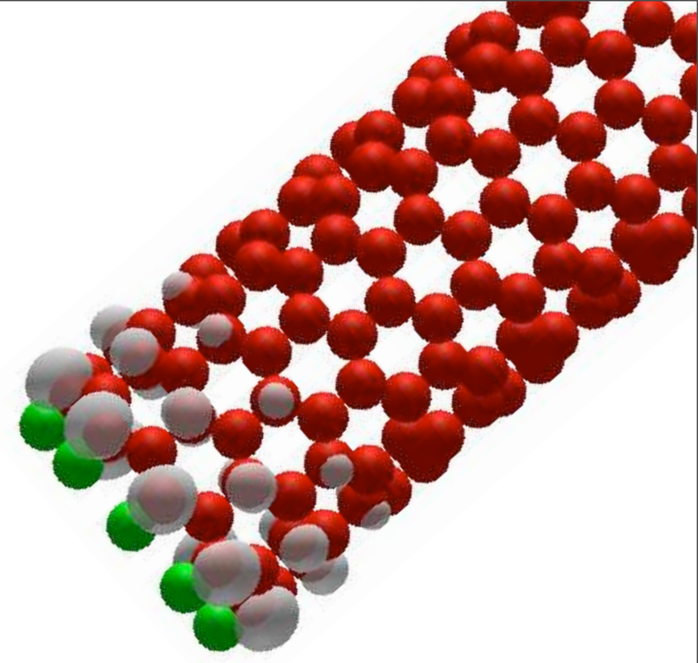
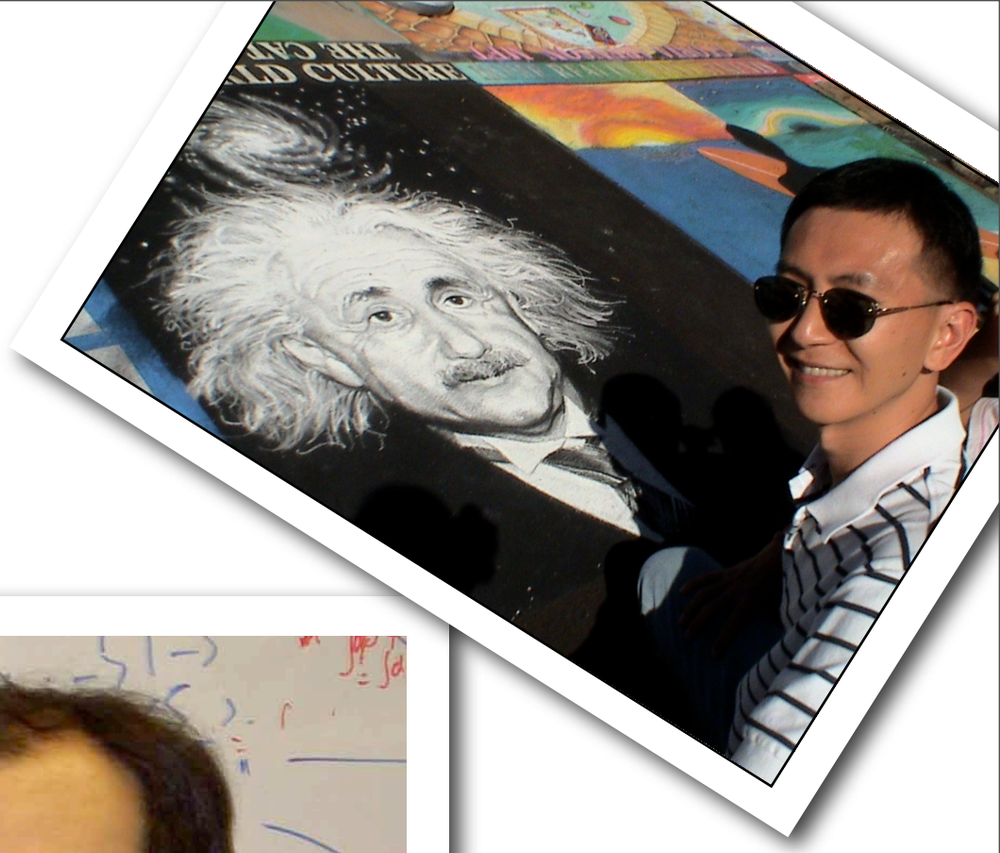


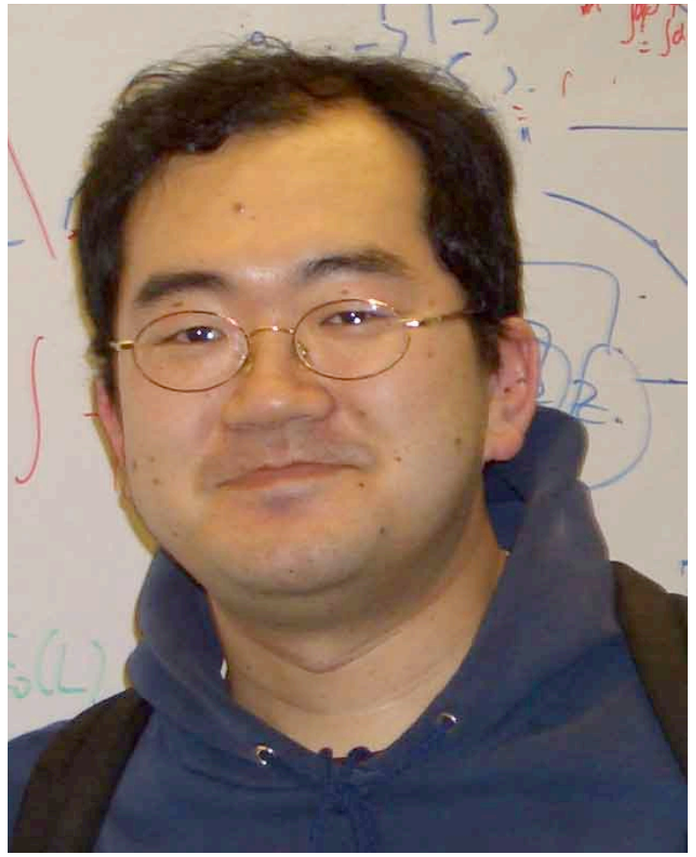
Quantized Nanomagnet

Hsiu-Hau Lin 林秀豪
National Tsing-Hua University
National Center for Theoretical Sciences

collaborators

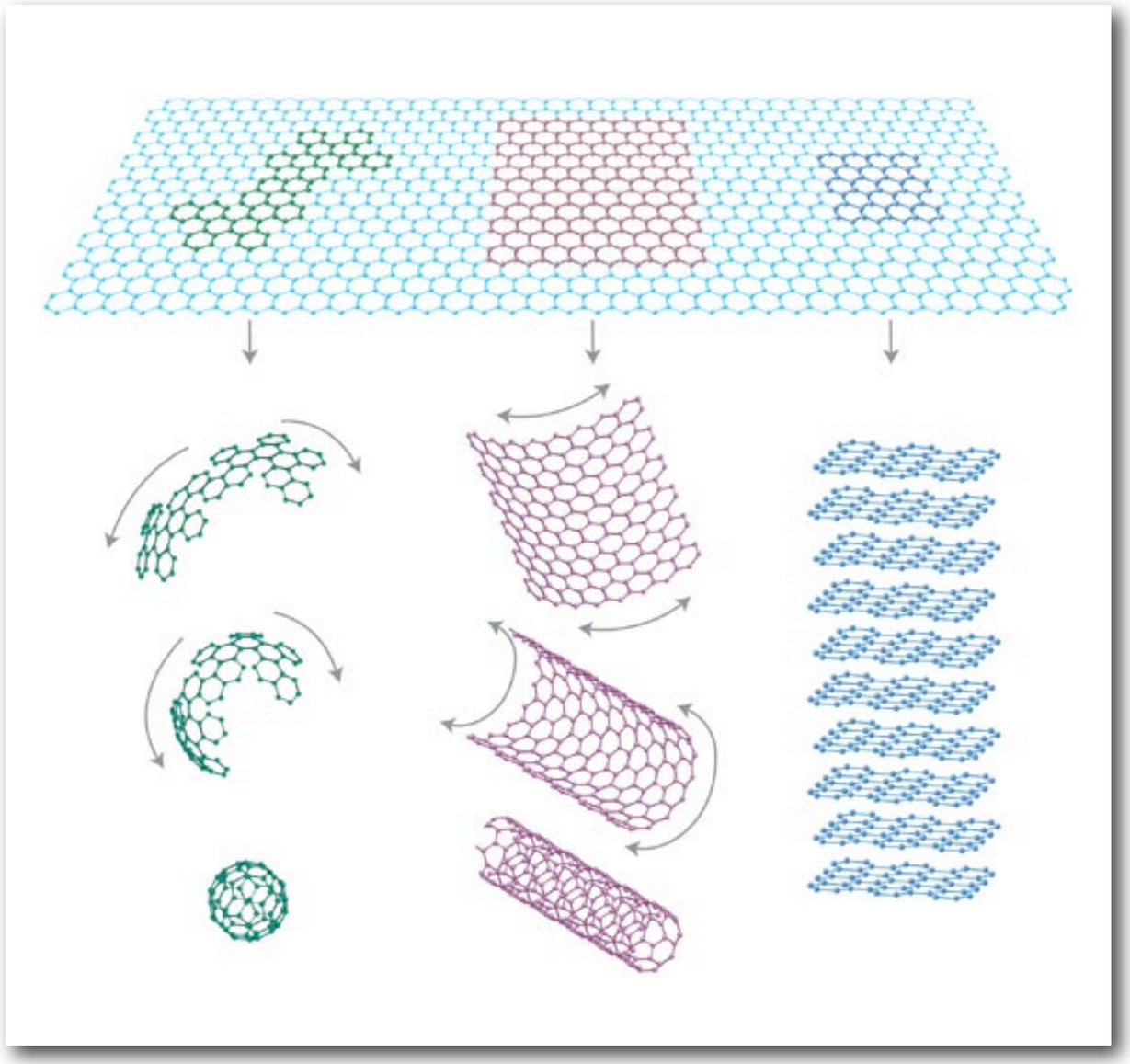


Horng-Tay Jeng
Academia Sinica



Masaki Oshikawa
Tokyo Univ

graphene family

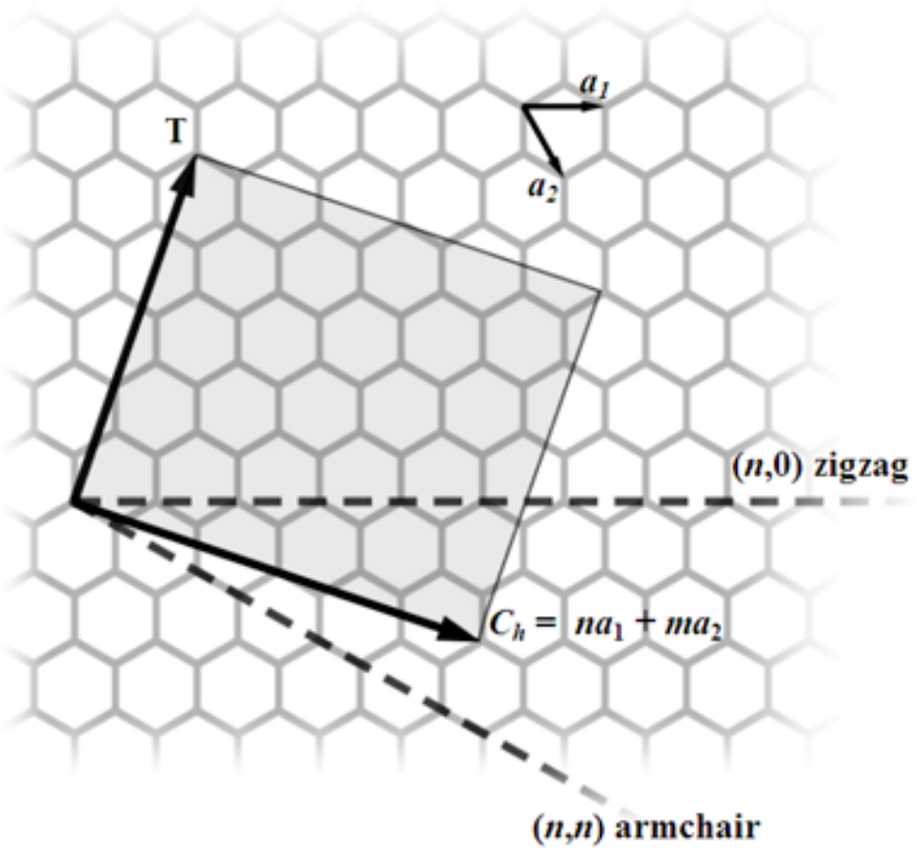


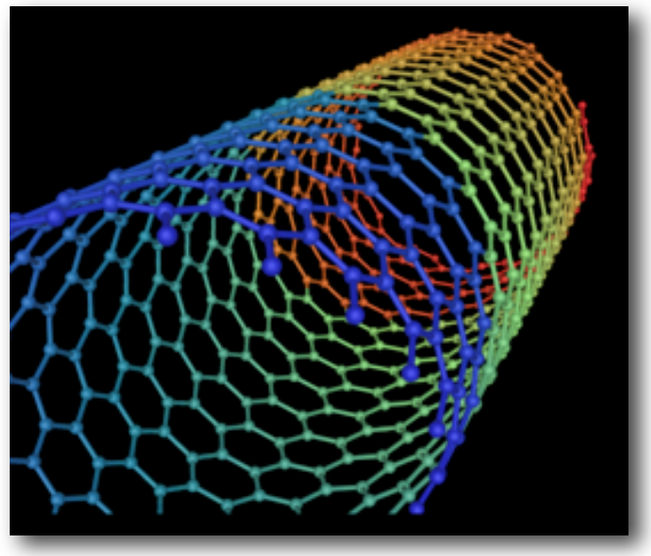
From the 2D graphene, "scissoring and wrapping" can give rise to 0D bucky balls, 1D nanotubes and 3D graphites.

Does the graphene family carry the blood of magnetism?

chiral vector

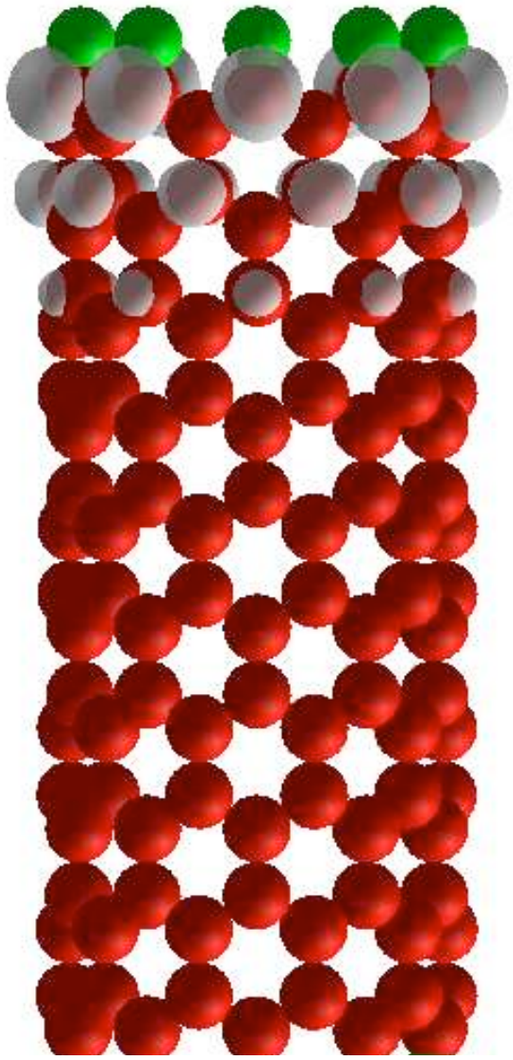
By identifying all lattice points related by integer multiples of the chiral vector $\boldsymbol{C} = (n_x, n_y)$ with $n_x \geq n_y \geq 0$, the two-dimensional graphene sheet is mapped to the chiral single-wall carbon nanotube.





Just roll it up!

topological nanomagnet



We found the ground state of the semi-infinite carbon nanotube has **quantized magnetic moment**,

$$M = \mu_B \left[\frac{n_x - n_y + 1}{3} \right],$$

which is dictated by the **chiral vector** but does not depends on the hopping amplitude and the interaction strength!

outline

- Motivations and hints
- Stage I: Lieb's theorem
- Stage II: weak-coupling analysis
- Stage III: counting edge states
- First-principles approach
- Conclusions

Hints and Motivations

VOLUME 84, NUMBER 8 PHYSICAL REVIEW LETTERS 21 FEBRUARY 2000

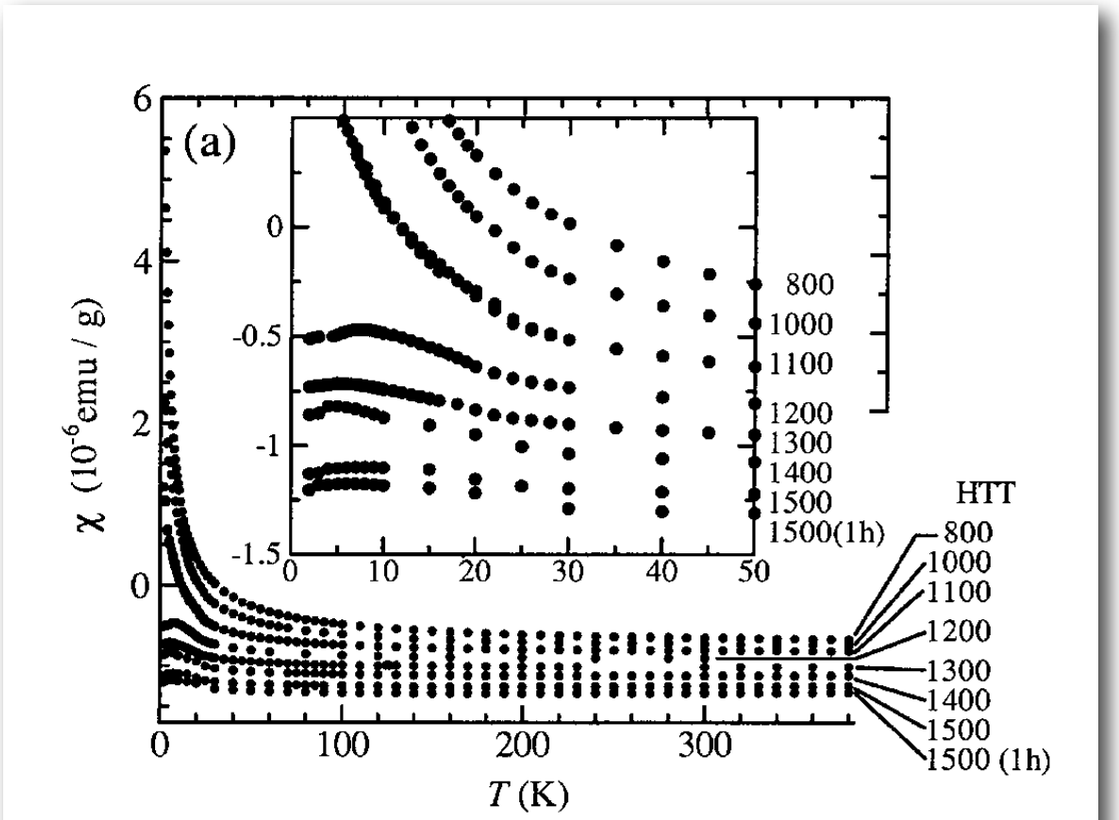
Disordered Magnetism at the Metal-Insulator Threshold in Nano-Graphite-Based Carbon Materials

Yoshiyuki Shibayama, Hirohiko Sato, and Toshiaki Enoki
Department of Chemistry, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo 152–8551, Japan

Morinobu Endo
Department of Electrical Engineering, Shinshu University, 500 Wakasato, Nagano-shi, Nagano 380–8553, Japan
(Received 26 April 1999)

Grinding up the graphite fibers gives rise to **magnetic moment** and thus Curie-like spin susceptibility.

With **heat treatment**, the broken edges glue back. Due to the suppression of open edges, the spin susceptibility becomes diamagnetic again.

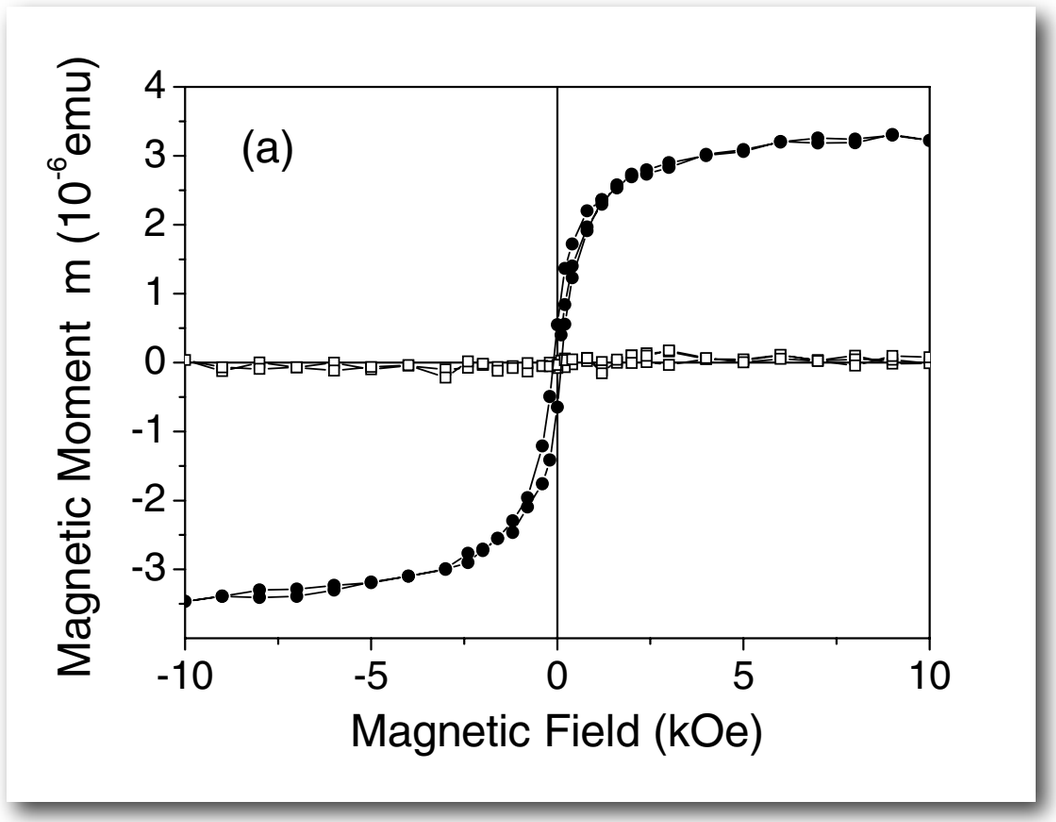


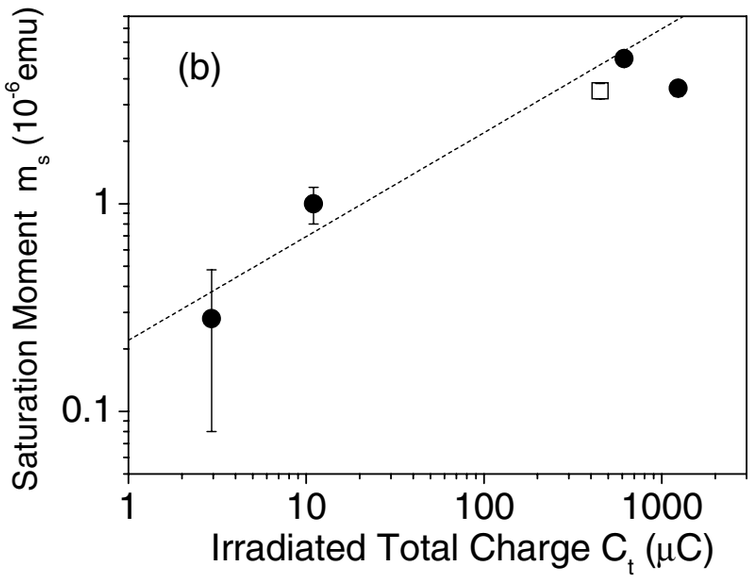
Shibayama et al.
Phys. Rev. Lett. **84**, 1744 (2000)

Induced Magnetic Ordering by Proton Irradiation in Graphite

P. Esquinazi,* D. Spemann, R. Höhne, A. Setzer, K.-H. Han, and T. Butz
Institut für Experimentelle Physik II, Universität Leipzig, Linnéstrasse 5, D-04103 Leipzig, Germany
(Received 1 July 2003; published 24 November 2003)

Magnetic moments show up in highly oriented polycrystalline graphite (HOPG) **after proton bombardment**. The magnitude of the moment is roughly proportional to the irradiation time.



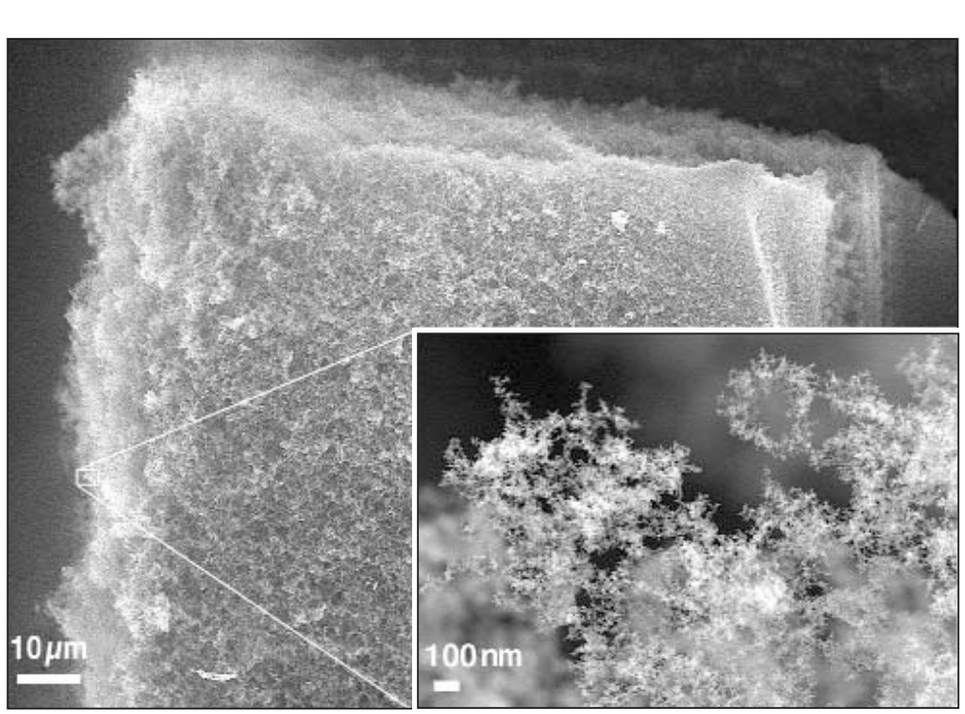


Esquinazi's group
Phys. Rev. Lett. **91**, 227101 (2003)

family blood?

Carbon Foam Reveals a Fleeting Magnetic Personality

2 APRIL 2004 VOL 304 SCIENCE



Attractive. One of the lightest substances ever made, nanoscale froth condensed from superheated carbon atoms is also magnetic at room temperature for a few hours.

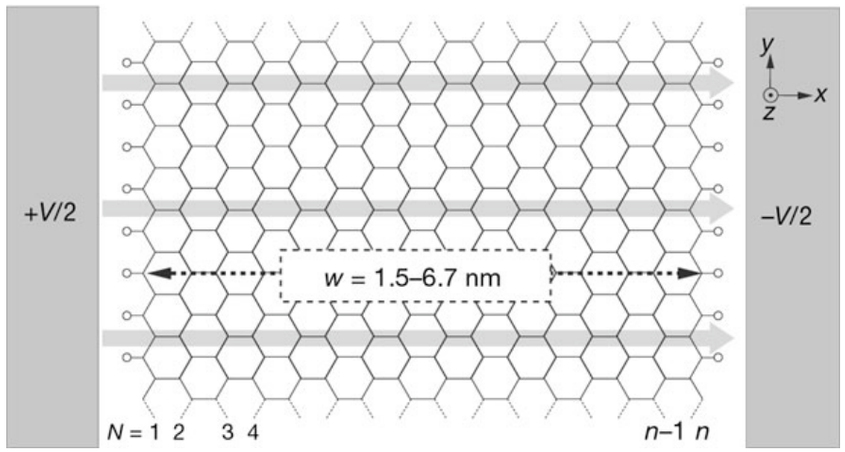
But Giapintzakis says extensive tests on the nanofoam show that **impurities** could account for at most 20% of the magnetism present. "We are sure we do not have an impurity effect," he says.

Bolstering the case, Giapintzakis adds, the team has found that other normally nonmagnetic materials such as **boron nitride** show similar properties when subjected to the same laser treatment.

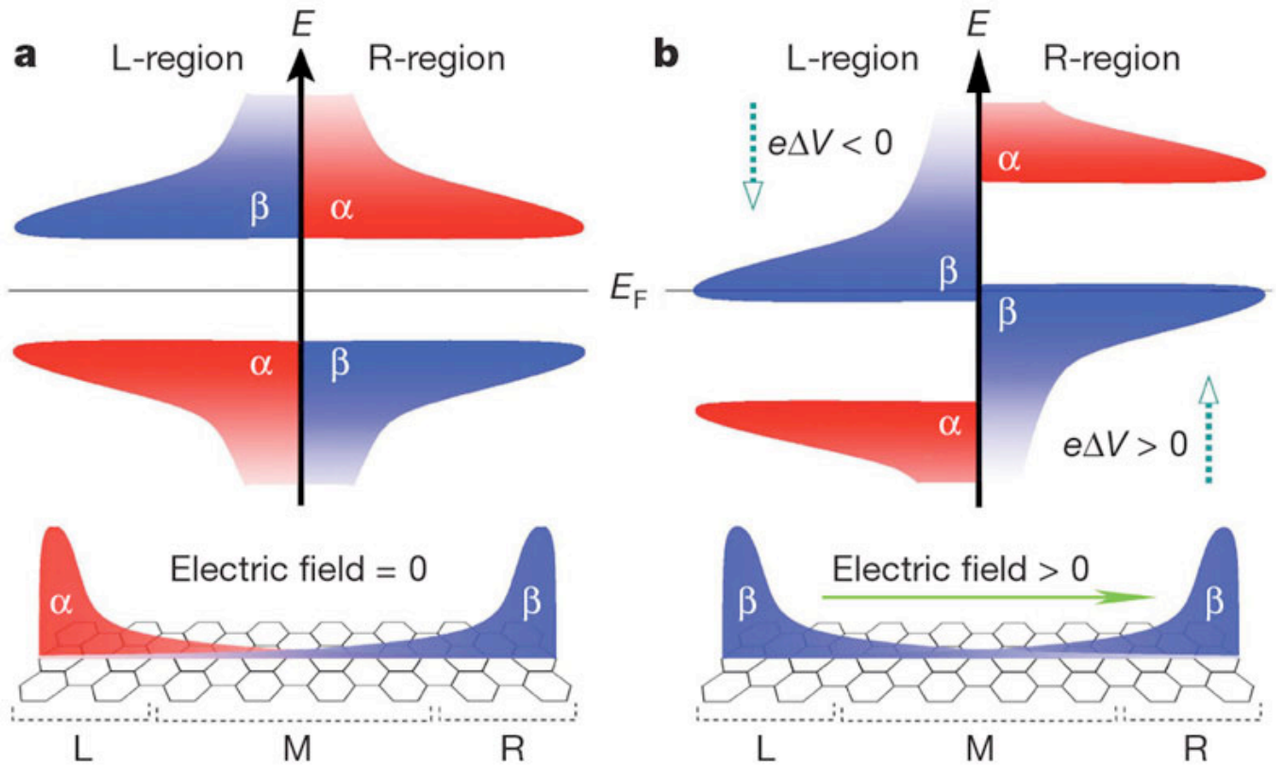
Science **304**, 42 (2004)

graphene nanoribbon

Louie's group
Nature **444**, 347 (2006)



electric field



Using electric field to manipulate the magnetic properties in zigzag graphene nanoribbon.

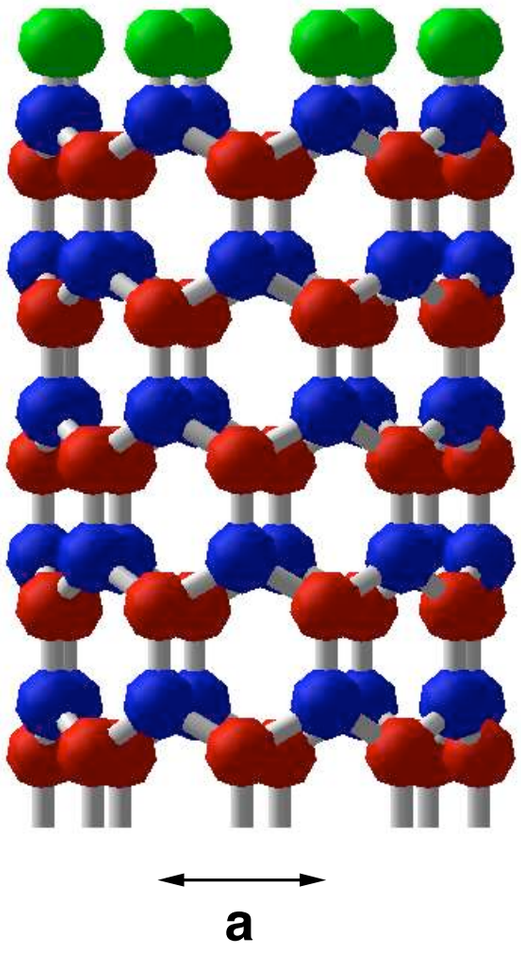
some observations...

- Electron-electron interaction can be important in graphene and related materials.
- The presence of open boundaries seems to enhance the magnetic instability and gives rise to ferromagnetic moments.
- How to quantify the moment then?

Stage I:
Lieb's Theorem

Hubbard model

To describe the electronic correlations in semi-infinite carbon nanotube, we start with the Hubbard model.

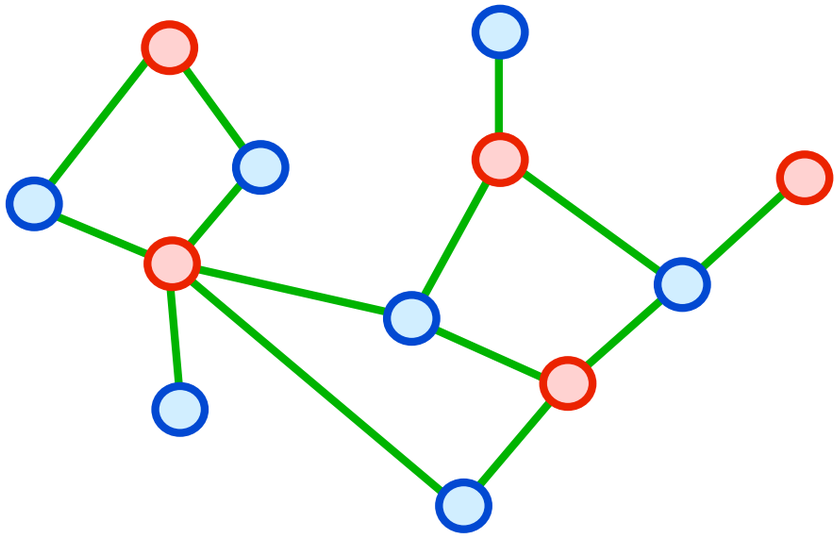


The Hubbard Hamiltonian contains two parts: hopping and on-site interaction,

$$\begin{aligned} H &= -t \sum_{\langle x,y \rangle} [c^{\dagger}_{x\sigma} c_{y\sigma} + c^{\dagger}_{y\sigma} c_{x\sigma}] \\ &+ U \sum_{x} (n_{x\uparrow} - \frac{1}{2})(n_{x\downarrow} - \frac{1}{2}), \end{aligned}$$

where t is the hopping amplitude between two sublattices and $U > 0$ is the on-site repulsive interaction.

Lieb's theorem



For the repulsive Hubbard model on bipartite lattice, there exists particle-hole symmetry at half filling.

E. H. Lieb
Phys. Rev. Lett. **62**, 1201 (1989)

Lieb showed that the ground state is unique up to the (2S+1)-fold degeneracy from the non-zero spin S.

finding S...

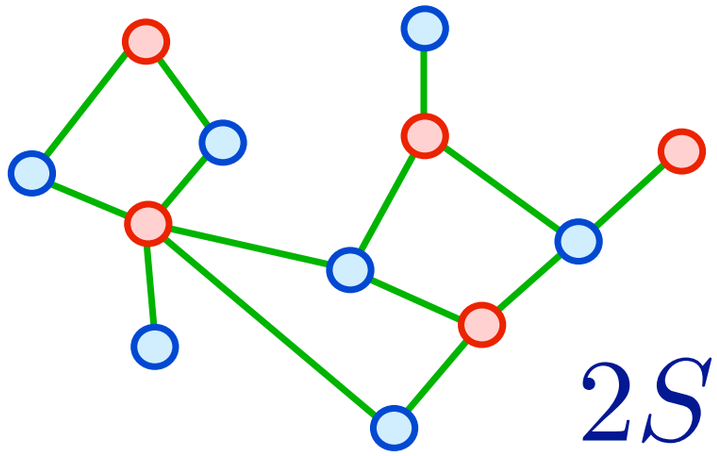
Since the previous conclusion is independent of the strength of interaction U and the hopping t, Lieb's obtained the ground-state spin S from the Heisenberg model in strong coupling

weak coupling

$$2S = N_A - N_B$$

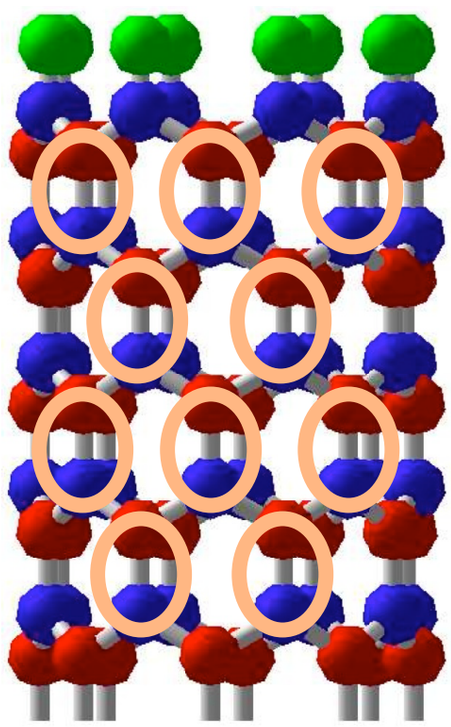
E. H. Lieb
Phys. Rev. Lett. **62**, 1201 (1989)

ambiguity arises?

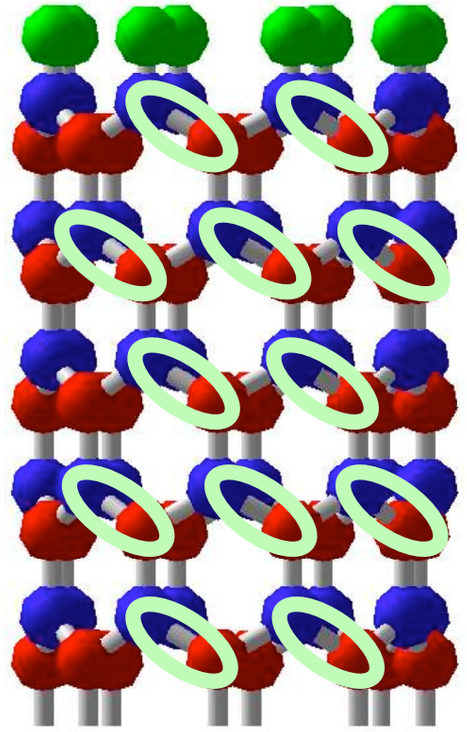


$2S = 2$

The counting is OK for finite bipartite lattices but becomes ambiguous when the lattice sites become infinite...



$2S = n$



$2S = 0$

Stage II:
Weak Coupling Analysis

weak coupling

Due to the lattice charge anomaly, we cannot determine the spin of the ground state. Thus, let's try the opposite weak-coupling limit.

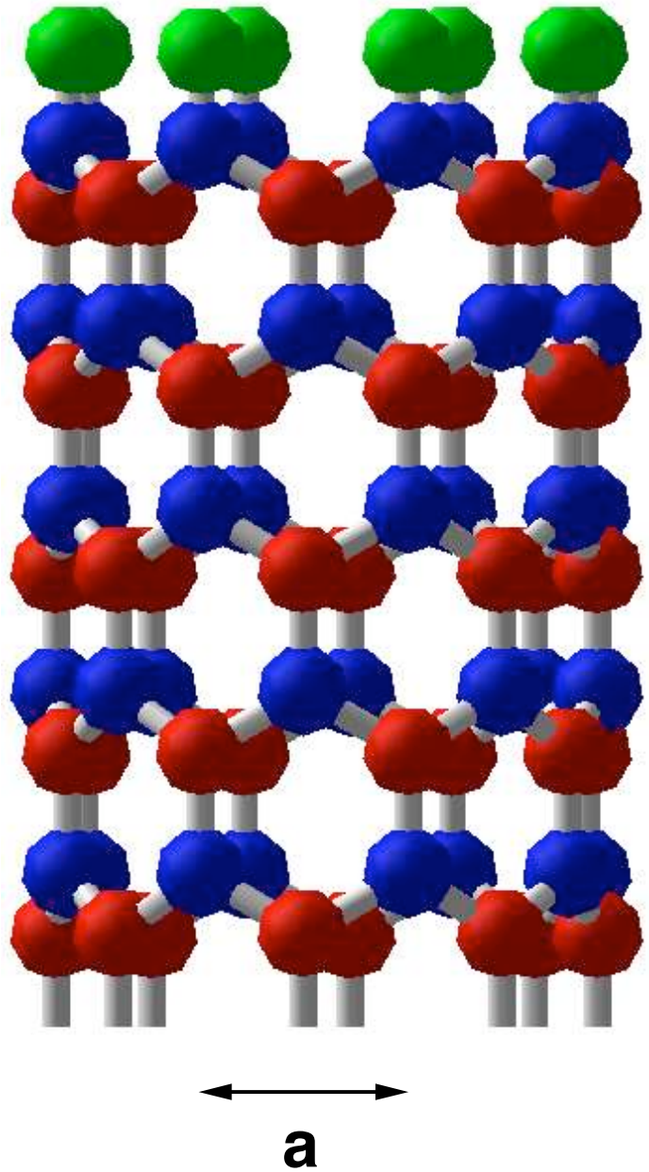
For simplicity, let me start with the semiconducting nanotubes.

Dirac Hamiltonian

Ignore the on-site interaction momentarily, the Schrödinger equation for the hopping part can be casted into the SUSY form

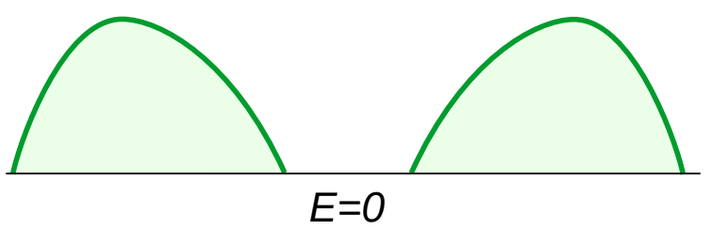
$$\begin{pmatrix} 0 & Q \\ Q^\dagger & 0 \end{pmatrix} \begin{pmatrix} \varphi_A \\ \varphi_B \end{pmatrix} = E \begin{pmatrix} \varphi_A \\ \varphi_B \end{pmatrix},$$

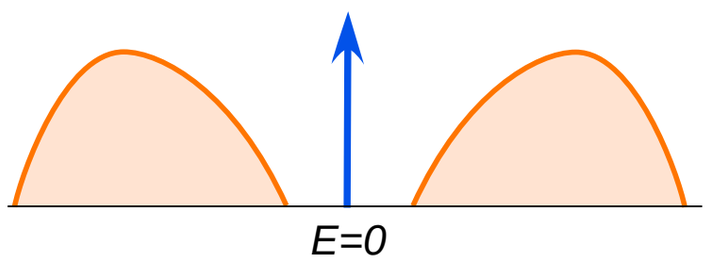
where $\varphi_{A/B}$ is the wave fucntion on sublattice A/B and $Q, Q^\dagger$ are the supercharge operators.



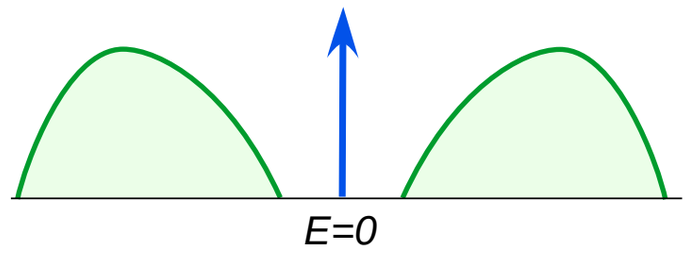
Huang, Wu and Mou
Phys. Rev. B **70**, 205408 (2004)

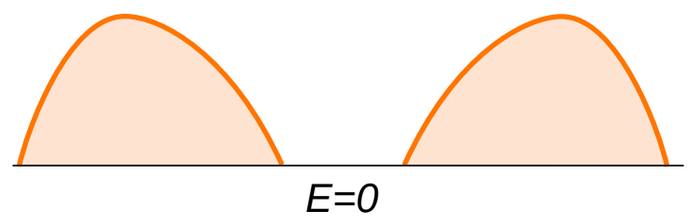
nodal structure





local density of states





$$\Psi_+ = \begin{pmatrix} 0 \\ \varphi_B \end{pmatrix}$$

$$Q\varphi_B = 0$$

$$\Psi_- = \begin{pmatrix} \varphi_A \\ 0 \end{pmatrix}$$

$$Q^\dagger \varphi_A = 0$$

Note that the wave functions have nodal structure since they are SUSY ground states.

projection onto the edge

In weak coupling, we can integrate out the bulk states and derive the effective theory by **projecting onto the edge states**. The ground state wave function takes the general form,

$$\Psi(x_1\alpha_1,...,x_{N_e}\alpha_{N_e}) = \sum_{i_a}\sum_{\beta_a} A(i_1\beta_1,...,i_{N_e}\beta_{N_e})$$
$$\times\Phi_{i_1\beta_1}(x_1,\alpha_1)...\Phi_{i_{N_e}\beta_{N_e}}(x_{N_e},\alpha_{N_e}),$$

where the summations carry over all possible edge configurations. Since all edge states are pinned at zero energy, the hopping Hamiltonian after projection vanishes. Only the interaction survives,

$$H_e = U\sum_x P_e[n_{x\uparrow}n_{x\downarrow}]P_e \geq 0.$$

fully polarized edge

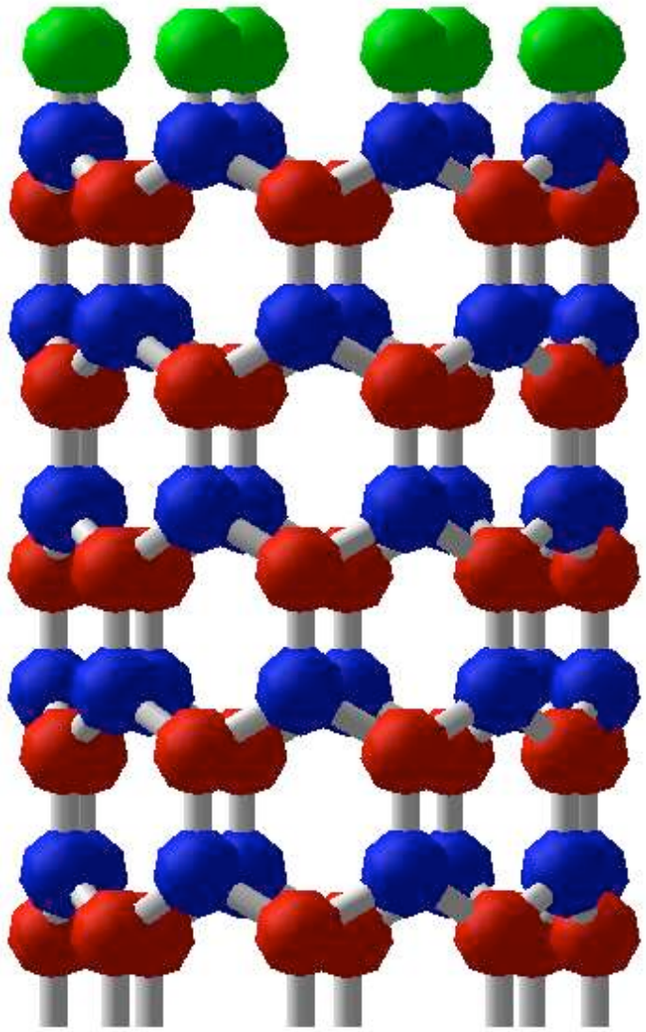
While the general form looks messy, the actual solution of the ground-state wave function turns out to be rather simple.

$$A(i_1\beta_1, ..., i_{N_e}\beta_{N_e}) = \epsilon_{i_1 ... i_{N_e}} \delta_{\beta_1\uparrow} ... \delta_{\beta_{N_e}\uparrow}$$

Since the spatial part is fully antisymmetric, the particles never meet each other on the same spot and achieve the minimal energy zero.

Stage III:
Counting Edge States

generalized Bloch theorem



Pereg-Barnea and Lin
Europhys. Lett. **69**, 791 (2005)

In the presence of open boundary, the **displacement operator** is no longer unitary $DD^\dagger \neq D^\dagger D$. Thus, the Bloch theorem needs some modification,

$$\Psi_+(n) = \sum_{i=1}^{N_<} c_i \, \mathbf{\Phi}_i \, z_i^n ,$$

where $\mathbf{\Phi}_i$ are $(n_x + n_y)$-dimensional eigenvectors and $N_<$ is the number of decaying modes with $|z_i| < 1$. The coefficients c_i must satisfy the n_y constraints due to the open boundary.

solving for z

Making use of the generalized Bloch theorem, the problem is reduced to the search for the **null space** of the (n_x+n_y)-dimensional supercharge,

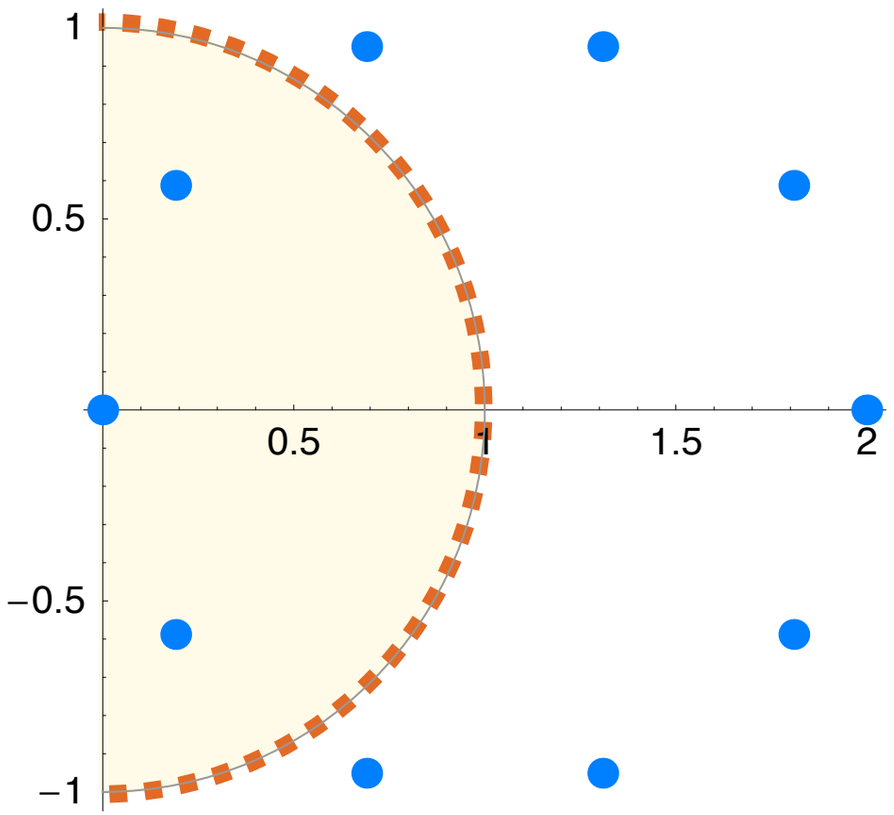
$$Q(z)\Phi = 0 \quad \rightarrow \quad \det Q(z) = 0.$$

In general, the determinant would give $(n_x + n_y)!$ terms of the polynomials of z. However, by appropriate choice of the unit cell, the determinant is greatly simplified.

$$\left(z-1\right)^{n_x}\left(\frac{1}{z}-1\right)^{n_y} = 1$$

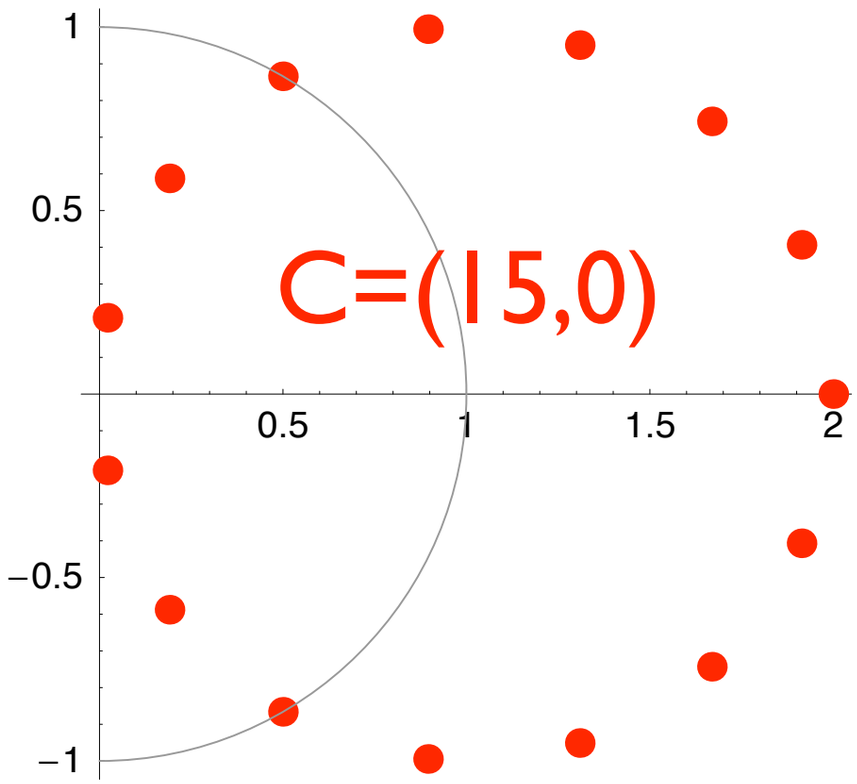
zigzag dots

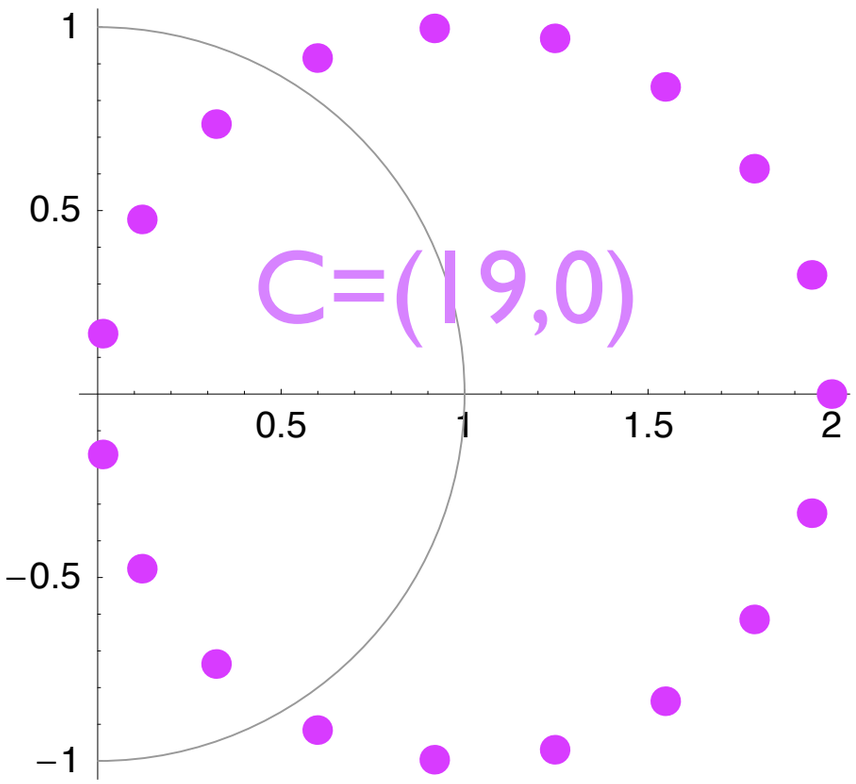
C=(10,0)



Taking (10,0) zigzag carbon nanotube as an example, there are ten solutions in the complex plane. Only three of them are decaying modes with $|z|<1$.

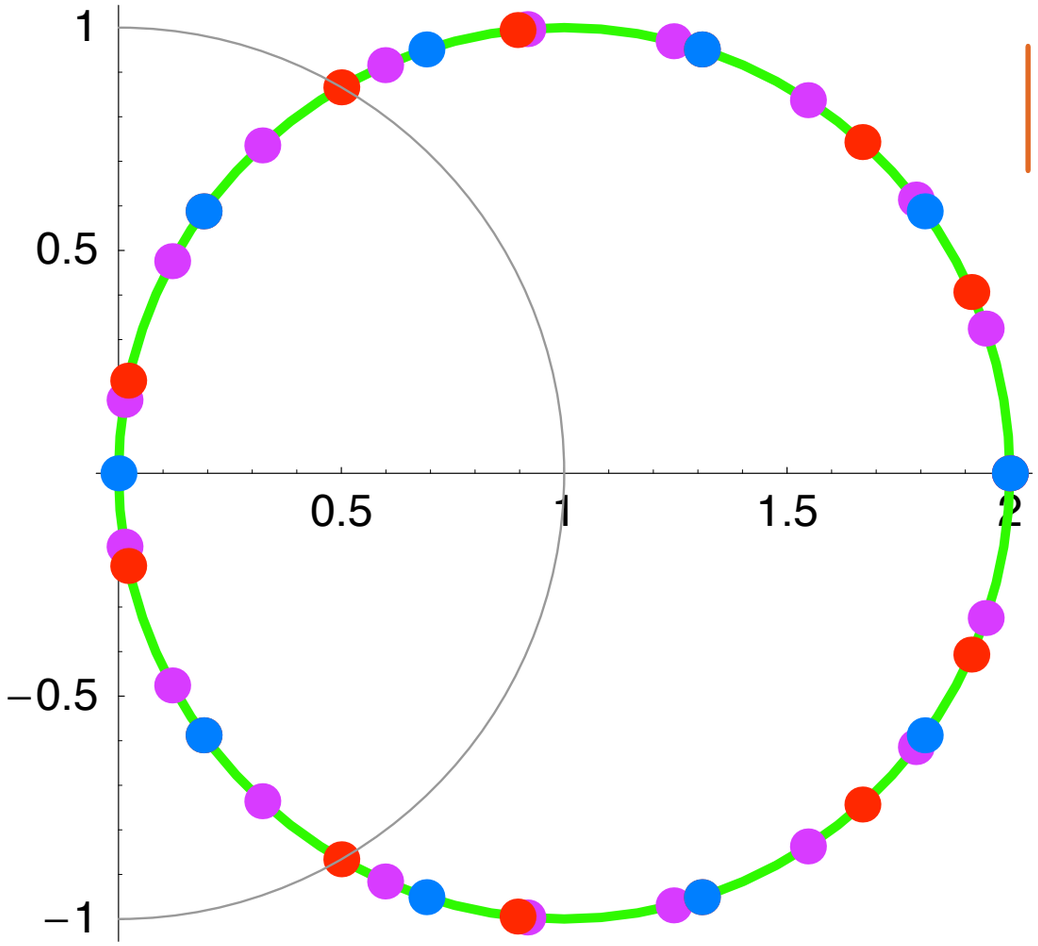
other family members





In fact, for nanotubes with parallel chiral vectors, the solutions fall onto the universal contour in the complex plane.

zigzag family contour

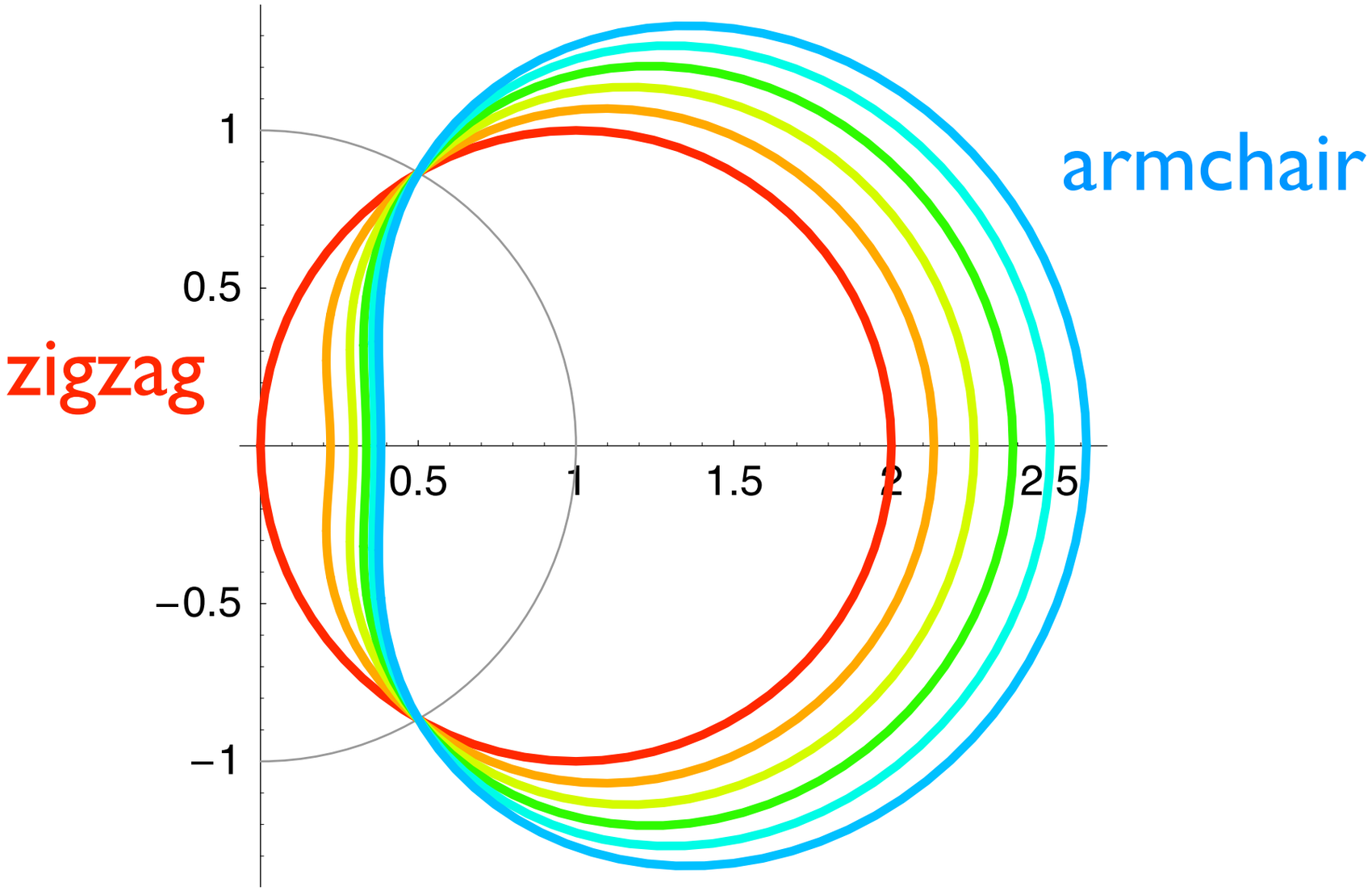


$$|z-1|=1$$

$$M=\mu_B\left[\frac{n_x+1}{3}\right]$$

The contour in the complex plane for the zigzag family with C=(nx,0).

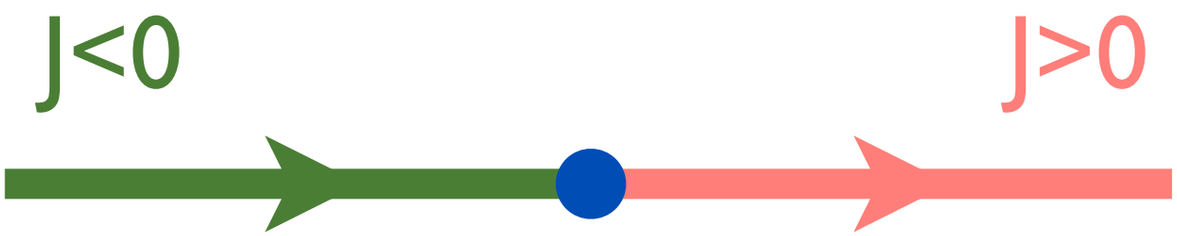
eye-ball contours

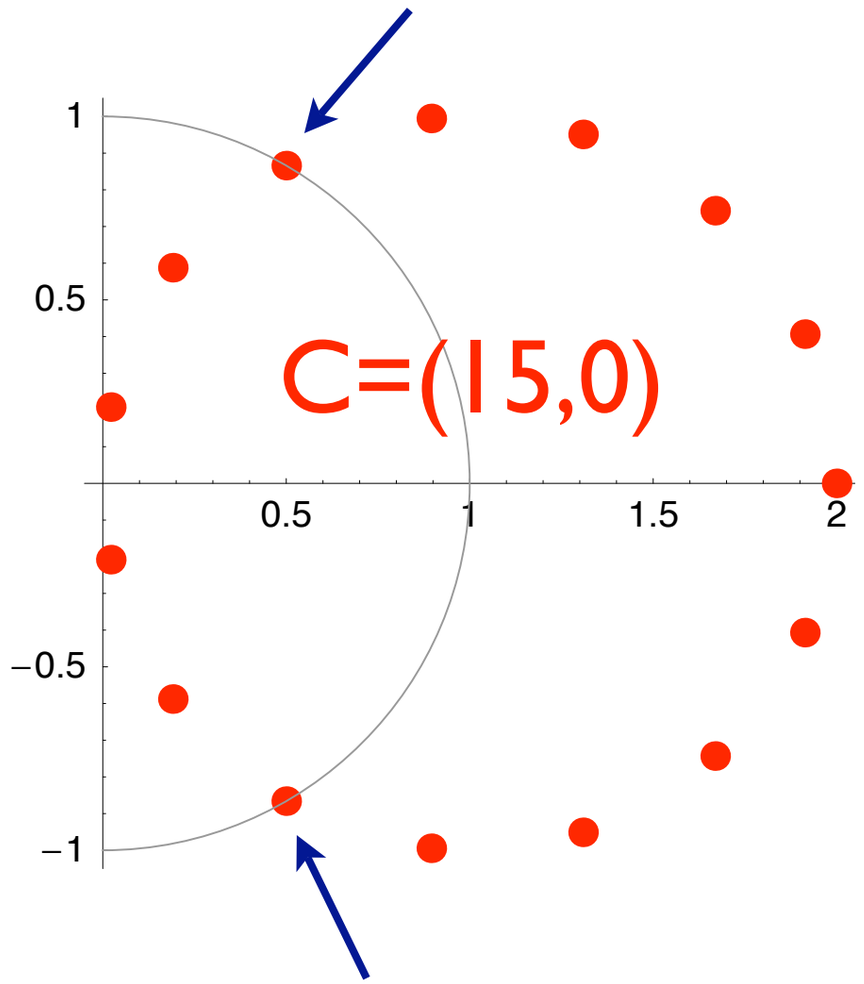


Contours in the complex z-plane for nanotubes with different chiralities (from zigzag to armchair).

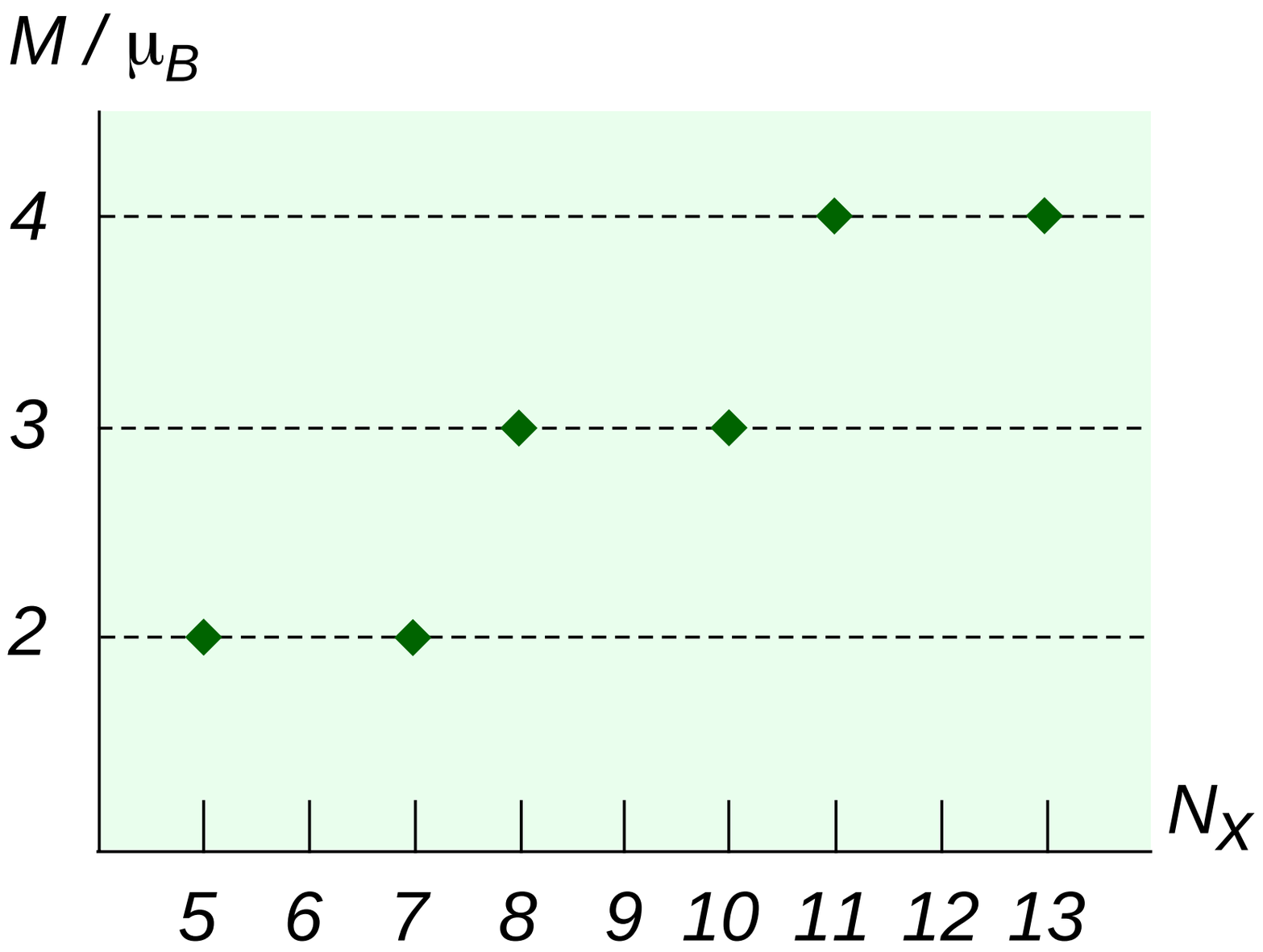
metallic nanotube

For (15,0)-zigzag nanotube, we have four edge states coupled to two conducting channels. The effective theory in the spin sector is similar to the Kondo model.





quantized moments



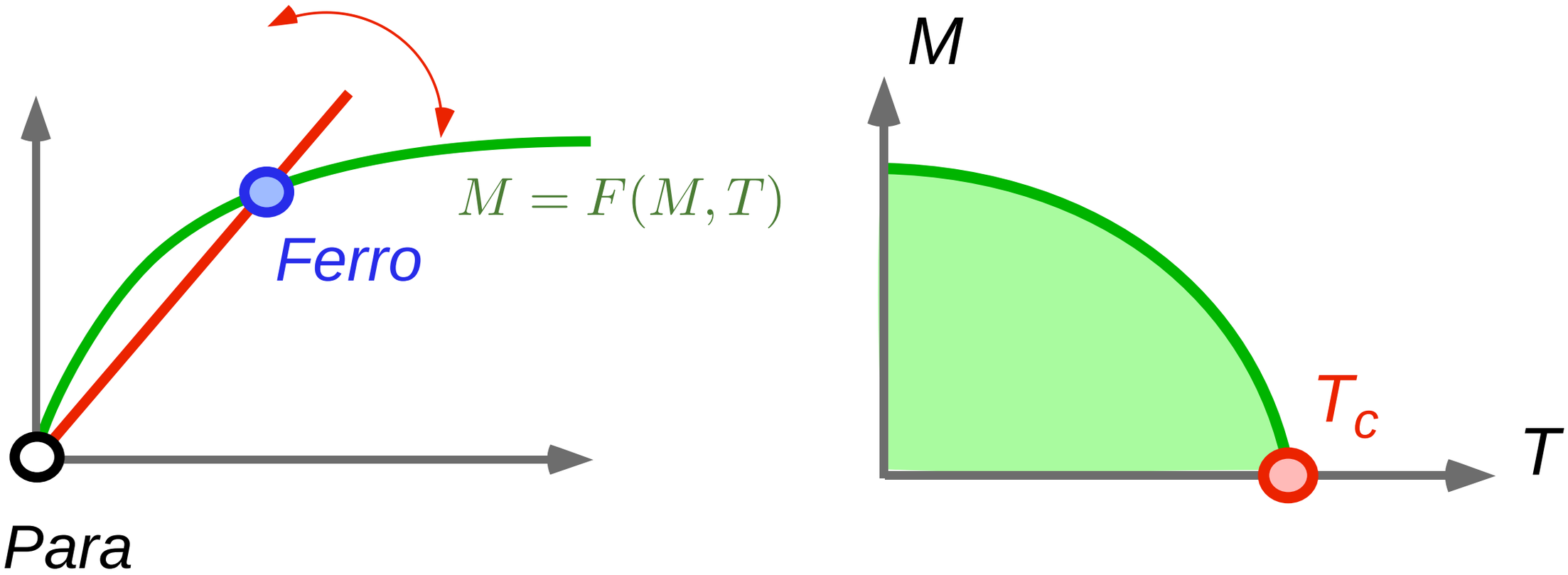
Making it Realistic...

first-principles approach

Can the beautiful quantization of magnetic moment be an artifact of Hubbard model? What happens in a more realistic carbon nanotube?

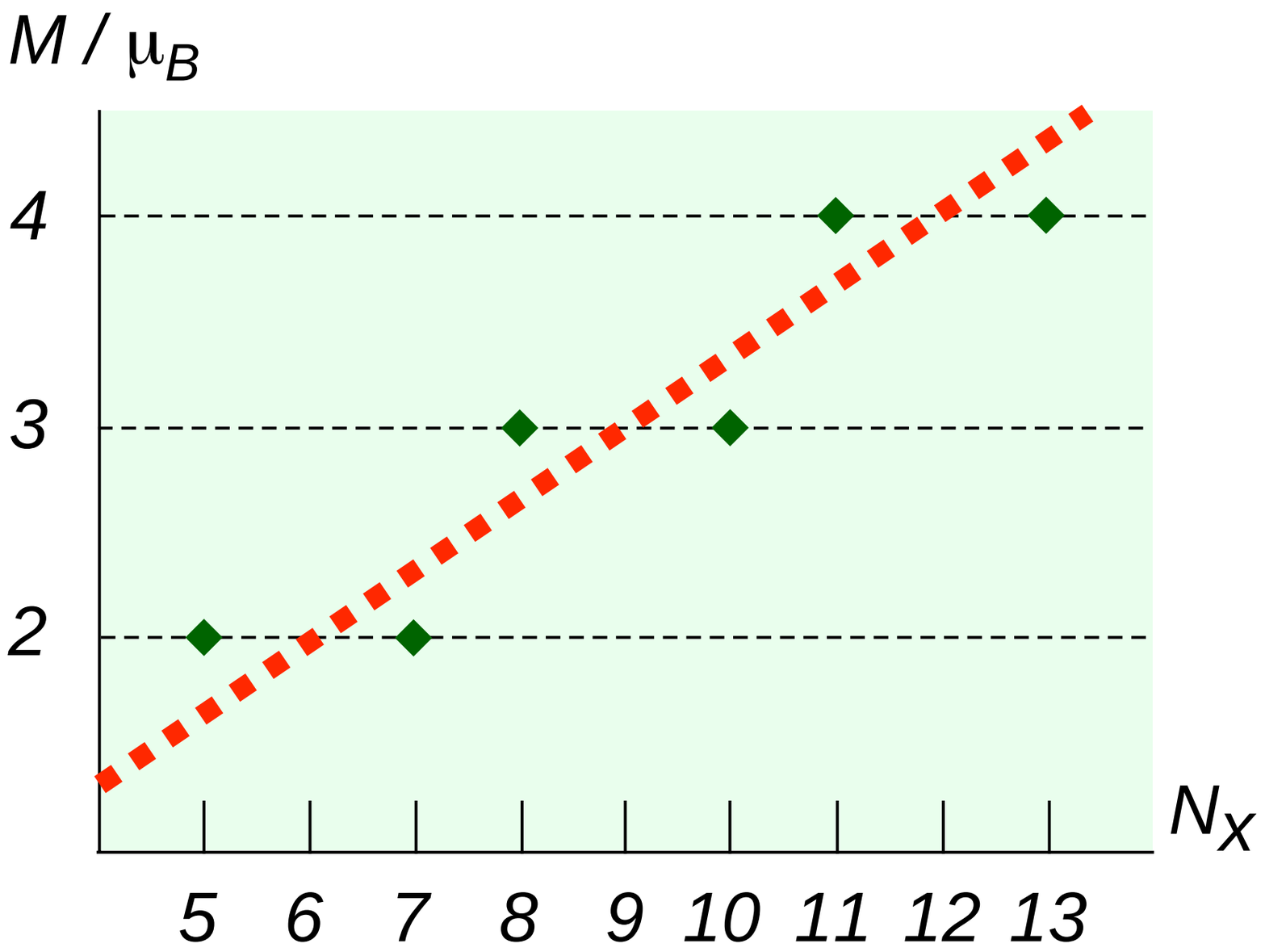
- Hopping in tight-binding limit may not be sufficient.
- Coublomb interaction is likely not screened and remained long-ranged.
- It's a generalized mean-field theory...

Weiss mean-field theory



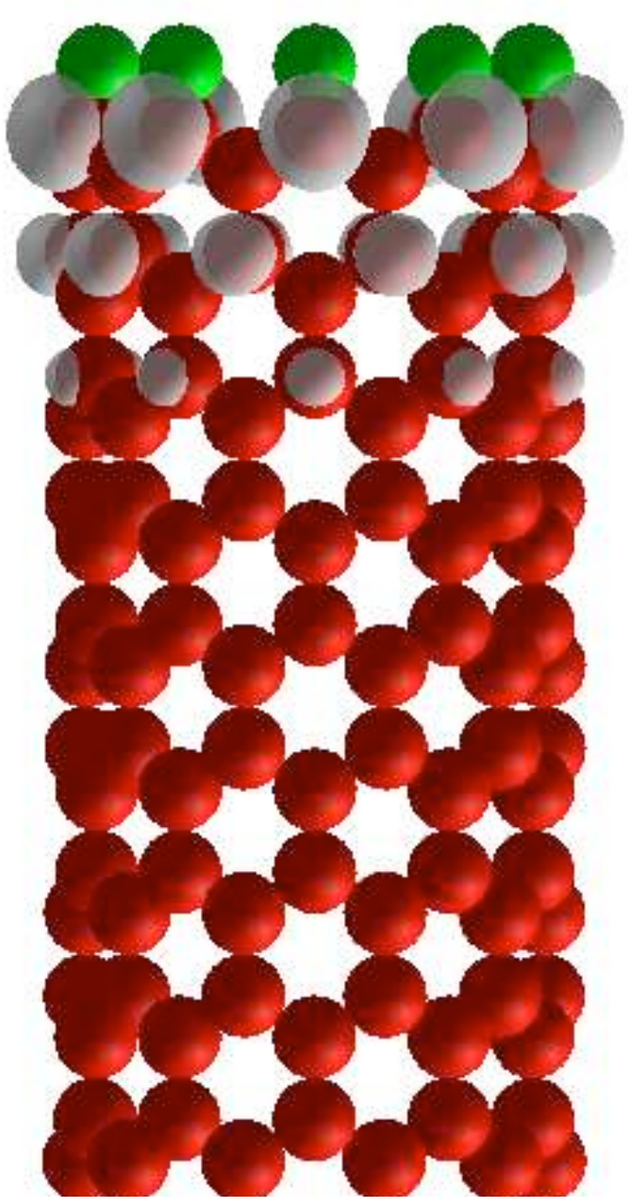
In Weiss mean-field theory, the magnetization is determined by some self-consistent equation and often changes continuously with temperature and other parameters.

mean-field expectation



magnetization profile

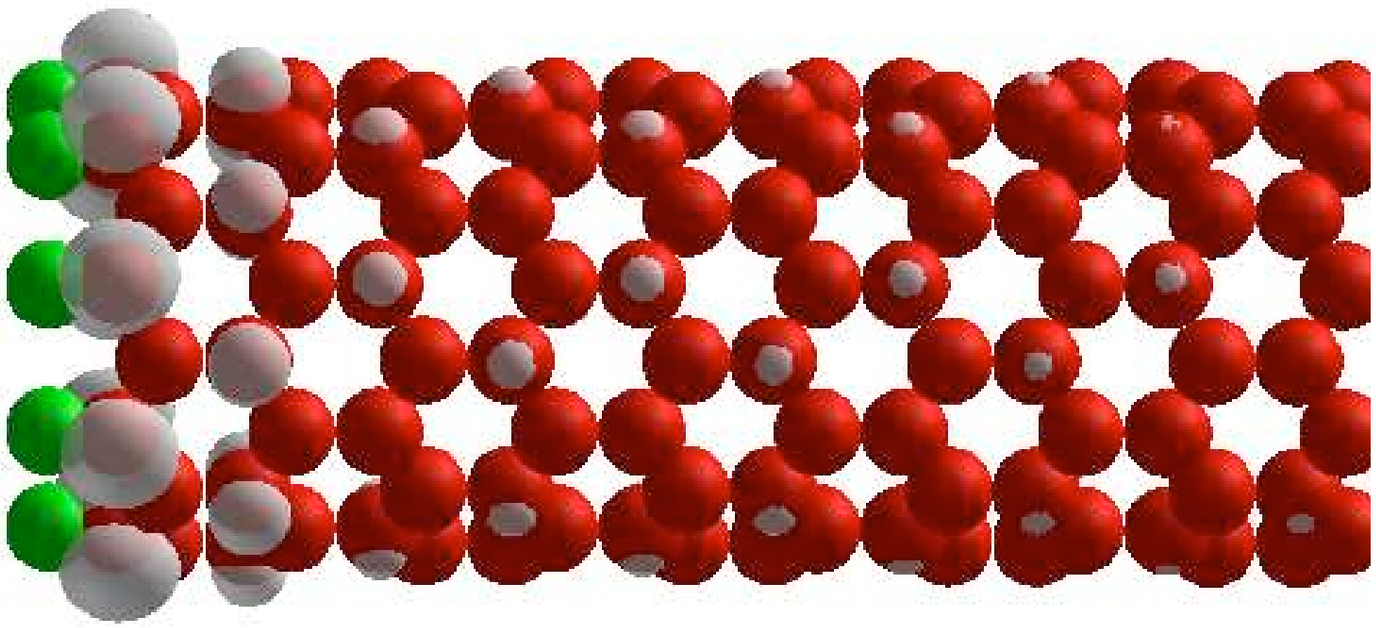
- LSDA calculations show that the ground state is ferromagnetic.
- Moments are localized near the edge.
- Nodal structure of the density profile is robust.



C=(10,0)

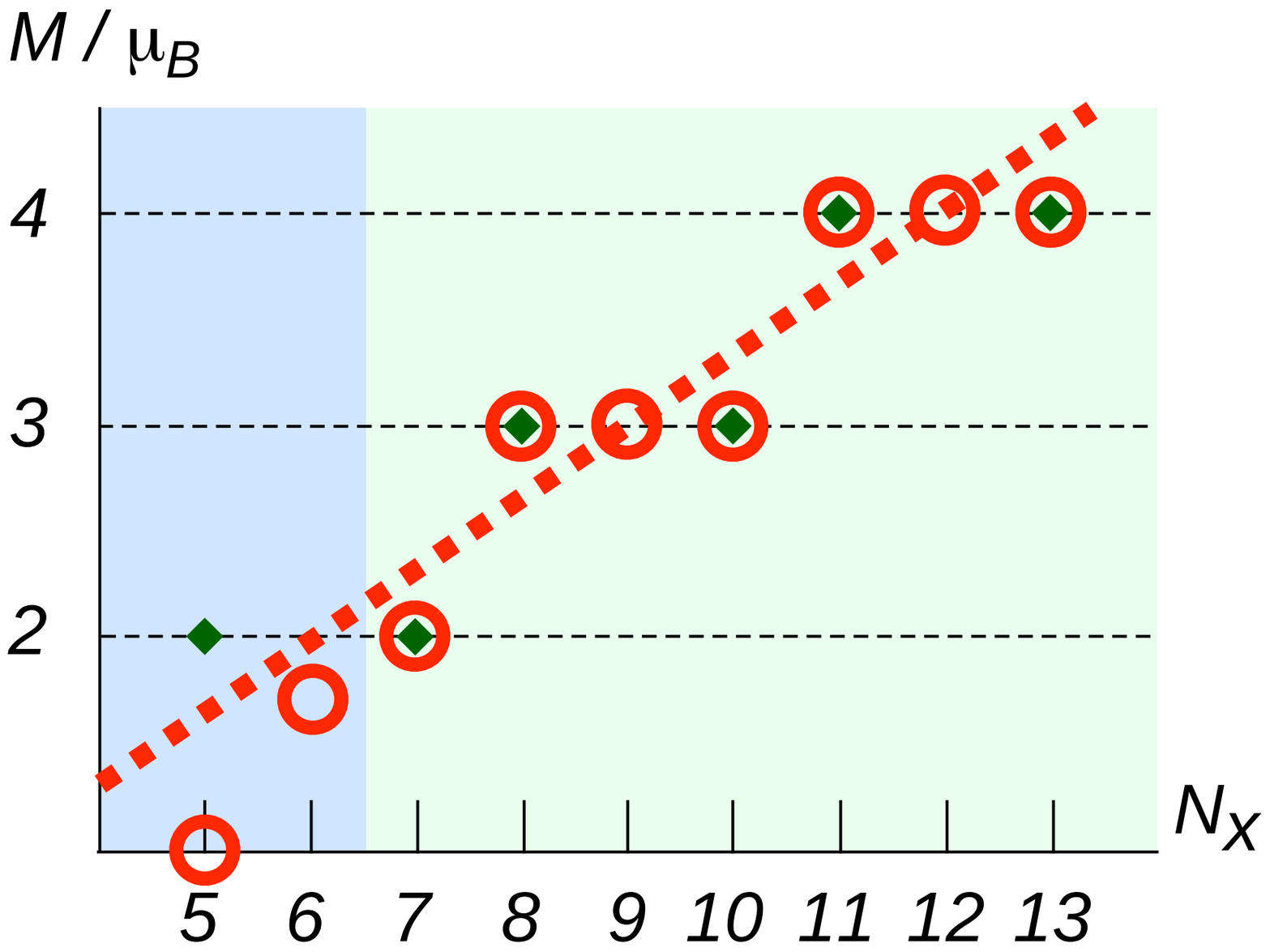
metallic nanotube

The conclusions remain the same except the magnetization extends further into the bulk as expected.



C=(9,0)

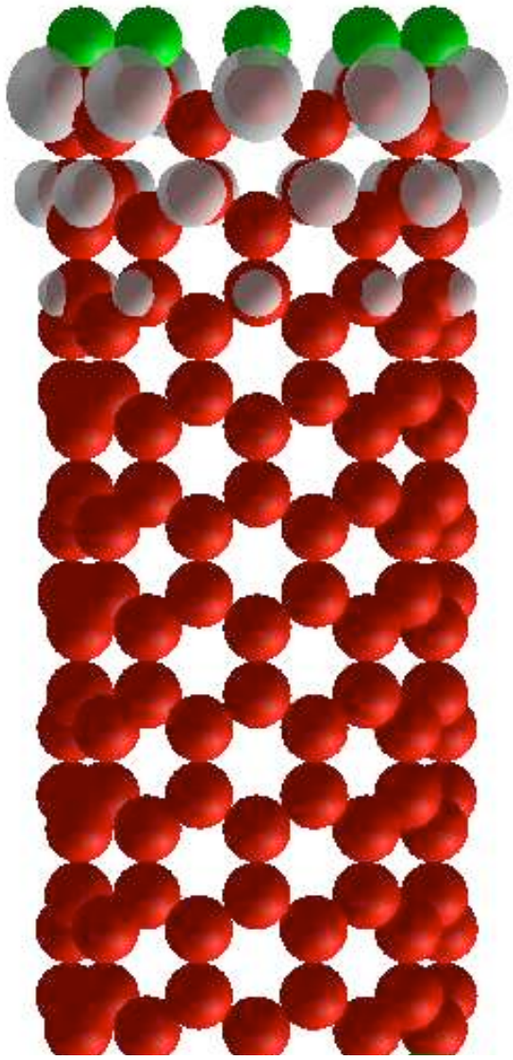
surprise!



Conclusions

- ***Magnetic moments*** spotted at the edge of generic single-wall carbon nanotubes.
- The moments are ***quantized*** and topologically robust.
- Time to rethink the physics in graphene related materials ***due to electronic correlations.***

topological nanomagnet



We found the ground state of the semi-infinite carbon nanotube has **quantized magnetic moment**,

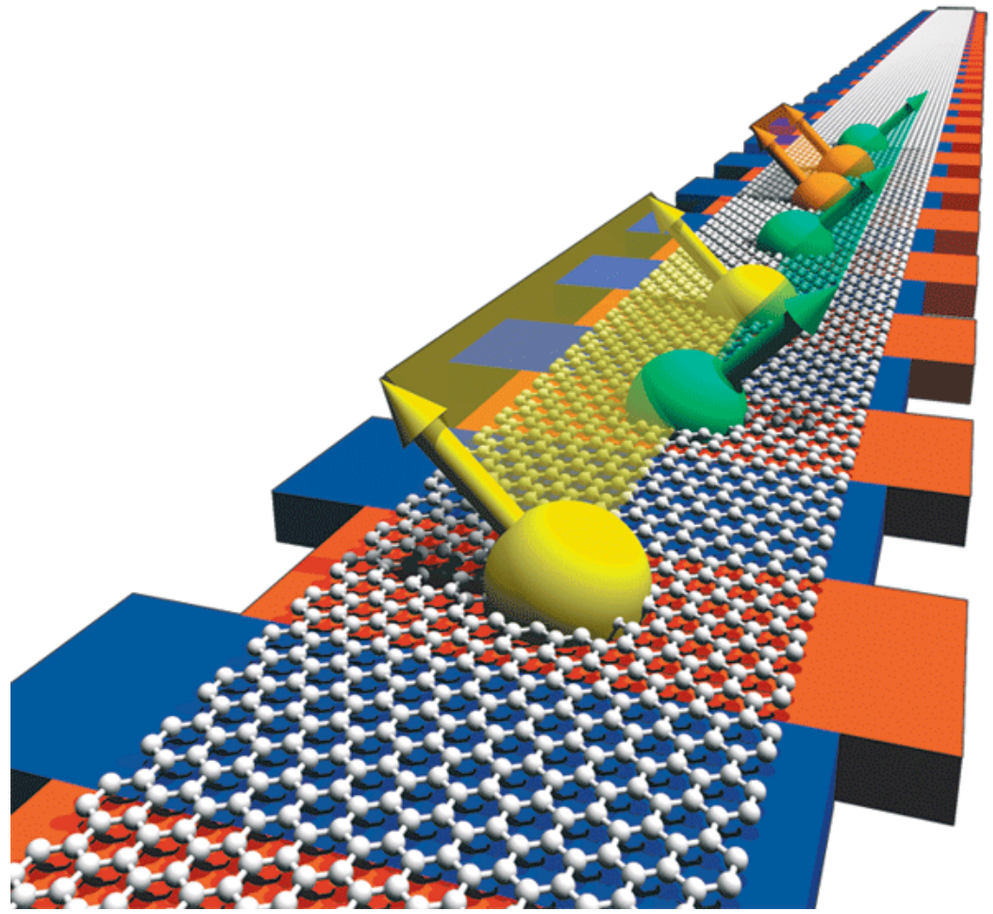
$$M = \mu_B \left[\frac{n_x - n_y + 1}{3} \right],$$

which is dictated by the **chiral vector** but does not depends on the hopping amplitude and the interaction strength!

Backup Slides

building qubits...

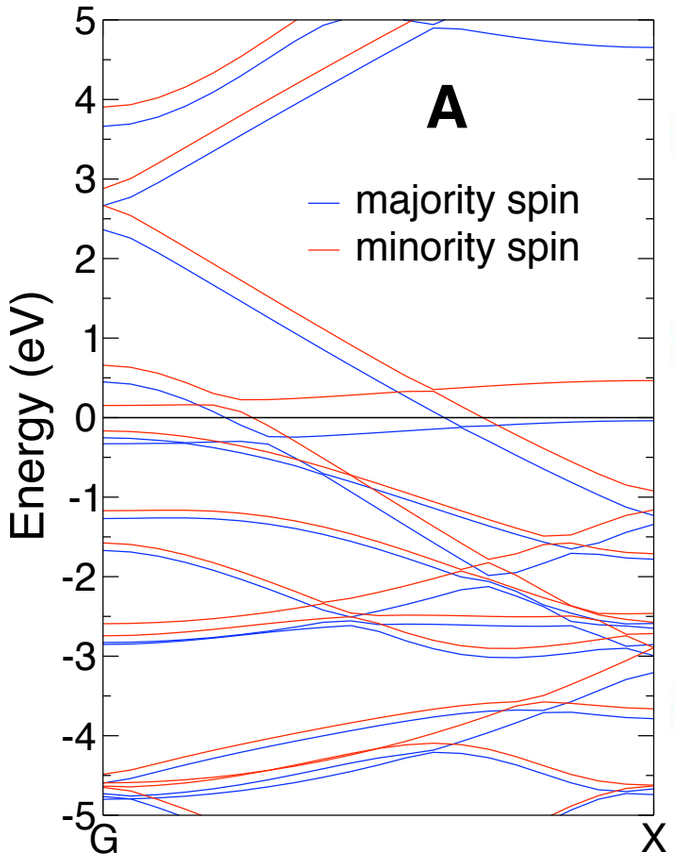
- Blue gates define the quantum dots and give rise to bound states.
- Carriers in the bound states act as qubits.
- Red gates control the exchange interaction between the qubits.

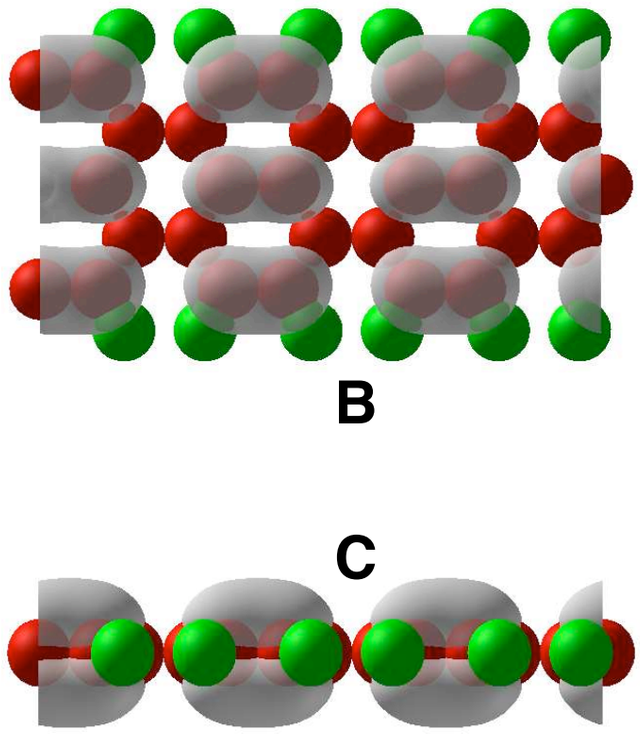


Burkard's group
Nature Phys. **3**, 192 (2007)

flat-band ferromagnetism

However, at nanoscale, edge topology and electronic correlations are important.





- Flat band comes from quantum caging effect!
- Ferromagnetic ground state arises from mutual interactions between electrons.

Lieb's theorem

Let us detour a bit and consider the **attractive** Hubbarde model. The hopping part is the same but the interaction changes sign,

$$H = -t \sum_{\langle x,y \rangle} [c^{\dagger}_{x\sigma} c_{y\sigma} + c^{\dagger}_{y\sigma} c_{x\sigma}] - |U| \sum_{x} (n_{x\uparrow} - \frac{1}{2})(n_{x\downarrow} - \frac{1}{2}).$$

Making use of the spin-reflection positivity from the on-site interaction, Lieb proved several interesting and important theorems. One of them reads:

E. H. Lieb
Phys. Rev. Lett. **62**, 1201 (1989)

For attractive on-site interaction U<0, the ground state is unique with S=0.

particle-hole symmetry

Now coming back to the repulsive Hubbard model. At half filling $\langle n \rangle = 1$, the model is particle-hole symmetric. Since all states with different S all have one representative in $S_z^{tot} = 0$ subspace, it is sufficient to consider states with $N_\uparrow = N_\downarrow = N_s/2$.

Perform the particle-hole transformation in the spin-up sector while leaving the spin-down intact, $c_{x\uparrow} \to \epsilon(x) c^\dagger_{x\uparrow}$, where $\epsilon(x) = 1$ for sublattice A and $\epsilon(x) = -1$ for sublattice B. Since the density operator transforms as $n_{x\uparrow} \to 1 - n_{x\uparrow}$,

$$U(n_{x\uparrow} - \frac{1}{2})(n_{x\downarrow} - \frac{1}{2}) \to -U(n_{x\uparrow} - \frac{1}{2})(n_{x\downarrow} - \frac{1}{2})$$

the **repulsive** interaction is mapped to the **attractive** one.

unique ground state

The particle-hole symmetry is crucially important to establish the link. After the transformation, the pseudo-spin operator is $\tau_z^{tot} = \frac{1}{2}(N_s - N_\uparrow - N_\downarrow) = 0$. Once the particle-symmetry is broken, the mapping generates a finite magnetic field and the ground state is no longer a pseudo-spin singlet $\tau^{tot} = 0$.

Therefore, Lieb's theorem also shows that the ground state of the repulsive Hubbard model at half filling is also unique except the total spin S is left to be determined.

lattice charge anomaly

This ambiguity is similar to the chiral anomaly we encounter in quantum field theory. Suppose we define the lattice charges $q_A(x), q_B(x)$ in the following way, $q_A(x) = \delta_{x,A}$ and $q_B(x) = \delta_{x,B}$. The Lieb theorem simply states that the total spin is

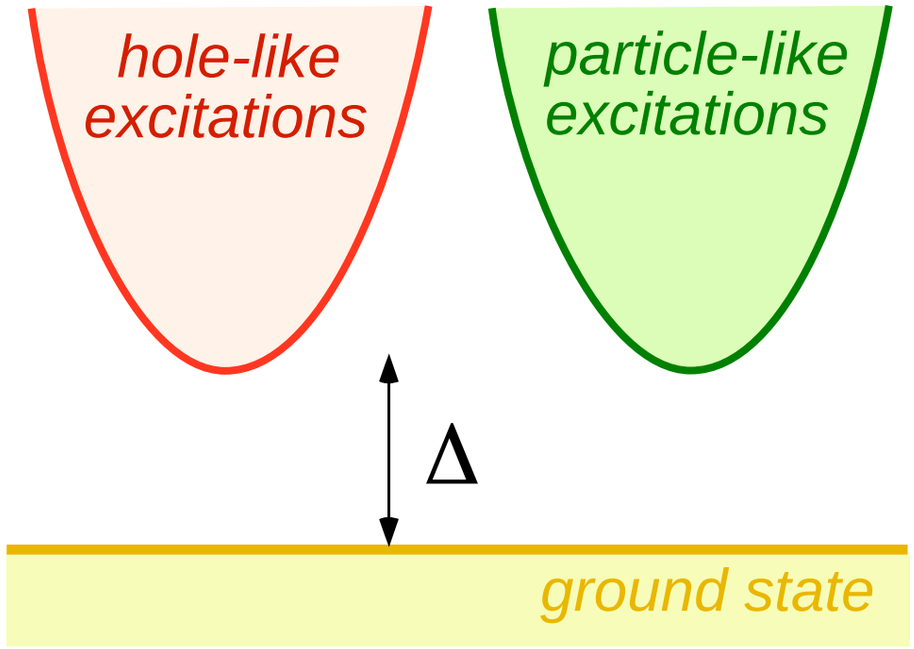
$$2S = \sum_x q_A(x) - \sum_x q_B(x).$$

Since the summation diverges, one needs to regulate the summations to obtain finite value. After regularization, the difference between the total lattice charge is

$$2S = \frac{n_x - n_y}{3}.$$

We know this cannot be the right answer since it is not an integer for semiconducting carbon nanotubes.

gapped spectrum



Consider the simple Dirac Hamiltonian with interactions coupling the fields with opposite chiralities. The spectrum $E(k) = \sqrt{k^2 + \Delta^2}$ is known to have **a finite gap** Δ to all excitations.

$$H_D = (\psi_+^\dagger \quad \psi_-^\dagger) \begin{pmatrix} 0 & p + i\Delta \\ p - i\Delta & 0 \end{pmatrix} \begin{pmatrix} \psi_+ \\ \psi_- \end{pmatrix}$$

midgap states

However, the Schrödinger equation allows midgap states with dispersion $E(\kappa) = \sqrt{\Delta^2 - \kappa^2} < \Delta$,

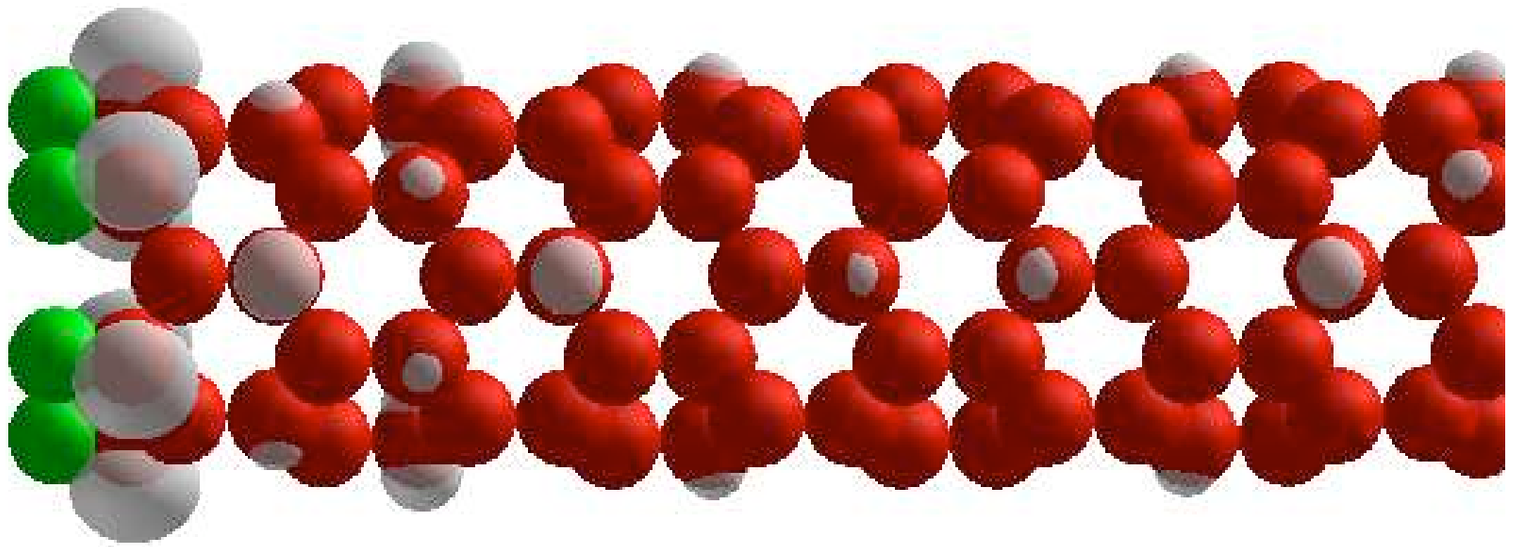
$$\psi_{R/L}(x) = C_{R/L}\ e^{\kappa x}.$$

While the Hamiltonian does support these midgap states, they do not satisfies the boundary condition $|\psi(\pm\infty)|^2 < \infty$.

Since the open boundary can be viewed as the impurity potential in the unitary limit, the midgap states occur right at the middle of the gap, i.e. at zero energy.

curvature effect

For nanotube with small radius, the curvature effect destroys the perfect quantization.



C=(6,0)

When cut into the flattened nanoribbon geometry, the quantization is recovered.