Symposium Progress and Challenges in Modelling Electron-Phonon Interaction in Solids (SYIS)

jointly organised by
the Semiconductor Physics Division (HL),
Crystalline Solids and their Microstructure Division (KFM), and
Surface Science Division (O)

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Electron-phonon interaction is crucial to predict and explain material behaviour under a variety of equilibrium and non-equilibrium conditions. Though recent developments in theoretical and computational methods have significantly advanced our capability, significant challenges still remain to be addressed. An accurate description of electron-phonon interactions in complex materials, including e.g., disordered perovskites, correlated oxides, superconductors, interfaces, and heterostructures, is often beyond the capability of existing methodologies. This symposium will bridge different communities working on electron-phonon interactions in solids and pinpoint common problems and open challenges in the field.

Overview of Invited Talks and Sessions

(Lecture hall H1)

Invited Talks

SYIS 1.1	Tue	9:30–10:00	H1	**Electron-phonon and exciton-phonon coupling in advanced materials** — •Claudia Draxl
SYIS 1.2	Tue	10:00–10:30	H1	**Exciton-phonon dynamics from first principles** — •Enrico Perfetto
SYIS 1.3	Tue	10:30–11:00	H1	**Polarons and exciton polarons from first principles** — •Feliciano Giustino
SYIS 1.4	Tue	11:15–11:45	H1	**Wannier-Function-Based First-principle Approach to Coupled Exciton-Phonon-Photon Dynamics in Two-Dimensional Semiconductors** — •Alexander Steinhoff
SYIS 1.5	Tue	11:45–12:15	H1	**Phonon influence on (cooperative) photon emission from quantum dots** — •Erik Gauger

Sessions

SYIS 1.1–1.5	Tue	9:30–12:15	H1	**Progress and Challenges in Modelling Electron-Phonon Interaction in Solids**

SYIS 1: Progress and Challenges in Modelling Electron-Phonon Interaction in Solids

Time: Tuesday 9:30–12:15 Location: H1

Invited Talk SYIS 1.1 Tue 9:30 H1

Electron-phonon and exciton-phonon coupling in advanced materials — •Claudia Draxl — Humboldt-Universität zu Berlin, Berlin, Germany

Treating various excitations in materials on equal footing allows us not only to achieve excellent agreement with experiment, but, importantly, to gain a deeper understanding of complex processes and materials. Prominent examples are organic-inorganic hybrid systems whose building blocks are very different in nature. I will discuss the critical role of both the mutual dynamical screening of the constituents and electron-phonon coupling (EPC) [1] to obtain the correct ground state and excitation spectra. Many-body perturbation theory (MBPT) is the state-of-the-art methodology for such problems. In particular, the Bethe-Salpeter equation (BSE) is the method of choice for computing optical excitations. For polar materials, the dielectric screening of both the electronic and vibrational degrees of freedom must be considered to obtain correct exciton binding energies and spectral features [2]. Finally, I will show, how the BSE together with EPC matrix elements can be used to treat exciton-phonon coupling and thus geometry relaxation in the excited state [3], giving a handle to the description of photoluminescence and exciton dynamics.

[1] I. Gonzalez Oliva, B. Maurer, B. Alex, S. Tillack, M. Schebek, and C. Draxl, phys. stat. sol. (a) 221, 2300170 (2024).

[2] M. Schebeck, P. Pavone, C. Draxl, and F. Caruso, J. Phys.: Condens. Matter (2024); https://arxiv.org/abs/2409.15099

[3] M. Yang and C. Draxl, https://arxiv.org/abs/2212.13645

Invited Talk SYIS 1.2 Tue 10:00 H1

Exciton-phonon dynamics from first principles — •Enrico Perfetto — Physics Department University of Rome Tor Vergata

Exciton dynamics, encompassing ultrafast photogeneration, diffusion, and thermalization, plays a fundamental role in optoelectronic, photovoltaic, and photocatalytic processes. In this talk we discuss a novel many-body approach to describe exciton dynamics from first-principles. We show that the introduction of an auxiliary exciton species, termed 'irreducible exciton', is crucial to formulate a theory free from overscreening of the electron-phonon interaction. The resulting Excitonic Bloch Equations, while having the same computational cost as the well-known Excitonic Boltzmann Equations, enable a comprehensive description of the temporal evolution of coherent, irreducible, and incoherent excitons during and after the optical excitation.

Within this framework, we explore the real-time dynamics of exciton formation, elucidating the mechanism by which quasi-free electron-hole pairs, generated by above-gap photoexcitation, are dynamically converted into bound excitons.

Invited Talk SYIS 1.3 Tue 10:30 H1

Polarons and exciton polarons from first principles — •Feliciano Giustino — The University of Texas at Austin

Polarons are quasiparticles formed when a charge carrier interacts with lattice vibrations. In materials with strong electron-phonon couplings, this phenomenon results in self-trapped polarons. Similarly, excitons, which are composite quasiparticles formed by the binding of an electron and a hole, can polarize the surrounding crystal lattice through spatial fluctuations in their charge density. This polarization, in turn, can promote the spatial localization of the exciton, leading to the formation of exciton polarons or even self-trapped excitons in the presence of strong exciton-phonon couplings. First-principles calculations of these effects are challenging because they require large supercells potentially involving hundreds or thousands of atoms. In this talk, I will discuss recent methodological developments that combine density-functional perturbation theory and the Bethe-Salpeter approach to compute polarons and exciton polarons from first principles. The main advantage of the present approach is that it does not require supercells, and all necessary information is generated via calculations in the crystal unit cell. To illustrate these developments, I will report on two recent applications: (i) the discovery of topological polarons in halide perovskites, wherein the distortion of the atomic lattice describes a vector field with definite topological invariants; and (ii) the discovery of large polarons and exciton polarons in rutile and anatase titanium dioxide, which provide a natural explanation for why anatase exhibits diffusive carrier transport while rutile supports thermally-activated transport.

15 min. break

Invited Talk SYIS 1.4 Tue 11:15 H1

Wannier-Function-Based First-principle Approach to Coupled Exciton-Phonon-Photon Dynamics in Two-Dimensional Semiconductors — •Alexander Steinhoff[1], Matthias Florian[2], and Frank Jahnke[1] — [1]Institut für Theoretische Physik, Universität Bremen, Bremen, Germany — [2]Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, USA

Marrying the predictive power of ab initio calculations with many-body effects of ever increasing complexity remains a challenging task. In particular, for van der Waals materials, understanding carrier-phonon interaction from first principles is a field of growing interest. Here, we present a many-body theory for coupled free-carrier, exciton, phonon and photon dynamics based on carrier-carrier and carrier-phonon interaction matrix elements obtained from first principles via projection on Wannier orbitals. We demonstrate the impact of carrier-two-phonon scattering processes on optical spectra and coupled nonequilibrium carrier-phonon kinetics in monolayer $MoSe_2$. Our studies open a perspective to advance the material-realistic description of nonequilibrium physics in two- dimensional nanostructures to new many-body levels.

Invited Talk SYIS 1.5 Tue 11:45 H1

Phonon influence on (cooperative) photon emission from quantum dots — •Erik Gauger[1], Julian Wiercinski[1], and Moritz Cygorek[2] — [1]Institute of Photonics and Quantum Sciences, Heriot Watt University — [2]Technische Universität Dortmund

Semiconductor quantum dots (QDs) provide an established on-demand single-photon source platform. Integrating such sources into quantum networks comes with the requirement of producing fundamentally indistinguishable photons, and the ability to generate and preserve coherence between different emitters.

The leading source of decoherence of solid state emitters such as quantum dots is typically their coupling to longitudinal acoustic lattice vibrations, as can be observed under coherent control experiments such as Rabi oscillations as well as in their emission resonance spectrum.

In this presentation, I will give an overview of our theoretical work on modelling, understanding, and mitigating the interaction between solid-state emitters and their vibrational environment, and how this interaction manifests in measurable signatures such as photon statistics and spectral properties.

With a view towards scaling up to networks of coupled emitters, a particular focus of this presentation will be our recent progress in understanding the interplay between collective light-matter coupling and phonon decoherence, by utilising intuitive and interpretable master equation approaches as well as state-of-the-art numerical process tensor methods. I will also make connections to experimental data for up to five quantum dots displaying signatures of cooperative behaviour.