RESEARCH CONFERENCES

ESF Conference in Partnership with LFUI

Charge Transfer in Biosystems

Universitätszentrum Obergurgl (Ötz Valley, near Innsbruck) • Austria
17-22 July 2011

Chaired by:
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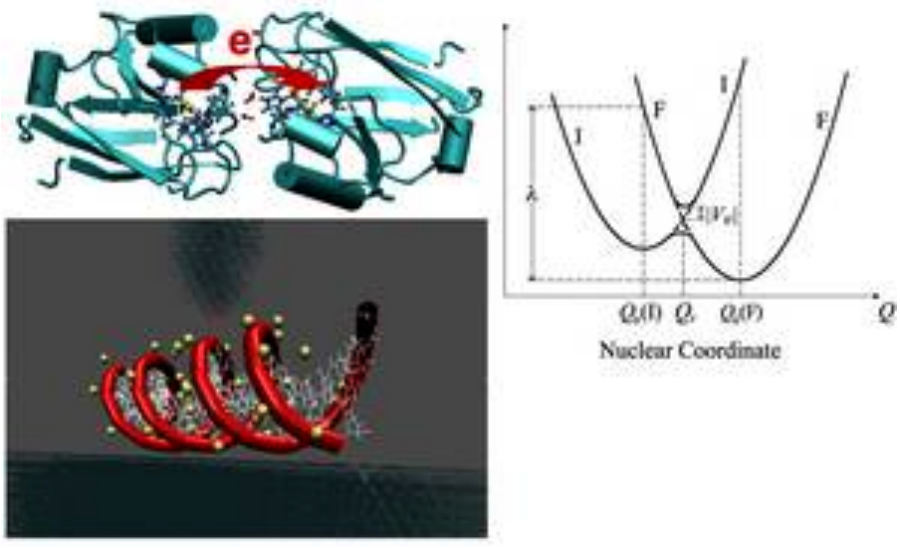

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Conference Highlights

Please provide a brief summary of the conference and its highlights in non-specialist terms (especially for highly technical subjects) for communication and publicity purposes. (ca. 400-500 words)

The issue of charge transfer in biosystems is a hot topic in the scientific community since decades, especially due to its relevance in biology and medicine. The advent of nanotechnology and in particular the use of biomolecules in devices has rekindled the interest in this field. This situation has generated the confluence of many different approaches to study the problem.

For instance, traditionally measurements of charge transfer were done on molecular ensembles in solution (chemistry groups), while now one can also measure electrical currents through single molecules between electrodes (physics groups): what is the relation between the transfer rates measured in solution and the electrical currents measured in devices? Theoretical methods are also merging and competing: is it better to compute transfer integrals at a high accuracy level for small frozen fragments or is it better to do simplified semi-empirical simulations of entire fluctuating molecules? What is the role of self-interaction corrections? Is it important to go beyond the two-state approximation? All these problems were debated during the conference.

A particularly relevant aspect, that was repeatedly raised during the plenary and informal discussions, is the relation between theory and experiment: how can the theorists communicate easily to the experimentalists what are the shortcomings of the existing theoretical methods for which the interpretation of experiments is not always clear and unanimous? How can experiments be employed to validate theories? Can we plan benchmark experiments that effectively assess theoretical/computational results? Can we identify small benchmark biosystems on which we can appraise different approximations in the existing methods? I remark the lively dialogue between theorists and experimentalists at the conference.

The conference was successful from several viewpoints:

- It really brought together different communities with diverse expertise and multi-disciplinary background (physics, biology, chemistry, engineering);
- It created an informal atmosphere that enabled unbiased discussions;
- It effectively brought into play early stage researchers, who animatedly participated not only in the oral/poster program but also in the discussions;
- It created new contacts between scientists, likely promoting future collaborative projects;
- There was a very high scientific quality of the presentations;
- The participants were quite satisfied of all the aspects of the conference, including the science, the organization, the venue, the social program, the professional response of the staff to any sudden request.

For me, as the conference Chair, it was particularly rewarding to clearly see the satisfaction of the participants, which is explicitly reflected in two initiatives that during the conference I was stimulated to undertake and are currently under review: a new ESF conference and an ESF-COST action. The latter can be the ideal framework for more regular meetings of research groups interested in charge transfer in biosystems.

I wish to express particular gratitude to the ESF representative Victoria Ibbertson. Besides the preliminary organizational work before the conference, she was also perfect on site: never redundant, always precise.

X ☐ I hereby authorize ESF – and the conference partners to use the information contained in the above section on 'Conference Highlights' in their communication on the scheme.

Scientific Report

Executive Summary

(2 pages max)

The conference program included 8 oral topical sessions and 1 poster session. The posters stayed on the boards for the duration of the conference. The oral sessions included invited lectures of 40 minutes each and short talks of 20 min each. The latter were selected among the submitted abstract, which were overall of high quality. In the details below, names of invited speakers are in bold characters. Each oral session was concluded with a discussion phase.

Session 1 was devoted to the fundamentals of electron transfer theory and the relation to charge transport. It included 2 invited lectures and 2 short talks. Moderator Yuri Berlin.

1. **Spiros Skourtis**, University of Cyprus, CY. *Modeling electron transfer and transport from the molecular to the cellular length scales*.
2. Emilie Cauët, University Libre de Bruxelles, BE. *Hole-trapping property of the human telomere sequence*.
3. Vladimir Egorov, Russian Academy of Science, RU. *Novel theory of charge transfers in condensed matter and its correlation with experiment: Optical line shapes for polymethine dyes and their aggregates*.
4. **Abraham Nitzan**, Tel Aviv University, IL. *Circular currents, current transfer and magnetic field effects in molecular wires*.

Session 2 was devoted to the development and application of computational schemes to determine the charge transfer poperties of materials, including biological materials. It included 3 invited lectures and 5 short talks. Moderator Rosa Di Felice.

5. **Nicola Marzari**, Oxford University, UK. *Charge transfer from first-principles: challenges and solutions*.
6. David Bowler, University College London, UK. *Charge transfer in large systems with linear scaling constrained DFT*.
7. Thorsten Hansen, Lund University, SE. *Non-equilibrium Green's function theory of 2D electronic spectroscopy*.
8. **Alessandro Troisi**, University of Warwick, UK. *What can we learn about charge transfer in biosystems from organic electronics?*
9. Michele Pavanello, Leiden University, NL. *Charge transfer in biological systems studied by subsystem density functional theory*.
10. Michael Zwolak, Los Alamos National Laboratory, US. *Rapid DNA sequencing via transverse electronic transport*.
11. Tomas Kubar, Karlsruhe Institute of Technology, DE. *Non-adiabatic simulation of charge transfer in DNA*.
12. **Gianaurelio Cuniberti**, TU Dresden, DE. *From molecular wires to organic semiconductors and back - some "don't ask, don't tell" of soft electronics*.

Session 3 was devoted to the investigation of charge transfer/transport in proteins and complex biological systems. It included 2 invited lectures and 5 short talks. Moderator Danny Porath.

13. **P. Leslie Dutton**, University of Pennsylvania, US. *Molecular engineering of photochemical charge separation*.
14. Nurit Ashkenasy, Ben Gurion University, IL. *Charge transfer through, and from, artificial proteins in solid state configurations*.
15. Samita Basu, Saha Institute of Nuclear Physics, IN. *Magnetic field effect on photoinduced electron transfer between calf thymus DNA and ternary copper complex*.
16. **Paolo Facci**, CNR-NANO-S3 Modena, IT. *ECSTM/STS investigation of single molecules bearing two redox levels*.
17. Eduardo Della Pia, Cardiff University, UK. *Observations of conductance gating for a single-redox engineered protein junction*.
18. Randall Thomas Irvin, University of Alberta, CA. *Spontaneous modulation of the electronic state of stainless steel via a novel synthetic bio-metallic interface*.
19. Liliana Radu, Ministry of Health Romania, RO. *Fluorescence resonance energy transfer in the investigation of normal and tumoral chromatin structure*.

Session 4 was devoted to the measurement and modeling of charge transport in DNA in molecular nano-junctions. It included 1 invited lecture and 4 short talks. Moderator Abraham Nitzan.

20. **Danny Porath**, Hebrew University of Jerusalem, IL. *Charge transport and spectroscopy in DNA molecules*.
21. Daria Brisker-Klaiman, Technion-Israel Institute of Technology, IL. *Coherent elastic transport contribution to currents through ordered DNA molecular junctions*.
22. Margarita Dimakogianni, University of Athens, GR. *On the conductivity behaviour of the DNA double helix*.
23. Orsolya Ujsághy, Budapest University of Economics and Technology, HU *Conductance of DNA molecules: Effects of*

decoherence and bonding.

24. Erika Penzo, Columbia University, US. *Directed biomolecular assembly of integrated single molecule devices: toward reliable transport measurements*.

Session 5, like session 3, was devoted to the investigation of charge transfer/transport in proteins and complex biological systems. It included 2 invited lectures and 6 short talks. Moderator Paolo Facci.

25. **Krzysztof Bobrowski**, Institute of Nuclear Chemistry & Technology, PL. *Radiation-induced electron transfer in enkephalins*.
26. Carlo Augusto Bortolotti, University of Modena, IT. *Transient open of solvent-accessible cavities in Yeast cytochrome c as a tool for fine-tuning of its redox potential*.
27. Lior Sepunaru, Weizmann Institute Rehovot, IL. *Temperature dependent electron transport in proteins*.
28. Moh El-Naggar, USC Los Angeles, US. *Electron Transfer across the Biotic-Abiotic Interface in Microbial Fuel Cells*.
29. **Bernd Giese**, University of Fribourg, CH. *Electron hopping through peptides: The role of side chains and the backbone*.
30. Stefano Corni, CNR-NANO-S3 Modena, IT. *Electron transfer proteins on gold surfaces investigated by molecular dynamics simulations*.
31. Gilbert Nöll, University of Siegen, DE. *Electrochemical switching of the flavoprotein dodecin on DNA-monolayers*.
32. Brotati Chakraborty, Saha Institute of Nuclear Physics, IN. *Magnetic field effect corroborated with docking study to explore photoinduced electron transfer in drug-protein interaction.*

Session 6 was devoted to charge migration and excitations in DNA, including excitations induced by light. It included 2 invited lectures and 2 short talks. Moderator Dimitra Markovitsi.

33. **Gary Schuster**, Georgia Institute of Technology, US. *Radical cation hopping and reaction in DNA*.
34. Irena Kratochvílová, Academy of Sciences of the Czech Republic, CZ. *Charge transport in DNA oligonucleotides with various base-pairing patterns*.
35. John M. Kelly, Trinity College Dublin, IE. *Dipyridophenazine metal complexes which undergo photo-induced electron transfer with DNA*.
36. **Torsten Fiebig**, Northwestern University, US. *Electronic Transfer Processes in Biological and Biomimetic Donor-Acceptor Systems*.

Session 7, like session 2, was devoted to the development, assessment and application of computational schemes for charge transfer parameters. It included 3 invited lectures and 2 short talks. Moderator Marcus Elstner.

37. **Agostino Migliore**, Tel Aviv University, IL. *Effective electronic coupling calculation using non-orthogonal diabatic states: application to charge transfer in !-stacks relevant to biochemistry and nano-electronics*.
38. Andrea Ferretti, CNR-NANO-S3 Modena, IT. *Hybrid functional and GW corrections to quantum transport calculations*.
39. **Ferdinand Grozema**, Delft University, NL. *Charges and excited states in DNA hairpins: a theoretical study*.
40. **Jochen Blumberger**, University of Cambridge, UK. *Electron transfer in cytochromes, oxidase and bacterial `wire'-proteins: Insights from molecular computations*.
41. George Kalosakas, University of Patras, GR. *Electronic parameters for charge transfer along DNA.*

Session 8, like session 6, was devoted to the investigation of charge migration and excitations in DNA. It included 4 invited lectures and 3 short talks. Moderator Gary Schuster.

42. **Thomas Carell**, Ludwig-Maximilians University, DE. *Metal-base pairs and metal containing DNA*.
43. **Dimitra Markovitsi**, CEA Saclay, FR. *Electronic excited states and reactivity of DNA*.
44. Frank Garwe, IPHT Jena, DE. *Long-range energy transfer in DNA after fs laser pulse excitation of silver nanoparticle neighboured to DNA*.
45. Rudy Schlaf, University of South Florida, US. *Electronic structure of self-assembled peptide nucleic acid thin films.*
46. **Jason Slinker**, University of Texas at Dallas, US. *Fundamentals of DNA-mediated electrochemistry.*
47. Marcos Brown Goncalves, University of Sao Paulo, BR. *Theoretical study of metal DNA structures.*
48. **Hans-Achim Wagenknecht**, University of Regensburg, DE. *Photoinduced electron transfer in synthetically modified DNA.*

The poster session was preceded by a flash presentation of posters, in which the early stage researchers who presented posters could illustrate with one slide the contents of their posters to the audience. There was an excursion that stimulated informal discussions among the participants: it was attended by almost all participants. There was a final Forward Look Plenary Discussion in the evening of the last program day (Thursday July 21), in which we discussed the stat-of-the-art and the challenges for the future, set few goals for the near future and planned a new conference on the same topics, chaired by Rosa Di Felice with co-chairs Bernd Giese and Spiros Skourtis.

Scientific Content of the Conference

(1 page min.)

- *Summary of the conference sessions focusing on the scientific highlights*
- *Assessment of the results and their potential impact on future research or applications*

Background

Charge transfer processes in biological molecules are important components of biochemical pathways in living organisms with potential applications in future medical, nano and energy science technologies. Studies of the charge transfer phenomena have been traditionally performed by chemists. Recently, with the advent of nanotechnology and the investigation of nucleic acids and proteins in the context of devices and single-molecule experiments, different communities have also come into play, notably physics and engineering. New theoretical and experimental methodologies have been proposed and successfully applied. The field is now mature to integrate the various approaches to get a deeper insight of the mechanisms of charge transfer in bio-systems both in solution and at the interface with solid materials. The conference has been an effective tool to confront different approaches and foster such integration.

Fields of interest:

- Nanomedicine (diagnostic and therapeutic tools, cancer treatment);
- Nanotechnology (future-generation electronic devices and computers, sensors and actuators);
- Toxicity of nanoparticles for the human body;
- Design of new materials guided by reactions with/between biological components;
- Energy transfer, artificial photosynthesis, energy production and storage.

Current knowledge

Charge transfer trough DNA and proteins has been explored in solution for several decades by introduction of a charge excitation at a specific point of the nucleic or amino acid chain, collecting it at another precise site and measuring the speed of propagation. These measurements are based on chemical alterations of the biomolecules. They are done on ensembles of molecules and involve the transfer of a single charge, be it positive or negative. Such experiments have revealed fast and slow charge transfer motions and different mechanisms have been proposed.

With the advent of nanotechnology and the need for finding new materials for device development, scientists have started to exploit in electronic applications the ability of biomolecules to transfer charge. The special self-assembly and structuring skills of biomolecules could be exploited to place them between electrodes and generate powerful self-forming devices and circuits at a high rate and low cost. Experiments on single molecules, rather than ensembles, are being carried out nowadays in many groups, with the objective of measuring the currents due to many electrons simultaneously, not one at a time. The electrode-molecule contact, under applied voltage, introduces a reservoir of mobile electrons into the molecule. The objective is to find molecules in which the mobility of these electrons can be exploited for practical purposes.

Theoretical efforts correspond to each of the above experimental approaches, with different methods. The first class of experiments is tackled on the basis of electron transfer theory. Within this theory, transfer integrals between a donor and an acceptor can be calculated as the splitting between adiabatic curves at the transition coordinate. This, in turn, can be computed in terms of energy level differences by Density Functional Theory (DFT), taking into account only the donor and acceptor sites, not the entire molecule. The second class of experiment is normally tackled by Landauer's transport theory, which allows us to compute the quantum conductance in a nano-junction. This can also be implemented in DFT, using Green function techniques.

Conference summary

The first introductory session was devised to set the **basics of charge transfer theory and its relation to the quantum theory of conductivity**. Two invited lectures fulfilled this scope.

- Spiros Skourtis emphasized the role of environmental fluctuations and the need for enhanced sampling techniques: not only time enhanced sampling, but general methods to improved tha statistics and account for rare events. He then discussed an application to bacteria nanowires that are essentially 2-dimensional networks of cytochromes with high packing and high order. These systems were also the topic of an experimental talk later in the program and a lively discussion soon arose.
- Abraham Nitzan talked about current transfer, explaining the relevance of transfer not only of the

position but also of the momentum. He introduced magnetic effects and the excitation of circular currents. He presented studies on simple model systems, namely benzene rings.

The two invited lectures were complemented by 2 short talks on applications of theory and computation. The discussion at the end of the session was characterized by the question: "What distinguished electron transfer in biological systems from electron transfer in other systems?" It emerged that model systems can be used to study the basics of charge transfer mechanisms, but different systems have peculiarities.

Two sessions (2 and 7) were devoted to **computational approaches to charge transfer**.

- Nicola Marzari (invited) talked about the relevance of the self-interaction correction. He discussed the oxidation of Fe ions in water and short-range self-interaction in transition-metal chemistry, where he showed that GGA+U theory works.
- Alessandro Troisi (invited) discussed the relation between charge transfer in biosystems and organic electronics. He insisted again on the importance of dynamical effects. He presented work on the charge separation at organic/inorganic interfaces, relevant for solar-cell applications.
- Gianaurelio Cuniberti (invited) talked about Landauer's theory on charge transport and applications to: molecules at surfaces, bioelectronics, molecular materials.
- Agostino Migliore (invited) presented new theoretical developments to compute thansfer integrals in DFT using non-orthogonal diabatic states.
- Ferdinand Grozema (invited) talked about the electronic structure and excited states in DNA hairpins from computational approaches.
- Jochen Blumberger (invited) presented large-scale molecular simulations of protein systems.

David Bowler presented linear scaling constrained density functional theory (DFT), with examples of scaling for Si and Ge and application to DNA and proteins. Thorsten Hansen's work was based on non-equilibrium Green's functions techniques. Michele Pavanello discussed the relevance of the amount of exact exchange contributions in the DFT computation of transfer integrals with hybrid functionals. Michael Zwolak talked about computational studies of DNA sequencing: he remarked, once again, the paramount importance of structural fluctuations. Tomas Kubar presented a scheme to effectively account for fluctuations in computational investigations, based on SCC-DFTB and QM/MM. Discussion on theory aspects.

- Transfer integrals calculations: need for benchmark systems to validate theories and experimental approaches
- λ, ΔG: need for polarizable force fields.
- Importance of the initial state for the charge-transfer reaction.

Two sessions (3 and 5) were devoted to **charge transfer/transport in proteins and complex biological systems**.

- Leslie Dutton's (invited) work is devoted to understanding elementary processes of oxidation-reduction and diverse biological events coupled to it. He explores biological redox reactions and possibility of engineering photochemistry.
- Paolo Facci (invited) presented experimental work done on single biomolecules with the electro-chemical scanning tunneling microscope.
- Krystof Bobrowski (invited) introduced the radiation-induced electron-transfer in enkephalins. He discussed experimental work on two different kinds of enkephalins, with either Leu or Met at the C terminal. Radiation is an alternative method to induce charge transfer, relative to the widely employed photochemistry.
- Bernd Giese (invited) gave an overview on the significant experimental contribution of his group to elucidate charge transfer in DNA through the years. Then he focused on new work on peptide assays for electron transfer. He discussed through-bond electron transfer, connection to photosystems, water mediation, the role of charges.

Among the short talks in these sessions, I point out the work of Moh El-Naggar on bacteria nanowires, who carried out transport measurements on lithographic electrodes, revealing high currents probably due to the network of cytochromes. Carlo Bortolotti presented a dynamical view of cytochrome C from electrochemistry measurements. Lior Sepunaru presented current measurements in protein layers between electrodes, revealing fingerprints of unfolding by varying the temperature.

The discussion on experiments focused on how to understand the mechanisms of charge transfer through DNA: how does the polaron model match with superexchange? What are good experiments to prove the existence of polarons? Though precise answers did not emerge, these critical issues were identified for future directives.

One session (4) was devoted to **DNA conductance and charge transport in DNA molecular junctions.** The only invited lectures, Danny Porath, reviewed the pioneering work done by him and his collaborators to enable and understand measurements of electrical currents through single DNA molecules between nanoscale electrodes. He then presented new developments on more complex biosystems.

Two sessions (6 and 8) were devoted to **charge migration and excitations in DNA.** All the invited lectures presented experimental work and discussed various methods and systems, including photochemistry and electrochemistry, as well as various DNA modifications with metal inclusion and with photoactive elements. It emerged that, despite the long years of investigation, a clear understanding of the charge transfer mechanisms still deserves attention, especially if DNA is to be exploited for nanotechnologies.

Forward Look

(1 page min.)

- *Assessment of the results*
- *Contribution to the future direction of the field – identification of issues in the 5-10 years & timeframe*
- *Identification of emerging topics*

Assessment of the results and need to go beyond the current knowledge

While the question of charge transfer in biosystems has been addressed by various experimental and theoretical tools as described above, full understanding of the complex processes encompassing this phenomenon is not yet available. This is due to the need for integration of the knowledge from the different disciplines described above. The conference provided a unique platform for this task of bringing together teams with different complementary backgrounds. Throughout the conference the need for a networking action emerged, either collaborative projects or training initiatives. Specifically, by networking we seek to understand how the outcome of one theory relates to another theory, how the outcome of an experiment relates to a different experiment, how theory relates to experiment. Specific questions and problems for future developments of the field are:

- what is the role of the environment and of intrinsic fluctuations on the charge transfer through biomolecules?
- what determines the current through a single biomolecule in a nano-junction and how can one compute it if the transfer integrals between donor and acceptor sites are known?
- what are the desired improvements of theoretical frameworks and experimental setups of charge transfer studies?
- how can the charge transfer phenomena be exploited in technological applications?

Progress in this context will boost knowledge in the suggested fields, with tremendous impact on the important applications listed above in the previous section of this report. We anticipate it will aid in establishing new directions for cooperative research in the 7th and 8th European Framework Programmes and will result in joint project proposals in the future. Furthermore, it will answer the need for development of strategies for future commercialization of research-produced knowledge.

The conference was extremely successful in stimulating on-site discussions and scientific contacts, as well as in achieving the active participation of early stage researchers.

Contribution to future directions

Impact on science and society: The main purpose of continued research in the field of the conference is to promote the development of methods and systems to study charge transfer in biological and organic molecules and molecular aggregates and between such molecules and inorganic substrates. The goal is to increase knowledge on basic principles of charge transfer to design and characterize systems that will meet future medical and industrial needs. Networking among active groups (e.g., through other conferences or a COST Action) is also aimed at improving international competitiveness and industrial relevance of EU research.

Main research objectives:

- understanding charge transfer mechanisms and resulting properties by combining experimental and theoretical approaches; providing tools for the design of new functional materials with structure-tailored

properties;
- transfer of people and knowledge between all research groups participating in the Action;
- establishing a link between physicists, chemists, biologists and engineers interested in charge transfer in organic and biological systems;
- identifying possible wide-scale cooperative work in the future and applying for joint research projects within EC Framework and beyond.

These objectives can be pursued through: international workshops, exchange visits, training of young researchers, scientific publications, proposals for international funding.

Identification of emerging topics

1. Development of electron-transfer theory. New computer implementations. Exploration of the limits of existing theoretical/computational methods.
2. Experimental investigations of charge transfer in bio/organic moleculesand supramolecular systems in solution.
3. Experimental investigation of charge transport through biomolecules between electrodes, development of setups to probe single molecules.
4. Theoretical and experimental investigation of charge transfer/transport between molecules and inorganic nanoparticles.
5. Applications in medicine, nanotechnology and energy science.

Benefits

The relevance of charge transfer phenomena in biosystems to both medicine and technology makes this field challenging, and exploratory research is quickly evolving. The conference helped correlate the work of European researchers and enhance their potential to unravel the mechanisms of charge transfer, which is a necessary step towards rational exploitation. Future collaborative work, also fostered by connections at the conference, is expected to have considerable scientific and also possible technological benefits for the know-how of this interesting field in Europe. In addition, many researchers working on charge transfer phenomena are young scientists, who will benefit from training visits and interactions with other groups.

▪ Is there a need for a foresight-type initiative?

YES. We have applied for a COST Action. However, thematic actions at the European level would also be necessary, either ESF program (e.g., RNP) or EC call.

Atmosphere and Infrastructure

▪ *The reaction of the participants to the location and the organization, including networking, and any other relevant comments*

The participants were enthusiast about the “geographic” location, due to the beautiful surrounding mountain landscape. Those driving through the Timmelsjoch also enjoyed the road trip. The weather was not good to enjoy the surroundings every day, but we managed to have a social excursion to the mountains and some participants organized other hikes in small groups. I should also note that the management at the venue was extremely collaborative when we wanted to shift the social trip from Wednesday to Tuesday due to the “occasionally” favorable weather conditions on Tuesday (contrary to the previous days) and we asked lunch bags at the last moment... Our secretary Victoria Ibbertson was also excellent, organizing last-minute cableway tickets for everybody.

The participants commented positively on the venue: the fact of being all together in a single place, for lodging, meals and conference, of course stimulated scientific discussion and friendship. In a similar situation, the risk is to feel “trapped”, so there is always some initiative to escape outdoors. These initiatives are indeed good, an ideal playground for informal brainstorming discussions, which are precious in scientific research. I think that in general in these conditions it is important to plan some long breaks in the program, to facilitate flexible actions beyond the program sessions. We were satisfied of the conference facilities and of the responsiveness of the staff in relation to the technical equipment.

Networking was effective: people always discussed, I barely saw somebody staying on his/her own. People met new

potential collaborators and colleagues with whom they already collaborate. The community planned a new conference and a networking action, which I think is a very good outcome for such an event. I am confident that collaborative projects and exchange visits will also emerge from the conference.
There was a good percentage of women scientists (27%) and young researchers (51%). There was lively interaction between young researchers and senior scientists. We had a very lively after-dinner outing on the last evening, with about 50 participants, both young and senior, in which the friendly atmosphere was really clear.

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Date & Author:

November 18, 2011. Rosa Di Felice