Ab initio description of spatiotemporal dynamics of multi-level atoms resonantly coupled to plasmonic materials

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Abstract

The research in plasmonics has been centered at linear optics of plasmonic structures with several attempts to go beyond linear regime and consider nonlinear properties of materials including noble metal nanoparticles aiming for a wide variety of nonlinear phenomena to appear. A newly emerging research field of nonlinear plasmonics takes two obvious routes, namely, combining linear plasmonic systems with highly nonlinear media or taking into account strongly inhomogeneous electromagnetic fields associated with surface plasmons that lead to spatial dependence of conductive electron density in metals and hence result in nonlinear phenomena such as second harmonic generation. The first route deals with nonlinear media at the macroscopic level while even a single atom or molecule located near plasmonic structure at resonant conditions may be considered as a nonlinear optical system due to high electromagnetic near-fields. For example, it is well known that a two-level atom exposed to strong resonant EM radiation has an average dipole moment that may be written as a sum over odd powers of the electric field amplitude. Such dipole coupled to plasmon fields via polarization currents in Maxwell equations results in a nonlinear optical system and obviously is able to lead to new venues of research now including nonlinear optics. It thus important to develop approach capable of capturing both size effects at the nanoscale and the time dynamics of electromagnetic fields. Generally speaking, such approach has to take into account Maxwell equations capturing electromagnetic waves and quantum dynamics of atoms or molecules in the vicinity of plasmonic materials. The latter can be accomplished by employing Bloch formalism. The resulting system of coupled Maxwell-Bloch equations contains the most accurate description of nonlinear phenomena driven by surface plasmon waves. I will discuss new concepts of atom-plasmon interactions at the nanoscale based on rigorous numerical solutions of self-consistent full-wave vector Maxwell-Bloch equations in two and three dimensions. Various nonlinear phenomena such as self-induced transparency will be considered. I will also discuss the Brumer-Shapiro scheme of coherent control implemented at the nanoscale.