Influence of direct dipole-dipole interaction on the optical response of 2D materials in inhomogeneous infrared cavity fields

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The interaction between light and matter can be strongly enhanced by using nanophotonic cavities that localize light at the nanoscale. For example, the strong coupling between the cavity modes and vibrational modes of molecules results in the formation of vibrational polaritons [1],[2],[3], which can have a significant impact on the physical and chemical properties of the system [4],[5].

Our work considers a 2D material formed, by a self-assembled molecular monolayer or by a single layer of a Van der Walls material, coupled to an infrared nanophotonic cavity, potentially reaching the strong coupling regime. These systems are often modelled using classical harmonic oscillator descriptions or c-QED Hamiltonians that neglect the direct dipole-dipole interactions within the 2D material [5]. However, important effects can arise from these direct interactions, such as the emergence of new collective modes. To include these effects, we diagonalize the full Hamiltonian of the system (2D material and nanophotonic cavity), including the direct dipole-dipole interactions within the 2D material and their interaction with the nanocavity.

The main effect of considering direct dipole-dipole interactions on the optical properties of the hybrid system for homogeneous or slowly varying cavity fields is the renormalization of the effective energy of the bright collective mode of the 2D material that couples with the nanophotonic mode. However, we find that, for situations of extreme field confinement, fully including the direct interactions within the 2D material becomes critical to correctly capture the optical response, with many collective vibrational states participating in the response. Further, we derive a simple analytical equation which establishes the criteria for the need of dipole-dipole interactions in the description of the hybrid system beyond the standard renormalization [6].

References

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