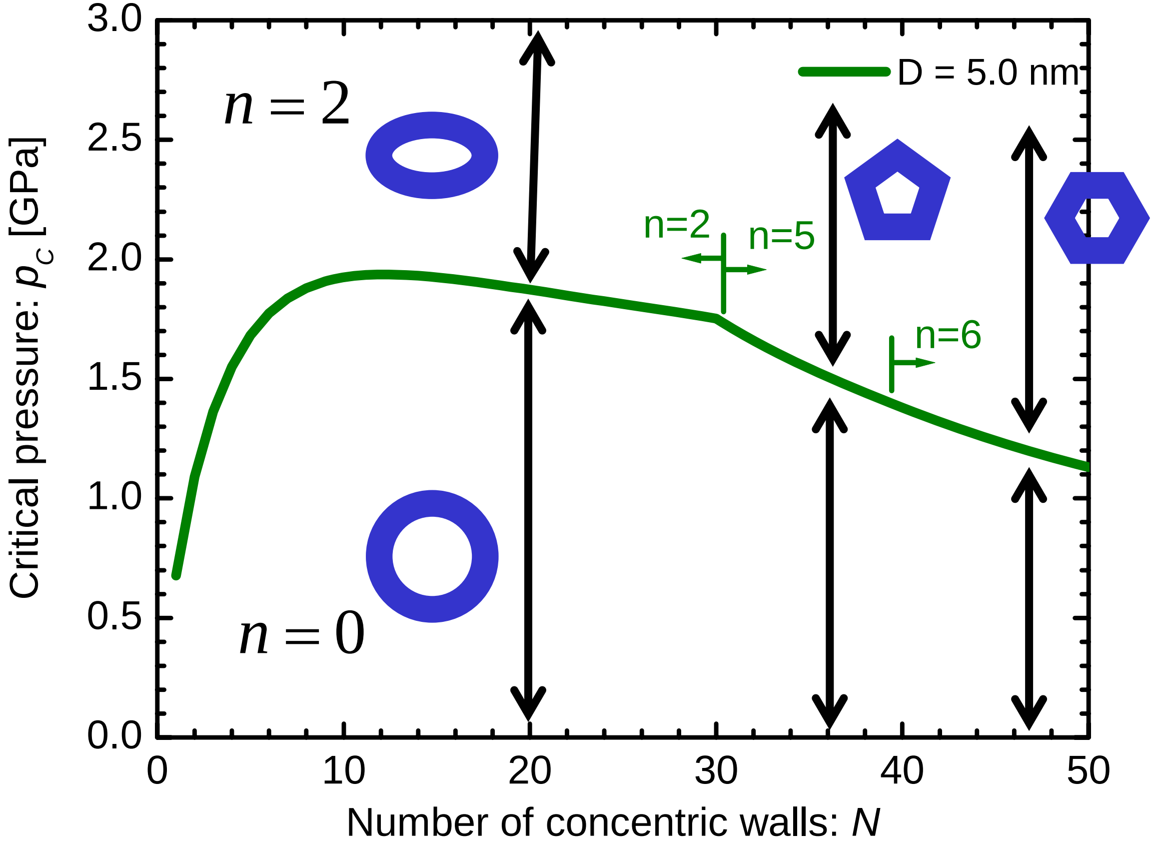
Inter-wall vdW energy $$U_I = \sum_{i=1}^{N-1} \frac{c_{i,i+1} r_i}{2} \int_0^{2\pi} (u_i - u_{i+1})^2 d\theta + \sum_{i=2}^{N} \frac{c_{i,i-1} r_i}{2} \int_0^{2\pi} (u_i - u_{i-1})^2 d\theta$$

Pressure-induced energy $$\Omega = p \int_0^{2\pi} \left(r_N u_N + \frac{u_N^2 + v_N^2 - u_N' v_N + u_N v_N'}{2} \right) d\theta$$

See H.Shima and M.Sato, *Nanotechnology in press*

4. Results

[1] Critical pressure curves



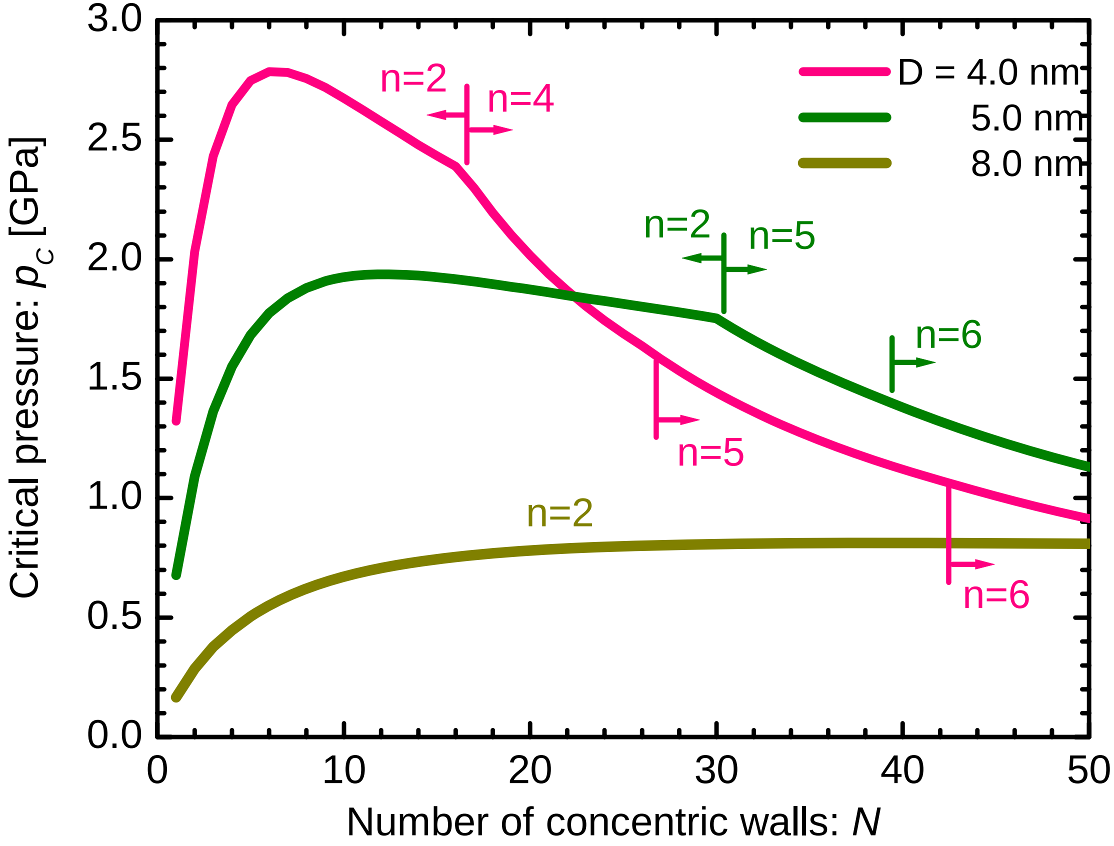
For given N and D, radial deformation occurs just above p_C

When N exceeds 30, radial corrugation is observed

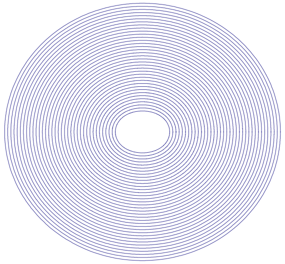
The wave number n of corrugation mode depends on N and D

4. Results

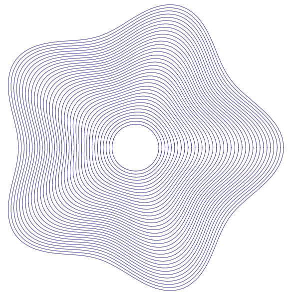
[1] Critical pressure curves



$n = 2$: Elliptic mode



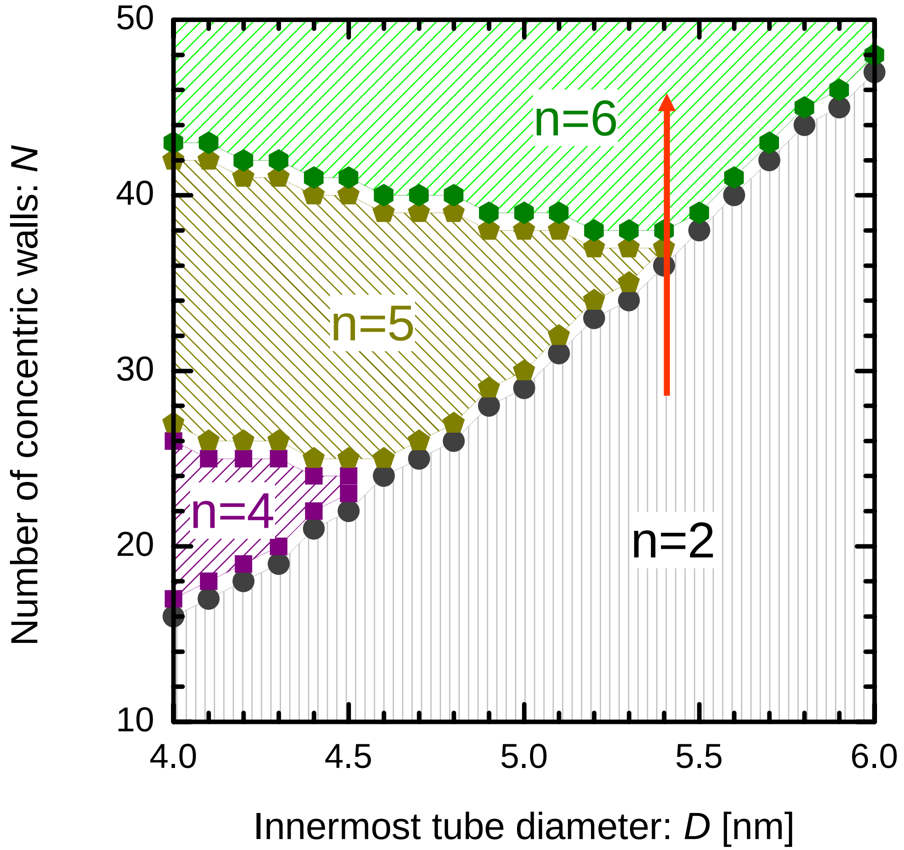
$n = 5$ **: Corrugation mode**



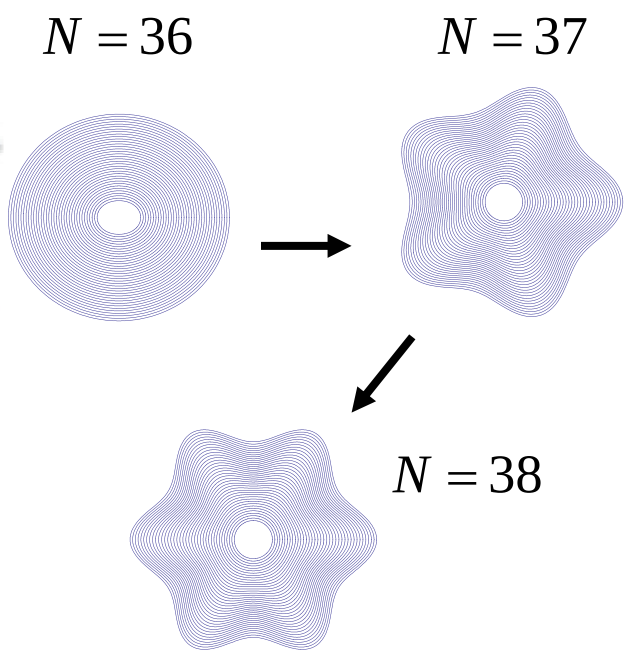
(= peculiar to MWNT with $N >> 1$ *)*

4. Results

[2] Phase diagram



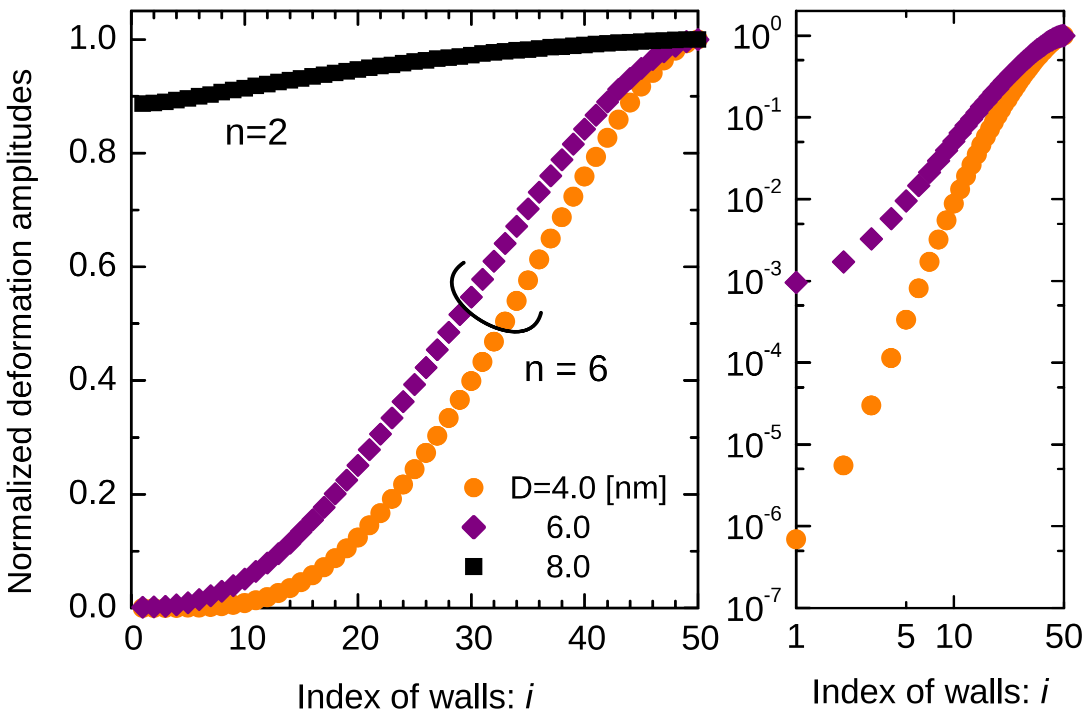
Increasing N with fixed $D = 5.4\,\mathrm{nm}$, then…



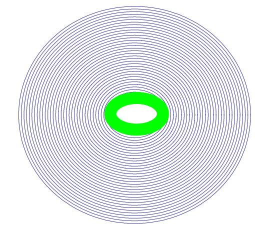
4. Results

[3] Geometric persistence of the innermost tube

In all corrugation modes, the circular shape of the innermost tube persists even under high pressure.

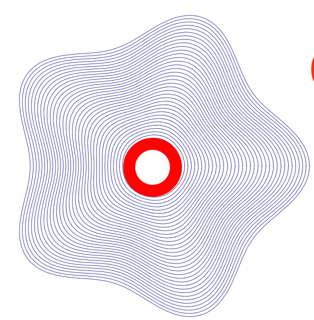


$n = 2$: Elliptic mode



Oval

$n = 5$ **: Corrugation mode**



Circular

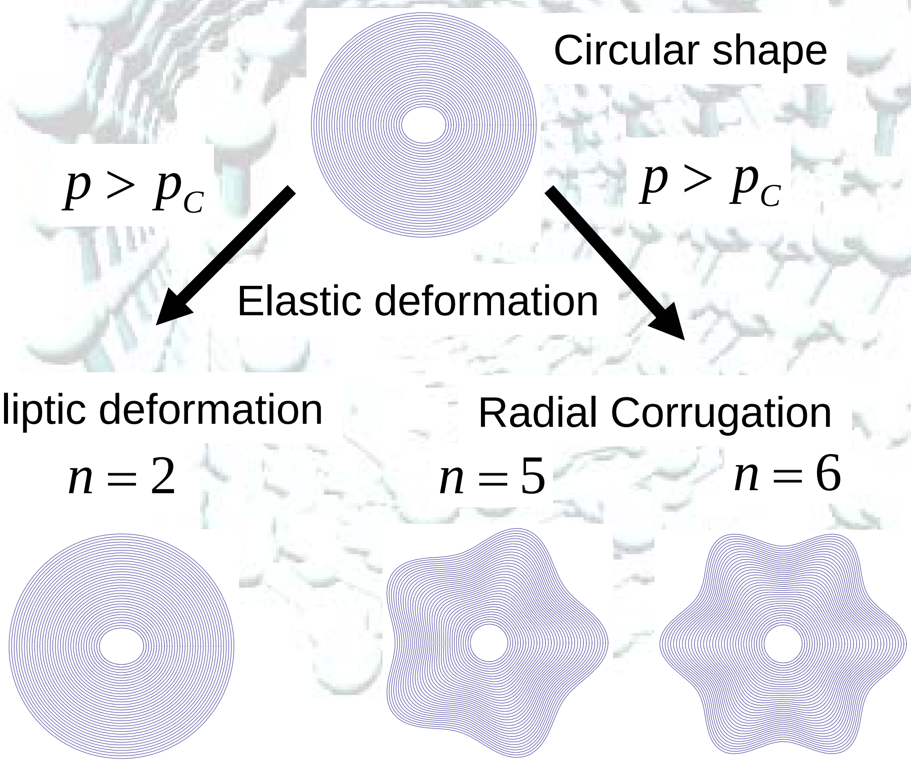
1-5 September 2008, Trend in NanoTechnology 2008 (TNT2008) in Oviedo, Spain

5. Summary

(*Contact to*: shima@eng.hokudai.ac.jp)

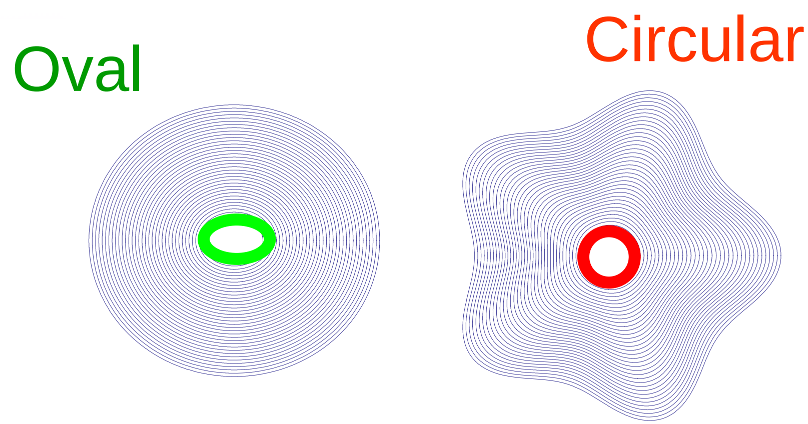
The main findings of this study are:

(1) Pressure-induced radial corrugation
in the cross-section of MWNT



(2) Mode index n depend on:
i) the tube diameter D and
ii) the number of concentric wall N

(3) Persistent cylindrical geometry
of the innermost tube of MWNT



5. Summary

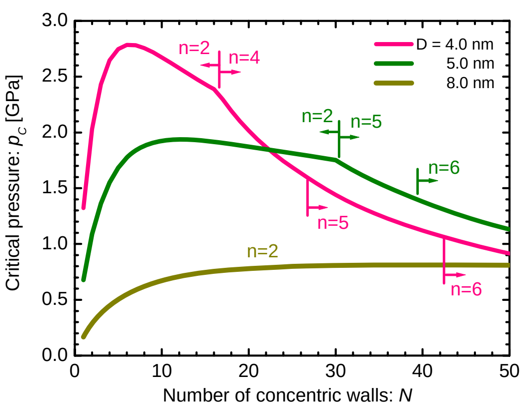
(*Contact to*: shima@eng.hokudai.ac.jp)

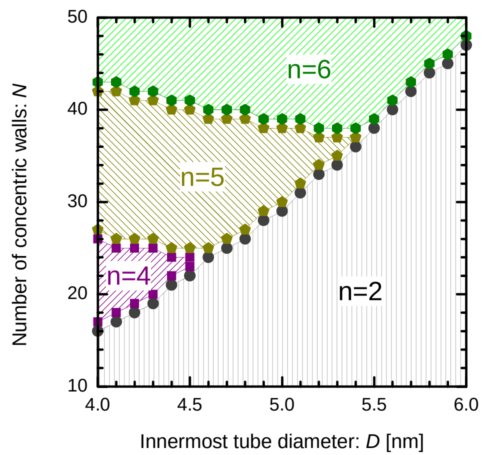
For multi-walled carbon nanotubes, we have demonstrated …

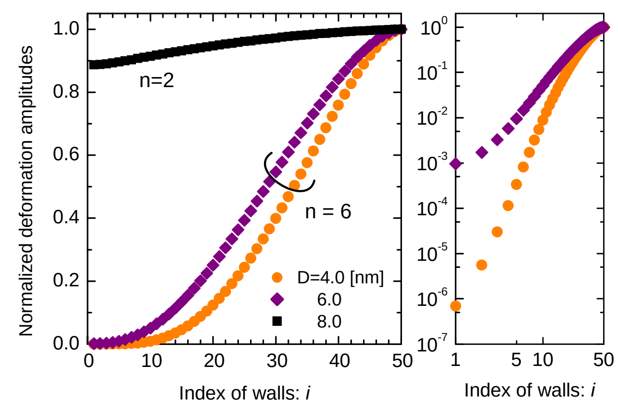
(1) Critical pressure curves

(2) Phase diagram of radial deformation

(3) Geometric persistence of the innermost tube







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