WORKSHOP #2

Electronic Structure of Nanomaterials: A Special Symposium in Honor of Dr. Mark Hybertsen

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This workshop has two goals. First, we will discuss recent progresses in first-principles modeling of nanomaterials, including exciting works in both methodology development and applications in real materials. Electronic structure methods have played a central role in fundamental understanding of the physical and chemical properties of nanomaterials and providing principles for materials design in photovoltaics, energy storage, catalysis, and quantum sensing / computing, to name a few. The workshop will bring together scientists working at the frontier of condensed matter physics, chemistry and materials science to seek for viable pathways to expedite the science at NSLS-II and CFN using theory and modeling.

This workshop will also be held in honor of the scientific contributions of Dr. Mark Hybertsen, especially his efforts to bridge theory and experiments in nanomaterials research. Dr. Hybertsen retired in early 2022 as the group leader of the Theory and Computation Group at CFN, after working at BNL for 15 years. He is widely recognized for his pioneering work in applying many-body perturbation theory to band structure in solids. His early work on semiconductors and insulators overcame the well-known "band gap problem" in density functional theory and demonstrated for the first time an excellent agreement with experimental data. The methodology developed by Hybertsen and Louie has since become the golden standard for obtaining accurate band structure of weakly correlated materials. Throughout his career, Dr. Hybertsen has made tremendous contributions to semiconductor physics and nanoscience research in a broad range of topics, including transport in nanojunctions, excitons in 2D materials, and defect physics, all in close collaborations with experimental colleagues. As a group leader of the Theory and Computation facility, he has dedicated his expertise to the CFN User Program and has supported many user collaborators with significant intellectual input. This special symposium will celebrate Mark's impact on both the scientific community and the CFN User Facility.

People are welcome to leave message to Mark using the online Kudoboard (https://www.kudoboard.com/boards/P0LA9EmS).

Start Time (ET)	Title	Speaker (Affiliation)
10:00 – 10:10 am	Opening Remarks	Charles Black
10:10 – 10:40 a.m.	Shift and Ballistic Currents from First Principles	Andrew Rappe University of Pennsylvania
10:40 – 11:10 a.m.	Exciton Complexes in Lead-halide Perovskites	Timothy C. Berkelbach Columbia University
11:10 – 11:40 a.m.	Towards 1D Topological Insulators	Latha Venkataraman Columbia University
11:40 – 12:10 p.m	Massively Parallel Cubic-scaling GW Calculations with the OpenAtom Software	Sohrab Ismail-Beigi Yale University
12:10-12:30 p.m.	LUNCH BREAK	
12:30 – 1:10 p.m.	Good Wishes to Mark	

1:10 – 1:40 p.m.	Defects in Materials for Energy Storage and Generation	Chris Van de Walle UC Santa Barbara
1:40 – 2:10 p.m.	Phonon, carrier, and cation screening of excited states in complex materials	Jeff Neaton Lawrence Berkeley National Laboratory and UC Berkeley
2:10 – 2:40 p.m.	KEYNOTE TALK: Single- and multi-particle excitations and time-dependent phenomena in nanomaterials: GW, GW-BSE, and beyond	Steve Louie UC Berkeley and Lawrence Berkeley National Laboratory
2:40 – 3:00 p.m.	COFFEE BREAK	
3:00 – 3:30 p.m.	Moire Physics in Layered Materials	Efthimios Kaxiras Harvard University
3:30 – 4:00 p.m.	Cooperative Interactions Between Surface Terminations Explain Photocatalytic Water Splitting Activity on SrTiO3	Marivi Fernandez-Serra Stony Brook University
4:00 – 4:30 p.m.	Catalysts for Selective CO2 Reduction: from Fundamental Understanding toward Rational Design	Ping Liu Brookhaven National Laboratory
4:30 – 5:00 p.m.	TBD	Tony Heinz Stanford University
5:00 – 5:10 p.m.	Closing Remarks	Mark Hybertsen

Abstracts

Shift and Ballistic Currents from First Principles

Andrew Rappe

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As the need for clean, safe, and sustainable energy increases, renewed focus on alternative energy sources such as photovoltaics and next-generation computing have become vital. This motivates study of the bulk photovoltaic effect (BPVE), a nonlinear optoelectronic property that can generate electricity without a p-n junction. To demonstrate the capability of first-principles BPVE theories to guide materials design, we outline an automated method to design distortions that enhance the shift current of monolayer MoS2 and use it to uncover a polar distortion that increases the integrated shift current more than ten-fold. Because the distortion can be driven by a static electric field via the converse piezoelectric effect, this finding shows that electric fields can be used to engineer the shift current response of a material and complements previous work showing that mechanical strain can also modulate the shift current response.

The ab initio methods commonly used to model the BPVE include only the shift current contribution to the BPVE. They have enabled significant increase in our understanding of the BPVE, but only explain part

of the experimentally observed photocurrent. To overcome this deficiency, we present a method that enables the ballistic current—a current resulting from asymmetric scattering—from first principles. We use a perturbative approach to express the ballistic current due to electron-phonon and electron-hole scattering in a form amenable for ab initio calculation, and then calculate the ballistic current for $BaTiO_3$ from first principles. The current due to electron-phonon scattering is comparable to the shift current, and is therefore experimentally relevant, while the current due to electron-hole scattering is much smaller in magnitude. This methodological development enables closer agreement between theory and experiments and lays the groundwork for further prediction and design of materials with large BPVE.

Related publications

[1] Z. Dai, A. M. Schankler, L. Gao, L. Z. Tan, and A. M. Rappe, "Phonon-Assisted Ballistic Current from First-Principles Calculations", Phys. Rev. Lett. 126, 177403 (2021).

[2] Z. Dai and A. M. Rappe, "First Principles Calculation of Ballistic Current from Electron-Hole Interaction", Phys. Rev. B 104, 235203 (2021).

[3] A. M. Schankler, L. Gao, and A. M. Rappe, "Large Bulk Piezophotovoltaic Effect of Monolayer 2H-MoS_2", J. Phys. Chem. Lett. 12, 1244 (2021).

(Keynote talk) Single- and multi-particle excitations and time-dependent phenomena in nanomaterials: GW, GW-BSE, and beyond

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Many phenomena in nature owe their emergence from interactions of a large number of particles. In particular, many-electron interactions are often dominant in excited-state phenomena of materials. We present in this symposium, in honor of Dr. Mark Hybertsen's career, some of our current understanding of excited-state properties of nanomaterials within the ab initio interacting Green's function approach using many-body perturbation theory. Different physical measurements and phenomena are connected to the different kinds of interacting Green's functions. Photoemission spectroscopy may be understood from the perspective of an interacting 1-particle Green's function (e.g., within the GW approximation for the electron self-energy). However, correlated multi-particle excitations such as excitons, trions, and bi-excitons require knowledge of the 2-, 3- and 4-particle interacting Green's functions, respectively. Moreover, nonequilibrium and time-dependent phenomena, such as field-driven phenomena and pump-probe experiments, require yet very different treatments from those of the equilibrium case. We discuss the concepts and methods behind the ab initio studies of these phenomena and give some illustrative examples including single- and multi-particle excitations, linear and nonlinear optical responses, as well as field-driven time-resolved and angle-resolved photoemission spectroscopy (tr-ARPES) computed using the BerkeleyGW code.