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Carbon atomic structures promoting twisted growth of second Graphene layer at magic angles 21.78° and 27.8°.

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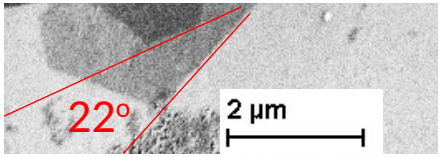
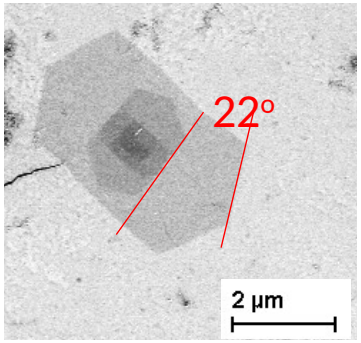
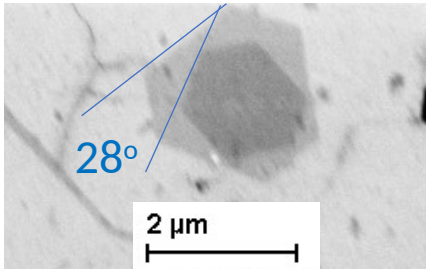
Turbostratic Graphene multilayer islands twisted at ~22° & ~28°

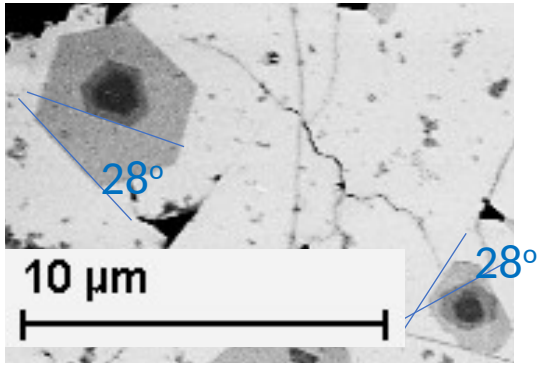
Observations

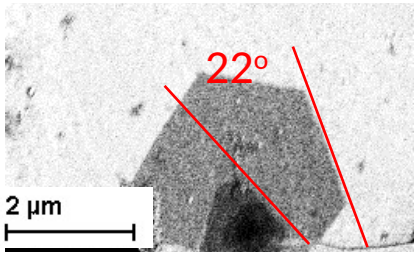
SEM micrographs

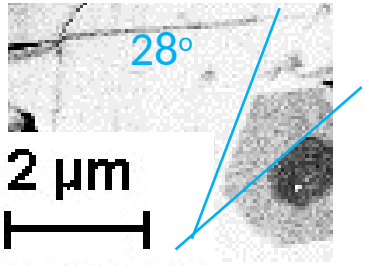


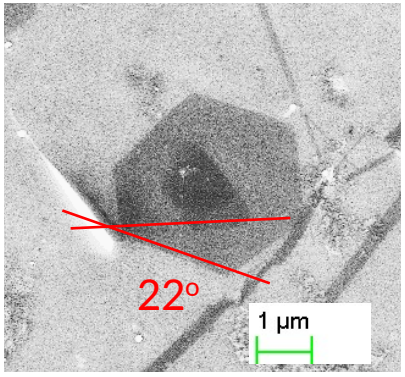


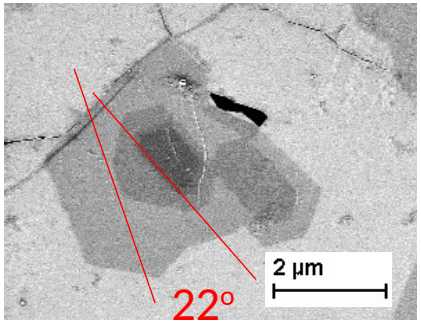


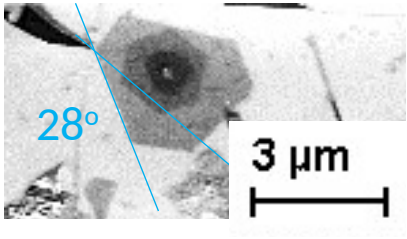


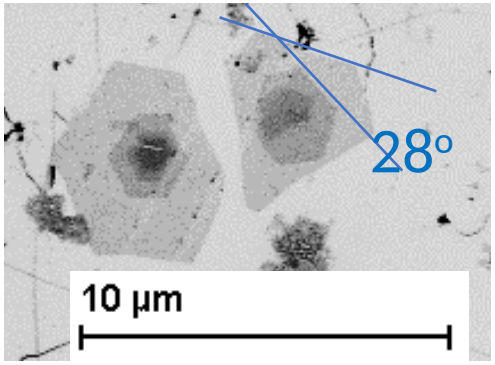






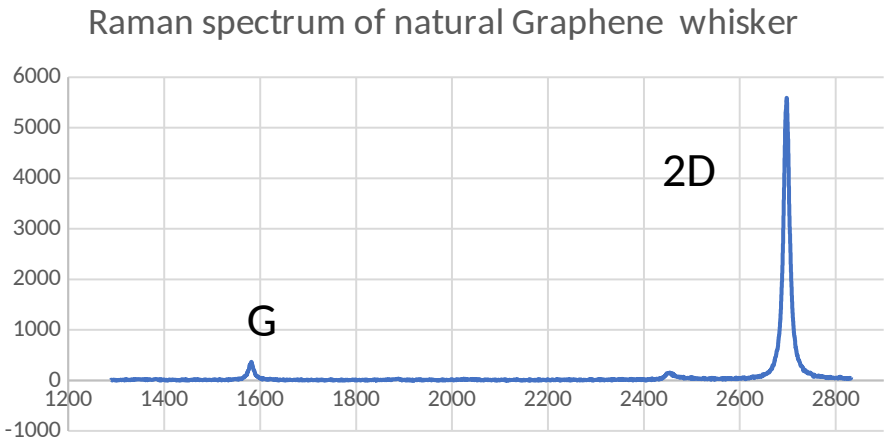


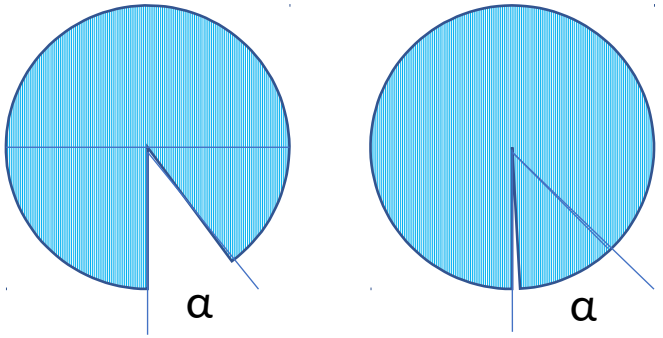


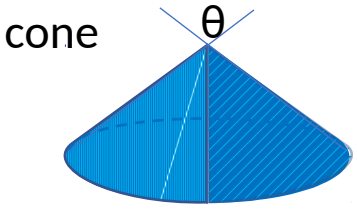


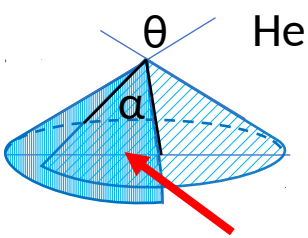
CVD Graphene grown on Cu foil and than transferred onto a glass substrate

Magic angles in Conical Graphene structures of biographene



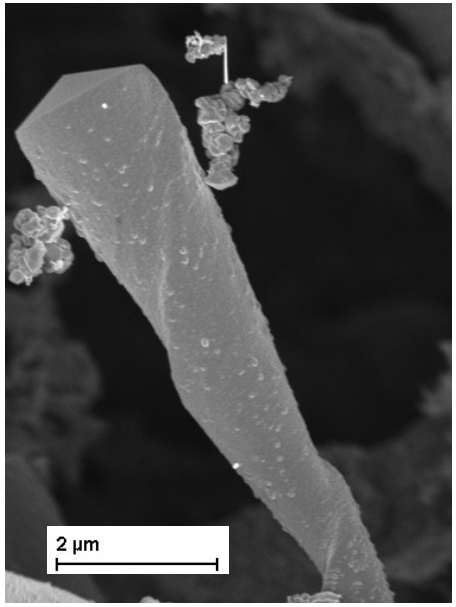


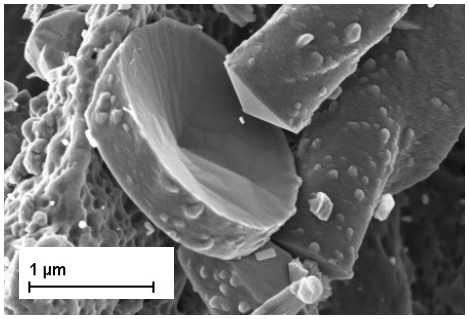


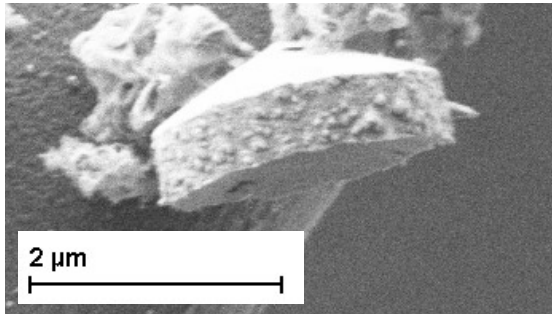


Bilayer twisted at angle α

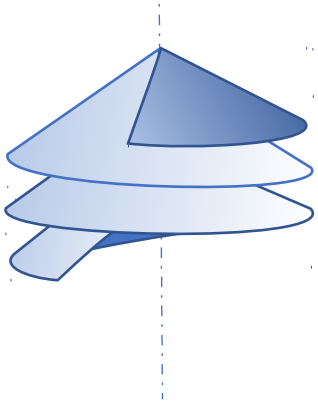
Graphene structures from natural biomaterials



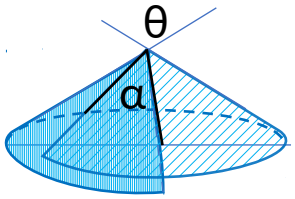




$$\Theta = 2 \times \arcsin(1-\alpha/360)$$

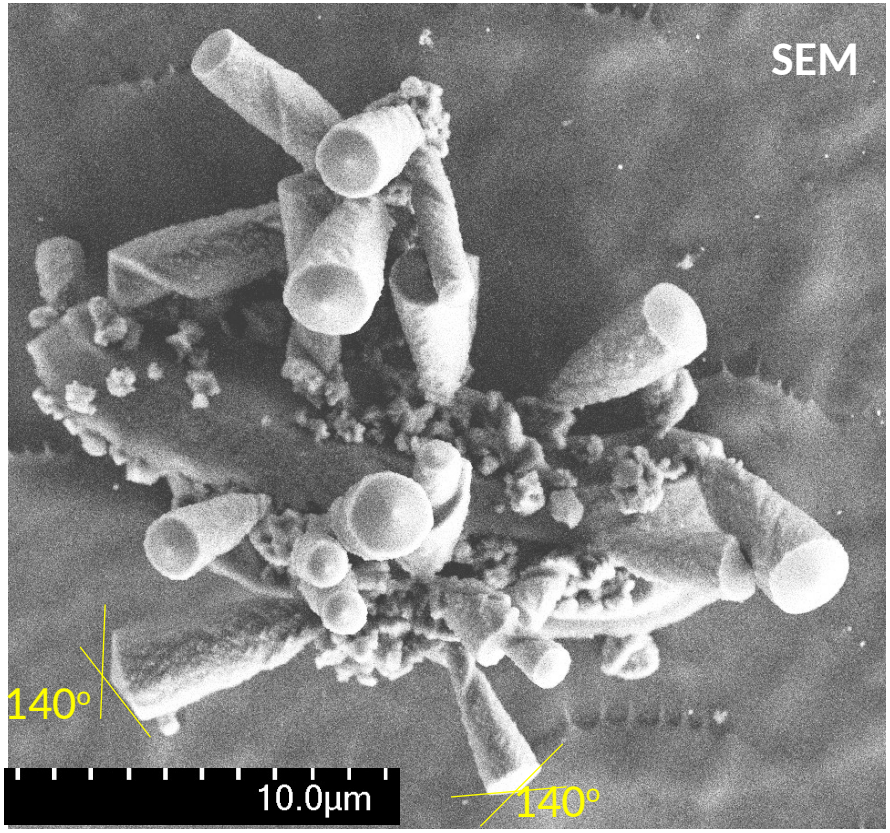


Whisker from helical cone

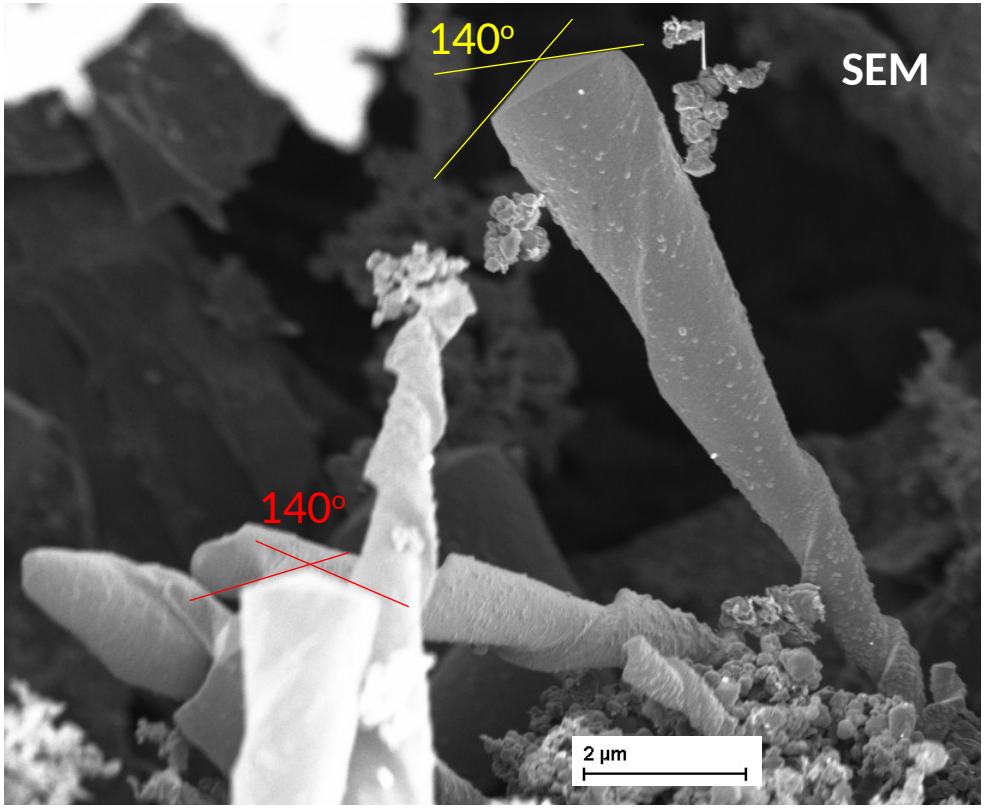


Graphene structures from natural biomaterials

Cone angle θ = 140° : Twist angle α = ~21.78°

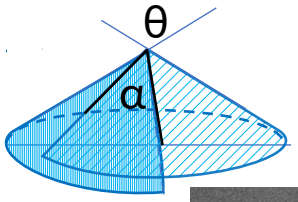


Dung Beetle leg



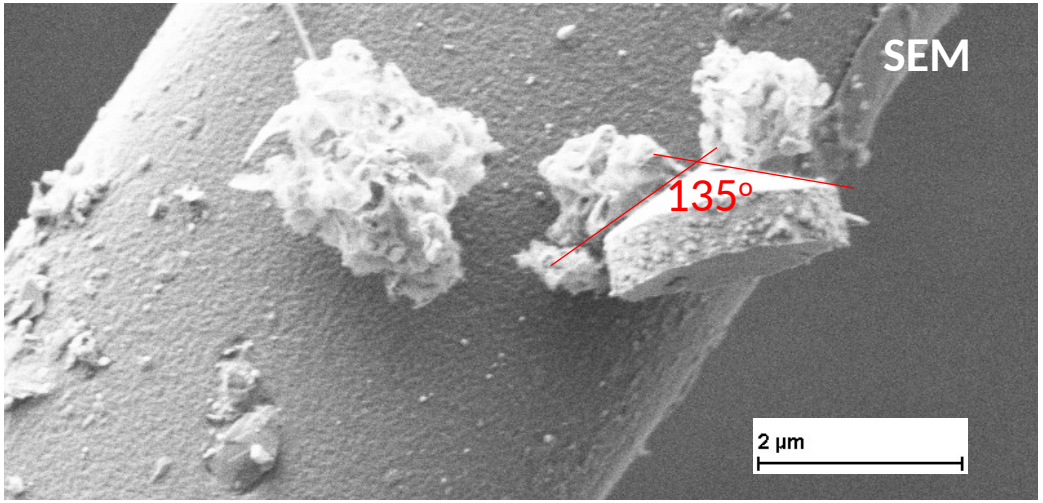
Anturium leaf

The same cone angles for different materials



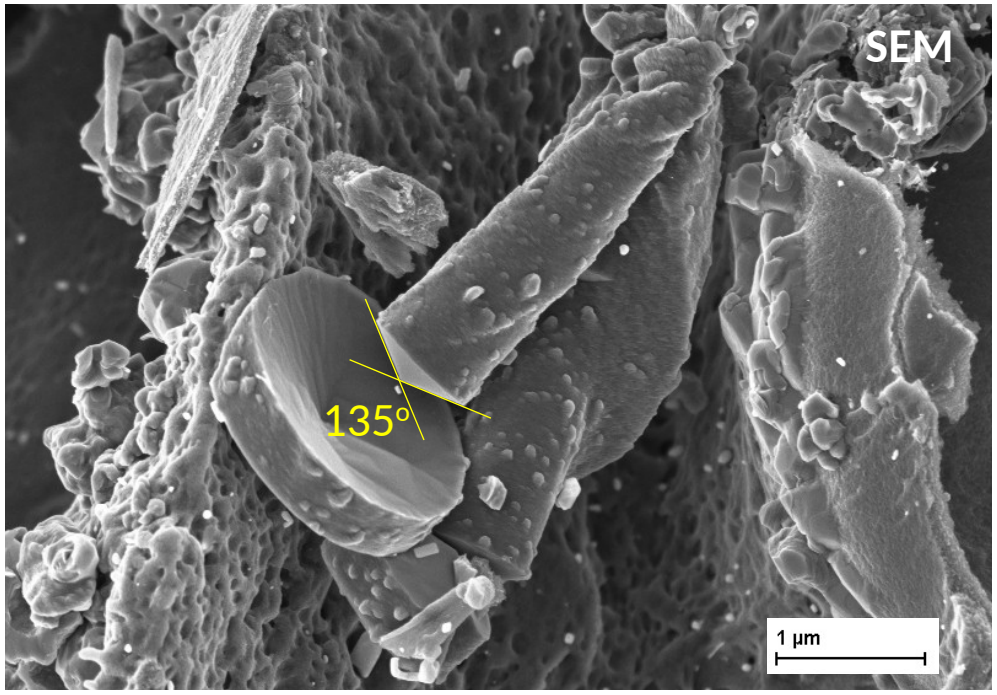
Graphene structures from natural biomaterials

Cone angle θ = 135° : Twist angle α = ~27.8°



silk

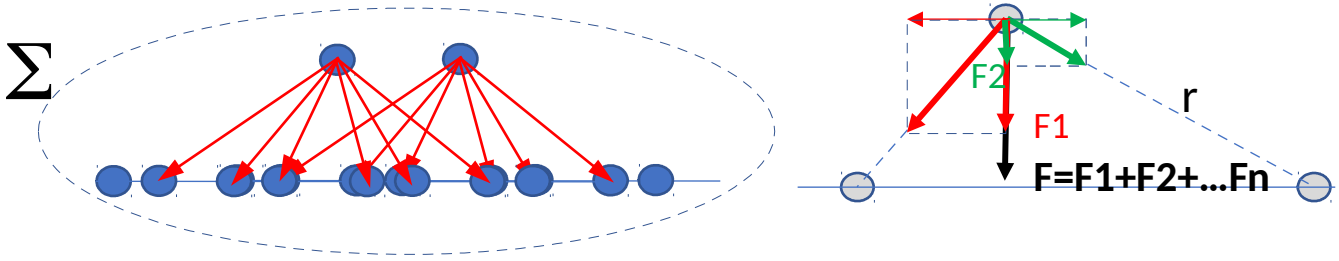
Repeatedly constant cone angles of 140° and 135°



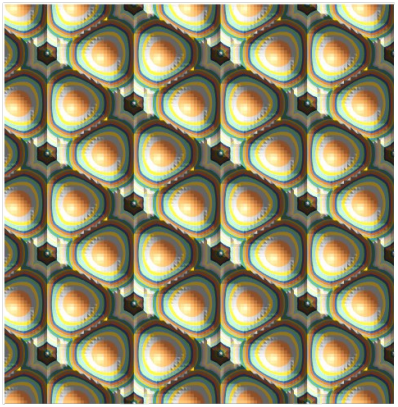
morning glory seed

Simulation methods

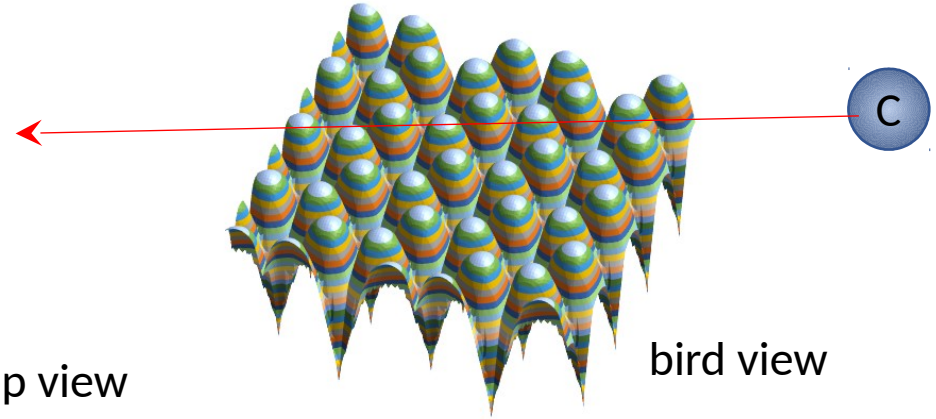
1. **LAMPS simulation with using Kolmogorov and Crispy potential (K-C) [*]**
2. **Summarization of attractive forces F normal to the Graphene plane acting on every carbon atom of the second layer by all atoms of the first Graphene layer (~ $1/r^7$)**



Calculated landscape of normal attractive force acting on carbon atom moving over Graphene



Top view



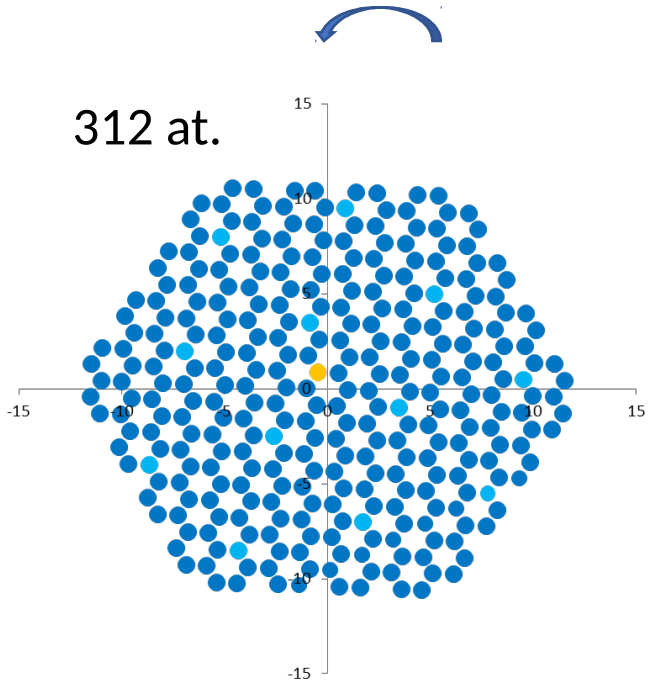
bird view

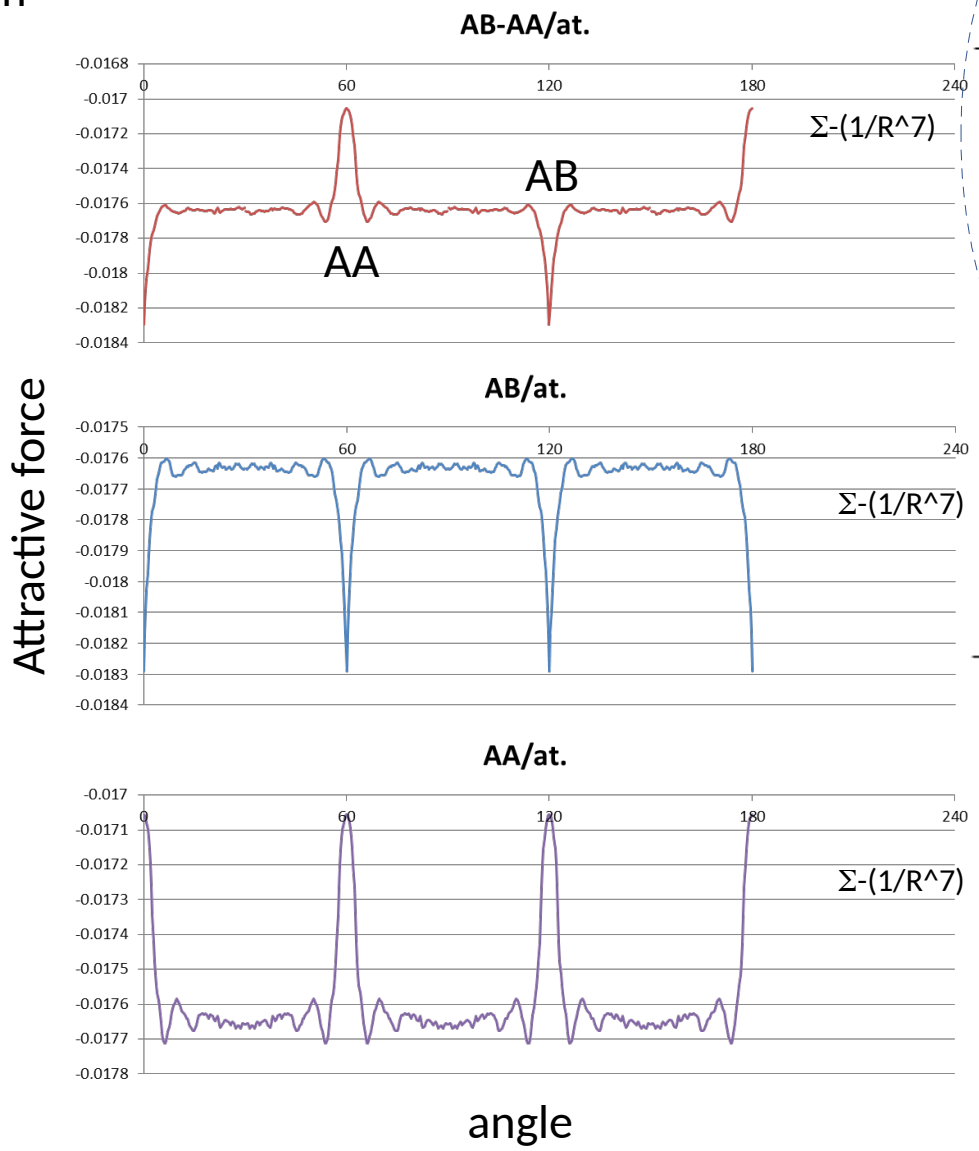
[*] A. N. Kolmogorov, V. H. Crespi, Registry-dependent interlayer potential for graphitic systems. Physical Review B, 71(23):235415, 2005

Position of rotation axis

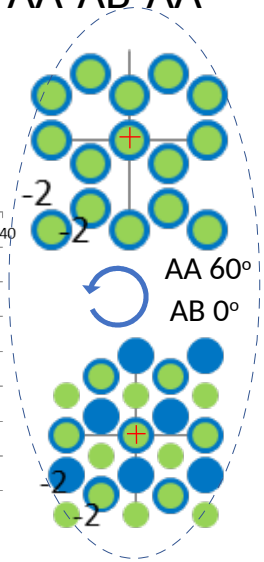
- Three basically different positions of rotation axis
- Stable AB configuration
- Unstable AA stacking

Simulated rotation of group of 312 atoms

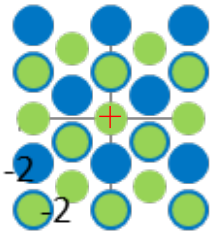




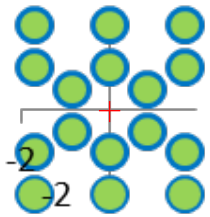
AA-AB-AA



AB-AB



AA-AA

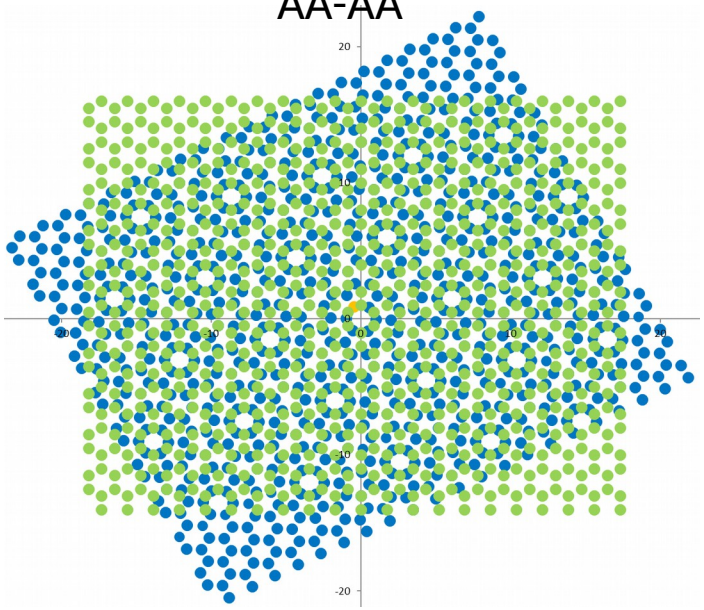


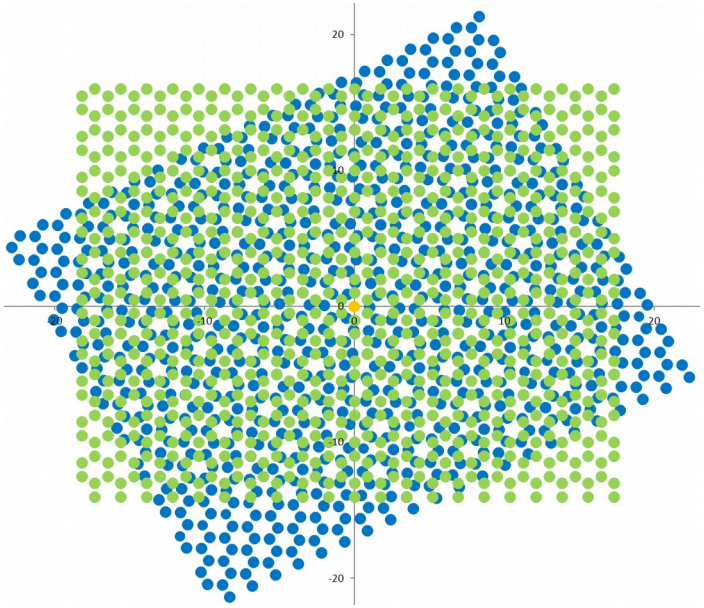
Moire patterns of twisted graphene bilayer

AA-AA

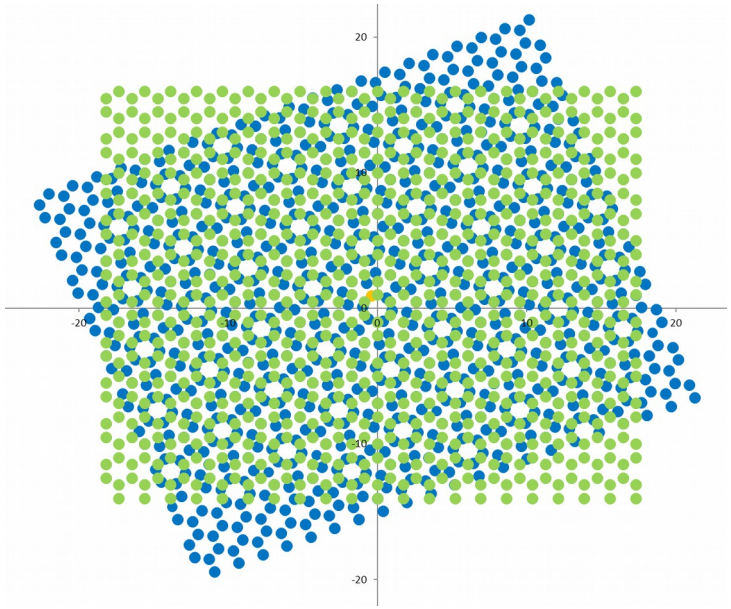
AB-AB

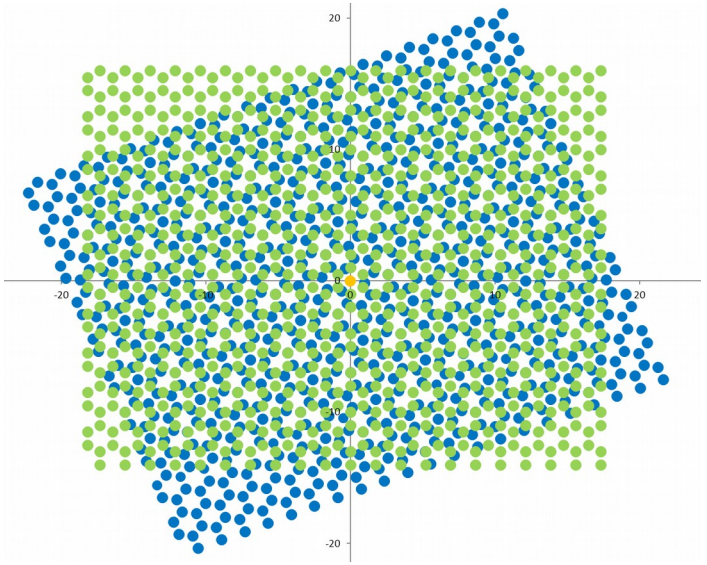
27.8°





21.78°

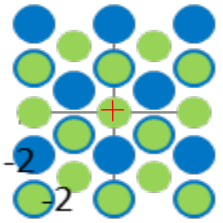


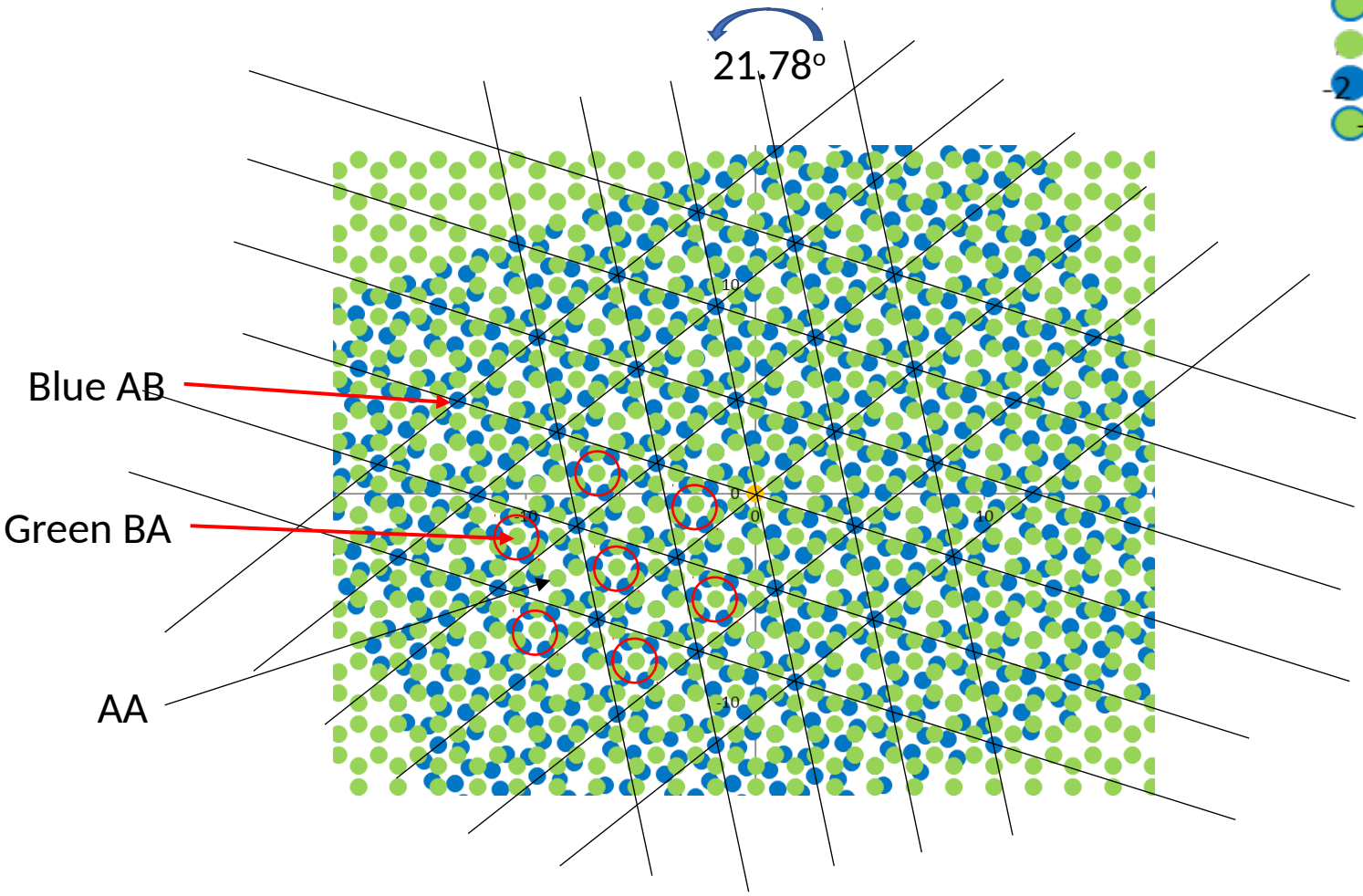


AB-AB rotation at 21.78°

- No distinct impressive Moire pattern visible
- Some atoms are located on stable AB or BA positions
- There is a periodical network of atoms at stable positions

AB-AB rotation axis position

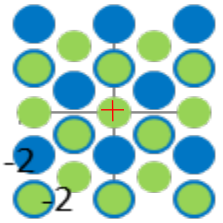


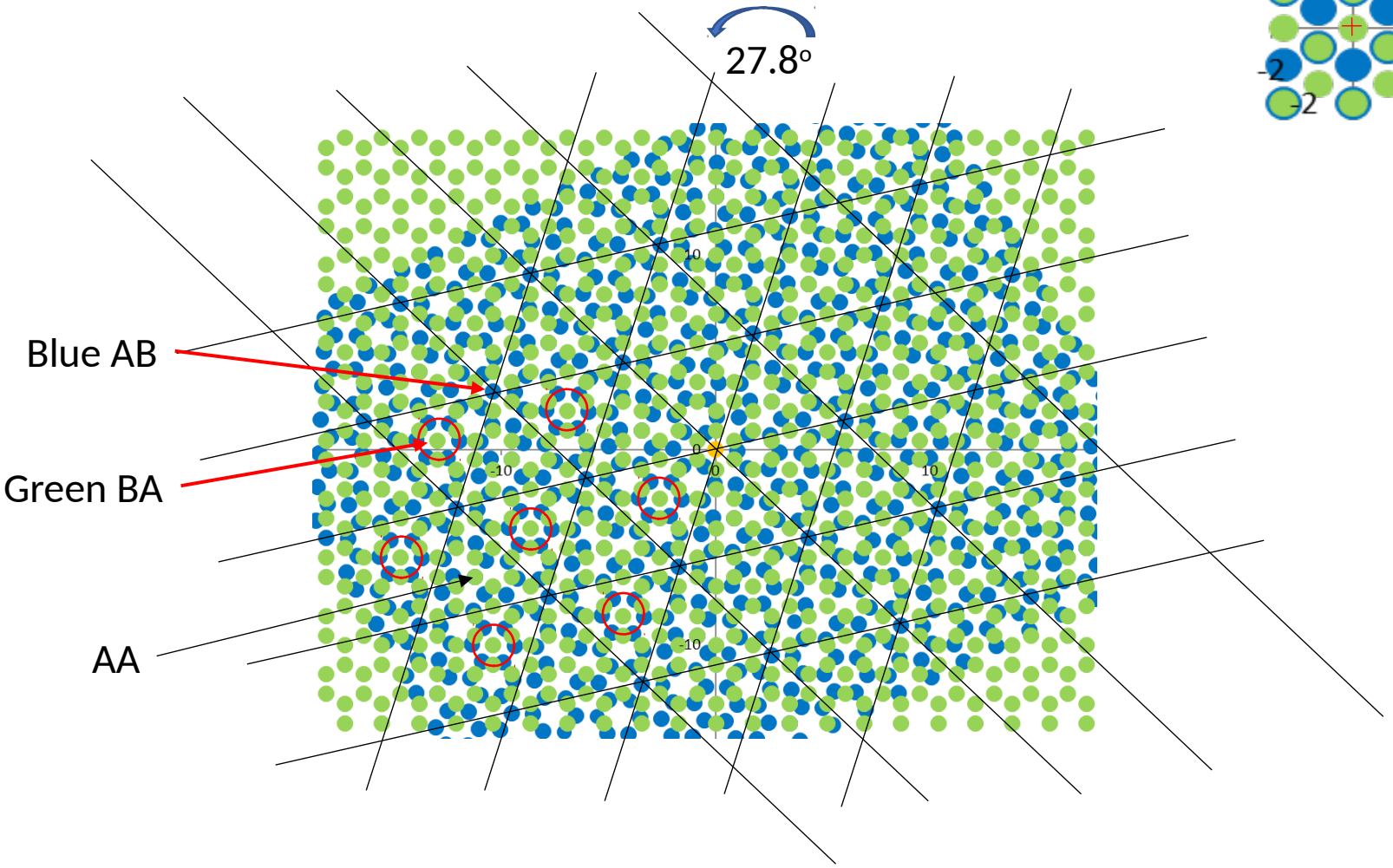


AB-AB rotation at 27.8°

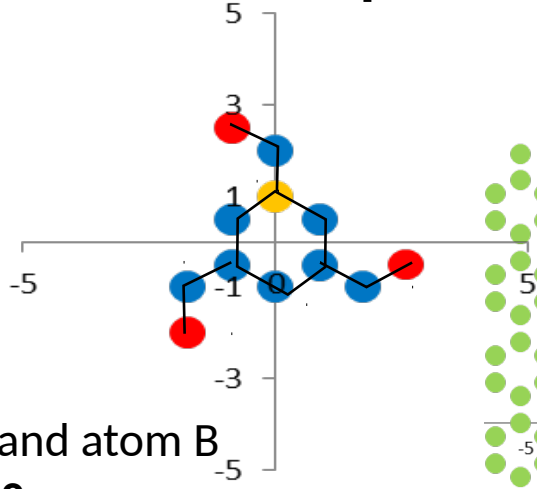
- Geometry very similar as for 21.78°
  - No distinct impressive Moire pattern visible
  - Some atoms are located on stable AB or BA positions
  - There is a periodical network of atoms at stable positions

AB-AB rotation axis position

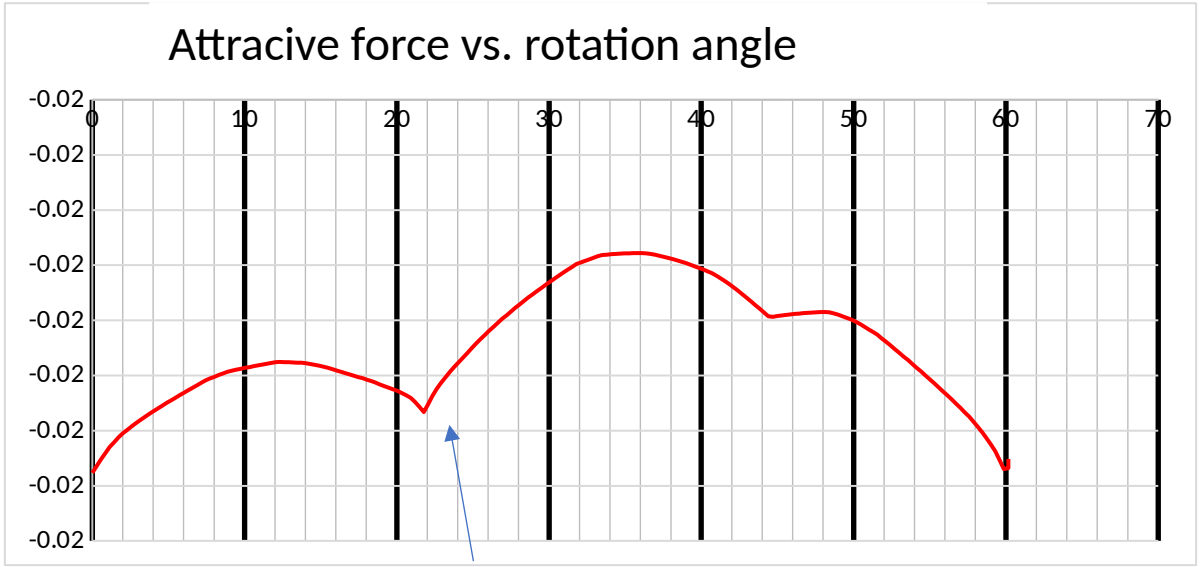




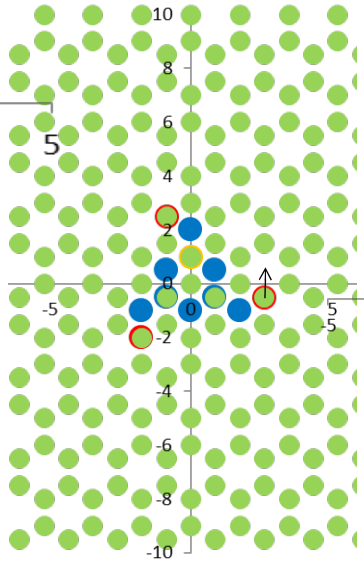
The basic structure with metastable position at 21.78° (AB)



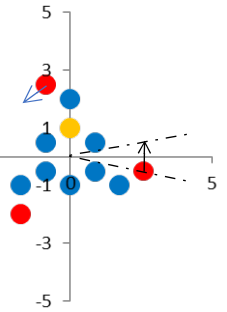
- Initial configuration AB
- Rotation around center of hexagon A and atom B
- „RED” atoms stabilize rotation at **21.78°**.
- There is 120° symmetry of the rotation of such structure

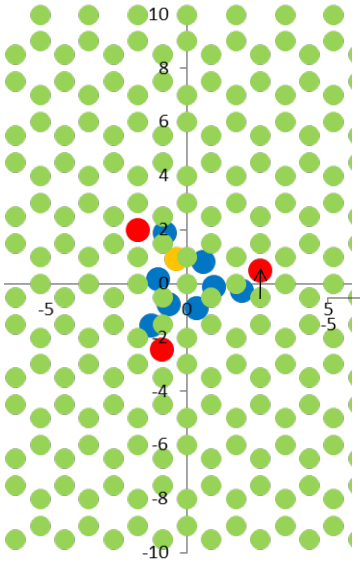


Metastable 21.78°

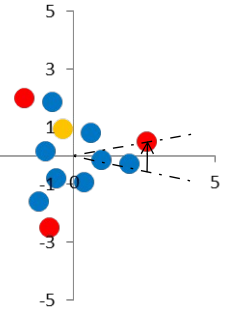


0°



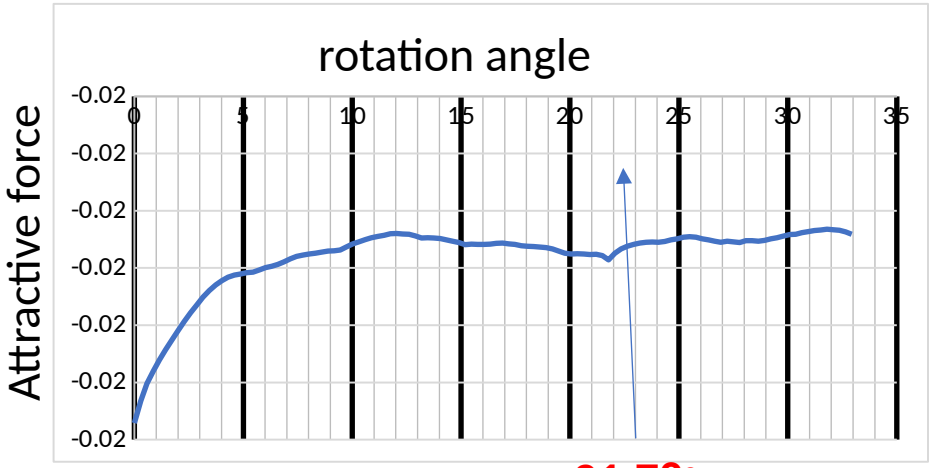


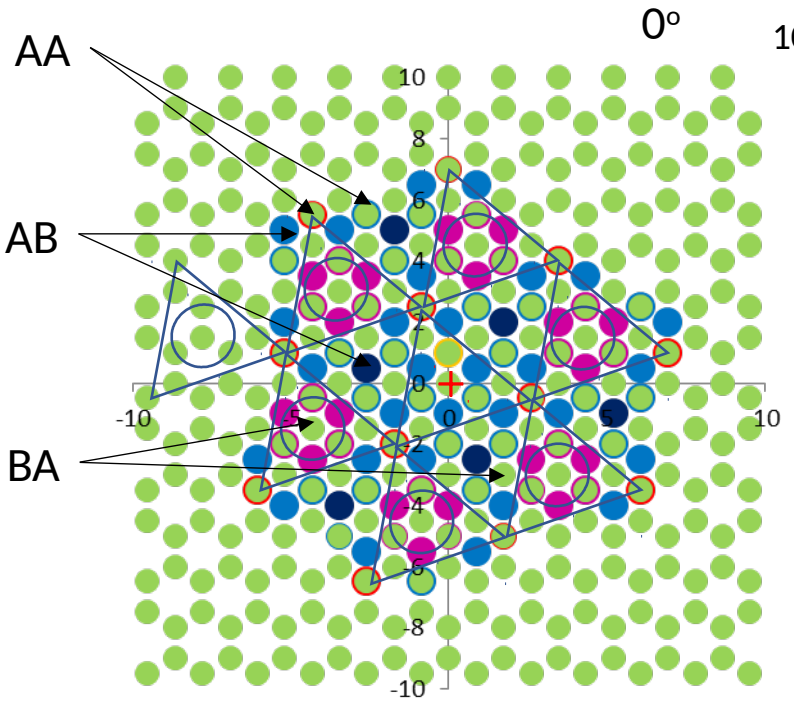
21.78°



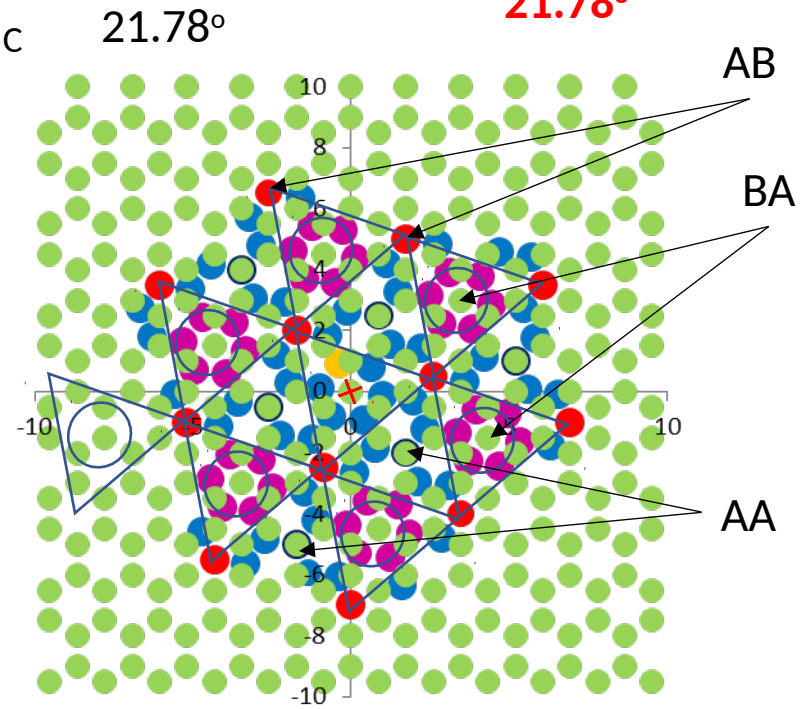
Bigger group of atoms (107 at.) at 21.78° (AB)

- Still metastable at 21.78°
- Diminishing of metastability with island growth



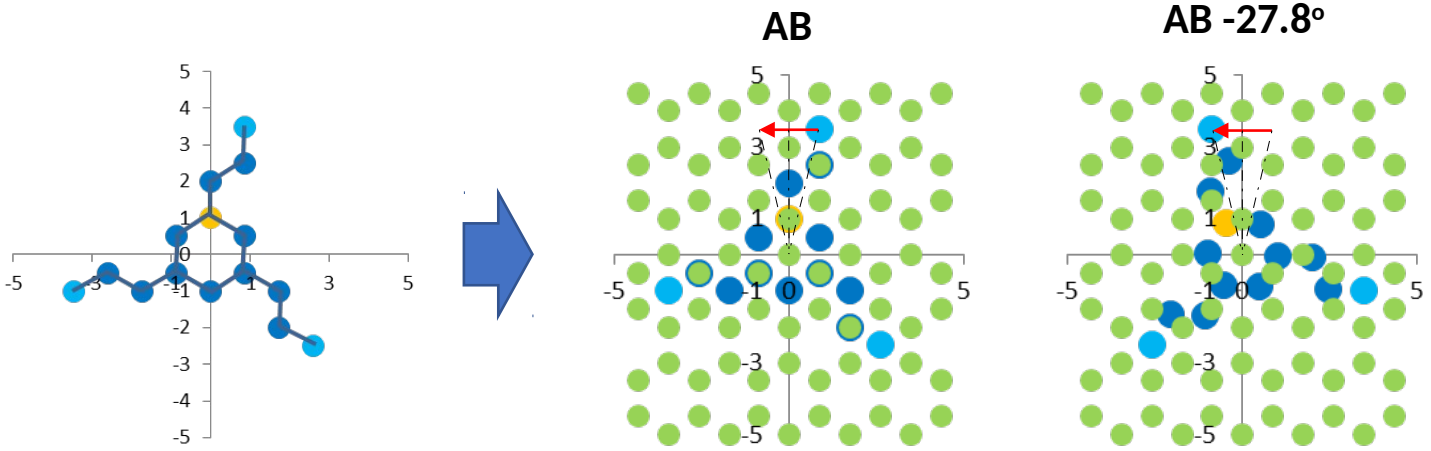


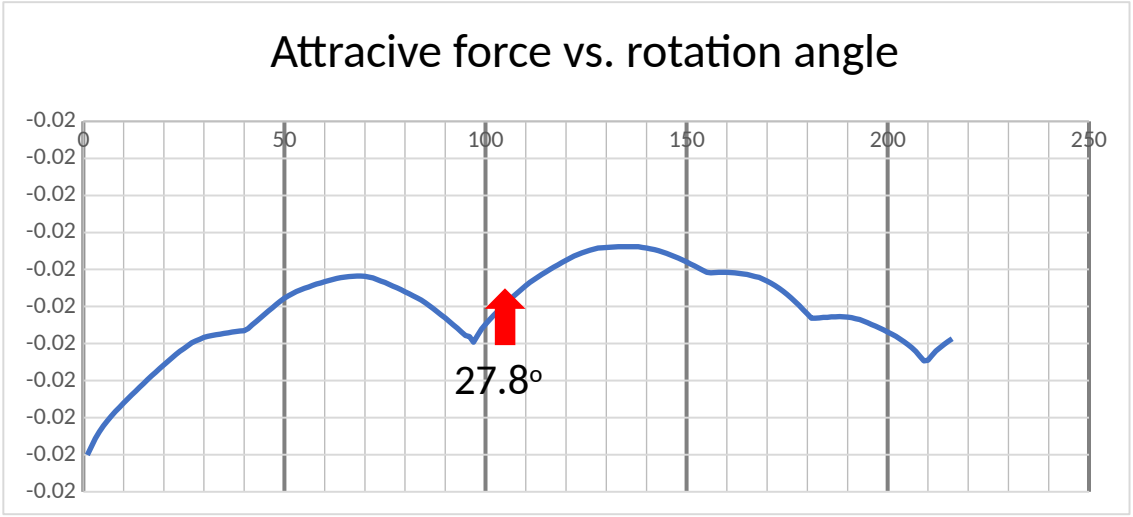
107 atomów C



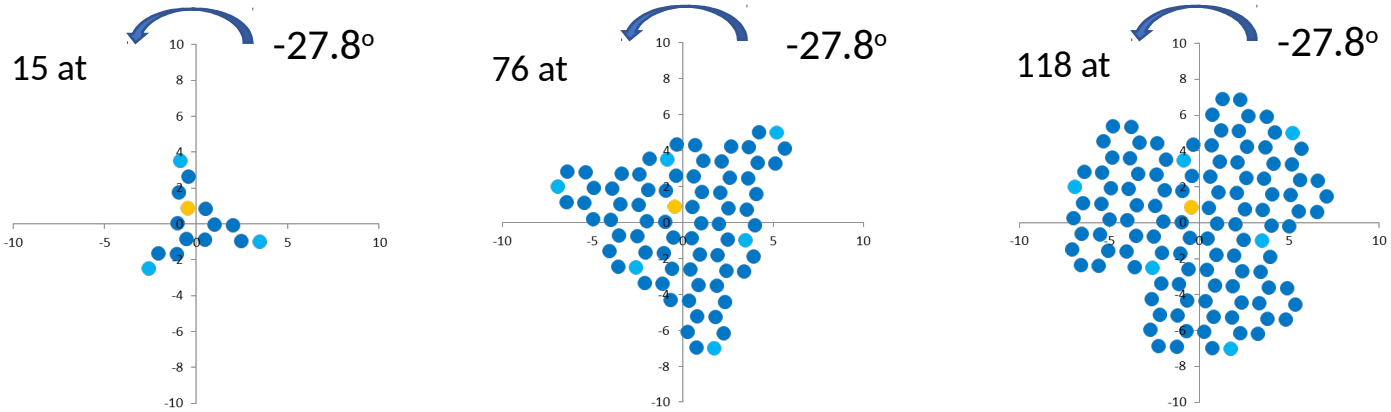
The basic structure with metastable position at 27.8° (AB) 15 atoms

- Initial configuration AB
- Rotation around center of hexagon A and atom B
- There is 120° symmetry of the rotation

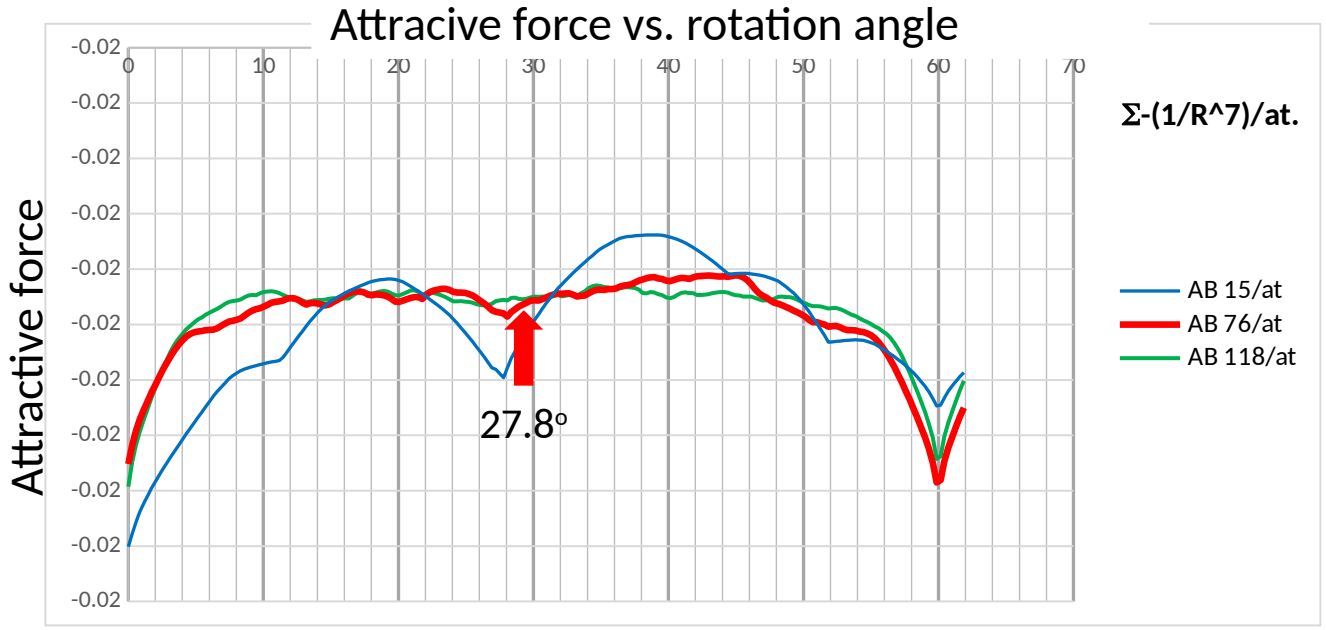




Metastability changes with island growth

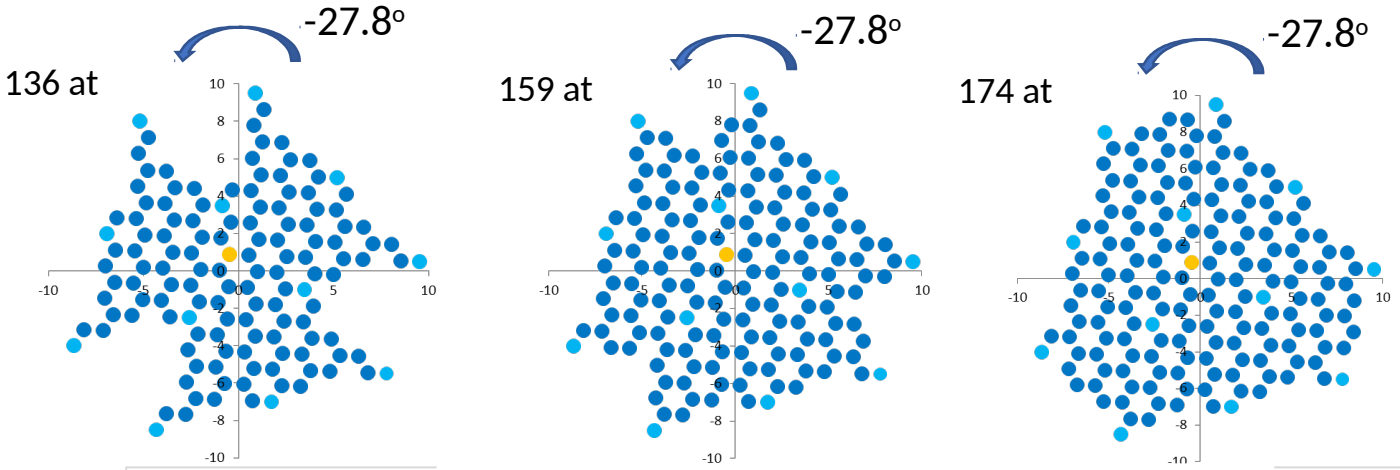


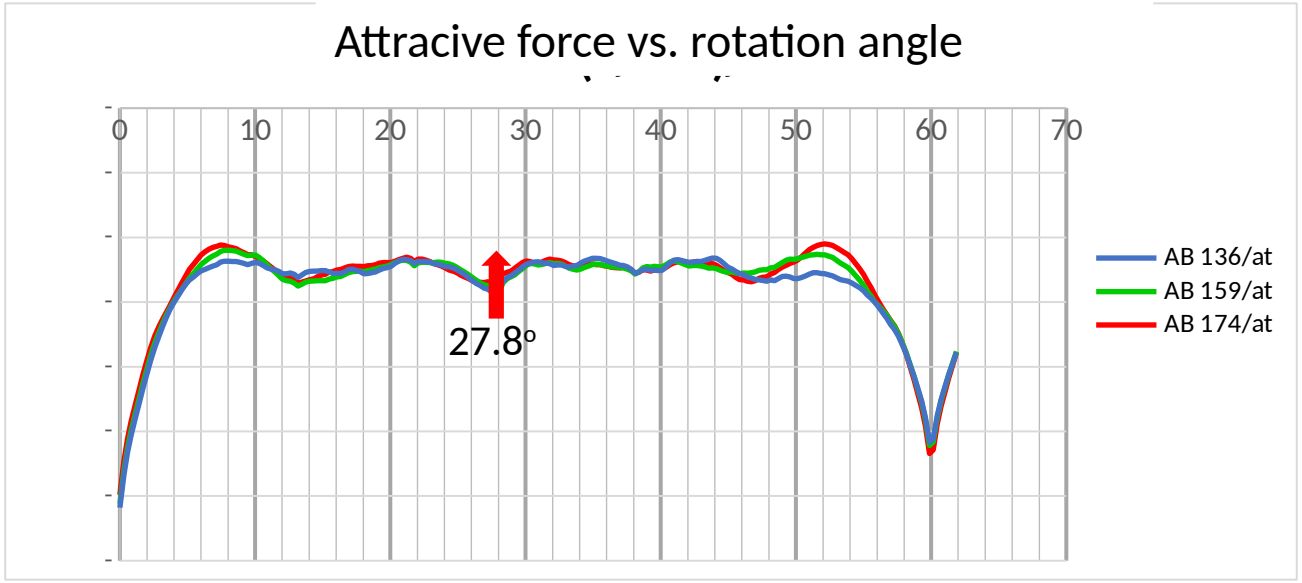
- Growth toward „pale blue” atoms stabilizes 27.8°
- Growth in other direction may diminishes metastability at 27.8° as shown for 118 atoms



Larger structures at 27.8°

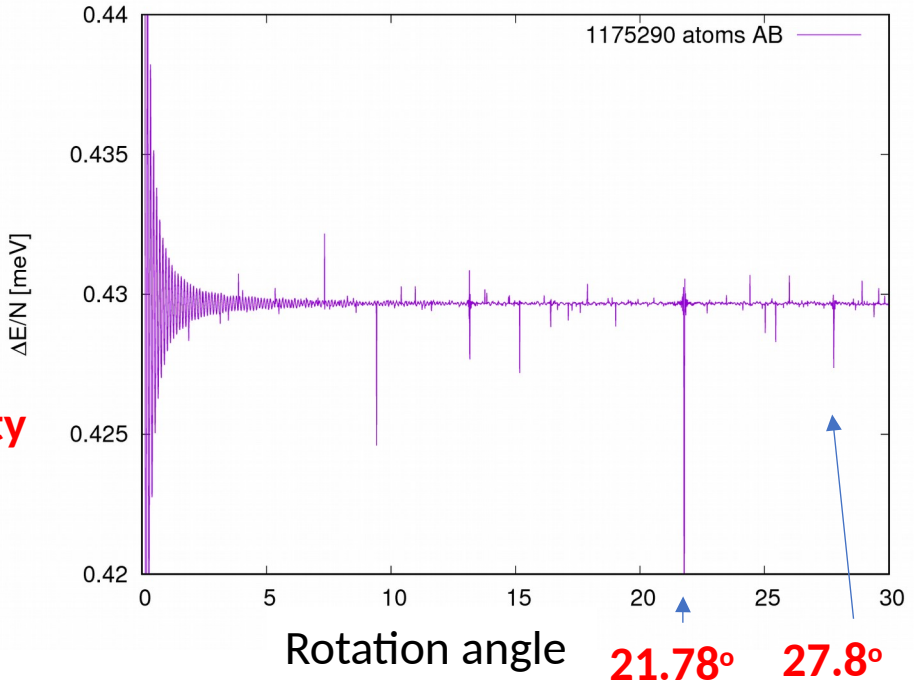
- Metastability may be preserved if „pale blue” atoms (at AB position after rotation) are on the tips of the Island with distinct triple symmetry 120°
- Preferred growth with triple symmetry 120°





LAMPS Simulation for 1 175 290 atoms

Kolmogorov and Crispy potential (K-C) [*] gave results closer to experimental data than Lenard-Jones potential



- **Extremly narrow divergence of stability angles of structures twisted at 21.78° and 27.8°**
- **Possibly less than 0.2°**

Simulated angles of stability compliant with angle of commensurate super-lattice structures by Castro [**]:

$$\cos\theta(m,n)=\frac{3m^2+3mn+n^2/2}{3m^2+3mn+n^2}$$

where (m,n) are pairs of co-prime numbers.
For example (m,n): **(1,1) – 21.787°**, (2,1) – 13.173°, (3,1) – 9.43°, **(2,3) – 27.795°**.

[*] A. N. Kolmogorov, V. H. Crespi, Registry-dependent interlayer potential for graphitic systems. Physical Review B, 71(23):235415, 2005
[**] A. H. Castro Neto J. M. B. Lopes dos Santos, N. M. R. Peres. Continuum model of the twisted graphene bilayer. Phys. Rev. B, 86:155449, 2012

Conclusions

- Growth of twisted Graphene layer starts after reaching some group of carbon atoms specific for given twist angle.
- Some atoms of twisted bilayer stabilize twisted configuration.
- Growth of twisted Graphene may diminish or enhance metastability of twisted configuration depending on growth direction
- Growth of twisted Graphene through preferential attaching atoms which stabilizes twisted configuration enhances metastability.
- The preferential growth of twisted bilayer with triple symmetry was simulated.

Thank you for your attention