Correlated Light-Matter Interactions in Cavity QED

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Many reactions in nature and engineering are catalyzed through the interaction between light and matter. Important examples include photosynthesis, the vision process, photo-chemical reactions, solar cells, and nanoplasmonics based on metamaterials. To better understand these complex processes, experimentalists in the field of cavity quantum electrodynamics (QED) have developed accurate methods to study quantum systems at the single-photon interaction limit (Nobel prize 2012). Further recent developments in this field are Fabry-Perot resonators with optical high-quality (high-Q) factors, circuit QED [1] and optomechanics [2].

In the electronic structure community, the quantized nature of the electrons is usually (approximately) incorporated, whereas the electromagnetic field is mostly treated classically. In contrast, in quantum optics, matter is typically simplified to models with a few levels, while the quantized nature of light is fully explored. In this work, we aim at treating both, matter and light, on an equal quantized footing.

To incorporate arbitrary geometries and matter distributions, we reformulate the Maxwell equations, by using the Riemann-Silberstein vector [3] into a matrix spinor representation similar to the Dirac equation. In the stationary limit, its eigenvalues and eigenmodes are the essential input for a quantized description of the electromagnetic field. Using this input, we present exact solutions for fully quantized prototype systems consisting of atoms or molecules placed in optical one-dimensional high-Q cavities and coupled to the quantized electromagnetic modes in the dipole or quadrupole coupling regime. We focus on spontaneous emission, atomic revivals, strong coupling phenomena, dipole-dipole couplings including van der Waals interactions, and Förster resonance energy transfer (FRET), all beyond the rotating wave approximation. These results are compared to a propagation of coupled Maxwell-Schrödinger systems, using the matrix spinor representation on a real-space grid.

The highlighted differences between these two schemes have implications for a future development of a time-dependent density functional theory formulation of quantum electrodynamics [4,5] and further allow to study modifications to the classical Maxwell equations for correlated multi-photon configurations. In the future, both can be used to open the new field of correlated spectroscopy [6].

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