8th RES Users' Conference 2014

Longitudinal and Transverse Electronic Transport in Atomically Doped Graphene – Towards the Quantum Hall Effect

Nicolas Leconte and Stephan Roche

September 23, 2014

Motivation

Push the limits of computational predictions further and further, to explore new physics

- Existing real space implementation of the Kubo-Greenwood formalism : longitudinal conductivity[1].
  - Scales linearly with system size
  - Desktop computer is sufficiently powerful
  - Parallel. on Tier1/2-type infrastructure (OMP, MPI,GPU[2])
- We developed a new expression for the transverse conductivity.
  - Scales linearly with system size.
  - However, Tier0 infrastructure is required (using MPI)
  - Large number of files have to be stored.

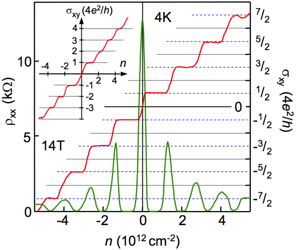
[1] H. Ishii, F. Triozon, N. Koboyashi, K. Hirose, and S. Roche, C.-R. Physique **10**, 283 (2009)

[2] Z. Fan *et al.*, Comput. Phys. Comm. **185**, 1 (2014)

Motivation

New Physics? . . . Quantum Hall Effect (QHE)!

- Specific impact of disorder on the QHE
- Lift degeneracies in the Landau Level spectrum
- Study the QHE in realistic samples (oxygenated graphene, polycrystalline graphene, hydrogenated graphene, Hofstadter spectrum,. . .)



Outline

Bottom-Up Approach

DFT

Ab initio calculations to describe the local potential induced by the impurity

⇓

DFT → TB

Extract sufficient TB parameters to reproduce the local potential

⇓

TB-parametrized Kubo Formalism

Allows us to simulate mesoscopic-sized systems (10^6 atoms) :

- comparison with experiment
- calculate transport properties to visualize quantum effects

Bottom-Up Approach

DFT

Ab initio calculations to describe the local potential induced by the impurity

$\Downarrow$

DFT $\rightarrow$ TB

Extract sufficient TB parameters to reproduce the local potential

$\Downarrow$

TB-parametrized Kubo Formalism

Allows us to simulate mesoscopic-sized systems (10^6 atoms) :

- comparison with experiment
- calculate transport properties to visualize quantum effects

Bottom-Up Approach

DFT

Ab initio calculations to describe the local potential induced by the impurity

⇓

DFT → TB

Extract sufficient TB parameters to reproduce the local potential

⇓

TB-parametrized Kubo Formalism

Allows us to simulate mesoscopic-sized systems (10^6 atoms) :

- comparison with experiment
- calculate transport properties to visualize quantum effects

Longitudinal conductivity

Kubo conductivity

$$\sigma_{DC} = \frac{1}{2} e^2 \rho(E_F) \lim_{t \to \infty} \frac{\partial}{\partial t} \Delta X^2(E_F, t)$$

Wave packet : mean quadratic displacement

$$\Delta X^2(E, t) = \frac{Tr\left[[\widehat{X}, \widehat{U}(t)]^\dagger \delta(E - \widehat{H}) [\widehat{X}, \widehat{U}(t)] \right]}{Tr\left[\delta(E - \widehat{H}) \right]}$$

Diffusion coefficient

$$D_x(t) = \frac{\Delta X^2(t)}{t}$$

Longitudinal conductivity

$$\left\langle \psi_1 \middle| \delta(E - \tilde{\hat{H}}) \middle| \psi_1 \right\rangle = \lim_{\eta \to 0} -\frac{1}{\pi} \mathrm{Im} \left(\left\langle \psi_1 \middle| \frac{1}{E + i\eta - \tilde{\hat{H}}} \middle| \psi_1 \right\rangle \right)$$
$$= \lim_{\eta \to 0} -\frac{1}{\pi} \mathrm{Im} \left(\kappa_1 \right)$$

where κ_1 is calculated using a continued fraction:

$$\kappa_1 = \cfrac{1}{E + i\eta - a_1 + \cfrac{b_1^2}{\ddots \cdots \cfrac{}{E + i\eta + a_{N-1} - \cfrac{b_{N-1}^2}{E + i\eta - a_N - b_N^2 \times \mathrm{Term}}}}}$$

Transverse conductivity

$$\sigma_{xy}(t') = \frac{2N_s}{V}\int_0^{t'} dt \int_{-\infty}^{\infty} dE f(E-\mu)$$

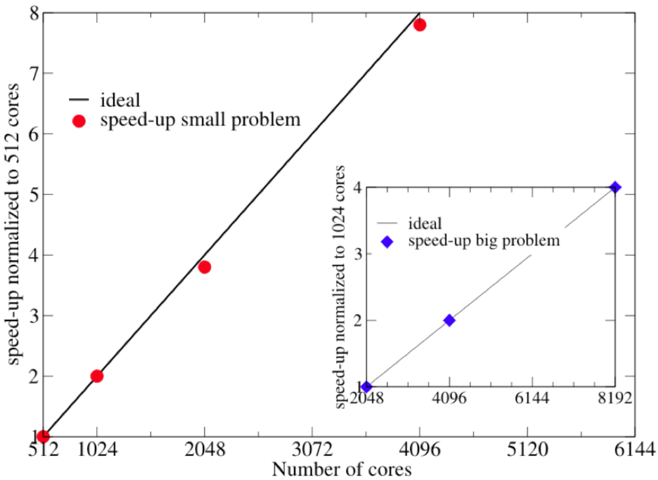
$$\lim_{\eta\to 0^+}\sum_{j=1}^{N_{\text{recurs}}} \text{Re}\left[\left\langle \Psi_1 \middle| \delta(E-H_0) \middle| \Psi_j \right\rangle \left\langle \Psi_j \middle| j_y \frac{1}{E-H_0+i\eta} j_x(t) \middle| \Psi_1 \right\rangle\right]$$

- (approximataly) resolved identity allows to separate the complicated product of two inverses of (sparse) matrices in two simpler factors
- These factors can be calculated with Lanczos recursion techniques allowing for linear scaling with Hamiltonian size
- However, this identity is the reason for the requirement of Tier0 infrastructure. . .
- For each time step, each j dependent term is done in parallel on each core before reducing everything on the masternode
- Very efficient parallelization: communications only take a few seconds at beginning and end of each serial $j-$run

Transverse conductivity

- Evolving and storing each Ψ_j ($\sim$ 100 MB)
- Up to 800 to 8000 recursion steps
- Example: 12 million atoms, 4000 recursion steps. We can do about 25 time steps in 24 hours (or 100K CPU-hours). For certain physics, 500 time steps are required (2M CPU-hours)
- Linear scaling with system size
- Quadratic scaling with number of recursion steps
- PRACE - 6th Call: $\sim$ 14M hours; 8th Call: $\sim$ 22M hours
- During 6th Call: further code optimization for use on Curie cluster (CEA), provided by PRACE technician

Prace Scaling



Prace Scaling

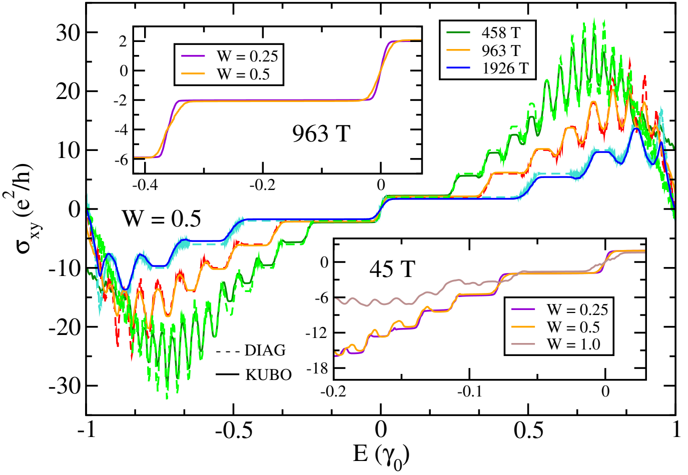
Table 1 : Scaling for a relatively small problem (720 000 sites, up to 4096 cores).

# cores	absolute timing (s)	speedup
512	1360	1
1024	689.7	2
2048	362.5	3.8
4096	174.5	7.8

Table 2 : Scaling for large system (2 000 000 sites, up to 8192 cores)

# cores	absolute timing (s)	speedup
2048	3799	1
4096	1879	2
8192	947.5	4

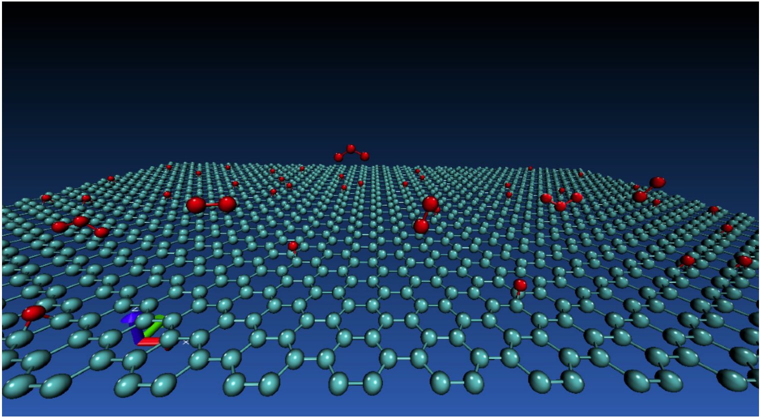
Transverse conductivity: validity of the method



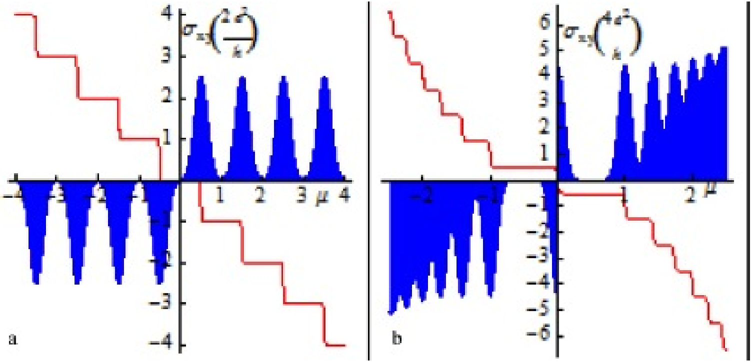
Outline

1. Motivation
2. Methodology
3. Results
4. Conclusions

Ozone Treated Graphene



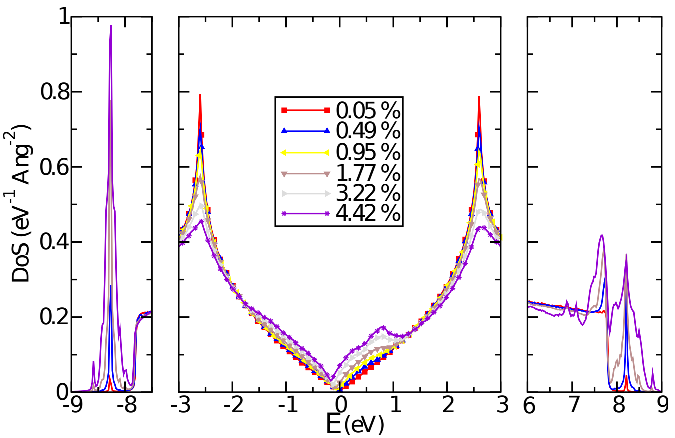
Landau levels : 2DEG *versus* Graphene



Graphene has zero energy states[3]

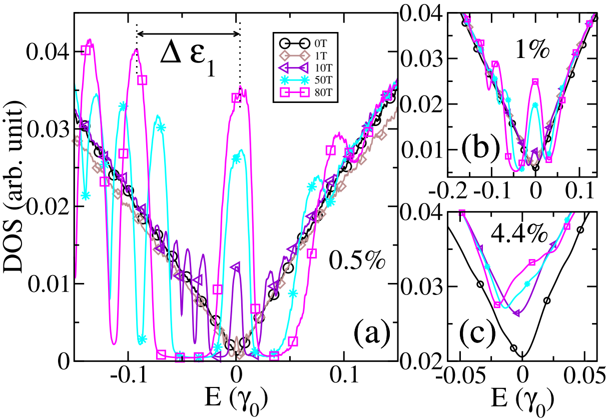
[3]Figure taken from Barlas *et al.*, Nanotechnology **23**, 5 (2012)

Density of States: without magnetic field



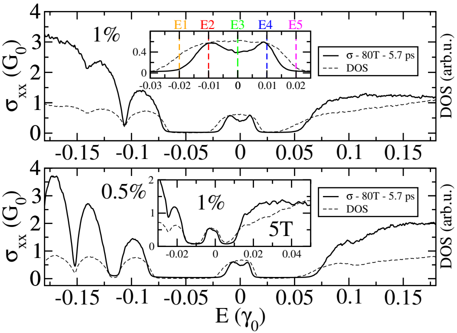
Effect of disorder more pronounced on electron side

Density of States: with magnetic field



Asymmetric Landau spectrum, increasing disorder destroys the Landau quantization

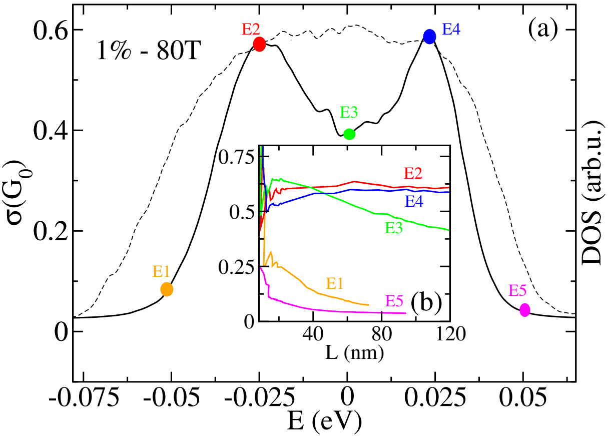
Longitudinal Conductivity



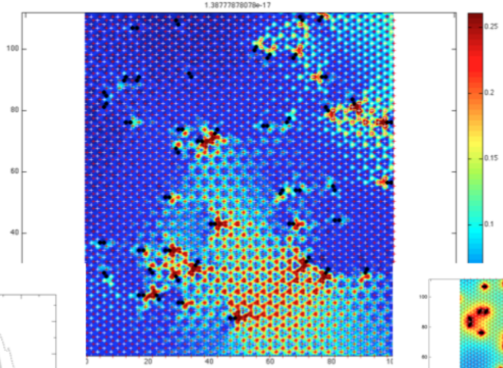
- Clear observation of mobility edges
- Splitting of Zero Landau Level (LL0)

N. Leconte, F. Ortmann, A. Cresti, J.-C. Charlier, and S. Roche, 2D Materials **1**, 021001 (2014)

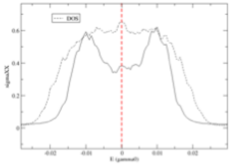
Longitudinal Conductivity

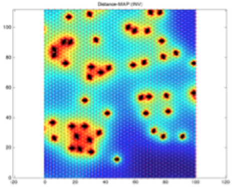


Real space projection of new states

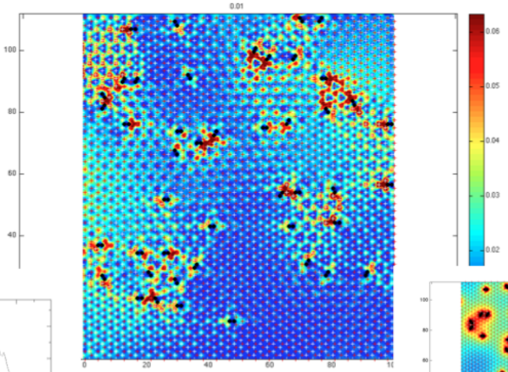


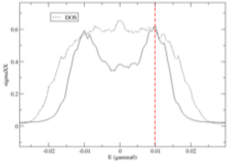
$$W_i = \sum_{j}^{N_{\mathrm{IMP}}} \frac{1}{d_{ij}}$$

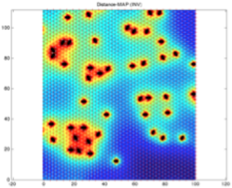




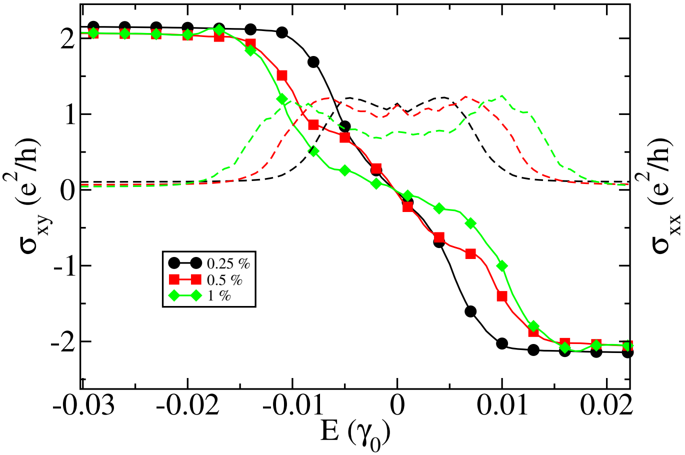
Real space projection of new states



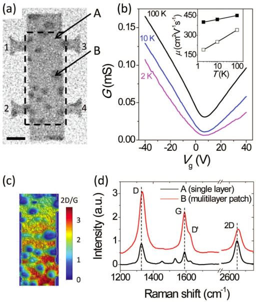


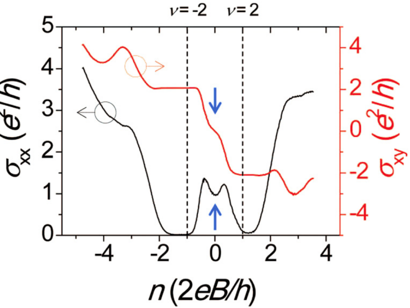


Confirmation of zero energy Hall plateau: σ_{XY}



Experimental Confirmation?



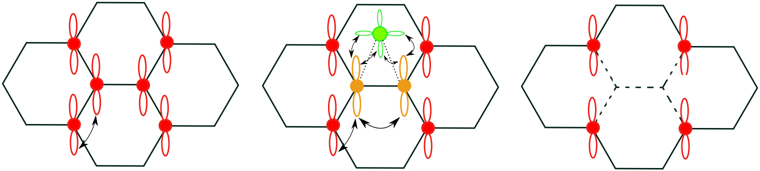


- Multilayer islands creating magnetic bound states?[4]
- Role of hydrogen?
- Evolution of splitting with magnetic field?
- Better characterization...

[4] Youngwoo Nam *et al.*, APL **103**, 233110 (2013)

Conclusions

- Without PRACE infrastructure, no transverse conductivity calculations. . .
- When you manage a large amount of hours, be flexible. . .



- Simplified model
- Similarity with oxygen, no electron-hole symmetry
- Computationnaly simpler
- Re-adjust initial guesses for allocation time

Acknowledgments + Questions

- Thank you for the attention
- Thanks to PRACE
- Thanks to collaborators : F. Ortmann, A. Cresti, J.-C. Charlier, and S. Roche,
- Questions?

N. Leconte, J. Moser, P. Ordejon, H.H. Tao, A. Lherbier, A. Bachtold, F. Alsina, C.M. Sotomayor Torres, J.-C. Charlier, and S. Roche
Damaging Graphene with Ozone Treatment: A Chemically Tunable Metal-Insulator Transition.
ACS Nano, **4** (7), 4033–4038 (2010).

N. Leconte, A. Lherbier, F. Varchon, P. Ordejon, S. Roche, and J.-C. Charlier
Quantum Transport in Chemically-modified Two-Dimensional Graphene: From Minimal Conductivity to Anderson Localization
Phys. Rev. B, **84**, 235420 (2011). *Editor's Suggestion*

N. Leconte, F. Ortmann, A. Cresti, J.-C. Charlier, and S. Roche
Quantum transport in chemically functionalized graphene at high magnetic field: Defect-Induced Critical States and Breakdown of Electron-Hole Symmetry
2D Materials, **1**, 021001

F. Ortmann, N. Leconte, S. Roche
Methodology paper on transverse conductivity implementation
Under preparation

N. Leconte, F. Ortmann, A. Cresti, S. Roche
Impurity engineered Landau Levels
Under preparation