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FLOWERED GRAPHENE: GROWTH, CHARGE AND THERMAL TRANSPORT

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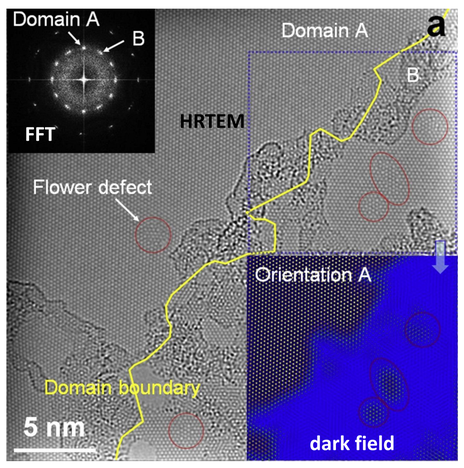
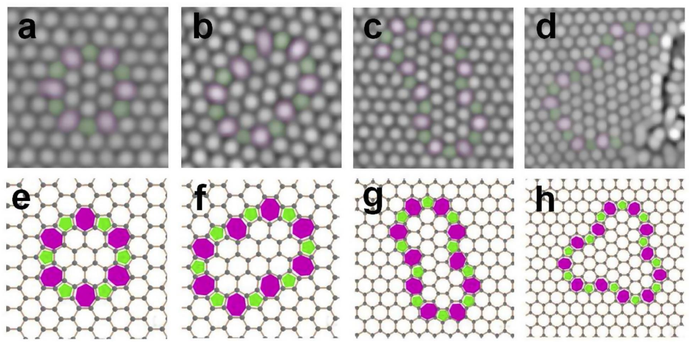
Structural properties and growth mechanism of flower defects

In recrystallized graphene monolayers grown by CVD on platinum, many flower-related defects appear. These flower-related defects are in fact rotated graphene domains as evidenced by a numerical dark-field image.

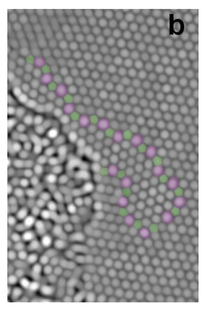




Flower defects are observed together with larger domains. The two main characteristics of these domains are, firstly, a 30° rotation of the inside with respect to the outside graphene matrix, and, secondly, a continuous grain boundary made of alternating pentagon-heptagon 5-7 pairs.

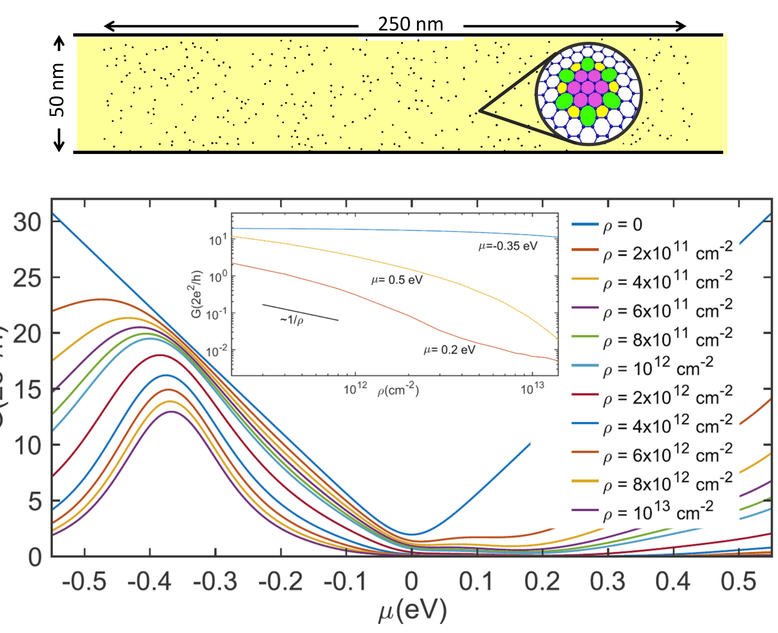
We suggest that the flowers originate from a **bulge nucleation mechanism**, which occurs during the dynamic recrystallization and is driven by the strain provided by the polycrystallinity of the Pt support. It consists of four main stages.

One of the main features of this mechanism is the presence of serrated grain boundaries and *left over* grains once the process has completed stage IV. Due to their dynamic character, stages II and III are rarely observed. The flower domains would be the grains *left over* by the dynamic recrystallization.



Electronic transport in flowered graphene ribbons by Green's function approach

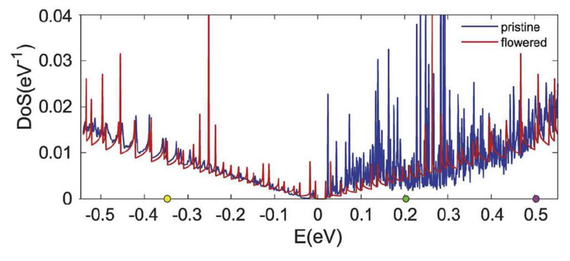
Room-temperature conductance G as a function of chemical potential μ for a 50nm x 250nm graphene ribbon with different flower concentration ρ.



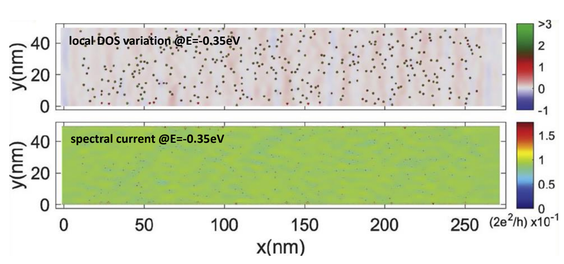
The **strong electron-hole asymmetry** around the Dirac point is due to break of chiral symmetry by odd carbon rings. We identify three different transport regimes:

- **Quasiballistic**: For holes close to the charge neutrality point (-0.4 eV<μ<0), the conductance is scarcely affected, at least for $\rho<10^{12}$ cm^2.
- **Localized**: For electrons close to the charge neutrality point (0<μ<0.2 eV) a transport gap develops and enlarges increasing ρ.
- **Diffusive**: For energies far away from the charge neutrality point (μ<-0.4 eV and μ>0,2 eV), the conductance decreases more moderately and in a fairly electron-hole symmetric way.

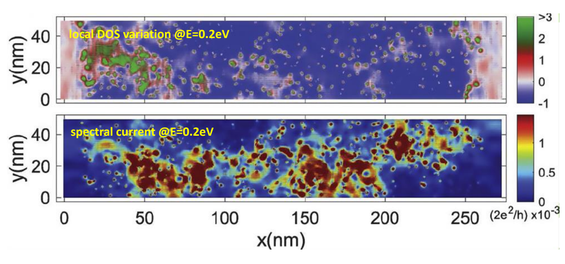
Further physical insight into these results can be gained by looking at the density of states (DoS) and local spectral current distribution as a function of the energy E. We consider $\rho=3x10^{12}$ cm^2.



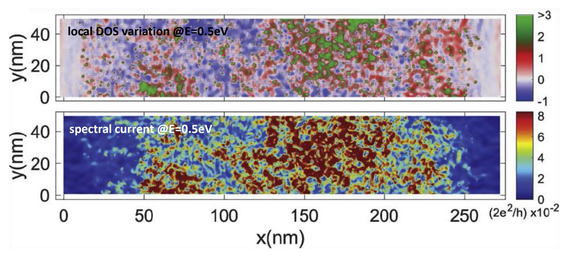
For holes, the density of states is only marginally affected by flowers, thus explaining the hole conductance robustness. For electrons, the density of states is strongly impacted. The spikes indicate the formation of additional states, which explains why the electron conductance is more strongly affected by flowers.



In the quasiballistic regime at E=-0.35 eV, the DoS varies more significantly on the flowers, but very weakly in the regions between them. As a consequence, transport is scarcely affected, as we can see from the fairly homogeneous distribution of the spectral currents.



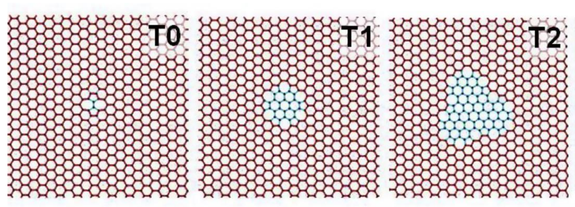
In the localized regime at E=0.2 eV, the DoS is strongly enhanced around the flowers and suppressed elsewhere. Therefore, the electron propagation is made difficult, as seen from the local distribution of spectral currents, where electrons appear to circulate in the high-DoS regions without being able to significantly cross the low-DoS areas.



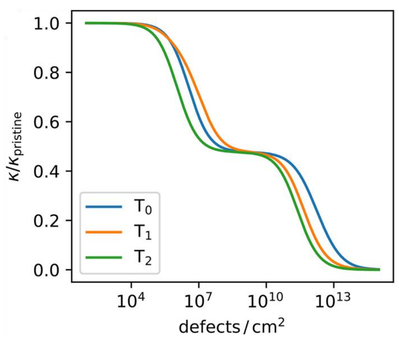
In the diffusive regime at E=0.5 eV, irregular variations of the DoS in the region between the defects are present, thus introducing scattering for electrons. Accordingly, the spectral current is fragmented.

Thermal transport in flowered graphene by Boltzmann-Peierls transport equation for phonons

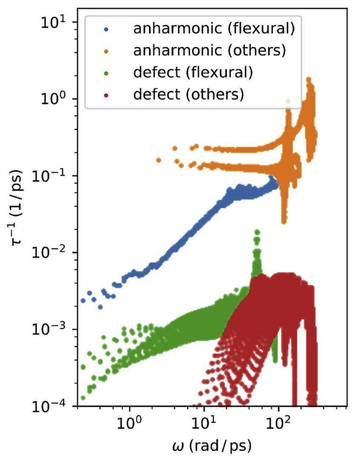
We compare the effects of three different kinds of crystallographic imperfections: Stone-Wales (T0), unitary flower (T1) and triple flower (T2) defects.



Room-temperature thermal conductivity κ of graphene in the presence of T0, T1 and T2 defects.



A salient feature of all curves is the **plateau at intermediate concentrations**, which interrupts the otherwise monotonic decrease of κ with increasing defect concentration. This turns out to be a manifestation of the peculiar physics of phonons in graphene and the singular importance of the flexural branch for transport.



The intrinsic scattering rates for the flexural branch are smaller than those of the other branches. This results from the branch quadratic dispersion and the symmetry-induced selection rule for three-phonon scattering processes. The elastic scattering rates for the flexural branch are higher and decay much more slowly when approaching zero-frequency. The presence of an interval of concentrations where elastic scattering is strong enough to drastically suppress the thermal transport by the low-frequency region of the flexural branch, but not enough to be relevant for all other branches, is the reason for the thermal conductivity plateaus.

Conclusions

The study of the structural properties of flower defects by HRTEM and *ab initio* simulations allowed us to propose a **new mechanism** for their growth. This represents a first step toward the experimental **control of flower density and position** by controlling the bulge nucleation mechanism during the recrystallization process.

The simulation of electron transport revealed a large transmission coefficient for holes and the development of a **transport gap** for electrons. This **asymmetry** originates from the odd-numbered carbon rings, which break the sublattice symmetry of graphene. Quasi-ballistic, diffusive and localized transport regimes are observed.

We observed a **strong reduction in room-temperature thermal conductivity** due to the presence of flower defects. The concentration required to drastically impede heat transport by flexural phonons is rather modest, around 10^{10}cm^{-2}. Higher concentrations affect all phonon branches and reduce the thermal conductivity by up to two orders of magnitude.

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REFERENCES

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