Quasi-2D Wannier excitons interacting with optical phonons via Fröhlich interaction as a prerequisite for high-temperature superfluorescence in perovskites

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The recent discovery of high-temperature superfluorescence in hybrid perovskite thin films [1] has opened new possibilities for harnessing macroscopic quantum phenomena in quantum technology. This study aimed to elucidate the mechanism that enables high-temperature superfluorescence in these systems [2]. The proposed model describes a quasi-2D Wannier exciton in a thin film that interacts with phonons via the longitudinal optical (LO) phonon–exciton Fröhlich interaction. We show that the superradiant properties of the coherent state in hybrid perovskites are stable against perturbations caused by the LO phonon–exciton Fröhlich interaction. Using the multiconfiguration Hartree approach [3], we derive equations of motion for a single-exciton wavefunction, where the vibrational degrees of freedom interact with the Wannier exciton through a mean-field Hartree term. Superradiance is effectively described by a non-Hermitian term in the Hamiltonian. The analysis was then extended to multiple excited states using the semiclassical Hamiltonian as the basic model. We demonstrate that the ground state of the model exciton Hamiltonian with long-range interactions is a symmetric Dicke superradiant state, where the Fröhlich interaction is nullified. The additional density matrix-based consideration draws an analogy between this system and stable systems, where the conservation laws determine the nullification of the constant (momentum-independent) decay rate part. In the Wannier exciton–LO phonon system, nullification is associated with the absence of a momentum-independent component in the Wannier exciton–LO phonon interaction coupling function (see Figure1). In contrast, the coupling function for the Frenkel exciton – local phonon interaction does have a momentum-independent component (black line) that leads to inhomogeneous broadening and consequent loss of coherence in the collective molecular excitation and suppression of super-radiant emission. Therefore, a prerequisite for the high-temperature superfluorescence in hybrid perovskite thin films is the formation of Wannier excitons interacting with LO phonons via Fröhlich interaction. We also discuss new manifestations of the exciton-phonon interaction, which are associated with nonlinearity (e.g., soliton formation, instability, etc.). Notably, there is a significant difference between the case of the LO phonon-Wannier exciton Fröhlich interaction and that of the Frenkel exciton-local phonon interaction. In the latter case, the nonlinearity is local, leading to the formation of Davydov's soliton in 1D molecular aggregates. In contrast, the LO phonon-Wannier exciton Fröhlich interaction results in an essentially nonlocal nonlinearity reflecting a significant increase in the range of interaction. Our analysis predicts the emergence of a novel class of 2D nonlocal solitons in Wannier exciton systems, arising from the interaction between excitons and phonons. This work elucidates the conditions that allow coherent states to retain their superradiant properties at high temperatures, providing guidance for designing new quantum technologies.

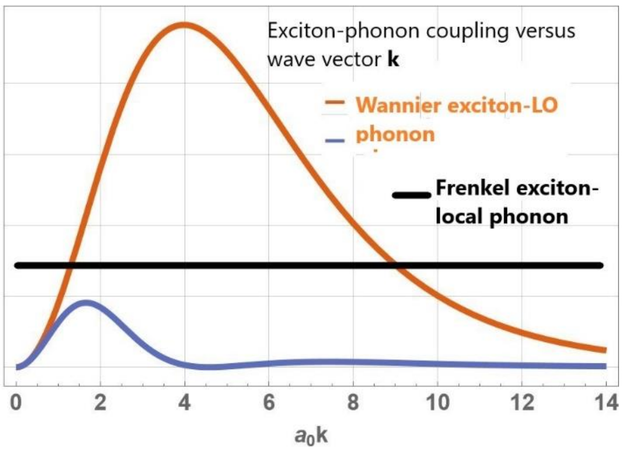

Figure 1: Exciton-phonon coupling versus wave vector. Orange and blue – Wannier exciton interacting with LO phonon, black - Frenkel exciton interacting with local phonon.

References

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