

## HL 91: Joint Focussed Session: Theory and Computation of Electronic Structure: New Frontiers VIII

Time: Friday 11:15–12:45

Location: TRE Phy

### Topical Talk

HL 91.1 Fri 11:15 TRE Phy  
**Tunable bandgaps and excitons in doped semiconducting carbon nanotubes made possible by acoustic plasmons** — ●CATALIN SPATARU and FRANCOIS LEONARD — Sandia National Laboratories, Livermore, California, USA

Doping of semiconductors is essential in modern electronic and photonic devices. While doping is well understood in bulk semiconductors, the advent of carbon nanotubes and nanowires for nanoelectronic and nanophotonic applications raises some key questions about the role and impact of doping at low dimensionality. Here we show that for semiconducting carbon nanotubes, bandgaps and exciton binding energies can be dramatically reduced upon experimentally relevant doping, and can be tuned gradually over a broad range of energies in contrast to higher dimensional systems. The later feature, made possible by a novel mechanism involving acoustic plasmons, establishes new paradigms for the understanding and design of nanoelectronic and nanophotonic devices.

HL 91.2 Fri 11:45 TRE Phy  
**Electronic Excitations in Single-Wall Carbon Nanotubes: Building-Block Approach** — ●RALF HAMBACH<sup>1,2</sup>, CHRISTINE GIORGETTI<sup>1,2</sup>, XOCHITL LOPEZ-LOZANO<sup>3</sup>, and LUCIA REINING<sup>1,2</sup> — <sup>1</sup>LSI, Ecole Polytechnique, CNRS, CEA/DSM, Palaiseau, France — <sup>2</sup>European Theoretical Spectroscopy Facility — <sup>3</sup>University of Texas at San Antonio, United States

Parameter-free calculations of electron energy-loss spectra for low-dimensional systems like single-wall carbon nanotubes can become numerically very demanding or even unfeasible for large diameters.

We overcome this problem by means of a building-block approach: Combining effective-medium theory and ab-initio calculations we can describe the collective excitations in nanostructures (like carbon nanotubes) starting from the microscopic polarisability of their building blocks (bulk graphite). To this end, Maxwell's equations are solved using the full frequency- and momentum-dependent microscopic dielectric function  $\epsilon(\mathbf{q}, \mathbf{q}', \omega)$  of the bulk material. The latter is calculated from first principles within the random phase approximation [1].

Besides an important gain in calculation time this method allows us to analyse the loss spectra of nanostructures in terms of their normal-mode excitations. We apply the building-block approach to study angular-resolved loss spectra for graphene and single-wall carbon nanotubes and find a very good agreement with full ab-initio calculations of these systems and corresponding experiments.

[1] AbInit: [www.abinit.org](http://www.abinit.org), DP-code: [www.dp-code.org](http://www.dp-code.org)

HL 91.3 Fri 12:00 TRE Phy  
**Functionalized Tips Leading to Atomic-Resolution Force Microscopy** — ●NIKOLAJ MOLL, LEO GROSS, FABIAN MOHN, ALESSANDRO CURIONI, and GERHARD MEYER — IBM Research – Zürich, Säumerstrasse 4, CH-8803 Rüschlikon, Switzerland

Performing atomic force microscopy (AFM) with a molecule or an atom at the tip the resolution can be dramatically enhanced as the resolution crucially depends on the chemical nature of the tip termination. A pentacene molecule is imaged with atomic resolution with a tip functionalized with a CO molecule. The interactions between the CO tip and the pentacene are studied with first principles calculations. The different energy contributions are analyzed, and the Pauli energy is computed. The source of the high resolution is Pauli repulsion, whereas van-der-Waals and electrostatic interactions only add a diffuse attractive background. To validate the usefulness of AFM with functionalized tips the natural product cephalandole A is studied. The measurements together with first principle calculations demonstrate that the direct imaging of an organic compound with AFM facilitates

the accurate determination of its chemical structure. The method might be developed further towards molecular imaging with chemical sensitivity, and could solve certain classes of natural product structures.

[1] Leo Gross, Fabian Mohn, Nikolaj Moll, Peter Liljeroth, and Gerhard Meyer, *Science* 325, 1110-1114 (2009).

[2] Leo Gross, Fabian Mohn, Nikolaj Moll, Gerhard Meyer, Rainer Ebel, Wael M. Abdel-Mageed, and Marcel Jaspars, *Nat. Chem.* 2, 821-825 (2010).

HL 91.4 Fri 12:15 TRE Phy  
**Impact ionization rates from ab initio calculations** — ●MARTON VOROS<sup>1</sup>, DARIO ROCCA<sup>2</sup>, GERGELY ZIMANYI<sup>3</sup>, GIULIA GALLI<sup>2,3</sup>, and ADAM GALI<sup>1,4</sup> — <sup>1</sup>Budapest University of Technology and Economics, Department of Atomic Physics — <sup>2</sup>UC Davis, Department of Chemistry — <sup>3</sup>UC Davis, Department of Physics — <sup>4</sup>Hungarian Academy of Sciences, Research Institute of Solid State Physics and Optics

Achieving multi exciton generation (MEG) in semiconducting nanocrystals may lead to overcome the well-known Shockley-Queisser limit when building semiconductor-based solar cells. A thorough, theoretical understanding of the experiments that reported MEG in e.g. Si and PbSe nanocrystals, is still missing and could significantly contribute to clarify the several controversial results in the field. Several theoretical and numerical studies have addressed the origin of the MEG formation, mostly supporting an impact ionization mechanism. However, impact ionization rates have only been evaluated for model nanocrystals by using empirical pseudopotentials fitted to bulk properties or by applying tight binding wavefunctions, and model dielectric functions to describe the screened Coulomb interaction. We present a full ab-initio scheme based on Density Functional Theory in a plane-wave pseudopotential implementation that includes static screening within the random-phase approximation. As a first application, we will discuss how impact ionization rates are affected by the size of small Si nanocrystals.

HL 91.5 Fri 12:30 TRE Phy  
**Selective Excitation of Molecular Vibrations by Tunneling Electrons** — ●JESSICA WALKENHORST<sup>1</sup>, MARIUS WANKO<sup>1</sup>, ALBERTO CASTRO<sup>2</sup>, and ANGEL RUBIO<sup>1</sup> — <sup>1</sup>Nano-Bio Spectroscopy group and ETSF Scientific Development Centre, Dpto. Física de Materiales, Universidad del País Vasco, Centro de Física de Materiales CSIC-UPV/EHU-MPC and DIPC, San Sebastián, Spain — <sup>2</sup>Institute for Biocomputation and Physics of Complex Systems (BIFI), University of Zaragoza, Spain

Tunneling electrons can be used to excite vibrations in molecules. By combining scanning electron microscopy (STM) with inelastic electron tunneling spectroscopy (IETS) one can obtain vibrational spectra of single molecules adsorbed on a surface. Interestingly in large molecules the vibrational spectrum depends on the impact point of the electrons on the molecular surface. Selective excitation of vibrational modes by the STM tip has been proposed to explain these experimental findings (the so-called 'excitation of local vibrations' model). Therefore, we want to simulate the dynamical response of Schiff base (CNH4+) to tunneling electrons. To this end, we perform molecular dynamics simulations based on time-dependent density functional theory (TDDFT). Vibrational spectra are then obtained by Fourier analysis of the velocity autocorrelation function. This way, we study the relative strength of the excited molecular vibrations in dependence on the impact point of the tunneling electrons. Finally, a comparison between numerical results and experimental findings is attempted and the validity of the 'excitation of local vibrations' model is discussed.