Simple Analytical description of CNT nonlinear response enhanced by metamaterials

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Abstract

We present an analytical model for describing complex dynamics of a hybrid system consisting of resonantly coupled classical resonator and quantum structures. Classical resonators in our model correspond to plasmonic nano-structures of various geometries, as well as other types of nano- and microstructures, optical response of which can be described classically. Quantum resonators are represented by atoms or molecules, or their aggregates (for example, quantum dots, carbon nanotubes, dye molecules, polymer or bio molecules etc), which can be accurately modeled only with the use of the quantum-mechanical approach. As a particular example of application of our model, we show that the saturation nonlinearity of carbon nanotubes increases multifold in the resonantly enhanced near field of a metamaterial.

1. Introduction

For describing classical and quantum systems coupled together a special approach is required. It was originally developed to model the dynamics of lasers where the classical system is normally represented by an optical (mirror) resonator, while the quantum system – by amplifying medium [1]. With the rapid development of nanotechnology it has become possible to engineer and study hybrid quantum-classical systems at the nanometer scale such as metallic nano-resonators and their arrays (i.e. metamaterials) combined with quantum dots, carbon nanotubes or dye molecules [2 - 5]. The optical response of the metallic nano-resonator can still be satisfactory modeled by the harmonic oscillator equations with appropriately chosen parameters [6], while quantum formalism using density matrix approach is to be used to describe quantum ingredients. This quantum-classical approach allows modeling analytically a wide range of optical and plasmonic effects in the hybrid quantum metamaterials, such as loss compensation, enhancement of nonlinear response and luminescence, etc.

2. Master set of equations

In this paper we consider a Quantum System (QS) placed in the near-field zone of a Classical electromagnetic System (CS). The field produced by the CS affects QS that in turn acts on CS with its field. In addition, there is an external field of the incident light, which interacts with both CS and QS. The actual number of the harmonic-oscillator equations required to adequately describe CS depends on its particular structure [6]. For the illustration purpose we will restrict our analysis to just one harmonic-oscillator equation, which should not limit the generality of our approach. The dynamics of QS is modelled using the density matrix formalism for 2 levels. The system of respective equations for slow amplitudes is:

$$\begin{cases} \dfrac{d\rho_{12}}{dt}+\rho_{12}\left(\dfrac{1}{\tau_2}+i\left(\omega-\omega_{21}\right)\right)=\dfrac{i\alpha_x x^* N}{\hbar}+\dfrac{i\mu_{QS}A^* N}{\hbar} \\ \dfrac{dN}{dt}+\dfrac{\left(N-N_0\right)}{\tau_1}=\dfrac{i\alpha_x\left(x\rho_{12}-x^*\rho_{12}^*\right)+i\mu_{QS}\left(A\rho_{12}-A^*\rho_{12}^*\right)}{2\hbar} \\ 2\left(\gamma-i\omega\right)\dfrac{dx}{dt}+\left(\omega_0^2-\omega^2-2i\omega\gamma\right)x=\alpha_\rho\rho_{12}^*+\chi A \\ N_0=\dfrac{\left(W\tau_1-1\right)}{\left(W\tau_1+1\right)},\quad \tau_1=\dfrac{\tilde{\tau}_1}{W\tilde{\tau}_1+1} \\ \alpha_\rho\sim\mu_{QS}\chi \\ \alpha_x\sim\mu_{QS}\mu_{CS} \end{cases} \tag{1}$$

Here μ_{QS} is the dipole moment of QS, μ_{CS} is the effective dipole moment of CS; $N=\rho_{22}-\rho_{11}$ is the population difference (in the absence of pump $N_0 = -1$, $N_0 > 0$ corresponds to the regime of amplification, $N_0 < 0$ - to losses); ρ_{22}, ρ_{11} and ρ_{12}, ρ^*_{12} are the diagonal and non-diagonal matrix density elements, respectively; τ_2 and $\tilde{\tau}_1$ are the constants describing phase and energy relaxation processes due to the interaction with a thermostat; $\omega_{21} = (E_2 - E_1)/\hbar$ is the transition frequency between levels 2 and 1; γ and ω_0 are the loss coefficient and resonance eigen-frequency, and χ^{-1} is the effective kinetic inductance of the nano-resonator. The dimensionless variable x corresponds here to one the dynamic characteristics of the oscillator.

3. CNTs combined with metamaterials

Here we illustrate application of our approach for modelling enhanced nonlinear optical response demonstrated recently in a plasmonic metamaterials combined with carbon nanotubes (CNT) [5]. In such a hybrid quantum-classical system the metamaterial structure works as a light concentrator enhancing optical fields locally. In the resonance case the intensity of the local fields can become significantly higher than that of the incident wave and therefore substantially affect the dynamics of CNT response. The nonlinearity of CNT appears due to the saturation induced by the direct pumping of such a two-level-like quantum system; the enhancement of the nonlinearity is caused by the addition field transfer of energy to CNT through the nano-resonator. This is described by the term $\dfrac{i\alpha_x x^* N}{\hbar}$ in the first equation of system (1). We introduce a relative transmission change according to the following expression:

$$\frac{\left(\dfrac{\Delta L_{CNT}}{L_{CNT}}\right)_{\sigma\neq 0,\,Resonance}}{\left(\dfrac{\Delta L_{CNT}}{L_{CNT}}\right)_{\sigma=0,\,Resonance}}=\frac{1+S_0\left(1+\dfrac{\sigma^2}{4\omega^2\gamma^2}\right)}{\left(1+S_0\right)\left(1+\dfrac{\sigma^2}{4\omega^2\gamma^2}\right)} \tag{2}$$

here $S_0=\dfrac{|A|^2}{|A_{s,0}|^2}$, $|A_{s,0}|^2=\dfrac{\hbar^2}{\mu\tau_1\tau_2}$, $\sigma=\dfrac{\alpha_x\chi}{\mu_{QS}}$; (2) clearly demonstrates that the relative increase of transmission for CNT in the presence of the metamaterial ($\sigma\neq 0$) is bigger than for CNT alone ($\sigma=0$) - see Fig. 1. In order to demonstrate the effect of the metamaterial on the absorption change due to saturation, we plotted the normalised absorption change $\dfrac{\Delta L_{CNT\&MM}}{L_{CNT\&MM}}$, shown in Fig. 2.

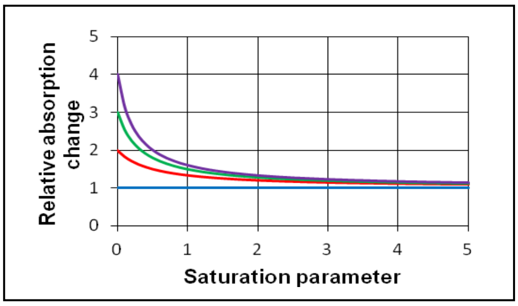

Fig. 1: Relative transmission (2) as a function of saturation parameter $S_0 = \frac{|A|^2}{|A_{s,0}|^2}$ for different values of coupling $\frac{\sigma^2}{4\omega^2\gamma^2} = 0,1,2,3$ („0“ - blue line, „1“ – red line, “2” – green line, “3” – violet line).

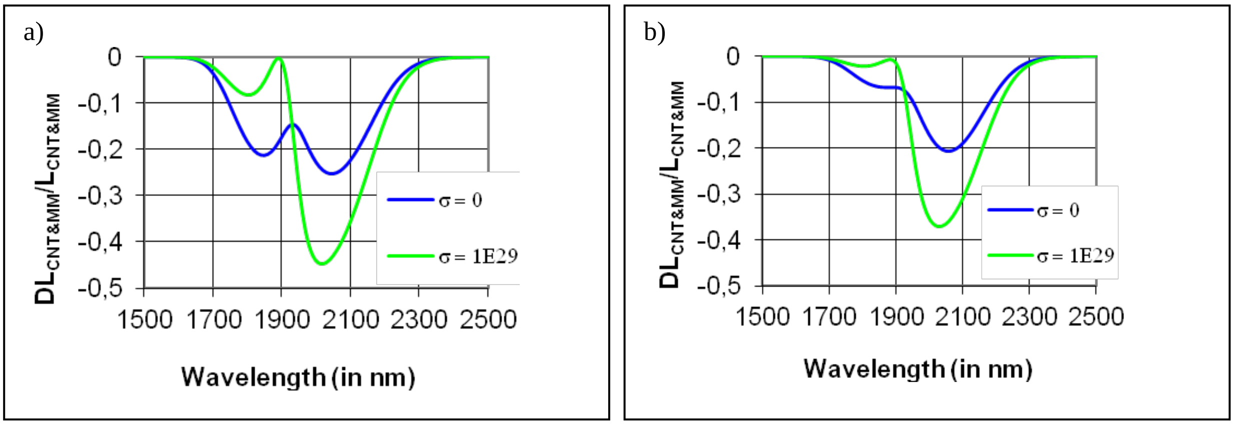

Fig. 2: Normalised absorption change spectrum of CNTs combined with metamaterial for a) homogeneous and b) inhomogeneous cases. Saturation parameter is $S_0 = \frac{|A|^2}{|A_{s,0}|^2} = 3$.

4. Conclusion

An analytical model for describing complex dynamics of a hybrid system consisting of interacting classical and quantum resonant structures has been developed. An application of this model for enhancement of saturation nonlinearity in the system of coupled CNT and metamaterial has been demonstrated. The model clearly demonstrates the effect of saturation nonlinearity enhancement due to the presence of the metamaterial.

References

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