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Extended Abstract

Nano science, Nanotechnology, Graphene & 2D material: Quantization of plasmon-polaritons with localized nanostructures- H R Jauslin - ICB-Laboratoire Interdisciplinaire Carnot de Bourgogne, Université Bourgogne Franche-Comte, France

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Abstract

We present a new approach to the quantization of the electromagnetic field in configurations involving nanostructures of finite size. The presence of dispersion and dissipation prevents the quantization of the phenomenological Maxwell equations with a space and frequency dependent dielectric coefficient, which is standardly used in the classical treatment of such systems. We follow the approach, started by Hopfield, in which the medium is described by a microscopic Hamiltonian system composed of a harmonic oscillator, which interacts with the electromagnetic field by a dipole coupling. This type of model was used by Huttner and Barnet to construct a quantum model for a bulk homogeneous. Using techniques developed by Ugo Fano, they diagonalized the Hamiltonian and characterized the plasmon-polariton excitations as the fundamental excitations of the system. This work was later extended to inhomogeneous media, and applied to the treatment of different phenomena, like the Purcell effect in spontaneous emission or like Casimir effects, among many others. It was noted by several authors that the diagonalization of inhomogeneous systems adapting the Fano technique leads to results that seem incomplete when the medium is of finite size. In particular, these results do not yield the correct properties when one takes the limit of vanishing size of the medium or of the coupling. It was however not clear what step in the diagonalization procedure is responsible to this apparent contradiction. We are going to present a different approach to the diagonalization that is conceptually close to the method of Bogoliubov transformations, and which leads to a diagonalization of the quantum plasmon-polariton model that gives a complete result and yields the correct limit when the coupling vanishes.

Localised surface plasmons (LSPs) are quantized plasma oscillations formed near the surfaces of metal nanoparticles. In contrast to propagating surface plasmons, which often require carefully-constructed optical arrangement for phase matching, LSPs can be excited easily by direct irradiation of light. It is well known that LSPs can focus light to nanometre scale and strongly enhance the electric field near nanometals, which is called the antenna effect. The research field dealing with such plasmon characteristics is called "plasmonics", and has been extensively investigated. In fact, many applications of LSPs, such as bio-sensing, solar cell and molecular fluorescence enhancement have been reported.

For several years, the studies focused on the quantum nature of plasmons are now rapidly evolving, in which the light field interacts with matter at nanometre scale and plasmons play a role in controlling the light-matter interactions at the quantum level. This new research field is called "quantum plasmonics". Quantum plasmonics has opened up a new frontier in the study of the fundamental physics of LSPs, namely vacuum Rabi splitting and the Fano effect, and now provide new potential applications of plasmons, such as single-photon sources, entangled-photon sources and quantum-controlled devices at the nanometre scale.

As is well known, however, the optical responses of LSPs are generally analysed using classical electromagnetism, in spite of the fact that LSPs are quantized plasma oscillations. This is because that the LSP antenna effect can be understood without the quantum nature of plasmon and can be explained simply by Maxwell's equations. However, quantum plasmonics requires a quantum-mechanical treatment of plasmons, in other words, the second quantization of plasmons,. In particular, the understanding of the quantum properties of a single LSP, namely dipole moment and its relaxation rate, is required to understand the vacuum Rabi splitting in LSP-matter interaction, because the strong coupling is realized when the coupling constant rate g of LSP-matter interaction is lager than the relaxation rates of the LSP and the matter.

Conventionally, the second quantization of surface plasmons is based on the framework of cavity QED theory and is described by the boson model. In general, the boson model is justified by the fact that the Pauli exclusion principle hardly affects collective excitation. For example, in a lattice model of condensed-matter physics, the prohibition of two excitations at the same lattice corresponds directly to the Pauli exclusion principle. This prohibition component constitutes only N−1 of a collective excitation mode, where N is the number of

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lattice, and therefore can be ignored for $N \gg 1$. This boson approximation is valid for collective excitations of general quasiparticles. Recently, however, it has been experimentally reported that LSPs are optically saturated by high-intensity light. Absorption saturation arises as a result of strong optical nonlinearity and cannot be explained by the above boson model. In contrast to propagating surface plasmons formed at the surface of planar metallic films, LSPs are localised in literally at nanometre scale. Therefore, the Pauli exclusion principle might be non-negligible, especially for small metal nanoparticles.

In this study, we propose a simple model of a saturable LSP for small metal nanoparticles using an effective dipole approximation. Our strategy is to directly compare the classical linear optical response of an LSP with that obtained from a saturable quantum two-level system. The second quantization is then performed by replacing a classical polarizability of the LSP with a quantum dipole operator described by the two-level system. Taking an ellipsoidal nanometal as an example, we introduce an optical response function of an LSP, based on the optical Bloch equations, and validate our method by analysing in detail the size dependence of the plasmon resonance frequency and the relaxation decay rate of a single LSP. Our numerical results show that the plasmon resonance frequency and spectral linewidth decrease as the aspect ratio of the ellipsoidal nanometal increases, which is similar to the size dependence observed in early experiments

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