# Report on the CECAM/Psi-k Workshop
# "Quantum Transport in Molecular Nanostructures"
# Trinity College, Dublin, Ireland
# May 22-25, 2012

## Organizers:

•Kristian Thygesen, Technical University, Lyngby, Denmark
•Stefano Sanvito, Trinity College, Dublin, Ireland
•Stefan Kurth, Univ. of the Basque Country UPV/EHU, San Sebastian, Spain, and IKERBASQUE, Basque Foundation for Science, Bilbao, Spain

### Summary of the workshop

The purpose of this workshop was to bring together experts working in the field of electron transport through single molecules both experimentally and theoretically. During the last decade, advances in various experimental techniques made it possible to probe transport properties at the level of single molecules. At the same time, a variety of theoretical methods have been developed to describe quantum transport at the molecular level. We have tried to find a balance in the workshop program to present the different areas of activity in the field, ranging from electronic charge and spin transport (both in the steady state and in the time domain), to heat transport through single molecules.
Although predominantly a theoretical workshop, we have had a significant number of presentations given by experimentalists (about 25% of the total). On the theoretical side, the methods presented to treat the transport problem ranged from an atomistic description using density functional theory (DFT) combined with the Landauer formalism or many-body methods such as GW to model studies focusing on various aspects of the transport problem.

### Scientific content and discussions at the workshop

The first experiments on transport through single molecules have been reported already quite some years ago. Since then a tremendous amount of progress has been made. In fact, initially there was some disappointment due to the difficulties in reproducing the experimental results on the same system by different techniques or groups. At the workshop it became clear that this issue now seems resolved and experimentalists can now reproduce each other's results.
Experimental studies have been presented using techniques such as mechanically controlled break junctions to transport spectroscopies using scanning tunneling and atomic force microscopes. Particularly impressive were the results on STM/AFM transport measurements with submolecular spatial resolution. Other investigations focused on the dependence of the linear conductance on molecular length or on variations in chemical composition as well as the effect of different anchoring groups. In several talks measurements or calculations of inelastic electron tunneling spectra where reported.
A recurring theme throughout the workshop, both on the experimental and the theoretical side, were the signature of quantum interference in quantum transport which show up as antiresonances in the transmission function or conductance. Quantum interference was found to crucially depend on the detailed topology of the molecule: while for linearly conjugated molecules the conductance tends to be rather high, for cross-conjugated molecules it tends to be rather low. Another interesting feature is that the antiresonances can split when interaction is included in the theoretical description.
Another issue which was discussed in several talks was the question of image charges which cannot be

described with local DFT approaches but are properly captured with the GW method. Variations on the theme of spin in transport ranged from the simulation of spin-resolved STM images of non-magnetic molecules on ferromagnetic surfaces to inelastic spin transitions, the Kondo effect and spin quadrupole accumulation.
On the fundamental side, the question of the applicability of the Landauer formalism using DFT has been addressed and how one can possibly go beyond that formalism by including dynamical exchange-correlation corrections. Finally, several speakers talked about time-dependent approaches to transport (time-dependent DFT and fully time-dependent many-body simulations based on the Kadanoff-Baym equations) which focus not only on the steady state but also on the transient regime or transport under AC bias.

During the workshop several issues were raised and discussed suggesting that further investigations are required. For instance, it became clear that there is a need for a better understanding of the gold-thiol contacts serving as the most commonly used anchoring groups in single-molecule transport measurements.
On the topic of quantum interference, which is apparently observed in experiments at room temperature, the effects of both electron-electron and electron-phonon interaction require further study.
On the methodological side , the GW approach to transport seems to be a promising route towards a quantitatively more accurate calculations on molecular junctions, at least for weakly correlated systems. In particular, GW seems to capture naturally the image charge effects. However, so far GW calculations have only been performed on relatively small systems and clearly the method needs to be developed further. The big question is, if it will eventually be possible to make GW scalable such that it can also be applied to bigger and potentially more interesting systems.
As a numerical methodology, the description of transport with the DFT+Landauer approach is certainly more developed. However, existing (semi-)local approximations are not capable to describe certain physical aspects such as image charge effects showing a need for better functionals. Moreover, although by now it is known that DFT+Landauer can even give the exact conductance under certain (limited) conditions, it also has been understood that even in the steady state, the DFT+Landauer approach in general needs to be augmented by dynamical exchange-correlation corrections of time-dependent DFT. For simple models these corrections can even be quantified but a practical way to express them, e.g., in terms of the density, is still lacking. Although maybe not surprisingly, it was interesting to see that the differences between the DFT and many-body steady-state approaches very much carry over to the time domain (TDDFT versus GW in the framework of the Kadanoff-Baym equations).
Many of these latter studies are done on simple model systems, typically described by lattice Hamiltonians, e.g., Anderson and Hubbard-like models. Of course, these models have extensively been studied in the community of people working on mesoscopic transport, I.e., transport through quantum dots. In fact, at the workshop also some speakers of this community were present (one of them even claimed “compared to quantum dots, molecules are simple!”) and it is quite clear that the theoretical description of transport through single molecules can benefit from the experience accumulated in the “mesoscopic” community through further interaction in the future.

**Assessment of the results and impact of the workshop**

In our opinion, the really strong point of the workshop was reflected by the breadth of the topics in the field of molecular transport covered in the talks. We not only had a significant number of talks presenting experimental results using various techniques (break junctions, STM/AFM, etc.) but also talks on a variety of theoretical approaches (GW, DFT+Landauer, spin transport, time-dependent with TDDFT or Kadanoff-Baym) addressing quite diverse physical questions (image charges, quantum

interference, correlation effects in transport, etc). We have received many positive feedback reactions from the participants on the form and the scope of the workshop. Participants were particularly positive about the breadth of the topics covered during the workshop. It was generally agreed upon the usefulness of this kind of workshop and further editions in the future will certainly be appreciated by the community of people working in the field of molecular transport.
The workshop in Dublin also served as a kick-off meeting for the newly established Psi-k working group “Quantum transport in nanostructures” which will, among other things, coordinate workshop proposals on the topic for future CECAM/Psi-k workshops. At this stage, there is already a CECAM workshop approved on transport through single molecules to take place in March 2013 in Bremen. However, at the meeting in Dublin there where already people interested in organizing a follow-up workshop beyond the one in Bremen.

**Annex**

- Program of the workshop
- List of participants

# Quantum transport in molecular nanostructures

May 22, 2012 to May 25, 2012

Location : Trinity College Dublin, Ireland

Details Participants Program Posters Abstracts Report

## Day 1 - May, 22nd 2012

Welcome

- 14:00 to 14:10 - Welcome

Electron transport through single molecules (Chair: Kristian Thygesen)

- 14:10 to 14:45 - **Latha Venkataraman**
  Electronics of Single Molecule Circuits
- 14:45 to 15:20 - **Nicolas Agrait**
  Unambiguous One-Molecule Transport Measurements under Ambient Conditions
- 15:20 to 15:55 - **Jascha Repp**
  Scanning Probe Spectroscopy of Individual Molecules on Thin Insulating Films
- 15:55 to 16:30 - **Fabian Pauly**
  Thermoelectric properties of molecular and metallic nanocontacts

Coffee break

- 16:30 to 17:00 - Coffee Break

Environmental and non-adiabatic effects (Chair: Colin Lambert)

- 17:00 to 17:35 - **Ivan Rungger**
  Transport properties of solvents and molecules in solution
- 17:35 to 18:10 - **Hannu Häkkinen**
  The gold-sulphur nano-interface: Self-assembled thiolate monolayers, thiolate-protected clusters and molecular junctions
- 18:10 to 18:45 - **Nicola Marzari**
  First-principles engineering of electrical and thermal transport in nanomaterials and devices

## Day 2 - May, 23rd 2012

DFT and beyond for ab-initio transport modelling I (Chair: Stefan Bluegel)

- 09:00 to 09:35 - **Ferdinand Evers**
  Fundamental aspects of density functional transport theories
- 09:35 to 10:10 - **Robert Stadler**
  Exchange and correlation effects in electron transfer through a single intra-molecular barrier for weak and intermediate coupling
- 10:10 to 10:45 - **Mikkel Strange**
  First-principles transport calculations: The GW method applied to small molecule/gold junctions

Coffee break

- 10:45 to 11:15 - Coffee Break

DFT and beyond for ab-initio transport modelling II (Chair: Stefan Bluegel):

- 11:15 to 11:50 - **Jeff Neaton**
  Quantitative First-Principles Approaches to Conductance, Thermopower, and IV Characteristics of Molecular Junctions
- 11:50 to 12:25 - **Andrea Ferretti**
  Hybrid functionals and GW corrections to quantum transport calculations

Lunch break

- 12:25 to 14:00 - Lunch Break

Controlling molecular transport I (Chair: Gemma Solomon)

- 14:00 to 14:35 - **Elke Scheer**
  Mechanical Control of the Electronic Transport through Individual Molecules
- 14:35 to 15:10 - **Colin Lambert**
  Controlled Electron Transport Through Single Molecules
- 15:10 to 15:45 - **Troels Markussen**
  Quantum interference in molecular junctions

Coffee break

- 15:45 to 16:15 - Coffee Break

Controlling molecular transport II (Chair: Gemma Solomon)

- 16:15 to 16:45 - **Herre van der Zant**
  Single-molecule three-terminal devices
- 16:45 to 17:20 - **Charles Stafford**
  Charging effects and thermoelectricity in molecular junctions
- 17:20 to 17:55 - **Sense Jan van der Molen**
  Quantum interference in molecular charge transport

Poster session

- 18:15 to 20:30 - Poster Session

## Day 3 - May, 24th 2012

Electron-ion interactions I (Chair: Latha Venkataraman)

- 09:00 to 09:35 - **Douglas Natelson**
  Molecular-scale junctions out of equilibrium: Noise and Raman Spectroscopy
- 09:35 to 10:10 - **Jing-Tao Lu**
  Current-induced atomic dynamics, instabilities, and Raman signals: Quasi-classical Langevin equation approach
- 10:10 to 10:45 - **Daniel Frisbie**
  Understanding the Role of Contacts in Molecular Junctions and Examining the Transition from Tunneling to Hopping Transport as a Function of Molecular Length

Coffee break

- 10:45 to 11:15 - Coffee Break

Electron-ion interactions II (Chair: Latha Venkataraman)

- 11:15 to 11:50 - **Gemma Solomon**
  Calculating Inelastic Electron Tunneling Spectra: Exploring Chemical Space
- 11:50 to 12:25 - **Tchavdar Todorov**
  Non-conservative forces under current: a simple interpretation

Lunch break

- 12:25 to 14:00 - Lunch Break

Spin transport I (Chair: Stefano Sanvito)

- 14:00 to 14:35 - **Nicolae Atodiresei**
  A First Principles Tour into Hybrid Organo-Metallic Interfaces
- 14:35 to 15:10 - **Nicolás Lorente**
  Kondo effects in inelastic spin transitions
- 15:10 to 15:45 - **Aaron Hurley**
  Spin inelastic electron transport through magnetic nanostructures

Coffee break

- 15:45 to 16:15 - Coffee Break

Spin transport II (Chair: Stefano Sanvito)

- 16:15 to 16:45 - **Maarten Wegewijs**
  Spin-anisotropy induced by quantum transport
- 16:45 to 17:20 - **Magnus Paulsson**
  Inelastic scattering from first principles calculations
- 17:20 to 17:55 - **Jens Paaske**
  Cotunneling fine-structure near a singlet-triplet transition

Social pub session

## Day 4 - May, 25th 2012

Time-dependent transport I (Chair: Stefan Kurth)

- 09:00 to 09:35 - **Robert van Leeuwen**
  Many-body effects in time-dependent quantum transport
- 09:35 to 10:10 - **Gianluca Stefanucci**
  Advances in non-equilibrium strongly correlated models using TDDFT

Coffee break

- 10:10 to 10:45 - Coffee Break

Time-dependent transport II (Chair: Stefan Kurth)

- 10:45 to 11:20 - **Thomas Niehaus**
  Time domain versus energy domain approaches in quantum transport
- 11:20 to 11:55 - **Peter Bokes**
  Quantum transport in stroboscopic wavepacket basis

Closing remarks

- 11:55 to 12:10 - Closing word

**CECAM - Centre Européen de Calcul Atomique et Moléculaire**
**Ecole Polytechnique Fédérale de Lausanne, Batochime (BCH), 1015 Lausanne, Switzerland**

# Quantum transport in molecular nanostructures

May 22, 2012 to May 25, 2012

Location : Trinity College Dublin, Ireland

Details Participants Program Posters Abstracts Report

- Jos Thijssen *(Kavli Institute of Nanoscience Delft University of Technology)*

## Austria

- Georg Kastlunger *(University of Vienna)*
- Robert Stadler **(invited speaker)** *(University of Vienna)*

## Czech Republic

- Tom‡ä Novotnù *(Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University in Prague, Czech Republic)*

## Denmark

- Morten Gjerding *(Technical University of Denmark)*
- Elvar Örn Jonsson *(CAMD, DTU Physics)*
- Jacob Lykkebo Jørgensen *(University of Copenhagen)*
- Jing-Tao Lu **(invited speaker)** *(Technical University of Denmark, Lyngby)*
- Troels Markussen **(invited speaker)** *(Technical University of Denmark)*
- Jens Paaske **(invited speaker)** *(The Niels Bohr Institute & Nano Science Center)*
- Gemma Solomon **(invited speaker)** *(University of Copenhagen)*
- Mikkel Strange **(invited speaker)** *(Technical university of Denmark, physics)*
- Marco Vanin *(University of Copenhagen)*

## Finland

- xi chen *(University of Jyvaskyla)*
- Hannu Häkkinen **(invited speaker)** *(University of Jyväskylä)*
- Robert van Leeuwen **(invited speaker)** *(University of Jyväskylä)*

## France

- Nicolas Génin *(Complex and Adaptative Systems Laboratory)*
- Raghvendra Pratap Singh *(IEMN, University of Science and Technology, Villeueve d"Ascq, Lille)*

## Germany

- Nicolae Atodiresei **(invited speaker)** *(Forschungszentrum Jülich )*
- Marius Buerkle *(University of Karlsruhe)*
- Ferdinand Evers **(invited speaker)** *(Karlsruhe Intitute of Technology (KIT))*

- Rico Friedrich *(TU Bergakademie Freiberg Institute of Theoretical Physics)*
- Rainer Haertle *(University of Erlangen-Nuernberg)*
- Thomas Hellmuth *(University of Karlsruhe)*
- Simon Liebing *(Institute of Theoretical Physics, TU Bergakademie Freiberg)*
- Thomas Niehaus **(invited speaker)** *(University of Regensburg)*
- Fabian Pauly **(invited speaker)** *(University of Karlsruhe)*
- Jascha Repp **(invited speaker)** *(University of Regensburg)*
- Elke Scheer **(invited speaker)** *(University of Konstanz)*
- Maarten Wegewijs **(invited speaker)** *(University of Aachen & FZ Juelich, Germany)*

## India

- Sandip Bhattacharya *(Trinity College Dublin and CRANN)*
- vijay lamba *(HCTM College Kaithal Haryana; India)*
- Dinesh Prasad *(Shri Venkteshwar Institute of technology , Indore )*

## Ireland

- Akinlolu Akande *(Trinity College Dublin)*
- Nadjib Baadji *(Trinity College Dublin)*
- Laila Balobaid *(university collage dublin)*
- Thomas Cathcart *(Trinity College Dublin)*
- Clotilde Cucinotta *(Physics, Trinity College Dublin)*
- Paul Gorman *(Trinity College Dublin)*
- Aaron Hurley **(invited speaker)** *(Trinity College Dublin)*
- James Lawlor *(TCD)*
- Donal Mackernan *(University College Dublin)*
- Devin Murray *(ACAM)*
- Awadhesh Narayan *(Trinity College Dublin)*
- Anna Pertsova *(Trinity College Dublin)*
- Stephen Power *(Trinity College Dublin)*
- Ivan Rungger **(invited speaker)** *(Trinity College Dublin)*
- Shourjya Sanyal *(CASL, School of Physics, UCD )*
- Amaury Souza *(Trinity College Dublin)*
- Maria Stamenova *(School of Physics, Trinity College Dublin )*
- Angelo Ziletti *(School of Physics - UCD - Dublin)*

## Italy

- Andrea Ferretti **(invited speaker)** *(CNR-Institute of Nanoscience, Modena)*
- Gianluca Stefanucci **(invited speaker)** *(University of Rome, Tor Vergata, Italy)*

## The Netherlands

- Everardus Hendrik Huisman *(University of Groningen)*
- Sense Jan van der Molen **(invited speaker)** *(Leiden University, The Netherlands)*
- Herre van der Zant **(invited speaker)** *(Delft University of Technology)*

## Saudi Arabia

- Konstantinos Gkionis *(King Abdullah University of Science and Technology)*
- Tobechukwu Obodo *(King Abdullah University of Science and Technology)*

## Slovakia

- Peter Bokes **(invited speaker)** *(Slovak University of Technology)*

## South Africa

- Kingsley Obodo *(University of Pretoria)*

## Spain

- Nicolas Agrait **(invited speaker)** *(Universidad Autonoma de Madrid)*
- Núria Aliaga-Alcalde *(ICREA Researcher-University of Barcelona)*
- Daniel Aravena *(Universitat de Barcelona)*
- Nicolás Lorente **(invited speaker)** *(Centro de Investigaciones en Nanociencia y Nanotecnologia)*
- Eliseo Ruiz *(Universitat de Barcelona)*

## Sweden

- Magnus Paulsson **(invited speaker)** *(Linnaeus University)*

## Switzerland

- Nicola Marzari **(invited speaker)** *(École Polytechnique Fédérale de Lausanne)*

## Taiwan, Republic of China

- Chao-Cheng Kaun *(Academia Sinica)*

## United Kingdom

- Colin Lambert **(invited speaker)** *(Lancaster University)*
- Emanuele Maggio *(University of Warwick Chemistry Department)*
- John McGrady *(Univ. Oxford)*
- Tchavdar Todorov **(invited speaker)** *(Queen's University Belfast)*

## USA

- Arunabh Batra *(Columbia University)*
- Daniel Frisbie **(invited speaker)** *(University of Minnesota)*
- Douglas Natelson **(invited speaker)** *(Rice University)*
- Jeff Neaton **(invited speaker)** *(Lawrence Berkeley National Laboratory)*
- Charles Stafford **(invited speaker)** *(University of Arizona)*
- Latha Venkataraman **(invited speaker)** *(Columbia University)*

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