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Quantum theory of radiative decay rate and frequency shift of surface plasmon modes

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In this work we study, in the time domain, the interaction between localized surface plasmons and photons in arbitrarily shaped metal nanoparticles, by using the Hopfield approach to quantize the plasmon modes, where the electron oscillations are represented by a harmonic matter field linearly coupled to the electromagnetic radiation. The plasmon-photon coupling gives rise to dressed plasmon modes. We have found that the radiation does not induce a significant coupling among the different quasielectrostatic plasmon modes for particles of size up to the plasma wavelength, but causes a frequency shift and an exponential decay in time of the modes. By solving the equations governing the expectation values of the plasmon creation and annihilation operators, we obtain a closed-form full-wave expression for the decay rate and for the frequency shift of the plasmon modes. It is nonperturbative and it only depends on the surface charge distribution of the quasielectrostatic plasmon modes. We validate the expression against the Mie theory for a nanosphere of radius comparable to the plasma wavelength. Eventually, we investigate the decay rate and the frequency shift of the plasmon modes in isolated and interacting nanoparticle of noncanonical shape, as their size increases up to the plasma wavelength.

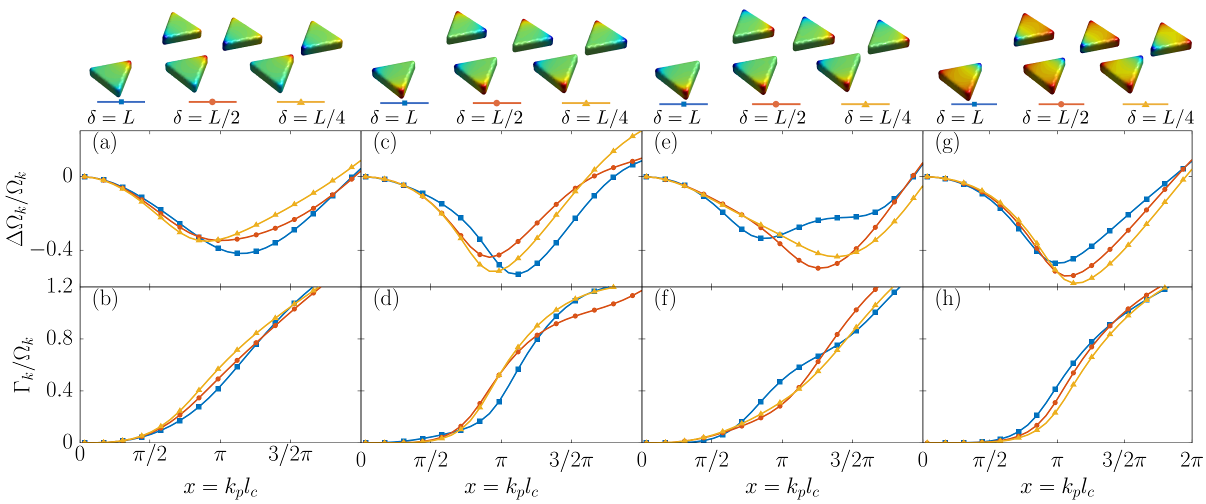

Figure 1. (a),(c),(e),(g) Frequency shift $\Delta\Omega_k$ (with respect to the quasistatic resonance frequency Ω_k), red normalized to Ω_k; and (b),(d),(f),(h) decay rate Γ_k, normalized to Ω_k, of the first four modes of a bow-tie antenna red (1-st mode (a-b), 2-nd mode (c-d), 3-rd mode (e-f), 4-th mode (g-h)) as a function of the size parameter $x = k_p l_c$, where $l_c = L/2$. Different edge-edge gap sizes δ have been considered, namely $\delta = 2l_c = L$ red (blue line with squares), $\delta = l_c = L/2$ (red red line with circles), $\delta = l_c/2 = L/4$ (red yellow line with triangles). The surface charge density distributions of the plasmon modes are shown above the corresponding panels.

The developed approach leads, as expected from the correspondence principle, to the same outcome obtained of the classical Hamiltonian equations. Nevertheless, it constitutes a first step toward a full-wave time-domain quantum framework based on a modal analysis for describing the interaction between a quantum emitter, an arbitrary shaped metal nanoparticle, and the electromagnetic radiation.

References

[1] C. Forestiere et al., "Magnetoquasistatic resonances of small dielectric objects" *Physical Review A* **102**, 043704, October 2020