THEORETICAL SPECTROSCOPY

The ETSF Users' Newsletter

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European Theoretical Spectroscopy Facility

Welcome to the seventh edition of the ETSF Users' Newsletter. In this issue, we chat to one of our users about his experiences with the ETSF, and also discover how understanding properties of materials can help to drive technological developments. Don't miss the upcoming events listed on page two.

Submission deadline for the ETSF's Spring call for proposals: 28 April, 17:00 (CET).

ETSF Scientist receives prestigious ERC Award

Angel Rubio, director of the NanoBio Spectroscopy group in San Sebastián, and ETSF Vice-president for Scientific Development, has received a prestigious European Research Council (ERC) Advanced Investigator grant.

Angel, along with two of his group members, Pierluigi Cudazzo and Matteo Gatti, will develop a theoretical framework which will allow prediction of new materials with tailored properties, either for the capture of energy, storage or transport, in an effort to develop alternative energy sources.

"With the ERC Advanced grant we will have the means to address successfully the ambitious objectives, including the control of the processes of dissipation and quantum coherence in nanostructures and biomolecules that are essential for designing new efficient artificial photosynthetic systems," Angel said.

User's Corner

Dr. Martin Knupfer is the head of the "Electronic and Optical Properties" department at the Leibniz Institute for Solid State Research (Dresden), and is one of our users. We spoke to him about his experiences working with the ETSF.

Dr. Knupfer, what scientific problem were you working on when you contacted the ETSF?

We carried out electron energy-loss spectroscopy and photoemission spectroscopy of pentacene and picene in the condensed phase. Both materials are aromatic hydrocarbons with quite similar molecular and crystal structures. For us, they are interesting due to rather fundamental issues, such as the question of the details in the density of states or the character of excitons in these materials. Moreover, in early 2010 it was reported that upon potassium doping, picene becomes superconducting, with a large transition temperature in the superconducting state of 18K. It is the organic semiconductor with the 2nd-highest Tc, only surpassed by the famous fullerenes. In order to understand the electronic properties of these interesting materials, we have been seeking for theoretical support that – together with our experimental results – can provide much more insight into the details.

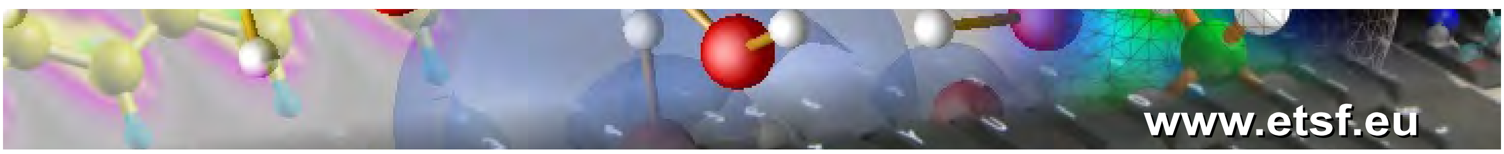
What were you expecting from your collaboration with the ETSF, and what was your experience?

My expectation mainly was the hope to be able to determine the density of states of picene and its excitation spectrum in a combined theoretical and experimental effort. Further, we are very much interested in the exact character and dispersion of excitons in pentacene single crystals. Our collaboration within the ETSF proposal was very close and fruitful, and we have published a first joint paper [1] on the electronic properties of undoped picene. This is the first paper that presents this kind of information based on sophisticated theory and experiment. We had regular contact via email and even met once with our theory collaborators from Angel Rubio's group, and I consider this project as very promising and successful, even at present where not all aspects have been finished.

What were you able to conclude from the project?

Within our joint work we have been able to (1) determine the occupied and the unoccupied electronic density of states of undoped picene close to the Fermi level. (2) analyse the electronic excitation spectrum and shed light on the importance of local field effects as well as electron-electron interactions for the understanding of picene. Both of these results would not have been possible to derive with the achieved depth without the theoretical part! Finally, our collaboration is still active.

[1] Roth et al, New J. Phys. 12 (2010) 103036

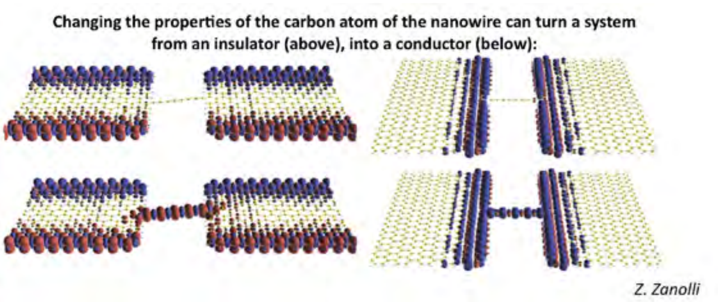


Theoretical Spectroscopy and the future of electronic devices

By G. Onida

Microelectronics, such as microchips and memory devices, rely almost exclusively on today's silicon-based technology. However, recent discoveries have demonstrated the possibility to conceive and to realize electronic devices made entirely of carbon, hence opening completely new perspectives in terms of possible applications.

One of the most important discoveries has been the production of carbon in the form of linear atomic chains, i.e. by carbon atoms displaying their so-called sp-hybridization, where atoms are bound to two neighbors oriented along a straight line (to be contrasted with the usual sp2 and sp3 hybridizations, where carbons are bound to three or four neighbors, as typically found in graphite and diamond, respectively), thus realizing one-dimensional nanowire structures.



The cluster beam deposition technique (used to produce amorphous cabon containing large fractions of linear chains) employed by the experimental group of Paolo Milani at the university of Milan, produced in 2002 some of the first experimental evidence of the existence of sp-hybridized carbon, by detecting its signature in the Raman spectra [1]. However, the measured spectra have a complex multi-peak structure, and show a nontrivial evolution with time, accompanying the progressive decomposition of linear chains induced by the exposure of the deposited samples to an oxidizing atmosphere. In order to achieve a systematic understanding of the spectra, and to connect the spectra with well-defined system properties, a thorough theoretical study was needed. This was the reason pushing Luca Ravagnan, one of the leaders of the experimental group in Milan, to submit a proposal to the ETSF.

Using a combination of Density Functional Theory (to determine structural properties, stability and electronic properties), Density Functional Perturbation Theory (to determine Raman spectra) and Green's Functions theories (to study quantum transport and conductance), the project provided several important insights [2-4]. First of all, it was found that sp-carbon chains are stabilized by bonding to the edges of graphitic nanofragments (which are present in cluster-deposited films). This has subsequently been confirmed in an important experimental paper [5], wherein high-resolution electron microscopy images showed the precise structures predicted by our calculations. As another important result, all nontrivial spectral features and decays of the experimental Raman spectra were consistently understood and interpreted. Moreover, the data for sp2-terminated chains point towards a rich phenomenology driven by even/odd alternation effects and by the effects of torsional strain. The latter modifies the electronic states near the Fermi level, suggesting the possibility to control the nanowire conductance, optical properties, and spin magnetization purely by twisting its sp2 termination, e.g. by coupling the terminating graphitic fragments with micromachined torsional actuators. Linear carbon chains bridging graphene nanogaps, recently proposed as an explanation of the conductance switching in two-terminal graphene devices [6], could hence acquire an important role in future carbon-based electronics.

[1] Ravagnan et al, Phys. Rev. Lett. 89, 285506 (2002).
[2] Ravagnan et al, Phys. Rev. Lett. 102, 245506 (2009).
[3] Manini and Onida, Phys. Rev. B. 81, 127401 (2010).
[4] Zanolli Z et al, ACS Nano. 9, 5174-80 (2010).
[5] Jin et al, Phys. Rev. Lett. 102, 205501 (2009); Meyer et al, New J. Phys. 11, 083019 (2009).
[6] Li et al, Nature Materials 7, 966 (2008); Standley B et al,

ETSF Agenda

2-6 May 2011 **CECAM school**: Theoretical Spectroscopy Lectures: theory and codes.

27-30 September 2011 **16th ETSF workshop on Electronic Excitations**: Besides addressing recent methodological advances within the various theoretical spectroscopy approaches, the workshop will aim for stronger and deeper exchange and dialogue with the community of experimentalists.

More information about these events at **www.etsf.eu**

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e-infrastructure

CAPACITIES