Department of Physics and Astronomy

UCL

Modelling the electrical conductivity of carbon nanotube films

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nanolayers

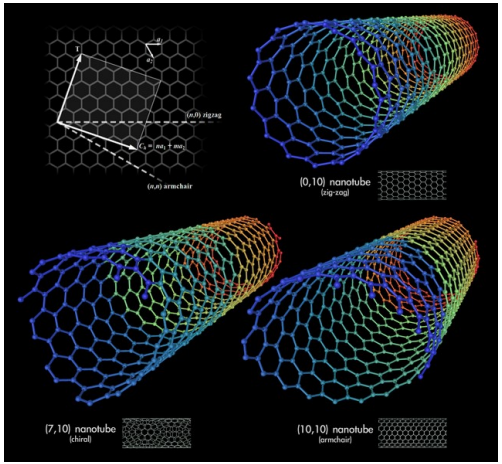
UKRI Engineering and Physical Sciences Research Council

THE THOMAS YOUNG CENTRE TyC

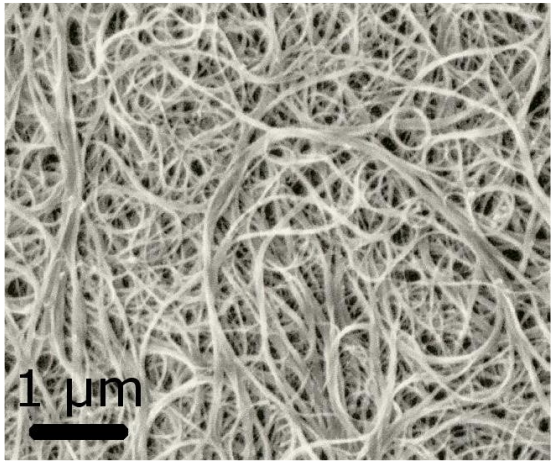
Carbon Nanotubes (CNTs)

Carbon nanotubes (CNTs) have many interesting properties

- Very high electrical conductivities (ballistic conductors)
- Many available nanotube structures (chiralities)
- Self organise into complex bundles and fabrics

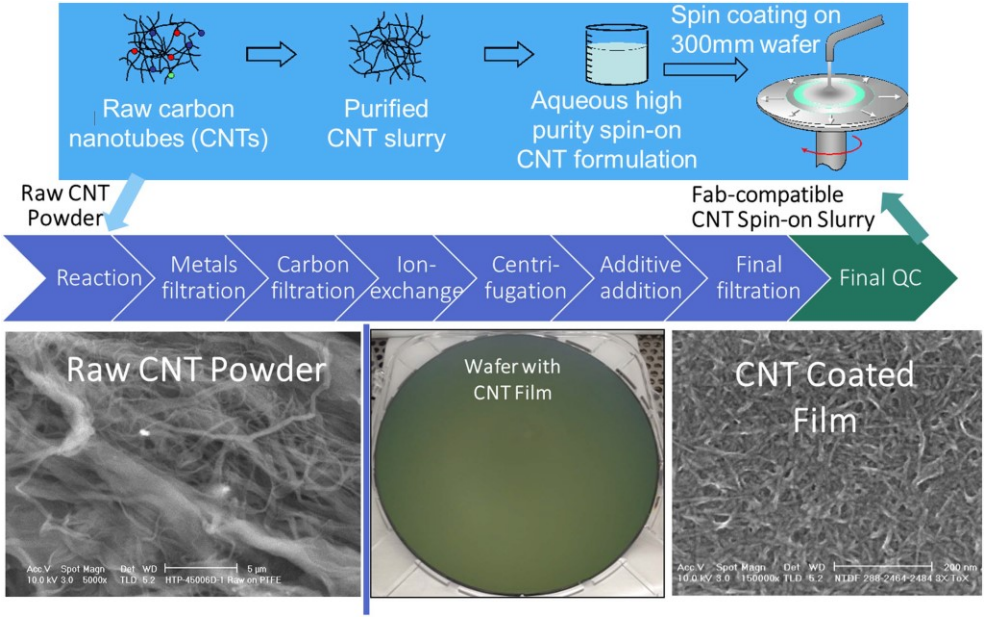


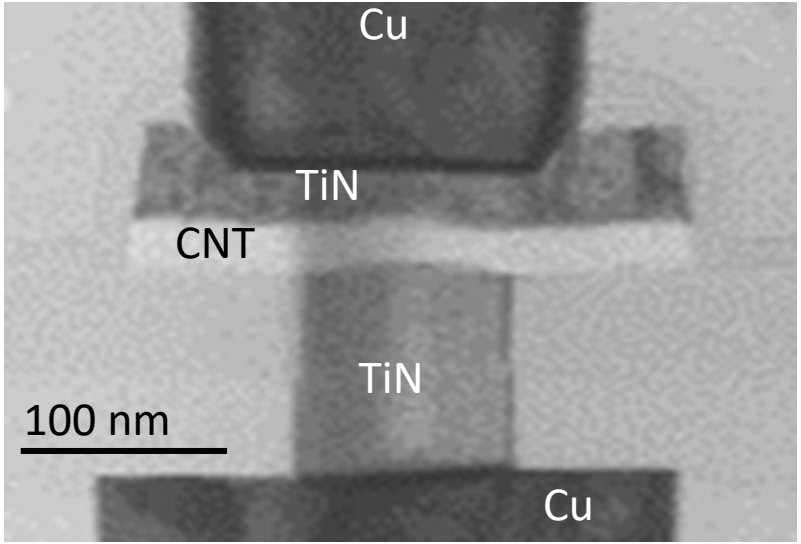
Example CNTs demonstrating the (n,m) notation



SEM image of a CNT fabric, demonstrating bundling and structural disorder

Motivation: NRAM cells

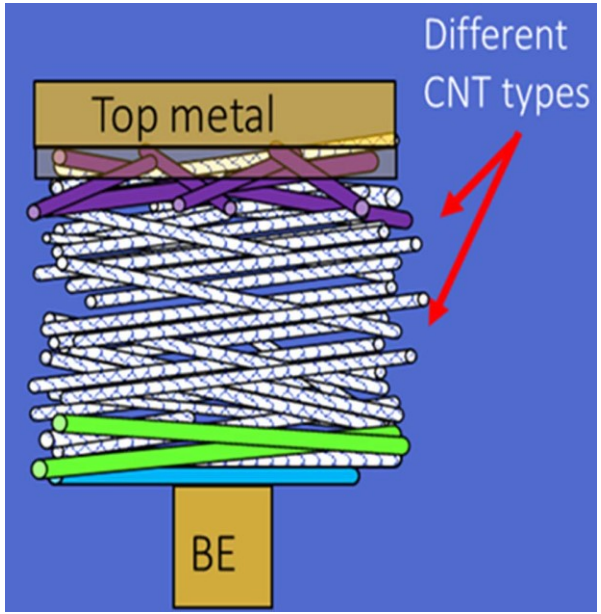


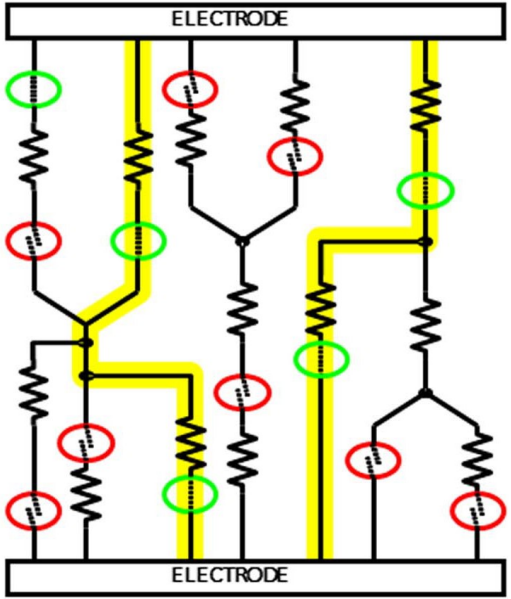


Gilmer, David C., T. Rueckes, and L. Cleveland. "NRAM: a disruptive carbon-nanotube resistance-change memory." Nanotechnology **29**.13 (2018): 134003.

Problem of scale

- The electrical properties of individual CNTs and CNT films are very different
- The primary source of resistance is electrons tunnelling from CNT to CNT





Gilmer, David C., T. Rueckes, and L. Cleveland. "NRAM: a disruptive carbon-nanotube resistance-change memory." Nanotechnology **29**.13 (2018): 134003.

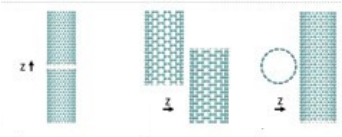
Levels of modeling

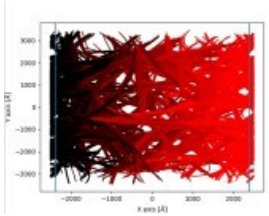
1) Mesoscopic structures of the device CNT fabric
2) Atomistic calculations of the conductivity of CNT junctions
3) Nodal current model of device simulation

The nodal model 3) uses the mesoscopic structures 2) and is parameterized from the atomistic calculations 1)

1) Mesoscopic structures

2) Atomistic calculations





3) Nodal model

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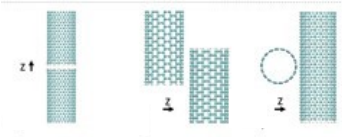
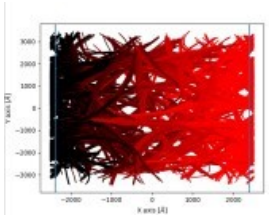
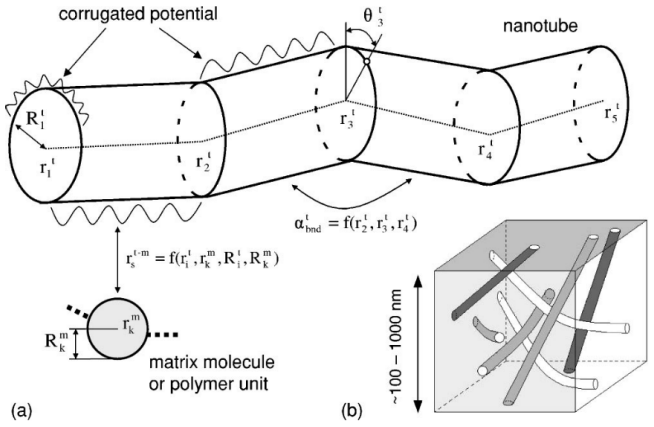
2) Atomistic calculations

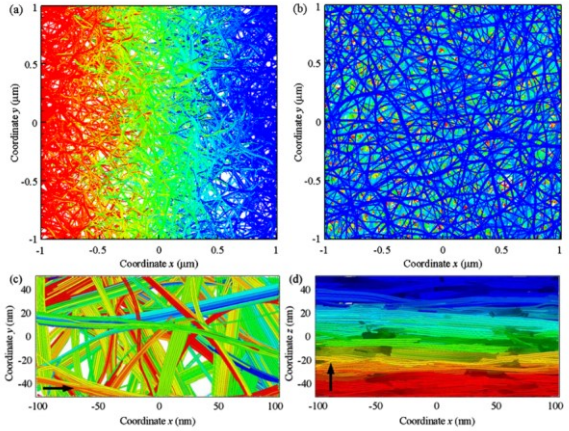


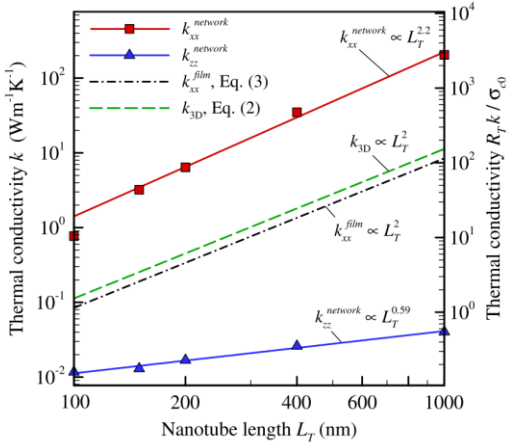


3) Nodal model

Mesoscopic modeling







Mesoscopic force field breaks down nanotube into segments. Parameterized to atomistic molecular dynamics model

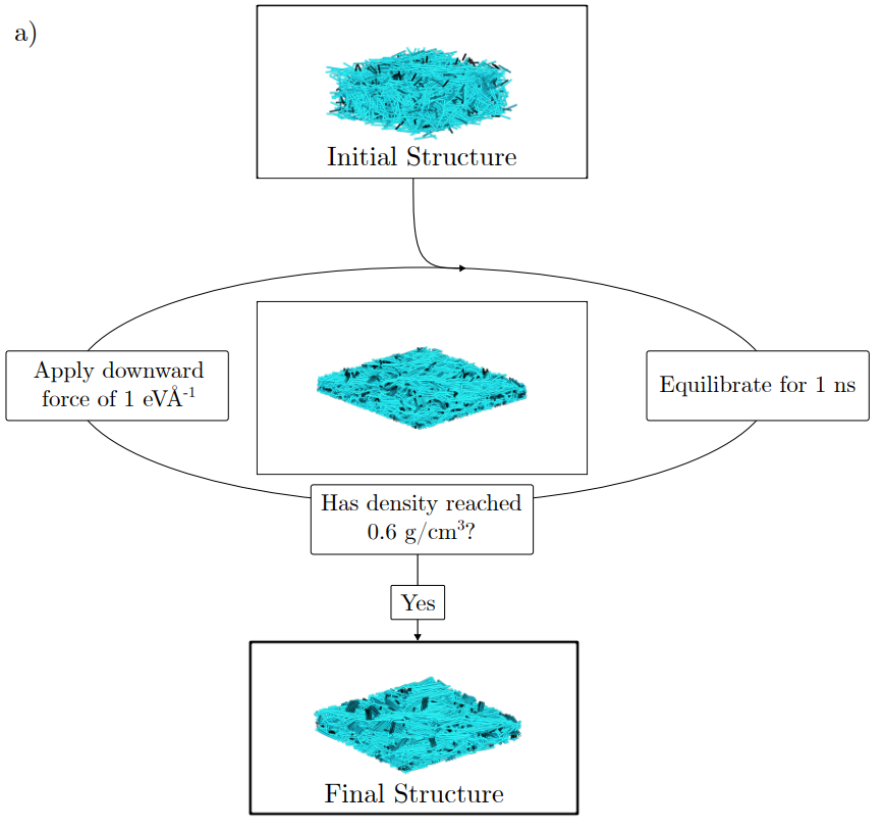
Can study dynamics of devices that contain many nanotubes of various lengths.

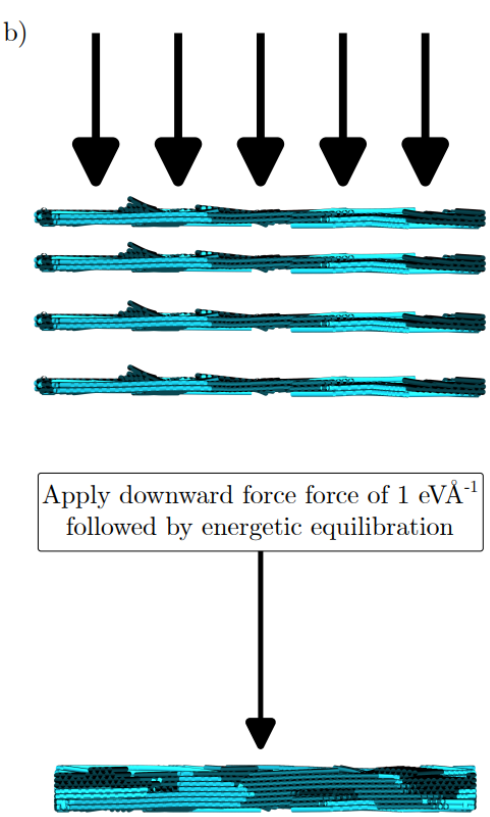
Provide input into key structural motifs and interactions within the system.

[1] A. N. Volkov and L. V. Zhigilei, J. Phys. Chem. C 114, 5513 (2010).
[2] L. V. Zhigilei, C. Wei, and D. Srivastava, Phys. Rev. B - Condens. Matter Mater. Phys. 71, 1 (2005).

Fabric model generation

- A library of CNT film structures was generated
- Length of the CNTs and the number of individual layers was varied
- High density structures were produced





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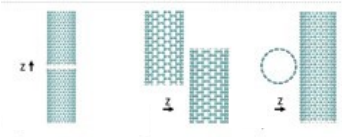
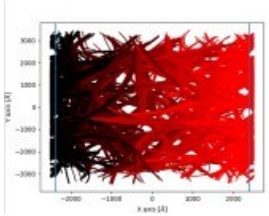
1) Mesoscopic structures

2) Atomistic calculations



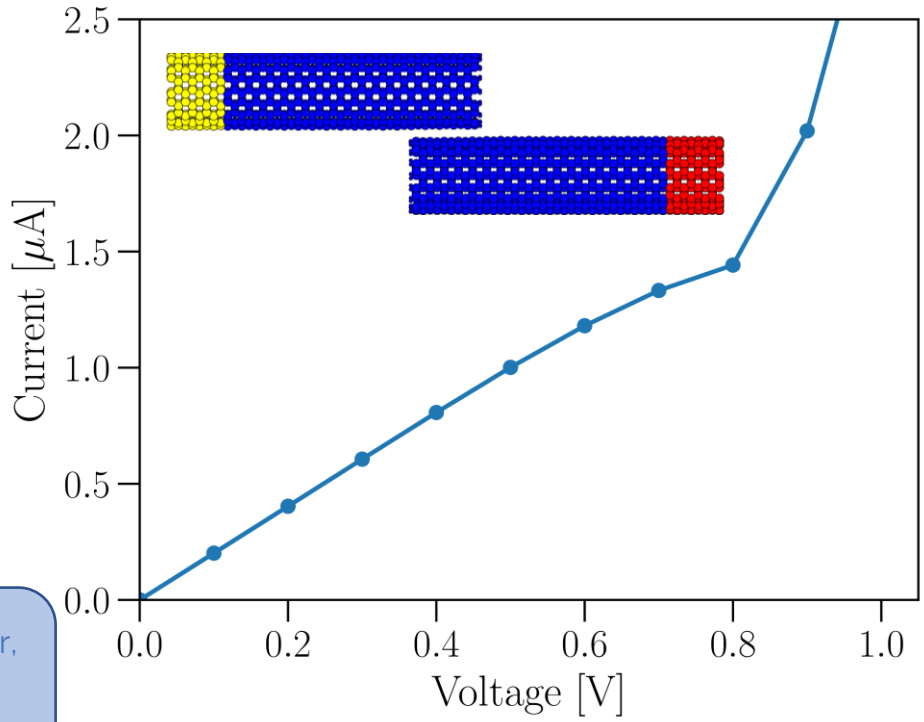


3) Nodal model

Atomistic modeling

We used density functional tight binding (DFTB) and the non-equilibrium Green's function (NEGF) method to look at junctions

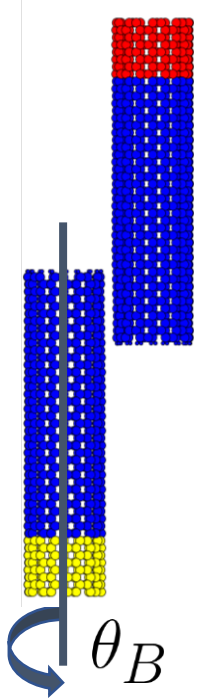
- 3OB Slater-Koster parameter set
- MBD dispersion correction
- Only metallic armchair CNTs considered (n,n)

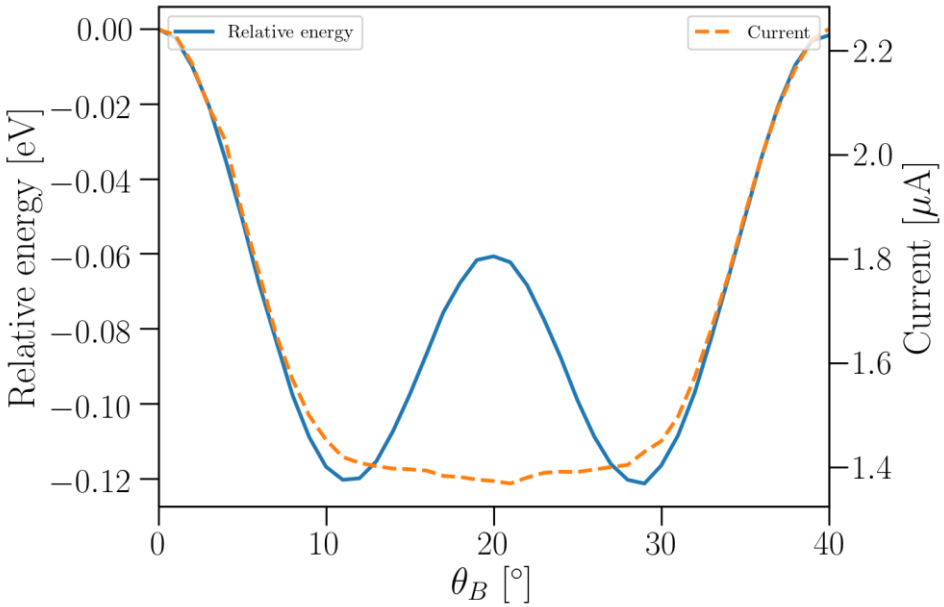


Durrant, T. R., El-Sayed, A. M., Gao, D. Z., Rueckes, T., Bersuker, G., & Shluger, A. L. Atomistic Modeling of the Electrical Conductivity of Single-Walled Carbon Nanotube Junctions. *Physica Status Solidi RRL (2022) 2200118*

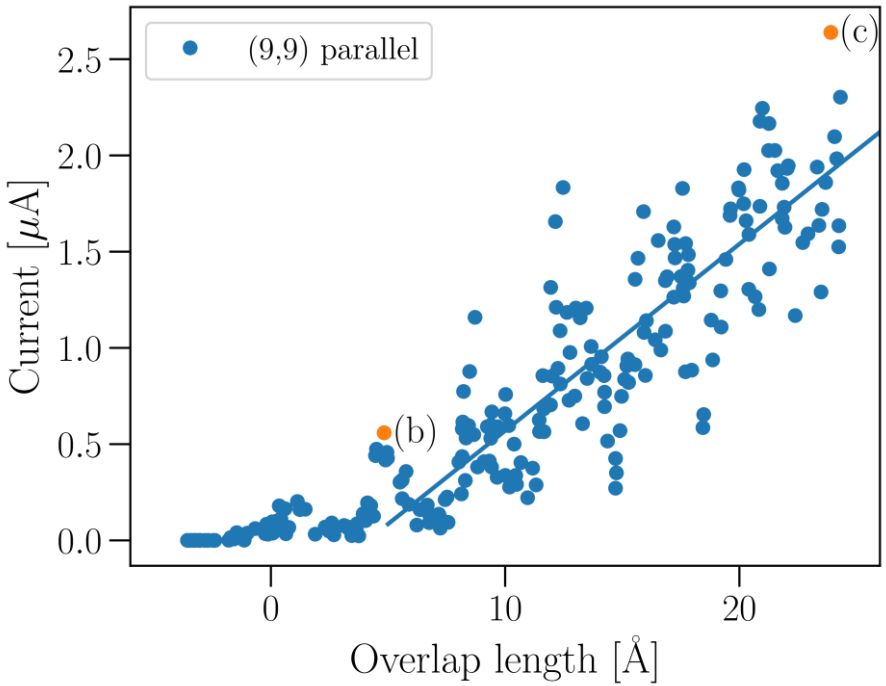
Rotational dependence

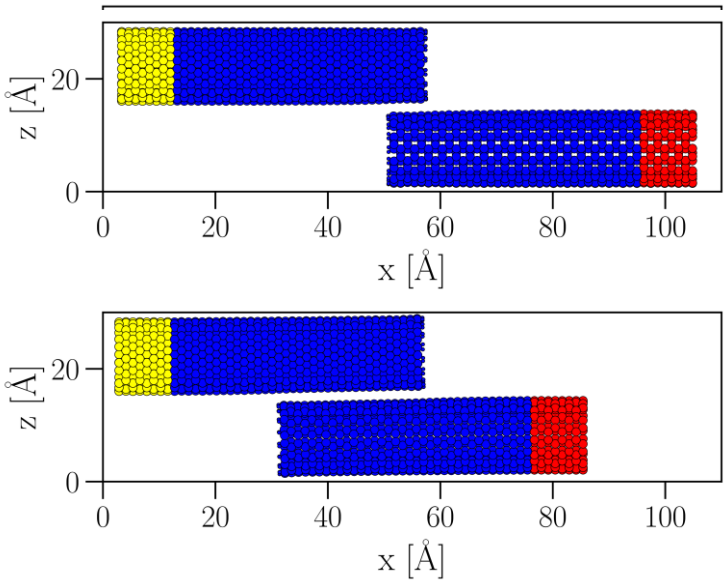
- The conductivity at a junction is strongly dependent on the local structure at the point of contact
- Strongest contributor to conductivity is the atomic registry at contact





Role of overlap

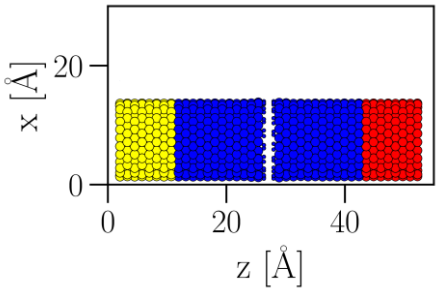




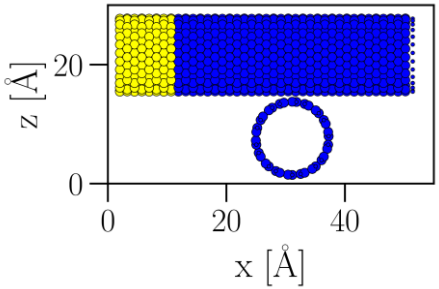
There is a linear relationship between overlap length and electric current

Generating junction structures

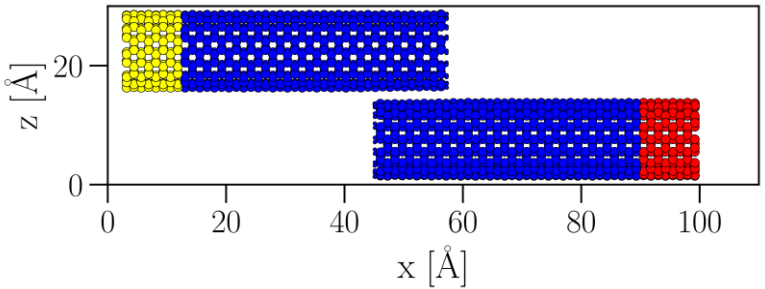
We applied random perturbations to three different template structures in order to produce a diverse range of junction structures (620 total)



a) Tip-to-tip



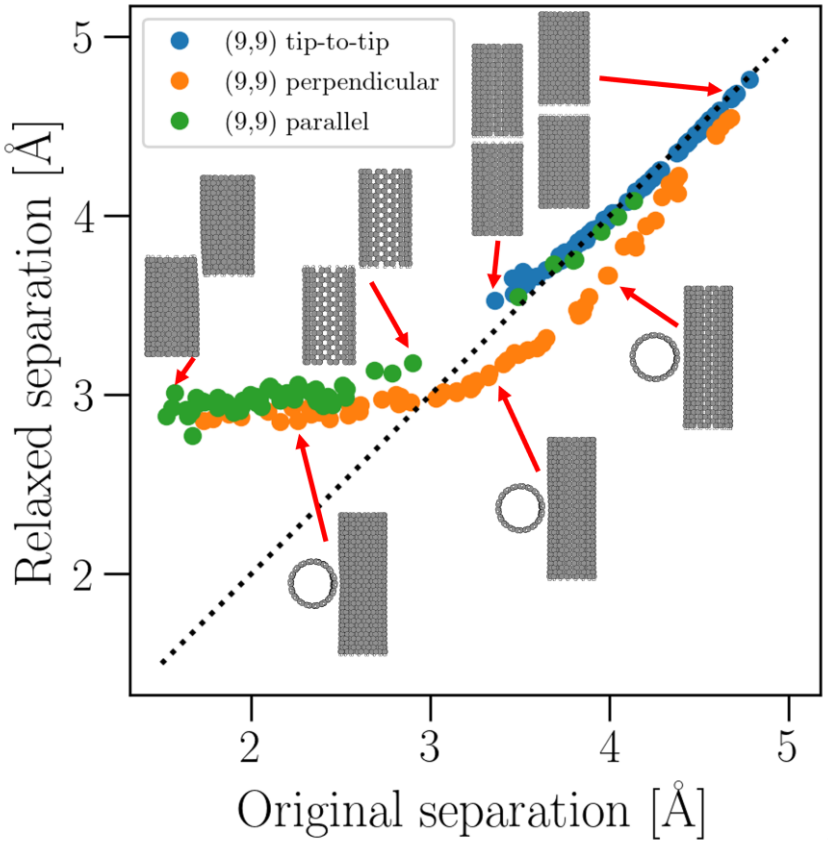
b) Perpendicular



c) Parallel

Junction structures

- (9,9) CNTs have an optimum van-Der Waals separation of 2.9 Å
- CNTs will deform rather than sit in closer contact
- Orientation of the CNTs controls how large the area of contact between the CNTs is



Electron tunneling

$$J = \frac{(2m\bar{\phi})^{\frac{1}{2}}}{\Delta r}\left(\frac{e}{h}\right)^2 V \exp\left(-\frac{4\pi\beta\Delta r}{h}(2m\bar{\phi})^{\frac{1}{2}}\right)$$

$$B = (2m\bar{\phi})^{\frac{1}{2}}\left(\frac{e}{h}\right)^2 S \quad \text{and} \quad C = \frac{4\pi\beta}{h}(2m\bar{\phi})^{\frac{1}{2}}$$

$$I = BV\frac{\exp(-Cr_{\min})}{r_{\min}}$$

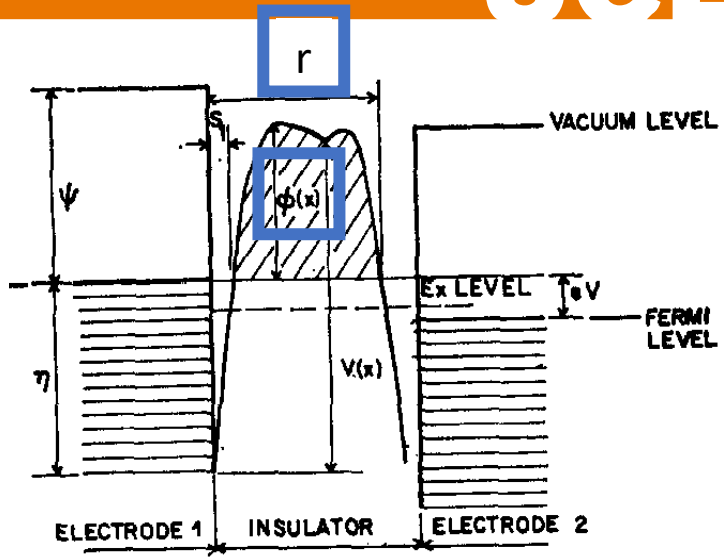


FIG. 1. General barrier in insulating film between two metal electrodes.

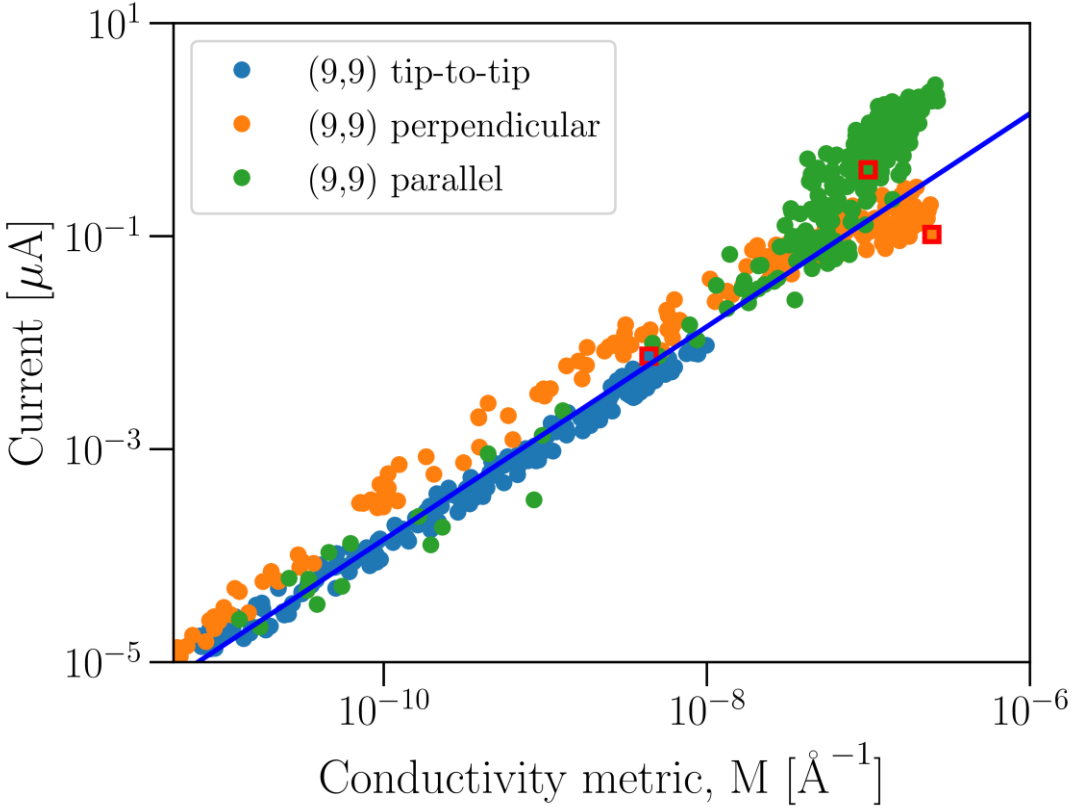
Simmons, John G. "Generalized formula for the electric tunnel effect between similar electrodes separated by a thin insulating film." *Journal of applied physics* **34**, no. 6 (1963): 1793-1803.

Sum of separations model

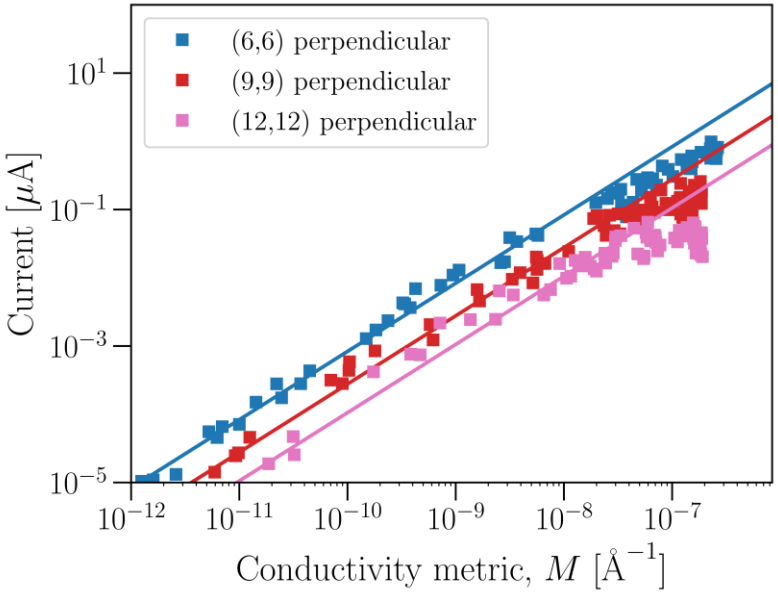
A simple model can be constructed by taking a sum of tunneling processes over all pairs of neighboring carbon atoms

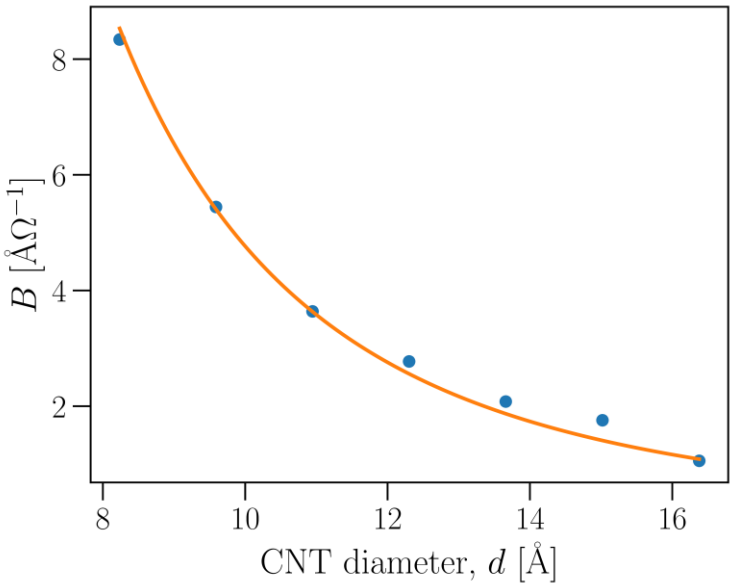
$$M = \sum_i \sum_j \frac{\exp\left(-C\, r_{ij}\right)}{r_{ij}}$$

$$I = MBV$$



Chirality dependence





Conductivity is chirality dependent and decreases with atomic radius

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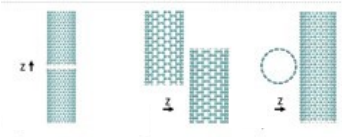
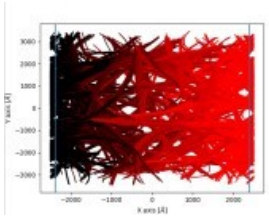


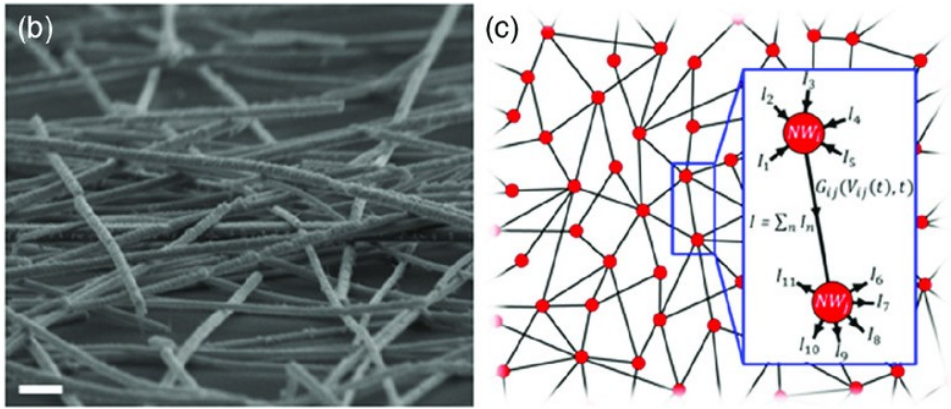


3) Nodal model

Nodal model

- The CNT fabric is abstracted as a network of nodes (in our case the beads of the mesoscopic model)

- The conductivity G_{ij} describes the electrical conductivity between node i and node j



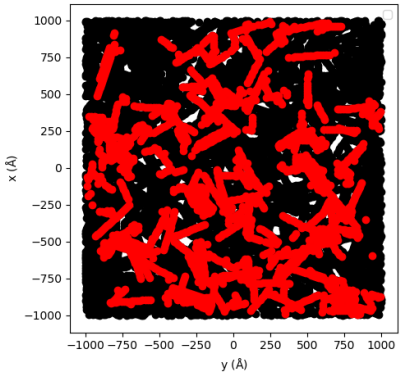
$$\mathbf{GV = S}\,, \quad \mathbf{I_{ij} = G_{ij}\,(V_j - V_i)}$$

Milano, G., Pedretti, G., Fretto, M., Boarino, L., Benfenati, F., Ielmini, D., ... & Ricciardi, C. (2020). Brain-Inspired Structural Plasticity through Reweighting and Rewiring in Multi-Terminal Self-Organizing Memristive Nanowire Networks. *Advanced Intelligent Systems*, *2*(8), 2000096.

Implementation details

Construct mesoscopic model (xyz) → Randomly allocate metallic CNTs → Build **G** and **S** matrix via model parameters → Solve simultaneous linear equations to yield **V** → Calculate current between every pair of nodes

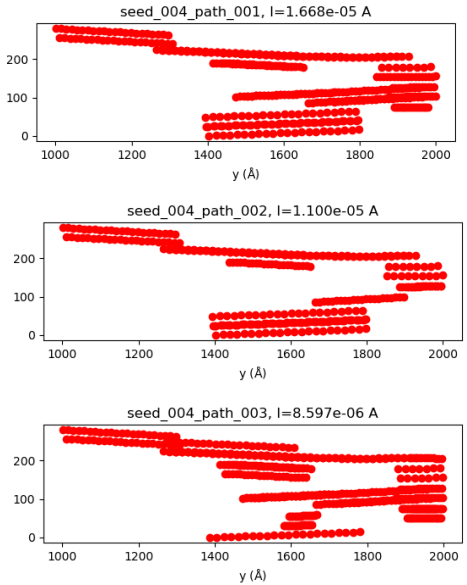
$$\begin{pmatrix} G_{11} & G_{12} & \cdots & G_{1N} \\ G_{21} & G_{22} & \cdots & G_{2N} \\ \vdots & \vdots & \ddots & \vdots \\ G_{N1} & G_{N2} & \cdots & G_{NN} \end{pmatrix} \begin{pmatrix} v_1 \\ v_2 \\ \vdots \\ v_N \end{pmatrix} = \begin{pmatrix} S_1 \\ S_2 \\ \vdots \\ S_N \end{pmatrix}$$



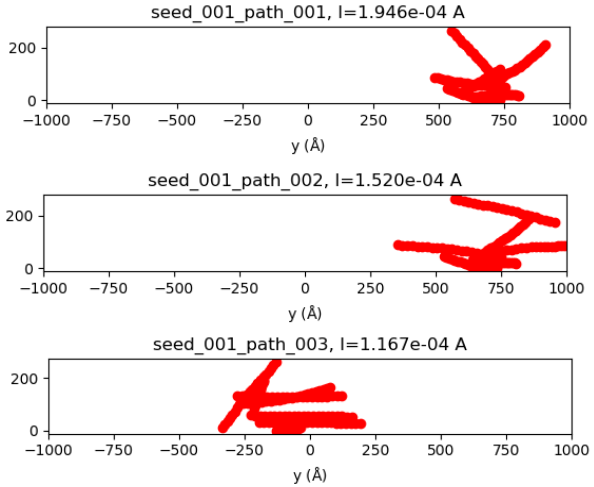
Pathway comparision

The distribution of conduction paths is quite different in comparison the layered and non-layered structures:

Layered examples



Nonlayered examples



Conclusions

- Large scale and dense CNT fabric models can be generated using mesoscopic forcefields
- Both the structure and electrical conductivity of CNT junctions are highly variable
- Simple conductivity models based only on the atomic structure can be constructed (although they lose accuracy for high overlap structures)
- These two components can be combined to model the total electrical conductivity of CNT films using nodal analysis

Thanks for listening

Acknowledgements

Prof Alex Shluger (UCL)
Dr David Gao (Nanolayers)
Dr Al-Moatasem El-Sayed (Nanolayers)
Dr Yvelin Giret (Nanolayers)
Dr Gennadi Bersuker (Aerospace)
Mr James Farmer (Aerospace)
Dr Dmitry Veksler (Aerospace)
Dr Thomas Rueckes (Nantero)
Mr Lee Cleveland (Nantero)
Dr Harry Luan (Nantero)
Dr Rahul Sen (Nantero)

Computational resources

Via our membership of the UK's HEC Materials Chemistry Consortium, which is funded by EPSRC (EP/R029431), this work used the ARCHER2 UK National Supercomputing Service (http://www.archer2.ac.uk) and the UK Materials and Molecular Modelling Hub for computational resources, MMM Hub, which is partially funded by EPSRC (EP/T022213)

Any questions?

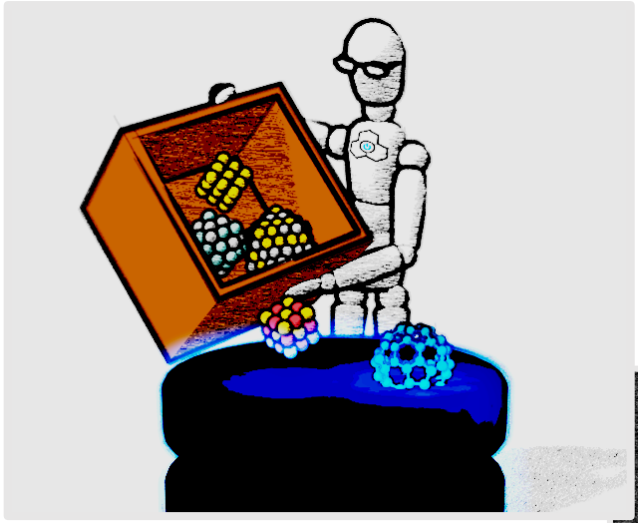
nanolayers

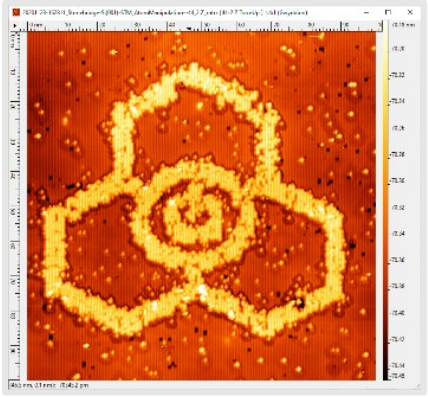
Materials Modelling, Machine Learning, and Research Digitalization

CritCat

PARTIAL-PGMs

MAST3RBoost





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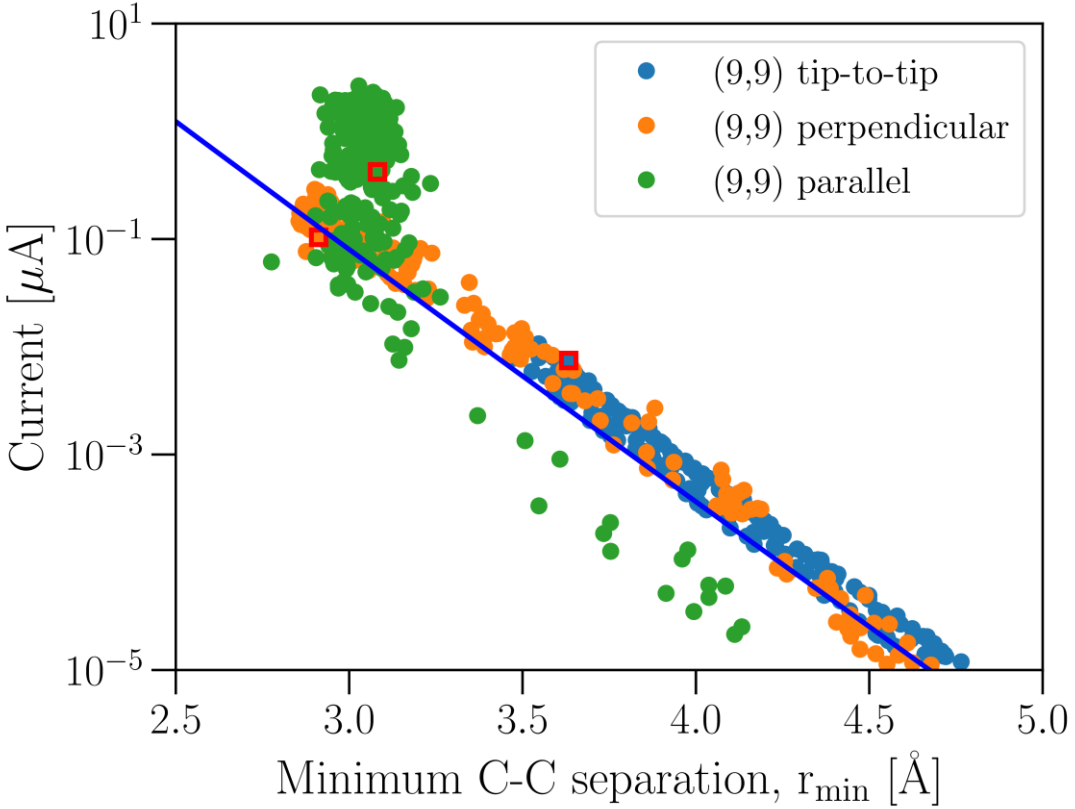
Separation model

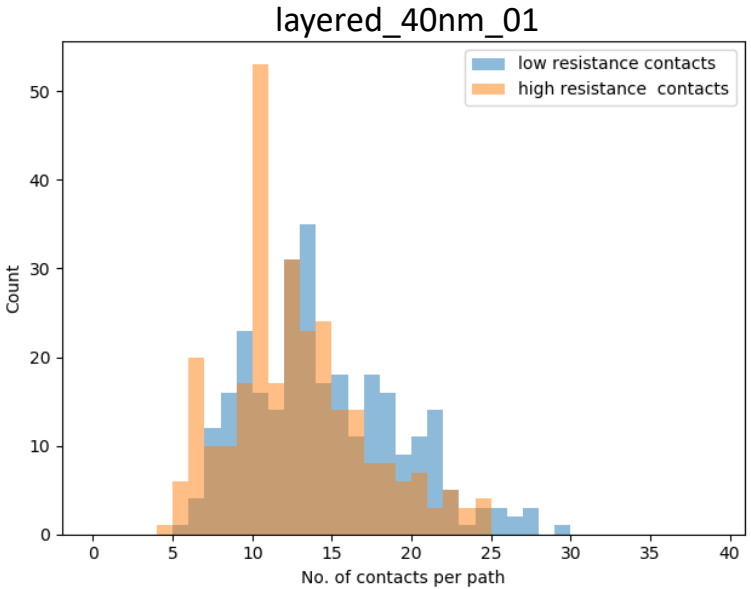
- Simplest model that can be fitted is to approximate the current from the smallest C-C separation, r_{min}

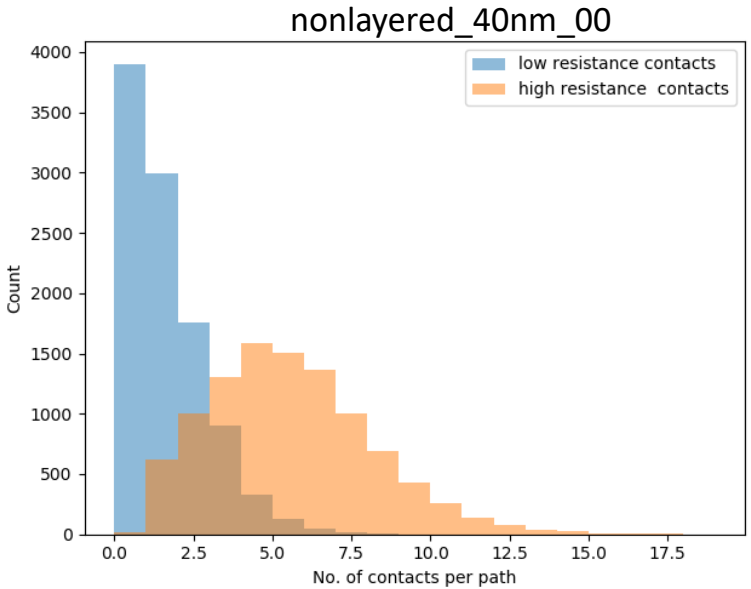
$$I = BV\frac{\exp\left(-C\,r_{\mathrm{min}}\right)}{r_{\mathrm{min}}}$$



Contact types

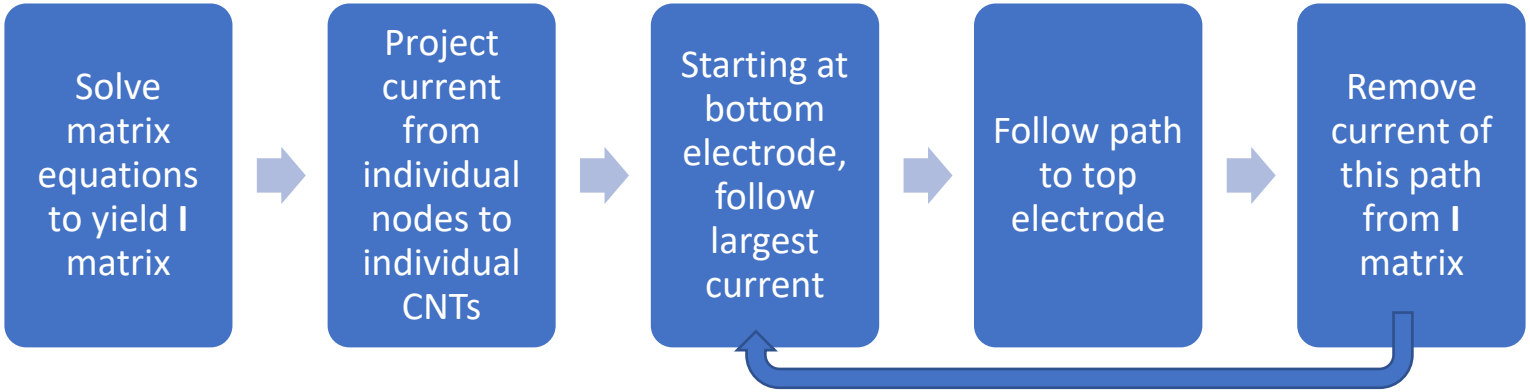
The distribution of contacts in the conduction paths is quite different in comparison between the layered and non-layered structures:





Post processing

Once the matrix equations have been solved, individual conductive paths through the CNT film can be extracted

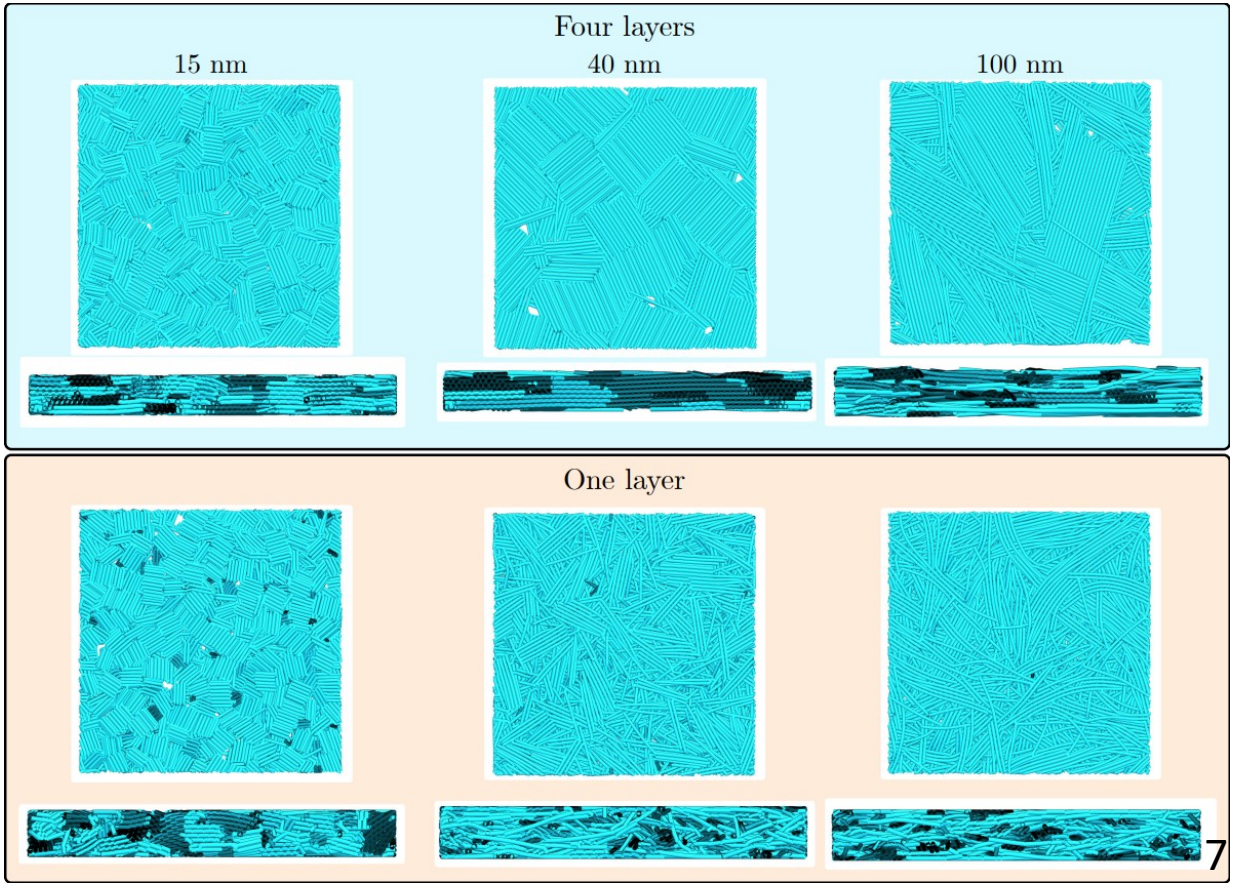


Path id	CNTs included	Total Current (μA)	Cumulative total (%)
1	1 12 5 9	10	33.33
2	7 10 9 4	5	50.00

Library of film structures

A library of CNT film structures was generated, where the length of the CNTs and the number of individual layers was varied

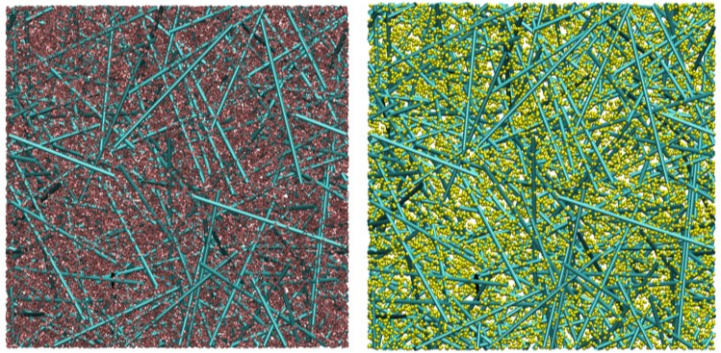
In this way, the links between mesoscopic structure and electrical conduction can be investigated



Extending the model

The developed framework can be extended with new components in order to explore potential switching mechanisms

- Introduction of amorphous carbon



- Trapping of charge in the CNT network