

Theory Colloquium

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“Cooperative quantum optics with molecules”

Abstract

Cooperative effects in complex, coupled quantum systems, cannot be understood by sole consideration of the individual constituents, as they arise from the interplay among them. Light-matter platforms provide an optimal playground for the observation and exploitation of quantum cooperative effects [1]. For example, structured subwavelength arrays of quantum emitters trapped in optical lattices, are ideal showcases of such cooperative behavior, as their optical response can be efficiently enhanced by controlling the hopping of surface excitations via the quantum electromagnetic vacuum induced dipole-dipole interactions.

While subwavelength separations are not easily achieved in standard quantum optics setups, molecular dimers and molecular aggregates (i.e. arrays of identical molecules, such as J- and H-Aggregates) can feature deeply subwavelength separations on the nanometer scale. The downside of such systems is the much more complex structure, which introduces coupling of electronic degrees of freedom with intra- and inter-molecular vibrations. We have introduced a quantum Langevin equations approach to electron-vibron interactions for single molecules subject to either classical or cavity quantum light fields [2]. The extension of this method to many particles allowed us to benchmark the scaling of cooperative effects such as super- and sub-radiance to molecular rings or chains, to quantify the effect of vibrations onto the operation of such systems as nanoscale coherent light sources [3] and to quantitatively describe couplings among collective electronic states via vibrations, in a process known as Kasha's rule [4].

[1] M. Reitz, C. Sommer, and C. Genes, Cooperative Quantum Phenomena in Light-Matter Platforms, PRX Quantum 3, 010201 (2022).

[2] M. Reitz, C. Sommer and C. Genes, Langevin approach to quantum optics with molecules, Phys. Rev. Lett. 122, 203602 (2019).

[3] R. Holzinger, S. Oh, M. Reitz, H. Ritsch and C. Genes, Cooperative subwavelength molecular quantum emitter arrays, Phys. Rev. Research 4, 033116 (2022).

[4] R. Holzinger, N. S. Bassler, H. Ritsch and C. Genes, Scaling law for Kasha's rule in photoexcited subwavelength molecular aggregates, arxiv: 2304.10236 (2023).

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