Quenching revisited: theory of strong coupling between quantum emitters and metal nanostructures

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Compared to dielectric cavities, the gain/loss balance which lies at the core of laser physics is shifted to new levels when using plasmon polaritons. These electromagnetic (EM) modes supported by metal nanostructures are deeply sub-wavelength, which enables a better coupling to the gain medium but comes along much higher ohmic losses. As a result, plasmonic nanolasers show potentially very high operating speeds and low thresholds. However, a sad truth in plasmonic nanolasers design is that the gain medium closest to the metal surface, supposedly the most coupled to the evanescent plasmons, is actually blind to the lasing mode. It is indeed quenched: all its energy is drained very efficiently into higher momentum, non-radiative EM excitations. Quenching is also detrimental in fields such as Surface-Enhanced Raman Scattering or optical antennas.

Here, we theoretically examine quenching from a quantum point of view, and show that the emergence of light-matter strong coupling can mitigate this issue. Using a macroscopic QED formalism based on multiple scattering techniques for the Green’s function [1, 2], we consider the two canonical problems of the metal sphere and plane. In both cases, it is found that because of their quasi-degenerate nature, the high momentum EM modes do not behave as an efficient Markovian bath, but rather as a single pseudo-mode with a limited decay efficiency.

FIG. 1. (a) Far-field spontaneous emission spectrum for a regular shell of dyes (see inset of (b)) at different distances around a silver nanoparticle (NP), illustrating the transition from quenching (broadening) to strong-coupling (splitting). (b) Inset, spatial-distribution of the light spectrum on the surface of the nanoparticle (NP). The spots underneath each dye follow the distribution of pseudo-modes with constrained draining efficiency, allowing for the build-up of a collective interaction with the dipole mode. Main frame: population dynamics of a single dye near the NP, showing a reversible exchange of energy with the corresponding pseudo-mode, signature of the strong coupling regime.