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Cavity quantum electrodynamics in application to plasmonics and metamaterials

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Frontier quantum engineering tasks require reliable control over light-matter interaction dynamics, which could be obtained by introducing electromagnetic structuring. Initiated by the Purcell's discovery of spontaneous emission acceleration in a cavity, the concept of electromagnetic modes' design have gained a considerable amount of attention due to development of photonic crystals, micro-resonators, plasmonic nanostructures and metamaterials. Those approaches, however, offer qualitatively different strategies for tailoring light-matter interactions and are based on either high quality factor modes shaping, near field control, or both. Remarkably, rigorous quantum mechanical description might address those processes in a different fashion. While traditional cavity quantum electrodynamics tools are commonly based on mode decomposition approach, few challenges rise once dispersive and lossy nanostructures, such as noble metals (plasmonic) antennas or metamaterials, are involved. The primary objective of this review is to introduce key methods and techniques while aiming to obtain comprehensive quantum mechanical description of spontaneous, stimulated and higher order emission and interaction processes, tailored by nanostructured material environment. The main challenge and the complexity here are set by the level of rigorousity, up to which materials should be treated. While relatively big nanostructured features (10 nm and larger) could be addressed by applying fluctuation–dissipation theorem and corresponding Green functions' analysis, smaller objects will require individual approach. Effects of material granularity, spatial dispersion, tunneling over small gaps, material memory and others will be reviewed. Quantum phenomena, inspired and tailored by nanostructured environment, plays a key role in development of quantum information devices and related technologies. Rigorous analysis is required for both examination of experimental observations and prediction of new effects.

1. Introduction

The ability to control light phenomena is essential for achieving various functionalities in majority of opto-electronic technologies, having inherent optical components. Continuous tendency in size reduction together with recent nanotechnological advances brought investigations of nanostructured devices into practical implementations. Noble metal-based

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structures, being the focus of this review, find use as auxiliary components in a large span of applications, among them biomedicine [1], photovoltaics [2], flat optical devices [3], sensing [4] and many others. The full potential of those elements has not yet been fully explored, especially with regards to the context of quantum applications [5,6,7,8]. One of the most prominent demands, to be achieved in order to enable those, is to confine and tailor propagation of electromagnetic energy at nanoscale.

One of the very promising and already proven approaches for controlling electromagnetic fields on nanoscale is to employ noble (plasmonic) metals, such as silver and gold[1]. The key optical property of those materials is their negative permittivity at the visible and infrared spectral ranges. Negative epsilon materials, being shaped into geometrical structures, have a unique property of supporting localized plasmon resonances (LPR) with the resulting phenomenon of nano-scale electromagnetic energy confinement [9]. The ability of plasmonic structures to concentrate light beyond classical diffraction limit [10–12] has the key advantage for tailoring light-matter interactions, opening roots for novel cavity quantum electrodynamics (CQED) phenomena - plasmonic CQED (PCQED). The major fundamental difference between traditional CQED and PCQED is in the way the interaction strength is controlled by an electromagnetic environment. The most prominent example, underlining differences between those approaches, is the change of spontaneous emission rates of an emitter in respect to a free space (Purcell effect [13]). While free space vacuum fluctuations of electromagnetic modes are solely dependent on fundamental physical constants, they can be modified by shaping surrounding material environment, in spite of the overall conservation of the total density of states available for radiation [14]. In fact, electromagnetic structuring leads to spectral and spatial redistribution of vacuum fluctuations. The relevant quantity here is the local density of states (LDOS) [9] – the proportionality constant, modifying interaction Hamiltonians and, consequently, resulting emission rates. The Purcell factor (the same quantity up to a constant) is proportional to the ratio of a quality factor of a confined mode to its volume (Q/V) and can be influenced by manipulating both of the quantities. Traditional CQED affects the interaction dynamics by exploring influences of quality factors, while PCQED acts via modal volumes for the most. This fundamental difference has to deal with optical properties of material components and geometries. While dielectric cavities could have quality factors as high as $\sim 10^{10}$ [15,16], their modal volumes are bounded from below by classical diffraction limit (roughly, a cubic wavelength in a material). On the other hand, plasmonic structures could confine energy to small regions, while associated quality factors are usually not exceeding hundreds [17], with few exceptions available [18]. From the theoretical standpoint, the difference between CQED and PCQED is in the way electromagnetic field is quantized. Furthermore, standard definitions of Purcell factors in PCQED could lead to ambiguous results and might be used only for preliminary estimations [19]. Main reasons, leading to the contradiction with classical Purcell theory, are presence of additional nonradiative decays channels and open nature of resonators, complicating rigorous definition of modal volumes. Those aspect were addressed in a series of recent works (e.g. [20,21]) and the revised formalism of Purcell factor was developed in [22]. Some of the above mentioned aspects along with other probable impacts of absorptive and dispersive nature of plasmonic materials on light-matter interaction dynamics will be discussed in details hereafter.

Large-scale nanostructures, having shaped plasmonic particles as unit cells[2], enables achieving modified interactions for macroscopic number of emitters. While, studies of modified light-matter interactions in CQED are typically restricted to single-mode/single-emitter scenarios, collective phenomena, tailored by nanostructured media (metamaterials [23]) gained a considerable attention due to a span of promising frontier opto-electronic applications. For example, so-called hyperbolic metamaterials (highly anisotropic artificially created crystals) hold a promise of broadband non-resonant enhancement of spontaneous emission rates [24] with few experimental demonstrations already available (e.g. [25,26]).

The main purpose of this review is to discuss few phenomena in the field of PCQED (including aspects of metamaterials) by introducing major theoretical tools and relating them to existing experimental reports. Light-matter interaction dynamics on nano-scale could be rather involved and, in contrary to well-defined isolated CQED-like problems, require conceptual mesoscopic type of treatment. A generic reason for this occurrence is related to the fact that nano-scale systems could not obey classical laws of electrodynamics (classical permittivity description breaks down), but yet too big to be treated with full atomistic modeling [27]. One of the major questions, to be addressed before approaching a mesoscopic problem of this kind, is whenever classical or quantum treatment of a phenomenon is required. In majority of cases, full quantum-mechanical treatment of problems, lacking of analytical solutions, requires heavy and even unobtainable computational resources. In order to bypass those limitations, capturing key mechanisms governing phenomena to the lowest level of sophistication is needed. Few examples, demonstrating this general concept, will be revised and discussed. It is worth noting few recent reviews on the topic of quantum plasmonics and metamaterials, giving a broad survey of theoretical and experimental works, e.g. [8,28,29–31]. The focus of this review, apart from revising a set of major tools, is to introduce approaches for mesoscopic models, which might require a unique treatment.

The review is organized as follows: first, general aspects of electromagnetic field quantization in the presents of material bodies will be presented. Next, main geometries, subject to PCQED studies, will be introduced and few recent achievements in the field will be reviewed. Mesoscopic models, enabling to bridge a gap between microscopic and macroscopic descriptions of PCQED, will follow. Concepts of emulation experiments, aiming to address complex phenomena by means of analog

[1] There is a venue for investigating and developing additional materials for plasmonics, such as aluminium [188] and transparent oxides [189].

[2] The concept of quantum metamaterial with semiconductor-based unit cells was introduced too (e.g. [190–192]).