

Luminescent Activities of a Two-level Molecule with Population Inversion

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In the previous works of our group, a stationary population inversion [1] and its up-converted luminescence [2] have been theoretically demonstrated for a two-level molecule near gold nano-particles sustaining localized surface plasmon-polaritons(LSPP). These phenomena result from the resonant coupling between the molecule and Mollow triplet sideband caused by the interaction between the strong coherent field and LSPP. We modeled LSPP by a two-level system, because the absorption saturation of LSPP has been observed [3]. However, the recent experiment reports the characteristic intensity dependence of the LSPP scattering that cannot be reproduced by the two-level model [4].

Thus, in this contribution, we employ an anharmonic three-level system as a model of LSPP considering the results in Ref. [4]. A two-level molecule is set to resonate with the upper sideband of the Mollow triplet by LSPP. The decay of each system is described by the Markovian master equation, and the output power spectra are obtained from the quantum regression theorem. The figure shows luminescence spectra for three-level model, in which we select parameters reproducing the scattering saturation in Ref. [4]. The molecular spectrum has a peak at its resonance energy (solid line). We can confirm the up-converted photoemission activity of the two-level molecule with population inversion in the case of the reasonable parameters of LSPP model for more realistic situation. The power spectrum of LSPP shows the down-conversion (dotted line) which guarantees the energy conservation.

The above results indicate potential applications of the present system for the novel up-converted coherent light source.

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[3] T. S. Ahmadi, *et. al.*, J. Phys. Chem. **100**, 8053 (1996).

[4] S. -W. Chu, *et. al.*, Phys. Rev. Lett. **112**, 0177402 (2014).

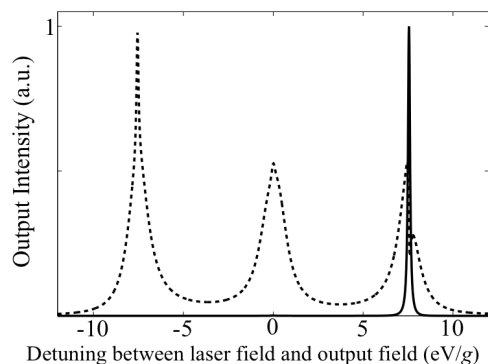


Figure: The output spectra of the molecule (solid line) and LSPP (dotted line)