Effects of a Plasmonic Nanostructure on Kerr Nonlinearity in a Four-Level Quantum System

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Abstract
Recent studies have revealed that several nonlinear optical phenomena can be strongly modified in quantum systems near plasmonic nanostructures. Examples of these phenomena include up-conversion processes [1], second harmonic generation [2], Kerr nonlinearity [3],four-wave mixing [4], nonlinear optical rectification [5], induced transparency [6,7], and gain without inversion [7,8].
In this work we continue our study on the effects of the presence of a plasmonic nanostructure (see Fig. 1, left) on the optical properties of a four-level double-V-type quantum system (see Fig. 1, right). In the quantum system under study the transitions $|2\rangle$, $|3\rangle$ to $|1\rangle$ are in energies that are influenced by the surface plasmons of the metallic nanostructure while the transitions $|2\rangle$, $|3\rangle$ to $|0\rangle$ are in quite different energies and are not influenced by the surface plasmons, so they interact with free space vacuum. This quantum system shows quantum interference in spontaneous emission near the plasmonic nanostructure [9-12]. In a previous study [6], we have shown that this system can also exhibit induced transparency and slow light when it interacts with a weak probe laser field. Here, we study the effects of the plasmonic nanostructure on Kerr nonlinearity.
For the plasmonic nanostructure we consider a two-dimensional array of metal-coated silica nanospheres (see Fig. 1, left), where the metallic part is described by a Drude-type dielectric function $\varepsilon(\omega) = 1 - \omega_p^2/(\omega^2+i\omega/\tau)$, where ω_p is the plasma frequency and τ is the relaxation time of the conduction-band electrons of the metal. We calculate the relevant decay rates with a rigorous electromagnetic Green's tensor technique, using a layer-multiple-scattering method for electromagnetic waves [9-11,13,14] (see results in Fig. 2). It is evident that the spontaneous decay rate for a dipole oriented parallel to the surface of the plasmonic nanostructure is suppressed relative to vacuum and exhibits significant suppression in the frequency region from $0.6\omega/\omega_p$ to $0.7\omega/\omega_p$, with the actual value becoming significantly smaller that the free space decay rate. In the same frequency region the spontaneous decay rate for a dipole oriented perpendicular to the surface of the plasmonic nanostructure is also suppressed but it remains larger than the free space decay rate.
The quantum system is initially in state $|0\rangle$ and interacts with a linearly polarized probe laser field, which couples the lowest state $|0\rangle$ with states $|2\rangle$ and $|3\rangle$. We use a density matrix methodology for the theoretical description of the optical properties of the laser field. Assuming a weak laser field, the density matrix elements can be calculated analytically up to third order with respect to the electric field amplitude. We use the relevant density matrix elements and determine the Kerr nonlinearity $\chi^{(3)}$. The imaginary and real part of $\chi^{(3)}$ are displayed in Fig. 3. Fig. (a) is in the absence of the plasmonic nanostructure, while figs. (b), (c) and (d) are in the presence of the plasmonic nanostructure. We see that due to the presence of the plasmonic nanostructure, both the real and imaginary part of $\chi^{(3)}$ are significantly modified. This modification depends on the distance of the quantum system from the plasmonic nanostructure.

References

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Figures

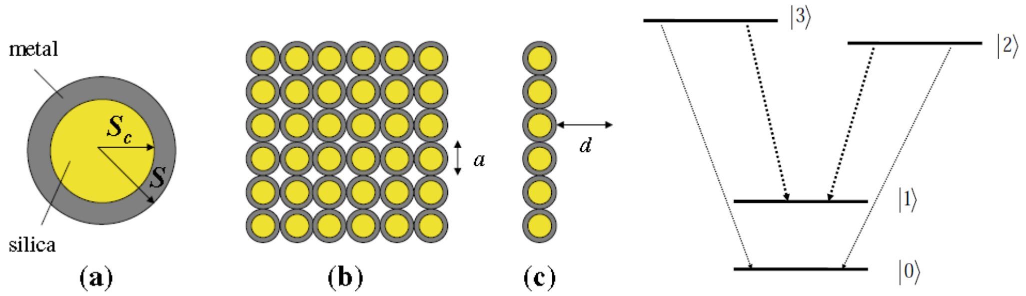

Figure 1: *Left:* (a) Metallic nanoshell made from a silica core of radius S_c and metal coating of thickness S-S_c. (b) Square lattice of metallic nanoshells (monolayer) with period α. (c) Side view of the monolayer where d is the distance of the quantum emitter from the surface of a nanoshell. *Right:* The quantum system under study is a double-V-type system.

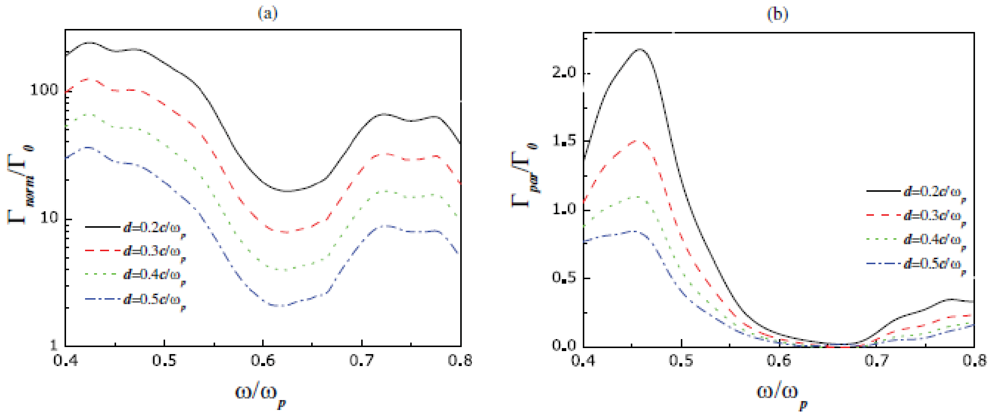

Figure 2: The spontaneous emission rate for a dipole which is normally oriented (a) [parallel oriented (b)] with respect to the plasmonic nanostructure as a function of the emitter frequency for various distances d from the plasmonic nanostructure (with exact values of d shown in the inset). Γ_0 is the decay rate in the vacuum.

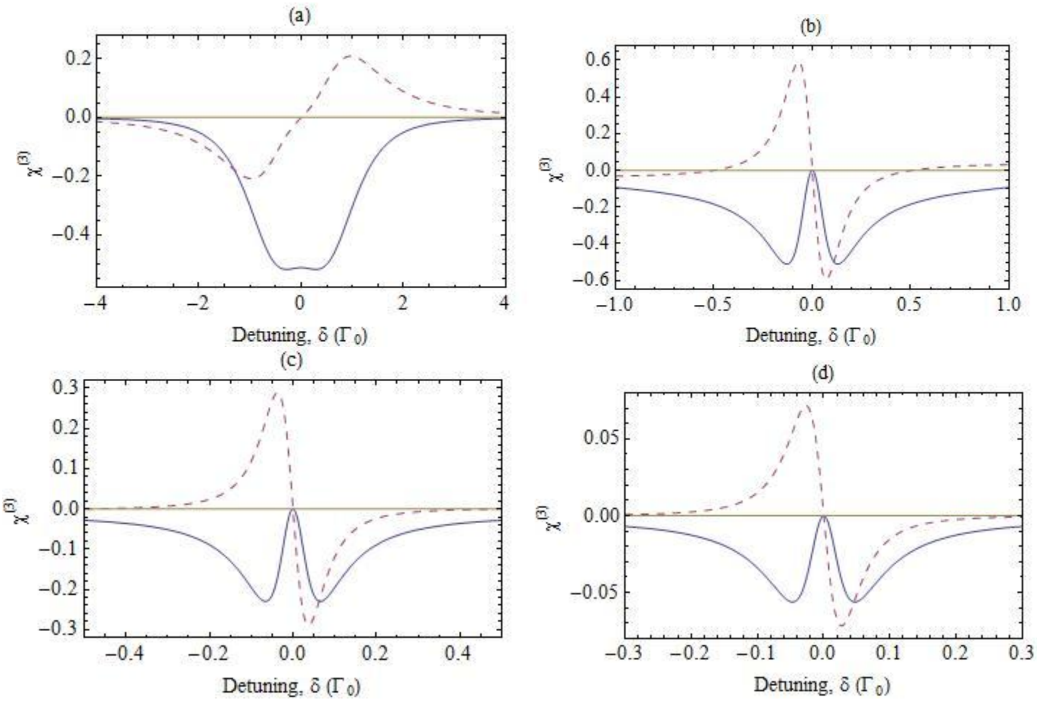

Figure 3: The real part (dashed curve) and imaginary part (solid curve) of Kerr nonlinearity [in units $2N\mu^4/(3\hbar^3\varepsilon_0\Gamma_0^{\ 3})$], where N is the atomic density. Here, the central Bohr frequency (calculated from state |0> to the middle of states |2> and |3>) is ω = 0.632ω_p, the frequency difference of the upper levels is $\omega_{32} = \Gamma_0$ and in (b) d = 0.5c/ω_p, (c) d = 0.4c/ω_p, and (d) d = 0.3c/ω_p. The detuning is defined as $\delta = \omega_{las} - \omega$. The spontaneous decay rates from states $|2\rangle$ and $|3\rangle$ to $|0\rangle$ are taken zero.