Q 37: Polaritonic Effects in Molecular Systems II (joint session MO/Q)

Time: Wednesday 11:00–13:00   Location: HS XV

Q 37.1 Wed 11:00 HS XV
Boundary conditions and violations of bulk-edge correspondence in a hydrodynamic model — Gian Michele Graf[1] and •Alessandro Tarantola[1,2] — [1]Institut für Theoretische Physik, Wolfgang-Pauli-Str. 27, 8093 Zürich, Switzerland — [2]German Aerospace Center (DLR), Institute of Quantum Technologies, 89081 Ulm, Germany

Bulk-edge correspondence is a wide-ranging principle that applies to topological matter. According to the principle, the distinctive topological properties of matter, thought of as extending indefinitely in space, are equivalently reflected in the excitations running along its boundary. Indices encode those properties, and their values, when differing, are witness to a violation of that correspondence. We address such violations, as they occur in a hydrodynamic context. The model concerns a shallow layer of fluid in a rotating frame and provides a local description of waves propagating either across the oceans or along a coastline; it becomes topological when suitably modified at short distances. The edge index is sensitive to boundary conditions, hence exhibiting a violation. Here we present a classification of all (local, self-adjoint) boundary conditions. They come in four families, distinguished in part by the degree of their underlying differential operators. Generally, both the correspondence and its violation are typical. Across families though, the maximally possible amount of violation can vary with their degree. Several indices of interest are charted for all boundary conditions. A single spectral mechanism for the onset of violations is furthermore identified, and the role of a symmetry investigated.

Q 37.2 Wed 11:15 HS XV
Cavity-mediated electron-electron interactions: Renormalizing Dirac states in graphene — •Hang Liu[1], Francesco Troisi[1], Hannes Huebener[1], Simone Latini[1,2], and Angel Rubio[1,3] — [1]Max Planck Institute for the Structure and Dynamics of Matter, Germany — [2]Technical University of Denmark, Denmark — [3]The Flatiron Institute, USA

Accurately modeling the interaction between electrons in materials and photon modes within dark cavities is crucial for predicting and understanding cavity-induced phenomena. In this work, we developed the photon-free quantum electrodynamics Hartree-Fock and configuration-interaction frameworks to model the coupling between electrons in crystalline materials and cavity photon modes. We applied these theoretical approaches to investigate the graphene coupled to different types of cavity modes. For a circularly polarized mode, a topological Dirac gap emerges due to cavity-mediated local and nonlocal electron interactions. In contrast, a linearly polarized mode induces a topologically trivial Dirac gap as a result of the cavity-mediated nonlocal electron interactions. Notably, when two cavity modes are introduced, the Dirac cones can remain gapless, but the Fermi velocity is renormalized through cavity-induced nonlocal electron interactions. Our nonperturbative approaches can capture the critical role of cavity-induced nonlocal electron-electron interactions in renormalizing Dirac states in graphene. These new theoretical frameworks pave the way for accurately predicting and exploring novel cavity-induced phenomena in a broader range of material systems.

Q 37.3 Wed 11:30 HS XV
Quantum algorithms for QED systems — •Francesco Troisi[1], Simone Latini[2], Heiko Appel[1], Ivano Tavernelli[4], and Angel Rubio[1,3] — [1]MPSD, Hamburg, Germany — [2]Department of Physics, DTU, Lyngby, Denmark — [3]CCQ, Flatiron Institute, Simons Foundation, NYC, USA — [4]IBM Quantum, IBM Research, Zurich, Säumerstrasse 4, 8803 Rüschlikon, Switzerland

Controlling the properties of matter is a central theme in modern science. Optical cavities provide a promising approach to controlling them by coupling the electronic transitions to the confined photons inside the cavity, making the photonic and electronic states inseparable. The polaritonic states are obtained, which due to the strong coupling regime, cannot be described by the perturbative approach. On a classical computer, this introduces big computational challenges as the QED matrix grows exponentially with the number of photonic modes and Fock states. Quantum Computing is a promising tool for studying such systems as adding one cavity mode requires as little as one qubit. Due to the complexity of the physics in materials or com-

plex molecules, we approach the cavity QED problem with a simpler system, such that we can learn the challenges in a controlled environment. In this work we couple a two-level matter system to many cavity modes, and we focus on studying a well-known physical phenomenon, the spontaneous emission, where excited atoms emit photons upon returning to their ground state. Despite its simplicity, one can still observe many features such as the Rabi oscillations and the decay rate making it an ideal candidate for approaching QED problems.

Q 37.4 Wed 11:45 HS XV
Control of cavity dissipations across the insulator-to-metal transition in 1T-TaS$_2$ — •Giacomo Jarc[1], Angela Montanaro[1], Shahla Yasmin Mathengattil[2,3], Enrico Rigoni[1,3], and Daniele Fausti[1] — [1]Department of Physics, FAU Erlangen-Nürnberg Erlangen, Germany — [2]Department of Physics, Università di Trieste, Trieste, Italy — [3]Elettra Sincrotrone, Basovizza (Trieste), Italy

Using optical cavities resonant with material excitations enables controlling light-matter interaction in both the regimes of weak and strong coupling. We study here the coupling of low-energy excitations in the charge-density-wave (CDW) material 1T-TaS$_2$ across its insulator-to-metal transition when embedded into tetahertz Fabry-Pérot cryogenic cavities. In the dielectric state, we reveal the signatures of a multimode vibro-polariton mixing, with the polariton modes inheriting character from all the CDW phonons as a consequence of the cavity-mediated hybridization. The multimode vibrational strong coupling is suppressed across the insulator-to-metal transition as a consequence of the optical dissipations introduced by the free charges, and a vibrational weak coupling regime is observed in proximity of the phase transition. When the cavity frequency is tuned within the spectral range of the continuum Drude excitation, we reveal that the quality factor of the cavity, which quantifies the dissipations of the coupled system, decreases passing from the insulating to the metallic state. Our evidences points to a scenario in which the free charges can effectively couple to the cavity field and subsequently modify the collective light-matter coupling.

Q 37.5 Wed 12:00 HS XV
Chirality and Dimensionality in the Ultrastrong Light-matter Coupling Regime — •Rémi Avriller[1] and Cyriaque Genet[2] — [1]University of Bordeaux, CNRS, LOMA, UMR 5798, F-33405 Talence, France. — [2]University of Strasbourg and CNRS, CESQ and ISIS, UMR 7006, F-67000 Strasbourg, France

We unveil the key-role of dimensionality in describing chiroptical properties of molecules embedded inside an optical Fabry-P*erot cavity.

For a 2D-layer configuration, we show that the interplay between molecular chirality and spatial dispersion of the cavity-modes, results in a gyrotropic coupling at the origin of a differential shift in polaritonic energy spectra. This differential shift is proportional to the gyrotropic coupling, while for 3D bulk-aggregate configurations it is shown to vanish.

We interpret physically the former 2D-chiral effect by analogy with the classical Newtonian motion of a fictitious particle in presence of 3D restoring force, and static magnetic field. The gyrotropic coupling is shown to directly perturbate the anholonomy angle of the classical trajectories, and the fictitious particle undergoes cyclotron gyrations upon entering the ultrastrong light-matter coupling regime.

Q 37.6 Wed 12:15 HS XV
The complex interplay of collectivity, locality and temperature in polaritonic chemistry — •Dominik Sidler[1,2,3], Michael Ruggenthaler[2,3], Jacob Horak[2,3], Thomas Schnappinger[4], and Angel Rubio[2,3,5] — [1]Paul Scherrer Institut, Villigen, Switzerland — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany — [3]The Hamburg Center for Ultrafast Imaging, Hamburg, Germany — [4]Stockholm University, Stockholm, Sweden — [5]The Flatiron Institute, New York, USA

Despite significant theoretical progress over the past years, still, no consensus has been achieved about the physically relevant mechanism in polaritonic chemistry. Based on ab initio simulations and analytic results, we will explore and identify physical mechanisms that shine light on the interplay of collective strong coupling with local chemical changes. For its detailed microscopic understanding, degeneracies and

cavity-induced local polarization patterns seem to play a crucial role. To capture those effects accurately, a fully self-consistent description is vital, since perturbation theory can lead to qualitatively erroneous predictions. Eventually, we demonstrate that the thermal statistic is altered non-trivially by collective strong coupling in optical cavities. Therefore, novel computational methods are required to simulate polaritonic chemistry accurately.

Q 37.7   Wed 12:30   HS XV

Controlling Plasmonic Catalysis via Strong Coupling with Electromagnetic Resonators — Jakub Fojt, Paul Erhart, and ●Christian Schäfer — Department of Physics, Chalmers University of Technology, 412 96 Göteborg, Sweden

Plasmonic excitations decay within femtoseconds, leaving nonthermal (often referred to as *hot*) charge carriers behind that can be injected into molecular structures to trigger chemical reactions that are otherwise out of reach - a process known as plasmonic catalysis. In this talk, we demonstrate that strong coupling between resonator structures and plasmonic nanoparticles can be used to control the spectral overlap between the plasmonic excitation energy and the charge injection energy into nearby molecules. Our atomistic description couples real-time density-functional theory self-consistently to an electromagnetic resonator structure via the radiation-reaction potential [1,2]. Control over the resonator provides then an additional knob for nonintrusively enhancing plasmonic catalysis [3], here more than 6-fold, and dynamically reacting to deterioration of the catalyst - a new facet of modern catalysis.

[1] C. Schäfer and G. Johansson, PRL 128, 156402 (2022). [2] C. Schäfer, J. Phys. Chem. Lett. 2022, 13, 6905-6911. [3] J. Fojt, P. Erhart, C. Schäfer, Nano Lett. 2024, 24, 11913-11920.

Q 37.8   Wed 12:45   HS XV

Role of Symmetry in Charge Transfer Complexation under Vibrational Strong Coupling — ●Anjali Jayachandran, Cyriaque Genet, and Thomas Ebbesen — University of Strasbourg, CNRS, ISIS and icFRC, 8 Allée Gaspard Monge, 67000 Strasbourg, France

The relation between symmetry and chemical reactivity has been explored for a long time. We reported earlier that symmetry also plays a key role in charge transfer (CT) complexation reactions under vibrational strong coupling (VSC) (1). We have now extended this study to a variety of donors and acceptors to gain further insight into how symmetry is acting on VSC. The experiments were conducted using the three isomers of trimethylbenzene (methyl groups in the 1,3,5; 1,2,4 and 1,2,3 positions on the benzene ring) as the donors with acceptors such as iodine, chloranil and 2,3-dichloro-5,6-dicyano-1,4-benzoquinone. It is observed that under vibrational strong coupling, there are large changes in the equilibrium constant, coefficient of absorption and the thermodynamic parameters for the different isomers. The changes seen in these parameters are dependent on the symmetry of the vibrational mode that is coupled to the IR cavity modes as well as the overall symmetry of the molecule. The result of this study confirms the relevance of symmetry in chemical reactivity under VSC and should be taken into consideration to steer reactions towards a desired outcome in this regime. 1. Y. Pang, A. Thomas, K. Nagarajan, R. M. A. Vergauwe, K. Joseph, B. Patrahau, K. Wang, C. Genet, T. W. Ebbesen, Angew. Chem. Int. Ed. 2020, 59, 10436.