ETSF Newsletter

Fall Edition 2021

Dear ETSFers! It is with a mixture of excitement and frustration that I contribute a few lines to this revamped ETSF Newsletter.

Excitement because against all odds and omicron I will be seeing some of you live next week to pump some calcium carbonate and ETSF adrenaline. Excited that Simona has proposed to edit a round of digital newsletters, to get some information flowing, allow you to advertise your latest work and collaborations, and highlight events and opportunities which will shape the future of our field, its funding, orientation and structure.

Frustration because as you all know we are still in a big covidy mess, which will have a lasting impact on our daily lives, comfort and democracy, and on the future of younger researchers, through lost opportunities for contacts and collaboration, and the further restriction of funding in future austerity measures. We have learned that digital tools and online access does not always produce equal opportunities - human interaction and networking is essential to learning, research, and to the progression of our work and careers.

This many-body interaction is the raison d'être of the ETSF, which has succeeded on many fronts, in dissemination, structuring training activities, and germinating collaborations. I want to ask all of you to (re)invest in our common social fabric in 2022. By joining and animating a collaboration or workshop organizing team. Or by publishing systematically with your ETSF affiliation: our numbers and publication records have grown immensely in the past 10 years, but articles acknowledging ETSF have stagnated after an initial exponential growth. By putting forward the names of new Research Team Leaders for the SC to consider, or simply by giving us straight feedback on what you want and need from the network, and what you can input.

This is an opportunity to thank the present Steering Committee for its dynamism: it is the first iteration with none of the original 10 founding PIs, and is demonstrating the energy and will to reinvent the network in order to serve our purposes best.

I wish you all an ambitious and productive 2022, with many new encounters, spectroscopic and otherwise

Matthieu

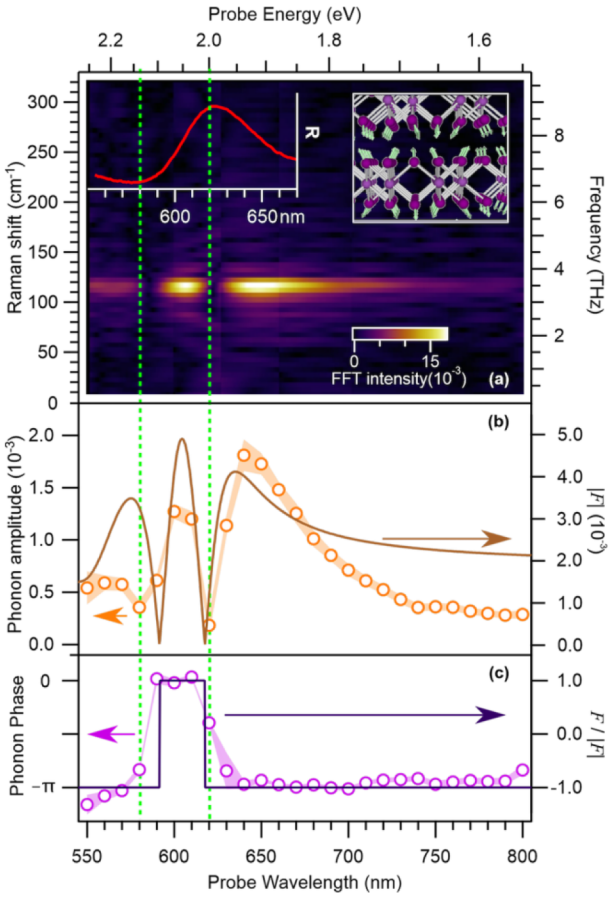
Focus Topic

Non equilibrium excitons and first principles modelling - by D. Sangalli

The implementation and use of the Bethe-Salpeter Equation to compute materials properties in the ab initio framework [1] is one of the most successful achievements of the ETSF Network. BSE describes the physics of the exciton. It has been implemented in many (ETSF and not) ab initio codes (Abinit, Yambo, EXC, BerkleyGW) and successfully applied to model a large variety of complex materials (bulk insulators and semiconductors, 2D materials, carbon nanotubes, etc..).

Experimentally excitons have been measured via absorption or photo-luminescence since many years ago. The advent of modern laser pulses opened the possibility to study coherent and non coherent excitons in the nonequilibrium (NEQ) regime [2]. Excitons can both be exploited for efficient optoelectronic devices and used to explore fascinating phenomena such as the generation of NEQ excitons Bose-Einstein condensate (BEC).

The implementation of the TD-HSEX scheme, or real-time BSE, [3] has made possible to model nonequilibrium excitons fully ab initio. Via real-time simulations, excitons can be explored in nonlinear optics [4], and dark excitons can be excited [5].



Coherent optical response in BiI_3. Theory and experiments. (Ref. 13)

Recent works discussed the intriguing possibility of generating NEQ-BEC of excitons, and of measuring the excitonic wave-function through time-resolved angle-resolved photo-emission experiments [6, 7]. This was also followed by a very reach experimental activity [8, 9]. The NEQ dynamics of excitons is dictated by their interaction with phonons. Different works have focused on modelling ab initio exciton–phonon interaction [10, 11]. Pump and probe experiments offer the opportunity to explore exciton–phonon in the real-time domain [12, 13].

Coupled equations for coherent and non-coherent exciton dynamics have been proposed [14, 15], and their feasibility in ab initio schemes is presently under study. Simpler schemes, based on coupling TD-HSEX with Ehrenfest dynamics could be adopted.

This is just one of the possible exciting developments we can expect in the next years.

Bibliography

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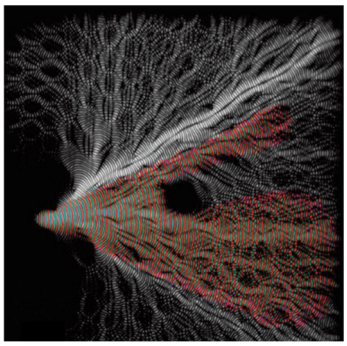
Scientific Highlights

Branched flow of electrons – by E. Rasanen



Branched flow is a common phenomenon in wave dynamics, where deflections of particles, rays or waves due to random fluctuations in the surrounding medium eventually lead to a chaotic pattern resembling the branches of a tree. Branched flow has been reported in a remarkable variety of physical systems covering, e.g., pulsar-generated microwaves, ocean waves, beams of light and two-dimensional (2D) electron gas [1,2]. We have discovered unexpected branched flow in electronic wave-packet dynamics of 2D periodic lattices without any randomized disorder [2].
The branching appears at wavelengths shorter than the typical length scale of the ordered periodic structure and for energies above the potential barrier. The branching is qualitatively similar in classical and quantum description. The strongest branches remain stable indefinitely and may create linear dynamical channels. In these channels the waves are not confined directly by potential walls as electrons in ordinary wires but rather, indirectly and more subtly by dynamical stability. We discuss the relevance of these "superwires" in 2D materials, especially in moiré superlattices, where the wavelengths are typically small compared to the lattice constant - thus creating an ideal system for branched flow.

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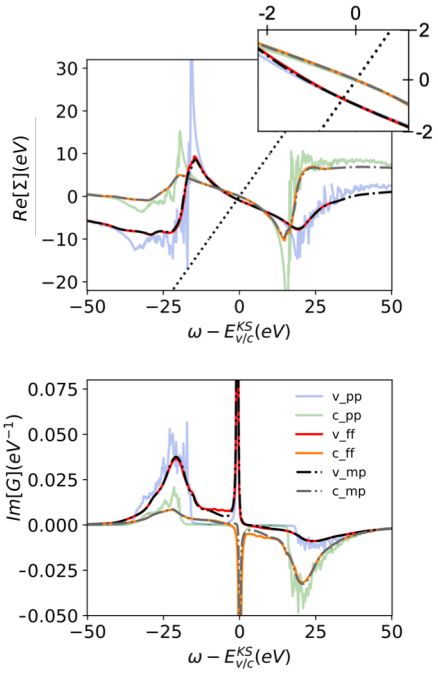
See also: https://www.tuni.fi/en/news/electrons-behave-tsunamis-findings-hold-promise-discovery-new-superconductors

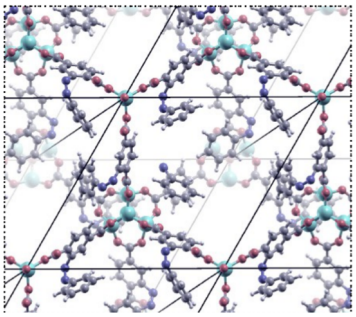
Frequency dependence in GW made simple using a multipole approximation - by D. A. Leon

The frequency integration in the evaluation of the self-energy is crucial to the deployment of the GW method. Full frequency (FF) approaches are accurate but computationally demanding, while the plasmon pole approximation (PPA), faster but limited in accuracy, is still commonly adopted.
We have explored [1] the analytic properties of the response function, X, and propose a multipole model (MPA) that naturally bridges from PPA to FF treatments. A key feature is the use of an optimal sampling of X in order to better interpolate the parameters of the model. The results are then used to obtain a simplified yet accurate description of W and to evaluate the GW self-energy in an efficient way. Overall, the proposed MPA technique leads to a level of accuracy comparable to FF methods at much lower cost. The MPA approach can be easily implemented in existing GW codes, making the method a good candidate to be potentially adopted by a large share of the theoretical spectroscopy community.



[1] D. A. Leon, C. Cardoso, T. Chiarotti, D. Varsano, E. Molinari and A. Ferretti, PRB 104, 115157 (2021)

Optical absorption spectra of MOF-5 and its photo-switching variant – by C. Attaccalite



Metal-organic frameworks (MOFs) have been attracting much attention in the past 20 years as possible candidate materials for a variety of applications, like in catalysis, gas capture and separation, drug delivery, or sensors. Among all families of MOFs, those with photoresponsive properties have been proposed as a potentially efficient technology for gas capture and release as light, possibly in the visible range can be used to change the gas adsorption properties of the material.

By employing the Bethe-Salpeter equation (BSE) in conjunction with the GW approximation we stuided the excited state properties of photoresponsive MOFs and their similarity with the molecular case.

We computed the optical absorption spectra of the *trans* and *cis* configurations using periodic and nonperiodic calculations with environment effects accounted for using a QM/MM approach. In the visible, near-UV, and mid-UV regions the optical excitations in the MOF are associated with the azobenzene functionalities and this results in spectral features similar to the case of gas-phase azobenzene and the azo-functionalized ligand. The most noticeable difference is the significantly more intense S1 band for *cis* in the MOF as compared to the free molecules, which points to a faster and more complete cis → trans isomerization in the framework, with strong implications for the design of MOFs with high photoconversion efficiencies. Consistent with these findings, all of the molecular models employed to represent the MOF are found to yield a reasonable description of the low-energy optical spectra between 2 and 5 eV of the periodic framework [1]. A similar result is found for MOF-5, the skeleton of the azobenzene-functionalized MOF, and one of the most studied MOFs. Our GW calculations show that MOF-5 is a wide gap insulator with a fundamental gap of 8 eV and not a semiconductor as identified in the last 15 year. The strong excitonic effects, arising from highly localized states and low screening in these materials, result in an optical gap of 4.5 eV and in an optical absorption spectrum in excellent agreement with recent experiments [2].

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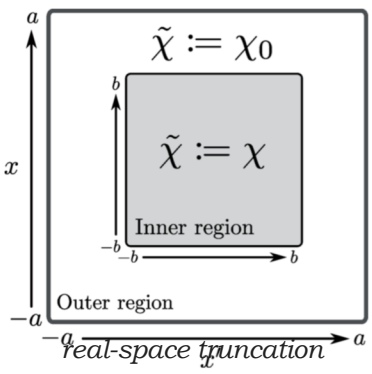
Exchange Correlation Kernel - by R. Godby

Nick Woods (Cambridge), Mike Entwistle (York/Berlin) and Rex Godby (York) have recently published two papers which employ robust numerical methods for determining the exact exchange-correlation kernel of TDDFT, fxc(x,x',ω) for prototype systems:

"Insights from exact exchange-correlation kernels",
https://doi.org/10.1103/PhysRevB.103.125155

"Accurate total energies from the adiabatic-connection fluctuation-dissipation theorem"
https://doi.org/10.1103/PhysRevB.104.125126



Upcoming Events

The **2021 Informal ETSF Workshop** is approaching.
Despite the pandemic situation still hinders in person meetings and travels, a significant group of ETSF members is going to meet in Coimbra to discuss challenging topics, open problems and future perpectives of ETSF.

All the info at the Webpage of the Workshop.

25th ETSF Workshop on Electronic Excitations: Fundamental challenges for theoretical spectroscopy from the frontier of technology to be held in Leuven, Belgium, from 13-17 June 2022

https://workshop.etsf.eu/

Deadline for *abstract submission* and *bursary applications*: **March 18th, 2022**

The 2022 edition of the workshop will include the traditional topics of interest of the ETSF community and focus on fundamental challenges for theoretical spectroscopy posed by cutting-edge present and future technologies, thereby promoting a fruitful exchange between academia and industry. The venue imec, an R&D hub for nano- and digital technologies, will serve as a gateway between industry and the academic world.

Topics covered by the workshop will include:

- Deep valence and upper core spectroscopy
- Time-resolved vibrational and electronic spectroscopy
- Dissipative quantum dynamics
- Photoresist radiation chemistry
- Multiscale and embedding methods
- Machine learning in theoretical spectroscopy

Introductory lectures:

- Geoffrey Pourtois, IMEC, Belgium
- Matthieu Verstraete, Université de Liège, Belgium

Invited speakers (theory):

- Alex Chin, Sorbonne Université, France
- Dorothea Golze, TU Dresden, Germany
- Miquel Huix-Rotllant, Aix Marseille Université, France
- Milica Todorovic, University of Turku, Finland
- Mariana Rossi, Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany
- Francesco Sottile, École Polytechnique, France
- Nicolas Tancogne-Dejean, Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany
- Vladislav S. Yakovlev, Max Planck Institute of Quantum Optics, Germany

Theoretical Spectroscopy Lectures - CECAM school - Lausanne, 21 March 2022.

The theoretical spectroscopy lectures are a core ETSF training event, since their creation, in 2006. Running every two years, they aim at introducing the basic concepts of electronic excitations and spectroscopy, from the ab initio theoretical point of view. The format is typical for this schools, with theoretical concepts in the morning, while the afternoons are devoted to hands-on with the different codes. After a brief reminder of Density Functional Theory, the courses will deal with Time Dependent DFT, Green's functions theory (GW approximation and solution of the Bethe-Salpeter equation), and non-linear approaches. The codes used during the hands-on are: Abinit (for ground state and GW calulations), DP (for TDDFT), EXC (for the BSE), 2light and Lumen (for non-linear spectroscopy within TDDFT and BSE, respectively). The next event will take place in Lausanne, on 21-25 March 2022. Among the novelties of this edition, an introduction of the experimental point of view in many spectroscopies and a detailed view on magnetic sytems. The organisers are Francesco Sottile (Ecole Polytechnique, Palaiseau), Valerio Olevano (Institut Neel, Grenoble) and Gian-Marco Rignanese (Universite de Louvain-la-Neuve). https://www.cecam.org/workshop-details/63

The **International Summer school in electronic structure Theory: electron correlation in Physics and Chemistry (ISTPC)** is ready for its 3rd edition. It is a two-week school that will take place in Aussois (Savoie, France) from the 19th of June to the 2nd of July 2022.
Pina Romaniello, for a long time responsible for the correlation team, is among the organisers. Other ETSF members will give lectures in this school (Francesco Sottile, Matteo Gatti, Fabien Bruneval). The topics covered are multiple: Second quantization, density functional theory (DFT), time-dependent DFT (TD-DFT), Hartree-Fock (HF) and post-HF methods, multi-configurational methods, multi-reference perturbation theory, model Hamiltonians, linear response theory, basics in solid-state physics, Green functions, Quantum Monte Carlo (QMC), density matrix renormalization group (DMRG), Random Phase Approximation (RPA), GW method, Bethe-Salpeter equation, Dynamical Mean Field Theory (DMFT), Mathematical aspects of electronic structure theory, reduced quantity functional theories. The school is supported by two French national research groups on electronic excitations and correlation: NBODY (https://wiki.lct.jussieu.fr/gdrnbody) and REST (http://gdr-rest.polytechnique.fr). The latter, in particular has a long-standing collaboration with the ETSF, via common seminars (next one will be January's webinar) and events, like the discussion meeting on electron correlation (https://www.etsf.eu/collaboration-teams/correlation-ct) and the one on Green's functions (http://gdr-rest.polytechnique.fr/functionals_greens_functions).
Important: since the school could not take place physically in 2021, it was, yes, postoponed to 2022, but a condensed version took place nontheless over zoom.
Materials and recordings of this event are accessible at this page:
https://quantique.u-strasbg.fr/ISTPC/doku.php?id=istpc2021:videoslides.
More info about registration for the 2022 edition, at the webpage of the school:
https://quantique.u-strasbg.fr/ISTPC/.

DPG Spring Meeting Frontiers of Electronic-Structure Theory: Focus on Artificial Intelligence applied to Real Materials

A symposium at the DPG Spring meeting, Regensburg, 6-11 March 2022 Abstract submissions for contributed talks and posters are open until December 22, 2021 https://www.dpg-tagung.de/r22/submission.html?language=en

Funding Opportunities

- COST-action TUMIEE (TOWARDS UNDERSTANDING AND MODELLING INTENSE ELECTRONIC EXCITATION) will close in September 2022, there are still money available of short term scientific missions https://www.cost-ca17126.industriales.upm.es/

- A more extensive list of funding opportunities of interest for ETSF community is reported in the webpage

News from Young Researchers

York:

Mike Entwistle is now a postdoc at FU Berlin.

Jack Wetherell, following a postdoctoral position in Lucia Reining's group, is now a data scientist at humn.ai.

Matt Hodgson has returned to York after postdoctoral spells in Halle and Durham, and is an Associate Lecturer, teaching courses in advanced theoretical techniques and computational continuum physics, and also leading the third-year advanced computational laboratory.

Toulouse:

Gabriele Riva and **Roberto Orlando** attended the last YRM and are now at the second year of their PhD. **Stefano di Sabatino** has started his second post-doc with Pina Romaniello.

Milan:

Marco Marino has recently joined the Solid State Theory Physics Group in Milan as PhD, working on photo-induced magnetic interaction betweeen organic molecules and antiferromagnetic substrates.

Strategy to improve the ETSF network

The ETSF Steering Committee has envisioned some actions on which Research Team Leaders and members should invest to strenghten the ETSF community:

- Promote **collaborations** between groups through existing or newly created *Collaboration Teams*
- Improve the **communication** inside the network through Newsletter, social media, meetings..
- Involve the **young researchers** (remember to *sign up for membership* in the ETSF Website!)
- Start collaborative actions to **get funding as ETSF (COST, Marie Curie...)**.
- Think at **ETSF as a live entity**, proposing new Research Team Leaders and research lines and considering how the individual research initiatives can spill over into the network activity.

How to **contribute** to ETSF Newsletter Fill the dedicated form
Write an email to simona.achilli@unimi.it