# Optical frequency conversion dynamics affected by inter-molecule correlations near a metallic nanostructure

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Localized surface plasmons on nanoscale metal nanostructures convert free-propagating light into strongly localized fields, which interact efficiently with molecules near the metal surface. This characteristic enables us to regard the metal nanostructures as "optical antenna" [1]. We have theoretically elucidated that quantum interference in an antenna-molecule coupled system suppresses the photon-dissipations by the antenna, and leads to efficient two-photon up-conversion in the molecule [2]. One of the key elements to control such quantum interference is the antenna-molecule coupling constant. Because these results are based on one-dimensional input-output theory, we have not assumed specific structures of the antenna-molecule coupling system. As the next phase, we will consider specific structures and their integration toward application to unprecedented optical devices.

In this work, we numerically demonstrated the optical response of a molecule located near gold nanoblocks with several nanometer separations by using the discrete dipole approximation (DDA) method. We estimate the effective antenna-molecule coupling constants and propose specific structures with optimal antenna-molecule coupling for efficient up-conversion. We also demonstrate optical responses on the periodically arranged nanoblocks and molecules. It is found that the inter-molecule correlations mediated by radiation fields shift the absorption peaks of the molecules. It is known that such correlations affect the emission characteristics of molecules [3]. Then, in order to discuss in detail the effects of inter-molecular correlations on the up-conversion processes, we analyze the up-conversion processes in more than one molecule. The analysis is based on quantum master equation considering specific metal structures in DDA, and showed that the inter-molecular correlations actually influence the up-conversion processes. In addition, we elucidate a dependence on the number and the position of the molecules. These results will guide us to realize efficient up-conversion devices.

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